PROCEEDINGS OF A SYMPOSIUM
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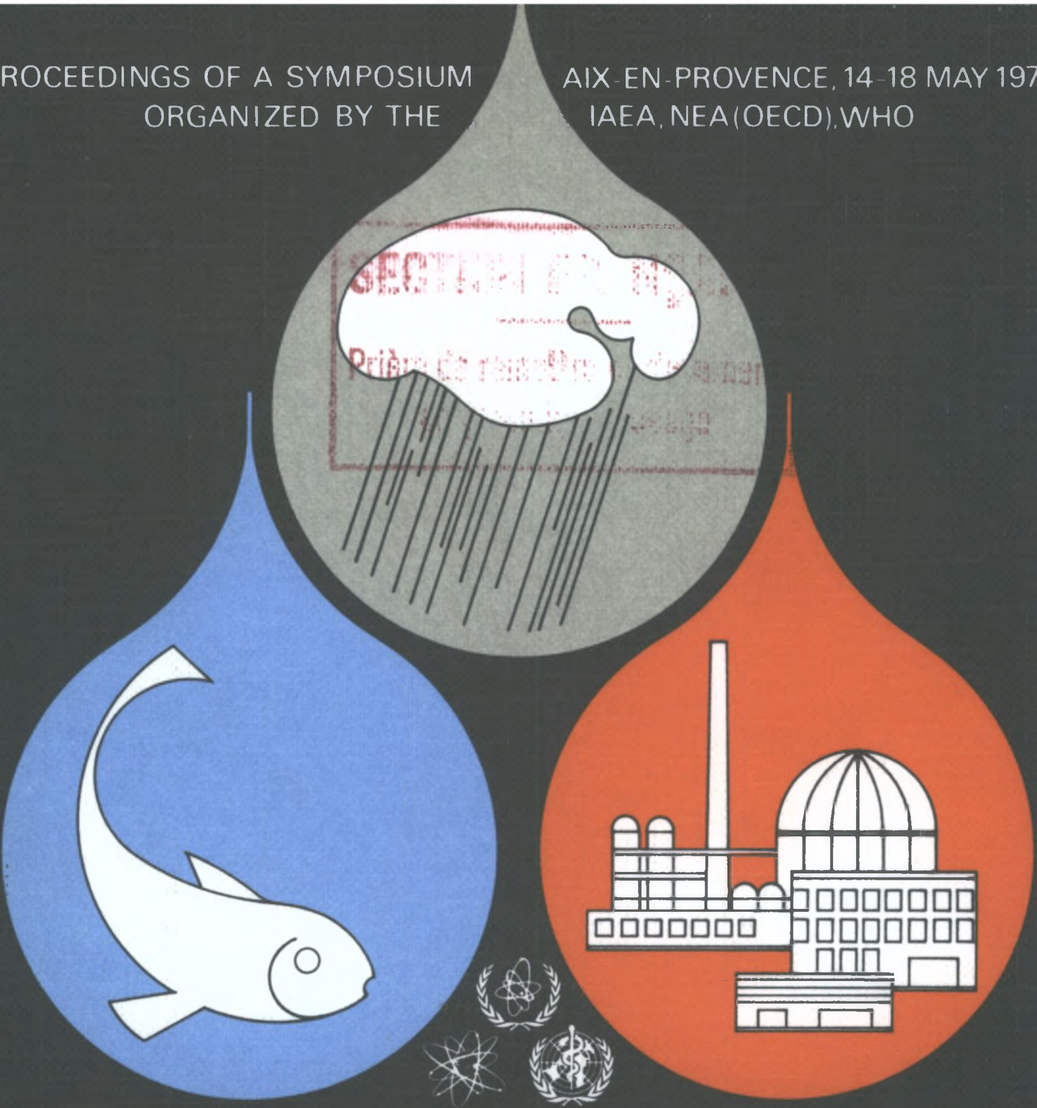
AIX-EN-PROVENCE, 14-18 MAY 1973



ENVIRONMENTAL BEHAVIOUR OF RADIONUCLIDES RELEASED IN THE NUCLEAR INDUSTRY

INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1973

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PROCEEDINGS SERIES

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ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY,
THE OECD NUCLEAR ENERGY AGENCY
AND THE
WORLD HEALTH ORGANIZATION
AND HELD IN
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FOREWORD

The release of radioactive materials in the nuclear industry and their behaviour in the environment have been the subject of two international symposia held respectively in New York in August 1970, on the Environmental Aspects of Nuclear Power Stations, and in Seattle, in July 1972, on the Interaction of Radioactive Contaminants with Constituents of the Marine Environment. Both these symposia were organized, and the proceedings published, by the International Atomic Energy Agency. A wide variety of topics were included, but relatively little attention was given to the environmental transfer processes other than those occurring in the seas.

The understanding of the behaviour and fate of radionuclides released in the nuclear industry is an essential step in a systematic assessment of the public health and environmental impacts of the growing use of nuclear energy for the production of electricity. It is expected that the number of nuclear facilities will increase markedly in the next few decades and that they will become widespread geographically. In spite of the fact that routine releases of radioactivity from various nuclear facilities are controlled to very low levels, it is appropriate that environmental behaviour of the radionuclides which are, or which potentially can be, released be understood well enough to permit a preliminary assessment of the impacts and development of adequate environmental monitoring systems.

To fulfil this need the International Atomic Energy Agency (IAEA), the OECD Nuclear Energy Agency (NEA) and the World Health Organization (WHO) convened a symposium in Aix-en-Provence from 14 to 18 May 1973. The Symposium attracted 210 participants representing 30 countries and 8 organizations. There were 49 papers presented from 12 countries and two international organizations and a panel discussion was held on the aims of and needs for environmental programs related to the nuclear industry.

The papers presented at the Symposium and the subsequent discussions emphasized the attention being given to the environmental aspects of nuclear programs. Although some participants were of the opinion that the degree of attention given to projections of potential radiation dose could be somewhat reduced owing to the small magnitude of the potential doses, there was widespread agreement that the systematic studies carried out with respect to releases of radioactive materials in the nuclear industry should be extended to releases of other pollutants so that meaningful comparisons of their impacts can be made. In such studies the sizeable interdisciplinary teams assembled at atomic energy research institutions in various countries could play a major role.

The present book contains the papers and the discussions. The joint organizers of the Symposium wish to acknowledge the co-operation of the French authorities, in particular the contribution made by the staff of the Commissariat à l'Energie Atomique.

EDITORIAL NOTE

The papers and discussions incorporated in the proceedings published by the International Atomic Energy Agency are edited by the Agency's editorial staff to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants.

For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies.

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Invited Paper

ENERGY, THE ENVIRONMENT AND NUCLEAR ENERGY LABORATORIES

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Abstract

ENERGY, THE ENVIRONMENT AND NUCLEAR ENERGY LABORATORIES.

Three issues that derive from the apparent conflict between the demand for more energy by the earth's people and the simultaneous requirement to maintain a healthy environment are at the core of the discussion here. The nature of the public concern for the environment and how it arose, the demand for energy, both in the United States of America and in the rest of the world, and the nature of the institutions seemingly best suited to understand and resolve these concerns are discussed.

INTRODUCTION

That we are on a planet is not new, but the full realization that we are riding on a truly isolated Spaceship Earth on an unaided voyage through the solar system came really only to the great mass of humanity with the look provided us by Spaceship Apollo on its way to the moon. Suddenly many of the earth's people realized that our resources are finite, that our environment is fragile, and that we must conserve the first and restore and enhance the quality of the latter if we are to survive. In the words of a current television advertisement in my country, "if you cross swords with Mother Nature, you will be punished."

Barbara Ward and Rene Dubos in their recent book "Only One Earth"[1] which was formulated for the United Nations Conference on the Human Environment[2] held in Stockholm last summer made an astute observation linking the environmental concern with the energy issue. We, here at this symposium, will do well to keep this thought continuously before us as we proceed through the week. I quote:

> "There exists a single unified system from one end of the cosmos to the other; in the last analysis, everything is energy. Its larger spirals are the galaxies, its smaller eddies suns and planets, its softest movement the atom and the gene. Under all forms of matter and manifestations of life there beats the unity of energy operating according to Einstein's law."

That conference threw together in even the most literal sense people from over the face of the globe - people from highly developed countries, people from countries only beginning to take their first steps toward becoming members of the world community, and people from all stages along the road between these two extremes. We, from the more developed countries, came to realize, often to our discomfort, that the human environment involves the total interplay and interaction of the technological, biological, social, and psychological factors around us. We found that two-thirds of the world's people consume nearly eight times less energy per capita than do citizens of more technologically developed countries. Such a discovery confronted us with some issues difficult to rationalize, and even painful to contemplate. On the one hand we must give extremely serious consideration to ways in which the deprived two-thirds would have open the option of a standard of living achieved by many and desired by most of the remaining one-third of the world population without at the same time degrading even further the already over polluted human environment. This is a particularly difficult problem because it is precisely all of those factors needed to improve the living standard that are the major polluters and the world population is rapidly increasing, particularly among the deprived two-thirds. Energy, already in short supply, is at the basis of economic growth; the world's store of non-renewable resources are rapidly being depleted, and the world's food supply is becoming less adequate. Thus when we view the world situation as it is and not as we would like it to be, it makes the nature of our task of reconciling an adequate energy, the physical environment and the quality of life one of challenge and urgency. The challenge is because of the innate difficulty of the task; the urgency because of the demands of unfulfilled expectations.

We are here this week to discuss these three matters- energy, the physical environment and the quality of life - as related to nuclear energy. We must however remember at least four things:

a) our less fortunate fellow earthpeople must use fossil fuel for some time yet because nuclear energy will still be uneconomically viable to them for many years;
b) sources of energy other than nuclear will constitute a major source of supply from now to year 2000 even in the most advanced countries;
c) there is an almost crisis demand for more energy;
d) at the level of the human environment the problems with developing a friendly nuclear energy are identical in a great many respects with those of fossil fuel. Thus by suitably conceptualizing the problems we can effectively contribute to a broader understanding of the total energy issue.

I want then to discuss with you three basic matters:

1. The development of public concern for protecting environmental quality and human health and what we in the United States are doing about it;

2. The nature of the energy problem and what we are doing about that;

3. Finally - as Ward and Dubos have said "Nature has so many unstable, unpredictable, and violent facets that man needs all his probing intelligence and enormous potential for understanding to enhance and stabilize its capricious bounties" the kind of institutional framework that might best get on with the job.

PUBLIC CONCERN ABOUT THE ENVIRONMENT AND ENERGY

In measuring progress, in the United States of America, we often refer to particularly significant achievements as milestones or highlights.

I will discuss three major environmental-energy categories indicating pertinent highlights and milestones of progress: 1970—a vintage environmental year, the rise of public concern for a quality environment, and the energy-environment crisis.

1970 - Beginning of an Environmental Decade

1970 was a vintage environmental year. An impressive series of highlights make it worthy of initiating an environmental decade. It was ushered in on 1 January when President Nixon signed into law the National Environmental Policy Act of 1969 (NEPA).[3] On 22 January, President Nixon followed by posing an environmental question which could become watchwords for the 70's:[4] "Shall we surrender to our surroundings or shall we make peace with nature and begin to make reparations for the damage we have done to our air, to our land, and to our water?" Problems of the human environment had now received official recognition at the highest governmental level. Then sequentially the following environmental highlights occurred:

Lengthy hearings on Environmental Effects of Producing Electric Power[5] were conducted before the Joint Committee on Atomic Energy, Congress of the United States during January and February. April 22, Earth Day, was observed with most major magazines devoting a large percentage of space or entire issues to environmental considerations. In June the President's newly appointed Council on Environmental Quality, which was authorized by NEPA, issued its first report.[6] The International Atomic Energy Agency conference on Environmental Aspects of Nuclear Power Stations was held in August in New York City.[7] This occurred during a temperature inversion period on the East Coast and hot, humid, smoggy weather added urgency to the meeting. The fall issue of the journal of the US National Wildlife Federation carried the results of their first full year of developing a nationwide Environmental Quality (EQ) index.[8] In October the Environmental Protection Agency (EPA) was formed and given responsibility for overall protection of the US environment. The first National Biological Congress,[9] held in November, emphasized areas of environmental concern by hearing agencies outline their procedures for preparing environmental impact statements. The year which had begun on a strong note from the President of the USA ended on a strong note with Congressional Hearings[10] with many heated exchanges concerning Federal Agency compliance with NEPA. Thus 1970 marked the point where public arousal had resulted in the formulation of legal mechanisms and vehicles by which the costs of environmental impact would become a part of the decision-making process. Let us review briefly how this came about.

Rise of public concern

Just over ten years ago Rachel Carson's book, an environmental-concern milestone, Silent Spring[11] appeared. The theme was that pesticide use had decimated bird populations - thus stopped their singing - and the mental image of the book's title jarred people into closer inspection of their immediate environment.

From this inspection came some who stoked the fires of environmental concern with environmental horror stories verified by "qualified experts and scientists." A few examples will illustrate: "In a decade, urban dwellers will have to wear gas masks to survive air pollution", "Rising noise levels will cause more heart disease and hearing loss", "Major ecological systems (water, soil or air) will soon break down", and many much worse. These stories carried by every imaginable type of news media and in comic strips were told so convincingly that soon most were accepting them as truths and repeated them accordingly and with euphoric enthusiasm. Nearly every community formed a citizens protest group to "fight" for saving marshes, trees, birds, and nature in general. Folk singers and ballad writers soon joined the scene with colorful anti-pollution songs. Radical ecology brochures were handed out at major science meeting places and environmental protesters interrupted proceedings. A lengthy environmental article by a Supreme Court judge successfully competed for space in Playboy—the country's leading male magazine, and in one case, a young lady campaigning by distributing litter bags with the slogan that "Dorothy was for the birds--and bees, and wildlife and for Montana" was elected to the state legislature.

Did we have concern? Yes, we had concern! So much in fact, that industry became quite defensive. We soon "learned" via public relations offices that many companies accused of polluting had officials who were avid bird watchers, gardeners, country boys, fishermen, or sportsmen. Also, many companies "confessed" they had been conservationists or environmentalists, "long before others were environmentally aware" or advertised "long before Earth Day we ordered: 'electrostatic precipitators', 'afterburners', 'protected the ducks', or 'cared for the birds around the plant'".

The crucial issue in this public concern is whether it will last. Despite an "underwhelming" Earth Day last year, the level of public interest still seems high: citizens environmental groups are still active (though the radical youth element is much lower). Environmental issues still compete effectively for the limited space in newspapers, journals, magazines of all types, and for time on radio and TV. Ecological cartoons and comic strip sequences are still plentiful. The League of Conservation Voters claimed major victories for candidates they backed in the 1972 election (43 out of 57). Bond issues for cleaner air and water were generally approved. A number of states passed measures recognizing environmental protection as a state duty. And in true "Western" style a children's toy set, the Johnny Horizon Environmental Test Kit, complete with air and water detection equipment to "trap polluters and hand them over to the law" was successfully marketed. Environmental consultation firms and pollution clean-up groups are still increasing.

At the same time the environmental ethic has been evolving in the USA there has been a growing international concern with world environmental problems culminating last year in the United Nations Conference On The Human Environment.[2] In addition, a number of substantial books and studies were published and a number of international conservations agreements including the joint USA and USSR environmental studies venture, were consumated.[12]

Let me elaborate on only two of these. From the meeting of 100 nations at the UN Conference came approval for a number of declarations to guide an international campaign against pollution as well as for creation of a permanent UN organization to work on world environmental questions. Plans were made for a $100 million environmental action fund and for creation of a global network of monitoring stations to be known as "Earth Watch" that would provide early warning of emerging threats to the environment.

Of the various published studies demonstrating international concern it probably was the controversial "Club of Rome" report, The Limits to Growth[13] which produced the most attention in the USA. This report based on systems analyses concluded that continuation of existing trends would lead to catastrophic scarcities and severe pollution by the year 2100. However, the report suggested that by implementing a policy which carefully balanced capital investment and population size, global equilibrium can be achieved. The present indications are encouraging that the global housekeeping concept is here to stay.

Energy-Environment Crisis (a Collision Course)

Though we appear to have one major crisis after the other the energy situation in the United States of America may now be truly approaching a crisis condition. In accordance with prediction, fuel shortage plagued much of the nation last winter: Many homeowners and schools ran short of heating oil, there were occasional factory shut downs, grain shipments were stranded on barges on the Mississippi and Ohio River, airplanes and railroads had to make changes in schedules, and currently many gasoline stations have closed and others have reduced operating hours due to gasoline shortages. Coal and electricity have also been in short supply from time to time with more shortages predicted for this summer. Whatever the general picture for the affected regions, our nation has an energy crisis.

It is true that only a part of environmental degradation is associated with energy development. Yet the clash between the nation's desire for additional power and its need to save the environment has become so intense that energy development, particularly for electricity in the eyes of many, is nearly synonymous with pollution.

Powerplants do pollute air and water that has already suffered extensive damage. Not only are large black clouds often visible to people in most parts of the country but the power lines intrude upon the dwindling land, especially in regard to beauty of the countryside. It is estimated that by the year 2000 almost ten million acres of land will be used in power line rights of way.[14] In addition, several hundred powerplants that also require large amounts of land are being planned or are under construction.[14] Hence each region of the country has a "symbol of pollution" near them. In the USA another stark reminder, the unreclaimed coal strip mines and uranium mine tailings, haunt wide expanses in many states.

This duality - desire for a clean environment and desire for more power - is ironic since the same environmental concerns that have decreased the availability of energy have also greatly increased our need for it. Power is needed in large quantities to recycle sewage and solid wastes, to gasify coal and to free it from sulfur. It has also been estimated that the elimination of lead from gasoline will cause a 15 percent increase in consumption. Electricity has a dual appearance also--to many homeowners it is a clean source of heat and is so advertised, yet to generate the electricity with low efficiency of conversion from coal to electric current much additional air and water pollution is generated.

Though nuclear powerplant siting has been vigorously protested, environmentalists have delayed only 1 or 2 plants from coming on line as planned. Delays have been due to equipment procurement and delivery, to slow downs in construction, and perhaps to other causes, yet this is not true for other potential energy sources. For instance, through court injunctions environmental groups continue to hold up the development of the Alaskan pipeline. While only three of the 16,000 existing off-shore oil wells have caused major pollution damage, drilling in California's Santa Barbara Channel has been stopped. Though at least 50 more petroleum refineries are needed, opposition has been so strong that not a single new installation is now under construction. Ports have not been developed to handle the giant supertankers even though such action could not only reduce oil transportation costs but would allow fuel delivery at a rate much nearer that needed to prevent severe shortages.

Environmental problem lists have been prepared by many and are of all lengths including some extremely long. Nearly everyone agrees that one of the most important energy-related environmental problems is air pollution from fossil fuel burning. Specific substances of concern are sulfur dioxide, nitrogen oxides and oxidants, carbon monoxide, particulates and unburned hydrocarbons and trace toxic substances.

There are, however, many other aspects of energy activities that may have serious environmental consequences, such as the insults that arise from each of the energy processes—from discovery of the raw fuel to ultimate disposal of the waste including waste heat (see Table I). Because of the sheer magnitude and complexity of the problem, especially when one examines all sources and options (see Figure 1), the impacts will not be fully known until much into the future.

The solutions to the problems of this energy-environmental confrontation may well alter the life-style in most of the developed world. Costs for electricity, heating, and gasoline will increase. Increased numbers of blackouts and brownouts will occur. The confrontation between environment and energy will intensify. Internationally the crisis could force a new order of priorities in diplomacy as attempts are made to improve relations with the mideastern countries that control so much of the world's oil reserves.

TABLE I. THE INTERACTING ELEMENTS IN STUDIES OF THE ENERGY ISSUE

SCIENCES	SOURCES	PROCESSES
PHYSICAL	NATURAL THERMAL GRADIENTS	RESERVES
BIOLOGICAL	OCEAN AND STREAM CURRENTS	LOCATION
ECOLOGICAL	ATMOSPHERIC CURRENTS	CONVERSION
ENGINEERING	TIDAL	STORAGE
SOCIAL	GEOTHERMAL	DISTRIBUTION
POLITICAL	SOLAR	USES
PSYCHOLOGICAL	NUCLEAR	DEMANDS
LAW / REGULATORY	HYDRO	WASTES
ECONOMICS	FOSSIL	
MEDICAL	GREEN PLANTS	

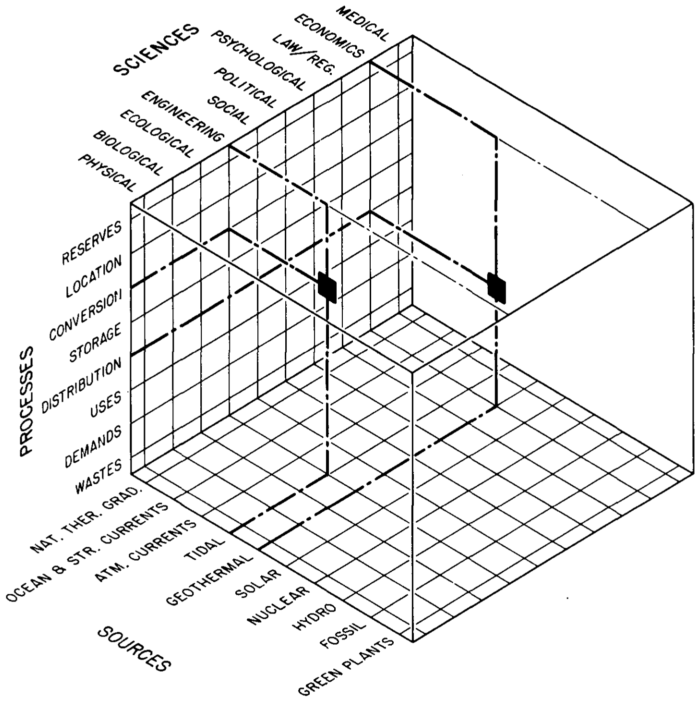


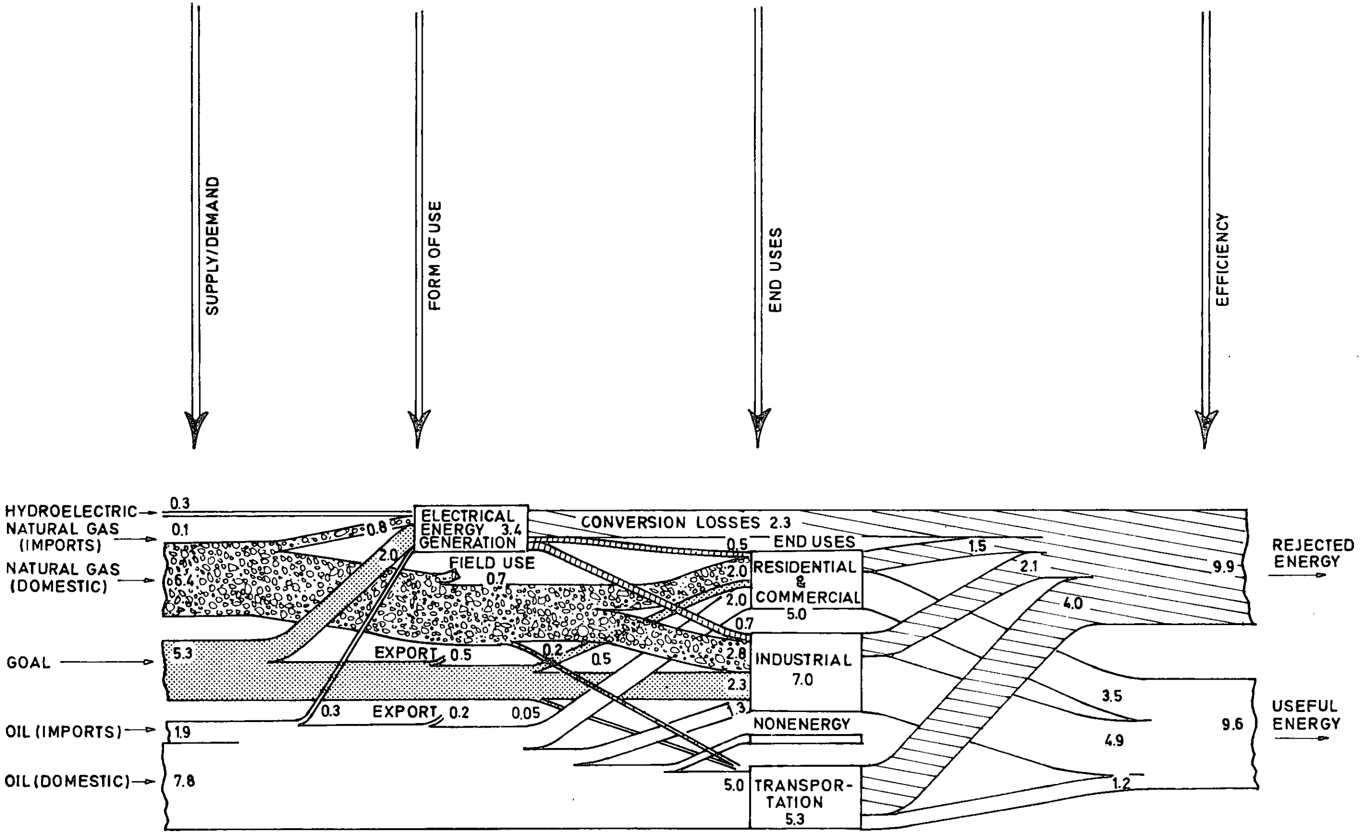
FIG.1. Complexities of the energy issue. The foremost intersecting point shows the engineering aspects of tidal energy conversion. For each source there could be as many as 80 different sources of impact.

ENERGY SUPPLY/DEMAND RELATIONSHIPS IN THE USA, PAST, PRESENT, AND FUTURE

Clearly then as stated by Senator Henry Jackson, co-author of the NEPA of 1969, the energy-environmental crunch is "the most critical problem facing the US today." How did this situation come about? What can be done?

It is relatively easy to trace many of the causes of the energy shortage particularly in the USA since we have become a prodigious energy guzzling machine. Much of the increasing demand is due to the increased per capita use as opposed to an increase due to population growth.

The present energy crisis has been brought about primarily by institutional weaknesses and not by a lack of potentially available resources or energy options. First, in the USA we do not have a comprehensive energy policy. For instance, fuel policy emanates from everywhere—the Bureau of Mines, the Atomic Energy Commission, the Environmental Protection Agency, in fact, from 64 different agencies in all. Secondly, we have had a long-standing policy that we had energy aplenty, that it was inexpensive and that we really did not need to do much to keep it that way. Oil, gas, and electric companies have



(UNITS: MILLION BBLS/DAY OIL EQUIVALENT)

FIG.2. Total energy flow pattern till 1960 [16].

until very recently advertised relentlessly to sell more and more of their product--"your best buy in modern living is electricity." Now information that the pilot light on a gas range consumes 1/3 of the fuel the appliance burns becomes hard to believe but does lend substance to the charge that we waste fully 50 percent of all energy we burn. In addition there is a lack of valid data on our resources and on demand with misdirected incentives and pricing policies on fuels and electric power.

These institutional weaknesses have been augmented by a real shortage of natural gas, by failure of the nuclear energy sector to meet its projection, somewhat due to the economics of the situation, to construction costs and delays and to a lesser degree due to the environmental standards that need to be met--for example, the Calvert Cliffs' decision[15] stopped the building of new nuclear plants until environmental needs are satisfied.

Until this point, we have dwelt essentially with generalities of the energy problems. Let us briefly explore some facts and some projections.

On 4 May of this year the US Congressional Joint Committee on Atomic Energy (JCAE) held a hearing and released a Committee Print entitled, "Certain Background Information for Consideration When Evaluating the 'National Energy Dilemma'".[16] The information is not new but is all in one place, is displayed for ease of comprehension, and is indeed most instructive.

The Print shows short-range projections of USA future energy supplies and requirements and also the consequences of possible alternative policy options for the longer range future. The document is sobering for us as I think it will be for you to. Let me summarize some of the main points.

The United States of America with about 6% of the world's population now consumes over 35% of the world's energy and minerals production. The average American uses in a few days as much energy as half of the world's people individually use in a year. The first series of slides represent US national energy flow patterns for 1960, 1970, 1980 (as projected in 1971), and 1985 (projected). The units are common for all supplies, uses, and time periods and I shall therefore simply speak in terms of energy flow units. Look at Figure 2 in some detail to learn how to read it, and because it represents a past year, 1960, for which data exist.

Note that about 80% of the USA oil supply came from domestic sources, 20% from imports. Note that about 3% of the supply was used to generate electricity, about 20% for residential and commercial end use (heating, mostly), about 13% in industrial use, but 50% in transportation, which includes autos and airplanes.

In 1960 domestic coal production was less, in energy flow units, than oil or gas. About 40% of the coal supply was used for electricity generation, but somewhat over 40% was used in industry, iron and steel production, for instance.

The gas supply was, as you might expect, nearly all domestic. About 12% was used to generate electricity, about 30% for heating, and about

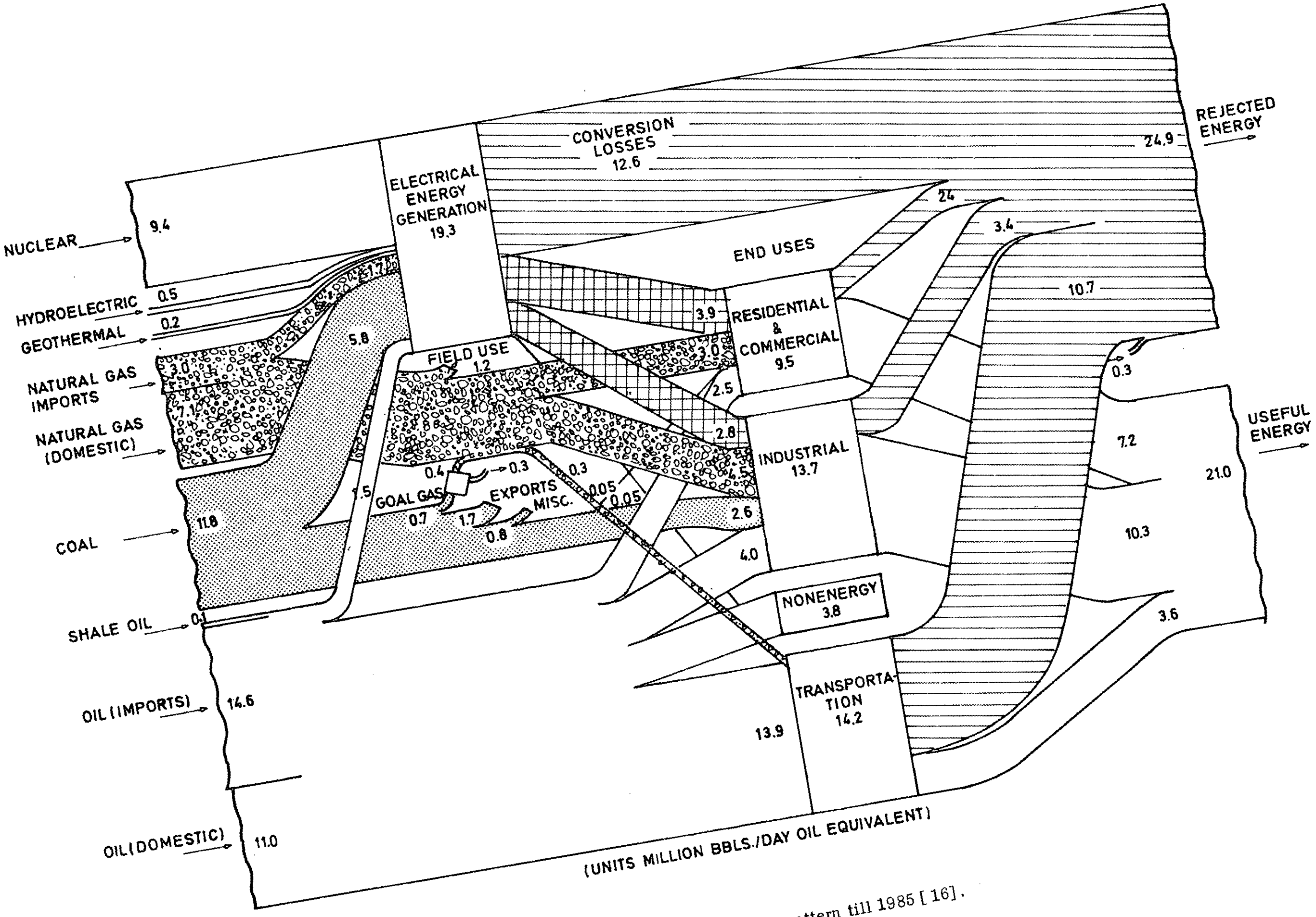


FIG.3. Total energy flow pattern till 1985 [16].

40% for industrial uses. In fact in 1960 the industrial sector of the USA economy already received more energy from gas than from coal.

Note that the hydroelectric contribution to the total, national energy supply was small, though of course the contribution to the electricity supply in some regions was not small. The USA had no significant production of energy from nuclear, geothermal or other sources in 1960.

Figure 2 shows plainly a familiar fact - that in the production of thermal electricity only about one-third of the fuel energy is recovered. Note also that the transportation sector of the economy rejected as waste energy over 75% of the heat energy supplied to it, the industrial sector rejected about 30%, as did the residential and commercial sector; overall, about 51% of the fuel energy was rejected.

For 1980 we anticipate that nearly 50% of our oil needs will be imported and that nuclear energy will be important. Coal gasification first appears. But overall conversion efficiency drops and at least some part of this drop is clearly associated with efforts to protect or improve the quality of the environment.

Figure 3 carried us out to 1985, a time far enough off to give the USA some flexibility now to change the supply or the use pattern that might occur then. But it should not be concluded that changes in policy could be made easily. Note that rejected energy is now 57% as compared to 51% in 1960. Note that imported oil has been projected to exceed that from domestic sources. Note also the growth of coal gasification, the first appearance of oil from shale, the anticipated significant dependence on imported natural gas, the continued increase in electricity generation, and increasing proportionate conversion losses up to nearly 50% rejection.

A comparison of the overall sizes of flow can be made, and comparisons for other years (not shown) can be made. Some interesting estimates emerge:

- the energy flow for 1919-1920 would be about 45% of that for 1960, that for 1940 about 53%, that for 1950 about 75%

- from 1960 to 1985 the electricity supply undergoes a five-fold increase

- from 1970 to 1985 the USA would use as much oil and gas as it had used throughout its history until 1970...but note as shown in Figure 4[17] that in general energy flows are increasing at a greater rate in much of the rest of the world than in the USA.

From the various materials in the JCAE print other kinds of time comparisons can be made such as the relative growth of useful versus rejected energy, the growth of energy demand according to a partial classification of end use, the growth of energy usage according to a classification of the physical form of the energy source and a supply/demand growth picture, for the different types of fuels and other energy sources.

Another type of analysis was made in the JCAE Print. It is possible that a US Atomic Energy Commission estimate[18] of 150,000 megawatts of nuclear power capacity by 1980 will not be made and that

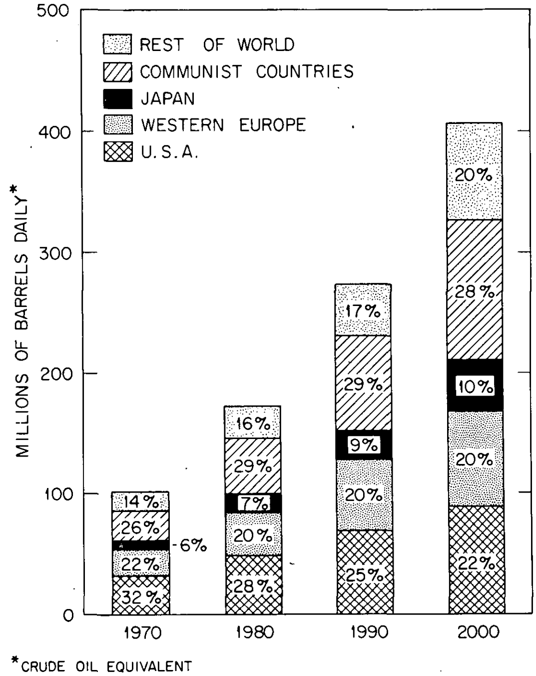


FIG.4. World energy consumption [17].

the shortage will be about 18,000 megawatts. This turns out to correspond to an additional requirement for oil of over one-half million barrels per day in 1980; at the present import price of $4/bbl, the foreign exchange amounts to about $1 billion for that year. Total dollar outlays for imported fuel, starting with about $7.5 billion in 1972, rise to $20 billion in 1980 and at least $30 billion by 1985 at present prices.

To provide for a consideration of energy policy options, the Committee Print also presents some estimates of energy supply and demand, carried out to the year 2000.

Figure 5 displays several of the more recent estimates of the USA's energy demand through the year 2000. Also shown is a USA population estimate. Figure 6 shows the results of one illustrative policy-option study/exercise, which I shall take a few moments to describe:

- Demand is assumed to be depressed about 25% below the demand that would occur if present growth rates in energy use were to continue.

- A certain magnitude of imports or shortages is assumed to be acceptable; in this example, the assumed magnitude exceeds that which some economists have considered acceptable for the financial security and stability of the USA.

- Production of oil from the 48 contiguous states is estimated based upon the assumption of a major, but not necessarily a maximum

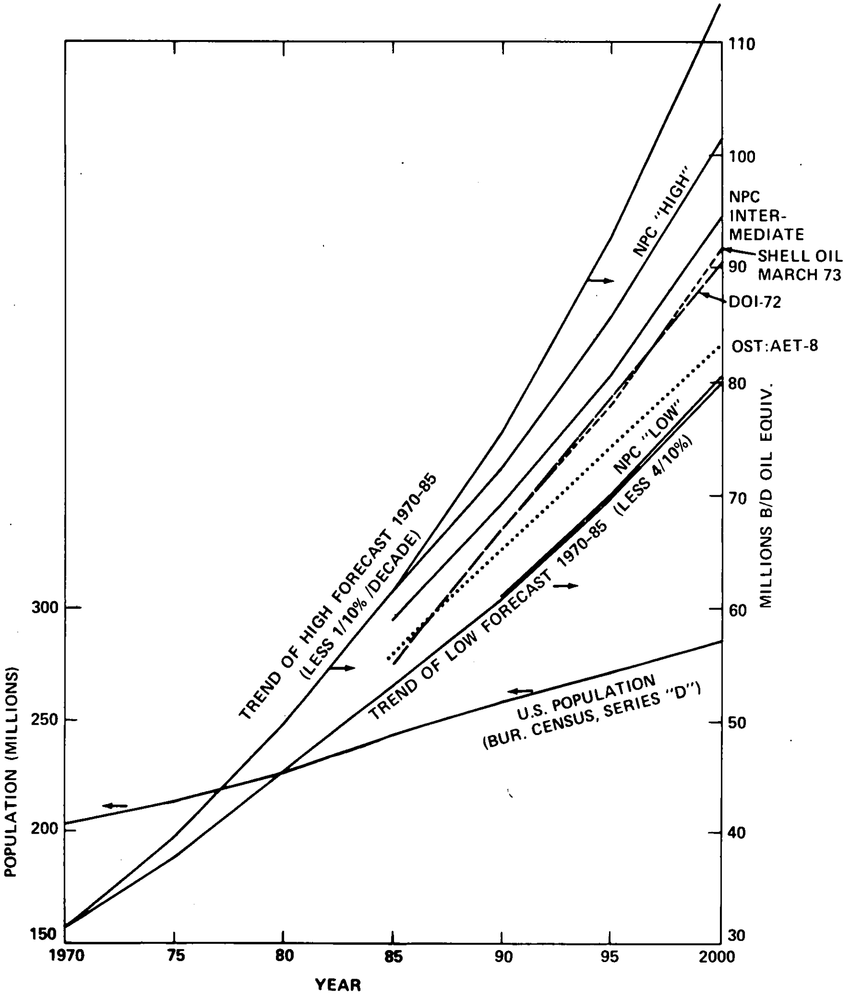


FIG. 5. Forecast of energy demand to 2000 [16].

effort to increase domestic oil production: at least a 50% increase in the price of domestic crude, immediate development of known off-shore oil deposits (e.g. Santa Barbara channel)...in short this is a optimistic forecast with possible environmental impacts.

- Coal production is assumed to triple, accomplished perhaps by a tripling of strip mining plus a 50% increase in underground mining above present rates...there is clearly an environmental impact here and the forecast is nonetheless possibly somewhat optimistic.

- Domestic gas is estimated quite uncertainly, but the assumption is that natural gas prices would be deregulated to try to sustain or increase production somewhat; if any major new sources of natural gas are discovered, these sources can be used soon afterwards.

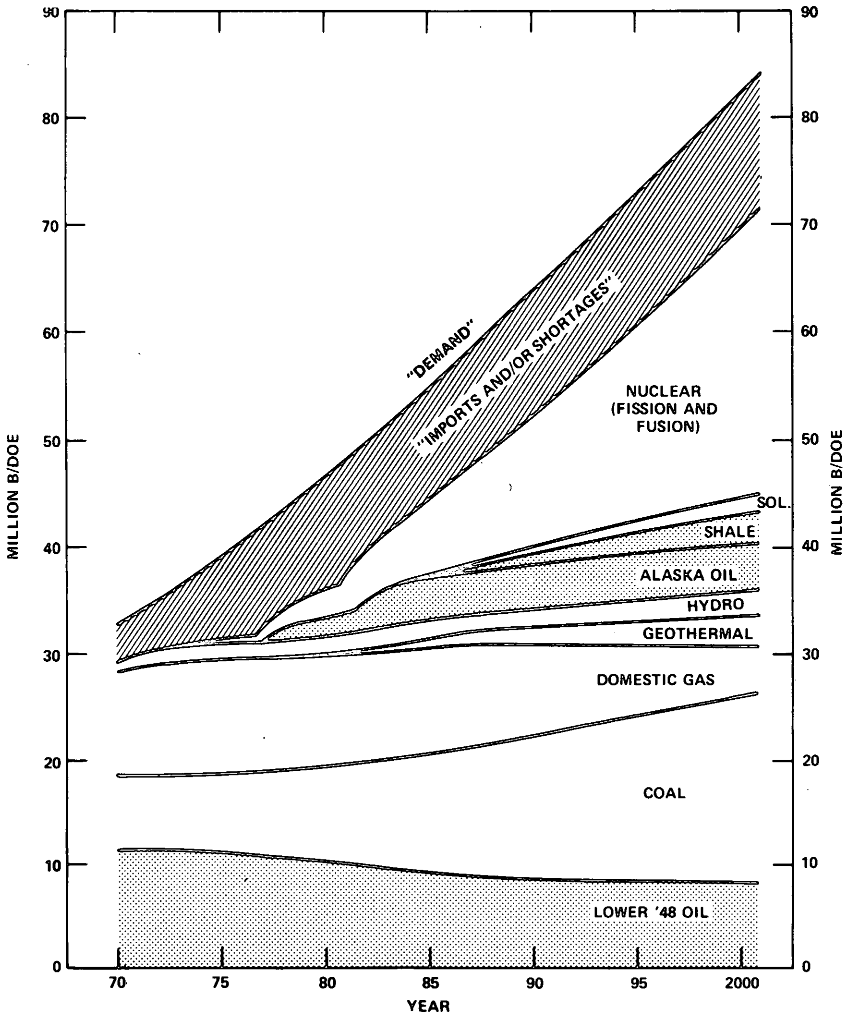


FIG. 6. 'Supply/demand' (JCAE "Option Exercise 7-A" 3-73): B/D oil equivalent plotted against years [16].

- Geothermal power of 100,000 megawatts by the year 2000 is assumed in this example, the electric equivalent of over 100 Hoover dams, with possible environmental impacts.

- Hydroelectric is projected for a 50% increase in present capacity, implying use of such sites as the Grand Canyon, with obvious environmental impacts.

- Alaska oil...the Valdez, Alaska pipeline is assumed to be in operation in late 1977 and another line through Canada by 1984, with obvious environmental impacts.

- Oil shale production is regarded as highly dependent on technology, availability of water, and legal factors.

- Solar technology is assumed to permit us to take over some of the heating and cooling loads in the Southwestern USA; in this example, the amount assumed would take care of more than all of the houses that would exist in New Mexico, Arizona, and Nevada by the year 2000, but technology is uncertain.

- Nuclear is shown to buildup to over 1 million megawatts by the year 2000, about in line with various estimates that have been made. A massive industrial effort is implied.

It seems to me that the material on energy supply/demand that I have just gone through portrays, highlights, dramatizes, and makes very specific the way in which policy on (i) energy development, (ii) quality of the environment, (iii) research and development and technology, (iv) capital investment, (v) land use, (vi) the economy and related financial matters is a tightly interwoven web.

AN APPROACH TO THE ENERGY/ENVIRONMENT DILEMMA: THE USA NATIONAL (NUCLEAR ENERGY) LABORATORIES

From the previous discussion the complexity of the problems associated with trying to develop a friendly energy and the urgency of resolving them now should be obvious. The focus has been largely on the biomedical-environmental problems concerned with the discovery, processing, development and use of nuclear and fossil fuel for central station power generation and disposal of the wastes. Since, however, the use of fossil fuel for residential, commercial, industrial, and transportation activities present the same general types of problems, solutions to problems in any one area have been generally applicable across the spectrum. Additionally the techniques and approaches useful for studying the energy-environment issue with fossil and nuclear fuels are proving equally applicable for evaluating the impacts of solar, hydroelectric, and geothermal sources, as well as for determining the biomedical-environmental impacts of other of man's activities--industrial and agricultural chemicals, drugs, and other physical insults to the environment.

Clearly the expertise needed to solve these problems where each partial problem is woven in a finite and often intricate way into the web of life must be quite varied with specialists drawn from almost every area of science and engineering. The issues are so complex, however, that there is an overriding requirement for the formation of teams of specialists who are willing to work with specialists from other fields to solve a common problem. Failure to view the problem as it is - whole and integrated - and not as the disciplinarians would like it to be - in discrete parts - can lead to disaster from the unexpected. There must, however, be more than the initial overall view, for unless the follow-on research continues to reflect integration of the various disciplinary views it in no sense can be considered interdisciplinary and will in all likelihood contribute to only a partial solution of the problem.

Teams alone, however, are not the total answer for there must be a fairly large component in the total effort wherein an individual scientist working alone but in constant contact with the team can explore many basic aspects of parts of the problem.

To build and mobilize teams around problems that change only slowly is difficult. To keep them functional on constantly changing problems is a major challenge for their leaders.

There are three major stumbling blocks to the development and maintenance of these interdisciplinary teams - individual, institutional, and funding agency concerns.

The individual concerns are a consequence of the narrow disciplinary training of our scientists in the United States of America which equips them for attacking problems of narrow scope. Additionally it instills in them the view that one must understand each of the minute processes in detail before any attempt can be made to comprehend even in barest outline the whole problem. The disciplines each have their own dogmas, their own reward systems, their own yardsticks for performance measurement, and their own methods of punishing the unfaithful who strays too far toward becoming a member of an interdisciplinary team. The resulting problem is that the major issues facing mankind - energy, the environment, health, housing, welfare, transportation, education and other problems - are so broad and comprehensive that no single discipline can encompass its intricacies let alone solve the problems associated with it. What is clearly necessary are team members who maintain their professional links so as to stay proficient in their specialities but who can be motivated by various incentives to change their conventional behavior patterns so as to become effective team members.

Institutional problems are just as difficult to overcome and in general the older and less flexible the institution the more difficult the problem becomes. Universities by the very nature of their primary responsibility are disciplinary oriented and by departments which usually have high walls in between. These facts coupled with the involvement of graduate students as a major portion of the research effort, their short stay and the resulting lack of significant institutional memory all work in the direction of discouraging deep involvement of universities in problem focused research. Many universities however, in response to the pressure of the times and the shortage of funds for unstructured research have recently created special projects for more mission oriented research. Even here however with rare exception the projects are narrow in scope, mainly short in duration and often do not involve principal professional academic staff.

The more classical research institute can create teams fairly diverse in nature and of considerable breadth but generally unless they were created in the first place with a broad mission, and unless they have a very large budget they will lack the depth and breadth of professional strength to launch comprehensive programs in the energy-environment area. They too therefore are in general not in a position to consider the total problem.

This then leaves us with the major federal government laboratories in the individual countries. Again the activities of most of these laboratories are rather narrow in scope. In the United States of America the Environmental Protection Agency has laboratories both narrow and broad in scope in the environmental area but they have little involvement in the many facets of the energy problem. The labs of the National Institutes of Health are primarily concerned with disease and only occasionally get involved in energy effluents research. The NASA labs are heavy in the engineering areas with little involvement in the more important energy areas except in special instances. The labs of the Department of Defense work largely on defense problems. Those of Interior, and other agencies in general are rather small and are narrowly focused on small parts of the energy problem.

A view of the competence of the above energy laboratories was recently stated by Monte Canfield, Jr., Deputy Director, Energy Policy Project, The Ford Foundation, Washington, D.C., in his recent 29 April 1973 keynote address to the American Medical Association's Congress of Environmental Health,[19] concerning a national commitment to an urgent effort for abundant clean energy: "Outside the Atomic Energy Commission, which deals with a limited area, there exists nowhere in government today that critical mass of technical talent we need to 'make it happen'". Furthermore, Anthony Roisman, who represents the Environmental Defense Fund, one of the intervenors in the Indian Point-2 Operating License Hearing, recently said: "The AEC has taken on responsibility (under the National Environmental Policy Act) and has acted responsibly. There is no doubt that the AEC is now doing the best job of any Federal agency in implementing NEPA."[20] It is of these laboratories then that I will talk for the next few minutes. Since until last November I was the Associate Laboratory Director for Biomedical and Environmental Sciences of the Oak Ridge National Laboratory (ORNL), I will take it for a more detailed discussion of the kinds of problems that can develop as well as some of the features which encourage the development of interdisciplinary teams so sorely needed to get on with resolving many of the energy issues...

Major programs of broad scientific scope and depth are conducted in the biomedical, environmental and physical sciences and in a broad spectrum of engineering sciences (see Figure 7). The engineering programs are heavily oriented toward reactor development and radioactive waste management. The biomedical and environmental sciences span the spectrum from the atom to gross regional environmental systems programs including at this stage of development demography, economics, regional planning and related activities. These programs have interests both in radiation areas and on the effects of other physical insults to the biosphere. The chemistry programs have a strong emphasis in the areas of inorganic chemistry and materials sciences. But in addition, they provide strong support to the engineering and bioenvironmental areas. Physics has a heavy commitment to nuclear fusion and to the use of accelerators and reactors for basic nuclear physics studies. Much of the work is done within one of the several disciplinary divisions of the laboratory. For tasks that are fairly complex, however, ORNL has historically mobilized teams of people from a few closely related disciplines under a project leader who gives both scientific guidance and exerts administrative control. Recently, however, as the problems needing attack have demanded input from many different disciplinary scientists a need for change has become apparent. Let me describe only a single example to illustrate the kinds of problems that can arise.

Two years ago the ORNL began, under National Science Foundation sponsorship,[21] a program on toxic materials which drew people from some ten laboratory groups—ecologists, chemists, biologists, analytical chemists, metallurgists, mathematicians, economists, regional planners, information specialists, reactor engineers, sociologists, demographers, and others. In order for the people to stay professionally expert, we felt they should remain physically in their disciplinary divisions (listed in Table II) and yet be members of the integrated team. While we found

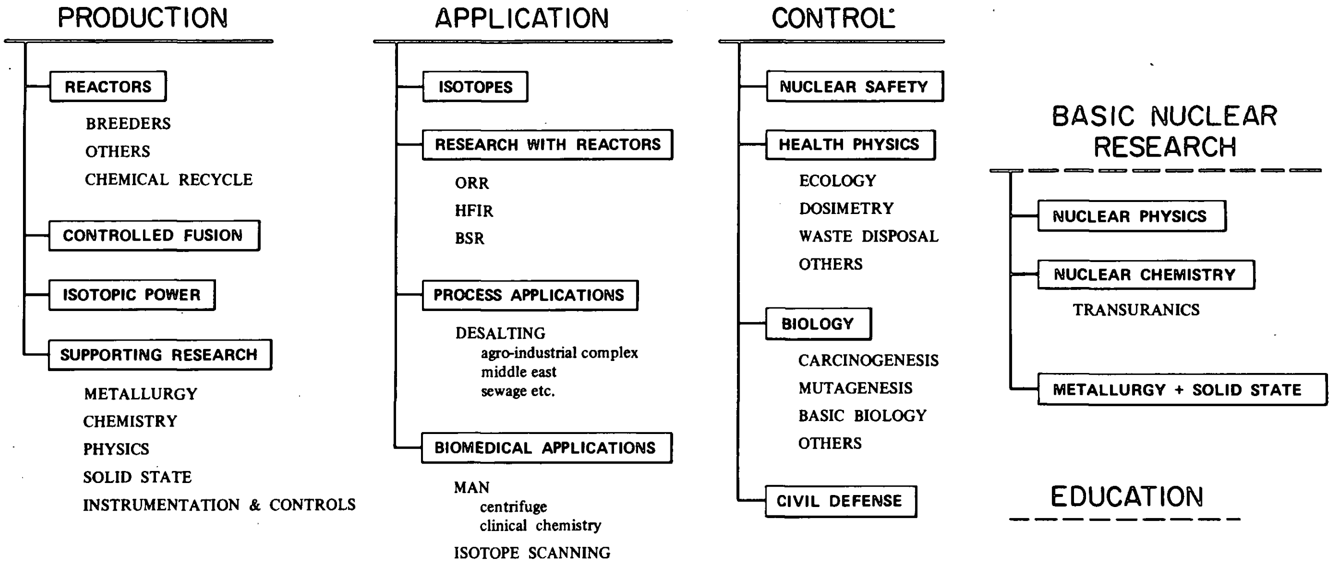


FIG. 7. ORNL — a nuclear energy laboratory.

TABLE II. DISCIPLINARY SCIENCE AND ENGINEERING DIVISIONS – ORNL

Analytical Chemistry
Chemistry
Chemical Technology
Reactor Chemistry
Physics
Solid State
Neutron Physics
Thermonuclear
Mathematics
Reactor
Metals and Ceramics
Instrumentation and Controls
General Engineering
Biology
Health Physics
Ecological Sciences
Molecular Anatomy
Civil Defense
Environmental Program-NSF
Environmental Information Systems Office

the people willing to be on the team, we found the division directors were resistant because they felt they were losing "control" of parts of their program—their territorial imperative was being violated.

We moved to solve the matter, as illustrated in Figure 8, by developing a series of task leaders, two subprogram coordinators and an overall program coordinator. These coordinators work closely with the division directors to insure excellence of the work, with the task leader to insure relevance to the broad objectives, and with the EP program leader whose responsibility it is to see that the technology is transferred as rapidly and effectively as possible.

Just in case there were perturbations in the system, the authority to insure compliance with the program objectives was placed in an ORNL associate director's office to insure an effective program irrespective of the home division. At times there is a bit of what I call "creative tension" between the program coordinators and the division directors about the use of people and facilities. This concern for territorial imperatives has insured a continued attention to the scientific problem and program by all concerned--each must stay alert or risk some loss of prerogative.

A problem still in the resolution stage concerns whether we can successfully build into the structure of a major R&D organization the needed social scientists and the professions of law planning and policy research because these latter types pose questions of a kind and in a way which scientists and technologists generally either resent or are unable to answer--resent because a "non-scientist," at least in their view, has the audacity to challenge the sacred dogma, and are unable to answer because the language is different.

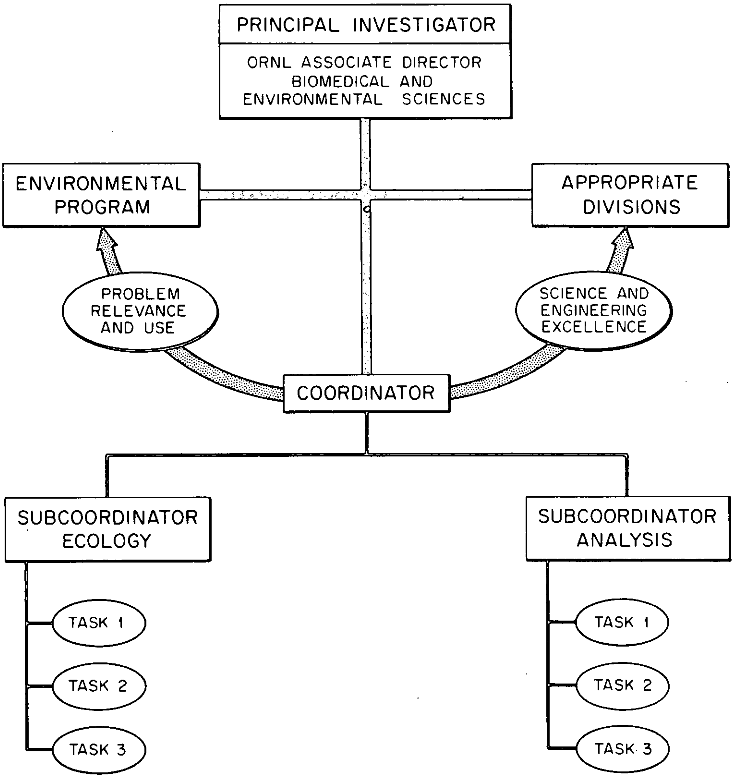


FIG. 8. Program team for ecology and analysis of trace contaminants.

The effort is beginning to work but I feel there will be much travail before any problem focused institution can really adjust to the mix. Yet, the mix must come either within the institutional framework or in the public forum.

Earlier I mentioned a third problem in trying to build an interdisciplinary team of any size and that is securing funding. The fact that there are very few "truly interdisciplinary" scientists makes the lot particularly hard. Because the number of such scientists is so small specialists in particular subject matter areas or disciplines are asked to pass judgment on interdisciplinary programs. But because of their disciplinary dogma concerns, they seldom take the trouble, or what is even worse, recognize the need to view the team or the problem as a whole. Another problem in the system is that often the Washington-based money tender feels that he must see to the "integration" of diverse pieces. I am one of a very few Washington bureaucrats who does not believe that is the way the system works best and, in fact, I think that if the science is not integrated at the bench, it will not really ever get integrated at all. Yet, it is the integration by the interdisciplinary team that is needed to solve society's problems. The R&D administrator then in search of funds for his problem focused team gets caught in the "commons" where the tyranny of small decisions by well-meaning bureaucrats erodes the very fabric of the interdisciplinary team. The total product of the team is seldom viewed or even looked for. This situation is much like that of the medical profession today wherein there is a specialist for everything. No one seems to take the health of the total patient into account. The garage situation where there are generator specialists, tire specialists, this and that specialists, with no one really checking into the general health of the car is equivalent. These matters we understand because we are the victims. Yet, as scientists, we are prone to the same fault of judging performance on only a part of the problem.

I feel that unless a way is devised to view and support these teams as a whole and to measure their total team output, we will never be able to get on with a solution of the major problems in the energy environment area. It goes without saying, however, that the team must be productive and earn the right to the broad gauge look.

Let me now conclude by saying that:

1. We have seen how public concern over degrading environmental quality arose and about some of the underlying causes.

2. We have discussed how while providing the energy we need we pollute the environment quite extensively. Yet the answer does not lie so much in slowing energy production and use as in making its use more efficient and by better control of its wastes. It then is left to those of us in the more developed countries to see that the lessons we learn are made available as economically as possible to those countries yet needing the energy to improve their standard of living.

3. We have explored briefly the complexities of the energy/environmental conflicts and hopefully recognize that to be effective in resolving them we must conceptually look at the whole problem as it is. Clearly large broad technological-based laboratories are needed to do this most effectively (Figure 9).

This alone, however, is not enough particularly for nuclear energy.

The ultimate success or failure of nuclear energy in my view will depend more upon the willingness of the public to accept it than upon its inherent safety, but I reach this conclusion only because I have spent the past 15 years being concerned with the health and safety aspects. There is the absolute necessity for candor and realism in discussions concerning nuclear energy especially in the area of being acutely aware of and sensitive to the varied aspects of public concern. This awareness alone is not enough. We must, in fact, be responsive to the needs and communicate with the public—communicate in a way that they understand, comprehend and believe. As scientists and perhaps also as research administrators many of us have come to place ourselves in the attitude of "we are scientists expert in the area and we know and understand! You who are not knowledgeable should believe us if we tell you you are safe. We are the protectors of the faith. We speak only truth!" That attitude is just plain nonsense! It is not an acceptable answer and unless scientists and technologists, in general, change from this view, we will be in difficulty.

Unfortunately, as I mentioned much earlier in my talk, most agencies are in trouble because of a crisis of confidence—scientists are in that

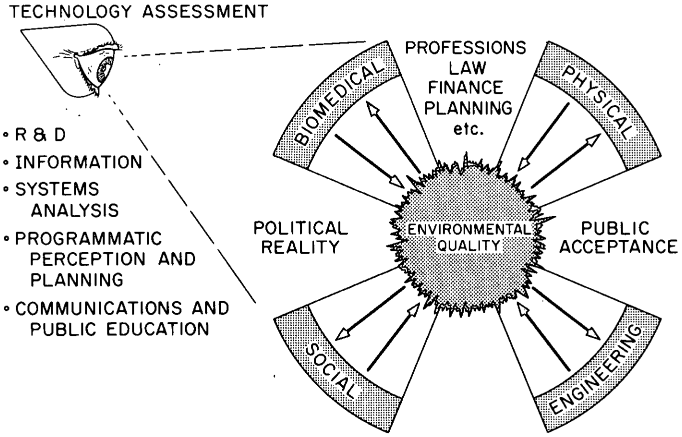


FIG.9. Components of a comprehensive multidisciplinary institution.

position also. To try to reverse this "I am more holy than thou" attitude, I remind my staff almost every day—talk to a new neighbor in his language about nuclear energy so that he can see it in perspective. It is a matter of history that in America we demand to be informed so that we can reach our own conclusions. Thus, if the American public is to accept nuclear energy, I feel it is incumbent upon those of us involved in getting answers to be candid, to be factual, and to be helpful in informing the public in language and terms they can understand. To do less is to court failure.

I suspect, whether or not you choose to believe it, that you soon will be facing these issues too. The virus of unrest is spreading and I do not believe national boundaries will prove to be able to stop it. Only the intensity in the separate countries will be different.

It behooves us then as we proceed through this week to keep the public interface constantly before us.

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Invited Paper

ASSESSMENT OF POPULATION EXPOSURES

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Abstract

ASSESSMENT OF POPULATION EXPOSURES.

The conventional method of relating assessments of radiation doses in the environment to dose limits applicable to the most highly exposed individuals (i.e. the 'critical group') will not be sufficient when the number of nuclear power reactors is rapidly increasing. Additional methods of assessment, designed to facilitate the control of the future situation, include concepts such as 'collective dose', 'population dose' and 'dose commitment' per unit practice (e.g. man-rems per MW · year). These concepts are discussed in relation to risk-benefit and cost-benefit evaluations in source-related assessments.

INTRODUCTION

Until recently the release of radioactive material into the environment from a nuclear reactor has been assessed mainly in relation to standards that apply to the most highly exposed individuals. This has been a quite satisfactory approach as long as the number of reactors has been small and the overlap of doses has been negligible. In future, however, this may not be sufficient. If we envisage a future, yet distant, where radiation sources are numerous, we would still wish to see it safe and well within the dose limits we have already found to be prudent. No authority can easily control a situation where individual dose contributions are not readily identifiable but the total is found to be too high. We have already had this experience with non-radioactive pollutants such as DDT, mercury and many other harmful substances. We would not wish to see it repeated with radionuclides and, indeed, there is no reason why we should with the good knowledge we have already acquired.

There is only one way in which the authorities can continuously control the development and shape the future situation to meet desired standards. The only way is by controlling each individual source so that the total future radiation level will be acceptable. If doses are expected to overlap, the dose limits must apply to the total and each source will have to be subject to more restrictive limitations than necessary had the particular source been the only one. Such extra caution is not necessary for radiation sources that are not expected to last until this future situation has developed, but it will be necessary for those sources, e.g. nuclear power stations, that may still be operating in the next century.

This cautiousness, aimed at the future, may often seem overscrupulous at the beginning when the sources are still few. However, gradually we have to adopt it and it may be wise to look for a policy and a protection system that is adapted to the future requirements.

In this paper I shall discuss assessments of population exposures not only in relation to individual dose limits in the simple situation where there is one single source of release of radioactive material and one easily identifiable critical group to which the dose limits apply, but also in relation to other requirements which become growingly important because of our concern of the future.

HISTORICAL DEVELOPMENT

Only few years ago it was still the practice in many nuclear installations to limit the release of radioactive material to the environment by limiting the concentrations of the various nuclides in air and water effluents, the limit usually being a fraction of the MPC value recommended by ICRP for application in radiological work. Obviously this limit did not prevent substantial amounts of radioactive material reaching the environment where the effluent rates were high, nor did it guarantee that ecological concentration processes might not cause high concentrations in certain food-chains.

It was therefore an obvious next step to assess the individual exposures by environmental monitoring to make certain that no individual would receive radiation doses in excess of the dose limits recommended by ICRP for individual members of the public. In its Publication 7 (1965) ICRP recommended a procedure for this assessment, introducing the concepts of 'critical nuclides', 'critical pathways' and 'critical groups'.

The common practice has been to refer to the ICRP dose limit of 0.5 rem per year for whole-body exposure of individual members of the public and to let the average dose to a representative sample of the critical group represent the highest dose to an individual. The maximum permissible release of radioactive material could then be calculated as the release that would make the annual dose to the critical group equal to the ICRP dose limit. This release limit has been referred to as the 'capacity' of the environment to accept radioactive material. The actual releases have usually been much below the limit and this 'margin of safety' has been considered reassuring.

With this policy, however, the 'margin of safety' could be misleading since an increased number of installations might well result in many sources exposing one and the same critical group. In fact, the true critical group in a large region may not even be any one of the groups that were found critical in relation to a single installation, but a group that receives some exposure from many of the installations.

Another problem has been caused by the understandable tendency to reduce the local exposures by making the releases through high stacks or long pipe-lines, or by retention of the most short-lived radionuclides. If these technical arrangements are not actually used to reduce the local exposure but instead to keep it at the same level, thus permitting more of the long-lived material to be released, the ratio between the population average dose and the local individual dose will increase and the average dose will become relatively more important.

The average dose in a large region or in a whole country may be of interest for at least three reasons. First, it must not reach such levels that the expected detriment becomes a significant burden to the society. Since each individual is protected by the requirement on an individual dose limit, however, it is not likely that any dose distribution subject to that limitation could possibly cause a significant social burden.

Secondly, the average dose to the whole population must not be so high that the genetic risk to the population becomes unacceptable. There is no indication that there is any threshold level of dose for a sudden change from acceptable to unacceptable genetic harm in a population, but the basic ICRP recommendations of 1965 (ICRP Publication 9) include a limit for the 'genetic dose' and it is usually the ambition of the competent authorities to limit the average dose in the whole population at a level much lower than this genetic dose limit.

The third reason is that the product of the number of exposed individuals and their average dose is a measure of the total detriment from the radiation exposure. An assessment of the total detriment is necessary for risk-benefit evaluations.

These considerations have made it clear that not only the highest individual doses but also the average doses in the population must be assessed.

During the last few years, gradually more emphasis has been put on the recommendation given in paragraph 52 of the basic ICRP recommendations (Publication 9, 1965):

> " ... the Commission recommends that any unnecessary exposures be avoided, and that all doses be kept as low as is readily achievable, economic and social considerations being taken into account"

The avoidance of unnecessary exposures implies risk-benefit evaluations. The expression "readily achievable" implies cost-benefit evaluation of further dose reductions.

Any risk-benefit evaluation must relate to the operation or practice that causes the radiation exposure. For that purpose it is necessary to assess the sum of all individual exposures because only that sum will represent the sum of all individual risks and hence be a measure of the total detriment. For the same purpose it is also necessary to add up not only the present but also the expected future exposures due to the given operation.

The summation of individual doses leads up to the concept of collective dose and the consideration of future doses leads up to the concept of dose commitment. Both these concepts will be defined and discussed in the following sections.

PRESENT OBJECTIVES OF DOSE ASSESSMENTS

The primary objective of the assessment of radiation exposures from radioactive material that has been released into the environment is to evaluate the radiation doses in relation to the relevant national or local requirements on radiation safety.

The conventional method has been to check that neither individual nor average doses in a given population exceed certain stipulated maximum values. The population may be the critical group, a local community or the population of the whole country. Assessments that are related to a specified group of people may be referred to as population-related assessments and their main objectives are to verify that:

(a) No individual in the given population is subject to a risk which is not acceptable and that

(b) The total deteriment within the population does not exceed acceptable levels.

The disadvantage with the population-related assessments is that they are only suited to check the acceptability of the current exposure situation and do not easily lend themselves to either risk-benefit evaluations or to predictions of future situations. For such purposes a source-related assessment is needed, its main objectives being to check that:

(c) The total detriment from the given source is not unjustifiable in relation to the expected benefit, and that

(d) The future situation will continue to be acceptable, even if the number of sources increases.

To obtain the necessary background for risk-benefit evaluations the total detriment from the source must be estimated. On the basis of a linear, non-threshold dose-effect relationship, the total detriment from a source is proportional to total population dose (the collective dose to the world population) from that source. To make the estimate useful also in the assessment of future situation, it may be related to a unit of practice, e.g. one year of operation or one MW · year of electric energy.

POPULATION-RELATED ASSESSMENTS

The most common population-related assessment is the assessment of the dose to the most highly exposed individual in the given population. The identification of a critical group in the ICRP sense is straightforward only if there is only one dominating source of exposure. In that case the population-related assessment of the highest dose becomes identical to the source-related assessment of the critical group exposure.

If there are many sources and, particularly, if the number of sources is expected to increase with time, it becomes impossible to draw any conclusions on the acceptability of the situation from dose assessments based on environmental measurements alone. One reason is that it will be increasingly difficult to identify the true critical group, another reason is that it will also be difficult to extrapolate the current observations in time.

The detriment to the given population may be assessed by multiplying its collective dose ($M = P\bar{D}$) by a risk coefficient (μ) derived on the assumption that there is a linear relationship between individual risk (R_i), and individual dose (D_i) and thus also between detriment (N) and collective dose:

$$\Delta R_i = \mu \Delta D_i$$

$$\Delta N = \mu \Delta M$$

the collective dose of the population being

$$M = \sum_{i=1}^{P} \sum_{k=1}^{S} D_{ik} = P\bar{D}$$

in the case of S sources (k) exposing P individuals (i). The collective dose is expressed in man-rads or, when used in relation to dose limits, in man-rems, i.e. derived from individual dose equivalents.

It is reasonable to assume that the average dose distribution from the k^{th} source, i.e. M_k/P, will be much smaller than the maximum individual dose $D_{i\max.k}$ from the same source. The overall average dose in the population, i.e. M/P, however, will be approximately S times higher than the average contribution from each source, if all sources are approximately equal:

$$M/P \approxeq S\,M_k/P$$

Even if the average dose from each source (M_k/P) is very much smaller than the corresponding maximum dose ($D_{i\max.k}$), it is not necessarily true that M/P is small in comparison with the highest individual dose ($D_{i\max}$) from the combined sources if the number of sources is high.

SOURCE-RELATED ASSESSMENTS

The total detriment from any given source may be estimated as

$$N_k = \mu\ M_{\infty k}\ .$$

where μ is the risk coefficient and M_∞ is the collective dose to the world population, sometimes referred to as the total <u>population dose.</u>

M_∞ can be assessed as the sum of the products $P_j\,D_j$ for a number (G) of population groups (j), each characterized by a constant value of the individual doses ($D_{ij} = D_j$) to the P_j individuals in the group. If the total population P can be divided into G such groups of equal size, the calculation becomes simpler, the extreme case being the summation over each individual (i), in which case G = P:

$$M_\infty = P\bar{D} = \sum_{j=1}^{G} P_j\,D_j \rightarrow P/G\sum_{j=1}^{G} D_j \rightarrow \sum_{i=1}^{P} D_i$$

If $P_x = dP/dx$ is the population density function versus any measurable quantity x related to the radiation dose (e.g. distance from the source), the general expression for the total population dose will be

$$M_\infty = P\bar{D} = \int_{x=0}^{\infty} P_x\,D(x)\,dx$$

a simple case being

$$M_\infty = \int_{D=0}^{\infty} P_D\,D\ dD$$

In principle the integration must be carried out in full, i.e. the individual doses must be added up even if they are assessed to be very low, since even very small doses to a large number of individuals may still contribute significantly to the total population dose. In practice, however, it is often acceptable to stop the integration when it is certain that the remaining contribution to the integral will not be needed for an estimate of the total with an accuracy within a factor of 2 or 3.

The collective dose to a local population or even to the whole population of a country must not be misused for the total population dose, because it is only the latter that, multiplied by the risk coefficient, gives a relevant measure of the total detriment from the given source.

Conceptually, a detriment is equally regrettable wherever it occurs and the actual geographical distribution of the components of the population dose may therefore be considered to be of secondary importance. This would not be true if a large population dose were limited to a small population, in which case the corresponding detriment might not be as acceptable as if it had been distributed over a larger population. However, it is extremely unlikely that any detriment from a single source would cause any significant burden to the population as long as the individual dose limits are not exceeded.

If the risk coefficient is μ rad^{-1} and the total population dose from a given practice is assessed to be m_∞ man-rads per unit practice, then the detriment would be expected to be μ m_∞ cases of death per unit practice (if the risk coefficient is related to cases of death and not to some other measure of detriment).

It is likely that nuclear power will be produced at a population dose (in addition to occupational exposures) of less than 1 man-rad per MW · year of installed electric power. This is the population dose from the full fuel cycle, with perhaps 0.3 man-rads per MW · year expected from reprocessing. This would mean, for example, that a 1000-MW power station might be expected to cause a detriment of up to one death in every ten years if the risk coefficient is assumed to be $\mu = 10^{-4}$ rad^{-1}. It should be noticed that the occupational collective dose in nuclear power stations has been found to range over two orders of magnitude, from 0.1 to 10 man-rads per MW · year.

When the population dose is assessed in relation to the electric power production it is easy to make assessments of future situations, once the plans for the future power production are known. If the total nuclear electric power production in a country with a population of P individuals is expected to be w MW (i.e. w MW · years per year if the MW · years are based upon the installed power) at a future time, and the electric power is expected to be produced at a population dose of m man-rads per MW · year, the total annual population dose at that future time would be expected to be $M_\infty = m_\infty w$ man-rads. The average annual individual dose in that population would be $\bar{D} = m_\infty w/P$, but only if the population dose were limited to the P individuals within the given population.

In practice, part of the population dose would fall in other populations and contributions from nuclear power stations in these other populations would add to the population dose in the first population. Only if (1) the population power density w/P is approximately the same in all populations; (2) the geographical population density is the same wherever the radioactive material may move; and (3) the activity transport follows a similar pattern irrespective of the location of the source, will the added contribution just compensate the losses.

These conditions are only approximately valid in the 30°-60° northern latitudes where most of the world's nuclear power stations are being built. They may still be assumed to apply for the global contamination with tritium and ^{85}Kr, but the third assumption is less valid for the relatively short-lived ^{133}Xe and doubtful for the even more short-lived nuclides, e.g.

those of elements such as iodine, krypton and xenon, which dominate the airborne pollution. It is also unlikely to apply in the case of contamination of rivers and estuaries, for which it is not easy to make generalized assumptions on the activity movement.

Fortunately, however, the population dose is most likely to be limited within the primary population for just those nuclides for which the assumption on global equilibrium is dubious. In the first approximation, therefore, it is not unlikely that the population dose may be approximated by the national community dose in each country where nuclear power is produced. This approximation must obviously be used with great caution because there may conceivably be countries with little power production that receive a significant community dose from their neighbours, and countries with a large power production but a substantial leakage of radioactive material to other countries. To the extent that the approximation is valid, however, the per caput dose in the primary country may be assessed at $\bar{D} = m_\infty w/P$.

The future total average annual individual dose from nuclear power in the 30°-60° northern latitudes may therefore be estimated at

$$\bar{D} = m_\infty \overline{w/P}$$

where $\overline{w/P}$ is the average installed electric power in MW per caput. For example, $m_\infty = 1$ man-rad per MW · year and a per caput power consumption of 0.001 MW in year 2000 would yield an annual average dose of

$$\bar{D}_{2000} = 0.001 \text{ rad}$$

For comparison, a high estimate (Sweden) with $\overline{w/P} = 0.005$ MW per caput in year 2000 would give $\bar{D}_{2000} = 0.005$ rad.

It should be recognized that there is a mathematical expectation of population dose also from accidental releases of radioactive material. Unless the expectation of population dose from accidents is an order of magnitude less than the population dose from regular operations, the specification of the latter alone as reflecting the total detriment from the source will be misleading.

"AS LOW AS READILY ACHIEVABLE"

Once the total detriment from a source has been estimated by a source-related assessment of the total population dose, it is possible to attempt a risk-benefit evaluation. This is a political evaluation, particularly since the risk and the benefit are not always shared by the same group of individuals. This is not an unusual situation as long as both the risk and the benefit are limited within the population of the primary country, which has full control of the source. It becomes increasingly difficult when some of the risk but little, if any, of the benefit occurs in other countries. What is then 'acceptable' or 'justifiable' on a risk-benefit basis must be subject to international agreements and universally accepted principles. A basic rule would seem to be that a population dose that would not have been judged acceptable on a risk-benefit basis had it fallen within the primary country, should neither be acceptable in any other country.

The fact that the population at risk is usually not identical with the population at benefit would make risk-benefit evaluations impossible on ethical grounds if the individual risks were not insignificant. It is only because each individual has the extra protection afforded by the individual dose limits that it is possible to use any actual distribution of benefits to justify the corresponding distribution of risks, even though different individuals are involved.

However, it is not sufficient to decide that a certain source is acceptable on the basis of a risk-benefit evaluation. To comply with the ICRP recommendation that all doses shall be kept 'as low as is readily achievable, economic and social considerations being taken into account'', an additional cost-benefit analysis of further dose reductions needs to be made.

Various authors have published estimates of the amount of money that is either currently paid per man-rem in reduction of radiation exposures or is judged to be a justified expense. The estimates usually fall between US $25 and US $250 per man-rem. The introduction of a monetary equivalent of one man-rem, say US $100, does not imply that a similarly simple evaluation can be made in risk-benefit evaluations. Both the detriment and the benefit from a certain practice or operation include intangible components that are very difficult to weigh. Even if society sometimes takes decisions on the basis of an actuarial value of human lives, it would often seem to be an unnecessary abstraction to make such calculations.

In cost-benefit evaluations, however, generalizations may be justified. Once the risk-benefit evaluation has established that a certain source is acceptable, it may be helpful to have a standardized approach to the decision on whether further dose reductions are appropriate. For this purpose a reference value of US $100 per man-rem may be used, but should be used with caution.

The sequence risk-benefit evaluation (i. e. to check whether the source is justified) — cost-benefit evaluation (i. e. to check if it might not be used with even lesser exposures) is often not a two-step procedure but a series of steps, which are not always clearly separated from each other.

THE DOSE COMMITMENT (Fig.1)

'Dose commitment' is a concept that has found use in the UNSCEAR assessment of global radiation levels. In mathematical terms, the dose commitment of a certain operation (e. g. 1 MW · year of electric power production) is the infinite time integral of the average dose rate ($\dot{D}$) from this unit of operation in a given population:

$$D_\infty = \int_{t=0}^{\infty} (\dot{D})_{av}\, dt$$

The term 'commitment' implies that the total integral is related to a specified practice or operation at a certain time. The product of the dose commitment from that practice or operation, the number of population and the risk coefficient is a measure of the total expected detriment.

$$N = \mu\ P\ D_\infty$$

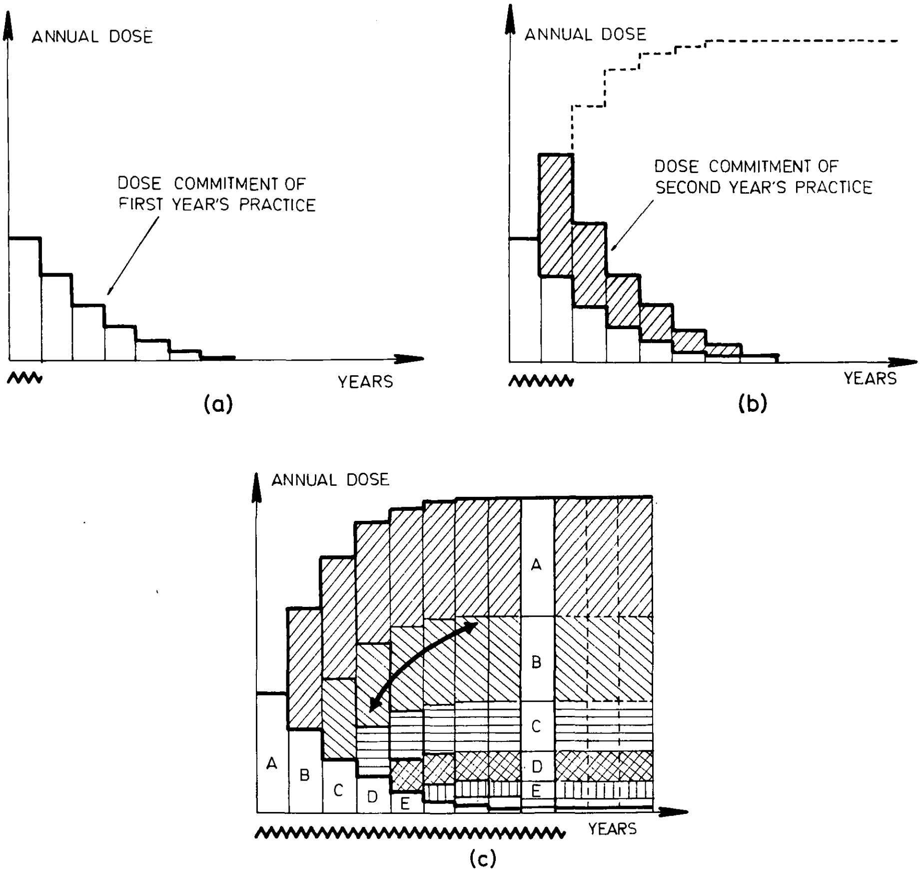

FIG. 1. Illustration of the relation between the annual dose commitment and the annual dose in an equilibrium situation: (a) decrease in annual dose from one year's practice, the sum of the annual doses being the dose commitment; (b) decrease in annual dose from two years' practice; (c) the annual dose is shown to be numerically equal to the dose commitment from one year of release.

Since it is irrelevant in this estimate whether the population P consists of the same individuals or different individuals over the integration period, the product PD_∞ is the collective dose of the given population as a group rather than as individuals. For example, the dose commitment to a critical group of infants near a nuclear power station, from the release of a certain quantity of radioactive material, is the sum of the resulting annual dose averages for that group over all future years, while the infants will be different each year.

The dose commitment makes it possible to give a measure of the total detriment per unit practice (e.g. 1 MW · year), including also the detriment from future exposure from long-lived radionuclides remaining in the environment. For each group (j) for which the dose commitment is assessed, the expected detriment will be $\mu\ P_j\, D_{\infty j}$ and the total detriment per unit practice will be $\mu\ m_\infty$, where

$$m_\infty = \sum_j P_j\, D_{\infty j}$$

If a source releases the same amount of radioactive material year after year, the following 'law' will apply:

> The annual dose at the time when equilibrium has been reached between annual release and annual decay will be numerically equal to the dose commitment for one year of release.

This means that the safest method to prevent high annual doses in the future from radioactive material of long half-lives is to let the annual dose limit apply not to the annual doses but to the annual dose commitments.

PRODUCTION AND RELEASE OF RADIONUCLIDES

(Session I)

Chairmen:

C.A. MAWSON (Canada)
Y. CHERNILIN (IAEA)

PRE-OPERATIONAL ASSESSMENT OF THE DISCHARGE LIMITS AND RELATIVE IMPORTANCE OF RADIOACTIVE AND OTHER WASTES FROM URANIUM PRODUCTION IN AUSTRALIA'S NORTHERN TERRITORY

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Abstract

PRE-OPERATIONAL ASSESSMENT OF THE DISCHARGE LIMITS AND RELATIVE IMPORTANCE OF RADIOACTIVE AND OTHER WASTES FROM URANIUM PRODUCTION IN AUSTRALIA'S NORTHERN TERRITORY.

The paper examines the sources and nature of the wastes likely to arise from the uranium extractive industry now being developed in the Northern Territory of Australia. The area in question is tropical and has a season of monsoonal rain with heavy run-off, which alternates with a long dry season. In the dry season river flow is minimal and parts of the river systems become isolated and subject to heavy loss from evaporation. In consequence, the environmental problems associated with the uranium mines and mills are unusual. Ecological mapping to define sensitive habitats and toxicity studies undertaken on sensitive species indicate that the levels that can be allowed for heavy metals will dictate criteria for treatment and containment of wastes from extraction plants. On the other hand, radiological considerations will govern disposal of pit water and run-off from storage and overburden dumps.

The naturally occurring levels and transfer mechanisms have been examined for uranium, radium and lead. These characteristics can be used to assess the environments of particular mines. For each of four probable mine locations four possible exposure routes for ^{226}Ra and ^{210}Pb can be identified and have been evaluated separately. Although their local environments are very similar, the critical exposure routes differ for at least three of the possible mine sites. This is because transfer and dispersion mechanisms, and land use, differ between the sites.

1. INTRODUCTION

Over the past two years substantial amounts of uranium mineralisation have been announced for the Alligator Rivers area of the Northern Territory of Australia. This region, about 10,000 km^2, is bounded by 131° 30' E to 133° 30' E and 12° S to 13° 30' S. It includes the catchments of Cooper's Creek, East Alligator River, Magela Creek and the eastern catchment of the South Alligator River (see figure 1).

The rainfall of the area is strongly seasonal. It is negligible between April and October, then builds up steeply to high monthly averages in January and February, and drops after March even more steeply than it rises. In contrast to the seasonal fluctuations of the rainfall, temperatures are relatively even and high. Evaporation is substantial and a free water surface would, averaged over a year, show a net loss of some 80 cm after a rainfall of 1.33 m.

Three major physiographic units are present; alluvial valleys and plains, low hills and ridges and the escarpment backed by the Arnhem land plateau.

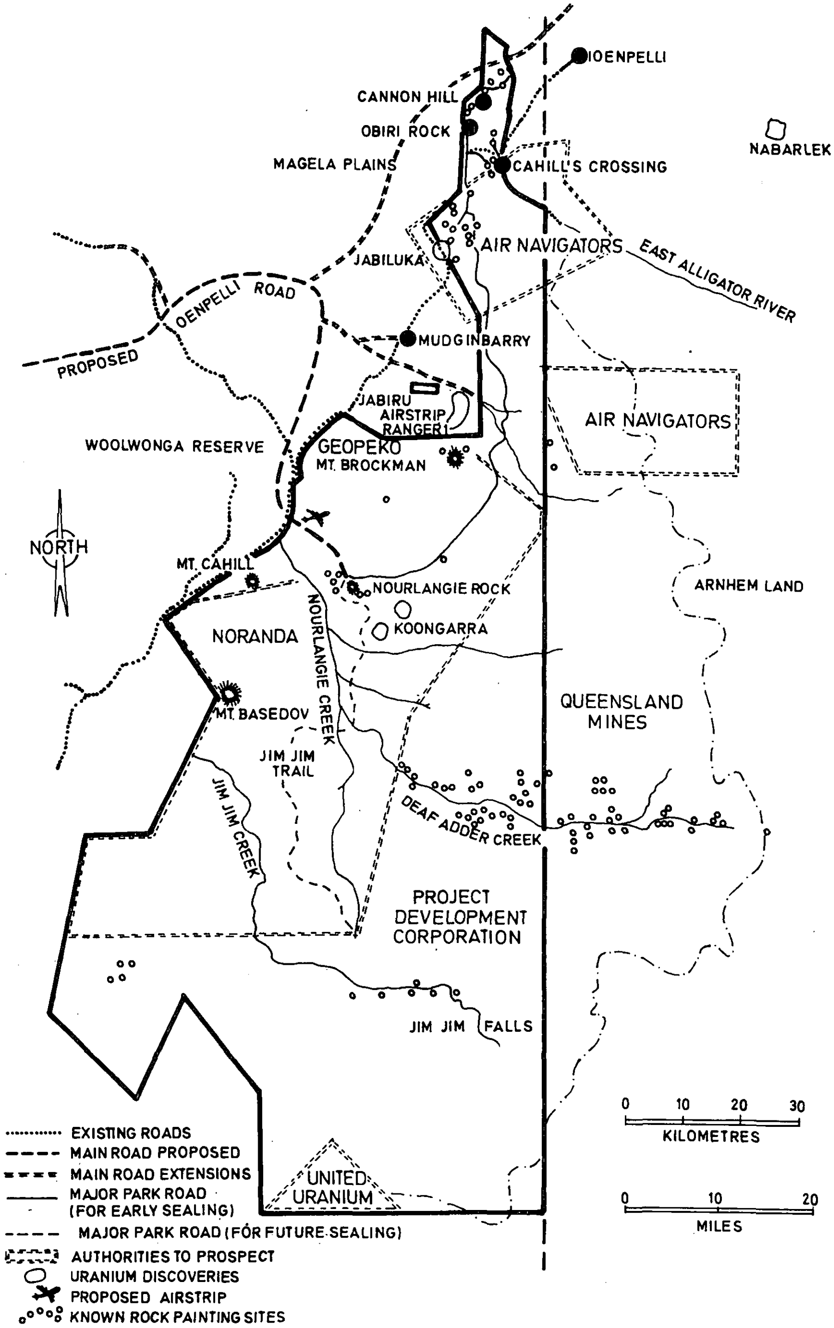

FIG. 1. The Alligator Rivers area of the Northern Territory of Australia.

The alluvial material varies from extensive sand deposits in the valleys of the rivers draining the sandstone plateau to subcoastal black soil plains in the lower reaches. The plains, of gradient 1 in 2000, are subject to annual flooding which in places, is to a depth of about 2 m for a period of up to 6 months.

On the steeper slopes and narrow alluvial flats, streams are markedly seasonal and during the 7 months dry season few continue to flow. Despite this, water tables remain high.

The dissected escarpment rises vertically up to 200 m forming steep sided gorges with numerous waterfalls during the wet season. On the plateau, the high and fluctuating water table causes marked sub-surface weathering of the rock. The land systems of the region have been described by Story et al. [1].

The drastic change in climate is a large factor in the ecology of the area. Plant species must be able to tolerate waterlogged conditions during the monsoons as well as the dry conditions which follow. On the plain reeds flourish during the wet season, but the ground subsequently becomes bare with severe cracking to polygons about 25-30 cm across, the cracks being up to 15 cm wide. The plains are of immense importance to magpie geese (Ansiranas semipalmata); the reeds growing through the floodwaters provide their breeding grounds and the wild rice that grows as the water recedes provides food for their young. Conservation of the black soil plains is a pre-requisite to the survival of this species [2]. Large herds of water buffalo also graze the plains which are held as pastoral leases and support two abattoirs - Mudginbarri and Oenpelli (see figure 1). Mudginbarri draws process water from Magella Creek.

Closely associated with the flood plains, but topographically separated from them, are long shallow depressions with uncoordinated drainage that are permanently waterlogged and under water weed and paper bark (Melaleuca leucadendron). These swamp areas provide valuable dry season refuge for waterfowl. The Woolwonga reserve (see figure 1) contains a large area of such swamps.

Further upstream discrete zones of vegetation can be seen clearly when the floodwaters recede. Away from the stream there is first a sedge meadow then a strip of paperbarks followed by a zone which includes Pandanus and Eugenia sp. (native apples). Beyond this is a small grassy plain which gives way to a Eucalyptus woodland as the ground rises.

Two main types of billabong (waterholes of about 0.15 km^2 area) are seen. Type 1 which has a substrate of fine particle black clay and gradually sloping sides exists most frequently when a condition of back flow of flood waters prevails. Type 2 has a sandy bottom, steep sides and is usually larger. It is generally parallel to the main river course and could be remnants of former river systems.

Type 1 billabongs develop more extensive sedge meadows on the margins during the dry season and the water becomes turbid and warmer. Commonly the bottom muds become anaerobic and a dense layer of micro-algae forms on the top in the warmer areas at temperatures from 32-37°C.

At the end of the dry season billabongs of type 2 are clearer and cooler and present a more moderate environment. Barramundi, a popular sporting and commercial fish, spawn in the estuaries and migrate to the fresh water river/billabong system as juveniles during the next wet.

They may remain in the billabongs for many years preying on a great variety of smaller fish and crustacea.

The upper reaches of the creek systems were once the hunting grounds of aborigines and family groups still live off the land during their frequent walkabouts. The early tribes had a rich culture and the gorges and overhangs of the escarpment abound with sacred sites and galleries of cave paintings of great anthropological and archaeological significance. To preserve these sites and the wilderness character of the area,

TABLE 1. ANALYTICAL RESULTS FOR SIGNIFICANT POLLUTANTS IN EACH WASTE STREAM (RADIOACTIVITY IS EXPRESSED IN UNITS OF pCi/l, ALL OTHER RESULTS ARE IN ppm)

Pollutant	Source of Pollutant			
	Overburden-ore dump	Pit water	Raffinate	
			pH 2	pH 7.8
Total hardness	170	70 - 170	-	-
Ca	1 - 2	2	260	450
Mg	3 - 4	16 - 40	3400	2270
HCO_3		75 - 200	-	-
Fe		1	1900	< 0.1
Zn	0.06 - 0.09	0.1 - 1.0	4.4	0.08
Cu	< 0.03	< 0.05	6.2	0.05
Pb	0.5 - 0.8	< 0.01	4.5	0.6
U	0.2 - 0.5	0.2	0.43	0.02
Co	0.005	< 0.006	2.4	
^{226}Ra	200 - 860	100	218	5.3
^{222}Rn	-	2×10^5	-	-
As	< 0.04	< 0.05	10	3.3
Mo			< 0.2	
Mn	< 0.08	0.1	1170	26
Ni	< 0.1	< 0.1	18.2	< 0.1
PO_4			700	8.7
Amine	-	-	13.6	1.9

TABLE 2. PIT WATER CHARACTERISTICS AT TWO POTENTIAL MINE LOCATIONS (UNITS ARE ppm AND pCi/l)

Location	Water Quality				
	Hardness	Zn	Pb	Cu	^{226}Ra
Nabarlek	~ 120	0.1	< 0.01	< 0.05	56
Ranger	~ 100	0.4	< 0.01	< 0.05	100

a National Park covering 3740 km^2 has been proposed [3]. The suggested border is shown in figure 1.

Economic quantities of uranium ore have been announced for four locations. The respective geology has been described by South [4]. For Ranger I, anomalies 1 and 3, the stated reserves are 82,500 tonnes of contained U_3O_8. The ore bodies are of the large disseminated type with fine grained pitchblende throughout the host rock. These veins also contain minor galena, pyrite, copper sulphide and haematite.

At the Nabarlek deposit, some 60 km N.E. of Ranger I, the uranium occurs in two high grade massive pitchblende cores. Stated reserves are 9,500 tonnes at an average grade of 24 kg/tonne. The pitchblende cores are coated by gummite and other secondary minerals, including curite and kasolite, that are lead-uranium complexes [5].

Limited information is available on the Koongarra uranium deposit, situated 20 km S-SW of Ranger I. Reserves have been stated as being intermediate in size and character between those of the Nabarlek and Ranger I deposits. Some of the secondary mineralisation contains lead.

Exploration of the Jabiluka ore body, 10 km N of Ranger I, is at an early stage and currently stated reserves are 3000 tonnes of contained U_3O_8. This deposit, as is the Koongarra one, is overlain with 3-10 m of sand. The overlay at Ranger I and Nabarlek is schist.

It is anticipated that the milling of ore from all deposits will involve sulphuric acid leaching followed by solvent extraction with alamine 336. Proposed production rates have been announced for only two of the deposits - 2.7 x 10^6 kg of uranium oxide a year for Ranger I and 10^6 kg a year from Nabarlek.

Within Australia over the last 2-3 years there has been a general awakening of the community's concern for the environmental aspect of continued industrial growth. This is now reflected by legislation and Government imposed guide-lines that parallel the environmental impact statement approach developed by the U.S.A. Environmental Protection Agency.

This paper describes work undertaken by the A.A.E.C. to assist Government in the specification of standards to be met by the mining companies. Our programme is a part of a much larger joint Industry-Government fact finding study directed towards the wider issues of land use and includes programmes on wildlife, entomology, anthropology and archaeology.

2. SOURCE AND NATURE OF POTENTIAL POLLUTANTS

Bearing in mind the types of secondary mineralisation and that each development would be an opencut operation, the source and nature of potential pollutants resulting from a uranium extractive industry are:

Source	Potential Pollutants
Top soil dump	Suspended solids in creek systems.
Overburden dump	Suspended solids, dissolved ^{226}Ra, U, Zn, Pb, Cu and other heavy metals.
Pit water	Dissolved ^{222}Rn, ^{226}Ra and heavy metals.
Raffinate (seepage and run-off from tailings dam)	^{226}Ra, uranium and other heavy metals, amines, acidity soluble sulphates and carbonates.

While metallurgical assessment of each of the deposits is at an early stage, the preliminary results from one of them (Ranger) is taken as being representative. Results of analyses for the range of potential pollutants is given in Table 1.

As discussed later, the potential environmental consequences of discharging untreated pit water are important. A comparison of its major characteristics at two of the ore body locations is given in Table 2.

3. LIMITING CRITERIA

In assessing the experimental work on aquatic organisms and plant species aimed at finding the allowable limit for each potential pollutant, the influence of the local hydrology has first to be appreciated. This is illustrated below, base line levels of water quality and of stable and radioactive elements being used to highlight the salient features.

3.1 Local Hydrology

3.1.1 Black soil plains

The geese and the buffalo ensure that the flood waters over the plains are turbid (suspended solids > 1,000 ppm). Conditions are ideal for adsorption on to the clay particles of any dissolved uranium [6], radium [7] and other heavy metals. We think that this adsorbed material will remain on the surface during the subsequent dry season but that the early showers of the next wet season will transport it into the large cracks that have developed in the soil profile by this time. In such a situation, pollutants introduced to the plains area do not accumulate near the surface but instead are fairly uniformly mixed through 1-3 m of black clay.

Table 3 presents some results on the vertical distribution of U, ^{210}Pb, ^{226}Ra and ^{137}Cs in soil samples from the Magela Plains. The results are fairly consistent with the hypothesis that a mixing mechanism is operative and that the mixing mechanism has a time period that is short in comparison with the time variation in fallout ^{137}Cs deposition.

TABLE 3. VERTICAL DISTRIBUTION OF RADIOACTIVE AND STABLE ELEMENTS IN SOIL SAMPLES FROM MAGELA PLAINS

Location (1:50,000 acres)	Depth	Concentration						
		pCi/ℓ				ppm		
		U	^{226}Ra	^{210}Pb	^{137}Cs	Cu	Pb	Zn
670240*	Surface	0.2		0.7	0.1	0.6	8.0	4.4
(West bank)	0.5 m	0.3	0.8	0.8	0.14	0.8	9.0	3.5
	1.0 m	0.6	0.7	0.8	0.1	0.6	9.0	4.0
	1.5 m	0.3		1.1	0.1	0.5	6.0	4.0
670240	Surface	0.4		2.0	0.27	0.9	6.0	3.5
(East bank)	0.5 m	0.3	0.5	1.3	0.17	0.6	7.0	3.5
677256	Surface	0.5		1.0	0.31	0.5	6.0	2.7
(East bank)	0.8 m	0.4	0.9	0.9	0.26	0.6	7.0	3.6
644273	Surface	0.5		1.7	0.27	1.1	7.0	4.9
(West bank)	0.8 m	0.4	0.3	0.9	0.22	0.5	6.0	2.5

* Levee bank

Some 40 locations on the Magela Plains (area 66 km^2) were sampled for surface soil and for a composite sample over a depth of 1 m. The average result and standard deviation of the γ spectroscopy analyses were:

Gamma Energy (MeV)	0.05	0.25	0.35	1.1
Radioactivity (arbitrary unit)	34	3.3	17.5	7.3
Standard deviation	4.6	0.5	2.7	0.8

The reasonably constant percentage standard deviation and its relatively low value indicate that the natural radioactivity entering the system is distributed fairly uniformly through the flood plain soil and that no preferential movement of any radon daughter occurs.

3.1.2 Swamp areas

For swamp areas that are deepest at the end nearest to the black soil plains it is possible to separate the elemental uptake from the flood water (by paperbarks) from the elemental cycling from water to sediments and water weeds following physical concentration by evaporation and related chemical precipitation. This is illustrated in Table 4.

3.1.3 Billabong systems

Some limnological aspects of the differing types of billabong are illustrated in Table 5. Boggy Creek has a black clay substrate, Little Nourlangie a sandy one. Georgetown is intermediate.

Quite marked changes in solute concentration result from more than solar evaporation. Boggy Creek waterhole is naturally eutrophic with very extensive water-lily growth by the end of the wet season. By August only macroalgae are evident and these too diminish in mass by the end of the dry season as a result of high temperatures and receding water line. Billabongs of this type are hydrographically isolated during the dry and lose water by evaporation at a rate of 5.5 - 6.5 cm/week from June to November.

Little Nourlangie appears to be directly connected to the ground water table but the regions of inter-connection could not be identified through seepage measurements (limit of detection was 0.2 cm/week). The drop in water level of this type of billabong is greater than that for type 1 (about 8 cm/week from August-November) but the relative consistency of solute concentration indicates a flowing underground system.

The intermediate type of billabong exhibits back flow during the early part of the wet season and have regions of sandy substrate. However the main difference to type 1 system appears to be that they receive a sub-surface influx of water during the early months of the dry season. For example, Georgetown billabong has an influx equivalent

TABLE 4. THE VARIATION IN ELEMENTAL URANIUM CONCENTRATION (μg/g ash) ALONG A SWAMP DEPRESSION

Map co-ordinates / Sample type	624279	618278	609277	600276*
Sediment	2.3	2.3	2.3	4.0
Bottom weed	3.5	4.2	5.9	-
Paperbark leaves	1.8	0.8	0.46	0.16

* Shallow end

TABLE 5. ELEMENTAL COMPOSITION OF WATERS FROM DIFFERENT BILLABONGS AND THEIR SEASONAL VARIATION (1971/72)

	BOGGY CREEK						GEORGETOWN							LITTLE NOURLANGIE							
	4/11	8/6	6/7	25/8	2/10	6/11	4/11	5/6	6/7	23/8	6/10	6/11	13/11	16/6	7/11	13/4	4/6	8/8	20/9	10/10	10/11
Hardness (ppm $CaCO_3$)	-	8	11	29	5	18	-	-	7	11	10	12	38	7	-	4	9	8	7	7	14
pH	-	4.9	6.2	6.6	5.6	5.4	-	-	6.7	5.7	5.7	5.8	5.4	6.0	-	6.5	6.4	6.4	6.2	6.2	5.8
Total Iron (ppm)	-	<.1	6.7	1.7	18	29	-	-	0.2	17	23	42	42	0.5	-	1.4	0.1	0.1	0.3	0.7	0.5
Uranium (ppb in water*)	1.6	0.6	0.3	3.4	68	49	6.6	-	0.4	0.9	1.1		12	-	1.8	0.2	0.6	0.1	0.5		21
Uranium (ppm in sand)	4.6	0.9	1.5	1.6		2.2	3.8	0.2	0.7			0.9		-	3.6	-	0.1	1.7		9.5	0.1
Radium (pCi/ℓ)	2.3	0.4	0.4	0.8	0.4	0.7	8.5	-	1.3	3.5	0.9	3.2	4.5	-	3.8	-	0.2	2.7	0.3	1.1	.7

* parts per 10^9

to about 3 cm/week through to September which then slowly drops to about 1 cm/week in November. Much of this sub-surface drainage has its origin in the Ranger I deposit and all samples from this region have U and Ra contents well above the average levels for the region.

The quite marked changes in total iron, that occur at Georgetown and Boggy Creek water holes towards the end of the dry season, are attributed to their turbidity. Decaying organic matter can transport substantial quantities of iron at these acidities particularly when the redox potential is low [8].

While these, and all other systems examined, showed increasing radium and uranium levels as the dry season progressed, the increases are not clearly related to changing hardness or decreasing redox potential (as inferred from Fe, Co and Mn determinations). The changes were however independent of pH.

3.1.4 Permanent streams

Many permanent small streams and springs exist at the face of the escarpment. The siliceous sandstone that they drain weathers only slowly and the related soils are well leached. Consequently the water contains very little dissolved inorganic salts (hardness is about 5 ppm $CaCO_3$ eq). However the water does contain large quantities of dissolved and suspended organic material which leads to a brown tannin colourisation and renders it very acidic. The water is poorly buffered so the pH can fluctuate rapidly. This is illustrated in Table 6.

When springs flow through thickly carpeted understory, selective precipitation of the uranium series occurs on the organic debris (peat anomalies) which show unsupported ^{226}Ra (probably through ^{230}Th deposition) to a depth of about 30 cm.

3.2 Experimental work on chemical pollutants

Little information was available on the responses of tropical species, plant or animal, to chemical pollutants. Hence it was necessary to undertake experimental work on the responses of local species to likely pollutants, before meaningful standards for them could be fixed. Local species were tested at a field station under conditions as natural as possible.

3.2.1 Aquatic

Using standard techniques, the 96 hour median tolerance level (96 hr TLm) of the hardihead (Craterocephalus marjorica) was determined for U, Cu, Zn and Pb. Hardiheads are a schooling species which grow to about 7 cm in length. Both the adult fish and 3-7 day old fry were tested. Each experiment was done in 40 ℓ of local creek water held at 25°C and a fresh polythene liner was used on each occasion.

Striped grunters (Amniataba percoides), spangled grunters (Madigania unicolor) and catfish (Neosiluris sp.) were also tested but for some metals testing was confined to two concentrations of toxicant to determine their sensitivity relative to that of hardiheads.

One crustacean (Macrobrachium sp.) was tested completely. The results of these experiments are shown in Table 7.

TABLE 6. TIME VARIATION IN ACIDITY AT BOWERBIRD WATER-HOLE

Date	pH	Date	pH
30.1.72	5.6	20.6.72	4.0
2.3.72	5.9	20.8.72	6.4
22.3.72	6.2		
10.4.72	5.8		
3.6.72	4.1		

TABLE 7. 96 hr TLm's FOR MAGELA CREEK ORGANISMS

Organism	96 hr TLm (ppm)				
	U	Cu	Pb	Zn	Co
Hardiheads (adult)	3.8	0.04	0.17	0.16	< 5
Hardiheads (fry)	3.6*	0.08*	2*	0.3*	
Striped grunter	2.4	>0.1	>0.3	0.2	
Spangled grunter	3.6	>0.1	>0.3	>0.2	
Catfish	72+				
Macrobrachium	> 5	0.12	0.5	0.34	

* 48 hr TLm.

\+ Catfish taken from Georgetown billabong.

TABLE 8. HEAVY METALS IN PIT WATER AND MAGELA WATER

Water Type	Cu(ppm)	Zn(ppm)	U(ppm)	Pb(ppm)	pH	Hardness as (ppm $CaCO_3$eq)
Magela Creek	< 0.04	0.035	0.0004	0.025	7.2	13
Ore Body						
Borehole	< 0.05	0.5	<0.02	0.4	7	99.2

TABLE 9. CHLAMYDOMONAS CELLS SURVIVING (AS A PERCENTAGE OF CONTROL) IN MIXTURES OF ALAMINE-336 (al), KERSENE AND 5% ALAMINE-336 IN KEROSENE (k+al) (ALL SOLUTIONS SHAKEN WITH CULTURE MEDIUM FOR 2 HOURS PRIOR TO USE)

Elapsed Time (hours) / Concentration (ppm V:V)	22			46			70			94			166		
	al	k^{+}	k+al^{++}	al	k	k+al	al	k	k+al	al	k	k+al	al	k	k+al
45	41			11*			9*			8*			8*		
40	43	68	49	11*	39	13*	9*	53	9*	8*	52	9*	8*	60	9*
35	39	59	41	13	31	11*	11	32	8*	29	42	8*	98*	46	8*
30	59	63	36	83	35	10*	81	36	7*	92	38	7*		45	7*
28	40			29			78			78			90		
25	69	70	65	97	59	51	92	69	37	106	71	39		68	
20	75	65	70	101	75	62	105	68	33	100	63	55			
15		74	66		99	74		75	47		88	64			
12	79			113			91			112					
10		68	65		94	66		81	46		88	60			

+ Kerosene volume equivalent to 5% Alamine -336 in Kerosene.

++ 5% Alamine in Kerosene (V:V) to give quoted concentration of Alamine -336 alone.

* Dead cells only remaining in culture.

Striped grunters were also exposed to ground water pumped from the ore body. These fish survived, unaffected, for 204 hours before the experiment was terminated. Table 8 gives a comparison of Magela Creek water and the pit water used.

It is most probable that the increase in hardiness in the pit water reduced the effectiveness of the heavy metals in this experiment.

In laboratory experiments at AAEC Research Establishment, the mosquito fish (*Gambusia* sp.) was exposed to 'mock' raffinate both untreated and neutralised. The 96 hr TLm values occurred at dilutions of 1:5,000 and 1:8 respectively. On the same species, the TLm value for alamine-336 was 3.5 ppm.

Papers containing the full experimental detail of this and further work plus an inventory of the fish species in the area will be published later this year.

3.2.2 Vegetation

As mentioned earlier, sedge meadows surround many of the billabongs and these may become exposed to toxicants as the floodwaters recede. The main plant cover consists of 1% water snowdrop (*Nymphoides minima*), 20% sedge (*Fimbristylis denudata*) and the remainder is the dominant grass (*Pseudoraphis spinescens*).

Plots with an area of 1 m^2 were laid out on a typical flood plain and subjected to fine sediment loads of 10 kg/m^2, 20 kg/m^2, 30 kg/m^2, 40 kg/m^2, 50 kg/m^2 and 60 kg/m^2 by grouting.

Plant growth was not impeded on 10 kg/m^2 and 20 kg/m^2 but was reduced on 30 kg/m^2 and 40 kg/m^2. 50 kg/m^2 and 60 kg/m^2 stopped growth completely.

Similar plots were exposed to U —— (50g, 10g and 1g/m^2), Cu —— (10g, 1g, 0.1g per m^2), Zn —— (10g, 1g and 0.1g per m^2) and Pb —— (100g, 10g, 1g per m^2). None of these plots showed any adverse reaction.

Plots were also watered with Alamine 336 dispersed in water so that doses equivalent to 50 ml, 10 ml and 1 ml per square metre were delivered. All three doses produced chlorosis in the plants, from which the 50 ml/m^2 and 10 ml/m^2 plots did not recover. Following this more quantitative experiments using Alamine 336 were done in the laboratory. *Chlamydormonas* sp. cultured in open flasks was used as the experimental material and the results are given in Table 9. Synergistic effects for the combination of alamine-336 and kerosene are indicated and remain important at alamine levels appropriate to waste streams within processing plants.

Full details of this and further work will also be published later this year.

4. RADIOLOGICAL ASPECTS

4.1 Disequilibrium in environmental radioactivity

Throughout the region disequilibrium exists between the ^{226}Ra and U-238 content for the range of environmental samples. For water samples from streams the Ra/U curie ratio averages at 8.0 with extreme

TABLE 10. DISEQUILIBRIUM RATIOS (Ra/U) FOR A RANGE OF ENVIRONMENTAL SAMPLES

Sample type	Disequilibrium ratio	
	Range	Average
Mussel flesh	$10^2 - 10^3$	300
Mussel shell	1 - 7	3.6
Bottom sediments	1 - 10	5.5
Eucalyptus leaves	1 - 20	9.0

values being two orders of magnitude on either side. Values on the high side appear to be related to streams that drain areas containing radiometric anomalies and that are relatively hard (30-50 ppm $CaCO_3$ eq.) and have a high (~10) Mg/Ca ratio. The lowest values relate to streams that are soft (3-5 ppm $CaCO_3$ eq.), have a Ca/Mg ratio of about one and drain small areas with reasonable slopes. Results for other types of samples are provided in Table 10.

If the drainage systems known to be influenced by nearby uranium mineralisation are excluded, the average ^{226}Ra level in water for the remainder of the area (60 sampling sites) is 0.46 pCi/ℓ. For the influenced systems the average is 4.3 pCi/ℓ.

To evaluate what restraints on the discharge of pit water will be determined by radiological considerations, it is first necessary to determine the concentrating mechanisms for uranium and radium by the various materials that are collected as food stuffs.

4.2 Concentration Factors for Uranium

4.2.1 Mussel

Measured concentration factors for uranium by mussels are presented in fig. 2. The slope of -1 on a log - log plot indicates that the concentrating mechanism is related to the mussel maintaining a regulated pool of uranium at a level of about 80 µg/kg. The radiological implications are therefore trivial.

4.2.2 Buffalo

Typically the uranium contents of soil, grass and water in the buffalo grazing areas are 2 µg/g, 0.01 µg/g and 0.002 µg/g. The uranium content of buffalo flesh is in the range 0.005-0.01 µg/g. There is then no indication of any significant concentrating mechanism for uranium by buffalo. The maximum permissable concentration of uranium in buffalo meat would, on strictly radiological considerations, be some 70 µg/g. It is inconceivable that the general environmental levels of uranium could be raised by a factor of 1000 without environmental degradation resulting from associated contaminants. A similar conclusion holds with respect to buffalo offal. The uranium contents of kidney and liver do not differ markedly from that of the flesh.

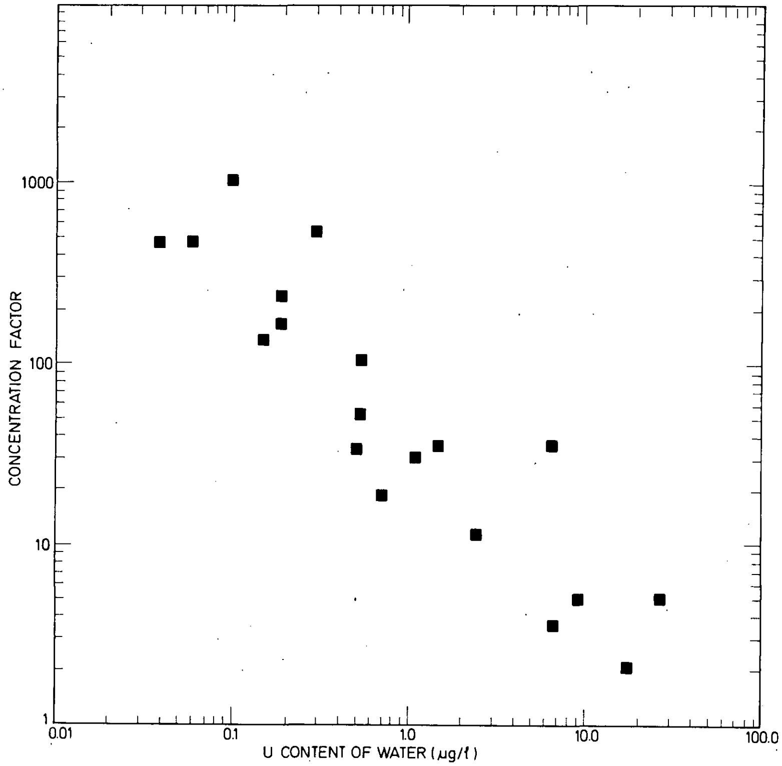

FIG.2. The variation of the concentration factor for uranium by mussels as a function of environmental levels of uranium.

4.2.3 Goose

Geese are more migratory than buffalo and their eating habits are such that their uranium intake, via sediments, would be greater. However, the uranium content of goose flesh is of the same order as that for buffalo.

4.2.4 Fish

Barramundi do concentrate uranium. Present data are not sufficient to define accurately the variation in concentration factor with the uranium content of the water but indications are that one is dealing with a regulated pool of about 0.02 ppm. Should subsequent work confirm this, then there would be no radiological implications.

Scavenger fish (e.g. catfish) if anything discriminate against uranium.

4.2.5 Native and European fruits and vegetables

Native apples, plums and mangoes show no significant concentrations of uranium (0.01 - 0.06 μg/g). The same is true for water-lilies that are eaten by aborigine children. Crops of peas, beans, carrots, cucumbers, cabbages, lomboks, beetroot, tomatoes and sweet corn were grown in the area and watered regularly from a source with known uranium and radium content. No significant concentrations of uranium were found in any of these vegetables, the range being 0.003 - 0.09 μg/g, with beetroot showing the highest concentration.

4.3 Concentrating Factors of Radium

4.3.1 Mussels

The concentration factors for radium by mussels are large. Measured values range from 10^2 to 10^4. An analysis of the data rules out the possibilities that one is dealing with either an exchangeable or a regulated pool of radium in mussels. Nor does the data support the concept that the radium is discriminated against during a metabolic concentrating process for iron, magnesium or calcium. The last mentioned, though not a major factor, is involved to some extent. The Ra/Ca ratio for water is typically 10^{-9}; for mussel flesh, 5.6×10^{-11} and for mussel shell, 1.3×10^{-12}. However, the data, rather than extrapolating to a concentration factor of 1 at a water calcium level of 1800 ppm (that of calcium in mussel flesh), simply shows a general trend to higher values as the dry season progresses. It is most likely more related to the Ra content of the bottom substrate than to anything else.

The preferred native protein food of aborigines consists of kangaroo, wallaby, possum, duck and goose. It would be exceptional indeed if groups of aborigines depended primarily on native aquatic foods for any length of time. A derived level for radium in mussels calculated on an assumed consumption of 2 kg/y is 4.5 pCi/g. There will be then some billabong systems for which the exposure route associated with mussels will be more limiting than that for drinking water by the end of the dry season.

4.3.2 Fish

Not enough data has been obtained for defining the nature of the concentrating mechanism of barramundi for radium. It appears that the radium content of barramundi is generally about 0.01 times that in mussels sampled at the same time. If further work confirms this then the exposure route associated with barramundi would be no more limiting than that related to mussels.

4.3.3 Goose and Buffalo

The radium level in buffalo and goose flesh sampled from the Magela plains area is, on average, 0.007 pCi/g. For an assumed consumption of 130 kg/y(350 g/d) the derived level for ^{226}Ra is 0.08 pCi/g. Assuming further that the exposure routes associated with meat consumption, drinking water and fish/mussel consumption are given equal weight, the maximum permissable concentration of ^{226}Ra in meat becomes 0.027 pCi/g i.e. about a factor of four up on naturally occurring levels.

To a first approximation the ^{226}Ra input to the Magela plains is simply the run-off from the catchment (870 km^2) of the Magela Creek system (10^{12} ℓ/y) by the average ^{226}Ra content of that water. Table 11 presents results for a sampling station on Magela Creek.

During this period the average suspended solids was 40 ppm and its ^{226}Ra content was equivalent to 0.07 pCi/ . The net annual input of ^{226}Ra to the Magela plains is of the order of 0.5 Ci.

TABLE 11. MEASURED ^{226}Ra CONTENT OF MAGELA CREEK WATER DURING THE WET SEASON

Date	pCi/ℓ	Date	pCi/ℓ
25/1/72	0.04	22/3/72	0.43
19/2/72	1.39	14/4/72	0.12
24/2/72	0.41	3/6/72	0.06
4/3/72	0.09	19/6/72	0.9

Table 11 is not weighted for river flow at time of sampling. The range of values indicate the need for proportional sampling devices.

An estimate, believed to be an upper value, for the volume of pit water from the Ranger I anomaly I open-cut, is 4.6×10^9 ℓ/y averaged over the expected life of the mine (20 years). Should this water continue to maintain a ^{226}Ra content of about 100 pCi/ℓ, the annual input of ^{226}Ra to the Magela plains from this source is of the same order as the natural input. Thus while there should not be any serious consequences in the long term, care will have to be exercised with respect to drinking water tolerances and more localised build-ups of ^{226}Ra in drying out swamps. This latter point can be emphasized by considering the flood plains of Ja-Ja lagoon near the Jabiluka ore body. Here the natural level of ^{226}Ra in surface soil is 20 pCi/g.

4.3.4 Native and European Fruits and Vegetables

Results for the ^{226}Ra content of the range of fruits and vegetables sampled are in the range 0.001 - 0.07 pCi/g with beans having the highest value. While no significant concentrating mechanisms are indicated, there is again a need to consider possible build-ups. What has been stated above for buffalo applies equally well here.

5. DISCUSSION

The sensitivity of the aquatic environment to the heavy metal contaminants produced during uranium milling will control certain aspects of the operation. Firstly, the raffinate should be discharged to a tailings storage dam. Secondly, to take advantage of lower initial concentrations and the exchange capacity of any aquifer involved in bedrock seepage from the tailings storage dam, the raffinate should be raised to a pH of 7-8. The combination of these requirements suggest that the final arrangement would involve maximum recirculation of water within the plant. For a scale of operation similar to that envisaged for Ranger I, this means a liquid discharge of about 4×10^8 ℓ/y to the storage dam.

Temporarily forgetting the threshold nature of chemical toxicity effects, the chemical pollution potential of the treated raffinate is not much lower than that contained in the pit water. For ^{226}Ra the position is quite different. The annual output of ^{226}Ra in pit water is about 200 times that in treated raffinate and about 4 times that of untreated raffinate.

Because of varying land use in the vicinity of each uranium deposit there will be differing restraints on the disposal of pit water. In all cases the barramundi/mussel exposure route exists but it involves a substantial dilution factor from the point of discharge to the point of interest for the Koongarra and Nabarlek deposits. As the Koongarra deposit is in a proposed National Park, there is no exposure route associated with market gardening or buffalo meat production (feral buffalo is used as pet food). The reverse is true for the RangerI and Jabiluka deposits.

Uncertainty exists on the mechanism of uptake of ^{226}Ra by barramundi and mussels. This is one case where cultivation under controlled conditions should yield more reliable information than that obtained in the field.

6. ACKNOWLEDGEMENTS

A large number of chemical analyses were carried out in connection with this study. Only a small fraction are presented here.

We gratefully acknowledge the efforts of Mr. J. Jones, Water Resources Branch, Department of the Northern Territory, and of the analysisists within the Chemical Technology and Health and Safety Division of the Australian Atomic Energy Commission.

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ОБ ЭФФЕКТИВНОСТИ ИСПОЛЬЗОВАНИЯ ОЧИСТКИ ВОЗДУШНЫХ ВЫБРОСОВ В ПРОИЗВОДСТВЕ ТЕПЛОВЫДЕЛЯЮЩИХ ЭЛЕМЕНТОВ ИЗ ОБОГАЩЕННОГО УРАНА

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Abstract–Аннотация

EFFECTIVENESS OF WASTE AIR CLEANING IN THE FABRICATION OF FUEL ELEMENTS FROM ENRICHED URANIUM.

The development of nuclear power engineering and the increase in the number of nuclear power stations have made it necessary both to enlarge fuel element fabrication capacity and to ensure the radiation safety of operating personnel and of the population living in the vicinity of such plants. From an examination of different methods of waste air cleaning it has been possible to determine the best way of removing alpha-active aerosols producted by ^{234}U, ^{235}U and ^{238}U. Local cleaning of the air removed from equipment (by a filter installed within the same housing) together with subsequent general cleaning provides for adequate radiological protection in operating the filters within the plant and reduces radioactive contamination outside to levels not exceeding the average permissible concentration for the atmosphere ($4,4 \times 10^{-15}$ Ci/litre).

ОБ ЭФФЕКТИВНОСТИ ИСПОЛЬЗОВАНИЯ ОЧИСТКИ ВОЗДУШНЫХ ВЫБРОСОВ В ПРОИЗВОДСТВЕ ТЕПЛОВЫДЕЛЯЮЩИХ ЭЛЕМЕНТОВ ИЗ ОБОГАЩЕННОГО УРАНА.

Развитие ядерной энергетики и рост числа атомных электростанций обусловливает необходимость как развития предприятий по выпуску твэлов, так и необходимость обеспечения радиационной безопасности для работающего персонала и населения, проживающего вблизи указанных предприятий. Оценка различных способов очистки воздушных выбросов позволила обосновать наиболее оптимальный вариант очистки воздуха от альфа-активных аэрозолей, обусловленных изотопами урана-234, урана-235 и урана-238. Применение локальной очистки воздуха, удаляемого от оборудования (при установке фильтра внутри укрытия) с последующей общей ступенью очистки обеспечивает должные меры радиационной безопасности как при эксплуатации фильтров в производственных помещениях, так и очистку выбросов до величин, не превышающих СДК для атмосферного воздуха ($4,4 \cdot 10^{-15}$ Ки/л).

Развитие ядерной энергетики и рост числа атомных электростанций обуславливает необходимость как развития предприятий по выпуску твэлов, так и необходимость обеспечения радиационной безопасности для работающего персонала и населения, проживающего вблизи указанных предприятий.

Одним из важных вопросов в этом направлении являются защита воздушной среды от загрязнения альфа-активными аэрозолями и достижение эффективной очистки выбросов до величин, не превышающих средне-годовой допустимой концентрации [1] для атмосферного воздуха.

Проведенные исследования позволили выявить источники образования альфа-активных аэрозолей, уровень концентраций их внутри укрытий

ТАБЛИЦА I. СРАВНИТЕЛЬНОЕ СОДЕРЖАНИЕ АЛЬФА-АКТИВНЫХ АЭРОЗОЛЕЙ ВНУТРИ БОКСОВ ПРИ ПРОВЕДЕНИИ ОТДЕЛЬНЫХ РАБОЧИХ ОПЕРАЦИЙ С ОБОГАЩЕННЫМ УРАНОМ

NN пп.	Наименование операций	Содержание аэрозолей в боксе, $n \cdot 10^{-15}$ Ки	Рабочее разряжение в боксе, мм.вод.ст.
1.	Пересыпка порошкообразных продуктов	1600	12-15
2.	Взвешивание	1200	10-12
3.	Просев	2000	7-9
4.	Дробление, просев	6000	10-15
5.	Обработка компактных изделий	300	7-10
6.	Механическая обработка	160	8-11
7.	Очистка	68	5-9
8.	Обрабогка шихты	40	20
9.	Контроль	30	10

технологического оборудования, а также сопоставить данные по содержанию аэрозолей в воздушных выбросах до и после очистки, т.е. оценить степень ее эффективности.

Было установлено, что концентрации альфа-активных аэрозолей, обусловленные изотопами урана-234, урана-235 и урана-238, могут значительно колебаться в зависимости от вида выполняемых операций (табл.I). Максимальные концентрации наблюдались внутри боксов при обработке порошкообразных продуктов, минимальные – при обработке компактных изделий.

Ввиду достаточной герметичности перчаточных боксов при разрежении 7-15 мм вод.ст., концентрации аэрозолей в воздухе в рабочих помещениях не превышали среднегодовых допустимых концентраций (СДК-1, $3 \cdot 10^{-13}$ Ки/л) и в большинстве случаев были значительно меньшими.

В целях предупреждения загрязнения атмосферного воздуха производственными выбросами, а также исключения потерь продукта, все местные вытяжные системы оборудованы фильтрами.

Исследования проводились на 2-х предприятиях. На одном из них воздух, удаляемый от боксов, где размещено различное производственное оборудование, подвергался двухступенчатой очистке: 1-ая ступень представляла собой ультратонкие волокна стекловолокна диаметром 1-1,5 мкм; 2-ая ступень – фильтр из ткани марки ФПП или ФПА, т.е. слой ультратонких волокон перхлорвинила или ацетилцеллюлозы, нанесенные на марлевую подложку [2]. При этом первая ступень очистки предназначалась для улавливания продуктов сгорания масел и других загрязнений с целью предотвращения "забивания" последующей ступени очистки.

Выброс осуществлялся через трубы высотой 20 и 15 метров. При указанном способе очистки эффективность составляла 95-99,4%.

На втором предприятии использовалась одноступенчатая очистка на фильтрах ФПП (на отдельных боксах или группе боксов) с последующим централизованным удалением выбросов через трубу, высотой 50 метров. Данные по эксплуатации различных очистных сооружений свидетельствуют о том, что в случае комбинированных загрязнений, где помимо аэрозолей обогащенного урана возможно поступление в выбросы различных продуктов сгорания, масел и проч., необходим тщательный контроль за работой фильтров и своевременная их замена во избежание повышения сопротивления фильтров и, следовательно, уменьшения производительности работы вентиляционных систем. Последнее может сказываться на на изменении кратности воздухообмена в помещениях. Так, в частности, до замены фильтров кратность воздухообмена в одном из рабочих помещений составляла 12,7 обмена в час, после замены фильтра повысилась до 17,0 обм/ч. Причинами снижения разрежения в боксах помимо повышения сопротивления фильтров могут быть и другие причины, как-то, негерметичность боксов после ремонта оборудования, открывание дверей форкамер и др.

Анализ полученных данных по эксплуатации различных фильтров показал, что наилучшим вариантом является применение локальной очистки воздуха, т.е. установка фильтра внутри укрытия или бокса. Это дает возможность следить за разрежением в боксе по манометру, установленному на каждом боксе, и, в случае необходимости, своевременно производить замену фильтра. При этом фильтр не вынимается из кожуха, а вместе с последним помещается в контейнер и удаляется из бокса взамен поставленного нового. При установке фильтров за пределами боксов не исключены случаи загрязнения воздуха рабочих помещений при операциях замены фильтров. При одноступенчатой очистке на фильтрах типа ФПП на другом обследованном предприятии была достигнута эффективная очистка удаляемого воздуха, которая составляла 96,6%-99,6% (табл.II).

Несмотря на высокую фильтрацию способность тканей типа ФПП, наличие проскока может обусловливаться некоторой негерметичностью между тканью и конструктивными элементами фильтра.

Воздух, удаляемый от боксов, подвергается дальнейшему разбавлению за счет выбросов, удаляемых системами общеобменной вентиляции.

Проведенные исследования позволили не только оценить эффективность очистки воздуха, как было показано выше, но определить величины валового выброса альфа-активных аэрозолей в воздухе, выбрасываемом вентиляционными системами после очистки и уровни загрязнения атмосферного воздуха в точке приземления факела.

Полученные среднемесячные величины валового выброса за годичный период наблюдения при одноступенчатой очистке на фильтрах типа ФПП, представлены в табл.III.

Из таблицы видно, что величины валового выброса находились в пределах $1{,}28 \cdot 10^{-2}$ Ки/мес – $3{,}96 \cdot 10^{-2}$ Ки/мес, при этом средняя величина составляла – $1{,}96 \cdot 10^{-2}$ Ки/мес.

С целью сопоставления фактических концентраций альфа-активных аэрозолей в точке приземления факела от централизованного выброса предприятия с среднегодовой допустимой концентрацией (СДК), рекомендуемой НРБ-69 [1] были проведены расчеты по методам, предложенным отечественными авторами С.А.Клюгиным [3], П.А.Андреевым [4] и В.М.Крупчатниковым [5].

ТАБЛИЦА II. НЕКОТОРЫЕ ДАННЫЕ ПО ЭФФЕКТИВНОСТИ ОЧИСТКИ ВОЗДУХА, ЗАГРЯЗНЕННОГО АЛЬФА-АКТИВНЫМИ АЭРОЗОЛЯМИ, УДАЛЯЕМОГО ОТ БОКСОВ

Условные наименования очистного устройства у бокса	Концентрация аэрозолей $n \cdot 10^{-16}$ Ки/л		Эффективность очистки, %
	До очистки	После очистки	
Ф-1	320	5,5	98,2
Ф-2	38	1,0	97,3
Ф-3	260	3,0	98,8
Ф-4	1040	5,0	99,5
Ф-5	91	3,0	96,6
Ф-6	1170	4,5	99,6
Ф-7	390	8,0	97,9
Ф-8	1040	6,0	99,4
Ф-9	650	6,5	99,0
Ф-10	390	4,5	98,8
Ф-11	326	8,0	97,5
Ф-12	110	2,5	97,8

ТАБЛИЦА III. СРЕДНЕМЕСЯЧНЫЕ ВАЛОВЫЕ ВЫБРОСЫ АЛЬФА-АКТИВНЫХ АЭРОЗОЛЕЙ

Период наблюдения	Суммарный выброс альфа-активных аэрозолей, $n \cdot 10^{-2}$ Ки/мес	Период наблюдения	Суммарный выброс альфа-активных аэрозолей, $n \cdot 10^{-2}$ Ки/мес
Январь	1,56	Июль	1,36
Февраль	1,86	Август	1,28
Март	1,9	Сентябрь	2,48
Апрель	2,32	Октябрь	2,16
Май	1,76	Ноябрь	3,96
Июнь	1,36	Декабрь	1,72

По С.А.Клюгину:

$$C_{\text{макс}} = \frac{G}{8{,}5\ W_{\text{в}}\ Z^2}\ \text{мг/м}^3\,(\text{мКи/м}^3),$$

где $C_{\text{макс}}$ – максимальная концентрация выбрасываемого вещества в воздухе у точки приземления воздушного факела, в мг/м^3 (мКи/м^3),
G – расчетное количество вещества, выбрасываемого из трубы в мг/с,
$W_{\text{в}}$ – скорость ветра на высоте выброса воздуха из трубы, в м/с,
Z – эффективная высота выброса, измеряемая от уровня земли у основания трубы, в м.

По П.А.Андрееву:

$$C_{\text{макс}} = \frac{285 \cdot G}{W_0 \cdot Z^2}\ \text{мг/м}^3\,(\text{мКи/м}^3),$$

где $C_{\text{макс}}$ – максимальная концентрация выбрасываемого вещества в воздухе у точки приземления воздушного факела в мг/м^3 (мКи/м^3),
G – количество выбрасываемого из трубы вещества, в г/с,
W_0 – скорость ветра на высоте 10 м от земли, в м/с.

Расчет приземных концентраций проведен при 3-х значениях суммарного выброса альфа-активных аэрозолей из трубы: $1{,}28 \cdot 10^{-2}$ Ки/мес, $1{,}96 \cdot 10^{-2}$ Ки/мес и $3{,}96 \cdot 10^{-2}$ Ки/мес, при условии действия точечного непрерывного источника, высотой 50 м и различных величин скорости ветра на высоте выброса.

Полученные данные отражены в табл.IV.

Из анализа представленных данных видно, что максимальная расчетная концентрация в точке приземления факела может составлять в условиях штилевой погоды (при скорости ветра – 1 м/с) – $14{,}4 \cdot 10^{-16}$ Ки/л.

При скорости ветра порядка 3-5 м/с максимальные концентрации альфа-активных аэрозолей в атмосферном воздухе составляли $2{,}8 \cdot 10^{-16}$ – $4{,}8 \cdot 10^{-16}$ Ки/л.

Расстояние от источника выброса на предприятии до места приземления воздушного факела в условиях горизонтальной местности рассчитывалось по формуле:

$$x_{C_{\text{макс}}} = \sqrt[m]{\frac{Z^2}{S^2}}\ \text{м},$$

где Z – высота от земли до оси потока, м;
m – коэффициент, определяющий влияние температуры атмосферного воздуха ($m = 2{,}0$);
S – коэффициент турбулентности атмосферы, зависящий от характера местности.

С учетом коэффициента S, равного для рассматриваемого случая 0,0832, расстояние от точечного источника до места приземления факела находится в пределах радиуса 750-800 м.

ТАБЛИЦА IV. СРАВНИТЕЛЬНЫЕ ДАННЫЕ ПО КОНЦЕНТРАЦИИ АЛЬФА АКТИВНЫХ АЭРОЗОЛЕЙ В АТМОСФЕРНОМ ВОЗДУХЕ В ТОЧКЕ ПРИЗЕМЛЕНИЯ ФАКЕЛА

Скорость ветра в м/с	Концентрация альфа-активных аэрозолей в атмосферном воздухе, $n \cdot 10^{-16}$ Ки/л		
	миним.	макс.	средн.
	I. По Клюгину С.А.		
1,0	2,4	7,2	3,6
3,0	0,8	2,4	1,2
5,0	0,4	1,4	0,7
10,0	0,2	0,7	0,3
	II. По Андрееву П.А.		
1,0	4,8	14,4	7,2
3,0	1,6	4,8	2,4
5,0	0,9	2,8	1,4
10,0	0,4	1,4	0,7

Замеренные в натурных условиях концентрации в указанном радиусе колебались в пределах $0{,}8 \cdot 10^{-16}$ – $2{,}1 \cdot 10^{-16}$ Ки/л, которые близки к значениям, полученным по теоретическим расчетам.

Таким образом, применение эффективной очистки вентилляционных выбросов в производстве тепловыделяющих элементов из обогащенного урана позволяет обеспечить радиационную безопасность для населения, проживающего вблизи указанных предприятий.

ВЫВОДЫ

1. При использовании фильтров из ткани ФПП для одноступенчатой очистки воздуха, загрязненного альфа-активными аэрозолями, была достигнута эффективность очистки 96,6-99,6%.

2. При наличии комбинированных загрязнений, где помимо альфа-активных аэрозолей имеют место продукты сгорания технических масел, целесообразно на отдельных вентилляционных системах или боксах использование двухступенчатой очистки в виде стекловолокна и ткани ФПП, что позволяет удлинить срок эксплуатации фильтров ФПП.

3. Использование локальной очистки воздуха, удаляемого от оборудования (при установке фильтра внутри укрытия), обеспечивает должные меры радиационной безопасности при эксплуатации и замене фильтров, а также позволяет организовать оперативный контроль за их работой.

4. Расчетные приземные концентрации в атмосферном воздухе с учетом среднегодового валового выброса активности из вентиляционных систем при одноступенчатой очистке на фильтрах типа ФПП составляли

$1{,}4\cdot10^{-16}$ – $2{,}4\cdot10^{-16}$ Ки/л, что значительно ниже среднегодовой допустимой концентрации (СДК) в атмосферном воздухе – $4{,}4\cdot10^{-15}$ Ки/л, принятой в НРБ-69.

5. Теоретически рассчитанные по формулам отечественных авторов концентрации альфа-активных аэрозолей в точке приземления факела удовлетворительно согласуются с измерениями, проведенными в натурных условиях.

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GASEOUS RADIOACTIVE EMISSIONS FROM REPROCESSING PLANTS AND THEIR POSSIBLE REDUCTION

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Abstract

GASEOUS RADIOACTIVE EMISSIONS FROM REPROCESSING PLANTS AND THEIR POSSIBLE REDUCTION.

The most important gaseous and volatile radioactive isotopes emitted from reprocessing plants are ^{3}H, ^{85}Kr, ^{129}I and ^{131}I. In addition, HTR plants also release ^{14}C resulting from the burning of the graphite matrix of the irradiated fuel elements.

To conform to the German policy to limit the dose rate to 30 mrem/yr in the vicinity of nuclear plants, the different gaseous and volatile radioisotopes must be retained to a greater or lesser extent. Decontamination factors of 100 are sufficient for the retention of ^{3}H and ^{85}Kr. For iodine, however, decontamination factors greater than 2000 may be necessary for FBR fuel. Since the separation of ^{14}C from inactive carbon dioxide or a quantitative retention of the burner off-gas is not economically feasible, ^{14}C will be quantitatively emitted. Methods for the decontamination of different types of off-gases are described. The processes under development will be able to fulfil the demand for the retention of ^{3}H and ^{85}Kr. However, further development is necessary to obtain adequate iodine retention.

1. Introduction

In considering different types of power plants for the production of electricity, technical, environmental and economic arguments in favour of nuclear energy have lead to the construction of an ever increasing number of nuclear power plants. As a consequence, increasing capacity for the reprocessing of spent fuel elements is required. This necessitates not only increasing the number of plants, but also their sizes, to obtain the lowest possible fuel cycle costs.

In general, the potential for the release of gaseous radioactive materials from reprocessing plants is much higher than for nuclear power stations. All of the ^{85}Kr present in the irradiated fuel is released to the plant off-gas, and currently to the atmosphere. A fraction of the tritium is also released to the atmosphere at this time, although most of it is transformed to tritiated water, incorporated into the liquid waste stream of the plant and discharged as such. Other isotopes like ^{131}I and ^{133}Xe have decayed in LWR and HTR fuel due to sufficiently long cooling times, or as in the case of long-lived ^{129}I their radioactivity is very low. Therefore these isotopes require no extra consideration for LWR and HTR fuel; adequate protection is achieved by dilution of the radioactivity. In reprocessing FBR fuel however, adequate iodine retention is still a problem to be solved.

There are now plans for a commercial reprocessing plant designed for light water reactor (LWR) fuel elements having a plant capacity of approximately 42 000 MWe of installed nuclear power, corresponding to about 1 500 t of heavy metal throughput per year, to go into operation in the early 1980's in the Federal Republic of Germany. In addition, a second plant of somewhat smaller size designed especially for the reprocessing of high-temperature reactor (HTR) fuel elements is planned for the last decade of this century. For future optimization of fuel cycle costs, cooling times for spent fuel elements must be fixed at approximately 150 days for the cases of LWR and HTR fuels. Fast breeder reactor (FBR) fuel elements require much shorter cooling times to guarantee a low inventory of stored fissile plutonium. The shortest practical time would be about 30 days, neglecting the tremendous technological problems of safety and heat removal encountered. A more realistic figure is based upon an upper limit of 90 days cooling.

Under both these conditions, a quantitative emission of the gaseous and volatile radionuclides would lead to an unacceptable radiation burden to the population living around the reprocessing plant. Dilution of the radioactivity is no longer adequate for safety. Therefore new methods for the separation of the gaseous and radioactive isotopes must be developed. To optimize the efficiency of the off-gas cleaning, the entire plant, but particularely several units of the reprocessing equipment (e. g. fuel dissolver), must be designed to minimize dilution of the off-gas streams prior to the cleanup systems.

The calculations and considerations for nuclear fuel reprocessing plant off-gas treatment and emissions presented in this paper are based upon an installed nuclear power of 50 000 MWe for the three different reactor types. The corresponding throughputs of heavy metal are:

1 750 t/yr for LWR
375 t/yr for HTR
500 t/yr for FBR.

The potential emission values and required decontamination factors for all important radioisotopes in reprocessing off-gases for the three reactor type fuel elements are summarized in table I. The dose rates at the most unfavourable point near the plant caused by quantitative emission, assuming a dispersion coefficient $\varkappa = 1 \cdot 10^{-7}$ sec $\cdot$ m^{-3}, are also included in table I. Selecting the meteorological conditions at the area of the Kernforschungsanlage Jülich as a model, this dispersion can be realized by a 150 m stack [1] .

2. Estimation of required decontamination factors

The layout and location of a reprocessing plant will be strongly influenced by existing regulations with regard to gaseous and liquid effluents. In the Federal Republic of Germany, the discharge of radioactive liquid and gaseous effluents must comply with § 34 of the First Radiation Protection Ordinance [2] . In an appendix to this ordinance, maximum permissible concentrations for all important radioisotopes in water and air are listed.

These data provide a basis for radioactive discharges from nuclear plants. Dilution factors may be calculated and applied for waste streams, if higher concentrations arise in the primary streams. In any case, the possible exposure of persons outside the controlled area around the plant is not allowed to exceed 0.5 rem/yr. However, to avoid continued surveillance of the neighbouring area according to § 22 No. 2 of the above mentioned ordinance, the possible exposure of persons outside the plant fence must be kept below 150 mrem/yr.

At present, a national recommendation is being considered to allocate to gaseous emissions from nuclear plants 1/5 of the maximum permissible radiation dose of 5 rem/30 yr, recommended by the International Commission for Radiation Protection (ICRP) for a genetic radiation exposure of the population from all sources; that means 1 rem/30 yr. This corresponds to a recommended maximum whole-body dose caused by radioactive gaseous

TABLE I. GASEOUS AND VOLATILE RADIOISOTOPES RELEASED IN REPROCESSING LWR, HTR AND FBR FUEL ELEMENTS AND THE MAXIMUM DOSE RATES EXPECTED AFTER QUANTITATIVE RELEASE FROM A 50 000-MW(e) PLANT $\kappa = 1 \times 10^{-7}$ s · m^{-3}

Reactor Type	LWR				HTR					FBR				
Burn-up [MWd /t]	30 000				100 000					80 000				
Throughput [t /yr]	1 750				375					500				
Throughput [t /d]	5.8				1.25					1.7				
Cooling Time [d]	150				150					90				
Radionuclides	^{3}H	^{85}Kr	^{129}I	^{131}I	^{3}H	^{14}C	^{85}Kr	^{129}I	^{131}I	^{3}H	^{85}Kr	^{129}I	^{131}I	^{133}Xe
Emission [Ci /yr]	$1.3 \cdot 10^{6}$	$2.7 \cdot 10^{7}$	73	$5.4 \cdot 10^{3}$	$1.6 \cdot 10^{6}$	$5.0 \cdot 10^{3}$	$1.6 \cdot 10^{7}$	40	$4.3 \cdot 10^{3}$	$1.5 \cdot 10^{6}$	$1.6 \cdot 10^{7}$	58	$1.2 \cdot 10^{6}$	$8.9 \cdot 10^{4}$
Inhalation Dose [mrem/yr]	12.8	-	0.95	416	15.7	0.3	-	0.5	330	15	-	0.8	$9.3 \cdot 10^{4}$	-
Submersion-β-Dose [mrem/yr]	0.3	196	-	-	0.4	$7.0 \cdot 10^{-3}$	115	-	-	0.4	120	-	-	1.3
Submersion-γ-Dose [mrem/yr]	-	132	-	-	-	-	78	-	-	-	79	-	-	2.5
Ingestion Dose [mrem/yr]	64	-	$1.9 \cdot 10^{4}$	$8.5 \cdot 10^{4}$	79	20	-	$1.0 \cdot 10^{4}$	$6.1 \cdot 10^{4}$	75	-	$1.5 \cdot 10^{4}$	$1.9 \cdot 10^{7}$	-
Total Exposure [mrem/yr]	77	328	$1.9 \cdot 10^{4}$	$8.6 \cdot 10^{4}$	95	20	193	$1.0 \cdot 10^{4}$	$6.1 \cdot 10^{4}$	90	200	$1.5 \cdot 10^{4}$	$2.0 \cdot 10^{7}$	3.8
Required Decontamination Factor	20	50	2 000		100	-	100	3 000		20	100	$5 \cdot 10^{5}$		-
Experimental Decontamination Factor	100	500	1 000		100	-	500	1 000		100	500	1 000		-

effluents in the vicinity of a fuel reprocessing plant of 30 mrem/yr. To meet these recommendations, the release of gaseous and volatile radioisotopes from a "50 000 MWe" reprocessing plant must be reduced considerably below the quantitative values.

The required decontamination factors are calculated assuming a dose rate

$$D = D(^{3}H) + D(^{85}Kr) + D(^{14}C) + 1/3\ D(^{129+131}I) \approx 30 \text{ mrem/yr.}$$

A further reduction is required, if the radiation burden caused by aerosols of nonvolatile isotopes like cesium, strontium and ruthenium must also be considered in the calculations of the 30 mrem/yr limit. The calculated data are listed in table I.

We are aware that this calculation may be subject to criticism, since various other modes of calculations are applied elsewhere. Therefore our proposal should only be considered as another attempt to obtain a better understanding of the true problems and as a basis for the succeeding considerations. It should not serve as a foundation for national regulations.

As may be deduced from our calculations, decontamination factors of 100 are sufficient for ^{3}H and ^{85}Kr. Applying state-of-the-art technology, they are realizable. Much higher decontamination factors than the presently achievable ones are required for iodine retainment in FBR fuel element reprocessing. No reasonable possibility exists for a separation of ^{14}C from the burner off-gas in HTR fuel reprocessing. Fortunately, the amounts of ^{14}C emitted to the atmosphere can be tolerated, if appropriate safety measures are applied.

The ^{14}C-data given in table I agree very well with recent calculations of BONKA, SCHWARZ, WIBBE and BÖHNERT [3] for its emission and resulting total exposure rate. As it can be seen from our calculations, only the ingestion dose is of importance. The value of 20 mrem/yr is based upon the most pessimistic assumptions; that means, the people exposed to this dose are permanently living in the critical place around the plant and feed themselves only with vegetables and water raised at the same spot.

3. Decontamination of process off-gases

A variety of separation processes are under development in several laboratories for the separation and retainment of noble gases. Good results have been reported for the FREON process [4], a cryogenic process [5] and the AKUT process [6]. Filtering, adsorbing and scrubbing techniques have been successfully applied for the cleanup of reprocessing off-gas streams from aerosols, iodine and tritium [7, 8, 9].

The choice of the appropriate process scheme depends on the type of fuel element, fuel composition, their chemical treatment, foreign matter in the system, process temperature, dilution of the off-gas, etc. Since our own R&D-work is mainly concentrated on the HTR fuel cycle, most experimental results given in this paper for an effective reduction of radioactive gaseous emissions from reprocessing plants are related to this particular fuel type. However, the procedures described may be applied with minor adaptations to fuel elements from LWR's and FBR's as well. Technical details are given in the following two chapters.

3.1 Decontamination of HTR reprocessing off-gases

In the course of reprocessing spent HTR fuel elements essentially all of the gaseous radioactive nuclides are released during the head-end treatment, which includes

- crushing and burning the graphite matrix of the fuel elements with oxygen, and
- dissolving the resulting oxide ash of uranium, thorium and fission products in THOREX-reagent (12 $\underline{M}$ HNO_3, 0.1 $\underline{M}$ HF, 0.3 $\underline{M}$ $Al(NO_3)_3$).

Only insignificant amounts of volatile radioisotopes are observed from the feed adjustment and solvent extraction steps. However, volatilization of ruthenium and iodine, mainly in the form of aerosols and tritiated water, occurs during the solidification or vitrification treatments of the aqueous fission product waste solutions originating from solvent extraction. Treatment of these off-gases does not require new technology and shall therefore not be further examined in this paper. Aqueous scrubbers and/or different types of solid adsorbers yield satisfactory decontamination factors of about 1 000.

Burner off-gas

In burning the graphite HTR fuel elements, large amounts of carbon dioxide and carbon monoxide are formed with a ratio of about 6 : 1. This off-gas is heavily contaminated with the radionuclides already mentioned and therefore suitable procedures are needed for their effective cleanup. The AKUT-process presently under development in our laboratory is promising. A block diagram together with mass flow data are shown in figure 1. The process is characterized by the following steps:

- coarse graphite particles and fines are removed by a cyclone and/or sintered metal filters
- aerosols are deposited by an electrostatic separator
- final decontamination from particulates is achieved with HEPA filters
- the remaining gas stream, containing the radionuclides ^{3}H, ^{14}C, ^{85}Kr and $^{129+131}I$, is passed through a catalytic oxidizer to convert to carbon dioxide and then liquefied by compressing to approximately 70 atm at room temperature
- the liquid is rectified and a krypton-rich fraction withdrawn from the top of the column for a final storage
- a second rectification of the bottom fraction of the first column yields an enriched fraction of tritiated water and iodine, which are then separated and fixed on suitable adsorbers for final storage.

The pilot plant test facility presently in hot cell operation is designed to achieve a decontamination factor of approximately 500 for krypton and iodine and 100 for tritium. Xenon is not separated from carbon dioxide under the selected conditions, since its thermodynamic behaviour is quite similar to that of carbon dioxide. However, 150 days of cooling time provides a nearly complete decay of its comparatively short-lived radioisotopes.

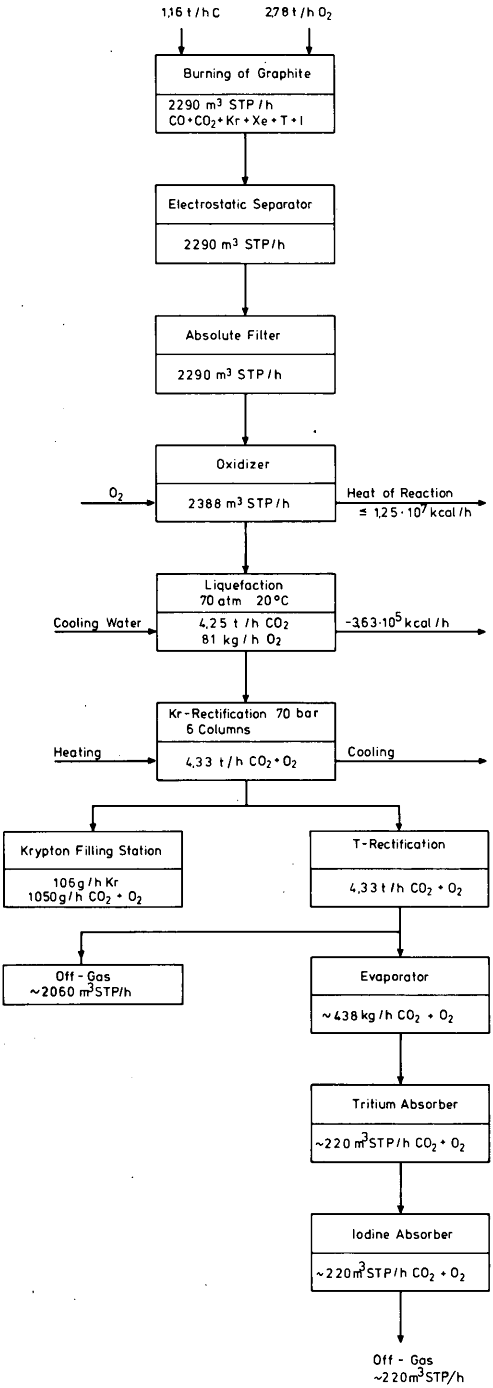

FIG. 1. Separation of krypton and tritium from burner off-gas by liquefaction and rectification.

The ^{14}C present in the fuel elements is discharged with the carbon dioxide off-gas, since a separation is not feasible.

Dissolver off-gas

Depending on the type of dissolver applied, e. g. continuous or batchwise operation [10], distinct off-gases are produced, each requiring a specific treatment.

A continuously operated dissolver yields an off-gas highly enriched with noble fission gases krypton and xenon. In order to prevent an unnecessary dilution by air, a helium purge gas cycle is recommended. The schematic flowsheet shown in figure 2 displays the required process steps:

- separation of the nitrous oxides and drying the gas stream with molecular sieves before entering the cryogenic unit
- deposition of xenon together with iodine in solid form at 82 °K and 1.6 bar
- purification of the contaminated xenon by rectification for possible further utilization
- deposition of krypton at 66 °K after compression to 6 bars
- collection of the krypton in steel cylinders for storage.

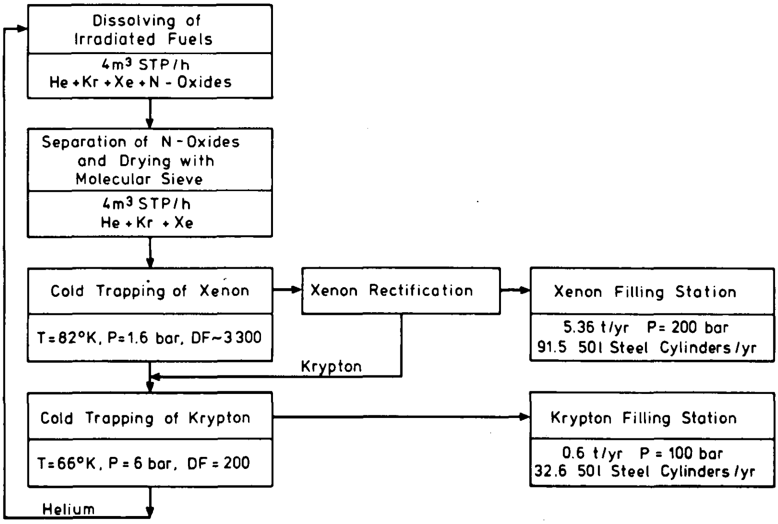

FIG.2. Separation of krypton and xenon from He purge gas cycle.

The purpose of the helium purge stream is to transport the noble gases, iodine and nitrous oxides from the dissolver to the gas treatment unit. The helium cycle is designed for a total throughput of 4 m^3 STP/h. Two deposition steps are required for the achievement of a coarse xenon/krypton separation. The helium gas is recycled.

Batchwise dissolving gives rise to air diluted off-gas, requiring a somewhat different treatment. The IDAHO cryogenic process flowsheet, modified at KFA, is shown in figure 3 and consists of the following steps:

- separation of nitrous oxides and the oxygen from air by reaction with hydrogen, thus forming elementary nitrogen and water, respectively
- drying the off-gas stream with molecular sieves
- low temperature deposition of xenon in a cryogenic unit, xenon rectification in a second unit for purification and possible utilization
- liquefaction of the remaining gas
- enrichment of krypton by rectification
- withdrawal of krypton at the bottom of the still and storage in steel tanks.

About 4 m^3 STP/h of air leaks into our batch dissolver.

3.2 Decontamination of LWR and FBR reprocessing off-gases

The favoured technology for reprocessing of LWR and FBR fuel elements is the "chop-leach" head-end followed by a separation of uranium, plutonium and fission products using solvent extraction. The fuel assembly is chopped or sheared mechanically. During this step only small amounts of krypton (approximately 1 % of the krypton inventory) are liberated from LWR fuels. This relatively low radioactivity may be discharged directly to the atmosphere.

From FBR fuel elements however, up to 90 % of the krypton is accumulated in the gas gap of the fuel pin. This gas should be withdrawn by suction in a suitable manner prior to chopping to avoid its dilution or loss.

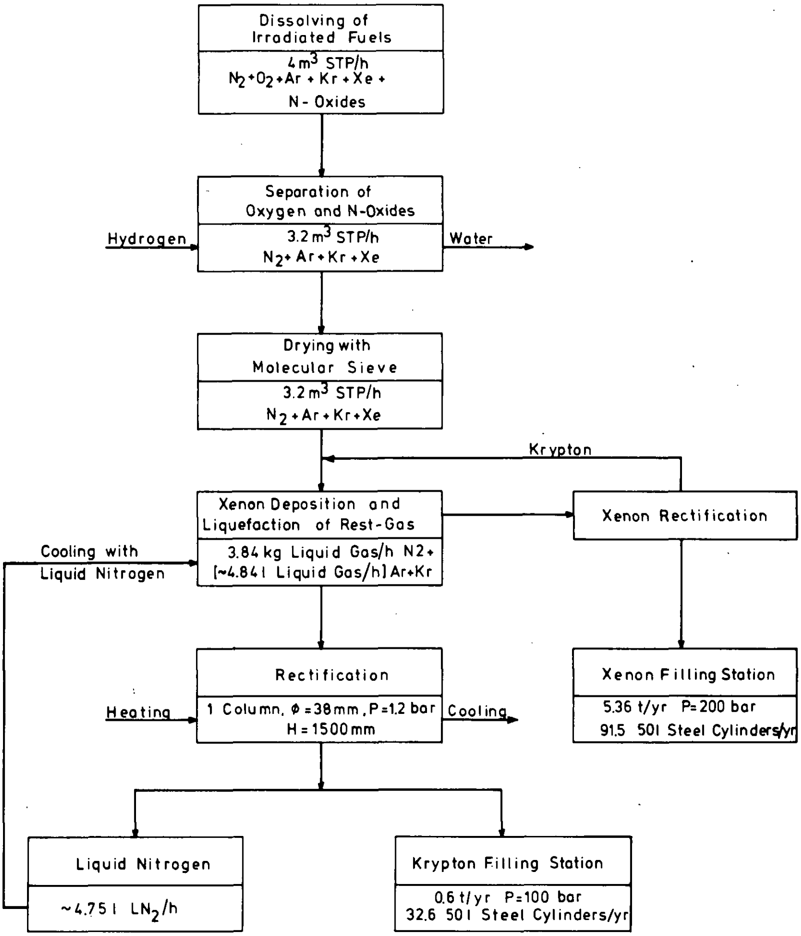

FIG.3. Separation of krypton and xenon from air-diluted dissolver off-gas.

To collect the gaseous and volatile fission products retained in the matrix of the oxidic fuel, the so-called VOLOXIDATION process is under development [11]. The chopped or sheared fuel elements containing $(U,Pu)O_2$ are oxidized with oxygen at temperatures between 450 and 600 °C. If under these conditions, the crystal lattice is transformed from a M_4O_9 to a M_3O_8 phase, the confined gas atoms are released.

Experimental measurements with FBR fuels indicate a more than 99.9 % release of tritium but only a 30 to 60 % release of krypton, applying this treatment. Similar results have been

TABLE II. PERCENTAGE RELEASE RATES OF GASEOUS RADIONUCLIDES IN VARIOUS STEPS OF REPROCESSING LWR, HTR AND FBR FUEL ELEMENTS

Reprocessing of	Process	Radionuclides	Amounts released to the off-gas stream during				Amounts found in aqueous phase
			Chopping [%]	Grinding [%]	Burning [%]	Dissolving [%]	[%]
LWR-fuel	PUREX	iodine	~ 1			90	< 5
		noble gases	1 - 5			95 - 99	0
		tritium	1 - 10			1 - 2	88 - 98
HTR-carbide type fuel	THOREX	iodine		< 1	30 - 50	40 - 60	< 10
		noble gases		< 1	95 ± 5	< 10	0
		tritium		< 1	> 97	< 1	< 2
HTR-oxide type fuel	THOREX	iodine		< 1	3 - 5	75 - 85	< 10
		noble gases		< 1	7 ± 5	~ 90	0
		tritium		< 5	10 - 20	< 2	70 - 85
FBR-fuel	PUREX	iodine	< 10			85	5
		noble gases	50 - 90			10 - 50	0
		tritium*	~ 5			~ 1	5 - 10

* about 90 % is released to the reactor through the stainless steel walls of the pins

obtained for LWR fuels, where more than 99 % tritium and up to 12 % krypton release have been measured. These results show that tritium volatilization is high enough, krypton however is retained to a relatively high percentage. A separation of krypton from dissolver off-gas presumably cannot be avoided.

A very serious problem arises, if short cooled FBR fuel elements have to be reprocessed, due to the very high ^{131}I radioactivity. Decontamination factors of 5 000 or higher are required. In the case of only 30 days cooling time, decontamination factors as high as 10^7 may even be necessary. No routine process is known today which can meet these high requirements. Much laboratory work in this field is done however, for instance at the Oak Ridge National Laboratory [11].

Table II represents a survey of percentage release rates for gaseous radionuclides in the various steps of reprocessing of fuel elements from LWR's, HTR's and FBR's. For completeness, also the amounts of radioactivity remaining in the aqueous processing streams are also included in the table. Particularly large amounts of tritium may be transformed to water and then be incorporated into the aqueous solutions. Since no post-separation of ordinary and tritiated water is economically feasible, it is most advantageous to have the tritium liberated in a gaseous state during the prime head-end treatment, in order to separate it in a concentrated form prior to an exchange or mixing with the aqueous phase.

4. Final storage of gaseous and volatile radioisotopes

Iodine and other volatile radioisotopes adsorbed on suitable solid adsorber materials are further treated for final storage as medium activity waste by incorporation in bitumen or concrete blocks, which then are safely deposited in salt caverns.

Tritium, in the form of water adsorbed on molecular sieves, poses some problems in long-term storage due to the fact that a slow evaporation of tritiated water occurs as a consequence of the existing solid/gaseous equilibrium at a given temperature. Low temperature storage suppresses this effect. Therefore storage of gastight containers in glacier or polar ice may be beneficial.

Radioactive noble gases are collected in steel tanks. Standard 50 litre cylinders can be filled up to pressures of 100 bars in the case of krypton and 200 bars in the case of xenon. For example, approximately 30 tanks for krypton and 90 for xenon per year are needed to serve a 50 000 MWe HTR reprocessing plant. The ^{85}Kr radioactivity per 50 litres tank amounts to about $5 \cdot 10^5$ Ci.

Engineered storage facilities may be used for about 150 years for long-term tank storage. A potential risk one is faced with, is a sudden accidental release.

We have proposed another potential method for final ^{85}Kr disposal, namely dumping the filled steel cylinders into the deep sea [6]. If the steel tanks are equipped with special valves which open at a predetermined depth, for instance below 2 000 m, the cylinders are partially filled with sea water forming crystallized noble gas hydrates under the existing high pressure. The same effect is achieved, if one waits until corrosion destroys the steel cylinders. Choosing a driftless deep sea area, the spreading of radioactivity is controlled by a comparatively slow diffusion process. Calculations have revealed that doses fall below dangerous concentrations at a distance of only 30 to 50m from the source. Additional safety is granted due to the chemical inertness of krypton. No assimilation or enrichment occurs in animals or plants. ^{85}Kr decays to stable ^{85}Rb, a harmless nuclide.

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DISCUSSION

W.O. SCHIKARSKI: What dose calculations did you apply in respect of ^{85}Kr? If you calculated the submersion dose, which represents only a theoretical approach, you will have got much more pessimistic values than would be yielded by consideration of the (realistic) skin dose.

Secondly, did you estimate the release of transuranium isotopes during reprocessing of the fuels for LWRs, HTRs and FBRs?

J. BOHNENSTINGL: For the calculation of ^{85}Kr dose rates the formula given by Vogt (JÜL-637-ST) were used, and Dr. Vogt will confirm, I believe, that the data concerned are skin dose values.

K.-J. VOGT: That is so.

J. BOHNENSTINGL: Regarding release of transuranium elements, we are at present performing calculations on the basis of some results from hot cell experiments. However, since they are not yet complete, these calculations do not figure in the paper.

A. BAYER: In what chemical form is iodine released from reprocessing plants?

M. LASER: This is rather a comprehensive question! In the case of HTR fuel elements, the iodine released appears to be mostly in elementary form. In the case of LWR fuel elements, work has been done by other authors on the form of the iodine liberated during reprocessing, and the subject is not in fact covered in our paper.

A. MERKEL: What was the source of the basic data you used to calculate FBR tritium inventory?

M. LASER: The data are based on an ORNL report on siting of reprocessing plants.

E. de RADEMAEKER: Do you feel entirely confident about the safety of storing gaseous and volatile radioisotopes (after solidification) in salt caverns? There has lately been a trend in the United States of America in favour of engineered storage rather than storage in salt formations.

M. LASER: Yes, I am quite confident. I understand from our experts on the subject that Germany is in a somewhat better position than the USA as far as safety of storage in salt formations is concerned. At Lyons Mine there are several unknown boreholes, so that water can penetrate in an uncontrolled manner. At the Asse salt mine, on the other hand, all the boreholes are known and insulated against water.

K. KREUZER: You reckon on a comparatively slow diffusion process in connection with the dumping of ^{85}Kr into the deep sea. However, there are plans in the offing for stirring up seawater in order to intensify nutritional protein production from the ocean. Might not this affect the speed of the diffusion process?

C.A. MAWSON (Chairman): Sympathizing with the authors of the paper if they are not certain how to reply to this question, may I offer a few remarks myself — without of course getting involved in a debate on deep-sea disposal as such.

Calculation of the amount of radionuclides that could safely be discharged into the deep sea gives a figure so astronomically large that the protagonists of ocean disposal feel that nobody will believe them — and hence recommend no disposal in the deep sea! In presenting the paper Mr. Bohnenstingl has said that the disposal of ^{85}Kr in cylinders in the deep ocean would have no effect on the seawater beyond 30 to 50 m from the point of disposal. He is faced with the same philosophical dilemma. The ocean is so big that people are loth to believe its capacity for waste absorption.

RELATIVE RADIOLOGICAL IMPORTANCE OF ENVIRONMENTALLY RELEASED TRITIUM AND KRYPTON-85[†]

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Presented by S.V. Kaye

Abstract

RELATIVE RADIOLOGICAL IMPORTANCE OF ENVIRONMENTALLY RELEASED TRITIUM AND KRYPTON-85.

Tritium (HTO) and ^{85}Kr are expected to continue as prominent components of the radioactive effluents from nuclear facilities. Simple comparison of air concentrations of HTO and ^{85}Kr with their respective maximum permissible concentrations does not provide a good indication of their relative radiological importance. Detailed considerations for each radionuclide emphasize differences which influence radiotoxicity. Dose estimates are presented, and the associated risks are compared for equal radionuclide concentration in air: somatic risk (HTO ~10 × ^{85}Kr) and genetic risk (HTO ~ 50 × ^{85}Kr). For populations in close proximity to the facility, ^{3}H will probably be the most significant dose contributor if nearly equal quantities of the two radionuclides are released. When projected radionuclide production and release figures are considered, the relative radiological impacts appear to be more nearly equal on a local basis for some facilities. However, in the latter case it is predicted that ^{85}Kr will contribute the greater dose to the global population.

1.0 INTRODUCTION

Tritium and ^{85}Kr are prominent in the radioactive effluent from a nuclear facility, because they are produced in large quantities and they have radioactive half-lives of moderate length (12.3 and 10.8 years, respectively). Based on the projected nuclear power economy in the United States and the free world, it has been estimated that by the year 2000 the annual production rates for ^{3}H and ^{85}Kr will be 14.6 and 520 MCi, respectively [1]. Estimated accumulations by that date are 96 MCi of ^{3}H and 3,150 MCi of ^{85}Kr [1]. Although in principle it is possible to design a nuclear facility to attain any desired low emission rate, it has not been practical to concentrate and retain the radionuclides. Continued release of ^{3}H and ^{85}Kr by the nuclear industry is anticipated [2]. The relative radiological significance of these radionuclides should be a consideration in decisions concerning capital outlays to construct new radwaste cleanup systems or to modify existing ones. Each nuclear facility has a responsibility to limit individual and population doses[1] resulting from its operation. Characteristics relevant to radiological assessment of environmental releases of ^{3}H and ^{85}Kr are discussed in detail in this paper, and estimated radiological insults to man are compared for equal concentrations of the two radionuclides.

[†] Research sponsored by the US Atomic Energy Commission under contract with the Union Carbide Corporation.

[1] The use of "dose" throughout this paper is actually "dose equivalent."

2.0 TRITIUM

2.1 Radiotoxicity

Tritium released to the environment is in the form of tritiated water (HTO) primarily, or it is soon oxidized to that form. As HTO it behaves as ordinary water throughout the hydrosphere. This is the most hazardous form for environmental ^{3}H, because it is metabolized like ordinary water and readily enters the body through the gastrointestinal tract, the lungs, and the skin. The International Commission on Radiological Protection (ICRP) has recommended for occupational workers exposed to HTO a maximum permissible body burden (MPBB) of 2000 μCi [3]. In other words, that quantity of HTO in a 70-kg reference man yields a dose rate to the whole body of 5 rems/year. The 2000-μCi MPBB, one of the largest MPBB values recommended for any radionuclide, is based on the fact that ^{3}H is a pure emitter with a low effective absorbed energy (0.006 MeV per disintegration) [4] and that as HTO it is distributed uniformly throughout the body water pool where it has a short effective half-time (10 days) [5]. A fraction of the ^{3}H taken in as HTO is known to become organically bound. The extent of such binding and its effect on ^{3}H retention will be discussed in the section giving dose estimates for ^{3}H.

2.2 Radionuclide Specific Characteristics

2.2.1 Transmutation and Quality Factor

Assuming that one can estimate the quantity of ^{3}H deposited in the reference tissue, assessment of the toxicity is complicated by questions of possible transmutation effects and of intrinsic biological effectiveness of the weak beta emissions (0.018 MeV maximum). When a ^{3}H atom undergoes radioactive decay, the molecular structure of which it is a part suddenly contains a helium atom where a hydrogen atom formerly existed. In his recent review of the radiobiology of ^{3}H, Thompson [6] concluded on the basis of cellular level studies and animal studies that transmutation effects do not seem to add significantly to the ionizing radiation effects. To adjust for the varying biological effectiveness of different irradiation conditions the ICRP recommends that a Quality Factor (QF) be used to account for differences in linear energy transfer (LET) [7]. The LET for a low-energy ^{3}H beta is higher than that for a more energetic beta, gamma, or x-irradiation. The ICRP at one time recommended a QF of 1.7 for a beta radiation having a maximum energy <0.03 MeV [7]. Some experimental evidence appeared to support the QF of 1.7 for ^{3}H, but questions of experimental endpoint and choice of reference radiation source tended to make interpretation difficult. Bond [8] evaluated the potential hazards from HTO and concluded from reviews of pertinent biological material that there appears to be little convincing evidence that the QF is significantly different from unity. The ICRP now recommends that a QF of 1.0 be used for all beta, gamma, and x-irradiation [4].

2.2.2 Bioaccumulation

The possibility of environmental ^{3}H concentrating in a particular organism, in dietary items, or in a reference tissue of man is always a concern. Although most chemical reactions discriminate against the incorporation of ^{3}H in favor of hydrogen [9], the possibility of some concentration of ^{3}H cannot be ruled out completely. For example, Bond [8] has suggested that discrimination against ^{3}H "leaving" a biochemical compound once it is incorporated might be greater than that for its "entering" the compound. That phenomenon would lead to concentration or "trapping" of ^{3}H in the compound in question. In reviewing ecological aspects of environmental ^{3}H behavior, Elwood [10] found that the isotopic effects of ^{3}H involved in most exchange reactions appear to be negligible. He states that "data from field studies in both acutely and chronically exposed ecosystems seem to support the idea that there is no concentration above that of environmental concentrations in food or water at any level of a food chain, although complex aquatic food chains and food chains in temperate climates have yet to be studied in detail." In fact, according to Elwood, "in most ecosystems, tritium concentrations would tend to become diluted at various stages of a food chain or in compartments of a coupled system."

2.2.3 Organic Binding

The human body is approximately 60% water and 10% hydrogen [3], thus two-thirds of the hydrogen in the body is present as water and one-third is present as organic hydrogen. Following the entry of HTO into the body, a fraction of the ^{3}H exchanges with hydrogen bound in organic molecules. One-third of the organically bound hydrogen in tissue is considered exchangeable, with that fraction decreasing as the fat content of the tissue increases [11]. Tritium present as HTO also can be incorporated by metabolic processes into nonexchangeable positions in tissue constituents. The rate and extent of incorporation of ^{3}H from HTO as tissue-bound ^{3}H is dependent on the metabolic activity of the tissue. Timing and duration of the exposure relative to the metabolic activity is also important. If the HTO is available for incorporation during formation and growth of a tissue (high metabolic rate), the probability of extensive organic labeling is enhanced. On this basis, the highest concentrations of organically bound ^{3}H are expected in those tissues and population segments which are in formative and growth stages at the time of an acute exposure. As the exposure duration increases to the chronic case, ^{3}H concentrations approach equilibrium values with a single tritium-to-hydrogen ratio common to all parts of the hydrogen pool. Organic binding would not be expected to cause significant bioaccumulation of ^{3}H from HTO, because water is approximately as hydrogen rich ($\sim$11% by weight) as any compound ever becomes (recall that the human body is $\sim$10% hydrogen). Another point is the influence of exposure pathway on the extent of organic binding. Experiments with rats chronically exposed to tritiated drinking water at a constant concentration have shown that the organically bound ^{3}H in tissue solids did not exceed 35% of the concentration maintained in the intake water [12]. The implication is that two-thirds of the organically bound hydrogen comes from organically bound hydrogen in food. Exposure pathway can be an important consideration in attempting to estimate the extent of organic binding.

Tritium loss from organic molecules, like its incorporation, is dependent on the metabolic activity. Thus mechanisms tending to retard ^{3}H incorporation also tend to retard its release. Tritium which becomes organically bound has a longer effective half-time, but it represents a small fraction of the total ^{3}H intake [11]. The role which organic binding plays in determining the radiological significance of HTO exposure is discussed in the following section.

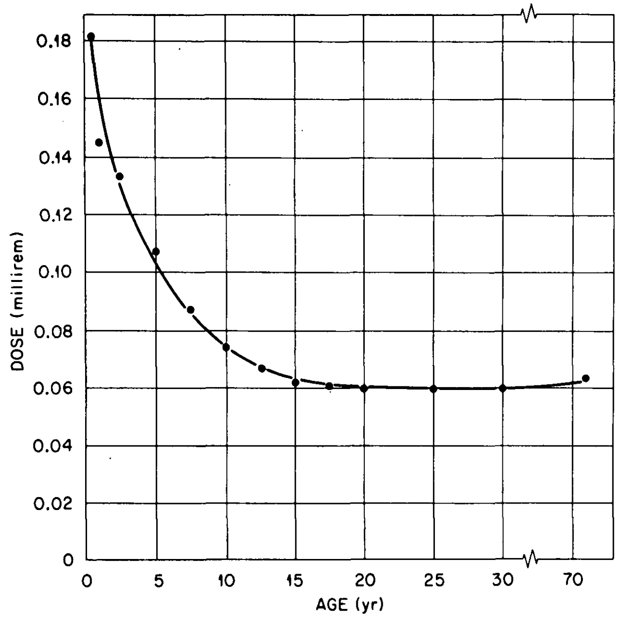

FIG.1. Dose to the whole body per microcurie intake of HTO as a function of age.

2.3 Dose Estimation

Dose estimates for ^3H exposures customarily consider only that ^3H entering the body water pool. Tritium is not an external exposure hazard, because the only radiation emission is the low-energy beta particle (0.018 MeV maximum) with a maximum range in water or soft tissue of only 0.005 mm (0.55 mg/cm^2) [13]. Recent skin measurements suggest 4 mg/cm^2 for average epidermal thickness [14], with the radiosensitive layer being below that depth. The radiation dose received following exposure to ^3H is therefore primarily internal, and the major portion of this dose is attributable to the ^3H present as HTO in the body water pool. Figure 1 presents an estimate of dose for a 1-µCi intake of HTO, with the dose presented as a function of the age of the individual exposed. Whenever a population is exposed it is desirable to be able to estimate the amount of variation in dose among subgroups, age groups in this example, and to identify the group receiving the highest dose (the critical group). The ^3H dose model from ICRP Publ. 10 was used to prepare Fig. 1, with the additional assumptions that the QF for ^3H betas is 1.0 and that the total body mass is the reference tissue. The estimated dose per microcurie of intake for an infant is approximately three times that for an adult. The curve in Fig. 1 reflects a range of effective half-times for HTO in the body water pool from 3.3 days for an infant to 10 days for an adult, and a range in body weights from 8 to 71 kg [15]. It is important to recognize that other factors such as ambient temperature, personal habits, and diet may alter the picture shown in Fig. 1. For example, an estimate of daily water intake as a function of age is shown in Fig. 2. In this case there is approximately a factor of 3 operating in the reverse direction,

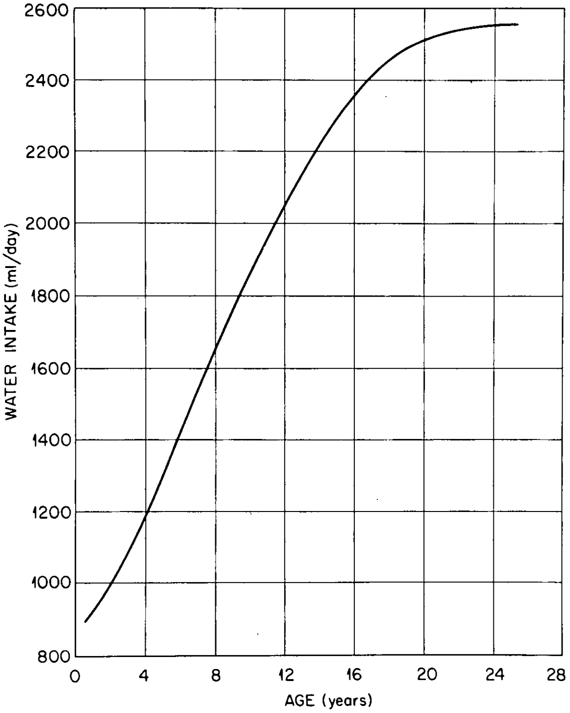

FIG.2. Daily water intake as a function of age.

thus if the exposure pathway is via water ingestion, the dose differential among age groups would be negligible. A similar situation exists if the primary exposure pathway is inhalation of HTO vapor [15]. If other exposure pathways are of primary importance, only a situation-specific evaluation can verify the absence of a critical population age group for HTO exposure. However, the expected ^3H release from nuclear facilities will be continuous. Since the ^3H will be present in the environment as HTO, thereby available to be ingested or inhaled in that form, little variation in radiation dose among population age groups is anticipated. Accidental or short-term releases of HTO in greater than usual quantities will increase the probability of there being critical groups within the exposed population which may receive a greater radiation dose because of one or more unique exposure pathways connecting them with the radioactivity source.

Returning to organic binding and its possible importance, the data of Snyder et al. [16], presented in Fig. 3, illustrate the excretion of ^3H by man following an acute exposure. Two components are clearly evident in the excretion curve with the hint of a third longer term component. The initial component was assigned a half-time of 8.7 days and the second a value of 34 days. Later data covering longer follow-up periods for other exposed individuals have verified the existence of the third component with a half-time in the range 300 to 600 days [17,18]. There is a tendency to ascribe the components of the excretion curve to various hydrogen pools in the body even though the compartments are chemical in nature rather than identifiable with specific organs or tissues.

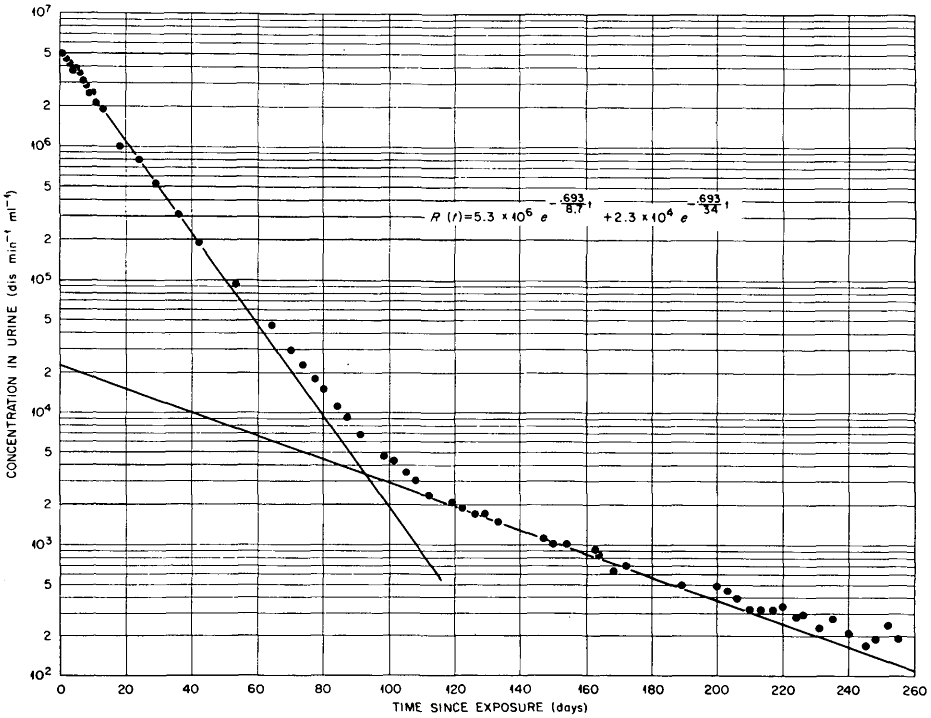

FIG.3. Tritium concentration in urine.

Typical suggestions are: 8.7 days--body water; 34 days--exchangeable ^{3}H in organic materials; and 300 to 600 days--nonexchangeable ^{3}H in organic materials, eventually freed by catabolic processes. The existence of permanently fixed hydrogen pools has also been suggested [19]. The probability of a very slowly replaced or a fixed hydrogen pool becoming heavily tritiated at the time of its formation due to a transient surge of environmental ^{3}H at a concentration significantly elevated above ambient levels is small. Thompson [6] has pointed out that exposures resulting as a consequence of such non-steady-state conditions might increase the dose to an individual, but would not increase the total population dose. Snyder et al. [16] estimated that the second component of their retention function increased the whole-body dose estimated for the first component by 1.7%. On the basis of their own data Sanders and Reinig [17] reported that bound ^{3}H could augment the dose they estimated for body water ^{3}H by up to 25%. Similar dose estimates by Bennett [20] for acute intake of ^{3}H indicate that 84% of the tissue dose is due to ^{3}H in body water and 16% is due to organically bound ^{3}H. A theoretical model for ^{3}H behavior in man was recently described by Croach [21]; the model is based on the assumption that ^{3}H enters the body in a manner typical for hydrogen and that once assimilated it behaves the same as ordinary hydrogen. Results obtained by Croach with that model suggest that very little ^{3}H from an acute exposure can become permanently bound, and even if that which became bound stays there until it decays, not enough can enter to make a significant contribution to total dose. With regard to the chronic exposure situation, Bond [8] has noted that since two-thirds of the body hydrogen is in body water, then if one assumes equilibrium with uniform labeling of all organic materials at the level existing in the body water, the body burden of ^3H, and hence the dose rate to the total body, can be increased no more than 50% over that which exists in the absence of organic binding. Based on his theoretical model, Croach states that "the energy deposition from permanently bound ^3H cannot increase the energy deposition (or whole-body dose) in the steady-state system with no permanently bound ^3H by more than ∿10%" [21]. Since dose estimates in Fig. 1 include the body water ^3H contribution only, it would be prudent to increase values taken from this figure by approximately 20% to include the dose contribution due to organically bound ^3H.

3.0 KRYPTON-85

3.1 Radiotoxicity

Krypton-85 is a noble gas and when it is released to the environment it tends to become distributed almost homogeneously over the surface of the globe and throughout the troposphere [1]. Environmental ^{85}Kr will accumulate in the atmosphere with little depletion by the biosphere. Thus the inventory will be determined by the release rate and the radioactive half-life of the radionuclide. Depletion of the atmospheric inventory of ^{85}Kr by washout, dry deposition, and deposition through adsorption on particles also will be negligible [22]. Krypton-85 generally is classified as a radionuclide of low radiotoxicity for a number of reasons: (1) it is primarily a beta emitter, β_1 - 0.672 MeV maximum with 0.996 abundance, β_2 - 0.16 MeV maximum with 0.004 abundance, and γ_1 - 0.514 MeV with 0.004 abundance; (2) it is poorly absorbed in the body, the blood/gas partition coefficient is 0.05 approximately, and the tissue/blood partition coefficient is 1 for all tissue except fat, for which it is 10 [23]; and (3) the principal radiation dose is delivered to the external body surface (skin), a tissue which is not particularly radiosensitive.

3.2 Dose Estimation

Unlike ^3H, ^{85}Kr dosimetry is not complicated by uncertainties concerning bioaccumulation, organic binding, and the quality factor. Dose estimates calculated from the work of Kirk [24] are presented in Table I. The data presented in Table I are for continuous exposure in an infinite cloud having a ^{85}Kr concentration of 1 pCi/cm^3, and the external dose calculations are for 50% of the point dose at the center of the cloud. The values in Table I illustrate the relative contribution of each ^{85}Kr emission to the total dose estimated for the respective reference tissues. The dose estimates in this table are in good agreement with those found in other detailed treatments of ^{85}Kr dosimetry [25]. As one would expect, the estimated dose is almost entirely due to external exposure; ∿95% of the whole body and gonad doses is due to external gamma and bremsstrahlung, and over 99% of the skin dose is due to external beta. The beta skin dose value given in Table I is applicable to the skin surface, with the more important dose being that received at the depth of the shallowest layer of live skin. This depth generally has been assumed to be 0.07 mm, and it is estimated that the beta dose at that depth is approximately 50% of the surface dose [26]. Recent measurements using a new

TABLE I. DOSE ESTIMATES FOR ^{85}Kr ASSUMING CONTINUOUS EXPOSURE IN AN INFINITE CLOUD (1 pCi/cm^3)

Origin	Dose (millirem/year)		
	Skin	Whole Body	Gonads
Gamma and Bremsstrahlung			
External	16.4	13.4	16.4
Internal		0.002	0.0007
Beta			
External	2076		
Internal		0.76	0.28
	2093	14.2	16.7

technique [14] suggest that a more reasonable value for radiological protection purposes is 0.04 mm, in which case the depth dose is estimated to be 70% of the surface dose [26].

4.0 RADIOLOGICAL COMPARISONS

4.1 For Unit Concentrations

4.1.1 Dose

Some confusion over the relative dose contributions of these two radionuclides may arise from a consideration of the maximum permissible concentrations in air (MPC$_a$) for unrestricted areas appearing in 10CFR20 [27]. The respective MPC$_a$'s are: 0.2 pCi/cm^3 for ^{3}H and 0.3 pCi/cm^3 for ^{85}Kr. It is generally erroneously assumed that these concentrations produce equivalent radiation doses to the individual, or doses of equal biological significance. This is not true for these two radionuclides. It has been stated [28] that future recommendations of the ICRP are expected to include an increase of the MPC$_a$ for ^{85}Kr by a factor of approximately 5. Although he does not suggest that such a revision of the MPC$_a$ is desirable, calculations by Kirk [24] illustrate that the MPC$_a$ for ^{85}Kr is conservative by at least a factor of 4.8, i.e., exposure at the MPC$_a$ results in doses which are less than one-fourth of the dose limit on which the MPC$_a$ is based. The preceding sections of this paper show that these two radionuclides are quite different in many characteristics which profoundly influence their respective radiological impacts. The effects of those differences are evident in the dose estimates presented in Table II. All of the dose estimates are for exposure to a unit concentration (1 pCi/cm^3 of air) of radioactivity. The ^{3}H values for whole body and gonads include dose contributions for inhalation and skin absorption, assuming HTO vapor absorption through the skin to be 75% of that via inhalation [31]. The values in Table II demonstrate the good agreement among dose estimates from various literature sources. The table bears out the fact that ^{3}H exposure results in essentially uniform irradiation of the whole body, while the skin dose is by far the largest one for ^{85}Kr. The whole-body dose for ^{85}Kr is approximately 1% of the skin dose.

TABLE II. ESTIMATED DOSE FOR ^{85}Kr AND HTO IN AIR AT A CONCENTRATION OF 1 pCi/cm^3

Exposure	Dose (millirems/year)	Reference
^{85}Kr at 1 pCi/cm^3		
Surface of skin or clothing	2093	Kirk [24]
	1690	Hendrickson [26]
	2080	Dunster and Warner [18]
	1664	Whitton [29]
	1830	Diethorn and Stockho [25]
Shallowest layer of live skin		
4 mm	1450^a	
7 mm	1040^b	
Whole body	14.2	Kirk [24]
	23.3	Hendrickson [26]
	14.1	Dunster and Warner [28]
	15.2	Diethorn and Stockho [25]
Gonads	16.7	Kirk [24]
	23.3	Hendrickson [26]
	17.3	Dunster and Warner [28]
	15.2	Diathorn and Stockho [25]
^{3}H (HTO) at 1 pCi/cm^3		
Surface of skin or clothing	∿0	Turner, Kaye, and Rohwer [30]
Whole body	960^c	
	1070^c	ICRP [4,5]
	1080^{c,d}	Bennett [20]
Gonads	960^c	
	1000^c	Osborne [31]

[a] Calculation based on the works of Kirk(24) Hendrickson(26), and Whitton(14).

[b] Calculation based on the works of Kirk(24) and Hendrickson(26).

[c] Intake via skin absorption assumed to be 75% of that due to inhalation(31).

[d] Calculated for acute intake only.

4.1.2 Risk

Considering somatic risk first, compare the 1,040- to 1,450-millirem skin dose for ^{85}Kr with the 960- to 1,080-millirem whole-body dose for ^{3}H. Although these dose estimates are of nearly equal magnitude, they are not of equal significance. The dose limit recommended for members of the public by the ICRP for skin (3 rems/year) is six times that recommended for the whole body (0.5 rem/year) [7]. If one accepts cancer induction as the radiological response and death as the endpoint, the reduced significance attached to skin irradiation is born out since 9 out of 10 skin cancers are curable [32]. Another intangible reduction exists since clothing provides protection against the low energy of the ^{85}Kr beta particle. It would appear that the somatic risk from ^{85}Kr is approximately one-tenth of the somatic risk from ^{3}H at equal radionuclide concentrations, a larger fraction than is obtained when whole-body estimates for the two radionuclides are compared.

The genetic risk from ^{85}Kr is estimated to be approximately one-fiftieth of that from ^{3}H at equal concentrations. In this case, the respective gonad dose estimates for the two radionuclides are compared. The genetic dose limit recommended by the ICRP for members of the public is 5 rems in 30 years [7].

4.2 For Local Populations

The comparisons in the preceding section (4.1) are useful in estimating the relative radiological impact of atmospheric releases on local populations when release rates for the two radionuclides are known. When a significant portion of the ^{3}H is released in liquid effluents rather than in gaseous effluents with the ^{85}Kr, exposure pathways other than inhalation and skin absorption may be the most important. In that case only site-specific evaluation is likely to provide an adequate estimate of the relative impacts of the released activities in terms of dose to the local population.

4.3 For Global Population

Released ^{3}H and ^{85}Kr each are freely dispersed in worldwide dilution pools: ^{3}H in the hydrosphere and ^{85}Kr in the atmosphere. Thus each is a potential source of irradiation for the world population. The United Nations Scientific Committee on the Effects of Atomic Radiation has provided information with which one can estimate doses to the world population for releases of various radionuclides, including ^{3}H and ^{85}Kr [33]. The estimated soft tissue dose (assumed to apply to whole body and gonads) to the world population for ^{3}H is from 0.38 to 1.6 μμrads/Ci released. For ^{85}Kr the estimated values are 0.067 μμrad/Ci for gonads, 0.053 μμrads/Ci for whole body, and 8.2 μμrads/Ci for skin. From these values, it is evident that on a unit basis ^{3}H dose to whole body and gonads is nearly an order of magnitude greater than the ^{85}Kr dose to those organs. Recalling the relative risks of whole-body and skin exposures as discussed in section 4.1.2, a unit release of ^{85}Kr yields an estimated skin dose which is approximately radiologically equivalent to the estimated whole-body dose from a unit release of ^{3}H. If we accept the estimate that annual ^{85}Kr production will be 35 times the ^{3}H production rate by the year 2000, and if equal fractions of each are released to the environment, it appears that of the two radionuclides ^{85}Kr will be the predominant contributor to world population dose so long as the nuclear power industry is the primary source of radioactivity release.

5.0 CONCLUSIONS

Tritium and ^{85}Kr probably will continue to be released to the environment from nuclear facilities. The two radionuclides each have unique environmental behavior and dosimetry characteristics which significantly influence radiological impact. The relative importance of the two radionuclides is dependent on the quantity of each released, the mode of release, the pathway of exposure, and the exposed population of interest. For populations located in close proximity to the nuclear facility and atmospheric releases of ^{3}H and ^{85}Kr of equal magnitude, it is estimated that the somatic insult from ^{3}H is 10 times that from ^{85}Kr and the genetic risk due to ^{3}H exceeds that of ^{85}Kr by a factor of 50. When a major portion of the ^{3}H is released directly to surface waters additional exposure pathways must be evaluated to determine the relative importance of ^{3}H and ^{85}Kr releases. When interest is shifted to the global population, ^{3}H and ^{85}Kr are of essentially equal radiological importance on a unit release basis. If one includes considerations for probable differences in production and release rates, ^{85}Kr becomes the more important of the two radionuclides in terms of dose to the global population. Placement of emphasis in designing radwaste treatment systems to remove ^{3}H or ^{85}Kr, or both, is therefore dependent on the identity of the primary population at risk as well as on our quest to reduce all environmental releases of radioactivity to the lowest practicable level.

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ESTIMATION OF RADIATION EXPOSURE ASSOCIATED WITH INERT GAS RADIONUCLIDES DISCHARGED TO THE ENVIRONMENT BY THE NUCLEAR POWER INDUSTRY

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Abstract

ESTIMATION OF RADIATION EXPOSURE ASSOCIATED WITH INERT GAS RADIONUCLIDES DISCHARGED TO THE ENVIRONMENT BY THE NUCLEAR POWER INDUSTRY.

Several fission product isotopes of krypton and xenon are formed during the operation of nuclear power stations, while other radioactive inert gases, notably isotopes of argon and nitrogen, are produced as neutron activation products. With the exception of ^{85}Kr these radionuclides are short-lived, and the containment and hold-up arrangements in different reactor systems influence the composition of the inert gas mixtures discharged to the environment. Cooling of irradiated fuel before chemical reprocessing reduces very substantially the amounts of the short-lived krypton and xenon isotopes available for discharge at reprocessing plants, but almost all the ^{85}Kr formed in the fuel is currently discharged to atmosphere from these plants.

Estimates are made of the radiation exposure of the public associated with these discharges to atmosphere taking into account the type of radiation emitted, radioactive half-life and the local, regional and world-wide populations concerned. Such estimates are often based on simple models in which activity is assumed to be distributed in a semi-infinite cloud. The model used in this assessment takes into account the finite cloud near the point of its discharge and its behaviour when dispersion in the atmosphere is affected by the presence of buildings. This is particularly important in the case of discharges from those reactors which do not have high stacks. The model also provides in detail for the continued world-wide circulation of the longer-lived ^{85}Kr.

1. INTRODUCTION

During irradiation of fuel at nuclear power stations, several fission product isotopes of the inert gases krypton and xenon are formed in the fuel. A large proportion of each of these radionuclides is retained in the fuel matrix; the small proportion which escapes through defects in the fuel cladding into the coolant contains relatively small amounts of the long-lived ^{85}Kr and much larger amounts of short-lived krypton and xenon isotopes. The composition and amount of fission-product inert gas activity discharged to atmosphere from reactor coolants can be controlled by means of delay systems. Storage of irradiated fuel for several months before reprocessing greatly reduces the amounts of short-lived krypton and xenon isotopes available for discharge at reprocessing plants; amounts of ^{85}Kr in fuel are not materially affected by this storage period and at present virtually all the ^{85}Kr formed in fuel is discharged to atmosphere from reprocessing plants. Other short-lived radioactive inert gases, principally ^{13}N, ^{16}N and ^{41}Ar, are produced by neutron activation of stable isotopes present in reactor coolant gases and air spaces within the reactor shielding. The composition and amount of activation-product inert-gas activity discharged depends upon reactor type and design.

To estimate radiation exposure of the public resulting from discharges of radioactive inert gases, it is necessary to consider three groups of people. These are local and regional groups, defined for present purposes as being within 6 km and 1000 km respectively of the discharge site, and the world-wide population. In a previous paper [1] the authors used this approach to estimate exposure of the public resulting from ^{85}Kr discharges from reprocessing plants. The model used to estimate exposure of the world-wide population is considered in greater detail in the present paper. Dose rates calculated using the revised model correspond, as before, to present United Kingdom and world-wide power programmes, and those predicted up to the end of the century. The model used previously to estimate exposure of local and regional groups is now applied to the other radioactive inert-gas nuclides discussed above. In view of the diversity in composition of discharges from reactors, emphasis is placed on the variation of dose rate with distance corresponding to unit discharge rate of individual nuclides. Examples are given of results for unit discharge rate of mixtures of krypton and xenon isotopes from water-cooled reactors and of ^{41}Ar from gas-cooled reactors.

Examination of decay schemes of the various nuclides concerned suggests that, in the majority of cases, irradiation of gonads by gamma rays is of greater importance from the public health point of view than irradiation of skin and surface tissues by beta rays. Beta exposure is important for local groups in the case of ^{85}Kr; the variation of skin dose rate with distance corresponding to unit discharge rate of ^{85}Kr is given in reference [1]. All the dose rates calculated by means of the models described in this paper refer to irradiation of gonads by gamma rays.

2. MODEL TO CALCULATE GONAD DOSE RATES FROM A FINITE CLOUD OF RADIOACTIVE INERT GASES

The simplest model for calculating the external gamma dose rate from a cloud of radioactive material assumes that the cloud is semi-infinite and of uniform concentration. This model overestimates dose rates when cloud dimensions are small compared with the mean free path of the gamma rays. In this case, a more accurate method is to integrate the flux density from a finite cloud with a defined concentration distribution and then convert the flux density to dose rate.

2.1. The concentration distribution within 1000 km of the discharge point

In order to define the concentration distribution within the finite cloud of radioactive gases, use is made of the model proposed by Pasquill [2] for the cloud resulting from a discharge of short duration. In this model, radioactive gas concentration distribution is assumed to be Gaussian both vertically and in the cross-wind direction; cloud dimensions are given for distances up to 100 km downwind in different conditions of atmospheric stability. This model has been extended to a uniform wind-direction distribution and a continuous discharge rate by replacing the Gaussian cross-wind distribution by a uniform distribution in a horizontal plane at a given distance. The concentration χ at the point of interest resulting from a continuous discharge rate in any given weather category is given by equation (1).

$$\chi = \frac{2.15Q}{(2\pi)^{3/2}suh}\left\{\exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right] + \exp\left[-\frac{(z+H)^2}{2\sigma_z^2}\right]\right\} \quad .... \qquad (1)$$

where Q = discharge rate

s = distance from the point of discharge to the point of interest in the cloud

u = mean wind speed for a given weather category

h = vertical spread from the cloud axis to the height at which the concentration is 1/10 of that on the axis

σ_z = standard deviation of the vertical Gaussian distribution (= $h/2.15$)

z = height above ground of the point for which the calculation is being made

H = effective height of the discharge point above ground.

The concentration at any point of interest in the cloud can be calculated from equation (1) for distances up to 100 km from the point of discharge. The model is extended to a distance of 1000 km by assuming that the upper edge of the cloud, defined by h, remains at a constant height above ground between 100 km and 1000 km, and that there is a steady transition from a Gaussian distribution to uniform distribution in the vertical plane over the same distance [3].

Modifying factors must be applied to the discharge rate, Q, to take account of radioactive decay of parent nuclides and the growth of daughters. Radioactive decay is accounted for by replacing Q by $Q \exp[-\lambda s/u]$, where λ is the decay constant of the nuclide concerned. Growth of any daughter product is accounted for by replacing Q by

$$\frac{Q_P \lambda_D}{\lambda_D - \lambda_P} \left\{ \exp\left[-\lambda_P\, s/u\right] - \exp\left[-\lambda_D\, s/u\right] \right\}$$

where Q_P = discharge rate of parent nuclide

λ_D and λ_P = decay constants for daughter and parent nuclides.

The daughter nuclide will only give a dose comparable to that of the parent if the half-life of the daughter is similar to or less than that of the parent. The decay schemes ^{88}Kr → ^{88}Rb, ^{89}Kr → ^{89}Rb, ^{133m}Xe → ^{133}Xe and ^{138}Xe → ^{138}Cs are of this type.

The concentration distribution defined by equation (1) applies essentially to discharges made into an undisturbed air flow in open country. In practice discharges are made in complex circumstances. One circumstance which deserves special consideration concerns stacks associated with reactor buildings; these stacks are rarely of sufficient height to prevent discharged radioactive gases from becoming entrained in the building wake and an estimate of the effect of this on subsequent dispersion is required. Examination of possible methods of accomplishing this suggests that a useful empirical method is that of defining a "virtual source" some distance upwind of the building. The distance from the building to the virtual source may be chosen such that the cross-sectional area of the virtual cloud at the building is equal to that of the building. The effective height of this virtual source is equal to half the building height [4]. Trial calculations show, however, that use of the virtual source method has little effect on results at distances greater than 1 km from the reactor. It is considered that entrainment in the building wake is adequately accounted for by assuming discharge from an effective height equal to half the actual height of the stack above ground. In contrast, reprocessing plants often have stacks which are sufficiently tall for discharges to be made into an undisturbed air flow; in this case the effective height is equal to the actual height.

2.2. Calculation of flux density and dose rate within 1000 km of the discharge point

Having defined the concentration at any point within the finite cloud, the gamma flux density at ground level resulting from this can now be calculated. The gamma flux density, F, at a distance r from a point source of q curies is given by equation (2).

$$F = 3.7 \times 10^{10} fq \frac{B(E,\mu r)\exp[-\mu r]}{4\pi r^2} \text{ photons/second.unit area} \quad .... \quad (2)$$

where μ = linear attenuation coefficient of the medium

$B(E,\mu r)$ = multiple scattering build-up factor (Berger's formula; data from Chilton [5])

f = branching ratio for the gamma decay

The flux density from a small volume δV of cloud having a concentration of radioactive material χ curies/unit volume is obtained by replacing q by $\chi\delta V$ in equation (2). The flux density, F_r, from the complete cloud is obtained by computer integration of equation (3) over all space.

$$F_r = \frac{3.7 \times 10^{10} f}{4\pi} \int \frac{B(E,\mu r)\exp[-\mu r]\chi}{r^2} dV \text{ photons/second. unit area} \quad .... \quad (3)$$

The gamma flux density at ground level at distances up to 1000 km from the point of discharge is calculated for each atmospheric stability category defined by Pasquill [2]. These values are weighted by the average frequency of occurrence in the United Kingdom of each stability category, as given by Bryant [6], to give annual average values for the gamma flux density.

The final step is to calculate the gonad dose rate corresponding to the gamma flux density. For this purpose it is assumed that dose rate to the body surface applies to the gonads, although it is strictly relevant only to male gonads [7]. The dose rate to the surface of the body may be calculated from the gamma flux density from the whole cloud if this is represented by four equal, horizontal, collimated beams incident on the front, back and both sides of the body [7]. This calculation has been carried out using Monte Carlo techniques for only a small number of gamma energies [8]. Interpolation between these energies is made by comparison with detailed calculations of dose rate per unit flux density to a small element of tissue-like material (water), which are available for a large number of gamma energies [9]. This comparison is justified because the ratio between the dose rates to a small tissue element and the body surface is almost constant over the total energy range considered.

2.3. Calculation of dose rates from a discharge of ^{16}N

Since the half-life of ^{16}N is only 7.2 s this nuclide is treated as a special case. The gas travels only 35 m during one half-life in the highest wind speed considered. For distances from the discharge point corresponding to several half-lives, the local concentration is very small, with most of the gamma flux density at these distances originating near the point of discharge. Thus, for distances greater than a few hundred metres from the discharge point, a cloud of ^{16}N can be treated as a point source and the dose rate calculated using equation (2). A steady discharge rate of R curies/second produces an inventory R/λ curies around the discharge point after a time long compared with the half-life. Thus, for a steady discharge rate of 1 Ci/s of ^{16}N, an inventory of 10.2 Ci would be maintained.

3. MODEL TO CALCULATE GONAD DOSE RATES RESULTING FROM CONTINUING DISPERSION OF RADIOACTIVE INERT GASES IN THE TROPOSPHERE

The concentration distribution in the finite cloud described in section 2.1 is applicable for the first few days after discharge while the radioactive gases travel over the first 1000 km. Constituent nuclides with half-lives longer than a few days continue to disperse more generally throughout the troposphere.

3.1. Dispersion in the northern hemisphere

Since most industrial regions are in the northern temperate zone, it is assumed that all radioactive gas discharges from sites of the nuclear power industry are made within this zone. The subsequent dispersion pattern of these gases in the northern hemisphere is based on the simple model proposed by Lal and Rama from data on global dispersion of man-made tritium, ^{14}C and ^{90}Sr [10]. This nuclear weapons test activity was initially introduced into the stratosphere at 50-75°N during late 1962 and appeared in the northern high latitude troposphere during mid-1963. The model incorporates two cells in the northern hemispheric troposphere because the evidence shows that the region around 30°N provides resistance to meridional exchange for the greater part of the year. Taking into account the seasonal variation in the exchange constant between the 30-90°N and 0-30°N cells, Lal and Rama estimated a mean residence time (reciprocal of the exchange constant) in each cell of about 3 months. Although there is some evidence that air in the 30-90°N cell is internally well-mixed [10], the model used in the present assessment assumes conservatively that the mean residence time of about 3 months applies to the latitude band where discharges are assumed to be made (35-60°N).

Measurements of the vertical distribution of radon in the atmosphere suggest that there is thorough and rapid mixing in the troposphere up to a height of about 10 km; there is very little radon at higher altitudes [11]. The model, therefore, incorporates uniform distribution of radioactive gas from the nuclear power industry per unit mass of air up to a height of 10 km in the latitude band and subsequently in the northern hemisphere as a whole. The assumption of uniform concentration is not applicable to nuclides such as ^{131m}Xe, ^{133m}Xe and ^{133}Xe which have short half-lives compared with the mean residence time in the latitude band. The contribution to dose from these nuclides in this region has not been estimated.

3.2. Transfer to the southern hemisphere

Transfer of tropospheric air between the two hemispheres is inhibited by wind patterns in equatorial regions. Exchange constants based on measured concentrations of man-made ^{14}C, ^{90}Sr, CO_2 and tritiated methane in the two hemispheres are low compared with exchange constants within individual hemispheres (section 3.1). Values for the mean residence time vary from 0.9 to 4 years with a value of about 2 years for average conditions [12]; a shorter mean residence time of about 7 months applies to the mixing of air between the tropics of the two hemispheres [10].

An exchange constant of 0.5 per year is used in the present model as a representative value. This behaviour is described mathematically in equations (4) and (5) for the case of transfer of radioactive inert gas from the northern hemisphere, where it is assumed to originate, to the southern.

$$\frac{d\ I_N(t)}{dt} = P(t) - \alpha(I_N(t) - I_S(t)) - \lambda I_N(t) \quad \ldots\ldots \quad (4)$$

$$\frac{d\ I_S(t)}{dt} = \alpha(I_N(t) - I_S(t)) - \lambda I_S(t) \quad \ldots\ldots \quad (5)$$

where I_N, I_S = inventories in the northern and southern hemisphere

P = discharge rate of a radionuclide

α = exchange constant across the equator

λ = radioactive decay constant

These equations make allowance only for radioactive decay as a means of reduction of the total airborne activity; it is known that in the particular case of ^{85}Kr deposition on land surfaces is negligible and solubility in the oceans is extremely low [13,14]. Equations (4) and (5) are used to calculate ^{85}Kr inventories in the two hemispheres corresponding to present and predicted discharge rates associated with the world power programme [1]. Dose rates from the corresponding uniform ^{85}Kr concentrations are calculated from the relationship that 1 pCi ^{85}Kr/g air gives 2.1×10^{-5} rad/yr to gonads [7].

4. ESTIMATED GONAD DOSE RATES FOR INDIVIDUAL NUCLIDES

Gonad dose rates are calculated as a function of distance for unit activity discharge rate of the inert gases produced either as fission or activation products in the nuclear power industry. Dose rates for the short-lived activation products ^{13}N, ^{16}N and ^{41}Ar to a distance of 100 km are shown in figure 1. Dose rates for krypton isotopes and daughters, excluding the long-lived ^{85}Kr, to the same distance are given in figures 2a and 2b. Dose rates for xenon isotopes and daughters to a distance of 1000 km are shown in figures 3a and 3b. All relate to a 30 m effective stack height which is taken as representative of typical reactor buildings. The finite cloud model described in sections 2.1 and 2.2 is used for all nuclides except ^{16}N and ^{83m}Kr. The model for ^{16}N is described in section 2.3. A semi-infinite cloud model is used

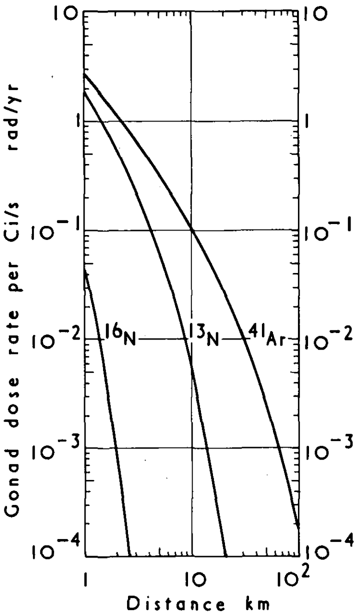

FIG.1. Gonad dose rates for unit discharge rate from a 30-m effective stack height, ^{13}N, ^{16}N, ^{41}Ar.

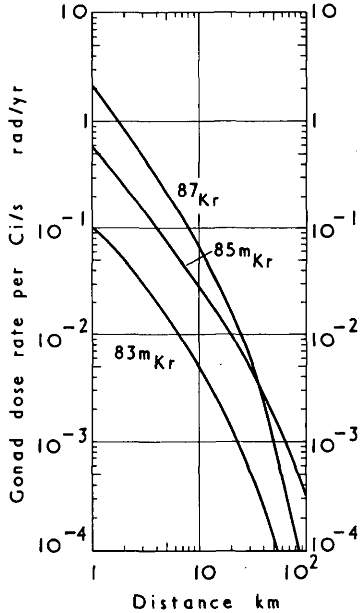

FIG.2a. Gonad dose rates for unit discharge rate from a 30-m effective stack height, krypton isotopes.

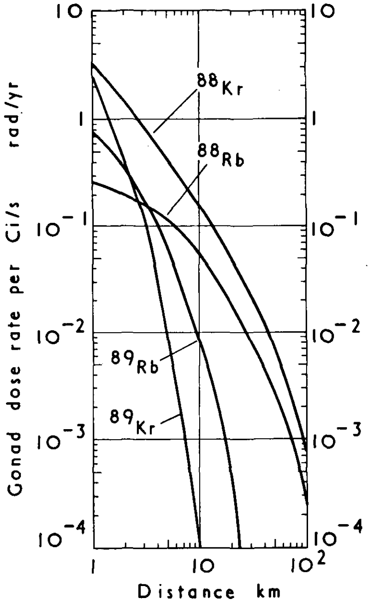

FIG.2b. Gonad dose rates for unit discharge rate from a 30-m effective stack height, krypton isotopes and daughters.

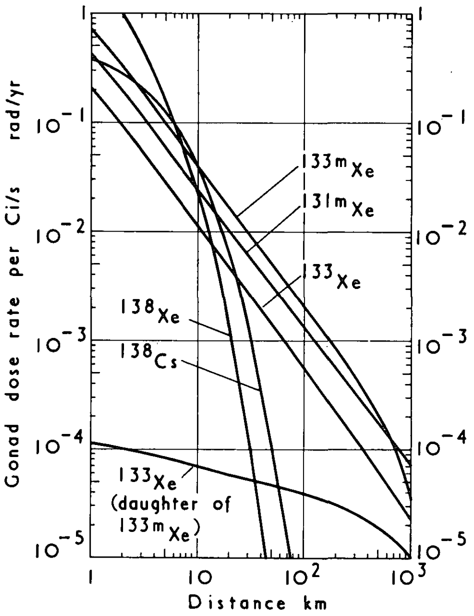

FIG.3a. Gonad dose rates for unit discharge rate from a 30-m effective stack height, xenon isotopes and daughters.

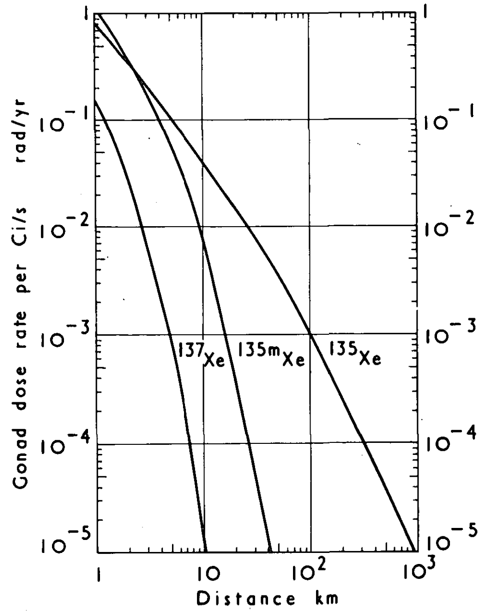

FIG.3b. Gonad dose rates for unit discharge rate from a 30-m effective stack height, xenon isotopes.

for ^{83m}Kr because the mean free path of its low energy gamma ray is only about 2 m. Decay schemes for the nuclides considered are given in table I [15,16]. Some of the decay schemes are very complex and gamma energies are grouped where appropriate. It is valid to use attenuation coefficients and relationships between photon flux density and dose rate for the average energy of each group because these parameters vary only slowly with photon energy.

The effect of varying stack height from 0-150 m is illustrated in figure 4 where gonad dose rates for unit discharge rate of ^{85}Kr are shown for distances up to 100 km. These values are calculated using the finite cloud model described in sections 2.1 and 2.2; this model is identical with that used previously [1].

Average gonad dose rates to the UK population arising from discharges of ^{85}Kr produced in present and predicted UK and world-wide power programmes [1] are shown in figure 5. This figure is a revised version of figure 3 in reference [1]. There is no change in the curve showing dose rates from the finite cloud ("first pass") arising from discharges at an effective height of 100 m; changes in the other curves result from the revised world-wide dispersion model described in section 3. Dose rates arising from circulation of ^{85}Kr in the latitude band are lower because the model now assumes uniform mixing to a height of 10 km instead of 3 km; dose rates arising from circulation of ^{85}Kr in the northern hemisphere are lower because the model now assumes slow transfer of ^{85}Kr to the southern hemisphere.

5. ESTIMATED GONAD DOSE RATES FROM MIXTURES OF RADIOACTIVE INERT GASES

Gonad dose rates as a function of distance for unit discharge rate of mixtures [17] of radioactive inert gases from three types of reactor are given in figure 6. Gas-cooled reactors (GCR), particularly those with steel pressure vessels, discharge mainly the activation product ^{41}Ar. The design of some boiling water reactors (BWR) includes a delay system whereby radioactive inert gases are held for a few tens of minutes before discharge to atmosphere; the discharges consist predominantly of very short-lived krypton and xenon isotopes. The design of pressurised water reactors (PWR) includes a delay system in which the gases are held for up to 120 days before discharge to atmosphere; measured discharges consist mainly of the longer-lived xenon isotopes, particularly ^{133}Xe.

6. DISCUSSION

The models used to calculate gonad dose rates arising from unit discharge rate to atmosphere of a wide range of radioactive inert gases provide more accurate results than earlier and more simple models. Present models take into account, inter alia, radioactive decay, the finite size of the cloud during the first stage of dispersion in the atmosphere, and subsequent circulation in a latitude band prior to more general circulation in the northern and southern hemispheres.

The results, displayed graphically, show that gonad dose rates for a large number of short-lived radionuclides considered decrease by at least four orders of magnitude between 1 km and 100 km from the discharge point. Unless amounts discharged were to be very large, only the longer-lived xenons (^{131m}Xe, ^{133m}Xe and ^{133}Xe) are likely to be of significance at distances greater than this. Gonad dose rates for these xenon isotopes are given to a distance of 1000 km.

TABLE I. DECAY SCHEMES USED IN THE CALCULATION [15, 16]

Nuclide	Half life	Mean beta energy MeV/disintegration	Representative gamma energy MeV	Representative branching ratio for gamma decay
^{13}N	10.0 min	0.40	0.511	2.00
^{16}N	7.2 s	1.87	6.18	0.73
^{41}Ar	1.83 h	0.40	1.29	0.992
^{83m}Kr	1.86 h	0.028	0.009	0.09
^{85m}Kr	4.4 h	0.28	0.238	0.77
^{85}Kr	10.76 yr	0.234	0.514	0.0043
^{87}Kr	76 min	1.05	0.405	0.92
			0.85	0.08
			2.26	0.22
^{88}Kr	2.8 h	0.34	0.19	0.38
			0.85	0.15
			2.16	0.68
^{88}Rb	17.8 min	1.53	1.00	0.21
			2.00	0.20
^{89}Kr	3.2 min	1.33	0.215	0.31
			0.50	0.99
			1.00	0.55
			2.00	0.70
			3.00	0.26
			4.00	0.16
^{89}Rb	15.4 min	0.51	1.00	0.68
			2.00	0.87
			3.00	0.29
^{131m}Xe	11.8 d	no beta	0.16	1.00
^{133m}Xe	2.26 d	no beta	0.23	1.00
^{133}Xe	5.27 d	0.115	0.081	1.00
^{135m}Xe	15.6 min	no beta	0.53	1.00
^{135}Xe	9.2 h	0.3	0.255	1.00
^{137}Xe	3.9 min	1.31	0.45	0.33
^{138}Xe	17 min	1.33	0.255	0.84
			0.405	0.48
			2.00	0.65
^{138}Cs	32.2 min	2.7	0.255	0.045
			0.50	0.34
			1.00	1.02
			2.20	0.18
			3.00	0.005

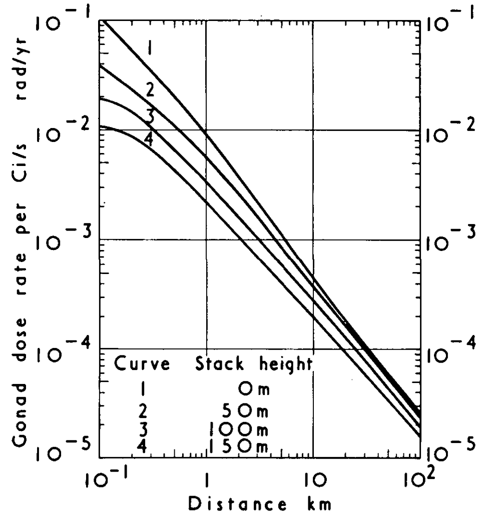

FIG.4. Gonad dose rates for unit discharge rate from stacks of different height, ^{85}Kr.

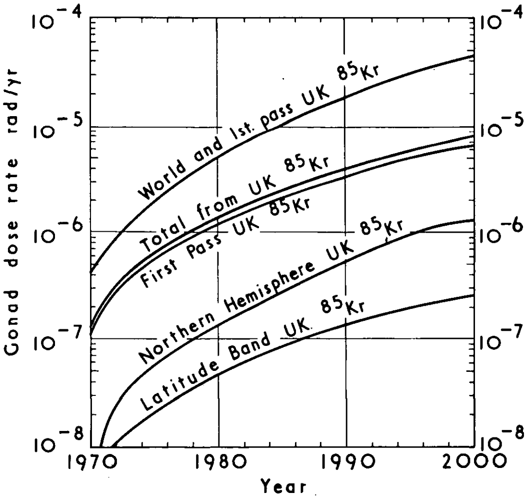

FIG.5. UK gonad dose rate due to ^{85}Kr from the UK and world power programs (revised version of Fig.3 of Ref. [1]).

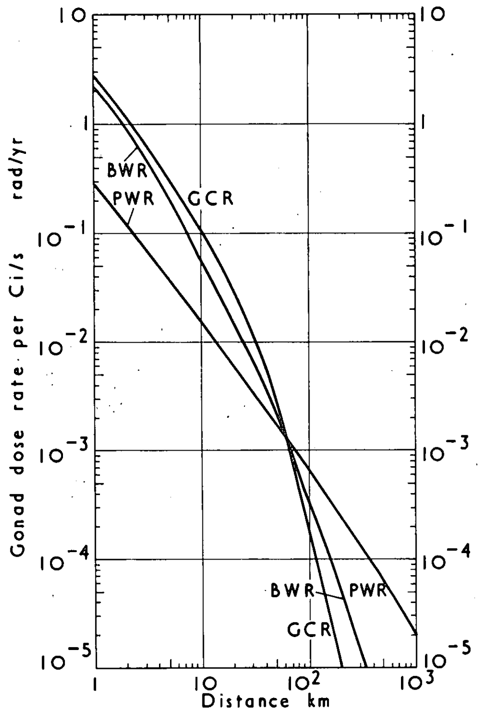

FIG.6. Gonad dose rates for a unit discharge rate of a typical mixture from a BWR and a PWR and of ^{41}Ar from a GCR.

Operational procedures, particularly the duration of delay before discharge, radically affect the composition of xenon and krypton isotope mixtures discharged from reactors. It is clear from examples of gonad dose rates for unit discharge rate of mixtures from two water-cooled reactors that dose rates fall off very much more slowly with distance when the delay is up to 120 days (PWR) than when it is only a few tens of minutes (BWR). The implication is that, for a given dose rate at the boundary of a reactor site, population radiation exposure, expressed as man-rads, is greater when radioactive gases are held for longer periods before discharge.

The estimation of total gonad dose rates for the long-lived ^{85}Kr includes contributions from each stage of the dispersion process. Dose rates for ^{85}Kr are expressed in terms of present and predicted UK and world-wide power programmes, on the assumption that all ^{85}Kr produced is discharged. At present most of the dose to the UK population from ^{85}Kr discharges results from "first pass" UK ^{85}Kr; the total dose rate arising from this source and from ^{85}Kr produced in the world-wide power programme is not affected by the model used for world-wide dispersion. By the year 2000, the contribution to dose to the UK population from ^{85}Kr produced world-wide is predicted to be much larger than that from "first pass" UK ^{85}Kr; the revised model predicts total dose-rates at the end of the century which are 50% of those calculated previously by the authors.

The effect of varying effective stack height on dose rates from the finite cloud is demonstrated by considering unit discharge rate of ^{85}Kr. Over the range of height 0–150 m dose rates differ by an order of magnitude at 0.1 km, falling to a factor of two at 10 km from the discharge point. The similarity in the dose rate curves for ground level discharge and a 50 m stack suggests that the choice of an effective height of 30 m as representative of typical reactor buildings is not critical.

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DISCUSSION

TO PAPERS IAEA-SM-172/76 AND IAEA-SM-172/5

A. BAYER: Miss Bryant, does Fig. 5 of your paper include the gonad dose due to ^{85}Kr from weapons tests? If not, what in your opinion is the size of this dose?

Pamela M. BRYANT: The figure does not include the gonad dose rate due to ^{85}Kr released in weapons tests, but this component at present constitutes only a small fraction of the gonad dose rate due to ^{85}Kr discharged by the nuclear power industry.

W. O. SCHIKARSKI: The values assumed for hold-up times for rare gases in LWR power plants are obviously important for your model. You mentioned 20 minutes' hold-up time for BWRs. In Germany, however, hold-up times for BWRs are of the order of 40 to 60 days. I take it that this would have some effect on your figures.

Hold-up times, incidentally, vary from one power plant to another and from one country to another, and depend on such factors as the date of plant construction, the type of off-gas system installed, and so on.

Pamela M. BRYANT: I agree that the results for mixtures of inert gases from power plants depend upon a number of factors, including hold-up time. We used published data which related to plants of early design. However, our values for the unit discharge rate of individual nuclides can be applied to any specified mixture of gases.

H. WIJKER: I have two questions and a comment for Miss Bryant. (1) Does she believe that it would be advisable to increase hold-up times of ^{85}Kr and other gases, especially with a view to decreasing the dose to the general public? (2) What assumptions were made in the calculational model concerning: (a) height distribution; (b) meteorological conditions; and (c) the time during which persons are shielded inside buildings?

Pamela M. BRYANT: In reply to your first question, it must be remembered that any benefit to the public from increased hold-up times of radioactive gases must be weighed against the possibility of increased doses to persons occupationally exposed.

The answers to your second group of questions are:

(a) We used a Gaussian distribution with height for the finite cloud model;

(b) We used weighted mean meteorological conditions by means of values for the annual average frequency of occurrence of Pasquill's stability categories in the United Kingdom;

(c) We made no allowance for shielding by buildings.

H. WIJKER: Starting from adiabatic changes with height changes (i.e. equilibrium) and using, instead of the adiabatic equation, the ideal gas equation and the equation that the pressure gradient balances the weight of each volume element, one can calculate temperature T(z), pressure p(z) and density $\rho(Z)$ versus height Z. Carrying out the calculation for air one finds a linear relation of temperature with height, viz. $dT/dz = 1/30°C/m$. This gives an atmospheric height of about 8 km, where T becomes 0°K. Inversions and mixing near the earth's surface, disturbing this overall temperature distribution to some extent, have here been ignored.

For the calculation of the ^{85}Kr distribution with height the above-mentioned temperature profile was used by us to replace the adiabatic equation. From these equations, viz. $dT/dz = -1/30°C/m$, $p = \rho RT$ and $dp/dz = -g\rho$, with $g = 9.8$ m/s^2, one can derive for ^{85}Kr $\rho(0)$ = about $3\overline{\rho}$ related to an equivalent height of about 3 km (i.e. the density at the earth's surface is about three times the average over the column). This is only one of the models that could be chosen, of course.

J. BOGEN: What is the reason for the different ^{85}Kr residence times you showed in a slide during your oral presentation (i.e. 3 months in the latitude band 35-60°N and 2 years in the "northern hemisphere")?

Pamela M. BRYANT: The residence time of 3 months represents the resistance to exchange between tropospheric air in the latitude band 35-60°N and that in the band 0-35°N. In other words the exchange constant governing horizontal transfer of air across the apparent barrier at about 30°N is 0.33 per month. The residence time of 2 years in the northern hemisphere represents the resistance to exchange of air between the northern hemisphere as a whole and the southern hemisphere. The exchange constant governing transfer of air across the equator is therefore 0.5 per year.

J. BOGEN: Is this hypothesis based on any measurements of your own?

Pamela M. BRYANT: We have not made measurements ourselves, but have used models proposed by other workers on the basis of data on global dispersion of man-made radionuclides.

J. BOGEN: But will there not be a difference in residence time as between radioactivity released by weapons tests and by nuclear reactors?

Pamela M. BRYANT: The rate of movement of weapons test activity between regions of the troposphere was used to quantify the rate of transfer of air that would carry airborne activity with it, irrespective of its origin.

K.-J. VOGT: I also have some questions for Miss Bryant. Regarding the global distribution of ^{85}Kr, does she take into account any removal processes, i.e. deposition onto the ground or into the ocean, in estimating the residence time in the troposphere? Secondly, on the subject of extension of the Pasquill model up to 1000 km, how did she extrapolate the standard deviations specified by Pasquill for distances up to 100 km?

Pamela M. BRYANT: We did not take removal processes into account in estimating gonad dose rates from ^{85}Kr in the troposphere because they have a minimal effect — the dry deposition rate and the solubility of Kr being very low. The mean residence time concerned is that governing interchange of air, and hence ^{85}Kr, between neighbouring regions in the troposphere.

We used Pasquill's values for standard deviations in the vertical direction for distances up to 100 km; we then assumed, after discussion with Pasquill himself, that the Gaussian distribution gradually breaks down until there is uniform concentration with height at 1000 km.

R.F. FOSTER: Owing to the low solubility of krypton, the ^{85}Kr content of the ocean must indeed be very low, but all the same it would be interesting to hear of any calculations that have been made of the fraction of the world inventory of ^{85}Kr that is in fact contained in the ocean or of the turnover time.

R. FUKAI: Several attempts have been made to estimate this. Preston at Lowestoft, UK, Volchuk at HASL, USA, and myself have all made independent estimates on the basis of slightly different assumptions, but have all come up with similar results, i.e. a figure of 0.1 - 0.2% for the fraction of the total inventory of krypton-85 present in the oceans. Incidentally, these results appear in the proceedings of a seminar on the pollution of the marine environment published by FAO in 1971.

H.F. MacDONALD: At the UK Central Electricity Generating Board we have used models similar to that described by Miss Bryant to evaluate doses due to airborne discharges from nuclear power reactors. At present we are engaged in folding in dose distributions obtained in these studies with population distributions in order to calculate the integrated population doses of the type discussed by Mr. Lindell this morning in his paper IAEA-SM-172/B. I would like to know if Miss Bryant's results enable her to say whether, on this basis, noble gas releases are likely to be limited by doses to individuals exposed close to the source, or by the integrated dose to the population of a region up to 100 km from the source.

Pamela M. BRYANT: Our work on the calculation of population doses, expressed in man-rads, has really only just started, but in the case of ^{85}Kr discharges skin dose to local population groups is the limiting criterion.

R. M. BARKHUDAROV: The following questions relate to the paper presented by Mr. Kaye. First, how is the tritium intake calculated — from drinking water only or from food also? Secondly, what exactly is shown in Fig.2 — the dependence of drinking water consumption on age or the dependence of total water consumption on age?

S. V. KAYE: The pathway considered is intake of tritiated water (HTO) from all sources; intake of tritiated organic molecules in food was not taken into account.

Figure 2 shows the age-dependent intake of total water.

R. LE QUINIO: Regarding the behaviour of krypton, I think there is no doubt that this gas diffuses in the same way as the other components of air; the relative homogeneity of the composition of air up to an altitude of 100 or even 200 km is a proof of good mixing of all the components.

On another point, it should be remembered that calculation of man-rems, without the concept of a threshold, results in infinite values. The population increases as the square of x (x = distance) and the individual doses decrease as $x^{-\alpha}$ (α being always less than 2), whatever the atmospheric diffusion model adopted.

R. FUKAI: Miss Bryant, may I revert to your statement that the residence time of ^{85}Kr in the troposphere is around 3 months (based on the data for ^{14}C and ^{90}Sr). Now, as we know, ^{85}Kr is not easily soluble in seawater, while ^{14}C and ^{90}Sr are fairly soluble therein. Under these circumstances, do you consider yourself justified in using the residence time for ^{14}C or ^{90}Sr in estimating ^{85}Kr distribution?

Pamela M. BRYANT: The mean residence time concerned refers to exchange of air in a horizontal direction between neighbouring regions of the troposphere, and not to transfer to seawater. The model is therefore appropriate to transfer of ^{85}Kr between neighbouring regions.

J. BOGEN: Mr. Kaye, in the paper you presented the dose rate was discussed only on the basis of HTO. However, in the atmosphere tritium also exists in the form of HT. Can you give any figure for the fractions of tritium in HTO and T in the form of HT, and their contribution to the dose?

S. V. KAYE: I do not recall the exact fractions of environmental tritium present as T_2 or HT, but I believe that they are small. Forms of tritium such as T_2 and HT are quickly oxidized to tritiated water, HTO. Since HTO is the principal form to which man is exposed, we based all our assessments on tritiated water.

ENVIRONMENTAL BEHAVIOUR OF RADIONUCLIDES

(Session II)

Chairman:

A.F. GIFFORD (United States of America)

Mémoire commandé

PRINCIPES DE LA SURVEILLANCE DE L'ENVIRONNEMENT D'UNE CENTRALE NUCLEAIRE

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Abstract–Résumé

PRINCIPLES OF ENVIRONMENTAL MONITORING AT NUCLEAR POWER STATIONS.

The authors review the development of radiation protection as an accompaniment to that of nuclear power generation, and compare the efforts in this field with those made to combat conventional forms of pollution. The principles of environmental monitoring at nuclear power stations that have emerged during the growth of nuclear power generation are set forth; they should serve as a model for the surveillance of conventional forms of pollution. The results of current environmental monitoring are discussed, it being noted that contamination of the environment and of man through the operation of nuclear power stations is at present so slight as to be virtually undetectable. The authors point to the need for a more objective assessment of the risks associated with nuclear power generation, which have been reduced to a minimum by – in particular – the control and monitoring arrangements instituted by public health authorities.

PRINCIPES DE LA SURVEILLANCE DE L'ENVIRONNEMENT D'UNE CENTRALE NUCLEAIRE.

Les auteurs envisagent d'un point de vue historique le développement de la radioprotection parallèlement à celui de l'énergie électro-nucléaire, en comparant les efforts mis en jeu dans ce domaine à ceux qui ont été développés dans la lutte contre les pollutions conventionnelles. Les principes de la surveillance des centres nucléaires, qui se sont dégagés au cours de cette évolution, sont exposés: ils devraient servir de modèle dans le domaine des pollutions conventionnelles. Les résultats actuels de la surveillance de l'environnement sont passés en revue: l'influence des centrales nucléaires sur la contamination du milieu et de l'homme demeure actuellement pratiquement indécelable. En conclusion, il apparaît la nécessité d'une appréciation désormais plus objective d'un risque rendu minime grâce, en particulier, aux structures de contrôle et de surveillance mises en place par les autorités de Santé publique.

1. INTRODUCTION

Le comportement des radioéléments dans l'environnement et leur surveillance ont fait, depuis vingt ans, l'objet de publications dont le moins qu'on puisse dire est qu'elles sont innombrables. Il faut cependant bien reconnaître que beaucoup d'entre elles apportent peu d'éléments réellement nouveaux.

Ainsi, pour la seule année 1971, nous avons compté plus de cinquante congrès divers sur la radioprotection, ce qui correspond à plus de 2 500 communications. Vingt de ces réunions avaient pour thème spécifique la contamination radioactive de l'homme et de l'environnement.

Parallèlement, les périodiques scientifiques publient en moyenne 5 000 articles par an sur la radioprotection. Pour la seule année 1970, on relève dans le Nuclear Science Abstracts plus de mille titres se rapportant spécifiquement à la contamination radioactive, dont 390 pour le seul Césium 137.

La bibliographie complète des travaux relatifs à la contamination radioactive nécessiterait à elle seule plusieurs volumes.

Le cheminement des radioéléments dans l'environnement n'est certes pas entièrement élucidé à l'heure actuelle. Mais on doit reconnaître qu'il a fait l'objet d'études, tant théoriques qu'expérimentales, dépassant largement en quantité comme en qualité tout ce qui a pu être fait au sujet des autres pollutions, dont plusieurs sont pourtant beaucoup plus graves.

Pendant ce temps-là, en effet, pour les nuisances traditionnelles, on ne note, par exemple pour le plomb, qu'un seul congrès à Amsterdam en 1972 avec quelques dizaines de communications....

Ce problème des pollutions graves est pourtant vieux comme l'humanité puisque, pour ne citer qu'un exemple, l'intoxication chronique par le plomb a pu être considérée par certains auteurs comme l'un des facteurs de la décadence de l'empire romain (31)! Or, de nos jours, la pollution par le plomb, à laquelle est venue s'ajouter la pollution par le mercure, reste toujours l'une des plus préoccupantes.

A notre époque, le taux de mortalité et de morbidité par cancer s'est accru dans des proportions alarmantes depuis le début du siècle (1) (2). On commence seulement à dresser l'inventaire des causes probables, qui sont innombrables (3), parmi lesquelles se trouvent la consommation excessive du tabac, l'usage de plus en plus répandu du goudron, les additifs alimentaires, l'exposition inconsidérée au soleil, l'amiante, pour ne citer que les plus connues. Si l'on voulait consacrer à ces nuisances indiscutables des efforts proportionnels à ceux qui ont été déployés pour les contaminations radioactives, on dépasserait certainement de très loin les possibilités technologiques et financières de la société dans laquelle nous vivons.

Il est donc grand temps de ramener l'activité dépensée dans les congrès et les publications sur la radioprotection à des proportions qui correspondent de façon plus objective à l'importance très relative du risque qu'elle a pour mission de contrôler.

Aussi, du point de vue de l'hygiène publique bien comprise, paraît-il plus utile de tirer les enseignements réalistes de l'expérience acquise dans ce domaine plutôt que de refaire, une fois de plus, l'énumération stérile d'éléments que tous les spécialistes n'ont déjà que trop entendue.

De toutes les sources de pollution, l'utilisation industrielle de l'énergie nucléaire est actuellement celle qui suscite les attaques

les plus violentes, alors que les niveaux de pollution correspondants sont si bas qu'ils échappent dans de nombreux cas à la détection par les techniques les plus fines. Il est classique d'invoquer, comme causes de cette inquiétude du public, la confusion entre énergie nucléaire et explosifs nucléaires d'une part, et la crainte des effets génétiques d'autre part (pour lesquels il serait d'ailleurs erroné de prétendre qu'ils constituent une exclusivité de la pollution radioactive).

Pour pouvoir mettre un terme à cette situation paradoxale, il faut d'abord en analyser les causes de manière beaucoup plus approfondie. C'est ce qui avait d'ailleurs été fait, dès 1958, dans le rapport n° 151 de l'OMS (4), intitulé "Questions de santé mentale que pose l'utilisation de l'énergie atomique à des fins pacifiques". Les conclusions de ce rapport traduisaient une grande sagesse et une grande clairvoyance de la part de ceux qui les avaient énoncées, mais aussi, il faut bien le dire, un certain courage, celui de confronter les moyens mis en oeuvre avec l'importance réelle du danger contre lequel on prétend les utiliser, surtout lorsqu'on le compare à celui des autres nuisances.

Lorsque l'énergie nucléaire a commencé à être utilisée à des fins pacifiques, études, exploitation et protection étaient généralement confiées à un organisme unique pour chaque pays. Cette situation, satisfaisante au départ, a cessé de l'être dès qu'a débuté la phase d'exploitation industrielle. En effet, il y a une vingtaine d'années, le schéma suivant s'offrait aux yeux d'un public qui commençait à se sentir concerné:

-d'une part, multiplication de services de radioprotection dépendant de l'exploitant: l'ampleur des études menées, l'attitude très circonspecte de ces services dont le public ne pouvait logiquement mettre le perfectionnisme que sur le compte d'un danger exceptionnel, ont contribué à susciter un climat de crainte amenant progressivement dans l'esprit du public, de façon totalement injustifiée, l'énergie nucléaire au premier rang des dangers créés par la civilisation. Il faut ajouter que la multiplication exagérée des congrès, symposiums, séminaires de radioprotection à tous les niveaux a encore aggravé cette situation, et continue d'ailleurs malheureusement à le faire.

-d'autre part, des services de santé publique alors presque inexistants, laissant pratiquement l'initiative et l'entière responsabilité de la protection aux exploitants, d'où l'impression d'insécurité et d'angoisse ressentie par le public.

Si l'on veut que régressent ces inquiétudes mal fondées, il convient de donner définitivement à la radioprotection une structure efficace à la fois sur les plans technologique et psychologique. Dans des pays de plus en plus nombreux, cette structure est désormais en place; mais l'opinion publique est en retard sur la réalité des faits. Une mise au point dans ce sens apparaît nécessaire, et les éléments essentiels à prendre en considération sont les suivants:

-responsabilité de l'exploitant qui doit prendre toutes mesures, tant au niveau de la prévision que du fonctionnement, pour ne porter atteinte à la santé de quiconque du fait de ses activités. Cette responsabilité implique qu'il soit parfaitement conscient de la situation qu'il crée et, par suite, qu'il vérifie lui-même constamment les conséquences de son activité pour l'environnement.

-contrôle indépendant exercé par la santé publique, seul organisme pouvant se porter garant, aux yeux des populations concernées, de l'efficacité des mesures prises par l'exploitant.

-réalisme à l'égard du très faible niveau de pollution radioactive actuel et des prévisions pour les années futures. Sans pour autant abandonner les études qui sont indispensables pour permettre à l'exploitant et aux organismes de santé publique d'assumer leurs responsabilités, il faut désormais résolument distinguer entre l'évaluation réaliste des niveaux de contamination, et les considérations académiques dans lesquelles trop nombreux sont ceux qui se perdent encore, au détriment d'une politique efficace de santé.

2. LA SITUATION ACTUELLE DE L'ENERGIE NUCLEAIRE

La production d'électricité d'origine nucléaire date d'il y a 20 ans; aujourd'hui, nous assistons au début des réalisations de grande puissance qui, selon les estimations actuelles, pourront permettre à la planète en l'an 2 000 de faire face à 60% de ses besoins en énergie électrique (plusieurs milliers de gigawatts) (5) (6) (7).

A la fin de l'année 1972, par exemple, la France disposait de 5 centrales, produisant au total 2 500 mégawatts électriques. Si l'on exprime le chiffre en mégawatts x ans, le recul dont nous disposons pour juger du retentissement de ces réalisations sur le milieu est, certes, encore modeste. A l'échelle mondiale, ce recul équivaut cependant au fonctionnement de cent centrales durant un an, et force est de reconnaître qu'elles n'ont suscité aucun souci majeur, comparable à ceux que donnent périodiquement, sans que l'on s'en alarme d'ailleurs, les autres sources d'énergie électrique.

Cependant, parallèlement au perfectionnement des moyens de production de l'énergie nucléaire, les techniques de radioprotection ont elles-mêmes évolué, et les bases théoriques et juridiques se sont clarifiées. Du point de vue du risque radioactif, on peut dire non seulement que la situation actuelle est très saine, mais encore que les dispositions prises constituent un modèle que la protection contre les autres nuisances adopterait avec profit.

Mais, nous l'avons dit, le grand public connaît mal cette remarquable évolution et ne retient malheureusement des échos qui lui parviennent que les hésitations de certains experts et les discussions byzantines qui contribuent à créer artificiellement un climat d'incertitude. Enfin, certaines presses, pour lesquelles le sensationnel dramatique représente un aliment de choix, les reprennent et les amplifient à l'envi.

Or, par delà les querelles économiques ou pseudo-philosophiques, et la plupart du temps exploitées à des fins politiques, l'humanité est maintenant confrontée avec un choix inévitable: ou bien elle décide (et c'est son droit a priori) le retour aux techniques ancestrales et l'abandon total du progrès, en acceptant en contre-partie la résurgence de fléaux que l'on a tendance à considérer trop légèrement comme définitivement abolis; ou bien la progression actuelle est maintenue et, dans ce cas, la contestation de l'énergie nucléaire est une imposture. En effet, l'on peut ergoter autant que l'on voudra sur l'exactitude des prévisions quant aux réserves de pétrole, une seule chose est certaine: seule l'énergie de fission permettra de passer le cap des quelques décennies nécessaires à l'acquisition d'autres sources d'énergie (6) (8).

3. LES PRINCIPES DE LA SURVEILLANCE

3.1. Définition des normes de sécurité

A l'inverse de ce qui s'est passé dans le cas de nombreuses autres nuisances beaucoup plus inquiétantes, des instances d'abord nationales, puis internationales ont, dès l'origine, étudié les effets des rayonnements ionisants et préconisé des règles.

Formée en 1928, la Commission Internationale de Protection Radiologique (CIPR) publiait dès cette époque des recommandations révisées tous les 3 ans. En 1956 apparaissait dans ces recommandations le concept de "niveau admissible" concrétisé par des limites chiffrées. Ces limites maximales admissibles étaient amenées à leur valeur actuelle en 1958; les recommandations publiées à cette date constituent la base aujourd'hui encore pratiquement inchangée des règles de radioprotection (9) (10).

A l'origine, les règles de sécurité ont été déduites de l'expérience fournie par l'observation clinique des patients et des médecins en contact avec les rayons X et le radium. Les limites maximales admissibles ont donc été fixées dès le départ comme des limites pratiques d'irradiation au-dessous desquelles l'expérience n'avait pu mettre en évidence aucun effet pathologique, ni à court ni à long terme.

C'est ainsi par exemple que la charge corporelle maximale admissible pour le Radium 226, qui a servi de base à la détermination des limites maximales admissibles dans le cas des contaminants ostéotropes, est fondée sur le fait qu'on n'a jamais observé de tumeurs chez des personnes dont l'organisme contenait une quantité de Radium de 0,1 µCi (11).

Cette expérience clinique porte sur une population plus importante qu'il n'est courant de le penser. En effet, la contamination à de faibles niveaux de masses importantes de population par les retombées radioactives ou par les centrales nucléaires est souvent considérée comme un phénomène nouveau. Il ne faut pourtant pas oublier que:

-aux premiers temps de leur utilisation, les rayonnements ionisants bénéficiaient de l'attrait de la nouveauté et passaient même pour un remède miracle (citons par exemple les "inhalateurs de radium", appareils contenant une source de radium de quelques millicuries qui avaient leur place dans les familles afin de permettre l'inhalation quotidienne d'une bouffée de radon auquel on prêtait de nombreuses vertus curatives et préventives!).

-l'irradiation à des fins médicales touche pratiquement l'ensemble de la population, et ce à des niveaux en moyenne cent fois supérieurs à ceux dont sont responsables les retombées radioactives et les centrales nucléaires réunies; ces irradiations atteignent d'ailleurs des valeurs encore beaucoup plus élevées lorsqu'elles sont appliquées à des fins thérapeutiques, sur un pourcentage non négligeable de la population (12).

A ces considérations pratiques fondées sur l'étude clinique s'ajoutent des travaux théoriques fondés d'une part sur le calcul et d'autre part sur les résultats de très nombreuses études statistiques (13) (14). Leurs résultats conjoints sont indiscutables si l'on considère les effets d'irradiations supérieures à 10 rems. Au-dessous de cette valeur, les résultats statistiques sont contradictoires. Mais la CIPR, devant ces incertitudes, a toujours retenu l'hypothèse la plus pessimiste: par exemple, le choix délibéré d'une relation linéaire entre les doses et les effets aux très faibles débits de dose. En fait, cette relation apparaît comme une limite très improbable. Elle recouvre en effet des phénomènes très complexes dans lesquels les pouvoirs colossaux de restauration cellulaire, qui commencent seulement à être soupçonnés, jouent un rôle prépondérant (15). Elle conduit certainement à retenir des limites maximales admissibles comportant un facteur de sécurité très grand (16).

Compte tenu du fait que la surveillance individuelle réglementaire à laquelle sont soumis les travailleurs, pour qui ont été définies ces limites, ne peut être raisonnablement étendue à chaque individu, la CIPR recommande, pour les membres du public, des doses limites moyennes, dérivées des doses maximales admissibles définies pour les travailleurs par application d'un facteur 10 de réduction. La CIPR confirme (publication 9, § 74) que "compte tenu du faible risque impliqué, les conséquences de dépassements même substantiels des doses limites seront vraisemblablement mineures" (10).

Enfin, l'irradiation des populations dans leur ensemble doit tenir compte à la fois des doses individuelles et du nombre de personnes impliquées, puisque le risque pris en considération est ici le risque génétique. La CIPR précise au § 86 de la publication 9: "La Commission recommande de maintenir la dose génétique de la population à un minimum compatible avec les nécessités et ne dépassant pas en tous cas 5 rems pour l'ensemble des sources s'ajoutant au fond naturel de rayonnement et aux utilisations médicales". Il s'agit bien entendu d'une dose moyenne, les limites individuelles demeurant celles fixées pour les "membres du public" (10).

Il est donc clair que, jamais pour aucune autre nuisance, des facteurs de sécurité aussi importants n'ont été adoptés. On entend parfois avancer que, pour la pollution chimique, les effets seraient mieux connus, et que les facteurs de sécurité seraient donc superflus. Or, il n'en est rien; en particulier les effets des rejets des centrales à charbon et à fuel sont beaucoup moins bien connus que ceux des rayonnements ionisants (17). Il ne saurait donc être question de remettre en cause ces normes; par contre, leur application aux cas concrets propres à chaque pays ne peut être faite sans adaptation, sans intégration dans une réglementation tenant compte des différents facteurs locaux (18) (19). Ces normes ne sont pas, non plus, un totem garantissant à elles seules l'efficacité de la protection. Seul le respect des règles élémentaires que nous exposerons ci-dessous peut permettre d'atteindre ces objectifs à travers le respect des normes.

C'est, peut-être, pour avoir compris trop tard qu'il était indispensable de se plier à ces exigences, que les promoteurs de l'énergie nucléaire doivent aujourd'hui faire face à une campagne d'opposition dont les effets se sont déjà révélés désastreux dans certains pays.

3.2. La répartition des responsabilités (20) (21)

Nul ne doit, volontairement ou non, porter atteinte au patrimoine ou à la santé d'autrui du fait de ses activités. Il s'agit là d'un principe de droit très général, et il est donc naturel que la responsabilité de l'exploitant d'une centrale nucléaire soit engagée au premier chef, dans la mesure où l'implantation d'une centrale dans un milieu donné risquerait, si l'on n'y prenait garde, de porter atteinte à l'intégrité de ce milieu et à la santé des populations avoisinantes. A la phase d'études, cette responsabilité lui impose de fournir aux autorités des documents complets concernant les conséquences du fonctionnement de la centrale en régime normal, la prévision des incidents et accidents possibles, et la justification des moyens mis en oeuvre pour en diminuer la probabilité et les conséquences. Lors du fonctionnement de la centrale, l'exploitant devra être en mesure d'effectuer toutes analyses lui permettant d'être parfaitement informé de la qualité et de la quantité des différents rejets opérés, afin de vérifier le respect des limites fixées par les autorités de santé publique et d'être en mesure de prendre toutes dispositions (notamment en étalant les rejets) pour que ces limites soient respectées.

La responsabilité de l'exploitant ne s'arrête pas, c'est évident, aux limites de la centrale. Il doit s'assurer, par tous contrôles qui s'avèrent nécessaires, que les limites d'irradiation fixées ne sont pas dépassées pour les populations avoisinantes. Quelles que soient les études préalables et les précautions prises, sa responsabilité se trouverait engagée si telle éventualité se produisait sans que les autorités de santé publique en soient immédiatement averties.

Si ces contrôles sont obligatoires pour l'exploitant, qui doit s'assurer de l'innocuité de son industrie et demeure de toutes façons responsable du dommage éventuel causé, il est évident qu'ils ne sauraient constituer une garantie aux yeux des individus qui pourraient se sentir menacés. L'exploitant ne peut donc être juge et partie aux regards de la population intéressée et celle-ci, lorsqu'elle est amenée à poser des questions, se retourne vers des autorités à la fois compétentes et indépendantes vis-à-vis de l'industrie en cause.

C'est en effet aux autorités de santé publique qu'il appartient de veiller à la sauvegarde de l'hygiène publique et de la santé des individus et c'est précisément dans le cas de l'énergie nucléaire que les rapports et les limites de la responsabilité de l'exploitant et de celle des autorités de santé publique ont été, pour la première fois, clairement dégagées et articulées avec précision (22) (23) (24) (25).

Pour exercer cette surveillance, les autorités de santé publique sont conduites à faire appel aux spécialistes compétents des différents maillons de la chaîne qui conduit de la source de rayonnements à l'homme: spécialistes de technologie nucléaire, de météorologie, de géologie, d'écologie, et bien entendu médecins radiobiologistes. Si la sauvegarde de la santé des individus passe ici, comme c'est souvent le cas, par celle du milieu, l'objectif n'en demeure pas moins la protection de la santé de l'homme, et la responsabilité appartient donc évidemment en dernier ressort au médecin, quels que soient les processus de dispersion de la pollution et les voies d'agression. Cette nécessité a été ressentie par toutes les instances internationales: Organisation Mondiale de la Santé, Agence Internationale de l'Energie Atomique, Comité Scientifique des Nations Unies pour l'Etude des Effets des Rayonnements Ionisants. Il en résulte que, dans les pays de technologie avancée, c'est sur les autorités de santé publique que repose la responsabilité dernière de la surveillance des moyens de production de l'énergie nucléaire.

Ce principe de la séparation des responsabilités, qui paraît parfois nouveau en milieu industriel, n'est pourtant pas d'une nature différente de celui qui est admis de longue date dans de nombreux domaines traditionnels. Pour donner un exemple banal, il appartient à l'automobiliste, et à lui seul, de veiller au maintien en bon état de marche de son véhicule et, de même, c'est lui qui doit surveiller son compteur de vitesse et ajuster celle-ci selon les obligations et règles de la circulation routière. Par contre, la surveillance de la circulation n'appartient ni à l'automobiliste, ni aux constructeurs, mais aux autorités de sécurité routière sans lesquelles les panneaux de limitation risqueraient de rester lettre-morte.

4. LES RESULTATS DE LA SURVEILLANCE

La surveillance de la contamination du milieu par les centrales nucléaires, en fonctionnement normal, doit porter sur l'ensemble des opérations jusqu'à la prise en charge des déchets.

On peut, d'un point de vue pratique, considérer quatre types de radioéléments libérés, en fonction de leur période radioactive:

-les éléments à périodes courtes sont essentiellement des gaz nobles, dont le principal est l'Argon 41; ne pouvant s'accumuler du fait de la brièveté de leur période, les seuls problèmes qu'ils posent en pratique sont ceux de l'irradiation externe des populations du voisinage immédiat (quelques kilomètres sous le vent). Les questions liées au cheminement dans le milieu, ainsi qu'au métabolisme, sont donc ici réduites au minimum, et l'on considère que le seul problème éventuel qu'ils posent pour ces populations très réduites est celui de l'irradiation externe qui doit être maintenue inférieure à la limite maximale admissible.

-les éléments à périodes moyennes qui sont rejetés dans le milieu présentent une assez grande variété car, aux produits de fission s'ajoutent les produits d'activation dépendant de la constitution du réacteur. Parmi les radioéléments habituellement rencontrés, citons: le Manganèse 54, le Cobalt 58, le Cobalt 60, le Zirconium 95, le Ruthénium 103 et le Ruthénium 106 (26).

Le Ruthénium 106 apparaît comme l'un des radioéléments prépondérants. Sa période étant d'un an, on peut considérer que la contamination du milieu par cet élément est uniquement fonction de la puissance électrique produite, puisqu'il n'y a pas à craindre de phénomènes importants d'accumulation ultérieure. Quant au Cobalt 60, que sa période de 5 ans rapproche du groupe suivant, les activités émises en demeurent très faibles.

-les éléments à périodes longues sont ceux qui, à moyen terme, sont susceptibles de poser quelques problèmes: ce sont essentiellement le Tritium, le Krypton 85, le Strontium 90 et le Césium 137.

-citons enfin, pour mémoire, un radioélément à vie très longue, l'Iode 129; les activités en cause sont très faibles, et la contamination résultante du milieu ne pourrait devenir significative que dans un avenir très lointain, en admettant que sa rétention, d'ores et déjà partiellement effective, ne puisse être encore améliorée.

Il convient,pour terminer, de ramener à ses justes proportions le problème de la contamination par le Plutonium 239, dont le seul nom évoque dans les esprits les méfaits de l'arme nucléaire! Le Plutonium présent dans le combustible retraité est récupéré dans sa quasi-totalité, le reste étant pratiquement entièrement confiné sous forme de déchets de très faible volume. Les activités rejetées dans l'atmosphère sont variables d'un centre à l'autre, et elles sont généralement très inférieures à 1 curie par an et par centre de retraitement, pour la totalité des actinides (5) (27).

Si l'on fait le bilan de la radiocontamination retrouvée actuellement dans l'environnement par les contrôles systématiques effectués par les services de santé publique, une première constatation s'impose: dans l'atmosphère, l'eau de pluie, les végétaux et dans une grande mesure également dans l'eau de rivière, il reste très difficile, dans l'hémisphère nord au moins, de distinguer la contamination revenant aux centrales nucléaires de celle des retombées radioactives remontant pourtant, pour l'essentiel, aux années 1961-62. (Dans l'hémisphère sud, elle est encore actuellement très inférieure à celle de l'hémisphère nord).

Mais, compte tenu d'une part du fait que les radioéléments retrouvés sont essentiellement des produits de fission, d'autre part que le taux de contamination radioactive décroît de manière continue actuellement, il y a tout lieu de penser que la contamination retrouvée, d'ailleurs à la limite des seuils de mesure avec les appareils les plus sensibles, est presqu'entièrement due aux retombées radioactives, sauf, à un très faible niveau, dans le voisinage immédiat des centrales nucléaires.

Actuellement, l'activité volumique du Strontium 90 est de l'ordre de quelques millièmes de picocurie par mètre cube dans l'atmosphère; le seul radiocontaminant spécifique de l'activité des réacteurs nucléaires retrouvé assez régulièrement à proximité des centrales est le Cobalt 60, mais les valeurs moyennes relevées après dilution ne dépassent habituellement pas quelques centièmes de picocurie par mètre cube.

L'activité surfacique du Strontium 90 déposé par les retombées en un mois est de l'ordre du centième de millicurie par kilomètre carré; l'activité de ce même élément dans les végétaux est de l'ordre de quelques dizaines de picocuries par gramme de Calcium et, en moyenne, d'une dizaine de picocuries par gramme de Calcium dans le lait (26) (28).

La contamination due aux centrales est bien entendu retrouvée dans les analyses d'eaux de rivière en aval immédiat des points de rejet ainsi que dans les sédiments de rivière, pour lesquels des activités en Ruthénium 106, en Césium 134 ou en Césium 137, de quelques dizaines de picocuries par gramme de boues calcinées représentent des valeurs non exceptionnelles.

En ce qui concerne enfin les aliments, si l'on met à part les contaminations du lait et des rations alimentaires par les retombées, bien connues et bien surveillées sur le plan mondial (26) (28) (29), seuls les poissons de mer présentent dans certains cas une faible contamination provenant des centres nucléaires côtiers. Ces activités sont de l'ordre de la centaine de picocuries de Césium 134 et du millier de picocuries de Césium 137 par kilogramme de poisson frais.

Il est certain de toutes façons que l'indicateur essentiel est la contamination du corps humain: par delà toutes les considérations théoriques, elle apporte une preuve tangible de l'exactitude des modèles de transfert. A l'heure actuelle, on peut affirmer que les seuls radioéléments artificiels retrouvés (en dehors des contaminations professionnelles), à des niveaux significatifs mais extrêmement faibles dans le corps humain sont le Tritium, le Strontium 90 et le Césium 137.

En ce qui concerne le Tritium, il est intéressant de noter que les valeurs trouvées dans les études systématiques d'échantillons urinaires se situent à deux niveaux: mille picocuries par litre au maximum d'une part, plusieurs milliers de picocuries par litre d'autre part, selon que les personnes sont ou non porteuses de montres lumineuses au Tritium (30). La cause essentielle de contamination corporelle semble donc être, ici, le port de montres radioluminescentes. Il serait d'ailleurs utile d'évaluer la part, certainement non négligeable étant donné la labilité du Tritium, de la contamination des personnes de l'entourage des porteurs de ces montres. Si l'on tient compte de cette source de contamination, l'apport de Tritium par les centrales nucléaires devient négligeable devant elle, et l'irradiation annuelle correspondante est certainement inférieure au dixième de millirem.

La contamination corporelle en Césium 137 n'a cessé de décroître depuis l'arrêt des grandes séries de tests nucléaires. En 1972, elle est revenue à des activités de l'ordre de quelques milliers de picocuries par sujet. Les études systématiques (29) permettent de comparer la charge individuelle mesurée chez les travailleurs des centrales nucléaires d'une part et chez les individus du public; en 1972, les valeurs trouvées sont voisines de 4 000 picocuries dans les deux cas, montrant bien que, même pour cette catégorie de population pourtant réputée particulièrement exposée que constituent les travailleurs des centrales nucléaires, la différence n'est pas significative.

Enfin, en ce qui concerne le Strontium 90, actuellement la contamination moyenne des os humains est de l'ordre de 2 picocuries par gramme de Calcium (5). (Dans les os de lapins utilisés comme indicateurs (26), les mesures effectuées récemment en France par exemple montrent un maximum (25 pCi/g Ca) dans une station éloignée de tout centre nucléaire mais où l'on note une abondance particulière des retombées radioactives, et un minimum (5 pCi/g Ca) dans la station la plus exposée à la contamination d'origine industrielle).

5. CONCLUSION

L'influence des centrales nucléaires existantes sur la contamination du milieu et de l'homme demeure actuellement pratiquement indécelable. Il est bon de l'affirmer lorsque l'on entend les contre-vérités diffusées par certains dans le public, mais aussi lorsque l'on voit parfois certains spécialistes de radioprotection prendre eux-mêmes le parti d'un pessimisme systématique injustifié.

Il est néanmoins certain que le milieu aura à faire face à un accroissement notable des rejets radioactifs dans les années à venir. Face à ce risque potentiel qu'il ne serait raisonnable ni de sous-estimer ni de craindre exagérément, des structures de contrôle et de surveillance ont été définies, et, pour une large part, mises en place sous la responsabilité des autorités de santé publique. Compte tenu de la garantie que représente leur caution, on ne peut que se réjouir de l'immense progrès qui résultera de la production nucléaire d'énergie électrique (tout comme on apprécie les progrès que l'utilisation des rayons X a fait faire à la médecine, et ce malgré l'irradiation moyenne de 100 millirems par an qui en résulte pour l'ensemble de la population).

Si les structures actuelles avaient été en place dès le début du développement de l'utilisation médicale des rayons X, il est certain que l'irradiation moyenne des populations serait demeurée à un niveau beaucoup plus bas, sans diminution du bénéfice retiré de ces techniques.

Compte tenu des précautions prises dans leur domaine, l'irradiation moyenne provenant des centrales nucléaires demeurera par contre à des niveaux incomparablement plus bas, même si l'on prend en considération le développement important qui leur est promis dans un proche avenir.

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DISCUSSION

J.M.M.A. STOLZ: You refer to low doses and to the possibility of somatic and genetic repair. What do you mean by "low" doses and dose rates in this context? Do you have in mind values of the order of one mrem, one rem or 100 rem, for example, and how do those values stand in relation to the occupational exposures and exposures of the general public likely to accrue from the operation of nuclear power stations?

P. PELLERIN: It is certain that at exposures of under 10 rem — delivered as a single acute dose — no clinical signs of damage to the organism have ever been detected, even using the most advanced methods of examination. From 25 rem upwards it is possible in some cases to demonstrate certain changes in the blood count — but in the light of all the factors that can cause changes in the blood picture this may not be very significant. As for the dose beyond which genetic changes — I don't say genetic damage — may occur in man, this has been put at 80 rem.

When one considers that doses of 10-20 rem are unhesitatingly administered to patients on important fractions of the body in the course of a complete radiological examination of the gastro-intestinal tract, then the 1-2 mrem per year resulting from the operation of nuclear power stations can only be regarded as negligible — and that figure of 1-2 mrem, let us remember, is based on the most pessimistic assumptions regarding the behaviour and habits of the population.

M. J. A. DELPLA: I had already read with interst the text of Mr. Pellerin's paper distributed before the meeting, and I must say that I have now listened to this presentation with even greater attention.

He has referred to the courage of the authors of Report No. 151 published in 1958 by the World Health Organization. May I in my turn refer to the courage that he himself has displayed in taking a firm stand, in this forum, against the present general trend of opinion. To fall in line with that trend would have been all too easy.

Our problem is indeed primarily a psychological one. Everyone talks of "risks" and strives to reduce the millirem figures. There is of course good reason for this: in the population context, the calculations yield man-rems, which are so easy to convert into cancers and genetic deaths. However, this conversion is based on a certain assumption, namely the assumption that the effect is proportional to the dose, the effect being obtained by a few tens of rems — or more — administered over a very short period and calculated in terms of a few-tens of millirems — or less — received in the course of a year.

Those of us who are operators of nuclear power stations are ready, if called upon to do so, to spend large sums on reducing still further the radioactivity of our plant effluents. However, each successive reduction is more and more costly. And it is largely in vain: even then the public, with its ingrained fear of cancer and genetic effects, is not reassured. No real medical expert will grant the validity of the concept of proportionality in connection with the radiation levels emitted by nuclear power stations, or even levels much higher than that.

What we have to do then, is to banish this impression of insecurity and anxiety from the public mind.

I should like, in conclusion, to ask Mr. Pellerin for elucidation of a particular point. The estimates for medical irradiation dose in France are a good deal higher than in various other countries, at least according to the literature available on the subject, and it would be interesting to hear whether he has any comments to offer on this situation.

P. PELLERIN: Mr. Delpla is quite right in calling attention to the uncertainties that at present prevail in this sphere. Those uncertainties are due, among other things, to the fact that in most countries the estimates of medical exposure of the population are based on rather limited enquiries and even depend to a considerable extent on voluntary statements by radiologists. The civilized world is divided into two different areas or domains in this respect. In one area — the smaller one, including notably Scandinavia — practically all medical irradiation is conducted under tight supervision in hospitals (which tend, incidentally, to be model public health institutions in other respects as well). In the other area (which, I may say, includes the United States of America) there is a large and, let us face the fact, virtually uncontrollable private radiological practice. In the latter area which, as I say, is large and radiologically important, estimates of medical irradiation tend to disregard the existence of this substantial uncontrolled practice, and so, contrary to the situation in respect of nuclear power stations, evaluations of medical exposure are liable to yield figures that are on the optimistic side. When we remember that overall radiological practice is expanding at about 10% per year, then theoretically in 10 years' time the medical exposure dose could double — during a decade in which nuclear power station operators may well have had to spend vast sums on reducing the dose rates due to their releases from 2 mrem to 1 mrem per year!

I therefore consider it high time for the adoption of a realistic attitude to nuclear power station releases, a greater effort at reduction being applied, if at all, in the field of medical exposure where, as I have just indicated, it would not so much come amiss.

ОСНОВНЫЕ ЗАКОНОМЕРНОСТИ ПОСТУПЛЕНИЯ СТРОНЦИЯ-90 С ДИЕТОЙ И ОСОБЕННОСТИ УСВОЕНИЯ ИЗ НЕЕ ДАННОГО ИЗОТОПА В СССР В 1964-1971 г.г.

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Abstract–Аннотация

BASIC PATTERN OF ^{90}Sr UPTAKE WITH FOOD AND FEATURES OF ^{90}Sr EXTRACTION FROM THE DIET IN THE USSR DURING THE PERIOD 1964-71.

The uptake with the diet of ^{90}Sr from all sources by the urban population of the Soviet Union fell from 61 pCi/d in 1964 to 16 pCi/d in 1969; during the period 1970-71 there was no further decline in the degree of foodstuff contamination. This phenomenon is attributable to the fact that the degree of atmospheric contamination declined while the concentration of ^{90}Sr in the soil remained stable. By studying foodstuff contamination levels and dietary patterns it has been possible to determine the role of individual foodstuffs as conveyors of ^{90}Sr. In particular, it has been established that ^{90}Sr uptake by the Soviet population occurs mainly through the consumption of bread and related foodstuffs rather than milk – in contrast to the United States of America and other countries with a 'western' dietary pattern. The paper contains comparative data on ^{90}Sr uptake and accumulation in bone tissue for inhabitants of the USSR and a number of other countries. The degree of ^{90}Sr extraction from foodstuffs is evaluated by means of two indices: the rate of accumulation and the observed ratio. In the Soviet Union these indices are about 1.5 times lower than in countries with a 'western' type of diet; for example, the rate of accumulation increased from 23 to 78 over the period 1964-1971 in the Soviet Union and from 50 to 83 over the same period in the United States of America, while the observed ratio varied from 0.015 to 0.058 in the Soviet Union and from 0.054 to 0.084 in the United States of America. It has been established that there is a tendency for the difference in ^{90}Sr extraction from the diet to decrease – a tendency connected with the decline in the role of bread and related products as the main conveyors of ^{90}Sr in the Soviet Union and an increase in the role of milk and vegetables. The authors consider possible reasons why and ways in which eating habits affect the extraction of ^{90}Sr from the diet.

ОСНОВНЫЕ ЗАКОНОМЕРНОСТИ ПОСТУПЛЕНИЯ СТРОНЦИЯ-90 С ДИЕТОЙ И ОСОБЕННОСТИ УСВОЕНИЯ ИЗ НЕЕ ДАННОГО ИЗОТОПА В СССР В 1964-1971 гг.

Поступление стронция-90 глобального происхождения городскому населению Советского Союза с пищевым рационом упало с 61 пКи/сут в 1964 г. до 16 пКи/сут в 1969 г. В течение 1970-1971 годов дальнейшего снижения загрязненности рациона не происходило. Указанное явление объясняется падением роли воздушного загрязнения при стабильном содержании стронция-90 в почве. Изучение уровней загрязнения пищевых продуктов, а также структуры питания населения позволило выявить роль отдельных пищевых компонентов как поставщиков стронция-90. Установлено, в частности, что основное количество изотопа поступает населению СССР с хлебопродуктами, а не молоком, как это имеет место в США и других странах с "западным" типом питания. Приводятся данные по сравнительному поступлению и накоплению стронция-90 в костной ткани у жителей СССР и ряда других стран. Степень усвоения стронция-90 из рациона оценивается с помощью двух показателей: кратности накопления и наблюдаемого отношения. В СССР эти показатели примерно в 1,5 раза ниже, чем в странах с "западным" типом питания. Так, в СССР кратность накопления изменялась от 23 в 1964 г. до 78 в 1971 г., а в США соответственно – от 50 до 83. Наблюдаемое отношение изменялось в этот период соответственно по СССР в пределах 0,015-0,058, а в США – 0,054-0,084. Установлено, что имеется тенденция к сглаживанию отмеченной разницы в усвоении стронция-90. Это сглаживание коррелирует с падением в СССР роли хлебопродуктов как основных поставщиков изотопа и возрастанием роли молока и овощей. Рассмотрены возможные причины и механизмы влияния особенностей питания на усвоение радиоизотопа.

Ранее сообщалось о результатах и методах изучения уровней и закономерностей поступления стронция-90 населению Советского Союза [1,2,3]. Отмечалось также, что из распространенного в СССР рациона стронций-90 усваивался в заметно меньших количествах, чем из рациона так называемого "западного" типа, распространенного в США [4]. В настоящем сообщении на основании материалов собственных многолетних наблюдений и анализа литературных данных по поступлению с пищей и накоплению в скелете стронция-90 "глобального" происхождения предпринята попытка вскрыть некоторые закономерности и механизмы, которые могут определять степень перехода стронция-90 из диеты в организм. Вскрытие подобных закономерностей, очевидно, могло бы сделать более точным прогнозирование накопления стронция-90 в костной ткани при загрязнении рациона данным изотопом как глобального, так и любого другого происхождения.

Фактические материалы собственных исследований, использованные в работе, относятся к данным по поступлению стронция-90 населению Советского Союза, к роли отдельных компонентов рациона, как источников поступления изотопа, к данным по уровню поступления кальция, а также, частично, к содержанию стронция-90 в скелете жителей СССР. В остальных случаях использования фактических данных, в том числе для получения производных и относительных величин, приведены ссылки на литературный источник, откуда взяты исходные значения.

Уровни поступления стронция-90 с пищевым рационом населению Советского Союза, обусловленные стратосферными выпадениями, представлены в табл.I. Данные табл.I свидетельствуют о том, что в 1969 году поступление стронция-90 по сравнению с 1964 годом, когда загрязнение диеты было максимальным, снизилось примерно в 4 раза. С 1969 года дальнейшего снижения загрязненности диеты не происходило. Напротив, в 1970-71 г.г. было отмечено некоторое увеличение уровня стратосферных выпадений [5] и загрязнения диеты. Показательно, что за период 1964-69 г.г. выпадения стронция-90 из атмосферы упали в 10-20 раз[6, 7]. Менее выраженное снижение загрязненности диеты объясняется тем, что содержание стронция-90 в почве в последние годы стабилизировалось и "почвенный" стронций-90 стал играть все более заметную роль в загрязнении продовольствия. Так, на основании данных по уровням стронция-90 в почве, атмосферном воздухе и пищевых продуктах [5,8,9] было определено, что в 1964 г. хлебопродукты на 84% загрязнялись за счет стронция-90, непосредственно оседавшего на поверхность зерна из воздуха, в то время как к 1969 году доля "воздушного" стронция-90 упала до 47% (несколько поднявшись снова в 1970-71 г.г. до 58-52%). Пищевые продукты на протяжении всего периода наблюдения являлись основным, решающим источником проникновения стронция-90 в организм жителей. Поступление "глобального" стронция-90 с питьевой водой не превышало 5%, а с воздухом – 1-2% от поступления изотопа с пищевым рационом [1,3].

ТАБЛИЦА I. ПОСТУПЛЕНИЕ СТРОНЦИЯ-90 С РАЦИОНОМ ГОРОДСКОМУ НАСЕЛЕНИЮ СОВЕТСКОГО СОЮЗА В 1964-1971 г.г.. (в среднем на человека)

Годы	1964	1965	1966	1967	1968	1969	1970	1971
пКи/сут	61	43	29	21	19	16	18	18
пКи/г кальция	92	61	41	30	27	21	23	23

Некоторые собственные и литературные исходные данные, использованные в работе для вычисления показателей, характеризующих усвоение и накопление стронция-90 в организме человека, приведены в табл.II. Примечания к табл.II указывают источник и способ получения соответствующих показателей. Дополнительно следует отметить, что для суждения о степени усвоения стронция-90 из рациона и его накопления в организме (скелете) используются два показателя — наблюдаемое отношение (Н.О.), которое представляет собой отношение концентрации стронция-90 в рационе к его концентрации в скелете (концентрации выражены в пКи/г кальция), — а также кратность накопления (К.Н.), которая показывает, сколько суточных "порций" изотопа содержится во всем скелете. При этом для определения величины содержания стронция-90 в скелете жителей США показатель его средней концентрации на 1 г кальция умножался на общее содержание кальция в скелете, принятое равным, согласно данным по "стандартному" человеку, 1000 г [15].

При определении соответствующего показателя по Советскому Союзу было принято, согласно данным Б.К.Борисова, полученным на 20 скелетах взрослых людей, что содержание кальция в скелете составляет 870 г.

При анализе данных табл.II представляется целесообразным отметить существенно более высокое загрязнение пищевого рациона в СССР по сравнению с США. Указанная разница не связана с различным уровнем выпадений, или разным загрязнением исходного пищевого сырья. Оно обусловлено разницей в структуре питания и, прежде всего, традиционно более высоким в СССР потреблением хлебопродуктов, причем из более темных сортов муки, чем в США [1,3]. Поскольку основное количество стронция-90, особенно в период интенсивных выпадений, содержится в оболочечной части зерна, темные сорта муки всегда содержат значительно больше стронция-90, чем высшие сорта, изготовленные из зерна аналогичной загрязненности. Следует добавить также, что особенно большое распространение хлебопродукты из темной муки имели в 1964 году [3,4].

В табл.III приведены данные, которые иллюстрируют относительную величину поступления и накопления стронция-90 в СССР и США. Материалы таблицы указывают на то, что поступление стронция-90 в СССР в течение ряда лет было в 1,5-2 раза выше, чем в США, а концентрация в костной ткани жителей была также выше, чем в США, однако не пропорционально превышению загрязненности рациона. Это несоответствие, наиболее резко выраженное в начальный период наблюдения

ТАБЛИЦА II. ИСПОЛЬЗОВАННЫЕ ДЛЯ РАСЧЕТОВ ДАННЫЕ ПО ПОСТУПЛЕНИЮ С РАЦИОНОМ СТРОНЦИЯ-90 И ЕГО СОДЕРЖАНИЮ В КОСТНОЙ ТКАНИ ВЗРОСЛОГО ГОРОДСКОГО НАСЕЛЕНИЯ СССР и США в 1964-71 годы

Годы		1964	1965	1966	1967	1968	1969	1970	1971
Стронций-90 в рационе; пКи/рацион	СССР	61	43	29	21	19	16	18	18
	США	26	24	18	13	14	12	12	13
Стронций-90 в рационе; пКи/г Са	СССР	92	61	41	30	27	21	23	23
	США	24	22	16	12	14	12	12	13
Стронций-90 в костной ткани; пКи/г Са	СССР	1,5	1,6	1,5	1,35	1,2	1,2	1,4	1,3
	США	1,3	1,1	1,05	0,9	1,1	1,1	1,0	-
Стронций-90 во всем скелете; пКи/скелет	СССР	1200	1300	1200	1100	960	960	1100	1000
	США	1300	1100	1050	900	1100	1100	1000	-

Примечания:
1. Данные по содержанию стронция-90 в рационе и костной ткани жителей США (города Нью-Йорк, Чикаго, Сан-Франциско) рассчитаны по материалам работ [10, 11, 12].
   Данные по США за 1968-1970 г.г. относятся только к Нью-Йорку.
2. Показатели содержания стронция-90 в костной ткани населения СССР представлены по материалам работ А.Н. Марея и др. [13,14]. За 1967 год концентрация изотопа в скелете определена как средняя величина между 1966 и 1968 г.г.
3. Данные по содержанию кальция в рационе жителей СССР – из работ [1-4].

ТАБЛИЦА III. СООТНОШЕНИЕ УРОВНЕЙ ПОСТУПЛЕНИЯ СТРОНЦИЯ-90 С РАЦИОНОМ И КОНЦЕНТРАЦИЙ ИЗОТОПА В КОСТНОЙ ТКАНИ ЖИТЕЛЕЙ СССР И США

Годы	1964	1965	1966	1967	1968	1969	1970
$\frac{\text{рацион СССР; пКи}}{\text{рацион США; пКи}}$	2,34	1,79	1,61	1,61	1,36	1,34	1,50
$\frac{\text{костная ткань в СССР; пКи/г Ca}}{\text{костная ткань в США; пКи/г Ca}}$	1,15	1,45	1,43	1,50	1,09	1,09	1,40
$\frac{\text{костная ткань; пКи/г Ca}\ \frac{\text{СССР}}{\text{США}}}{\text{рацион; пКи}\ \frac{\text{СССР}}{\text{США}}}$	0,49	0,81	0,89	0,93	0,80	0,82	0,93

(1964 год), совпадающий с периодом максимальных выпадений, затем существенно сгладилось, хотя и продолжает закономерно прослеживаться в течение всего периода наблюдений.

В качестве причин несоответствия между поступлением стронция-90 с рационом и его концентрацией в костной ткани могут выступать или факторы, связанные с особенностями диеты и формой нахождения в ней стронция-90 (влияние на усвоение изотопа) или же то обстоятельство, что различна средняя масса скелета и, следовательно, количество кальция, которое участвует в "разбавлении" усвоенного стронция-90 и, тем самым, отражается на значениях его концентраций в скелете (влияние на показатели накопления). В свою очередь, к числу факторов, связанных с особенностями диеты, можно, очевидно, отнести следующее:

- уровень содержания в диете кальция;
- наличие веществ, прежде всего фитиновых соединений, способных связывать стронций-90 (и кальций) в желудочно-кишечном тракте и, тем самым, снижать его усвоение;
- более затрудненное усвоение из диеты "воздушного" стронция-90, т.е. попавшего на продовольствие непосредственно из воздуха, по сравнению с "невоздушным", т.е. попавшим туда из почвы, или из воздуха, но опосредованно, пройдя предварительно в организм сельскохозяйственных животных (например, стронций-90 в молоке).

К "невоздушному" стронцию-90 мы условно относим здесь все виды структурного, неповерхностного, загрязнения, включая сюда загрязнение изотопом, проникшим внутрь растений не из почвы, а с загрязненной поверхности листьев.

Для того, чтобы подойти к характеристике возможной роли особенностей диеты и к оценке влияния указанных выше факторов на усвоение изотопа, в таблицах IV и V приведены материалы, позволяющие отметить некоторые существенные особенности пищевого рациона и его отдельных компонентов, как "поставщиков" стронция-90, в СССР и США.

ТАБЛИЦА IV. ВКЛАД ОТДЕЛЬНЫХ КОМПОНЕНТОВ РАЦИОНА В ОБЩЕЕ ПОСТУПЛЕНИЕ СТРОНЦИЯ-90 ГОРОДСКОМУ НАСЕЛЕНИЮ СОВЕТСКОГО СОЮЗА В 1964-71 г.г. (в среднем по стране; в % от поступления со всем рационом)

Годы	Хлебопродукты	Молоко и молочные продукты	Картофель	Мясо и мясопродукты	Прочие
1964	60,4	15,7	6,7	4,0	13,2
1965	59,0	15,6	7,1	4,0	14,3
1966	48,8	17,4	8,5	5,0	20,3
1967	40,4	17,1	10,6	6,2	25,7
1968	33,7	17,5	11,7	5,9	31,2
1969	35,9	20,5	12,2	7,1	24,3
1970	34,3	19,4	13,1	7,4	25,8
1971	35,6	20,1	12,6	7,5	24,2

ТАБЛИЦА V. ВКЛАД ОТДЕЛЬНЫХ КОМПОНЕНТОВ РАЦИОНА В ОБЩЕЕ ПОСТУПЛЕНИЕ СТРОНЦИЯ-90 НАСЕЛЕНИЮ США в 1964-1970 годы (в % от общего поступления) [16] *

Годы	Молочные продукты	Зерновые	Овощи	Фрукты	Мясо, рыба, яйцо
1964	43	34	13	7	4,5
1965	44	26	18	8	5,5
1966	36	28	20	8	7
1967	32	20	28	14	5,5
1968	32	19	27	18	5
1969	34	20	26	15	4,5
1970	34	21	27	14	4,5

* В среднем населению по городам Нью-Йорк и Сан-Франциско.

В частности, материалы таблиц IV и V позволяют отметить, что в СССР основное количество стронция-90 поступает населению с хлебопродуктами, а в США – с молочными продуктами. В течение всего периода наблюдений хлеб в СССР имеет большее значение, чем в США, что объясняется его относительно большим потреблением и привычкой населения к включению в рацион ржаного и темных сортов пшеничного хлеба. Материалы таблиц IV и V, а также приведенные выше данные по характеристике загрязненности темных и светлых сортов хлеба и их распространенности в СССР и США позволяют считать, что в США основное количество или даже практически весь стронций-90 в диете был представлен структурным, "невоздушным" изотопом, в то время как в СССР, где значительная часть стронция-90, особенно в первые годы, поступала с темными сортами хлебопродуктов, заметная доля изотопа в рационе неизбежно была непосредственно воздушного происхождения. Эта доля изотопа в диете СССР была рассчитана и ниже будет приведена в целях определения возможной разницы в доступности для усвоения структурного и воздушного стронция-90.

В таблицах VI и VII представлены материалы, характеризующие усвоение и накопление стронция-90 в СССР и США, причем в табл. VII приведены данные, учитывающие содержание в диете лишь "невоздушного" стронция-90 (в целях проверки допущения, что "воздушный" стронций-90 не усваивается или усваивается хуже).

Материалы табл. VI указывают на возрастание со временем показателей К.Н. и Н.О., как в СССР, так и в США. Следует отметить, что увеличение данных показателей объясняется не возрастанием содержания стронция-90 в скелете, а падением загрязненности рациона. Со временем разница между показателями К.Н. и Н.О. в СССР и США сглаживалась, что свидетельствует о влиянии каких-то факторов, изменявшихся со временем. К числу таких факторов могут относиться какие-то элементы диеты и явно не относится влияние разбавления стронция в большей массе костного кальция у жителей США, поскольку, хотя сам факт влияния данного фактора очевиден, масса кальция в скелете является величиной весьма стабильной и не может объяснить отмеченной динамики в соотношении показателей К.Н. и Н.О. Величины, приведенные в последней, нижней строке табл. VI, указывают на то, что между показателями Н.О. в СССР и США существует большая разница, чем между показателями К.Н. Причина этого явления лишь отчасти может объясняться большим количеством кальция в скелете американцев, поскольку эта разница не столь значительна (870 г и 1000), как разница показателей в 1964-1967 г.г.

ТАБЛИЦА VI. ПОКАЗАТЕЛИ, ХАРАКТЕРИЗУЮЩИЕ УСВОЕНИЕ И НАКОПЛЕНИЕ СТРОНЦИЯ-90 В СССР И США в 1964-1970 годы

Годы		1964	1965	1966	1967	1968	1969	1970
Кратность накопления (К.Н.)	СССР	20	30	41	52	51	60	61
	США	50	46	58	69	78	92	83
	$\frac{\text{СССР}}{\text{США}}$	0,40	0,65	0,71	0,75	0,65	0,65	0,74
Наблюдаемое отношение (Н.О.)	СССР	0,016	0,026	0,037	0,045	0,045	0,057	0,058
	США	0,054	0,050	0,066	0,075	0,078	0,092	0,088
	$\frac{\text{СССР}}{\text{США}}$	0,30	0,52	0,56	0,60	0,58	0,62	0,66
$\frac{\text{Н.О. в СССР}}{\text{Н.О. в США}} : \frac{\text{К.Н. в СССР}}{\text{К.Н. в США}}$		0,75	0,80	0,79	0,80	0,89	0,96	0,89

ТАБЛИЦА VII. ПОКАЗАТЕЛИ ПОСТУПЛЕНИЯ И НАКОПЛЕНИЯ У ЖИТЕЛЕЙ СССР СТРОНЦИЯ-90 "НЕВОЗДУШНОГО" ПРОИСХОЖДЕНИЯ

Годы	1964	1965	1966	1967	1968	1969	1970	1971
Поступление с рационом; пКи/сут	30	25	21	17	16	13	14	14
Кратность накопления (К.Н.) в СССР	40	52	57	65	60	74	79	72
Поступление с рационом; пКи/г Са	45	35	30	24	23	17	18	18
Наблюдаемое отношение (Н.О.) в СССР	0,033	0,046	0,050	0,056	0,052	0,071	0,078	0,072
$\frac{\text{К.Н. в СССР}}{\text{К.Н. в США}}$	0,80	0,13	0,98	0,94	0,77	0,81	0,95	-
$\frac{\text{Н.О. в СССР}}{\text{Н.О. в США}}$	0,61	0,92	0,76	0,75	0,67	0,77	0,89	-
$\frac{\text{Н.О. СССР/США}}{\text{К.Н. СССР/США}}$	0,77	0,81	0,78	0,80	0,87	0,95	0,94	-

ТАБЛИЦА VIII. НЕКОТОРЫЕ ИТОГОВЫЕ ПОКАЗАТЕЛИ, ХАРАКТЕРИЗУЮЩИЕ УСВОЕНИЕ И НАКОПЛЕНИЕ СТРОНЦИЯ-90 В СССР И США

Годы	1964	1965	1966	1967	1968	1969	1970
$\frac{\text{К.Н. в СССР}}{\text{К.Н. в США}}$ "общего" стронция-90	0,40	0,65	0,71	0,75	0,65	0,65	0,74
$\frac{\text{К.Н. в СССР}}{\text{К.Н. в США}}$ "невоздушного" стронция-90	0,80	1,13	0,98	0,94	0,77	0,81	0,95
Процент "невоздушного" стронция-90 в рационе	50	58	72	81	81	84	78
$\frac{\text{Н.О. в СССР}}{\text{Н.О. в США}}$	0,30	0,52	0,56	0,60	0,58	0,62	0,66
Кальций в рационе $\frac{\text{СССР}}{\text{США}}$	0,61	0,65	0,63	0,65	0,70	0,76	0,78
$\dfrac{\text{Костная ткань; пКи/г Ca } \frac{\text{СССР}}{\text{США}}}{\text{Рацион; пКи } \frac{\text{СССР}}{\text{США}}}$	0,49	0,81	0,89	0,93	0,80	0,82	0,93
$\frac{\text{Н.О. в СССР}}{\text{Н.О. в США}} : \frac{\text{К.Н. в СССР}}{\text{К.Н. в США}}$	0,75	0,80	0,79	0,80	0,89	0,96	0,89

Очевидно, определенное значение здесь имеет факт более высокого поступления американцам кальция с рационом. Как известно, кальций способен снижать усвоение стронция-90 [4], однако, как видно из соотношения показателей К.Н. и Н.О. в последней строке табл.VI, не прямо пропорционально его количеству в рационе, а заметно слабее.

Материалы таблиц VII и VIII позволяют заключить, что при допущении, согласно которому "воздушный" стронций-90 не усваивается, разница между СССР и США оказывается менее выраженной. Сглаживание разницы показателей К.Н. со временем по "общему" стронцию-90, которое происходит одновременно с падением вклада "воздушной" компоненты в загрязненность рациона также указывает на связь последнего обстоятельства с усвоением стронция-90. Все это указывает на справедливость предположения о худшем усвоении из рациона "воздушного" стронция-90. Однако и этот фактор не объясняет полностью различий в усвоении стронция-90 между СССР и США, прежде всего в 1964 году. Мы полагаем, что в этот период в СССР усвоение стронция-90 из рациона тормозилось также вследствие повышенного потребления хлебопродуктов из темных сортов муки, которые богаты фитиновыми соединениями, способными связывать в желудочно-кишечном тракте кальций и стронций-90.

Таким образом, анализ материалов по поступлению и накоплению стронция-90 у жителей СССР и США позволяет отметить, что особенности диеты способны существенным образом влиять на показатели усвоения и накопления стронция-90 из рациона. На этих показателях сказывается также тот обычно неучитываемый факт, что среднее общее содержание кальция в скелете жителей разных стран существенным образом варьирует.

ВЫВОДЫ

1. Поступление стронция-90 с диетой населению Советского Союза в 1969 году по сравнению с 1964 годом – периодом максимального поступления – упало в 4 раза. В 1970-71 г.г. дальнейшего снижения загрязненности рациона не происходило.

2. В период 1963-1971 г.г. роль отдельных компонентов рациона, как "поставщиков" стронция-90, существенно изменилась, причем значение хлебопродуктов упало, а молока, мяса – выросло.

3. Изменения значимости отдельных компонентов рациона как поставщиков стронция-90 обусловлены, главным образом, снижением "воздушной" компоненты загрязненности диеты и возрастанием роли "почвенной".

4. Показатели усвоения стронция-90 из распространенной в СССР диеты (кратность накопления в организме и наблюдаемое отношение "скелет-рацион") существенно отличаются от таковых в странах с "западным" типом питания (США), причем имеется тенденция к сглаживанию отмеченной разницы.

5. На показатели усвоения стронция-90 из диеты и его накопления в организме способны влиять такие факторы, как характер загрязнения продовольствия стронцием-90 (структурное, или поверхностное), количество фитиновых соединений в диете (вклад темных сортов муки), количество кальция. Определенное влияние на широко употребляемый показатель наблюдаемого отношения оказывает также общее содержание кальция в скелете.

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THE US ATOMIC ENERGY COMMISSION PROGRAM FOR MONITORING THE BEHAVIOUR OF RADIONUCLIDES RELEASED TO THE ENVIRONMENT

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Abstract

THE US ATOMIC ENERGY COMMISSION PROGRAM FOR MONITORING THE BEHAVIOUR OF RADIONUCLIDES RELEASED TO THE ENVIRONMENT.

The US Atomic Energy Commission (AEC) conducts major nuclear energy programs at about 30 sites. The environmental monitoring programs at these sites are conducted by AEC or its operation contractors and are governed by general requirements set forth in AEC management directives. The objective of these directives is to ensure that environmental monitoring is adequate to determine the extent to which levels of radioactivity released from AEC facilities comply with applicable environmental quality standards and to evaluate the effects, if any, of planned and unplanned releases on the environment. Emphasis has been placed on the monitoring of pathways of human exposure, although environmental media such as soil and sediment have been monitored to detect significant trends in radioactivity levels. In some instances direct gamma body counts on members of the public and hypothetical dose estimates for selected individuals and population groups have been made to define more accurately the risks of public exposure in the vicinity of AEC facilities.

Environmental monitoring programs have generally demonstrated public exposures in the vicinity of AEC facilities to be less than one per cent of the ICRP recommended standards. Some operations, however, have resulted in potential public exposures representing somewhat larger fractions of the ICRP recommended standards. The levels of exposure that resulted from these and other AEC operations will be discussed. With few exceptions, exposures to individuals and groups in the population have not ranged above a few per cent of the ICRP standards.

Comprehensive soil and sediment monitoring is being accomplished in the vicinity of a number of AEC facilities and sites to determine the degree of build-up of plutonium in the environment due to routine and accidental releases. There is no evidence that build-up of plutonium in soil and other media present a public health hazard. The quantities of plutonium released and the levels of plutonium detected in the environment will be discussed.

1. INTRODUCTION

Environmental monitoring programs of varying scope and complexity are conducted at approximately 30 U.S. Atomic Energy Commission (AEC) installations located throughout the United States. These installations are operated for the AEC by universities and industrial firms commonly referred to as AEC contractors. There is great diversity in the activities conducted at these sites, in the site environs and in the types and quantities of radionuclides potentially available for release to the environment. Thus, environmental monitoring needs differ greatly from site to site.

Although the scope of this paper is limited to the monitoring practices at AEC sites, the AEC also regulates the radioactivity emission control and monitoring practices of privately owned and operated facilities such as AEC-licensed nuclear power reactors and nuclear fuel reprocessing plants. This licensing and regulatory authority encompasses effluent monitoring and preoperational and routine environmental monitoring practices to assure compliance with applicable AEC regulations and requirements. In addition to the preoperational and routine environmental monitoring programs conducted by the AEC licensees, the AEC and U.S. Environmental Protection Agency (EPA) and the state within which an AEC-licensed facility is located, conduct special surveys, independently and cooperatively, to provide further assurance that the public and environment are adequately protected.

The environmental monitoring programs at AEC sites are conducted by AEC or its operating contractors and are governed by general requirements set forth in an AEC management directive. The purpose of the AEC directive is to assure that environmental monitoring is adequate to determine the degree of compliance with applicable environmental radiation, air and water quality standards, and to evaluate the effects, if any, of planned and unplanned releases on the environment and the public. In some instances independent environmental radiation monitoring surveys are conducted in the vicinity of AEC sites by state and local agencies and by the EPA.

The concentrations of radionuclides in the environs of AEC sites and the associated exposures to the public have generally been a few percent or less of the appropriate population exposure standards expressed as annual dose limits or annual average concentrations in air and water. AEC standards for individuals in the public are one-tenth of the International Commission on Radiological Protection (ICRP) continuous exposure limits for radiation workers and, for the general public, are one-thirtieth of the ICRP continuous exposure limits for radiation workers.

2. AEC ENVIRONMENTAL MONITORING AND REPORTING REQUIREMENTS

The environmental monitoring requirements for AEC sites apply to both radioactive and nonradioactive pollutant materials although the latter materials are not discussed in this paper. The requirements call for environmental monitoring as appropriate to (a) verify the adequacy of effluent and operational controls, (b) determine compliance with applicable standards, and (c) detect environmental trends related to site operations. The basic objective of these requirements is to assure that AEC operations are so conducted that the public and environment are protected. The environmental monitoring program, along with other data systems such as stack monitoring, provides information feedback on the adequacy of operational practices and controls for minimizing, treating and disposing of radioactive wastes. A secondary objective is to provide a data and information base adequate for determining that radiation standards are met and for informing AEC management and the public of effects, if any, that AEC operations have on the public and environment. Environmental monitoring data have proved to be vital to the preparation of responsive replies to public inquiries and expressions of concern regarding possible environmental effects of AEC operations. Each contractor is expected to develop and conduct an environmental monitoring program consistent with these AEC objectives. The environmental monitoring program at each site is periodically evaluated by AEC health and safety personnel to assure that objectives are met.

Each site required to have an environmental monitoring program also prepares and distributes an annual report describing the monitoring program, and summarizing and interpreting the results. General guidance is provided for report format and content (see Appendix). The reports are widely distributed to local, state and Federal health and environmental protection agencies and to the public. In addition, each report is summarized in an issue of the EPA monthly publication, ''Radiation Data and Reports.'' In 1971, environmental monitoring reports were prepared and distributed for 27 AEC sites.

3. MONITORING PRACTICES AT AEC SITES

3.1 General

In the first decade or more following the birth of the atomic energy program in the U.S., environmental monitoring was considered necessary, yet there was little experience in how to conduct an adequate program. Generally, most varieties of animals, plants, produce and other environmental media in the vicinity of the site were sampled or measured. As experience was gained in conducting and interpreting results of environmental monitoring programs, the emphasis shifted to the sampling of media that are sensitive indicators of human food chain contamination and accumulation of radionuclides in the environment. At the same time, greater reliance was being placed on effluent monitoring with environmental monitoring providing confirmatory evidence that standards were being met.

In the past few years, with the heightened interest in the quality of the environment, it has not been sufficient to simply demonstrate that radiation concentration and dose standards have been met; it has been necessary also to define the potential exposures to the public and impact on the environment in the vicinity of nuclear installations. This necessitates the measurement of environmental levels of specific radionuclides and potential public exposure down to background levels, including the levels of radioactivity in soil and other media not directly related to routes of human exposure. Even the presence of detectable levels of man-made radionuclides in environmental media has become an area of major concern for some sectors of the public. For example, it has been necessary for AEC contractors to do extensive soil sampling to determine the cumulative deposition of persistent radionuclides in offsite areas. The emphasis that is being placed on soil sampling at AEC sites is evidenced in the establishment of soil sampling programs at 13 AEC sites in the past 4 years. The assessment, however, of the significance of soil contamination is difficult, since radionuclide levels in soil are not directly translatable to potential human exposure. Soil contamination data are considered only as indicators of long-term trends. Radionuclide levels in media (air, water, etc.) more directly related to human exposure are used to evaluate potential exposure to the public.

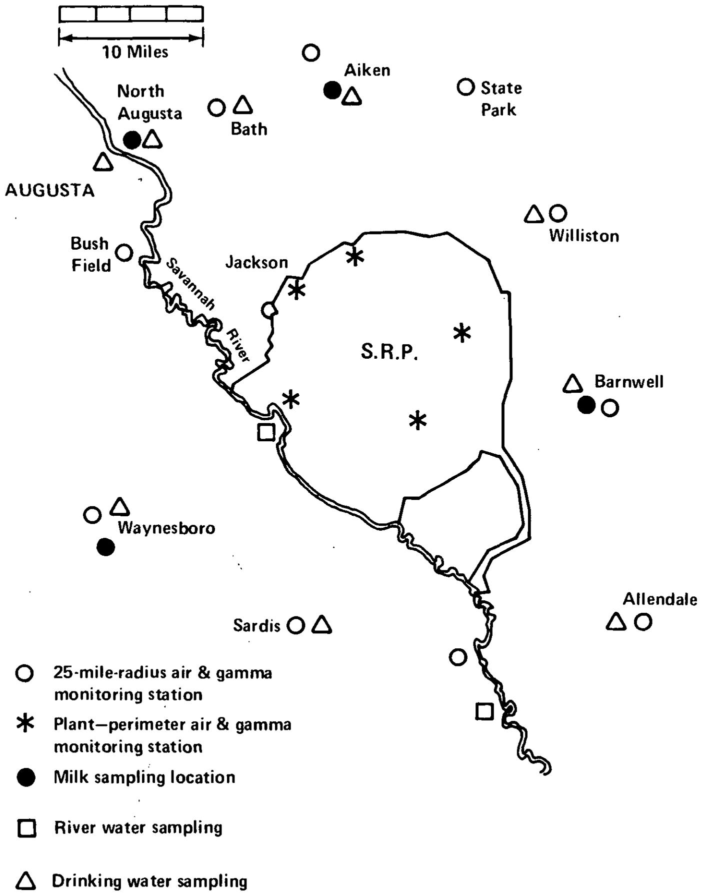
With the increased emphasis on the environment, the AEC's policy on the control of radioactivity releases from AEC sites has shifted from primarily an effort to restrict public exposure to some small fraction of applicable dose standards, to reducing releases still further to achieve the lowest levels feasible, considering cost, benefit and availability of control technology. With the decreasing quantities of radioactivity in effluents, the task of monitoring environmental media and estimating public exposure has become increasingly more difficult. Monitoring techniques and equipment must be capable of collecting larger and more representative samples and analytical procedures must be sufficiently sensitive to permit the identification and quantification of radionuclides at these diminishing levels in the vicinity of AEC sites. The normal fluctuations in background levels of radionuclides in the environment further compound the task of assessing the decreasing increments of such radionuclides released from AEC sites. For some radionuclides, particularly those that do not persist or concentrate in environmental media (i.e., noble gases), environmental levels are generally so low they must be calculated using empirical models.

TABLE I. ENVIRONMENTAL MEDIA AND LOCATIONS MONITORED AT SEVERAL AEC SITES

	ENVIRONMENTAL MEDIA MONITORED AND NUMBER OF MONITORING LOCATIONS									
AEC SITE	AIR	WATER	SOIL	FOOD	MAN	CROP PLANTS	FISH & WILDLIFE	AMBIENT RADIATION	VEGETATION	SEDIMENT
ARGONNE NATIONAL LABORATORY	12	6	50	1				16	7	
BROOKHAVEN NATIONAL LABORATORY	4	17	12	2			5	4	12	14
HANFORD SITE	23	11	13	4			3	2	13	
LAWRENCE LIVERMORE LABORATORY	16	24	59	2				52	9	
LOS ALAMOS SCIENTIFIC LABORATORY	27	10	9				3	22		9
NATIONAL ACCELERATOR LABORATORY		8						1		
NATIONAL REACTOR TEST STATION	8	12		10		7		11		
NEVADA TEST SITE	104	99		41	60			97		
OAK RIDGE SITE	17	10	9	1			1		5	
ROCKY FLATS PLANT	33	46	75						75	6
SAVANNAH RIVER PLANT	15	16		20		14	4	15	14	

3.2 Usual Monitoring Practices

Because of the varying operational and environmental conditions from site to site, it has not been practical for the AEC to adopt standard monitoring practices and procedures to be followed at each site, although consideration is being given to the development of guidelines for certain practices (e.g., soil sampling) that can be made more uniform. As a result of the good communications among the various AEC and contractor organizations, many monitoring practices and procedures have been widely and uniformly adopted. The offsite media most commonly sampled in the vicinity of AEC sites fall into two groups: one including air, water, foodstuffs and ambient gamma radiation which directly relate to human exposure and established radiation standards; and a group including soil, vegetation and sediment which are indicators of the accumulation of radionuclides in the environment and for which there are no radiation standards. Table I indicates the types of environmental media



Four 100-mile-radius air and gamma monitoring stations not shown.

FIG. 1. Sample collection points at the AEC's Savannah River Plant (SRP).

TABLE II. CHARACTERISTICS OF THE AERIAL RADIOLOGICAL MEASURING SYSTEM (ARMS)

MEASUREMENT	CHARACTERISTICS
GROUND POSITION ACCURACY	DATA POINTS PLOTTED WITHIN ±0.001° (LONGITUDE AND LATITUDE)
DETECTABLE DIFFERENCES IN REPEAT SURVEYS OVER LARGE AREAS (625 Square miles)	0.75 μ R/h AT 3 ft FOR A ONE SIGMA LEVEL OF CONFIDENCE
MINIMUM DETECTABLE GROUND CONCENTRATION	0.1 μ Ci/m^2 (^{60}Co AS AN EXAMPLE)
EXPOSURE RATE CONVERSION	392 counts/s AT 500 ft TERRAIN CLEARANCE REPRESENTS 1 μ R/h AT 3 ft
GAMMA-RAY SPECTRAL RESOLUTION	8.5% AT 1.00 MeV (FULL WIDTH AT HALF MAXIMUM)

routinely monitored for radioactivity and the number of sampling locations per type of media at each of 11 major AEC sites. The radionuclides for which these various media are most commonly analyzed are strontium-90, cesium-137, iodine-131, tritium and plutonium-239. Although the radioactive noble gases (e.g., argon-41) generally cannot be detected in the ambient air, they are of considerable interest since they contribute the major portion of the offsite exposure in the vicinity of several AEC sites.

The environmental monitoring program at the AEC's Savannah River Plant (SRP) is illustrated in Figure 1 as an example of environmental monitoring in the vicinity of AEC sites. Typical of most AEC sites, the SRP operates peripheral, intermediate and distant (background) air, water, and gamma monitoring stations and water monitoring stations upstream and downstream of the site.

The frequency with which various environmental media are sampled and analyzed is as dependent on the nature of the media as on the characteristics of the radionuclide(s) of interest. The dynamic systems, such as streams, ambient air, milk, aquatic biota and wildlife, are continuously or frequently monitored or sampled regardless of the physical decay rate of the radionuclides of interest. However, compositing of samples over a long period of time is often practiced when long-lived radionuclides are measured. The more static systems, such as lake water, soil and sediment, are less frequently sampled and analyzed unless the radionuclide of interest has a short half-life.

Normally, environmental monitoring at AEC sites begins at least 1 year prior to startup of nuclear activities and continues until the site has been fully decommissioned, including all necessary decontamination. Thus, in the active lifetime of a given site, the environmental monitoring program will proceed through three phases; preoperational, operational and postoperational. In addition, a complete radiation survey is required prior to release of any AEC facility for uncontrolled or public use.

3.3 Special Programs and Surveys

At times, it is necessary to augment the routine effluent and environmental monitoring programs conducted in the vicinity of nuclear installations by special radiation surveys. Such surveys make use of AEC laboratories and aerial monitoring capabilities and the services of independent laboratories within state and local agencies and the EPA.

3.3.1 AEC Aerial Radiological Measuring Program

The AEC maintains an Aerial Radiological Measuring System (ARMS) as an around-the-clock accident response capability. In order to provide this continuous response capability, it is necessary to maintain the airplane, specialized instrumentation, pilots, technicians, navigators, and data processing specialists in a high state of readiness. Through the continued utilization of ARMS for large land area surveys and frequent special surveys, resource maps and documents are kept up to date and the overall proficiency of the system is maintained.

The ARMS capability includes highly sensitive instrumentation for gross gamma radiation measurement and spectral analysis, air sampling, meteorological conditions, airplane position data, general purpose and multispectral photography in black and white and color, infrared scanning, and scientific

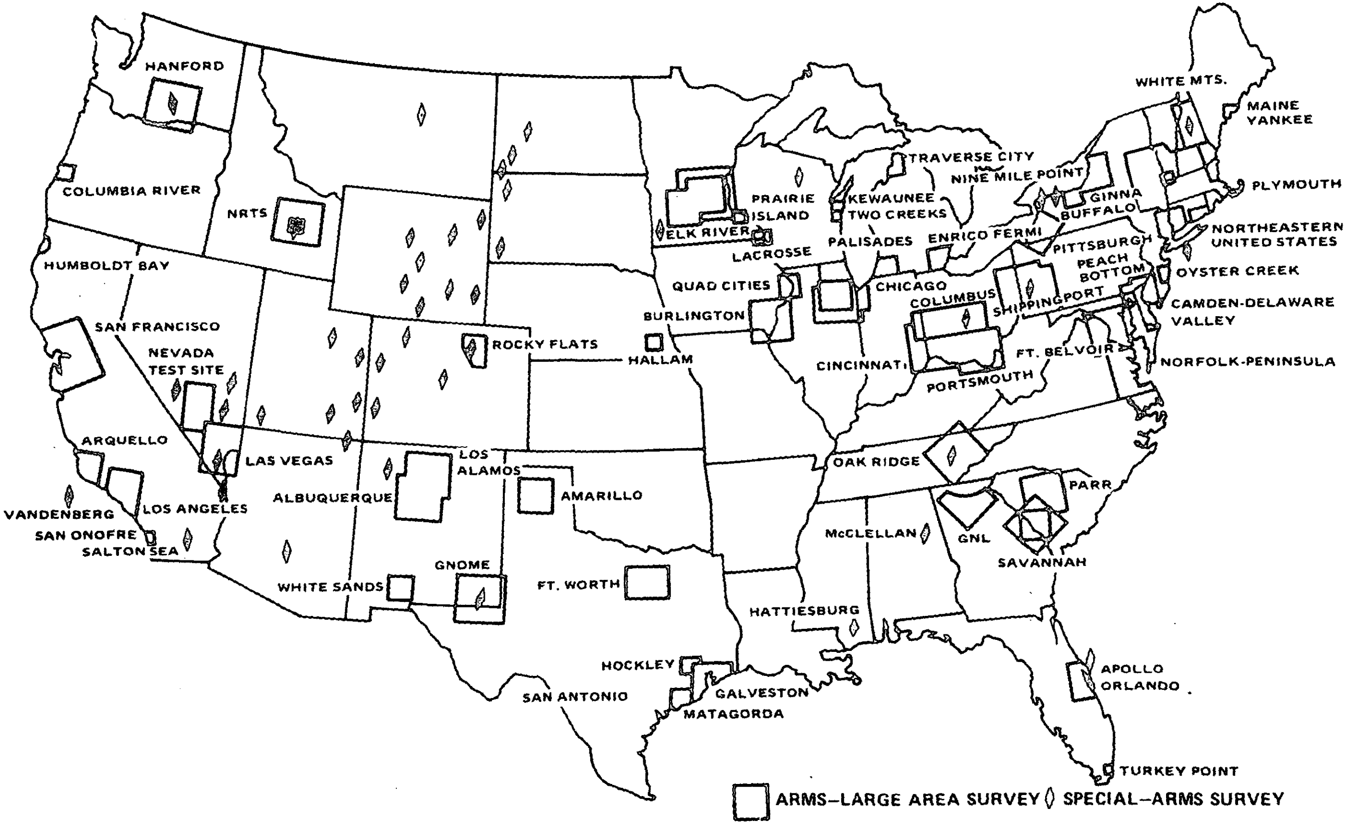
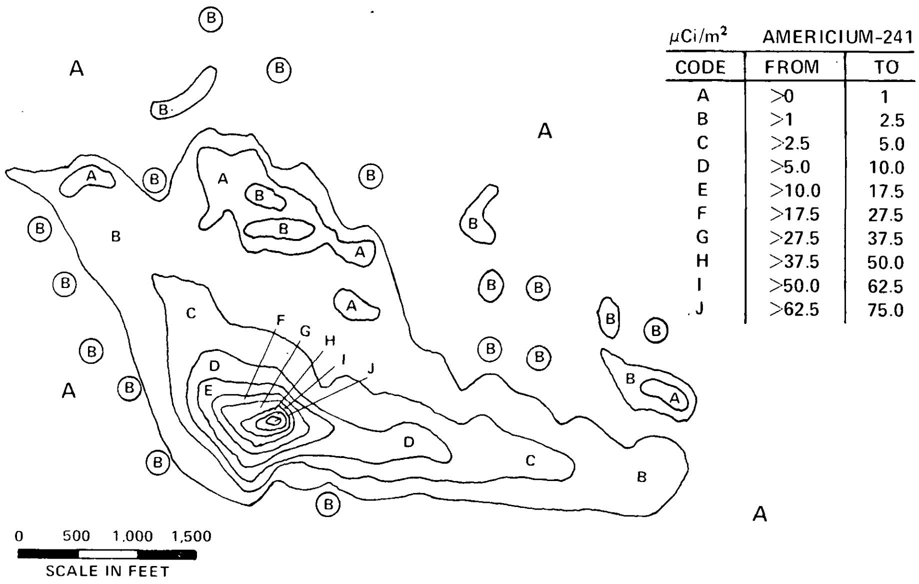


FIG.2. Surveys performed by AEC's aerial radiological measuring system (ARMS).



$\mu Ci/m^2$	AMERICIUM-241	
CODE	FROM	TO
A	>0	1
B	>1	2.5
C	>2.5	5.0
D	>5.0	10.0
E	>10.0	17.5
F	>17.5	27.5
G	>27.5	37.5
H	>37.5	50.0
I	>50.0	62.5
J	>62.5	75.0

FIG.3. Aerial radiological survey by helicopter for ^{241}Am gamma emissions.

backup in the application of this system to a wide variety of radiological measurements. Table II summarizes the characteristics of the standard ARMS instrumentation.

ARMS was designed, developed and is operated for the AEC by EG&G, Inc., Las Vegas, Nevada. In addition to background surveys that have documented more than 10 percent of the nation's land area (see Figure 2), ARMS has been used for locating lost radiation sources and investigating special contamination situations. A source search that led to the development of an expanded capability was the location by ARMS of a test missile that malfunctioned and landed in Mexico. The missile payload included as part of the instrumentation two cobalt-57 sources of approximately 400 mCi each. The ARMS instrumentation was successfully adapted to respond to the low energy cobalt source which led to development of a capability for detection of americium-241 found in conjunction with plutonium-239. Figure 3 shows the americium-241 fingerprint from a single nonnuclear plutonium dispersal test as detected in a helicopter survey at the Nevada Test Site.

In June 1972 the ARMS airplane was used in a typical mission, to perform an aerial radiological survey of the AEC's Lake Ontario Ordnance Works to determine residual levels of radium-226 in onsite areas that had been used for storage of wastes from uranium processing. Figure 4 depicts the site and vicinity, the dose contours, and the waste storage areas. The raw data from gross gamma count and gamma spectral measurements were recorded on paper tape to permit computer processing. From calibration measurements over a land area of uniformly dispersed natural radioactivity, the altitude dependence of the gross gamma count rate was determined and a conversion factor computed. Using this conversion factor and the survey altitude data, the raw gross count rate was converted to exposure rate in microroentgens per hour at 3 feet above ground, the ''terrestrial exposure rate.''

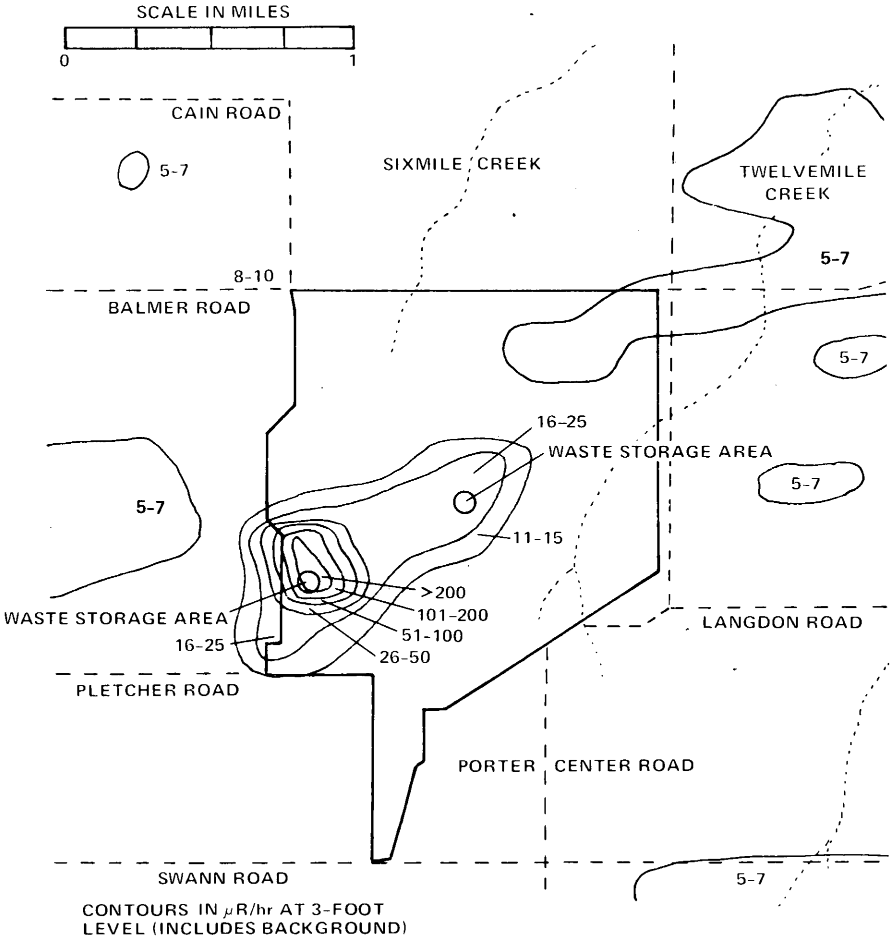


FIG. 4. Aerial radiological survey of AEC's Lake Ontario ordnance works.

Recently, the ARMS airplane was used to survey radiation levels in the vicinity of the AEC's Shippingport Atomic Power Station in Pennsylvania in response to allegations by members of the public that the Station had contaminated the surrounding environment. The survey results demonstrated ambient radiation levels in the vicinity of the Station to be no greater than 12 μR per hour (105 mR/year), and within normal ranges of background for the area.

The ARMS has proved to be extremely useful in determining the levels and distribution of radioactivity at nuclear facilities and locating lost gamma radiation sources. Its capabilities also permit evaluation of geological and biological conditions and thermal effects in addition to radiological conditions.

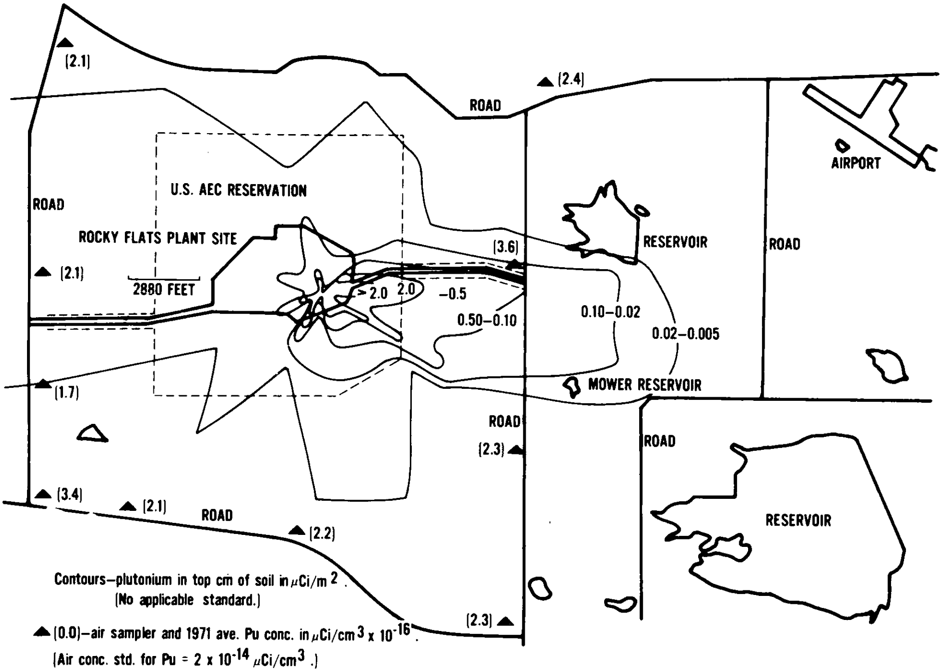


FIG. 5. Plutonium in soil and air in vicinity of the AEC Rocky Flats Plant.

3.3.2 AEC's Health and Safety Laboratory

The AEC's Health and Safety Laboratory (HASL) in New York City is uniquely staffed and equipped to collect and analyze environmental media and foods. The services of HASL are frequently used to evaluate environmental conditions in the vicinity of nuclear facilities. For example, HASL was called on to do extensive soil sampling in the vicinity of the AEC's Rocky Flats Plant in Colorado after it was learned that plutonium from leaking drums of contaminated waste oil had been spread by strong gusty winds to adjacent areas onsite and offsite. Over 50 soil samples were collected by HASL in the vicinity of the plant in areas undisturbed since the Plant started operations in 1953. Each sample consisted of several 20 cm deep soil columns along a 5-meter long transect. The soil and associated vegetation in each sample were air dried, crushed and blended. Plutonium was leached from soil aliquots, isolated by anion exchange, electrodeposited on a platinum disc and counted by alpha spectrometry.

The HASL survey resulted in approximations of total plutonium-239 in offsite areas, and its general distribution and vertical movement in soil. Figure 5 is a recent refinement by the Rocky Flats Plant of the original HASL plutonium deposition contour lines. These contours were drawn by computer and are determined on the basis of the HASL soil sampling results plus more recent samples collected and analyzed by the Rocky Flats Plant. It is now estimated that a total of 3 to 5 curies of plutonium are in the soil in the general vicinity of the Plant, essentially all of which is attributed to wind-blown plutonium from the leaking oil drums.

3.3.3 EPA and State and Local Agencies

A number of state and local agencies have established radioactivity monitoring programs in the vicinity of AEC sites to develop independent environmental data. Results from these programs have compared well with AEC contractor results and provide confirmation of contractor data.

Although the AEC promulgates and enforces radioactivity emission standards for all nuclear installations, EPA is responsible for establishing generally applicable environmental radiation standards for the public and is therefore interested in the concentrations of radionuclides in the vicinity of nuclear installations. The EPA has, on its own initiative, sampled environmental media in the vicinity of AEC sites to provide an independent assessment of radioactivity levels. On a number of occasions the AEC has requested that the EPA assist by conducting a special independent radiation survey in the vicinity of an AEC site. The most recent such survey was made at the Shippingport Nuclear Power Station to ascertain whether elevated strontium-90 concentrations existed in soil and milk, and elevated iodine levels existed in milk as a result of Station operation. The EPA survey, conducted in February and March of this year, confirmed that the Station had not contributed detectable levels of radioactivity to the environment and substantiated Station effluent monitoring results and findings of the ARMS aerial survey mentioned above.

4. EXPERIENCE AT AEC SITES

4.1 General

All significant levels of radioactivity in effluents to the environment from AEC sites are routinely monitored, providing data on the types, concentrations and quantities of radionuclides entering the environment. Currently, well over 99 percent of the gross curies of radioactivity released to the environment from AEC sites consists of radioactive noble gases and tritium, most of which is released to the atmosphere. The current releases of noble gases and tritium from AEC sites consist of approximately 10 percent argon-41, 25 percent tritium and 65 percent krypton and xenon. Since krypton is predominantly a low energy beta emitter, it is not a significant contributor to public dose in the vicinity of AEC sites even though it may be the predominant radionuclide in terms of gross curies released. Tritium and argon-41 are the major contributors to radiation exposure at several AEC sites.

The other radionuclides of environmental interest released from AEC sites consist primarily of low curie quantities of various fission products, activation products, uranium and iodine, and subcurie quantities of plutonium and transplutonium elements.

Since noble gases are not generally measured in ambient air, their offsite impact is usually calculated on the basis of quantities discharged, diffusion models and supplemental thermoluminescent dosimeter (TLD) measurements. Tritium and other radionuclides commonly released from AEC sites are much more susceptible to detection and measurement in environmental media, though in some instances, offsite exposures are calculated to supplement environmental monitoring results.

4.2 Airborne Radioactivity

The airborne radionuclides collectively are responsible for the major portion of the potential exposure to the public near AEC sites. The noble gases released from nuclear reactors result in potential doses ranging to a few milliroentgens per year to individuals in the vicinity of AEC sites. Argon-41, a major contributor to this exposure, is being reduced by modifications to displace the air in reactor voids. Ranking high with the noble gases as a major source of exposure near AEC sites is tritium released to the atmosphere from heavy water reactor and tritium processing operations. Though tritium does not concentrate in environmental media, it enters milk, other foods and water resulting in exposure to members of the public ranging below a milliroentgen per year near AEC sites. Most tritium, when released to the atmosphere, is in the gaseous form and has a minimal contribution to exposures in the vicinity of AEC sites. The AEC's Mound Laboratory, Miamisburg, Ohio, is developing instrumentation to simultaneously monitor for tritium gas and tritiated water vapor.

Perhaps the most difficult airborne radionuclides to monitor and evaluate are the plutonium isotopes. High-volume air sampling equipment having a minimum detection level in the vicinity of 10^{-18} to 10^{-17} microcuries of plutonium per cm^3 of air is used at a number of AEC sites to detect plutonium concentrations in the region between background levels and the maximum permissible concentration applicable to the general public. At the Rocky Flats Plant, Golden, Colorado, wind entrained plutonium from areas of contaminated soil is believed to be the major contributor to offsite annual average air concentrations which have ranged up to 1 to 2 percent of the maximum permissible concentration for the general public for airborne soluble plutonium. The soil became contaminated when drums containing plutonium contaminated waste oils leaked onto a dirt storage pad. The storage area has been stabilized with an asphalt covering to prevent further spread of contamination by wind action. Offsite soil contamination levels range from background (<0.01 $\mu Ci/m^2$) to 2 $\mu Ci/m^2$ at the plant perimeter and offset air concentrations averaged over a year range from 1.7 to 3.6×10^{-16} $\mu Ci/cm^3$ of air (maximum permissible concentration = 2×10^{-14} $\mu Ci/cm^3$).

Occasional airborne releases of iodine-131 from nuclear fuel dissolution activities are reflected in air and milk sampling programs, but do not result in calculated doses of more than a few milliroentgens per year to the thyroids of children. Milk is usually sampled from individual farms to avoid iodine dilution with the pooling of milk from more than one farm. Generally, iodine in milk contributed by nuclear weapons testing debris can be distinguished from AEC site-contributed radioiodine by the accompanying fission and activation products. On occasion, radioiodine has been released from inadvertent dissolution of nuclear fuels not adequately decayed, resulting in elevated radioiodine levels in milk. Perhaps the largest single such release from an AEC facility occurred in May and June 1961. A total of 153 curies was released resulting in milk concentrations ranging up to 6000 pCi/liter. The empirical relationships relating the transfer of radioiodine from air to grass to milk indicated that the ratio of the dose from milk intake to the dose from direct inhalation of the airborne iodine was approximately 700. This has been generally described as the ''reconcentration factor'' and has varied from 100 or 200 to over 1000 in various studies of accidental releases, controlled releases and fallout. There are many variables (e.g., chemical form of the radioiodine, weather and livestock feeding practices) involved in the process of reconcentration, which must be evaluated on a case-by-case basis.

Strontium-90 and cesium-137 are not released to the atmosphere at levels detectable beyond AEC site boundaries, nor are they routinely monitored in ambient air. They are monitored in other media, particularly in milk, since they are important constituents of fallout.

Most AEC sites monitor ambient air for gross beta and alpha activity to detect any major perturbations in atmospheric radioactivity. Such data have little value in the assessment of the environmental impact of radioactivity routinely released from AEC sites.

4.3 Waterborne Radioactivity

Activation products (e.g., phosphorus-32 and zinc-65) in once-through cooling water from the Hanford, Washington, production reactors constituted the greatest source of waterborne radioactivity released from AEC sites until the last of these reactors was shut down early in 1971. Calculated maximum potential doses to individuals in the public decreased with the gradual shutdown of these Hanford reactors from about 25 percent of radiation dose standards in 1967 to near zero dose in late 1971. Presently, the major source of waterborne radioactivity at AEC sites is the tritium released from various facilities. However, tritium discharges have not caused offsite surface water concentrations to exceed 1 percent of the maximum permissible concentration for the general population.

Plutonium-239 and plutonium-238 in liquid effluents are of concern because of their persistance in the environment and evidence that plutonium is concentrated significantly by algae. All AEC sites, however, have been able to maintain plutonium releases at sufficiently low levels that concentrations, including background levels, in offsite waters are about 0.1 percent or less of the maximum permissible concentration for the general public. AEC sites are increasingly recycling plutonium-contaminated waste water to minimize plutonium releases to the environment. In addition, a project is currently underway at the Hanford site to remove plutonium-contaminated soil from a trench which received contaminated waste discharges during the period 1955 to 1962.

Strontium-90 and cesium-137 are not currently released in significant quantities in liquid effluents from AEC operations. However, low levels of these radionuclides have been observed in some environmental media as the result of past waste disposal practices. Strontium-90 concentrations in stream water on one AEC site range around one maximum permissible concentration due to leaching of strontium from old disposal ponds and burial areas. However, the strontium-90 is diluted to less than 1 percent of maximum permissible concentration following mixing in river water beyond the site boundary.

Cesium-137 levels, distinguishable above fallout levels, have been observed in a stream area at one AEC site. Discharges of cesium into the stream over a long period of time were considered to be acceptable because the concentration of cesium in the water was within the maximum permissible concentration for discharges to offsite areas. However, after reactor coolant water ceased to be discharged to the creek, thus decreasing stream flow, cesium from creek sediments appeared in measurable levels in fish taken from the creek and deer that feed in the stream flood plain. Deer flesh was found to contain up to 48 picocuries per gram and fish flesh up to tens of picocuries per gram.

4.4 Ambient Radiation Levels

In addition to the above-mentioned exposures from effluent plumes containing noble gases, offsite radiation fields due to accelerator operations and gamma irradiation sources are monitored using thermoluminescent dosimeters (TLD), ionization chambers and sometimes special equipment to detect high energy radiation. Such offsite radiation fields have ranged up to about 10 percent of the public dose standards at the site perimeter. Ambient radiation measurements are generally difficult to perform reliably and accurately

TABLE III. OBSERVED BEHAVIOR OF SELECTED RADIONUCLIDES AT INDIVIDUAL AEC SITES

SITE	ISOTOPE	SITE RELEASE RATE–Ci/YR AIR	SITE RELEASE RATE–Ci/YR WATER	OBSERVED CONDITIONS IN VICINITY OF SITE
A	^{41}Ar	20,000	–	SITE BOUNDARY – <5% OF STANDARD RESIDENTIAL AREAS – ≈ BACKGROUND
B	^{137}Cs	15.3	–	AIR – NOT DETECTABLE (DETECTABLE IN AIR AND SOIL ONSITE) (DETECTABLE IN WILD ANIMALS ONSITE) MILK – BACKGROUND
C	^{3}H	545,800	124,600	AIR – LESS THAN 0.2% OF STANDARD RIVER – ≈ 0.2% OF STANDARD MILK – ≈ 0.04% OF STANDARD FARM PRODUCE – BACKGROUND
D	^{131}I	3.5	0.2	AIR – <0.01% OF INHALATION STANDARD MILK – <10% OF STANDARD
E	^{238}Pu	(0.4 mCi)	(15 mCi)	AIR – <0.05% OF STANDARD RIVER – BACKGROUND SOIL – LIGHT CONTAMINATION FARM PRODUCE – BACKGROUND
F	^{90}Sr	–	3.4	DISCHARGE TO RIVER – ~100% OF STANDARD RIVER – <1% OF STANDARD FISH – BACKGROUND
G	URANIUM	(2644 lb)	(2618 lb)	AIR – <1% OF STANDARD RIVER – <0.1% OF STANDARD SOIL – LIGHT CONTAMINATION

due to the small increments of dose that must be distinguished from the highly fluctuating background levels.

5. SUMMARY

The two to three decades of operating experience at AEC-owned sites have clearly demonstrated the value of environmental monitoring programs. These programs have provided a feedback of valuable information concerning site operations, site environs, the behavior of radionuclides and compliance with radiation standards. This information has been particularly valuable to the AEC in keeping other Federal and state agencies and the public informed on the extent to which AEC installations affect the environment and the public.

The site environmental monitoring programs have generally provided reliable data for assessment of potential environmental impact and confirmation and documentation of the absence of unacceptable radiological conditions. Table III provides a summary of radiological conditions observed for selected radionuclides in the vicinity of a number of AEC sites from which quantities of the radionuclides have been released. Although the observations are not precise, they provide substantial documentation that radioactivity levels and potential public exposure do not exceed small fractions of the applicable radiation standards. They also substantiate that the policy and practice of controlling effluent radioactivity to the lowest practical levels is resulting in steadily diminishing levels of environmental radioactivity due to the operation of AEC installations.

APPENDIX

FORMAT FOR A TYPICAL ENVIRONMENTAL MONITORING REPORT

I. **Title Page.** Include the title, report period, site or facility, address, and operating contractor. A notice shall be provided which states that the site or facility is an AEC-owned facility.

II. **Introduction.** Provide a brief description of the site or facility and the nature of its primary operations or activities.

III. **Summary.** Provide a concise evaluation and interpretation of the monitoring data contained in the report in relation to applicable standards and requirements with explanation, as appropriate, of unusual incidents or releases. This section should include discussion of any abnormal natural occurrences such as flooding, forest fires, fish kills, altered land use, etc., which could have resulted from or have some impact upon the facility or its operation, and should be written in a manner understandable by the interested layman.

IV. **Monitoring Data Collection, Analysis, and Evaluation.** Include a brief description of the type and frequency and sampling, methods of analysis, and accompanying tables and graphs which clearly and accurately present the monitoring results. A map showing the location of monitoring stations and sampling points shall also be included.

As a general rule, data should only be presented for radioactivity and pollutants in media for which there are applicable standards or other meaningful bases for interpreting the results (e.g., background levels, upstream versus downstream concentration) and interpretations shall be made, as appropriate, of how the environmental levels resulting from plant operations compare to relevant parameters such as background, natural radioactivity, and environmental quality standards.

V. **References.** List and explain, as appropriate, the effluent, air and water quality, and other standards or documents cited in the body of the report.

DISCUSSION

W.O. SCHIKARSKI: According to your measurements, what are the most important radionuclides contributing to the population dose and contained in (1) airborne effluents and (2) waterborne effluents?

C.G. WELTY, Jr.: Of the radionuclides released to the atmosphere from AEC installations, ^{41}Ar probably contributes the greatest portion of population dose in the vicinity of the installations; tritium, we believe, contributes the greatest portion of man-rem dose to the public at large.

Among those radionuclides released in liquid effluents, tritium is considered to be the greatest contributor of exposure, both in the vicinity of the site and as regards the general public.

G.H. PALMER: Can you give any idea of the cost of the Aerial Radiological Measuring System? It is presumably expensive, and since it can only provide radiation data and must be followed up by ground surveys for specific nuclides, is it really justified in terms of cost?

C.G. WELTY, Jr.: It is very difficult to assign monetary values to the individual aerial surveys, and therefore I cannot provide meaningful cost data for surveys described in our paper. I wish to re-emphasize, however, that the Aerial Radiological Measuring System is an emergency response system and the planned land area surveys described in the paper constitute a side benefit resulting from the necessity frequently to use the system and to maintain it in a high state of readiness. It must be remembered that the system can be used to investigate thermal effects and other physical parameters, apart from gamma radiation. I agree, of course, that ground measurements are needed as well.

H. WIJKER: In your measurements of environmental exposure rates, what percentage of background do you hope to attain, i.e. what is your target sensitivity?

C.G. WELTY, Jr.: Our environmental monitoring programs indicate that, generally speaking, AEC sites are maintaining radioactivity and public exposure levels within one per cent of the applicable ICRP standards. We believe that it is desirable to develop quantitative information, when feasible, on radioactivity and exposures in this region below one per cent of the applicable standards. For some radionuclides, concentrations in the vicinity of one per cent of the standard can be quantitatively measured, e.g. tritium in surface waters. The levels of other radionuclides in environmental media, and their contribution to public exposure, cannot be directly or indirectly measured within the low ranges of interest and must be estimated by calculational methods, as is the case with ^{41}Ar.

N.T. MITCHELL: My comments relate to the design of your monitoring programs and particularly that element thereof which has no direct relevance to public radiation exposure. Our policy in the United Kingdom has been largely to confine ourselves to critical exposure pathways, or those which are potentially critical. Even where monitoring could be described as being conducted for public relations purposes — where exposure is so infinitesimally low that on scientific grounds one could discount any need for monitoring — we aim to link our surveys to the pathways that would be critical, so as to establish the public-exposure significance of any contamination we might find. Could you comment on the extent to which you find yourself obliged to perform monitoring which has no direct bearing on exposure of the public?

C.G. WELTY, Jr.: In the United States of America we also believe that it is necessary to concentrate monitoring efforts on the levels of radioactivity in the important human exposure pathways. The public, however, is expressing considerable interest in the extent to which nuclear facilities contribute to the presence of radionuclides in all environmental media. We therefore find ourselves called upon to determine the levels of radionuclides in all off-site media of public interest, such as soil and sediments, and to publish our findings.

W.K.G. KÜHN: It is, I believe, estimated that the tritium activity in the atmosphere will reach 10^8 Ci by the year 2000. Have you measured any general increase of ^3H in the atmosphere?

C.G. WELTY, Jr.: The AEC site environmental monitoring programs are not sufficiently sensitive to indicate short-term changes in tritium background levels in the atmosphere. Use of moisture condensing samplers at AEC sites permits the detection of tritiated water vapour in the atmosphere at levels well below the applicable standard. Milk sampling has also proved to be a sensitive indicator of elevated tritiated water vapour in the atmosphere. Instrumentation capable of simultaneously monitoring the oxidized and elemental forms of tritium in the atmosphere is at present being designed and developed.

A PRELIMINARY EXPERIMENT ON THE DEPOSITION OF GASEOUS RADIOIODINE ONTO ENVIRONMENTAL MATERIALS

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Abstract

A PRELIMINARY EXPERIMENT ON THE DEPOSITION OF GASEOUS RADIOIODINE ONTO ENVIRONMENTAL MATERIALS.

The deposition rates of gaseous radioiodine onto some environmental materials were measured experimentally with the use of an air-circulating chamber of dimensions 1.0 m × 1.5 m × 2.0 m. The circulating air speed and relative humidity are controllable in the ranges of 0 to 5 m/s and 50 to 100%, respectively, and a water spray device was set in the ceiling. To simulate the composition of airborne radioiodine released from the stack of a fuel reprocessing plant or radioisotope production plant, carrier-free Na ^{131}I was oxidized in 10N HNO_3 solution and the gas generated was introduced into the chamber after being scrubbed in NaOH solution by continuously bubbling N_2 carrier gas. The composition of ^{131}I compounds in the chamber was analysed by radio-gas-chromatography. At the start of injection, dominant components were the inorganic iodides, HIO_3 and HIO_4, and then the fraction of organic iodides increased and remained at about 80% after about four hours had elapsed. Averaged concentrations in the chamber were in the range 10^{-6} to 10^{-7} μCi/ml and exposure durations were 6 to 7 hours. The environmental materials used were soil, sand, water, vegetables and pine tree leaves. The deposition velocity was determined as the ratio of the deposit per unit area of the surface of the materials to the accumulated concentration in air. The values obtained were in the range 10^{-2} to 10^{-3} cm/s and they decreased in the sequence soil, sand, vegetables, water. Converting the values for the vegetables to values for vegetation per unit area of the ground by using data on area density of vegetation, the deposition velocity to the vegetation was estimated to be almost the same as that to soil.

1. INTRODUCTION

Radioiodine is often a critical nuclide in nuclear accidents. The thryroid dose resulting from the ingestion of vegetables contaminated with deposited radioiodine is considered to be important as well as that resulting from the inhalation of airborne radioiodine. The chemical composition of radioiodine released into the atmosphere from the stacks of nuclear facilities differs according to its origin, the time elapsed from generation, the surface-to-volume ratios of the containments, and the type and efficiency of the filters. The behaviour of airborne radioiodine in reactor containment systems has been summarized [1, 2] and the reported fractions of organic iodide, which is little removed after generation of iodine gases, are distributed over the wide range of 0.002 to 30%. Analysis by radio-gas-chromatography of airborne radioiodine in the exhaust air of an ^{131}I production plant or a fuel reprocessing plant showed a distinct difference in composition between before and after passing a filter system, i.e. the fraction of $H^{131}IO_3$, which was 80% before filtering, decreased to 17% after filtering, while that of $CH^{131}I$, which was 15%, relatively increased to 69% [3]. These results suggest that the chemical form of iodine released to the atmosphere depends on the type and efficiency of the filter, and that most of the airborne radioiodine would be in organic form.

Deposition in the field is a very complicated phenomenon, and the deposition velocity introduced by Chamberlain is used as a parameter for giving an overall estimate of the transfer of radioiodine from air to vegetation. Many experimental determinations of the deposition velocity of radioiodine have been made both in laboratory experiments and in field tests. Most of the experiments, however, concern elemental iodine and few deal with organic iodide. The deposition velocity of methyl iodide is two orders of magnitude (in field experiments) or three orders (in laboratory experiments) less than that of elemental iodine [4]. In general, experimental conditions involve many parameters, most of which are not clear, particularly for field experiments. In addition, data reported in the literature cannot always be applied to other cases, even under comparable conditions. It is most desirable to do field experiments using actual environmental samples, the radioiodine compositions of which are similar to those in the actual release, but it is not easy to carry out a field test with release of radioactive materials because of the difficulty in getting a test area sufficiently large and radiologically safe. Hence the authors constructed a chamber to simulate the natural conditions to some extent, and a preliminary experiment with ^{131}I and environmental samples was made.

2. METHODS

Apparatus

The experimental apparatus, shown in Fig. 1, consists of a chamber, an air-circulating duct, a blower with ventilating rate up to 90 m^3/min, a humidifier, and an ^{131}I gas generating device. The chamber, 2 m long, 1 m wide by 1.5 m high, has 2 pairs of gloves, 6 air-sampling pipes at the side wall and a set of water sprays in the ceiling. The wind speed distribution in the chamber was measured with a thermister anemometer

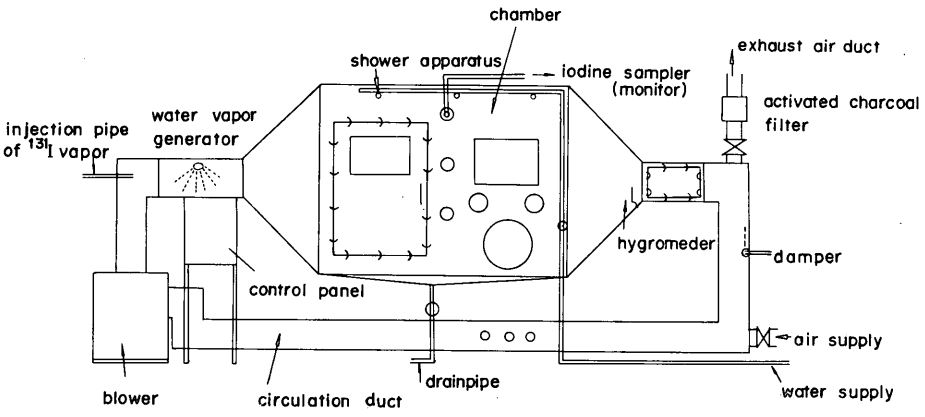

FIG. 1. Experimental apparatus.

at various ventilating rates, and wind flow was visualized with pieces of wool string attached on a frame. The flows were rather irregular and weak backflows were observed at the ceiling and bottom surfaces. Therefore, the mean wind speed in the middle of the chamber was used only as a qualitative index in the present experiment.

Peparation of airborne radioiodine

Carrier-free $Na^{131}I$ solution of about 0.5 ml was added in 10N HNO_3 (100 ml) kept at 100°C in a reaction flask and the iodine produced was scrubbed with 10N NaOH (20 ml) solution by bubbling N_2 carrier gas continuously at a flow rate of 100 ml/min. The gas was subsequently introduced into the chamber. Samples of airborne radioiodine were taken at various periods after injection into the chamber and analysed by radio-gas-chromatography; the separation column kept at 120°C was of silicon-oil-coated celite base (60-80 mesh) packed in a glass tube of 4 mm inner diameter by 3 m length, and carrier gas (He) was made to flow at 30 ml/min. The results of the three runs for determining the composition of ^{131}I in the chamber, which were made under the same conditions prior to the deposition runs, are shown in Fig. 2. The detected iodine compounds were $H^{131}IO_3$, $H^{131}IO_4$ and $CH_3{}^{131}I$ and $C_2H_5{}^{131}I$. The fraction of HIO_3 was 25% at the initial stage and decreased rapidly owing to deposition on the wall. It was not detected after 4 hours. HIO_4 was 40% at first and decreased to 20% after 4 hours, then remaining constant. Organic component (CH_3I 85%, C_2H_5I 15%) increased up to 80% and reached equilibrium in 4 hours. The fractions obtained were assumed to be the same in the deposition runs.

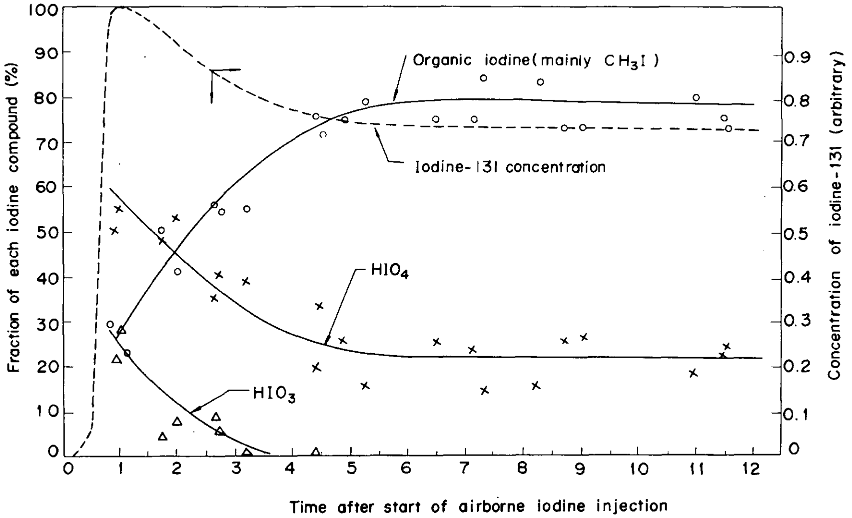

FIG. 2. Variation in the fraction of airborne iodine components in the chamber.

Measurement of ^{131}I

Airborne ^{131}I in the chamber was sampled with three cartridges of activated charcoal (5 cm in diameter by 2 cm thick each) in series for a 30-minute or one-hour period each, and measured with a 5 in. ø × 4 in. NaI(Tl) scintillation detector and a 400-channel pulse height analyser. The variation of concentration with time was monitored continuously with an activated charcoal cartridge and a 2 in. ø × 2 in. NaI(Tl) detector.

Environmental samples

Soil (humus soil passed through a sifter of 2-mm mesh), sand (mass mean dia. 0.5 mm) and water were put in trays of 750-cm^2 width and 5-cm depth. Potted leafy vegetables were spinach (Spinica oleracea L.), leaf beet (Beta vulgaris L.) and a variety of Chinese cabbage. Fresh pine twigs were put in a pot. Pine needles have been chosen as an index material in environmental monitoring because of their abundance in all seasons in our country.

Experiment

The samples were arranged on the floor of the chamber. The radioiodine, which was generated as described above, was then injected continuously and air was circulated for several hours. After the injection was stopped, the air containing radioiodine was exhausted for 30 min and the samples taken out. The pine needles were separated from the twigs and the vegetables chopped up. Each sample was placed in a plastic case, 13 cm ø by 7 cm high, and the γ-spectrum was measured with a 5 in. ø × 4 in. NaI(Tl) scintillation detector and pulse height analyser. The amount of ^{131}I was obtained from the photopeak area at 0.36 MeV.

Three runs were carried out under similar conditions. The wind speed was about 2 m/s and the relative humidity about 80%. The amount of ^{131}I used was about 0.5 mCi in each run. The fraction of the injected ^{131}I of the total amount was estimated to be a few per cent, although no detailed estimation of balance of the iodine amounts was made. The concentration of airborne ^{131}I was 10^{-7} to 10^{-6} μCi/ml and the duration of exposure about 6 to 7 hours.

3. RESULTS AND DISCUSSION

Airborne iodine

Figure 3 gives an example of the variation in the concentration of airborne ^{131}I in the chamber. The charcoal cartridge used in the continuous monitoring had a sampling efficiency of 60%, and the sampling efficiency of the three-cartridge sampler was nearly 100% for the sampling period of less than 2 hours. The time integrated concentration of airborne ^{131}I in each run was evaluated by the sampling method.

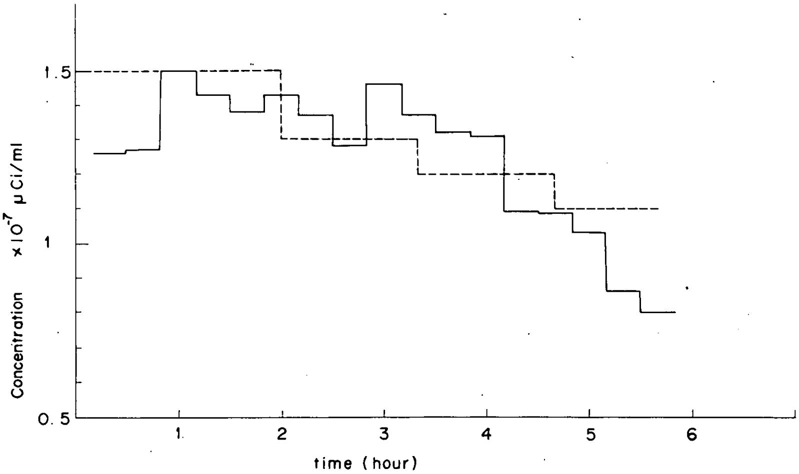

FIG. 3. Variation in the concentration of airborne radioiodine in the chamber (Run 2): Solid line, the value of continuous monitoring; broken line, the value obtained by sampling.

Deposition velocity

To get the deposition amount per unit area of the leaves from the measured activity of ^{131}I per gram of sample, surface areas of leaves per unit weight of the plants were obtained. They were 160, 60, 75 and 40 cm^2/g for spinach, leaf beet, Chinese cabbage and pine needles, respectively. The deposition velocity was determined according to the definition [5]:

$$\text{Deposition velocity} = \frac{\text{Amount deposited per cm}^2\text{ of surface}}{\text{Time integral of volumetric activity per cm}^3}$$

The results are shown in Table I. The value for water is the smallest and that for sand is one order of magnitude less than that for soil. The difference may be attributed to the difference in surface area in contact with air.

Values of deposition velocity reported in the literature are usually for vegetation per unit area of the ground. To convert the deposition velocity obtained for the surface of a sample itself to that for vegetation, the area density of vegetation is needed. This depends on the method of cultivation and the state of growth of the plant. Mean values based on agricultural statistics in Japan are 1.5 kg/m^2 for spinach and 3.3 kg/m^2 for Chinese cabbage, both in the harvesting stage. The corresponding value for leaf beet is considered to be nearly the same as that for spinach. These values are applied to the calculation. The converted deposition velocities are shown in Table II. The values for vegetation are around 0.02 cm/s and are nearly the same as that obtained for soil.

The values obtained are one or two orders less than those of elemental iodine and seem to correspond to those for organic iodides. As seen in

TABLE I. DEPOSITION VELOCITIES OBTAINED FROM THE EXPERIMENT

Sample	Number of samples	Average (cm/s)	Standard deviation (cm/s)
Pine needle	15	2.0×10^{-3}	0.92×10^{-3}
Spinach	4	1.1×10^{-3}	0.32×10^{-3}
Leaf beet	6	2.1×10^{-3}	0.40×10^{-3}
Chinese cabbage	6	0.69×10^{-3}	0.29×10^{-3}
Soil	6	27×10^{-3}	6.3×10^{-3}
Sand	6	2.5×10^{-3}	1.0×10^{-3}
Water	4	1.0×10^{-3}	0.99×10^{-3}

TABLE II. DEPOSITION VELOCITIES CONVERTED TO NATURAL CONDITIONS OF THE GROUND SURFACE

Samples	Average (cm/s)	Standard deviation (cm/s)
Spinach	0.026	0.008
Leaf beet	0.024	0.005
Chinese cabbage	0.014	0.006
Soil	0.027	0.006
Sand	0.0025	0.001
Water	0.0010	0.0008

Fig. 2, however, the fraction of inorganic component is rather large in the early stage of a run. As deposition is additive, the contributions of the organic and inorganic components to deposition may be expressed as:

$$V_g C = V_{go} C_o + V_{gi} C_i$$

where V_g and C denote deposition velocity and time integrated concentration of ^{131}I respectively and subscripts o and i denote the organic and the inorganic respectively. Fractions C_o/C and C_i/C are obtained from the data of 10-min mean concentration using the curves shown in Fig. 2. The fractions of the organic component in the time integral of concentration were 52, 52 and 60% in the three runs, i.e. the contribution of the inorganic component is of the same order as the organic one in the present experiment, although the later predominates after 4 hours. The observed deposition velocity values are very small. The deposition velocity of the inorganic compound, which has a small deposition velocity, was identified as HIO_4 by radio-gas-chromatography, as mentioned above.

Importance of vegetable ingestion relative to inhalation

The thyroid dose received by a child continuously inhaling an iodine isotope of C μCi/cm^3 can be estimated as follows;

$$D_{inh} = \frac{4k\epsilon f_a}{m\lambda_e} F_{inh} C \quad (\text{rem/h})$$

where

k is the conversion factor from activity unit to dose unit
ϵ is the effective energy of the isotope for the thyroid
λ_e is the effective decay constant of the isotope in the thyroid
m, f_a and F_{inh} are the thyroid mass, the fraction of uptake defined in the ICRP internal dose calculation and breathing rate of an adult (standard man)
C is the concentration of the isotope above the ground.

The same concentration of the isotope, C, is considered, taking into account the difference in thyroid mass and breathing rate, as giving about a four-fold dose to a child as to an adult [6, 7] so that the factor of 4 is here used in the equation. In the case of ingestion of leafy vegetables the adult is considered, though it is not known yet whether adults are more critical than children, and the thyroid dose is calculated by:

$$D_{ing} = \frac{k\epsilon f_w}{m\lambda_e} F_{ing} C_v$$

$$C_v = \frac{S}{\lambda_{eff}} V_g C$$

where

f_w is the fraction of uptake also defined by the ICRP
F_{ing}, λ_{eff}, S and V_g are the daily ingestion, the effective decay constant of the isotope on the vegetable, the reciprocal of the area density of vegetation and the deposition velocity, respectively.

The ratio of D_{ing} to D_{inh} shows which pathway gives more dose to the thyroid:

$$R = \frac{D_{ing}}{D_{inh}} = \frac{f_w F_{ing} S V_g}{4 f_a F_{inh} \lambda_{eff}}$$

According to the ICRP, $f_w/f_a = 1.3$ and $F_{inh} = 2 \times 10^7$ cm^3/d. Experimental data on the effective half-life of ^{131}I on vegetation range from 5 to 7 days. Therefore, λ_{eff} would be about 1.3×10^{-6} s^{-1}. A figure of 6.7 cm^2/g for S the reciprocal of the area density may be considered as a fairly good figure on the basis of national statistical investigations in Japan. The daily intake of leafy vegetables F_{ing} is on average 60 g/d. Though the deviation of this average is wide, it is not more than one order of magnitude. Accordingly, for ^{131}I taken with spinach

$$R \approx 5V_g$$

Consequently, the ratio depends strongly on the deposition velocity, which varies very widely; taking a deposition velocity of 10^{-1} cm/s, which is overestimated on the basis of the present study, the importance of vegetable ingestion is the same as that of inhalation. When choosing a velocity available to elemental iodines, the ingestion pathway would become more important.

4. SUMMARY

The deposition velocities of ^{131}I gas (organic component 50% and inorganic component 50%) to selected environmental samples were determined under conditions of little wind and high humidity in an air-circulation chamber. The values for vegetables were converted to those for vegetation and were around 0.02 cm/s, which is nearly the same as those obtained for soil. The values are considered to be reasonable for gaseous radioiodine and are one or two orders less than those reported for elemental iodine. They show that the dose from the ingestion of leafy vegetables is as important as that due to inhalation.

ACKNOWLEDGEMENTS

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DISCUSSION

C.A. MAWSON: As I understand it, you introduced elemental iodine into the chamber and after 4 hours 80% of the iodine was in the form of methyl and ethyl iodide. Where did the methyl and ethyl groups come from?

K. IMAI: As a matter of fact we did not introduce elemental iodine into the chamber. The Na^{131}I was oxidized in a reaction flask, and the gas generated was scrubbed with an NaOH solution and then injected into the chamber.

The mechanism of generation of the methyl and ethyl groups is not clear; they may be generated in the oxidation stage or they may predominate in the generated gas itself after 2 hours.

In the chamber the initially predominant inorganic component is removed by deposition onto the wall, and the organic component attains 80% in the equilibrium state after 4 hours.

P. REINIGER: I have two questions on your paper. First, the deposition velocity on plants, at least over longer periods, may be influenced by their physiological status. In this connection I should like to ask whether your experimental chamber was illuminated. Secondly, iodine being very reactive, do you not think it possible that interaction with ozone, light and H_2SO_4 (from SO_2) would have a considerable effect on your results?

K. IMAI: I agree with you as regards the importance of the physiological status, though in these preliminary experiments we did not bother about it much. The chamber was illuminated by lamps at the top. Regarding your second question, I doubt whether there is any general agreement on the subject. I personally feel that some considerable part of the organic iodide (HIO_4 would be the same) would be decomposed by the factors you mentioned and attached to the aerosol in the atmosphere. However, our results could not be extended to problems in the atmosphere without careful re-working.

K.-J. VOGT: At the Jülich Nuclear Research Establishment we last year performed some field experiments to measure the deposition velocity of elemental iodine on vegetation (grass, clover). The deposition velocity was found to be proportional to the vegetation density and also to be dependent on biological and meteorological parameters. I think it is difficult to simulate meteorological conditions, such as friction velocity, inside deposition chambers.

We intend to present the results of our deposition experiments at the IAEA/WMO Symposium on the Physical Behaviour of Radioactive Contamination in the Atmosphere to be held in Vienna in November 1973.

J. BOGEN: What is the volume of your reaction chamber?

K. IMAI: The volume is 3 m^3 (2 m×1 m×1.5 m).

J. BOGEN: Are you quite sure that deposition takes place in the gaseous phase, and that there is no possibility of transfer from the gaseous to the aerosol phase?

K. IMAI: To separate the aerosol we placed a paper filter in front of the charcoal cartridge. We found no significant ^{131}I activity on the filter. This makes us confident that most of the airborne iodine is in the gaseous phase, and that deposition takes place in this phase in our experiments.

WIND-DRIVEN REDISTRIBUTION OF SURFACE-DEPOSITED RADIOACTIVITY*

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Abstract

WIND-DRIVEN REDISTRIBUTION OF SURFACE-DEPOSITED RADIOACTIVITY.

The deposition of radionuclides on a terrestrial surface can result in the delivery of dose to man through external radiation exposure, food chain contamination or inhalation of suspended particles. ^{239}Pu is one of the few radionuclides generally regarded as constituting an inhalation hazard through wind-driven resuspension. No adequate models of resuspension exist. Two major problem areas have been identified: (1) the rate at which initially deposited debris weathers into a less erodible state as a function of time, and (2) a source term factor in material removed per unit time per unit area, applicable to a source of any configuration. In experiments conducted at the US Atomic Energy Commission's Nevada Test Site, concentrations in air of particles moving in suspension were studied over a period of six weeks following the nuclear cratering event Project Schooner and over a 10-month period following accidental venting of an underground nuclear explosion. Suspended air activity was observed to decrease in time with half-times of 35-80 days; this factor appears to be much more important than variations in meteorological parameters over these relatively short time periods. Movement of small particles in suspension accounts for only a minor fraction of the total mass movement. As much as 50% of the initially deposited debris was moved by saltation within a 24-hour period. Such movement can result in extensive micro-inhomogeneities with accumulation of debris under bushes or other places affording shelter from erosive forces. More detailed study of the resuspension process in an aged ^{239}Pu field is now in progress. Preliminary data are given on the particle size distribution of the total mass and the ^{239}Pu moving in suspension.

INTRODUCTION

The deposition of radionuclides on a terrestrial surface can result in the delivery of radiation dose to man through several mechanisms including external radiation exposure, terrestrial food-chain contamination, and the inhalation of small particles which have been resuspended from the original deposition area. The latter pathway can become relatively important if the radionuclide of concern is a long-lived alpha-emitter, is of low biological availability through food-chain pathways, and is retained in the lung (or pulmonary lymph nodes) over long time periods. ^{239}Pu is one of the few radionuclides that are generally regarded as constituting an inhalation hazard through wind-driven resuspension.

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* Work performed under the auspices of the US Atomic Energy Commission.

No adequate models of the resuspension process presently exist. Early attempts to define the magnitude of the problem used a "resuspension factor" which was defined as [1]

$$S_f = \frac{\text{Air Concentration (in activity/m}^3\text{)}}{\text{Surface Deposition (in activity/m}^2\text{)}}$$

This factor, coupled with an "attenuation factor" to account for the decrease with time of the fraction of the source apparently available for resuspension, has been used to model the resuspension process with emphasis on deriving a permissible surface deposition level [1,2]. However, this approach suffers from several serious drawbacks. For example, it presumes that measured levels of air activity are directly relatable to the ground deposition at the point where air measurements are made. This presumption is frequently found to be unjustified; in large fallout fields, for example, resuspension factors are commonly much larger in areas of low deposition than they are in areas of relatively higher deposition [3 – 5]. A logical explanation is that resuspended air activity levels are more strongly related to surface deposition levels upwind of the point of measurement than they are to the deposition level at the actual point of measurement.

Other objections may be raised to the "resuspension factor" approach primarily due to the lack of consideration of many variables which one intuitively feels should be significant and/or which have been shown to be significant in the related problems of wind-driven sand and soil erosion [6 – 8]. Examples are wind velocity, surface roughness, physical and chemical characteristics of the soil surface, and vegetation cover.

The authors are currently engaged in an effort to develop a more generally applicable model which can be used to predict resuspended air activity levels as a function of time and distance from a surface deposit of any geometrical configuration and of varying soil characteristics. This broad problem may be broken down into at least two major problem areas: 1) the rate at which an initial surface deposit weathers down into the soil surface and becomes relatively nonerodible and 2) the rate at which a surface contaminant at a given point in time is resuspended in terms of the fractional activity removed per unit time per unit area. A complete understanding of these two processes and the parameters upon which they depend should allow the construction of a completely general resuspension model.

A complete description of the redistribution process (as opposed to resuspension) must also include the movement of particles too large to be moved in suspension, but which move by the processes of saltation and surface creep [6]. Although such particles are too large to be respirable, they do account for the majority of the total mass movement of soil by the wind. It has also been shown that small particles are much more susceptible to movement in suspension if there is concomitant movement of larger particles in saltation [7].

This paper concerns our initial experiments to delineate the factors pertinent to the redistribution process. More elaborate experiments currently in progress are briefly discussed.

METHODS

Two experiments were carried out at the U.S. Atomic Energy Commission's Nevada Test Site. The first was performed following the nuclear cratering event, Project Schooner, which was detonated on 8 December 1968. Fourteen sequentially operated air sampling stations had been established prior to the event, and were automatically turned on by the presence of the elevated radiation field produced by the radioactive cloud. Sampling was continued for six weeks following the event. Additional details concerning this experiment have been published previously [5].

A second experiment was carried out following the accidental venting of an underground nuclear explosion on 18 December 1970. Six sequentially operated stations equipped to sample both air activity and ground surface deposition were established three months after the venting and were operated over a 9 to 10 month period. The locations of the sampling stations are shown in Fig. 1.

Descriptions and functional characteristics of the sampling stations have been reported previously [9,10], and will only be summarized briefly here. Each air sampling station consisted of a bank of seven sampling heads, six of which were activated in sequence. The seventh sampling head served as a control; it was not actuated but its filter was changed and analyzed as though a regular sample to monitor the possibility of surface contamination of the filters. A convoluted fiberglass filter with a collection efficiency of 99% for particles as small as 0.025 μm was used for both experiments. Air flow rates were approximately 20 ft^3/min for the Schooner experiment and 10 ft^3/min for the Baneberry experiment. Sampling periods were adjusted as necessary. At early times in the Schooner experiment, stations were programmed so that each succceeding sampler ran twice as long the immediately preceding sampler, with the first sampler running for one hour. During the Baneberry experiment, samplers at Station 4 were on a 24 hour time period; other stations were generally on a 96 hour period. Filters were oriented upwards thereby prejudicing the collection of particles towards the smaller sizes particularly during periods of high wind speed.

Field calibration of air flow rates for each sample head was performed at the beginning and end of each run using a manometer to measure the pressure drop across the filter. A running-time meter for each sample head allowed precise determination of the actual time period that each sampler ran. In some cases due to equipment malfunction or power failures, this information could not be matched to field observations and the samples involved were not included in further data analysis.

In addition to the air samplers, three automatically opening and closing fallout trays [9,10] were deployed at each of the six stations established for the Baneberry experiment. Inserts were placed in each tray which presented a collection surface of 2.35 ft^2 of adhesive-coated paper. The opening and closing of the trays was synchronized with the air samplers such that each tray was open for the running time of two air samplers. The fallout trays were deployed primarily to catch larger particles moving in saltation.

Filters and adhesive-coated papers were hermetically sealed in thin-walled aluminum cans in a constant-geometry configuration [10]. For the Schooner experiment, one of the most prominent radionuclides of fairly long half-life was ^{181}W. The Ta K X-rays produced by its decay dominated the low energy spectra of these samples, and it was possible to reliably quantitate this radionuclide using a NaI(Tl) spectrometer. All results were decay corrected to detonation time.

Samples from the Baneberry experiment were of much lower total activity. These samples were set aside for several weeks after sealing to allow for the reestablishment of secular equilibrium in the ^{238}U natural radioactivity

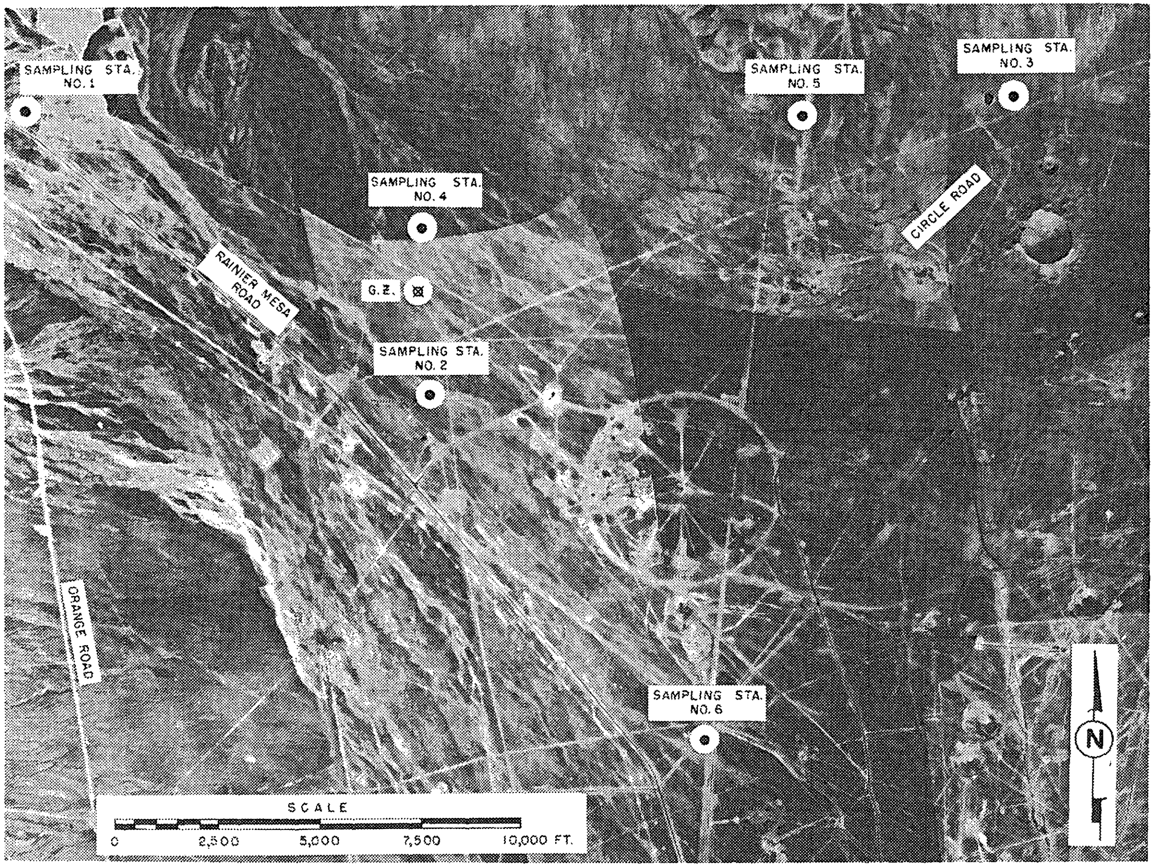

FIG. 1. Station locations for the experiment conducted following the Baneberry venting.

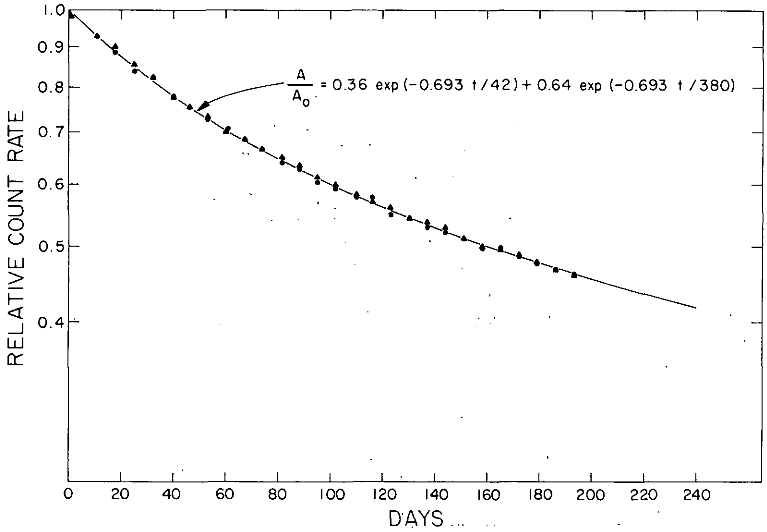

FIG.2. The decrease of activity with time of two individual deposition samples. The line is a least-squares fit of the data.

chain and then assayed for gross-gamma activity using a NaI(Tl) crystal. In order to correct for the influence of radioactive decay on the gross gamma measurements over the extended period of this experiment, six of the earliest, more active samples were counted each day the counter was used. Results for two of these samples are shown in Fig. 2. Data from all six samples were well fit by a two component exponential curve corresponding to half-lives of 42 ± 1 and 380 ± 7 days. Analysis of these same samples by Ge(Li) spectrometry indicated that the most prominent radionuclides were ^{103}Ru and ^{106}Ru/^{106}Rh with half-lives of 39.6 and 369 days. All samples were therefore decay corrected to a common point in time using this derived relationship.

Only limited meteorological data were available in conjunction with the Schooner experiment. Several 30 ft towers with wind speed and direction sensors were located in the vicinity of the Schooner surface ground zero preceding the event for other purposes, but were inactivated two days after the event. One other wind sensor in the area was operating between the fifth and the twelfth day after the event. During the Baneberry experiment, wind sensors mounted on 2 m towers were deployed specifically to support this study. Wind speed and direction data were continuously recorded by strip chart recorders. Precipitation data were available from three previously established stations in the vicinity.

The gross effects of the redistribution processes were examined in the Schooner fallout field two years after the event (October, 1970). Areas affording shelter from the erosive forces of the wind were examined in comparison to locations affording little or no shelter. This was accomplished by the use of a FIDLER instrument, a portable scintillation counter designed for the detection of low-energy gamma- or X-rays [11], with pulse height selection optimized for the detection of the Ta K X-rays accompanying the decay of ^{181}W.

RESULTS AND DISCUSSION

Project Schooner

Results for two of the 14 individual air sampling stations are shown in Figs. 3 and 4. The length of the horizontal bars represents the time period that each sampler was operating; all data were corrected for radioactive decay to the time of detonation. Station 25 (Fig. 3) was located 6 miles north of surface ground zero and was downwind at detonation time. Of particular interest is the marked secondary peak of activity which was also recorded at several additional stations during the time period of 30 to 50 hours following the detonation. At Station 25, this secondary peak reached 30% of the maximum activity recorded during cloud passage, and also corresponds to the first period following the detonation of strong southerly winds.

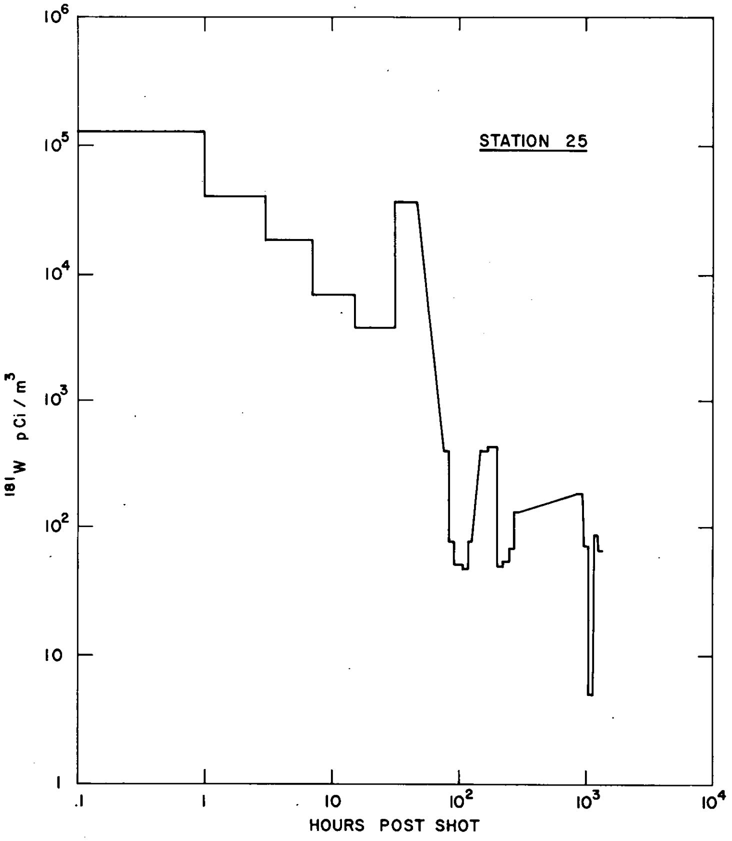

FIG. 3. ^{181}W air activity as a function of time at Station 25 following Project Schooner, a nuclear cratering experiment. Station 25 was 6 miles north of surface ground zero, and was downwind at detonation time.

Station 8 (Fig. 4) was located 7500 ft south of surface ground zero, was upwind at detonation time, and recorded relatively low activities at early times. However, during the first time period of strong northerly winds, air activity levels increased substantially and at 90 hours post detonation Station 8 recorded the highest air activity level of any of the 14 stations.

Both Stations 8 and 25 are fairly typical of the results for this experiment in that resuspended air activity levels were quite high within the first few days following the initial deposition of the fallout field, and exhibited an easily identified response to significant changes in wind direction or speed.

The variation of air activity with time is more readily identified in the semi-logarithmic plots of Figs. 5 and 6. Both Figures are plots of composite normalized data from the five downwind stations within six miles

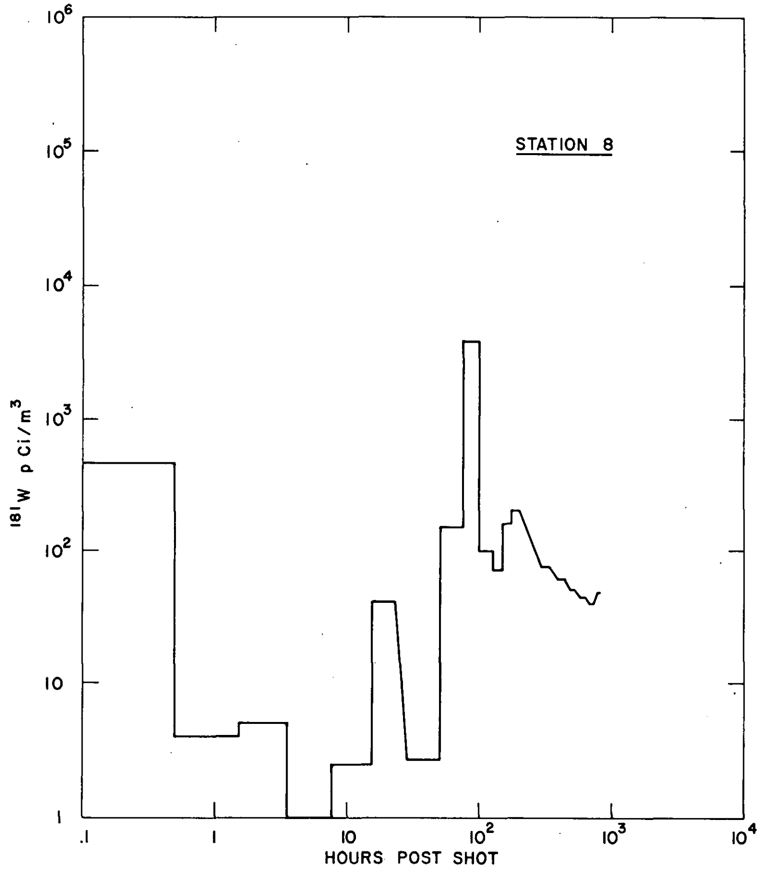

FIG.4. ^{181}W air activity as a function of time at Station 8 following Project Schooner, a nuclear cratering experiment. Station 8 was 7500 ft south of surface ground zero, and was upwind at detonation time.

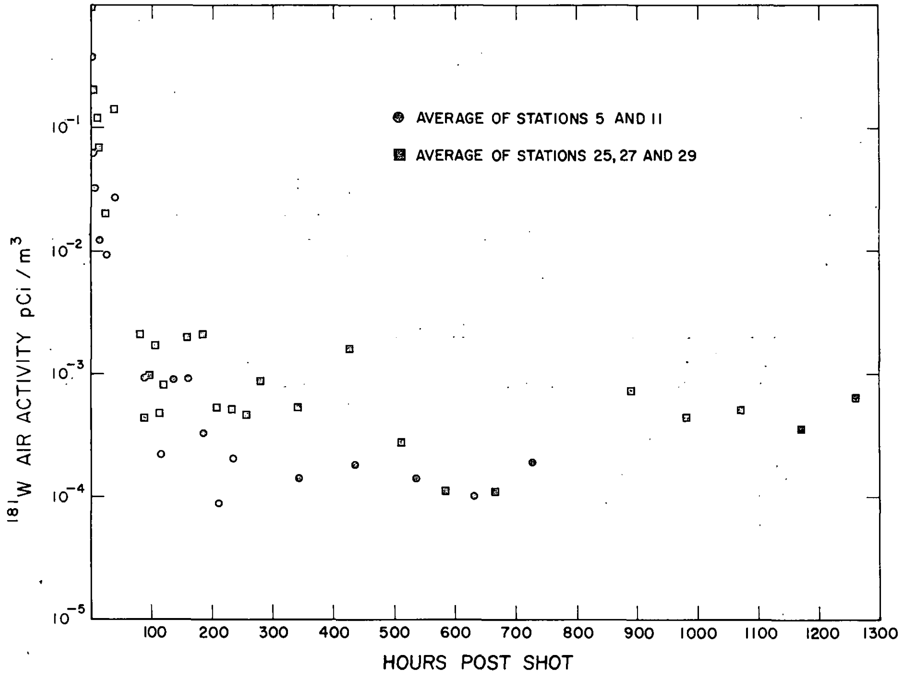

FIG. 5. ^{181}W air activity as a function of time at the five downwind stations within six miles that were closest to the line of maximum deposition from Project Schooner. Data were normalized to the first sample taken at the time of cloud passage.

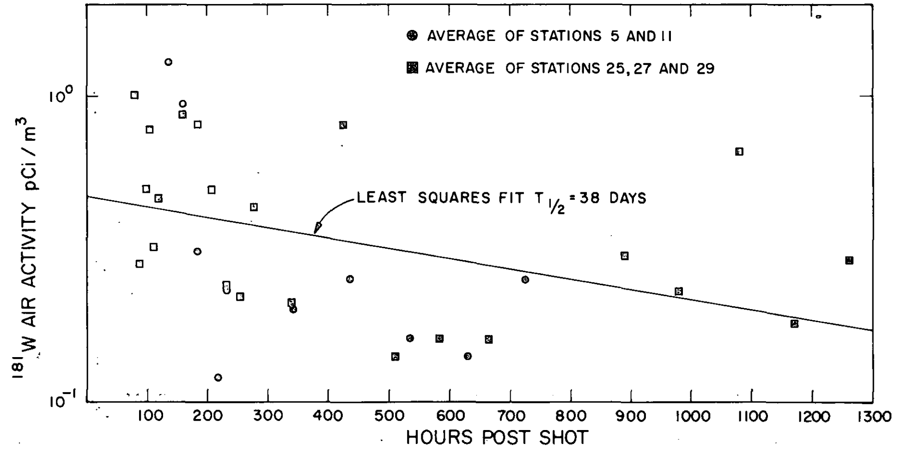

FIG. 6. ^{181}W air activity as a function of time at the five downwind stations within six miles that were closest to the line of maximum deposition from Project Schooner. Data were normalized to the first sample taken after 70 hours had elapsed following the detonation.

TABLE I. DISTRIBUTION OF ^{181}W ACTIVITY BETWEEN AREAS EXPOSED TO AND SHELTERED FROM WIND EROSION

Activity was measured using a FIDLER instrument [11]. The measurements were replicated 51 times.

Measurement point	Mean	Standard error
5 in. above grass clumps	45,800 counts/min	380 counts/min
5 in. above exposed soil, 2 ft from nearest grass	35,300	670
Difference	10,500	750 ($p = < 0.0001$)

of surface ground zero which were closest to the line of maximum deposition. Fig. 5 is normalized to the first sample; the initial three orders of magnitude decline is presumed to reflect the disappearance of the initial cloud produced by the explosion although the secondary peak between 30 and 50 hours post detonation is a major perturbation. The second phase of the decline is better illustrated in Fig. 6 where the data have been renormalized to the first sample taken after 70 hours had elapsed following the detonation. A least-squares fit of these data indicates a "weathering-in" rate or "attenuation factor" with a half-time of 38 days. This value is in good agreement with the half-time of 5 weeks measured by Wilson, et al. [4] during the period of 4 to 24 weeks following a plutonium release at the Nevada Test Site in 1957, although the data fit in both cases is poor.

Results of the survey with the FIDLER instrument to examine the extent of the gross movement of material within the Schooner fallout field approximately two years after the detonation are summarized in Table I, where the existence of major inhomogeneities is clearly established. The 30% excess material found to be associated with the presence of grass clumps was not removed when the grass itself was cut away. Such inhomogeneities were not found in a nearby location where vegetation was larger, but where the soil surface was covered with rocks of about 4 cm diameter. This implies that there has in fact been a substantial net movement of deposited radioactive material to positions of shelter against erosive forces as opposed to the other possibility that the debris was originally deposited in such a manner.

Baneberry Venting

The fallout pattern from the Baneberry venting was primarily to north-northeast of surface ground zero (Fig. 1). Station 4 was located as close as practical to the area of maximum deposition; Station 6 was chosen as a background sampling location. A total of 467 air filter samples were analyzed with 150 exposed at Station 4. A Kruskal-Wallis one-way analysis of variance by ranks [12] was performed to determine if the samples from the six stations or all possible groupings of stations taken 3, 4, or 5 at a time were drawn from the same or identical populations of air activity values.

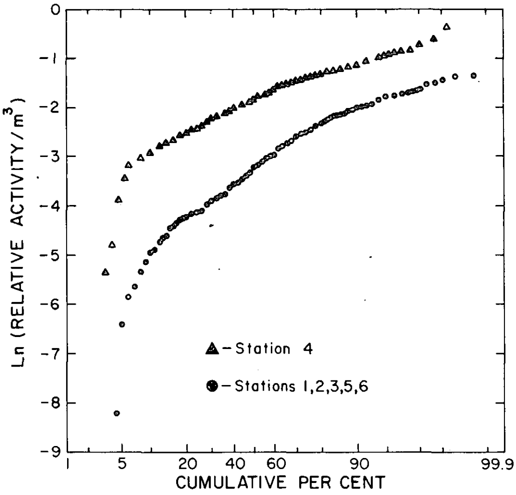

FIG.7. Log-probability plot of the air activity levels measured three to twelve months after the Baneberry venting.

The results are unequivocal that only the air activity values measured at Station 4 were different from the others ($p = < 0.001$), or stated another way, air activity levels at Stations 1, 2, 3, and 5 could not be distinguished from those measured at the background location, Station 6.

The measured air activity values are shown in Fig. 7 where the natural logarithms of the activity values are plotted against the cumulative percent of the samples on a probability scale. The medians of the two distributions differ by a factor of 4; the "background" distribution has a greater variance than does the distribution of samples from Station 4.

A similar analysis was performed for the 243 deposition measurements made at the six stations. Results are that the samples collected at each station were not drawn from the same or identical populations ($p = < 0.05$). A similar plot of these data is shown in Fig. 8. For clarity, only three stations are plotted. Values for the remaining three stations would lie between those shown for Stations 5 and 6. There is a much greater station difference for deposition values than was observed for air activity levels. The median values for Stations 1 through 6 have the approximate ratios of 2:3:4:160:8:1.

Several factors were investigated for their influence upon the measured air activity and deposition levels. Precipitation occurred while 30 of the air filter samples were running. These 30 values were classified into two categories according to whether they exceeded the median value of their respective group. The 30 values were evenly divided into two groups of 15 by this method, so it was concluded that precipitation had no significant effect on the measured air activity levels. Precipitation did, however, have a substantial effect on measured deposition levels. Of the 27 cases during which precipitation fell, 26 were above the median of their respective station. The one-tailed probability of observing this and even more extreme outcomes was calculated by use of the Fisher exact probability test [12]; the result is 5×10^{-8}. This effect, however, probably has negligible bearing on the question at hand,

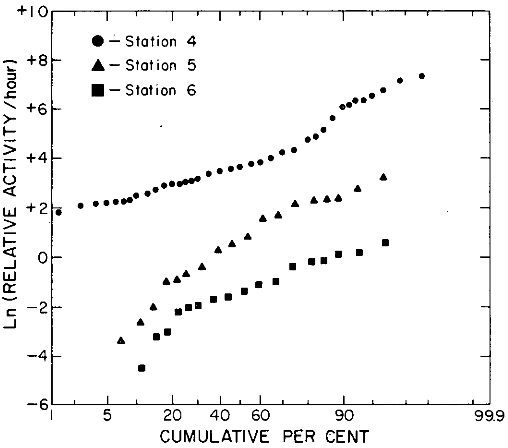

FIG.8. Log-probability plot of the deposition levels (primarily due to saltation) measured three to twelve months after the Baneberry venting. For clarity, levels measured at three of the six stations have been omitted from the plot.

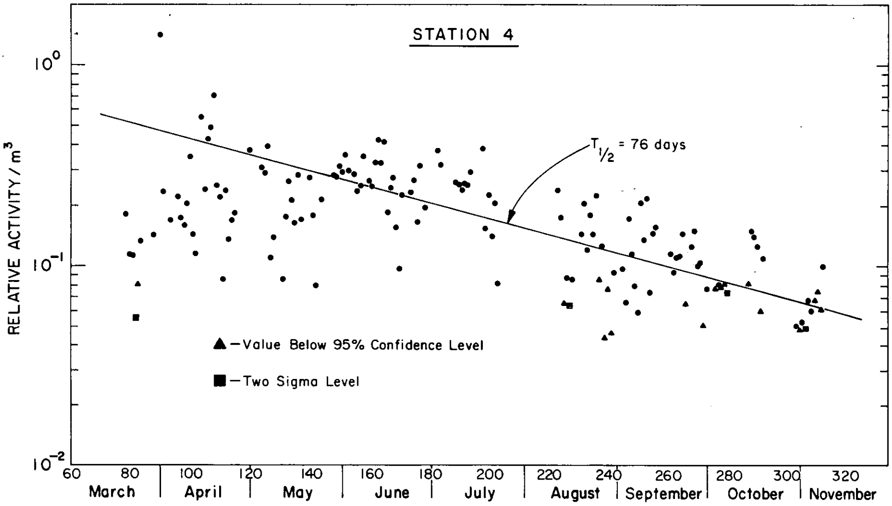

FIG.9. Gross-gamma air activity levels as a function of time at Station 4 three to eleven months after the Baneberry venting. The solid line is a least-squares fit to the data points.

i.e., the movement of surface-deposited radioactivity in suspension or saltation, but reflects the well known cleansing effect of precipitation on tropospheric air masses. For this reason, those deposition samples so affected were not considered in further data analysis.

The air activity levels measured at Station 4 are plotted in Fig. 9 as a function of time during the experiment. Fig. 10 is a similar plot of the deposition (saltation) measurements recorded at Station 4.

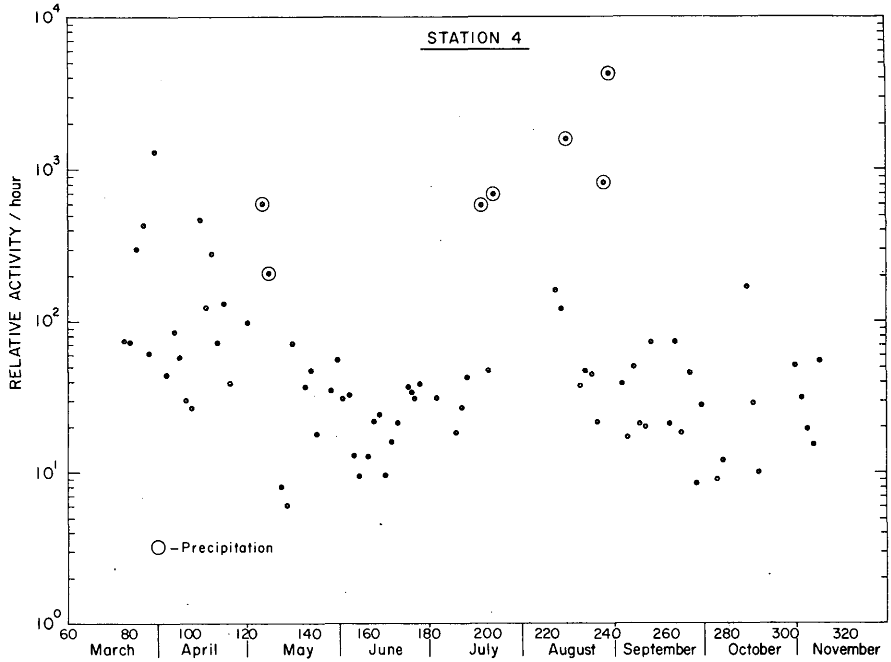

FIG. 10. Gross-gamma deposition levels as a function of time at Station 4 three to eleven months after the Baneberry venting.

The wind measurements were processed in several ways for later correlation studies. Fig. 11 is a plot of the overall directional dependence of the wind during the period of the experiment. The angle plotted is the direction from which the wind blew. The scale, however, has been rotated 180° so that if one mentally places north in the usual position at the top of the plot, the result is the direction in which the winds were blowing. The magnitude scale is in percent of the time that the winds were in that direction. A large fraction of the time the winds were blowing towards the southeast; the strong winds, however, blew predominately towards the north.

Several correlation studies were performed in an effort to identify predictors of resuspended air activity and saltation movement. Nonparametric statistical tests [12] were used throughout as none of the variables under study were distributed normally or lognormally. Independent variables investigated were time and the following hourly summaries of wind measurements averaged over the appropriate sampling periods: wind speed (u), u^2, u^3, peak gust (g), g^2, g^3, resultant vector wind speed (vu), vu^2, vu^3, resultant vector peak gust (vg), vg^2, and vg^3.

Results of correlation calculations with time are given in Table II. The significant correlations obtained for the air activity levels at Stations 1, 2, 3, 5, and 6, which represent background values of atmospheric radioactivity, are fortuitous due to the sampling period involved which

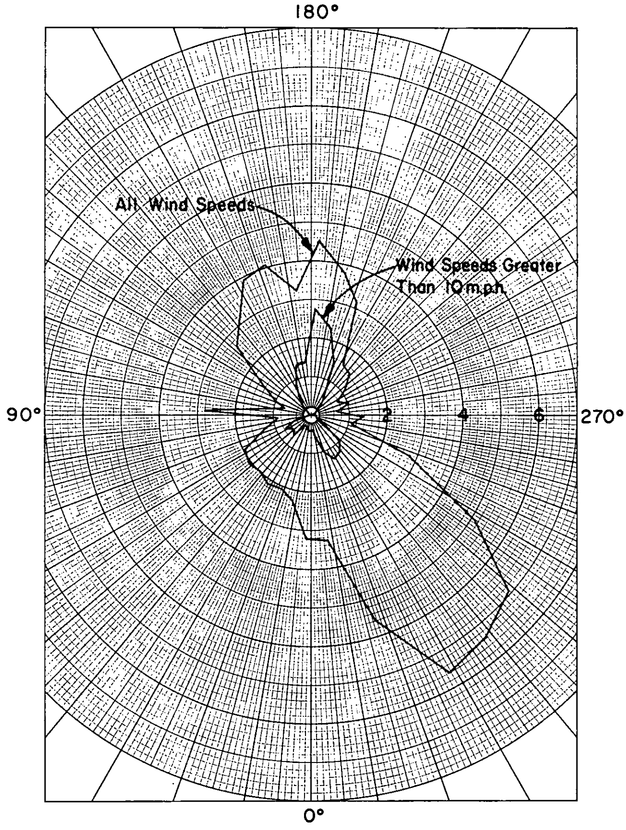

FIG.11. Wind patterns observed in the Baneberry experimental area during the months of March through December, 1971.

coincided with the descending side of the annual cycle of tropospheric radioactivity levels. The result for Station 4, -0.56, indicates that time is a powerful indicator of resuspended air activity as was observed following Project Schooner and Operation Plumbbob [4]. A least-squares fit of the Station 4 air activity data to a single exponential function was made using Bevington's CURFIT routine [13]. The result is the solid line shown in Fig. 9; a half-time of 76 days was calculated which is twice as long as the value derived from the Project Schooner data. As the median of the air activity values at Station 4 was only a factor of four above the median of the background stations, a correction to the above fit was calculated to account for the simultaneous decrease with time of the background. To accomplish this, the pooled data from Stations 1, 2, 3, 5, and 6 were fit to a single exponential function. A good fit to the data is provided by the equation:

$$A(t) = 0.14 \exp(-0.693\ t/110\ \text{days})\ \text{counts/min}\ \text{m}^3$$

TABLE II. SPEARMAN RANK CORRELATION COEFFICIENTS BETWEEN TIME AND THE VARIABLES INDICATED

Station	Air Activity		Deposition	
	r_s	p	r_s	p
1	-0.58	< 0.00003	-0.76	< 0.00003
2	-0.57	< 0.00003	a	
3	-0.35	0.0019	-0.36	0.019
4	-0.56	< 0.00003	-0.38	0.0008
5	-0.39	0.0021	a	
6	-0.35	0.0021	a	
1,2,3,5,6	-0.46	< 0.00003	b	

[a] $p = > 0.05$

[b] Not calculated

The Station 4 data were then refit to a two component exponential function with one component as above. The new result, thereby corrected for background variation, gives a half-time of 66 days which is still substantially higher than that derived from the Schooner data.

Although not shown in Table II, the twelve wind variables listed previously were also tested for correlation with time. None of them were significantly correlated even at the $p = 0.10$ level.

Spearman correlation coefficients were calculated for the air activity levels measured at each station versus the twelve wind parameters. Only one coefficient was significant at the 0.05 level: the correlation with vu at Station 5. However, since a total of 72 coefficients were calculated we expect three or four coefficients to be significant at the 0.05 level due to chance fluctuations alone. Although these correlations were unrevealing, there are some individual indications of the influence of wind parameters on the resuspended air activity measurements at Station 4. The highest value recorded during the study was on the 90th day of the year, and this value exceeded by an order of magnitude each of the seven previous measurements in the study. This was also the first period of strong winds in the study; the average wind speed while this sample was collected was 20 miles/hour with an average hourly peak gust of 30 miles/hour. This type of volatile response was typical of the results during the Project Schooner measurements. However, the highest wind speed during all of the Baneberry measurements was recorded on day 289 when the average wind speed was 22 miles/hour with an average hourly gust of 36 miles/hour. In contrast to the previously mentioned measurement, the air activity level during this time period was essentially the same as the measurement which preceded it when the average wind speed was only 11 miles/hour.

TABLE III. SPEARMAN RANK CORRELATION COEFFICIENTS BETWEEN THE AMOUNT OF MATERIAL MOVING IN SALTATION AT STATION 4 AND WIND PARAMETERS

Parameter	r_s	p	Parameter	r_s	p
u	0.32	0.004	g	0.22	0.03
u^2	0.39	0.0008	g^2	0.31	0.007
u^3	0.44	0.0002	g^3	0.38	0.002
vu	a		vg	a	
vu^2	0.23	0.03	vg^2	a	
vu^3	0.35	0.002	vg^3	0.23	0.03

[a] $p = > 0.05$

TABLE IV. SPEARMAN RANK CORRELATION COEFFICIENTS BETWEEN THE AMOUNT OF MATERIAL MOVING IN SALTATION AT ALL SIX STATIONS AND THE MOST SIGNIFICANT WIND PARAMETERS

Station	u		u^2		u^3	
	r_s	p	r_s	p	r_s	p
1	0.38	0.03	0.41	0.02	0.43	0.01
2	0.49	0.007	0.52	0.002	0.53	0.003
3	0.40	0.02	0.39	0.02	0.39	0.01
4	0.32	0.004	0.39	0.0008	0.44	0.0002
5	0.53	0.009	0.54	0.006	0.57	0.003
6	0.40	0.06	0.49	0.03	0.49	0.03

Correlation studies with the saltation (or deposition) measurements were more rewarding. At Station 4 a highly significant negative correlation with time was observed (Table II). As is shown in Fig. 10, however, the time dependence is quite different from that of the air activity measurements. The amount of material moving in saltation decreased much more rapidly and then leveled off for the remainder of the experiment. Significant correlations were obtained between the saltation measurements and wind parameters which are tabulated in Table III. The highest and most significant correlation is with the average of the cube of the wind speed. This is the functional relationship that had been previously established by investigators studying the erosion of sand and soil [6,7]. Similar results were obtained at all six sampling stations; the relevant data are given in Table IV.

Bagnold [6], in his studies on sand movement, showed that the flow of particles moving in suspension should also be related to u^3. The flow of particles moving in suspension is not readily measured in an open field; measurements of concentration, however, are presumably equal to the flow divided by the wind speed. Therefore, under conditions of constant meteorology at least, the concentration of material moving in suspension should be related to u^2. Healy and Fuquay [14] have done an approximate theoretical analysis of this problem; for an area source infinite in extent crosswind but finite in extent downwind, the concentration of particles moving in suspension was derived to be a function of u^2/Λ where Λ is the deposition velocity. Λ, however, is also a function of u [15]. We would therefore expect, under the given assumptions concerning the source configuration, that the concentration of resuspended material should be a function of u.

Station 4 was located as close as possible to the center of the Baneberry fallout field in an attempt to approximate an infinite area source and to remove the dependence upon wind direction. The predicted relationship between u and resuspended air activity was not observed. It is not known, however, how well the location truly approximated an infinite source.

Although no meteorological variables were identified as predictors of resuspended air activity, it was established that the time since deposition is a valid predictor over periods at least as long as a year following the deposition. For estimating average air concentrations, it is undoubtedly far more important than any meteorological variable. Although specific data are lacking, this is very likely not true several years post deposition.

We are presently starting a much more elaborate resuspension study in another area at the Nevada Test Site which was contaminated with ^{239}Pu fifteen years ago. In the center of this area, the concentration in air of ^{239}Pu is 10^2 to 10^3 times higher than that due to world-wide fallout. The current existence of such levels is presumptive evidence that the apparent exponential decrease of the concentration of material moving in

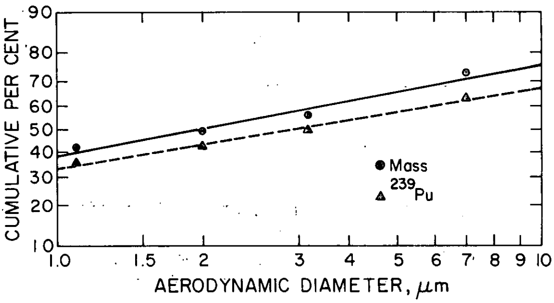

FIG.12. Log-probability plot of the aerodynamic particle size distributions of the total mass and the ^{239}Pu trapped by a 20 ft^3/min five-stage cascade impactor. Sampling was done in an open desert area at the Nevada Test Site which was contaminated with plutonium fifteen years ago.

suspension does not operate indefinitely. (A half-time of 100 days over a fifteen year period would reduce initial concentrations by some sixteen orders of magnitude.)

An ultra-high-volume (2000 m^3/h) air sampler has been developed for the ^{239}Pu resuspension program to meet the objective that quantifiable air samples be obtained on the same time scale that major diurnal meteorological changes take place. This will greatly enhance the ability to investigate the influence of a variety of meteorological variables on the rate at which material is suspended from the soil surface.

Some initial data on the particle size distributions of the total mass and the ^{239}Pu moving in suspension in the plutonium study area are shown in Fig. 12. These data were obtained by operating a 20 ft^3/min five-stage cascade impactor for a 35 day period. The median diameters for mass and ^{239}Pu are 2.0 and 3.2 μm, well within the respirable range.

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DISCUSSION

W.O. SCHIKARSKI: Do you see any possibility of extrapolating your resuspension measurements from the desert pavement to the surface conditions normally obtaining in the surroundings of nuclear installations?

P.L. PHELPS: We certainly hope to develop a generalized model, for which we shall naturally have to consider a variety of soil types. At the Nevada Test Site we are carrying out, or planning to carry out, experiments in five areas with differing soil and plutonium characteristics. In addition, if our desert experiments are successful, we shall perform studies on more typical soils using tracer techniques.

EQUATION EMPIRIQUE TRADUISANT L'EPURATION DES EAUX DE SURFACE

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Abstract–Résumé

AN EMPIRICAL EQUATION FOR THE PURIFICATION OF SURFACE WATERS.

An empirical equation describing the kinetics of surface water purification on sediments is given. The equation is of the type $C = C_0 (1 + t/a)^n$, where C_0 and C are respectively the concentration of the radioelement in water initially and after time t; a and n are parameters estimated by the least squares method and based on data from different model experiments, suspended sediments, stagnant water tanks and large pools. Values of these parameters are given for ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{85}Sr, ^{106}Ru, ^{137}Cs, ^{141}Ce and ^{203}Hg. The advantages of this simple empirical equation are discussed. Applications to other pollutants are foreseen.

EQUATION EMPIRIQUE TRADUISANT L'EPURATION DES EAUX DE SURFACE.

On rend compte de la cinétique d'épuration des eaux de surface sur les sédiments par une équation empirique de la forme $C = C_0 (1 + t/a)^n$ où C_0 et C sont respectivement les concentrations initiale et au temps t du radioélément dans l'eau; a et n sont des paramètres ajustés par la méthode des moindres carrés et déterminés à partir de modèles expérimentaux divers: sédiments en suspension, bacs d'eau stagnante, bassins de grandes dimensions. On donne les valeurs de ces paramètres pour ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{85}Sr, ^{106}Ru, ^{137}Cs, ^{141}Ce, ^{203}Hg. Les avantages de cette équation empirique simple sont discutés. Etant donné son vaste champ d'application, son extension à d'autres polluants est envisagée.

INTRODUCTION

Pour chiffrer un phénomène aussi complexe que la cinétique de l'épuration d'une eau de surface comprenant toutes les espèces dissoutes, les sédiments en suspension, les microfaune et microflore habituels et toutes les pollutions en solution ou particulaires d'origine industrielle, urbaine et agricole, il est illusoire de se reporter aux équations de Langmuir, de Freundlich ou de Brunauer, Emmet et Teller (B.E.T.), même avec l'aide d'un puissant ordinateur, sous peine de voir les résultats obtenus entachés de 1000% d'erreur. Une voie qui permet d'aborder globalement, dans toute leur complexité, de tels phénomènes est la recherche d'une formulation purement empirique qui, s'affranchissant de toute loi physique établie, devra offrir en compensation l'avantage d'un très vaste champ d'application couvrant la totalité des cas de figure que peut présenter le sujet étudié. C'est une telle approche que nous avons choisie pour établir une équation empirique de la cinétique de fixation de neuf radioéléments à partir de nos expériences de laboratoire.

DISPOSITIFS EXPERIMENTAUX

Des expériences radioécologiques ont été réalisées dans des modèles réduits de dimensions très diverses comportant dans tous les cas de l'eau et des sédiments de l'Isère (caractéristiques physico-chimiques déjà publiées [1]) et dans presque tous les cas des plantes aquatiques ou semi-aquatiques. Les radioéléments dont nous avons voulu suivre le comportement sont les suivants: ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{85}Sr, ^{106}Ru, ^{137}Cs, ^{141}Ce, ^{203}Hg. Ces systèmes expérimentaux sont fermés, ce qui minimise l'importance des problèmes de dispersion dans la phase aqueuse. Nous avons pu vérifier en effet que l'homogénéisation du radioélément dans l'eau est immédiate en regard de la cinétique d'épuration.

Sédiments en suspension

Le plus petit dispositif est aussi le plus précis: il consiste en erlenmeyers de 50 ml contenant 40 ml d'eau et des sédiments maintenus en suspension par agitation vigoureuse sur plateau rotatif à 350 tours/min durant des temps croissants mesurés à partir de l'instant de la contamination par 1 ml de solution radioactive [1]. Un dispositif de filtration rapide sous vide permet de recueillir la phase aqueuse directement dans les flacons de comptage par spectrométrie γ en fin d'agitation. La durée de ces expériences varie de 0,5 à 192 min. Certaines ont été menées avec des sédiments non tamisés, d'autres avec les fractions des limons ou des sables fins. Nous avons volontairement diversifié les anions associés pour disposer d'un champ d'application de l'équation proposée le plus vaste possible.

Bacs d'eau stagnante

Des bacs en matière plastique de 30 litres contenant 6 cm de sédiments surmontés de 10 cm d'eau ont été utilisés pour suivre le comportement des radioéléments en présence de diverses espèces végétales aquatiques ou semi-aquatiques (Myriophyllum verticillatum, Elodea canadensis, Mentha aquatica, Potomogeton densus, Sium erectum, Arundo phragmites communis, Lemma minor, Ulothrix zonata, Callitriche polymorpha, Chara foetida, Iris pseudacorus) et animales (Asellus aquaticus, Planorbis, Gammarus pulex, Limnea stagnalis) [2]. Les sédiments ne sont pas en suspension et l'eau est aérée par un léger barbotage d'air. La durée de ces essais varie de 30 à 40 jours.

Bassins de grandes dimensions

^{60}Co, ^{85}Sr, ^{137}Cs ont été suivis dans une «piscine écologique» reproduisant une berge de l'Isère (fig. 1). Les caractéristiques de cet écosystème sont les suivantes: diamètre = 6 m, hauteur = 1 m, sédiments et terre: 32 t, eau: 3 m^3. En un an, quatre rejets ont été effectués dans la phase aqueuse et suivis jusque dans les nombreuses espèces aquatiques ou ligneuses implantées [2].

De même, un bassin de plus petite taille (3 m^3 avec 3 t de sédiments et terre et 135 l d'eau) [3] a permis l'étude du cheminement du nitrato-nitrosyl-^{106}Ru le long d'un profil de berge.

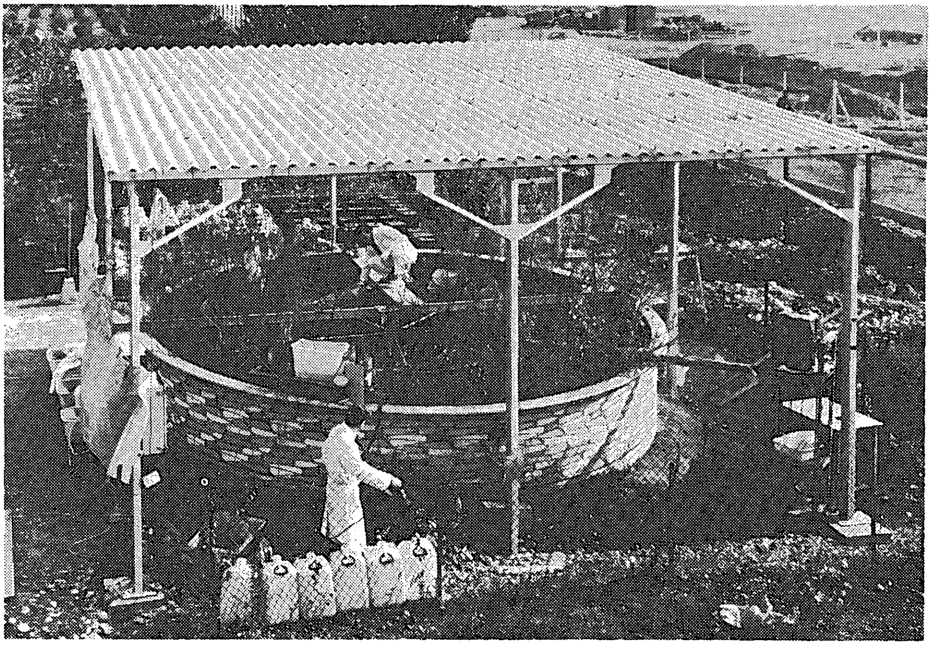

FIG. 1. Modèle de berge de l'Isère: piscine écologique.

EQUATION EMPIRIQUE

Allure des courbes d'épuration

Dans tous les cas étudiés, la courbe de la radioactivité de l'eau (ou de son logarithme) en fonction du temps présente une concavité tournée vers le haut. Des cas très différents sont représentés sur la figure 2: il s'agit d'une part de radioéléments s'adsorbant faiblement (^{60}Co et ^{85}Sr) et d'autre part de radioéléments s'adsorbant beaucoup et très rapidement (^{106}Ru, ^{137}Cs et ^{141}Ce). Sur des courbes très régulières présentant une concavité intermédiaire, nous avons remarqué une propriété attribuable à une fonction hyperbolique au sens large, ce qui nous a conduits, compte tenu de la valeur finie de l'ordonnée à l'origine, à une équation de la forme

$$C = C_0 \left[1 + \frac{t}{a} \right]^n$$

où C est la concentration du radioélément au temps t dans l'eau, C_0 est la concentration initiale du radioélément supposé instantanément homogénéisé dans l'eau au temps t = 0, t est le temps écoulé depuis la contamination, $a > 0$ et $n < 0$ sont deux paramètres à ajuster.

Un troisième paramètre nous aurait permis de satisfaire la condition $C \to C_\infty > 0$ quand $t \to \infty$. En fait, in situ, les matières adsorbantes sont loin d'être saturées [1] et nous pouvons toujours admettre soit que la variable t est convenablement bornée supérieurement, soit que C_∞ est négligeable.

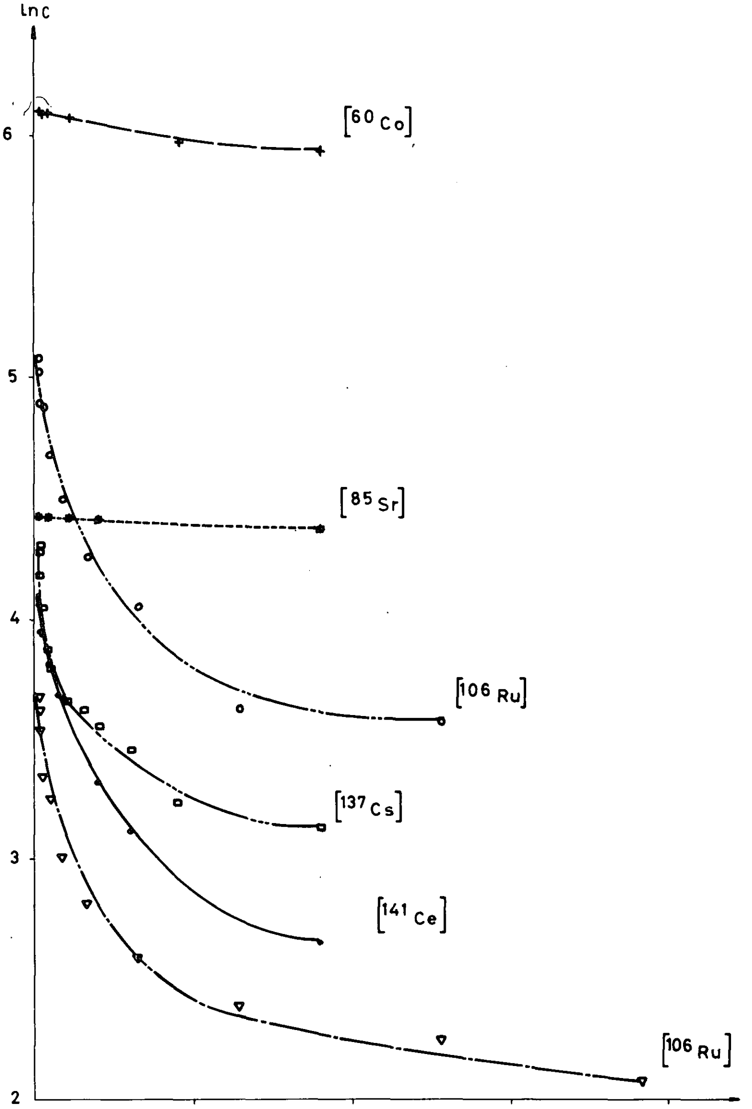

FIG. 2. Epuration de l'eau par des sédiments en suspension [1].

Ajustement par la méthode des moindres carrés

L'écriture de l'équation ci-dessus en deux points (t_1, C_1) et (t_2, C_2) conduit à un système de deux équations transcendantes à deux inconnues et ne permet donc pas d'évaluer les paramètres a et n. Par contre, considérant que

$$\ln C = \ln C_0 + n \ln \left[1 + \frac{t}{a}\right]$$

a pour dérivée

$$\frac{d(\ln C)}{dt} = \frac{n}{a+t} = p$$

les tangentes en deux points de la courbe ln C = f (t) que nous pouvons tracer ont pour pente

$$p_1 = \frac{n}{a+t_1} \quad \text{et} \quad p_2 = \frac{n}{a+t_2}$$

d'où les estimations graphiques assez grossières des paramètres:

$$a = \frac{p_2 t_2 - p_1 t_1}{p_1 - p_2} \quad \text{et} \quad n = \frac{p_1 p_2 (t_2 - t_1)}{p_1 - p_2}$$

Partant de cette première approximation, les paramètres non linéaires a et n voient leur valeur affinée par la méthode des moindres carrés. Cet ajustement se fait à l'ordinateur (IBM 360/50) par le sous-programme AMAGT (écrit en Fortran IV, niveau G) [4] qui réalise un compromis entre la méthode de Gauss et la méthode du gradient [5] en tenant compte du poids statistique (inverse de la variance) de chaque point expérimental. A chaque itération, les dérivées partielles de la fonction sont calculées:

$$\frac{\partial C}{\partial n} = C_0 \left[1 + \frac{t}{a}\right] \ln \left[1 + \frac{t}{a}\right] \quad \text{et} \quad \frac{\partial C}{\partial a} = - \frac{n\, t\, C_0}{a^2} \left[1 + \frac{t}{a}\right]^{n-1}$$

Les ordonnées ajustées, les transformées des ordonnées expérimentales sous la forme ln C, les transformées de la variable sous la forme ln (1 + t/a), les paramètres ajustés a et n ainsi que la matrice de covariance sont imprimés afin de permettre le contrôle de la validité de l'ajustement obtenu.

Contrôle de la validité de l'ajustement

Ayant obtenu le meilleur ajustement possible, nous devons ensuite vérifier s'il est satisfaisant. Les poids statistiques attribués à chacun des points expérimentaux peuvent parfois augmenter ou diminuer systématiquement en fonction du temps, conduisant à des valeurs des paramètres a et n parfaitement correctes mais telles que la régression linéaire entre ordonnées expérimentales et ordonnées ajustées ait une pente non homogène à l'unité. Nous ne baserons donc pas forcément notre contrôle sur cette régression qui ne peut s'appliquer qu'aux cas où l'expérience a conduit à l'homoscedasticité des ordonnées. Par contre, considérant que $\ln C = \ln C_0 + n \ln (1 + t/a)$, la régression entre les transformées des ordonnées expérimentales sous la forme de leurs logarithmes népériens et la transformée de la variable sous

TABLEAU I. PRESENTATION DE QUELQUES-UNS DES AJUSTEMENTS OBTENUS

Elément	Forme Chimique	Type d'expérience	Végétaux aquatiques	a	n	Contrôle [a]
^{51}Cr	EDTA	Suspension	Néant	6,17	- 0,005	***
^{59}Fe	Citrate	Suspension	Néant	91	- 0,056	**
^{60}Co	Chlorure	Suspension	Néant	15,8	- 0,085	***
^{85}Sr	Nitrate	Suspension	Néant	92	- 0,076	**
^{106}Ru	Nitrato - nitrosyle	Suspension	Néant	0,39	- 0,299	***
^{137}Cs	Chlorure	Suspension	Néant	0,34	- 0,245	***
^{141}Ce	Chlorure	Suspension	Néant	1,52	- 0,339	***
^{203}Hg	Acétate	Suspension	Néant	16,8	- 8,060	***
^{54}Mn	Chlorure	Bacs	Ulothrix	0,017	- 1,307	**
^{54}Mn	Chlorure	Bacs	Plusieurs [2]	0,058	- 1,196	**
^{85}Sr	Nitrate	Bacs	Néant	1,78	- 0,306	***
^{85}Sr	Nitrate	Bacs	Roseaux	2,19	- 0,318	***
^{106}Ru	Nitrato - nitrosyle	Bacs	Potamots	0,36	- 0,929	***
^{106}Ru	Nitrato - nitrosyle	Bacs	Ulothrix	0,12	- 1,285	***
^{144}Ce	Chlorure	Bacs	Néant	3,03	- 1,586	***
^{144}Ce	Chlorure	Bacs	Roseaux	0,94	- 1,959	***
^{203}Hg	Chlorure	Bacs	Mille - feuilles	14,2	- 1,512	***
^{203}Hg	Chlorure	Bacs	Callitriches	14,6	- 1,453	***
^{203}Hg	H_3C Hg Cl	Bacs	Callitriches	38	- 2,783	***
^{203}Hg	H_3C Hg Cl	Bacs	Elodées	17,5	- 1,528	***
^{60}Co	Chlorure	Bassins	Plusieurs [2]	0,012	- 0,476	**
^{85}Sr	Nitrate	Bassins	Plusieurs [2]	2,46	- 0,451	**
^{137}Cs	Chlorure	Bassins	Plusieurs [2]	0,157	- 0,702	***
^{106}Ru	Nitrato - nitrosyle	Bassins	Plusieurs [3]	0,049	- 0,629	***

[a] *** : ajustement très hautement significatif, $\alpha = 0{,}001$; ** : ajustement hautement significatif, $\alpha = 0{,}01$; * : ajustement significatif, $\alpha = 0{,}05$ (deux cas de courbes à 5 et 6 points seulement, non présentées dans ce tableau).

la forme ln $(1 + t/a)$ doit toujours être linéaire et de pente homogène au paramètre n. Le premier point est vérifié par un test de F, rapport du carré moyen dû à la régression au carré moyen résiduel, et le second par le fait que le paramètre n se trouve effectivement dans l'intervalle de confiance du coefficient de cette régression. De plus, le coefficient de corrélation linéaire nous garantit une bonne intensité de la liaison entre ordonnées expérimentales et ajustées.

RESULTATS ET DISCUSSION

Tableau récapitulatif (tableau I)

Il serait surabondant de présenter ici la totalité de nos résultats (soixante-cinq expériences très variées). Nous en avons choisi vingt-quatre pour témoigner du vaste champ d'application de l'équation proposée. Nous stipulerons en outre que le seul cas où l'ajustement obtenu ne s'est pas avéré statistiquement significatif au seuil de probabilité $\alpha = 0,05$ a été celui d'une courbe qui n'était construite que sur quatre points expérimentaux seulement.

Caractère général et prévisionnel de l'équation

Parmi les soixante-cinq cas que nous avons mis en équation, il en est qui correspondent à des systèmes expérimentaux de tailles très diverses. Ainsi, le rapport de la masse active de sédiments (premier centimètre seulement) au volume d'eau surnageante est de 100 grammes secs par litre pour les bacs, de 60 g/l pour le bassin et de 40 g/l pour la piscine. Il est de 1 g/l pour les sédiments en suspension. Il nous faut également rappeler que les charges en entraîneur s'étageaient de la micromole à la picomole par litre et que les conditions climatiques des expériences étaient soit celles du laboratoire, soit celle de la Cluse de Grenoble en été comme en hiver.

Ainsi, la démarche que nous proposons pour prévoir l'épuration d'une eau de surface polluée commence par une étude hydrologique du site pour définir un modèle représentatif; c'est dans ce modèle que la cinétique d'épuration sera suivie avec la plus grande précision possible et c'est à partir des données expérimentales recueillies que l'ajustement par moindres carrés sera conduit, définissant les deux paramètres de l'équation empirique proposée qui, son vaste champ d'application nous en apporte la conviction, restera valable dans tous les cas possibles, alors que, comme le signalent Picat et coll. [6], le modèle d'adsorption monocouche de Freundlich valable pour ^{137}Cs ne l'est plus pour ^{85}Sr.

S'agissant de définir la concentration du polluant à la distance x du point de rejet, deux cas sont à considérer dans l'utilisation de notre information à des fins prévisionnelles:

L'hypothèse d'une homogénéisation immédiate est acceptable et C_0 représente alors la concentration du polluant après dilution instantanée dans le cours d'eau, la correspondance entre les variables temps t et distance x étant directe moyennant la connaissance du temps moyen de séjour de la veine fluide. Notons l'intérêt qu'il y aurait à connaître et prendre en compte la distribution des temps de séjour des particules pour pouvoir ultérieurement estimer le transport de la fraction du polluant fixé sur les sédiments ainsi que son intervalle de confiance. Guizerix et coll. [7] donnent des exemples d'évaluation d'une telle distribution.

L'hypothèse ci-dessus est à écarter et, C_0 étant la concentration en polluant dans la bouche de rejet, nous devons pondérer la valeur de C par un terme de dilution directement fonction de la distance x pour $0 < x \leq D$, distance dite de bon mélange. Ce terme s'écrit $qD/(qD + Qd)$, où q et Q sont respectivement les débits de l'égout de rejet et de la rivière et où la distance d s'exprime en fonction de la vitesse d'écoulement de la rivière V par la relation $d = Vt$. Pour évaluer D, plutôt que l'équation de diffusion classique qui donne des solutions particulières fort complexes [8, 9], nous préférons retenir, comme le font les praticiens de la Section d'application des radioéléments du Centre d'études nucléaires de Grenoble [10], la formule expirique proposée par André, Mazeran et Dutillet (Electricité de France) : $D = 10\, b\, \sqrt[3]{Q}$ où D est la distance minimale de bon mélange en m, b la largeur moyenne de la rivière en m et Q le débit de la rivière en m^3/s. Cette équation, malheureusement non homogène, a cependant été établie sur un très grand nombre de mesures et convient parfaitement pour des rivières de 50 à 200 m de large à régime non torrentiel. Pour $x > D$, le terme de dilution sera supprimé et notre équation sera reprise à partir du point de bon mélange pris comme nouvelle origine avec C_0 concentration du polluant en ce point.

CONCLUSION

Chaque fois qu'une étude du comportement d'une pollution in situ est soit trop imprécise, soit trop onéreuse, soit le plus souvent impossible du fait du danger qu'elle représenterait en elle-même, nous suggérons d'accomplir une telle démarche. Nous soulignerons enfin l'intérêt qu'elle présente pour l'étude de la cinétique de fixation d'autres polluants tels que les toxiques, notamment les organométalliques (grâce à l'utilisation de molécules marquées, par exemple). La variabilité des paramètres a et n est essentiellement dépendante du modèle choisi et non du radioélément considéré sous une forme chimique donnée. L'équation empirique proposée pose de toute façon le problème du choix d'un modèle hydrologique adapté aux conditions particulières que l'on veut considérer.

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BEHAVIOUR OF RADIONUCLIDES IN AQUATIC SYSTEMS

(Session III)

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THE FATE OF RADIONUCLIDES DISCHARGED FROM A PWR NUCLEAR POWER STATION INTO A RIVER

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Abstract

THE FATE OF RADIONUCLIDES DISCHARGED FROM A PWR NUCLEAR POWER STATION INTO A RIVER.

A detailed radiological study of a river in the vicinity of a large PWR nuclear station was conducted to determine the impact of the station on the environment and surrounding population. The wastes from the reactor are discharged into a 1.8-km long canal that empties into a large river. During a 1-yr period samples of algae, plankton, aquatic grasses, fish, bottom sediment and water were collected from the discharge canal and from the river below and above the canal. Because the temperature of the canal water can be as much as 12.7°C higher than the river water, normal habits and growth characteristics of fish and other biolological life in the canal and surrounding river waters are altered. The influence of this atypical situation on the uptake and concentration of radionuclides in the fish and aquatic plants is discussed.

Radionuclides found in samples of algae, grasses, and plankton as a consequence of the discharges from the nuclear power station included ^{3}H, ^{14}C, ^{54}Mn, ^{57}Co, ^{60}Co, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{131}I, ^{134}Cs, ^{137}Cs and ^{144}Ce. The fish were dissected for muscle, heart area, stomach, gut, bones, and liver plus kidney. Special care was exercised to remove all bones from the muscle. Radionuclides detected in the fish samples were ^{3}H, ^{14}C, ^{58}Co, ^{60}Co, ^{131}I, ^{134}Cs and ^{137}Cs. Concentration factors for water to organism were calculated and their implications are discussed. The radiation dose to people eating the fish was estimated to be below 10 mrem/yr.

Introduction

Radionuclides discharged from a nuclear power station into the aqueous environment may result in some radiation exposure to the surrounding population. As the number of large nuclear power stations increase along rivers, lakes, and oceans, the need to understand better the fate of these radionuclides in the environment and to determine the resulting population exposures becomes increasingly important. Hence, a 1-year radiological study was conducted on the aqueous environment of a large pressurized water reactor (PWR) during 1970-1971 to determine the impact of liquid effluents from the station on the environment and the surrounding population.

The nuclear power station chosen has a generating capacity of 590 megawatts electrical (MWe) and had been operating since August 1967. The station is located on the Connecticut River 35 km south of Hartford and 26 km above the mouth of the river. Tidal effects on the river are approximately 1 m at the station, and salt water approaches to within about 6.5 km downstream of the station at high tide. The liquid effluents from the station are discharged into a canal in which water flows at the rate of 1.41×10^{6} liters/min. This coolant water empties into the Connecticut River 1.8 km below the station.

The canal offers an unusual habitat for studying radionuclide uptake of fish. Fish are attracted to and remain in the heated effluent canal

(Δ T 12.7°C) during the colder months of the year. Occupancy of this "warm-water" habitat results in an abnormal seasonal metabolism as well as feeding and activity behavior. Thus, it has been shown that brown bullheads (catfish) in a control area (a cove 3.2 km below any detectable heated water from the plant) all had empty stomachs in December when the water temperature was 2°C; a comparable sample from the canal at this time showed that 30% of the fish had fed [1]. In March when the control area was 4°C, 23% of the bullheads had fed, while on the same date 51% of those in the canal were feeding actively. During the spring and early summer months when the water temperatures increase, fish begin to leave the canal and at temperatures above 35°C practically none remain. Due to their abnormal habits and the crowding of the fish in the canal (an average of 30,000 has been estimated to be in residence during the winter), their physical condition is somewhat impaired and the uptake by them of radionuclides through the food chain may be atypical [1].

Methods

The fish species that were collected were white catfish (*Ictalurus catus*), brown bullhead (*Ictalurus nebulosus*), and carp (*Cyprinus carpio*). These composed more than 95% of the winter fish species population in the canal [2]. Between 4 and 10 fish per species were collected twice during the winter, in December and March. The size of these catfish and bullheads ranged from 250 g to 1000 g, and the carp from 300 g to 4000 g. Additional biological samples collected from the aquatic environment of the station included aquatic plants (*Vallisneria americana*), algae, and plankton. The algae consisted mostly of blue-green algae (*Lyngbya*, *Oscillatoria*, *Phormidium*) with some green algae (*Cosmarium*) and diatoms (*Nitzchia*, *Melosira*). The plankton samples, including some detritus, were not identified.

The procedures and methods employed for the radionuclide analysis of biological samples have been described in detail [3]. Briefly, fish samples were dissected for muscle, kidney and liver, thyroid (head kidney and heart area), and bone, and the tissues were combined by species of fish for each sampling period. All samples were analyzed directly for photon emitters by spectrometry with a 10 x 10-cm NaI(Tl) detector or an 11-cm^3 Ge(Li) detector. Samples were also analyzed with a NaI(Tl) gamma-ray coincidence/anticoincidence system. Radiochemical analyses were performed to measure ^{90}Sr and ^{32}P; ^{3}H and ^{14}C were determined by treating samples in a combustion train, collecting water and CO_2, and measuring the radioactivity with liquid scintillation and gas counting techniques.(1)

Results

Because concentrations of radionuclides discharged by the plant were in many cases below minimum detectable levels in canal and river water, concentrations were determined from concentrations in waste tank samples before discharge. In Table I are listed the average radionuclide concentrations calculated to be present in the discharge canal during the period of study. The average concentrations listed in the first data column are based on monthly discharge volumes and activities reported by the station [4]. The average concentrations listed in the second data column were calculated from analyses of samples from the primary system (test tanks) and the secondary system (steam generator blowdown and leakage), the volume

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(1) We thank E. J. Troianello, U. S. Environmental Protection Agency, Winchester, Mass., for the ^{3}H and ^{14}C analyses.

TABLE I. AVERAGE RADIONUCLIDE CONCENTRATION IN DISCHARGE CANAL (pCi/liter)

Isotope		Calculated from Station Reported Values(a)	Calculated from Measured in-plant Samples(b,c)
12.3 -yr	^{3}H	10,400	19,400
5730 -yr	^{14}C	NR(d)	0.074
15 -h	^{24}Na	NR	0.086
14.3 -d	^{32}P	NR	0.043
27.8 -d	^{51}Cr	NR	0.023
313 -d	^{54}Mn	0.48	0.31
2.7 -yr	^{55}Fe	NR	0.80
270 -d	^{57}Co	NR	0.013
71.3 -d	^{58}Co	4.1	1.1
44.6 -d	^{59}Fe	0.04(e)	0.021
5.26-yr	^{60}Co	0.66	0.49
28.5 -yr	^{90}Sr	NR	0.0015
65 -d	^{95}Zr	NR	0.0046
35.1 -d	^{95}Nb	NR	0.0065
66.2 -h	^{99}Mo	NR	0.13
253 -d	^{110m}Ag	NR	0.0075
8.06-d	^{131}I	2.3	19.4
20.9 -h	^{133}I	1.7	9.6
2.07-yr	^{134}Cs	NR	0.40
6.7 -h	^{135}I	NR	4.0
13 -d	^{136}Cs	NR	0.094
30.2 -yr	^{137}Cs	0.58	0.42

Note: Approximately 23 µCi of ^{133}Xe and 0.10 µCi of ^{135}Xe are discharged annually in the water, but aeration would be expected to expel these nuclides; ^{89}Sr was not detected in the discharge canal (< 0.1 pCi/liter).

(a) Average for 1970 and 1971, based on discharges reported by the station and a total dilution of 6.3×10^{11} liters/yr.

(b) Average of concentration in two samples from primary system discharge (7.2×10^{6} liters/yr) and in six samples from secondary system discharge (1.8×10^{7} liters/yr), with a total dilution volume of 6.3×10^{11} liters/yr.

(c) To calculate average concentration in Connecticut River, divide by 25.

(d) NR - Not reported.

(e) Reported for only one 6-month period.

TABLE II. RADIONUCLIDE CONCENTRATION MEASURED IN FISH (pCi/kg)

Isotope	Muscle Measured	Muscle Predicted	Gut	Kidney + Liver
		Canal Fish		
^{3}H	2600-7600	10,400(0.90)[a]	NM[c]	700-1200
^{14}C	320-440[b]	630(4500)	NM	400-625[b]
^{55}Fe	< 90	80(100)	NM	600-3600
^{58}Co	< 25	80(20)	< 20-290	< 40
^{60}Co	< 15	13(20)	< 20-140	< 25
^{131}I	< 40	34(15)	< 60-1500	< 50-1800
^{134}Cs	< 20-145	160(400)	< 20-200	ND
^{137}Cs	110-320	250(400)	75-380	150-430
		Control Fish		
^{3}H	570		NM	NM
^{14}C	300[b]		NM	300[b]
^{55}Fe	< 90		NM	3800
^{137}Cs	80		70	130

(a) Concentration factors: see ref. [5].
(b) Assumes fish tissue to be 5% carbon [5].
(c) NM - Not measured; ND - not detected.

discharged of each and the total available dilution volume. Five of the eight values obtained by the two procedures agreed within a factor of two, but ^{131}I, ^{133}I, and ^{58}Co values differed considerably. Because the average values based on monitoring all discharges should be superior to those based on the occasional periodic samples, concentrations for the radionuclides given in data column 1 were used in the calculations. For radionuclides not reported by the station, however, the concentrations given in data column 2 were used. Concentrations at any time would differ considerably from the averages in Table I because primary system wastes are only discharged periodically, and the radionuclide contents of both wastes change with time.

The concentration of radionuclides measured in fish from the canal and in control fish (collected concurrently and 9 km up river from the discharge canal) are listed in Table II. The concentrations given for the canal fish result from both station discharge and fallout. No significant differences in concentrations of radionuclides were observed in the three fish species. No radioactivity was observed in the heart area (thyroid); ^{131}I was the only significant radionuclide related to plant discharge observed in the kidney and liver. The gut generally contained the highest activities of all dissected tissues. This might be due in some cases to the contents of the gut, although, as mentioned above, the gut was often empty. Muscle, the most important tissue because it is eaten, contained a measurable excess above background of ^{3}H, ^{14}C, and ^{137}Cs in all fish. The muscle of some fish also contained measurable quantities of ^{134}Cs.

To determine the feasibility of predicting the concentration of radionuclides in fish muscle from the concentration in water, the average water concentrations listed in Table I for these nuclides were added to the background concentrations measured in a control water sample,(2) and the sums were multiplied by the appropriate concentration factors (CF). The resulting predicted radionuclide concentrations in fish muscle are given in the second data column in Table II. The concentration factors are given in parentheses. The predicted values in all cases are of the correct magnitude, and, except for ^{58}Co, within a factor of two of the measured ranges. Some disparity may be a result of the radionuclide concentrations in the fish not being in equilibrium with those in the water, the uptake of radionuclides from foods rather than water, and the atypical aquatic environment of the canal, which was discussed above. Considering these factors and the uncertainty in the concentration factors, which are based on numerous values obtained under a variety of conditions, agreement between measured and predicted concentration is reasonable and illustrates the feasibility of predicting concentrations in fish muscle, which is an important pathway for radionuclides to reach man.

In Table III are presented the radionuclide concentrations measured in algae, water grass, plankton, and bottom sediment. The concentrations given for the water grass and plankton are the results of plant discharge, in that the concentrations measured in control samples (shown in parentheses) have been subtracted. Because algae grew only in the discharge canal, no control sample was available for analysis. Consequently, the concentrations of ^{55}Fe, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{137}Cs, and ^{144}Ce include the contribution from fallout.

The plankton is exposed to plant discharges for a relatively short time--about 80 min during transit down the 1.8-km canal. Hence, even though equilibrium would not be realized between the radionuclide content of the plankton and the water in the canal, the results do illustrate the strong tendency of plankton, a principal food for fish, to concentrate certain radionuclides discharged by nuclear power stations. The water grass (_V. americana_) was collected a short distance up-river from the mouth of the canal and was exposed to canal discharges only at high tide. Consequently, it is not feasible to relate the radionuclide content of the canal water to that measured in the grass. The water grass does concentrate certain of the discharged radionuclides, principally ^{54}Mn, ^{58}Co, ^{60}Co, ^{90}Sr, and ^{131}I, and thus acts as a reservoir for these radionuclides in the aquatic environment. On the other hand, the algae had grown in the canal water for their lifetime and, if it can be assumed that the calculated water concentrations listed in Table I are applicable, concentration factors can be calculated for this mixture of green and blue-green algae. The concentration factors, pCi/kg(algae) per pCi/kg(water), are presented below. For those radionuclides present in fallout, the background water concentrations given above have been added to the concentrations listed in Table I.

$^{3}\mathrm{H} = 0.9$
$^{14}\mathrm{C} = 7 \times 10^{3}$
$^{54}\mathrm{Mn} = 23 \times 10^{3}$
$^{57}\mathrm{Co} = 28 \times 10^{3}$
$^{58}\mathrm{Co} = 10 \times 10^{3}$
$^{60}\mathrm{Co} = 32 \times 10^{3}$
$^{131}\mathrm{I} = 5 \times 10^{3}$
$^{134}\mathrm{Cs} = 3.5 \times 10^{3}$
$^{137}\mathrm{Cs} = 3.2 \times 10^{3}$

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(2) Concentrations measured in the control water sample were 0.05 pCi ^{137}Cs/liter and 1,200 pCi ^{3}H/liter. Carbon-14 was not measured, but was estimated by assuming there are 11 ppm carbon in fresh water with a specific activity of 6.08 pCi ^{14}C/g C [5].

TABLE III. RADIONUCLIDE CONTENT OF SAMPLES COLLECTED AT MOUTH OF DISCHARGE CANAL (pCi/kg)

Isotope	Algae	Water Grass	Plankton	Bottom Sediment
^{3}H	9,100	NM	NM	
^{14}C	1,200(b)	NM	210	
^{32}P	< 50	31	< 50	
^{54}Mn	11,000	140	1,240	200-1,400
^{55}Fe	41,000(a)	< 300(600)(c)	NM	
^{57}Co	370	< 5	< 40	
^{58}Co	39,000	120	3,600	800-11,900
^{60}Co	21,000	25	1,560	300-2,600
^{90}Sr	42(a)	26(< 10)	< 30(< 40)	
^{95}Zr	1,800(a)	< 15(< 20)	< 90(< 190)	
^{95}Nb	1,300(a)	< 30(85)	400(< 125)	
^{131}I	11,000	250	8,800	
^{134}Cs	1,400	< 15	500	300-1,500
^{137}Cs	2,000(a)	< 25(30)	< 150(340)	400-2,600
^{144}Ce	7,600(a)	< 80(230)	< 400(1100)	

(a) Background from fallout was subtracted from all values except these.
(b) Assumes algae to be 5% carbon [5].
(c) Background concentrations measured in control samples of water grass and plankton are given in parentheses.
(d) Strontium-89 was not observed in any samples.

The CF calculated for the cesium isotopes and the cobalt isotopes are in good agreement, and the CF calculated for ^{3}H is near the expected value of one. The lower CF calculated for ^{58}Co relative to that for the other cobalt isotopes may be the result of its much shorter half life and the time that had elapsed between plant discharge and sample collection. In addition to the role algae play as a food for fish in the food chain of man, the large concentration factors calculated above, as well as the ease of sample collection and analysis, make this type sample useful as an indicator of radionuclides in the aqueous environment.

Radionuclides that are discharged by the plant also accumulated in bottom sediments of the canal, particularly at the mouth where the velocity of the water is reduced and small particulate matter is deposited. The ranges in concentration measured at eight points in the mouth of the canal are given in the last column of Table III. The radioactivity was found, in general, to be associated with the fine particulate matter and organic materials. The extent of leaching or dissolution under conditions present in the river are not well understood, and the significance of these sediment deposits as to their contribution of radionuclides to man is not known. However, these deposits in the sediments of streams and rivers are a potential source of radionuclides to the aquatic environment and should not be ignored.

At this nuclear power station, the only pathway by which radionuclides discharged into the aquatic environment can reach man is through the consumption of fish. No other aquatic life in the immediate area is consumed by man nor is the water down river from the station used for drinking or public swimming. Shellfish that exist in the estuary and shad that briefly swim past the station each April to spawn up river were analyzed, but no radionuclides discharged by the station were detected.

For ^{90}Sr and ^{131}I, the daily intake corresponding to the population exposure of 170 mrem/yr for bone and 500 mrem/yr for thyroid is given directly by the Federal Radiation Council (FRC) [6]. For all other radionuclides, doses were computed from occupational exposure recommendations by the International Commission on Radiation Protection (ICRP) [7]. In this computation, the $(MPC)_w$ value given by the ICRP for a 168 hour week is converted to a daily intake by assuming an average water consumption of 2.2 liters/d [7]. These daily intakes, so calculated, would lead to doses of 5 rem/yr if the whole body is the critical organ, 30 rem/yr if the bone is the critical organ, and 15 rem/yr when any other organ, except gonads, is the critical organ [7]. When the daily intake is divided by these respective doses, a daily intake-dose rate relationship of pCi/d per rem/yr is obtained--the relationship used in this paper to estimate dose. An example of calculating this relationship is given below for ^{54}Mn.

The ICRP lists the GI(LLI) as the critical organ for ^{54}Mn and 10^{-3} μCi/cm^3 as the $(MPC)_w$ value [7]. Hence, the relationship will be

$$\frac{10^3 \text{ pCi/cm}^3 \times 2200 \text{ cm}^3\text{/d}}{15 \text{ rem/yr}} = 147{,}000 \ \frac{\text{pCi/d}}{\text{rem/yr}} \qquad (1)$$

Dividing this value into the daily intake will give the dose to the GI(LLI) from ^{54}Mn. Each nuclide is treated similarly, and a list of these values has been published [8].

In Table IV are listed the principal radionuclides, the critical organ for each, and the concentration factor which, when multiplied by the concentration in canal water (see Table I) or river water,(3) will give the estimated radionuclide concentration in canal- and river-caught fish, respectively, due to plant discharges. These concentrations are listed in columns 4 and 5 of Table IV. In the last column of Table IV are the dose rates calculated using the above relationship (pCi/d per rem/yr) and assuming a person will eat 50 g of fish per day [10], which are caught 6 months per year from the canal and 6 months per year from the river. A recent creel survey of the canal and river reasonably supports this daily intake [11].

Phosphorus-32 and the radioisotopes of iodine, principally ^{131}I, are the critical radionuclides discharged by the station into the aquatic pathway and contribute about 98% of the total dose. Although the estimated ^{32}P concentration of 0.043 pCi/liter in the water is low, the large concentration factor results in a high estimated concentration in the fish muscle,

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(3) The dilution factor for river water is 25 times that for canal water, based on an average fresh water flow past the station of 1.57×10^{13} liters/yr and a total average discharge in the canal of 6.3×10^{11} liters/yr [9].

TABLE IV. ESTIMATED DOSE TO CRITICAL ORGANS FROM EATING FISH(a)

Isotope	Critical Organ	CF[5](b)	Est. Conc. in Canal Fish (pCi/kg)	Est. Conc. in River Fish (pCi/kg)	Dose Rate(c) (mrem/yr)
^{3}H	Total Body	0.90	9,360	374	0.02
^{32}P	Bone	1×10^{5}	4,300	172	7.5
^{32}P	Total Body	1×10^{5}	4,300	172	0.3
^{32}P	GI(LLI)	1×10^{5}	4,300	172	0.8
^{54}Mn	GI(LLI)	100	48	2	0.01
^{58}Co	GI(LLI)	20	82	3	0.02
^{60}Co	GI(LLI)	20	13	0.5	0.005
^{95}Nb	GI(LLI)	3×10^{4}	195	8	0.04
^{131}I	Thyroid	15	34	1.4	5.6
^{133}I	Thyroid	15	25	1.0	1.2
^{135}I	Thyroid	15	60	2.4	1.0
^{134}Cs	Total Body	400	160	6	0.11
^{137}Cs	Total Body	400	230	9	0.07

(a) The sum of the doses from all other nuclides considered (^{14}C, ^{24}Na, ^{51}Cr, ^{55}Fe, ^{59}Fe, ^{57}Co, ^{90}Sr, ^{95}Zr, ^{99}Mo, ^{110m}Ag, ^{136}Cs) was 0.010 mrem/yr.

(b) CF used with radionuclide concentrations in water (see Table I) to estimate concentration in fish muscle due to plant discharges.

(c) Annual dose rate based on fishing the canal for 6 months and the river for 6 months.

and hence, a high dose rate. Regrettably, a delay in the analysis of the fish muscle precluded the possibility of measuring the ^{32}P present in the muscle. Additional fish samples will be collected to evaluate the calculated ^{32}P value.

The total dose rates to each critical organ, estimated by summing the dose from each radionuclide, are listed in Table V. The highest dose is to the bone from ^{32}P and to the thyroid from the radioisotopes of iodine. These estimated dose rates probably represent maximum exposures that will be received. Few people will continuously consume 50 g of canal fish per day, and consume them so promptly that little radioactive decay has occurred. Nearly 96% of these calculated dose rates are due to eating fish caught from the canal, which is located on station property.

In the last column of Table V is listed for each critical organ the fraction of the Radiation Protection Guide (RPG) for a "suitable sample" of the exposed population as recommended by the FRC [6]. For no organ is the estimated dose near the RPG. The most critical organ appears to be the bone, which is about 5% of the RPG.

TABLE V. THE ESTIMATED TOTAL DOSE RATE TO THE CRITICAL ORGANS

Critical Organ	Dose Rate (mrem/yr)	Fraction of RPG(a)
Bone	8	0.05
Thyroid	8	0.02
GI(LLI)	1	0.006
Internal Whole Body	0.5	0.003

(a) Radiation Protection Guides recommended by the FRC [6]; the RPG are 500 mrem/yr for thyroid and 170 mrem/yr for all other critical organs.

Conclusions

This environmental surveillance study at a large commercial PWR nuclear power station demonstrated the feasibility of tracing effluent radionuclides through the nearby aquatic environment to sediment and biota, including food fish. Sampling and analysis of fish were relatively convenient because liquid effluents are discharged into a coolant-water canal that attracts and retains fish during the winter because of its higher water temperature. For the four radionuclides that were detected in fish muscle, concentration factors from water to fish computed from annual radionuclide discharges and average dilution volumes were in reasonable agreement with recently tabulated values. The radionuclides that, according to discharge data and tabulated concentration factors, cause the highest radiation exposures to fish eaters, could not, however, be measured in the fish muscle.

The computed radiation dose rates to man were 8 mrem/yr to the thyroid from ^{131}I, ^{133}I, and ^{135}I, and 8 mrem/yr to the bone from ^{32}P if 9 kg of the consumed fish had lived in the coolant canal during winter and the other 9 kg in the river during summer. To obtain a more precise dose rate value, the concentrations of radioiodine and ^{32}P in fish muscle, the actual fish catch in the canal and river, and the maximum consumption of these fish should be measured.

It is recommended that the demonstrated calculations of concentrations in fish muscle based on effluent data be undertaken at nuclear power stations, followed by direct measurements of the critical radionuclides whenever possible. The latter will reduce uncertainty caused by the effect of variable radionuclide discharges throughout the year, as well as the effects of local aquatic environmental characteristics on uptake by fish.

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DISCUSSION

F. O. HOFFMAN: Did you consider age-dependence in your estimation of thyroid dose? It appears to me that a 'critical group' might be constituted by young fishermen under the age of 16 years, whose thyroids would be significantly smaller than the 20 g thyroid of the adult.

R. L. BLANCHARD: No, we did not consider children in our estimate of thyroid dose — only the adult thyroid as defined by the ICRP. Your point is valid as some children are extremely partial to fish and may consume as much of it as an adult.

R. F. FOSTER: I have a question in connection with Table V of the paper: could you comment on the use of RPG values for populations rather than for individuals?

R. L. BLANCHARD: Because we are speaking of an avid fisherman, the RPG value for an individual may indeed be more appropriate than that for populations. However, because the fisherman population in the area is substantial, we did not think it incorrect to use the lower value, which gives a more conservative estimate of the fraction of the RPG — i.e. a higher fraction.

TRANSFER MECHANISM AND CONCENTRATING PROCESSES OF CERTAIN RADIONUCLIDES IN THE AQUATIC ECOLOGY OF THE ISMAILIA CANAL

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Abstract

TRANSFER MECHANISM AND CONCENTRATING PROCESSES OF CERTAIN RADIONUCLIDES IN THE AQUATIC ECOLOGY OF THE ISMAILIA CANAL.

The behaviour of certain radionuclides in the aquatic ecology of the Ismailia Canal is of great interest for the evaluation of the possible hazards that may occur to man through the movement of such radionuclides via the food chain. Laboratory investigations have been carried out to understand the accumulation and release of ^{134}Cs, ^{60}Co and ^{89}Sr by some of the biota, such as phytoplankton organisms, certain types of aquatic plants, snails and fish, the hydrological characteristics of the Ismailia Canal and the uptake by bottom sediments.

The physico-chemical characteristics of Ismailia Canal water were performed periodically for 18 months to determine the seasonal variations in the conductivity, pH, total hardness, alkalinity, organic matter content, anion and cation content, etc. Laboratory investigations of the hydrological characteristics of Ismailia Canal water with respect to the relative distribution of radionuclides between water and bottom sediments were carried out. The distribution of the radioactivity in the bottom sediments was measured and computed and the results presented in the form of histograms for both flowing and non-flowing ecosystems. Possible applications of the experimental laboratory data to the natural conditions prevailing in the Ismailia Canal are also discussed.

INTRODUCTION

The behaviour of radionuclides in the aquatic ecology of the Ismailia Canal is of great interest for the evaluation of the possible hazards that may occur to man from the movement of such radionuclides via the food chain [1, 2].

The radionuclides selected for the detailed studies presented here were; strontium, cobalt and caesium (e.g. ^{89}Sr, ^{60}Co and ^{134}Cs) because of their relatively long half-lives and their presence in relatively large amounts in the reactor water coolant wastes. Certain investigations that could help an understanding of the transfer mechanism and concentrating processes of these radionuclides in the aquatic ecology of the canal were necessary. These investigations included:

(1) The physico-chemical characteristics of Ismailia Canal water;
(2) The hydrological characteristics of Ismailia Canal water;

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(3) The uptake of these radionuclides by bottom sediments;
(4) The distribution of the radioactivity in the bottom sediments; and
(5) The accumulation and release of these radionuclides by some of the biota.

PHYSICO-CHEMICAL CHARACTERISTICS OF ISMAILIA CANAL WATER

The physico-chemical characteristics of Ismailia Canal water were tested periodically for 18 months to determine the seasonal variation in the specific conductivity, pH, total hardness, alkalinity, organic matter content, Na^+, K^+, Ca^{++}, Mg^{++}, Fe^{3+}, Zn^{++}, Pb^{++}, Cl^-, SO_4^{--}, PO_4^{--} and SiO_2 content [3]. These investigations indicated that Ismailia Canal water is slightly basic with a limited variation in pH (8-8.6), moderately hard and with an average total solid content of about 260 mg/litre. In general, the Ismailia Canal could be considered as a bicarbonate stream of limited mineral content with slight seasonal variation.

HYDROLOGICAL CHARACTERISTICS OF ISMAILIA CANAL WATER

Laboratory investigations of the hydrological characteristics of Ismailia Canal water with regard to the relative distribution of radionuclides between water and bottom sediments were carried out by using aquaria containing samples of the bottom sediments and canal water. For the non-flowing ecosystem the canal water was left with the bottom sediments inside the aquaria for two weeks before the start of the experiment. For the flowing ecosystem the canal water was changed continuously at predetermined rates of displacement totalling 4 litre/d. Radionuclides were added to both ecosystems in order to maintain concentration levels of about 2 to 4 μCi/litre [4].

The results indicated the existence of various distinct phases in the aqueous environment, which were found to follow the generalized equations [5]:

For the non-flowing ecosystem

$$Y = a\,T^b$$

For the flowing ecosystem

$$Y = a\,X^b$$

i.e.

$$\text{Log } Y = \text{Log } a + b \ \log T$$

and

$$\text{Log } Y = \log a + b \ \log X$$

where $Y = C/C_0 = \dfrac{\text{Activity of water in the aquarium}}{\text{Activity of water in the influent tank}}$

T = Contact time

X = Number of displacements of aquarium water

$$= \frac{\text{Effluent volume of water}}{\text{Initial volume of aquarium water}}$$

and a and b are constants.

The relationships between fractions of ^{89}Sr, ^{60}Co and ^{134}Cs activity in the canal water (C/C_0) with respect to contact time and aquarium volumes in non-flowing and flowing ecosystems respectively are given in Tables I and II.

TABLE I. LINEAR RELATIONSHIP BETWEEN PERCENTAGE ACTIVITY RETAINED IN AQUEOUS SOLUTION AND CONTACT TIME IN A NON-FLOWING ECOSYSTEM

Phase	Radioisotope	Linear relationship log Y = log a + b log T	Slope	Correlation coefficient
I	^{89}Sr	1.82 - 0.065 log T	-0.065	0.94
	^{60}Co	1.86 - 0.063 log T	-0.063	0.83
	^{134}Cs	1.79 - 0.12 log T	-0.12	0.81
II	^{89}Sr	2.00 - 0.27 log T	-0.27	0.94
	^{60}Co	2.87 - 1.19 log T	-1.19	0.98
	^{134}Cs	2.65 - 1.13 log T	-1.13	0.98

TABLE II. LINEAR RELATIONSHIP BETWEEN FRACTION OF ACTIVITY IN THE CANAL WATER (C/C_0) WITH RESPECT TO AQUARIUM VOLUMES IN A FLOWING ECOSYSTEM

Phase	Radioisotope	Linear relationship log Y = log a + b log X	Slope	Correlation coefficient
I	^{89}Sr	-0.23 + 0.59 log X	0.59	0.98
	^{60}Co	-0.13 + 1.24 log X	1.24	0.81
	^{134}Cs	-0.17 + 1.26 log X	1.26	0.98
II	^{89}Sr	+0.58 + 0.05 log X	0.05	0.94
	^{60}Co	-0.47 + 0.160 log X	0.160	0.72
	^{134}Cs	-0.64 - 0.016 log X	-0.016	0.64
III	^{89}Sr	-1.11 + 1.13 log X	1.23	0.64
	^{60}Co	-2.44 + 4.80 log X	4.80	0.75
	^{134}Cs	-1.18 + 1.66 log X	1.66	0.71

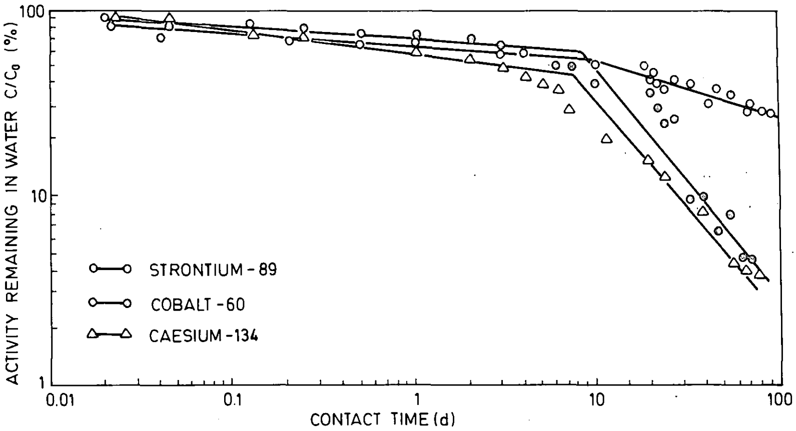


FIG. 1. Linear relationship of decrease in activity with respect to time in a non-flowing ecosystem.

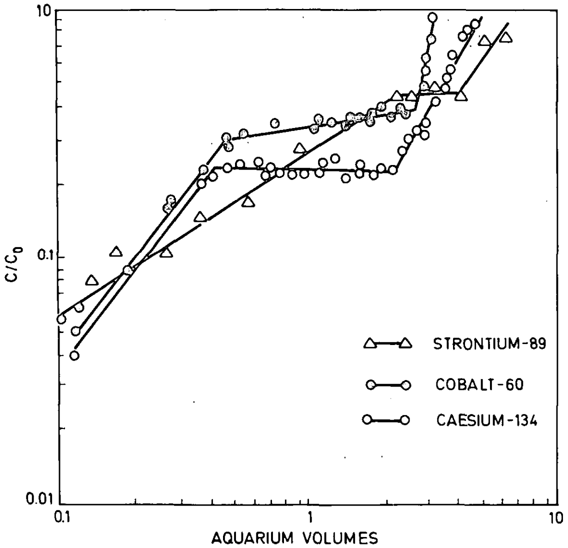


FIG. 2. Increase of activity in the aqueous phase with respect to continuous flowing radioactive influent in a flowing ecosystem.

There were two phases in an impounded ecosystem, while in a flowing ecosystem three phases occurred, as shown in Figs 1 and 2. These phases were found to be independent of the contact time, the rate of displacement of Ismailia Canal water, the size of the aquaria and the nature of the bottom sediments. Moreover, for the flowing ecosystem it was noticed that the clay content of the bottom sediments mainly affected the number of displacements necessary to produce a phase transition. This means that the increase in the clay content was always followed by an increase in the number of displacements necessary for phase transition. This phenomenon was found to exist regardless of the rate of displacement, size of aquarium and nature of radionuclide.

UPTAKE OF RADIONUCLIDES BY ISMAILIA CANAL BOTTOM SEDIMENTS

The sorption capacity of the bottom sediments of Ismailia Canal towards ^{89}Sr, ^{60}Co and ^{134}Cs was measured by batch and column techniques [6, 7]. The results showed that this sorption capacity was largely influenced by grain size fraction, carrier concentration and contact time. In the presence of Ismailia Canal water the apparent capacity for caesium varied from 5.4×10^{-3} to 4.3×10^{-3} meq./100 g, for strontium from 6.5×10^{-3} to 6.7×10^{-3} meq./100 g and for cobalt from 3.2×10^{-3} to 7.3×10^{-3} meq./100 g.

DISTRIBUTION OF THE RADIOACTIVITY IN ISMAILIA CANAL BOTTOM SEDIMENTS

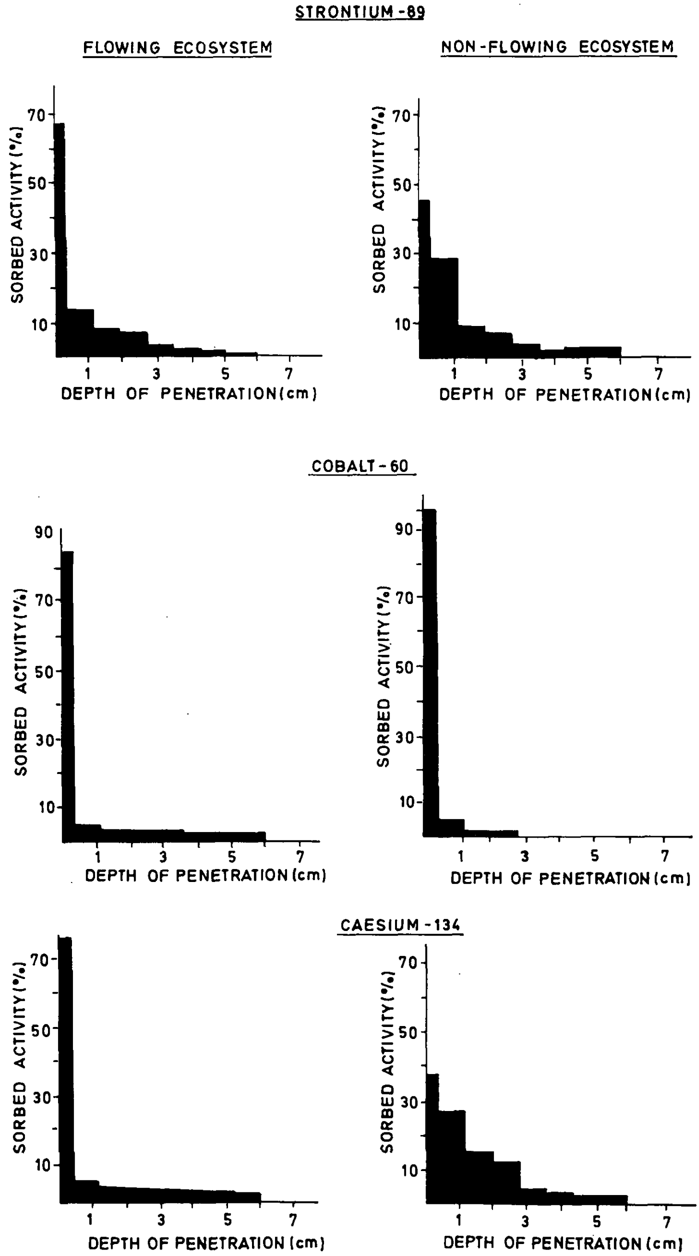
The distribution of the radioactivity in the bottom sediments was measured and computed and the results were presented in the form of histograms for both flowing and non-flowing ecosystems [8], as shown in Fig. 3. These radioactivity measurements were carried out either by an end-window GM counter set up for β-emitters or by a NaI(Tl) gamma-ray spectrometer for γ-emitters. Corrections during radioactivity measurements were always taken into consideration whenever necessary.

In both ecosystems the cobalt incorporated in the bottom sediments was detected mainly in the upper layer, while caesium and strontium migrated to deeper layers, regardless of the size of aquarium. It was also noticed that the cobalt was acid soluble and only 14% was removed by exposure to canal water. On the other hand, the caesium was tightly fixed, while 60% of strontium was easily removed by canal water. These results are in agreement with previous experimental findings [9].

ACCUMULATION AND RELEASE OF RADIONUCLIDES BY SOME OF THE BIOTA

Various laboratory investigations were carried out to study the accumulation and release of the selected radionuclides by some of the biota, such as phytoplankton, aquatic plants, snails and fish. The experimental procedures necessary for investigating the accumulation and release of these biota can be summarized as follows.

The survey of the phytoplankton organisms present in the Ismailia Canal was carried out with the Sedgwick-Rafter method [3]. The aquatic plants under investigation were Elodea densa, Ceratophyllum demersum, Potamogeton pictenatus and Chara sp. For uptake studies they were immersed in aquaria containing solutions of various concentrations (from 0.5 to 10 μCi/litre) of ^{60}Co, ^{134}Cs and ^{89}Sr at pH 8.2. At predetermined intervals portions of these plants were removed and treated for radioassay. The correlation between the concentration of radionuclides in the canal water and its uptake by these plants after various contamination periods


STRONTIUM-89

FLOWING ECOSYSTEM

NON-FLOWING ECOSYSTEM

COBALT-60

CAESIUM-134

FIG. 3. Comparison of distribution of different radionuclides through bottom sediments for flowing and non-flowing ecosystems.

was tested for significance using the 'T test at the 95% confidence level'. For release studies of the plants the previously immersed plants were transferred to aquaria containing fresh water that was changed frequently. At predetermined intervals plant samples were removed and treated for radioassay. The data were presented in terms of the percentage release of the initial uptake.

The snail Biomphalaria alexandrina and the fish Tilapia zillii were studied. Specimens were placed in an aerated aquarium containing an aqueous solution of these radionuclides at concentrations ranging from 9 to 10 μCi/litre. For uptake studies samples were removed at various intervals and measured for radioactivity. For release studies these samples were transferred to aquaria containing running fresh tap water, then removed and treated for radioassay. The uptake studies were used to calculate the concentration factor of the snails and fish.

The phytoplankton present in Ismailia Canal water showed a variety of microorganisms, representing the diatoms and green algae at a concentration of 150-300 organisms/ml of canal water.

The uptake of radionuclides of strontium, cobalt and caesium by certain types of aquatic plants present in the Ismailia Canal was found to increase on increasing the initial concentration of these radionuclides in the canal water. The maximum uptake was reached by these plants after various periods of contamination ranging from 1 to 4 days. A major portion, ranging from 45 to 85% of the sorbed radionuclides, was released after 2 to 4 days. Examples of uptake and release data by these aquatic plants are shown in Figs 4 and 5. Statistical evaluations of the significance of the correlation coefficient between the uptake of radionuclides by these plants and their concentration in canal water made it possible to select the following aquatic plants which could serve as reliable indicators.

(1) Ceratophyllum was found to be the most favourable biological indicator for the concentration of strontium radioisotopes in the Ismailia Canal, which ranged from 0.5 to 10 μCi/litre, and for contamination periods up to 15 days.

(2) Elodea was found to serve as a biological indicator for the strontium radioisotopes for contamination periods of 2 to 16 days.

(3) Potamogeton was also found to serve as a biological indicator for short periods of contamination by strontium and caesium radioisotopes (0 to 2 days).

Moreover, the snail B. alexandrina concentrated these radionuclides from the aqueous environment with a concentration factor of 1600 for cobalt and 15 for caesium or strontium after 3 weeks contact time. The percentage release was 35% of the sorbed caesium and cobalt and 70% of the sorbed strontium after one day. This percentage release reached 95% for caesium, 70% for cobalt and 80% for strontium after two weeks.

Tilapia zillii fingerling fish concentrated these radionuclides with a concentration factor, after 3 weeks contact time, of 50 for cobalt, 29 for strontium and 11 for caesium. After one day the percentage release was 50% of the sorbed caesium, 60% of the sorbed cobalt and none of the sorbed strontium. This percentage release reached 96% for caesium, 80% for cobalt and only 10% for strontium after 3 weeks. This means that the strontium sorbed by Tilapia fish was firmly fixed. The variation of percentage release from this T. zillii fish with contact time is illustrated in Fig.6.

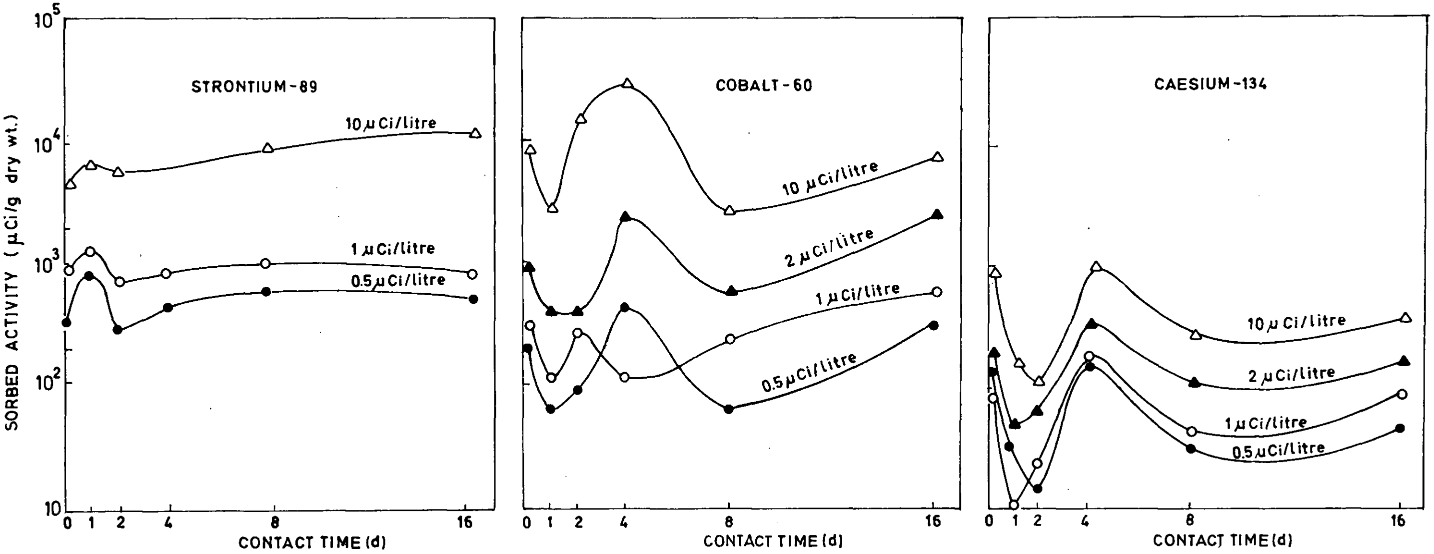


FIG.4. Uptake of radionuclides by Elodea densa plant.

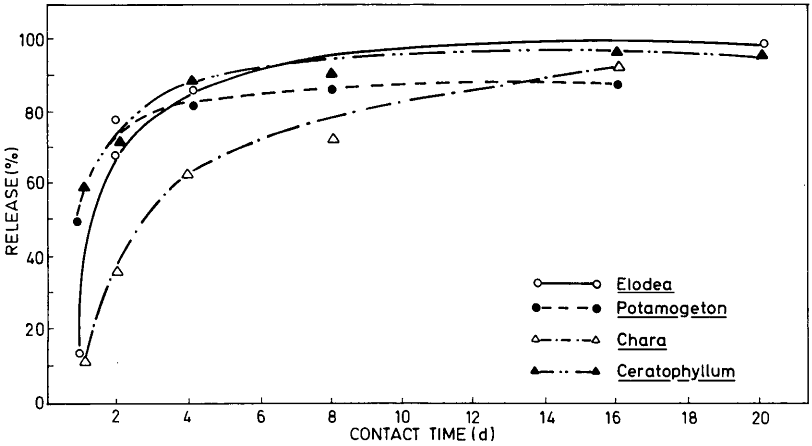


FIG.5. Release of strontium-89 from different aquatic plants present in Ismailia Canal.

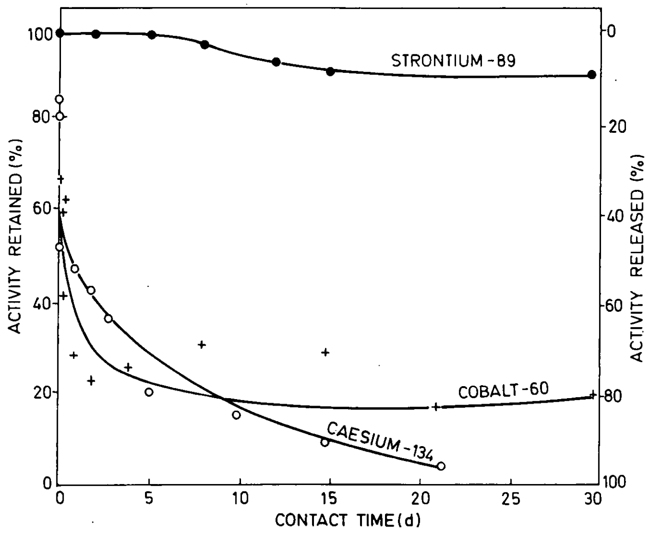


FIG.6. Activity released from contaminated *Tilapia zillii* fish.

CONCLUSIONS

Laboratory experiments carried out on aquatic plants indicated their possible application as biological indicators for certain radionuclides and for certain contamination periods (i.e. *Elodea densa* and *Ceratophyllum demersum* for strontium radioisotopes and *Potamogeton pictenatus* for strontium and caesium radioisotopes).

The initial uptake of ^{89}Sr by snails was higher than that by the fish studied. However, the strontium sorbed by the fish was firmly fixed, while that by the snails was easily released after immersion in clean tap water. The ^{60}Co uptake of the snails was significantly higher than that of the fish, though the cobalt retained was in the same range. The behaviour of ^{134}Cs was the same for both snails and fish.

From the laboratory hydrological studies carried out on Ismailia Canal water and bottom sediments and considering that the average speed of Ismailia Canal water is about 2. 2 km/h, sampling at short intervals will definitely give an indication of the magnitude of contamination of the Ismailia Canal water under conditions of continuous disposal.

In general, it could be stated that the magnitude of contamination of the aquatic environment of the Ismailia Canal by either strontium, caesium or cobalt is difficult to assess unless radioactive equilibrium is achieved between the level of radioactive contamination of the canal water and the corresponding concentration levels in the existing biota, suspended matter and bottom sediments. Because of the high rate of flow of the Ismailia Canal, it is presumed that such equilibrium can be only achieved (a) for bottom sediments, surface and bottom biota under conditions of continuous disposal, and (b) for surface biota under conditions of accidental release.

However, this is only a predictive picture of the fate of these radionuclides when disposed into the Ismailia Canal. Typical field experiments remain a problem because of the difficulty in convincing the public of the absence of possible radiation hazards from such experiments. In other words, it is probably feasible to apply the detailed laboratory experiments presented in this study to the natural conditions prevailing in the Ismailia Canal water without exaggeration.

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ENVIRONMENTAL BEHAVIOUR OF RADIOCOBALT AND RADIOSILVER RELEASED FROM NUCLEAR POWER STATIONS INTO AQUATIC SYSTEMS

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Abstract

ENVIRONMENTAL BEHAVIOUR OF RADIOCOBALT AND RADIOSILVER RELEASED FROM NUCLEAR POWER STATIONS INTO AQUATIC SYSTEMS.

To deduce the general characteristics of the behaviour of radiocobalt and radiosilver released from nuclear power stations into the aquatic environment, the release situations of several existing power stations together with field data obtained to date on these and other releases are systematically assembled and reviewed. These data indicate that, although the major sink for radiocobalt is bottom sediments, some fractions tend to recycle into a soluble phase and go through complicated processes such as hydrolysis, inorganic and organic complex formation, adsorption-desorption with suspended particles and bottom sediments, etc. In the case of radiosilver the complex formation with chloride ions in the seawater medium seems to be important in maintaining this element in solution, while there are practically no data available for freshwater systems.

Since these reactions have pronounced effects on the availability of the radionuclides to aquatic biota, possible speciation equilibria and related geochemical behaviour of cobalt and silver radionuclides in natural waters are discussed in detail on the basis of the data obtained by various workers as well as by the present authors from laboratory experiments and field measurements. An attempt is made to evaluate and compare the importance of various processes under natural conditions.

Finally, the accumulation of radiocobalt by the aquatic biota is reexamined from the view point of concentration factor. Revised concentration factors are presented for some groups of marine organisms.

INTRODUCTION

It is known that cobalt-58 and cobalt-60 are among the major radionuclides in waste effluents released from most boiling-water and pressurized-water nuclear power stations. Silver-110m is also released from some types of reactors. Due to the cooling-water requirements, the siting of nuclear power stations has been and will be closely connected with the availability of water resources, freshwater and seawater, which in turn bring radioactive substances back to the aquatic environment. Cobalt-60 and silver-110m have relatively long half-lives of 5.3 years and 253 days respectively and, through the accumulation by aquatic biota, these radionuclides might possibly become critical in radioactive waste disposal operations. Although the impact of radioactive waste effluents on aquatic ecosystems and human health has been carefully assessed for each nuclear reactor site and appropriate control practices are applied to various in situ conditions, the detailed behaviour of the above-mentioned radionuclides in the aquatic environment is still not fully understood.

The present paper attempts to systematize the information obtained so far on the environmental behaviour of radiocobalt and radiosilver in order to aid a better understanding of the geochemistry of these elements in the

TABLE I. EXAMPLES OF ESTIMATED RELEASES OF RADIOCOBALT AND RADIOSILVER INTO THE AQUATIC ENVIRONMENT FROM NUCLEAR POWER STATIONS

Power station	Tarapur (India)	Garigliano (Italy)	Dresden Unit I (USA)	Humboldt Bay (USA)	Trino Vercellese (Italy)	Indian Point Unit I (USA)	Latina (Italy)	Bradwell (UK)
Type of reactor	Boiling water				Pressurized water		Graphite-moderated gas-cooled	
Capacity (MW(e))	400	150	210	65	252	265	200	300
Recipient area for liquid waste release	Arabian Sea Coast	Garigliano River	Illinois River	Humboldt Bay	Po River	Hudson River	Mediterranean coast	Blackwater Estuary
Period considered	1969[a]	1969	1968 - 1969	1971 - 1972	1967	1969	1969	1969 - 1970
Estimated release rate (Ci/yr)								
^{3}H	5	~ 10	5	12	600	1100	40	140
^{58}Co	0.2	< 5	0.8	0.001	2.2	6	-	-
^{60}Co	0.1	< 5	1.1	0.1	1.7	5	0.05[b]	0.5
^{110m}Ag	-	-	-	0.001	-	-	-	0.02
^{137}Cs	0.3	~ 2	0.2	0.1	1.1	3	8	30
Source of data	[1]	[2]	[3,4]	[5]	[2]	[6]	[2]	[7 - 10]

[a] The period for pre-commercial operation.

[b] Including iron-59.

hydrosphere and to indicate problem areas that should attract attention in future studies. The results of laboratory experiments on the chemical speciation and sorption characteristics of cobalt and silver, together with the data obtained on the stable isotope concentrations of these elements in natural waters around Monaco are presented and used in discussing the behaviour of these elements in the aquatic environment.

The concentration factors of cobalt for marine organisms, which have been computed on the basis of stable element analysis of biota and ambient waters, can be in error due to analytical uncertainty in the determination of cobalt. Therefore, revised concentration factors are presented in this paper for some marine organisms.

RELEASE SITUATIONS

To obtain a general idea of release situations of radiocobalt and radiosilver in actual operations of nuclear power stations, examples of release of these radionuclides from existing power stations in several countries are summarized in Table I. The information was taken from the open literature and in some cases release figures were estimated on the basis of related data given. The sources of data are indicated in the table [1-10]. In addition to release figures of radiocobalt and radiosilver, those for tritium and caesium-137 are also given in Table I to show relative quantities of radiocobalt and radiosilver to other components of the waste effluents. As shown in Table I, the annual release ranges from 1 mCi to 6 Ci for cobalt-58, 0.05 - 5 Ci for cobalt-60 and 1 - 20 mCi for silver-110m. Although the capacities of those power stations listed that had been operational during the 1960s are relatively small compared to those of larger reactors of recent and future construction, it is not expected that the release of radiocobalt and radiosilver from a specific type of power station will differ dramatically from the figures cited in Table I. The increase of release by a factor of up to 2 normally, 3 at maximum in very unfavourable conditions, may be expected for large capacity power reactors. Since the release rates at present from all of the stations listed in Table I have been proved to be well below the authorized limits, it is unlikely that even large capacity power reactors will cause any significant radiological impact on the aquatic environment, except for cases of multiple siting of nuclear reactors along a river system or a limited stretch of sea coast. Generally speaking, among several radionuclides that will be released, the only radiologically significant radionuclide would be tritium, which is due to the relatively large release rates, even though the radiotoxicity of tritium is low. Nevertheless, a relatively small introduction of radiocobalt and radiosilver into the aquatic environment should not be completely neglected since the recent example of silver-110m in the Blackwater Estuary [9] shows that this radionuclide has now become that with the highest radiological significance because of its accumulation by oysters. This is due to operational changes of the power station during 1970. As shown by this example, environmental monitoring is always desirable to ensure safe operation of nuclear installations as well as to investigate the fate of radionuclides in the environment.

FIELD DATA AVAILABLE

To systematize the information obtained so far on the environmental behaviour of radiocobalt and radiosilver, field data obtained in release areas of these radionuclides from various nuclear installations were assembled. Not only the data obtained for releases from nuclear power stations, but also those for releases from other types of nuclear facilities are considered here since the chemical forms of these radionuclides in waste effluents of these latter facilities do not differ substantially from those in effluents of power reactors. In addition to these data, measurements made so far on radiocobalt and radiosilver of fall-out origin in environmental samples are also reviewed briefly in order to indicate available sources of data for these radionuclides. In cases of fall-out, however, the chemical forms of radiocobalt and radiosilver introduced into aquatic systems may be quite different from those in waste effluents and, consequently, the availability of these radionuclides to aquatic organisms may not be similar.

1. Fall-out origin

In the early period of nuclear explosion tests in the Pacific during the 1950s Lowman [11,12] reported the occurrence of cobalt-57, -58 and -60 in water, plankton and fish and Seymour [13] that of cobalt-57 and -60 in marine invertebrates (clam, snail and lobster) and fish[1]. The latter author also measured silver-110m in a lobster. These samples were collected from close-in fall-out areas from the explosion tests. In this connection Lowman [11] suggested that practically all radionuclides, except for the caesium, strontium and iodine isotopes, would be present in the particulate form when they are introduced into the sea as fall-out.

Folsom and Young [14] measured low-level cobalt-60 and silver-110m in livers of squids, mussels, barnacles and fishes collected in 1964 from various localities of the Pacific Ocean, proving wide-spread distribution of these radionuclides through radioactive fall-out and advection of surface currents.

In 1967 Robertson et al. [15] collected seawater and marine organisms from various depths at a station 350 miles off Newport, Oregon, and determined both cobalt-60 and stable cobalt in these samples. The concentrations of cobalt-60 at this station were estimated to be $0.2 - 3\times10^{-3}$ pCi/litre at the surface, 0.1×10^{-3} pCi/litre at about 700 m depth, and less than 0.1×10^{-3} pCi/litre below 700 m. The percentage of particulate fraction (retained by 0.3 μm pore-size membrane filter) was 20 - 60% at the surface and slightly lower at depths, being 10 - 30%. At a station 10 miles offshore from Coos Bay, Oregon, where the effect of the Columbia River discharge was recognizable, cobalt-60 was present at a concentration of 4×10^{-3} pCi/litre (35% particulate) in the surface water, while at 30 m depth 6×10^{-3} pCi/litre (5% particulate) were found. At this station 0.3×10^{-3} pCi/litre of silver-110m was also found at the surface in the particulate fraction. One important finding made at the offshore Newport station was that the specific activity of cobalt-60 in fish species and a shrimp was much higher than that of seawater, the ratios of which ranged from 40 - 100, while the specific activity ratio for caesium-137 was equal to unity. This indicated that either cobalt-60 was in a form more readily available for biological assimilation than its

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[1] More recent data on ^{60}Co and other radionuclides in various marine organisms in the Bikini area have been published in Ref. [78].

stable counterpart in seawater, or that these organisms had resided for some time in waters influenced by the Columbia River discharge, where much higher cobalt-60 concentrations were present.

Jenkins [16] also reported low-level occurrence of cobalt-60 and silver-110m in Pacific salmon collected mainly from the Alaskan coast, finding a systematic decrease of cobalt-60 in the muscle tissues of salmon from north to south along the Alaska-Washington-Oregon coast.

In 1970 Folsom et al. [17] detected and measured another silver radionuclide, silver-108m, in the livers of squids, tunas, albacores, lobsters and mussels, and suggested using the pair of silver isotopes, silver-108m and -110m to date the time of the original activation as having considerably different half-lives, 127 years and 253 days. Independently, silver-108m was also measured by Cooper et al. [18] in Pacific salmon.

On the basis of measurements made in tunas for cobalt-60, silver-108m and -110m among other radionuclides, Folsom et al. [19] tried to understand the variation in these radionuclides in connection with the transport by the Californian current along the Oregon-California coast. Based on cobalt-60 and silver-108m data for albacores during the period of 1964-1971 Hodge et al. [20] demonstrated that these fall-out radionuclides tended to be retained in the upper layers of the Pacific Ocean, having half-times of decrease of 2.6 and 7.1 years, respectively, which are considerably longer than the half-time of decrease of fall-out itself. Young and Folsom [21] suggested the use of mussels and barnacles as indicators to follow variation in manganese-54, cobalt-60 and zinc-65 in the marine environment.

The above-mentioned information obtained to date for radiocobalt and radiosilver of fall-out origin demonstrates the usefulness of these data as tags for following processes taking place in water masses in connection with the variations of these radionuclides. As has been pointed out, situations in waste disposal operations may be quite different from those in fall-out.

2. Columbia River

The discharge of reactor effluents from the Hanford reactors to the Columbia River began with the first reactor operation in 1944 and ended with the closure of the KE reactor in 1971, although a dual-purpose reactor for power and plutonium production still operates at present and discharges small amounts of its primary and secondary coolant water through a seepage trench. During this period an environmental co-ordinated study program was established and in the framework of this program comprehensive studies in various fields, biology, radioecology, radiogeochemistry, sedimentology, hydrology, oceanography, etc., of the Columbia River, its estuary and adjacent ocean areas were completed, the major findings of which were summarized in a book edited by Pruter and Alverson [22]. Since the amount of information obtained in this study program that is relevant to the behaviour of radionuclides in water, sediments and biota of this complex water system, is tremendous, only direct information concerning the behaviour of radiocobalt in Columbia River water and its interaction and distribution in suspended particles and sediments will be considered here.

Cobalt-58 and -60 were measurable by multidimensional gamma-spectrometry in Columbia River water throughout the operational period of the Hanford reactors, even though their levels were relatively low compared to those of 'Columbia River radionuclides', such as phosphorus-32, scandium-46, chromium-51, zinc-65, etc. [23]. The behaviour of several radionuclides in the waters of the Columbia River, its estuary and adjacent sea areas was reviewed by Forster [24] and distributions of radionuclides in river and shelf sediments were summarized by Forster [25] and Gross [26]. During the period of 1964-65 Perkins et al. [27] completed weekly measurements of 11 radionuclides including cobalt-58 and -60 in Columbia River water at Pasco, Washington (50 km downstream from the Hanford reactor area) and at Vancouver, Washington (350 km downstream from Pasco). The levels of cobalt-58 measured were approximately 1 - 50 pCi/litre (10-80% particulate) and 0.1 - 5 pCi/litre (80-95% particulate) at Pasco and Vancouver, respectively, and those for cobalt-60 were 0.5 - 5 pCi/litre (20-80% particulate) and 0.1 - 3 pCi/litre (80-95% particulate) at the respective stations. Since the effluent water contained only 4.2% and 1.8% particulate fractions for cobalt-58 and -60, respectively, the above data indicate that cobalt-58 and -60 were readily sorbed by particulate materials in the river. Nevertheless, the large increase in the total content of these radionuclides in the river, observed after the spring freshet, was considered to have resulted not only from the resuspension of radioactive sediments, but also from redissolution of resuspended radionuclides. Based on transport data of radionuclides by the river at Pasco and Vancouver, the percentage depletion of the radionuclides between these stations was computed. The values computed for cobalt-58 and -60 were normally in the range of 50-80%, except for the period of spring freshet, during which negative values were obtained. These negative values indicate that the scouring action of the river exceeded the deposition rate during this period. Assuming that the depletion and input rates of the radionuclides into the river were reasonably constant for several years, the total inventory of each radionuclide considered was estimated. These estimates for cobalt-58 and -60 showed that 140 Ci of cobalt-58 and 450 Ci of cobalt-60 were found in the bottom of the Columbia River between Pasco and Vancouver at the beginning of 1965. The major deposition area was indicated to be McNary Dam.

By analysing sediment cores taken from McNary Dam for cobalt-60 and zinc-65 and assuming the difference in radioactive decay to be the principal cause for changes of the ratio of these two isotopes in the cores, Nelson et al. [28] estimated an average sedimentation rate at a specific station in McNary Dam to be 12.5 cm/yr. They also estimated, using the ratio of zinc-65 to chromium-51 in core samples, that about 30% of the annual deposit was scoured downstream during the spring freshet. They treated streambed sediments with various chemical reactants, finding that while cobalt-60 was not released from the sediments by treating with 1N ammonium acetate solution, acetic acid (pH 3) and bromoethanol treatments removed from the sediments approximately 30 and 40% of cobalt-60, respectively. Sodium hyposulphite treatments also removed up to 70% of cobalt-60, although the mechanism of the removal was not definitely established. The authors concluded that sizable fractions of the radionuclides sorbed on the Columbia River sediments were removed by the treatments with oxidizing agents, reducing agents and acidic solutions, although the radionuclides were tightly bound to the sediments in conditions in situ and were not readily replaced by contact with salt solutions.

Based on the data obtained by Johnson [29] on the replacement of zinc-65 sorbed on Columbia River sediments by other metal ions, Osterberg et al.

[30] pointed out that only a minor fraction of zinc-65 in Columbia River sediments could be displaced by ion-exchange, while a much larger portion appeared to be bound in complexes, perhaps with organic matter. Since cobalt-60 in the sediments was eluted to a similar extent as zinc-65 with EDTA solutions [29], it is reasonable to assume that a situation similar to that of zinc-65 applies also to cobalt-60 in the sediments.

Elution studies on Columbia River sediments with seawater made by Robertson et al. [31] revealed that a considerable portion of manganese-54 (40%), cobalt-60 (5%), europium-152 (9%) and caesium-137 (15%) was eluted when 30 g of the McNary Dam sediment was treated with 8 litres of seawater. On the basis of these data, the authors concluded that a substantial fraction of the stated radionuclides will be leached from the sediments in a relatively short period of time when Columbia River sediments are introduced into the Pacific Ocean, even though the remaining radionuclides in the Columbia River during the post-operational period of the Hanford reactors (July 1971) are tightly bound to the sediments and enter the river mainly by resuspension, especially during the high river flow in spring and early summer. The partitioning of cobalt-60 in Columbia River water in July 1971 showed that 98% was in particulate form, while some 1% was cationic, less than 0.3% was anionic, and 0.6 - 0.7% in a non-ionic form. An increase in the particulate fraction of cobalt-60 is recognizable, compared with the values of 70 - 90% of cobalt-60 in the particulate form when the Hanford reactors had been operating [27].

The fate of cobalt-60 in Columbia River water after entering the estuary and the Pacific Ocean has not been clearly demonstrated by the field data, even though the distribution of this nuclide in estuarine and continental-shelf sediments has been extensively investigated. Forster [25] found that the cobalt-60 content of bulk sediment samples collected along an east-west transect between 40 km upstream of the Columbia River and 100 km seaward showed a minimum at the mouth of the river, a maximum at 25 km upstream from the mouth and several maxima between 8 and 40 km seawards. The seaward maxima may be explained by flocculation and subsequent deposition of this radionuclide beyond the heavy mixing area. As to the distribution of cobalt-60 in shelf sediments, Gross [26] and Barnes and Gross [32] found that cobalt-60 was distributed in a band nearly parallel to the coast-line at depths between 60 and 90 m throughout the adjacent part of continental shelf to the Columbia River mouth, having the centre for distribution at about 10 km west of the river mouth.

On the basis of the data reviewed above, it can be said that a major portion of the radionuclides of cobalt released from the Hanford reactors to the Columbia River becomes bound to the river sediments rather quickly and then behaves with the sediment movements. The mechanism of the sorption of radiocobalt, however, has not been clearly understood, although several studies on this subject suggested that ion-exchange may be a minor mechanism and perhaps complex formation with organic matter may be involved. When the Columbia River water enters into the estuary and sea some sizable fraction of radiocobalt is likely to be released from the suspended particles and to become involved in various types of processes operational in the marine environment. The identity of these processes and time factor involved are not known. Although radiocobalt is likely to deposit eventually to the bottom due to the downward vector of processes involved, there is a tendency for radiocobalt to be retained in the upper layer of the sea for a certain time.

3. Clinch and Tennessee River

Besides the Columbia River, another well-studied freshwater environment for low-level radioactive waste disposal is the Clinch-Tennessee River system. This system at present still receives the waste effluents of complex facilities of the Oak Ridge National Laboratory. As was done in the case of the Columbia River, a co-ordinated study program was established and operated during the period 1957-1964. Detailed release figures of gross beta activity and 9 individual radionuclides including cobalt-60 during 1944 and 1963 were given by Parker et al. [33]. Based on the weekly monitoring data of water for radionuclides obtained along a 250 km reach of the Clinch-Tennessee River system, the same authors made mass-balance analysis of major radionuclides introduced into the Clinch River. They concluded that the radionuclides including cobalt-60, except for caesium-137, which entered the Clinch River in the water phase, left the Clinch River in the water phase. The levels of cobalt-60 measured at several monitoring stations during 1961-1962 were 20 - 50 pCi/litre in the Clinch River and 3-30 pCi/litre in the Tennessee River. Cobalt-60 in suspended solids represented 2-30% of these concentrations. These percentages are considerably less than those in the Columbia River, in spite of the fact that the monthly average of suspended solids, approximately 200 ppm, in the Clinch-Tennessee rivers is much higher than a suspended load of 8-40 ppm (lithogenous particles: 85-95%) in the Columbia River [34].

The radioactivity in the bottom sediments of the Clinch and Tennessee rivers was studied by Pickering et al. [35]. By analysing the sediment cores taken from 33 km reach of the Clinch River, they found that in July 1962 150 Ci of caesium-137 and 18 Ci of cobalt-60, which respectively represented 21% and 9% of the total amounts released during nearly 20 years of the laboratory operation, were present in the sediment of this part of the river. The vertical distribution of cobalt-60 in the sediment cores was strongly correlated with that of caesium-137, suggesting that these radionuclides had been incorporated into the sediments before entering the river. Leaching studies of the sediments with acid, base and salt solutions revealed that salt solutions in natural pH were not effective in removing cobalt-60 from the sediments, thus indicating that simple ion-exchange was probably not the primary cause of the uptake of cobalt-60 by the sediments. They suggested the association of cobalt-60 with manganese oxide in the deposition process.

Nelson et al. [36] observed that cobalt-60 in White Oak Lake was usually found in algae and organic detritus rather than in sediments or other inorganic components. The specific mechanism involved in the accumulation was not clear. Gloyna et al. [37] also demonstrated by simulated stream experiments that transport of radionuclides including cobalt-58 by a stream could be substantially influenced by plant growth, both rooted and floating. Although the retention of radiocobalt by plant growth may be temporary, association of radiocobalt with organic substances may affect the fate of radiocobalt considerably in the stream environment. Quantitative estimates of this effect are not known.

Based on the data reviewed above, it can be concluded that the major part of the cobalt-60 in the Clinch and Tennessee rivers is in the water phase and is transported rapidly downstream, where dilution by inflowing water and sediments from other rivers obscure the fate of this radionuclide.

4. Blackwater Estuary

The monitoring data of the release of cobalt-60 and silver-110m from the Bradwell nuclear power station to the Blackwater Estuary were reported by Mitchell [8,9,38,39] with those of phosphorus-32, iron-55, zinc-65, caesium-134 and caesium-137. The levels of cobalt-60 and silver-110m measured in oysters collected from the Blackwater Estuary in 1967-68 varied in the ranges of 0.007 to 1.9 pCi/g (wet) and 0.07 to 0.38 pCi/g (wet) respectively, depending on distances from the waste effluent outfall [38,39]. In 1969-1970 the silver-110m content of oysters increased, ranging from 0.06 to 2.3 pCi/g (wet) [8,9]. As already mentioned, silver-110m in the Blackwater Estuary recently became the radionuclide of the highest radiological significance due to operational changes of the power station during 1970 [9]. Preston et al. [10] found that the ratio of cobalt-60 between oysters taken from the immediate vicinity of the effluent outfall and approximately 500 m from this point was 12, while the ratios were 5.4 for silver-110m and 3.6 for caesium-137. This indicated that the gradient of the decrease in silver-110m and caesium-137 with distance from the effluent outfall was much smaller than that of cobalt-60. Mitchell [39], on the basis of similar observations, concluded that silver-110m and caesium-137 were relatively evenly dispersed in the estuary in the aqueous phase and were not to any great extent associated with particulate matter. This may be due to a strong tendency of silver to form complexes with chloride ions in brackish water or normal seawater.

Silver-110m was also found in oysters collected from the vicinity of the Windscale discharge [10], although this radionuclide was only a minor component of radioactive wastes released from the Windscale facilities. Cobalt-60 was also measurable in various monitoring samples taken from effluent discharge areas of other nuclear power stations operating in the United Kingdom, such as in seaweed from Hinkley Point, in freshwater moss from Trawsfynydd, in winkles from Winfrith, etc. [8,9,38,39].

The above observations made in the United Kingdom indicate the sediment-seeking tendency of cobalt-60 and the complex-forming tendency of silver-110m, possibly with chloride ions, in the marine environment.

5. Humboldt Bay

As shown in Table I, among other radionuclides cobalt-60 and silver-110m were released into Humboldt Bay from the nuclear power station. The transport of radionuclides in Humboldt Bay was studied by Heft et al. [5]. Based on measurements of cobalt-60 in water, sediments, algae and eel grass in various parts of the bay, they computed the inventory of cobalt-60 in the bay. Cobalt-60 in the sediments represented normally more than 95% of the inventory, except for August 1971 when it was only 63%. Nevertheless, they found that no correlation existed between the standing concentration of cobalt-60 in the bay and current release amounts, and that the amount to be found in the bay represented only about one-fourth of the annual release amount.

Although the forms of cobalt-60 transported out of Humboldt Bay are not known, the above data show that the turnover rate of cobalt-60 in and out of the bay environment is reasonably rapid.

6. Other power stations

While there are many other nuclear power stations that are supposed to release radiocobalt, sometimes with radiosilver, to the environment, the available field data for these stations are rather scarce in the open literature. Only a few examples are given below in order to obtain some idea on the situations in different geographical areas from those reviewed above.

Ophel and Fraser [40] studied the fate of cobalt-60 in Perch Lake, a small lake, where low-level radioactive liquid wastes from the Chalk River nuclear facilities were released. Based on the measurements of cobalt-60 in water, sediment and food-chain components, they estimated a material balance of cobalt-60 in this lake as of September 1969: 140 mCi in the bottom mud, 9 mCi in the lake water and 0.08 mCi in aquatic plants. These figures indicated that more than 90% of cobalt-60 in Perch Lake was to be found in the bottom sediment and that approximately 30% of cobalt-60 added into the lake prior to September 1969 was retained in the lake at that date. As opposed to the observations of Nelson et al. [36], Ophel and Fraser [40] did not find any significant difference in cobalt-60 concentrations between organically poor sediment (2% organic matter on dried basis) and organically rich sediment (55% organic matter on dried basis).

The marine environment in the vicinity of the Tarapur power station, India, was studied by Kamath et al. [1]. They pointed out the effective scavenging of activation products including cobalt-58 and -60 by suspended silt, which characterizes the local water and amounts to 100 ppm in discharge water. In the same locality Patel et al. [41] found that a sea-hare species (Aplysia benedicti) shows a specificity for the accumulation of cobalt radionuclides and suggested the use of this species as an indicator organism for the environmental monitoring.

Umezu et al. [42,43] measured cobalt-58 and -60 in various marine organisms collected from the vicinity of the Mihama nuclear power station, Japan. Maximum concentrations of cobalt-58 and -60 were observed in mussel tissues, being 4 pCi/g (ash) for cobalt-58 and 34 pCi/g (ash) for cobalt-60.

The accelerating developments in the nuclear industry, especially in electrical power generation, require a broadening of existing monitoring networks for environmental radioactivity, especially at new sites of power stations. This will lead to further accumulation of field data on the behaviour of radionuclides including radiocobalt and radiosilver in the environment. This may be considered to be an advantage in the sense of permitting the study of the behaviour of various elements in radiologically labelled environments, if appropriate action is taken to safeguard the environment and human life from the radiological impacts of such contaminants.

CHEMICAL SPECIATION IN THE AQUATIC ENVIRONMENT

In the foregoing review the following points concerning the behaviour of radiocobalt and radiosilver become apparent:

(1) The major sink of radiocobalt in both freshwater and seawater systems is bottom sediments.

(2) Although a major portion of radiocobalt in a stream environment becomes eventually incorporated into bottom sediments, the rate of incorporation may vary considerably depending on the conditions of the stream, such as pH, sediment load, organic content of water and sediments, discharge volume, stream velocity, degree of plant growth, etc. (cf. the Columbia River versus the Clinch-Tennessee rivers).

(3) The mechanism of the sorption of radiocobalt onto suspended particles and bottom sediments has not been well understood; coprecipitation with or adsorption on manganese dioxide (in the case of the Clinch River) or specific sorption due to complex formation probably with organic matter (in the case of the Columbia River) may be involved.

(4) The retention of radiocobalt in the aquatic environment by biota, especially by plants, may affect, at least temporarily, the transport and fate of the radionuclide. The quantitative importance of this effect, however, has not been demonstrated in field conditions.

(5) When stream water loaded with radiocobalt adsorbed on suspended solids enters into marine conditions, a sizable fraction of radiocobalt is released from the suspended solids and becomes involved in various types of processes operational in the marine environment. The identity of these processes and time factor involved are not known.

(6) Silver-110m released into the marine environment is not to any great extent associated with particulate matter and tends to distribute evenly in the aqueous phase due, perhaps, to a strong tendency of silver to form complexes with chloride ions present in vast amounts in the marine media.

These points indicate that chemical forms of radiocobalt and radiosilver in natural waters and interactions of various forms of these radionuclides with suspended particles of inorganic and organic nature play a decisive role in governing the fate of these radionuclides in the aquatic environment. To supplement the information obtained so far on these subjects, several types of laboratory experiments were carried out by the present authors. The results of these experiments are discussed below in connection with the findings already made by other workers.

1. Precipitation

One of the mechanisms of bringing an element dissolved in the aqueous phase into the solid phase is precipitation of the element itself and/or coprecipitation of the element with others. The precipitate formation of several radionuclides including cobalt-60 in distilled water and seawater media over a wide range of pH values was studied by Fukai and Huynh-Ngoc [44]; Figure 1 shows the results obtained. The experimental curves in Fig. 1 indicate that in the pH range of natural waters (pH 5-9) slight precipitate formation up to 15% was observed in the distilled water medium, which was not exclusively attributed to wall-effect of the glass vessels used in the experiments. On the other hand, the precipitate formation of cobaltous ions as hydroxide seems negligible in the seawater medium with or without the presence of iron hydroxide, in accordance with the theoretical curve for the formation of cobaltous hydroxide, shown in Fig. 1 as a broken line. The dash-dotted curve in Fig. 1 corresponds to the theoretical formation of the solid phase CoOOH at a cobalt concentration of 10 μg Co/litre in the seawater medium, where electron activity change with that of pH was considered. Comparison of this theoretical curve with the experimental data

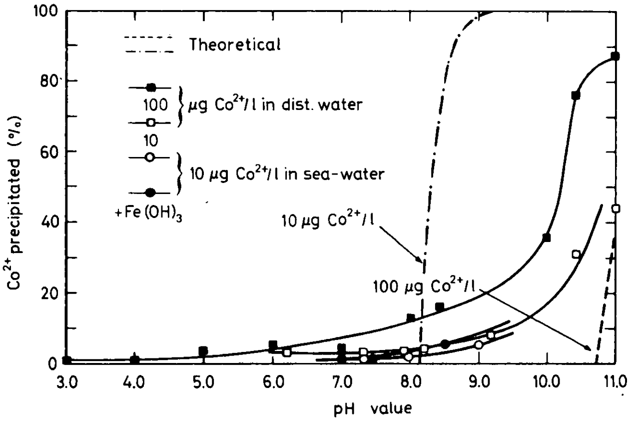

FIG. 1. Precipitate formation of divalent cobalt, Co^{2+}, in distilled-water and seawater media [44].

indicates a considerably retarded formation of the solid phase, if any, under actual conditions. On the basis of these observations, the authors concluded that the precipitation of cobalt under natural conditions is a minor factor in controlling cobalt concentrations in natural waters, unless significant amounts of manganese oxide are present, which is a rare case under normal conditions. Since the solubility product of cobaltous carbonate is at least 2 orders of magnitude higher than cobaltous hydroxide, the possibility of a formation of carbonate precipitate is unlikely, even in waters having a high bicarbonate content.

By mixing equal volumes of filtered seawater with filtered river water in the laboratory, Lowman et al. [45] found that 100% of cobalt was precipitated in 2 hours after the mixing. Since the precipitation of calcium within 3 hours after mixing was also observed in the experiments, either the condition of the experiment represented very special circumstances or, more likely, the 'precipitation' represented the coagulation of fine particles in the freshwater that passed through the filter, under the contact with the salt water.

The particulate formation of silver as well as cobalt in the freshwater and seawater medium was studied by Murray and Murray [46]. They found that in Var River water medium at pH 8 silver formed 50% particulate at a concentration level of 10 μg Ag/litre, while only 5% particulate was formed in the seawater medium. They confirmed the results of Fukai and Huynh-Ngoc [44] for cobalt.

On the basis of these data it is reasonable to conclude that precipitation is a minor mechanism to bring cobalt in natural waters into the solid phase, while the precipitation of silver in slightly basic freshwater may affect considerably the fate of this element in natural waters, though chemical reactions involved are not clearly demonstrated.

2. Complex formation with inorganic ligands

In the seawater medium it is known that trace amounts of metals occur frequently in the form of complex-ions and/or uncharged ion-pairs with inorganic ligands of anions, such as hydroxide, chloride, sulphate, carbonate, etc. [47]. In the freshwater medium, however, the inorganic complex formation is expected to be much less pronounced since the reaction is governed by the law of mass action, where the concentrations of anionic ligands have to be sufficiently high to form any measurable quantities of the complexes. Except for carbonate in some special conditions, the concentrations of anions in freshwaters are normally more than 1000-fold less than those in seawater and not sufficient to form any sizable quantities of inorganic complexes in solution.

Marazović and Pučar [48] studied the complex formation of the mixture of cobalt-56, -57 and -58 in a seawater medium by means of electrodialysis, finding that trace amounts of cobalt ions behaved as typical divalent cations consisting of only one chemical species. The results of this work indicate that even in a seawater medium cobalt ions were not significantly complexed with any ligands, though the possibility of the formation of labile complexes was not completely excluded. In any case, the formation of labile complexes does not affect the equilibria of reactions of cobalt ions with other reactants since the dissociation of these complexes is easy and rapid. However, as shown by the experimental results obtained by the present authors on the complex formation of cobalt with EDTA, which are given below, the apparent activity coefficient of cobalt in the seawater medium seems to be much smaller than that expected for divalent ions in the ionic strength of seawater. This may be attributed to possible hydrolysis of cobaltous ions in the seawater medium.

In the case of silver the prevention of precipitate formation in a seawater medium, reported by Murray and Murray [46], should be attributed to the complex formation of silver with chloride ions. Another set of results obtained by the present authors on the particulate formation of silver in varying mixtures of seawater and distilled water showed that the precipitate formation of silver was 85% in a mixture of 10% seawater and 90% distilled water (by volume) at pH 8.4, while in a mixture of 30% seawater and 70% distilled water the precipitate formation was decreased to approximately 25%. These results confirm the possible complex formation of silver with chloride ions in the seawater medium.

Based on these data the inorganic complex formation of cobalt and silver in natural waters is not expected to be significant, except for the case of silver in the seawater medium. As has been shown, the latter case affects considerably the fate of silver in the marine environment.

3. Complex formation with organic ligands

There is evidence that some fractions of trace metals in seawater occur in the form of organic complexes of various kinds. Reviewing critically the evidence obtained up to 1970, Fukai [49] concluded that the occurrence of organically bound trace elements in seawater could be regarded as an established fact, although the chemical nature of organic compounds involved and the mechanisms of the formation of organic-metal compounds were not clearly understood. Especially for cobalt and silver no evidence from field data has been obtained.

Proposing a method for the estimation of total cobalt in seawater, Krishnamoorthy et al. [50] implied the presence of organically bound cobalt in seawater, though they did not give any proof of the in situ occurrence of

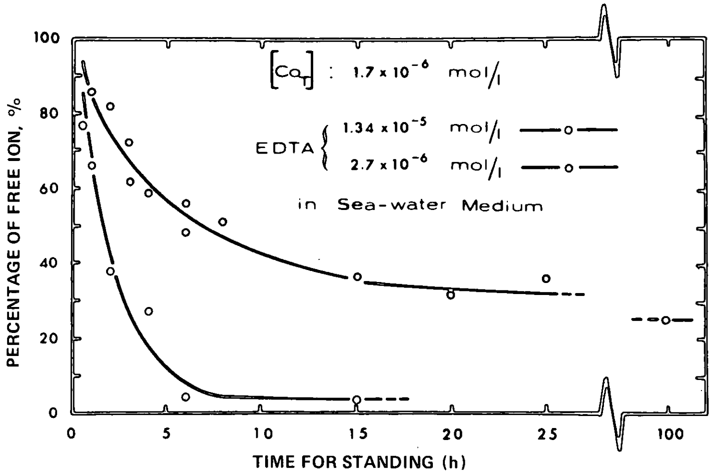

FIG. 2. Chelate formation of cobalt with EDTA in the course of time at two different concentrations of EDTA in the seawater medium.

such compounds. Koshy et al. [51] studied the association in the seawater medium of various trace elements including cobalt with humic acid extracted from a marine sediment, observing slow formation of non-ionic and anionic complexes of cobalt in laboratory conditions. By using dissolved organic matter in a seawater medium obtained from phytoplankton culture solutions, Pillai et al. [52] demonstrated that 72% of cationic cobalt added to this organic solution was transformed to non-ionic forms during 4 weeks and then remained unchanged up to 55 days. These results suggest that the in situ association of ionic cobalt with soluble organic matter is possible if sufficient concentrations of soluble organic matter are maintained under natural conditions.

On the other hand, on the basis of their results of precipitation experiments of metals in the presence of EDTA and other organic compounds, Duursma and Sevenhuysen [53] denied the possibility of the chelate formation of metals including cobalt in natural seawater. Based on further experimental results concerning the effect of dissolved organic matter on the sorption of cobalt-60 and zinc-65 onto freshwater and marine sediments, Duursma [54] concluded that no chelation of divalent cobalt and zinc is possible at the concentration level of 1 μg/litre of leucine; these elements could be chelated if chelating agents stronger than leucine occur in seawater at a concentration of some tenths of a milligram per litre as organic carbon.

Using dialytic equilibria between radionuclide-labelled and non-labelled sample waters, Barsdate [55] found that well over 50% of cobalt together with manganese and zinc were in complexation or association with highly dispersed organic colloids in the waters of distrophic lakes in Alaska, while complexation of the type detected by this specific method was unimportant in the marine environment.

A well-known organic-cobalt compound in natural waters is cobalamin. Lowman and Ting [56] stated that 35-50% of the total cobalt, which occurred at concentration levels of 2-5 μg/litre in the estuarine water of Puerto Rico,

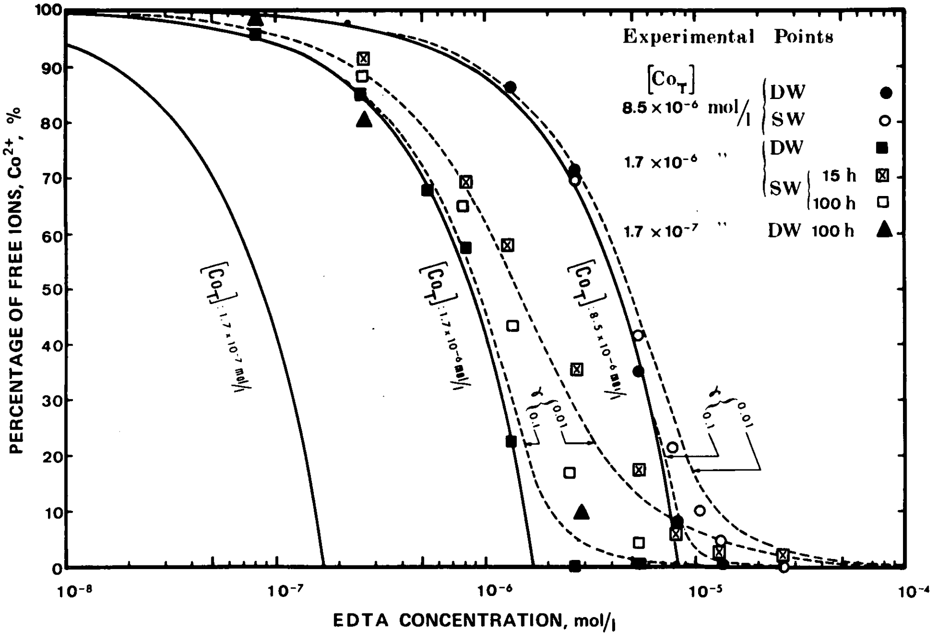

FIG. 3. Chelate formation of cobalt with varying concentrations of EDTA. DW: In a distilled-water medium; SW: in a seawater medium; γ: activity coefficient of cobalt.

was in a complexed form with organic matter, and used cobalt-57 labelled cobalamin and ionic cobalt-58 in the experiments designed for demonstrating different uptake patterns between these chemical forms by marine organisms. This gave an impression that cobalamin is a representative organic compound in the marine environment. As has already been pointed out by Fukai [57], the quantitative contribution of cyanocobalamin to the total cobalt in seawater cannot exceed a few per cent at maximum, since the concentration level of cyanocobalamin and its analogues is in the order of magnitude of 10 ng/litre and the cobalt fraction of cyanocobalamin amounts only to 4% of this compound.

To obtain some idea on the kinetics of the chelate formation of cobalt in natural waters, a series of model experiments were carried out by the present authors using EDTA as an example of organic ligands. In these experiments the degree of chelation was examined by the adsorption of free cobalt ions onto solid manganese dioxide prepared according to Fukai [57]. In Fig. 2 the chelate formation of an exaggerated concentration of divalent cobalt ions, 1.7×10^{-6} mole/litre (100 μg Co/litre) with 2 concentration levels of EDTA is shown. In spite of the fact that the chelate formation of divalent metals with EDTA is known to be a rapid reaction in normal laboratory conditions, the results given in Fig. 2 indicate that 2.7×10^{-6} mole/litre of EDTA chelates cobalt ions only partially after 100 hours contact, and approximately 10-fold equivalent of EDTA to cobalt, 1.34×10^{-5} mole/litre, takes more than 5 hours to chelate cobalt ions. The chelate formations of various concentrations of cobalt with varying quantities of EDTA are shown in Fig. 3. In this figure solid curves represent theoretical computations of the chelate formations without taking the activity coefficient of cobalt into account, while broken curves correspond to those based on an activity coefficient of 0.1 or 0.01, respectively. It is apparent from Fig. 3 that in higher cobalt concentrations (1.7×10^{-6} mole/litre = 100 μg Co/litre <) the solid curves agree with observed values in a distilled water medium after a few hours' duration of the experiments, while in a seawater medium the observed points for 100 hours' duration fall between the two broken curves that correspond to the assumed cobalt activity coefficients 0.1 and 0.01. For more diluted cobalt concentrations (1.7×10^{-7} mole/litre = 10 μg Co/litre >) the experimental points for 100 hours' duration deviate widely from the theoretical curve even in a distilled water medium. These experimental results indicate that the rate of the chelate formation reaction of cobalt with EDTA should very much depend on the concentrations of both cobalt and EDTA, especially in a medium of a solution of strong electrolytes such as seawater. If this can be generalized to actual chelating agents occurring in situ, the chelate formation of cobalt in the extremely diluted concentration in natural waters should be a very slow process.

Reviewing the above data, the picture of the complex formation of ionic cobalt with soluble organic matter in natural waters is not yet conclusive. Taking the kinetic aspect of complex forming reactions discussed above into account, however, it is reasonable to conclude that this kind of reaction between the reactants in extremely diluted concentrations encountered in natural waters proceeds at a slow rate, unless heterogeneous surfaces, such as inorganic particles or detritus where the reactants are highly concentrated, are involved.

Practically nothing is known about the organic complex formation of silver. Considering the fact that stability constants of various chelate compounds of monovalent silver are much smaller than those of divalent cobalt, the organic complex formation of silver in natural waters is supposed to be much less effective than that of cobalt.

4. Metabolic and decomposition products

The suggested slow rate of organic complex formation of cobalt does not necessarily imply a less probable occurrence of organically bound cobalt in natural waters. Metabolic products of living organisms and decomposition products of dead bodies may be other sources of organically bound cobalt or similar compounds of other trace elements. Actually, the above-mentioned cobalamin is one of the compounds that belong to this category. Fukai et al. [58] demonstrated the in situ transformation of zinc extractable with dithizone into an unextractable form, which might be attributed to biological activity.

A series of experiments were conducted by the present authors on the formation of organically bound cobalt from the decomposition of marine organisms. After 10 g (wet) of living seaweed was kept for 10 days in a culture medium spiked with cobalt-60, it was killed by immersing it into hot water and letting it decompose in a seawater medium at room temperature (15-18°C). At certain time intervals a portion of the medium water was taken, millipore filtered and then dialysed against seawater. The process of dialysis was followed by measuring the cobalt-60 activity and organic matter remaining in the dialysis bag respectively by γ-scintillation counting and permanganate titration. The results for the interaction of ionic cobalt and EDTA are given

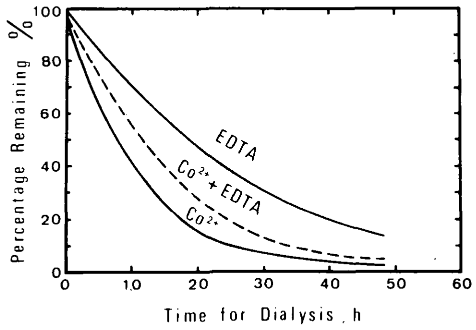

FIG.4. Dialysis of cobalt or EDTA in the seawater medium as well as the mixture of these two components.

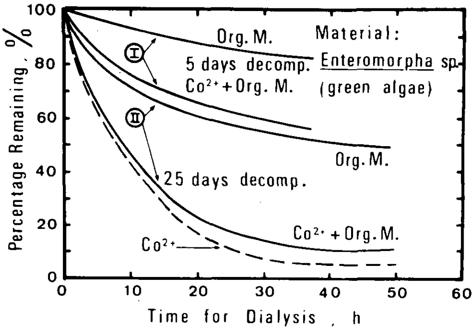

FIG. 5. Dialysis of decomposition products of cobalt-labelled seaweed.

in Fig. 4 to illustrate how organically bound cobalt can be identified by the dialysis experiments. The solid curves in Fig. 4 represent the process of dialysis with time for pure ionic cobalt and EDTA in a seawater medium, while the decrease of cobalt-60 activity in the dialysis bag with time in the presence of EDTA is shown as a broken curve. The retardation of the rate of dialysis of cobalt-60 in the presence of EDTA is a semi-quantitative measure of the association of cobalt-60 with EDTA. In a similar way the association of cobalt-60 with organic molecules in actual samples can be estimated. Figure 5 gives an example of the decomposition experiment with green algae (Enteromorpha sp.). From Fig. 5 it can be seen that 5 days after the start of the decomposition considerable parts of cobalt-60 were bound with organic matter, while after 25 days only a small fraction of cobalt-60 was in association with organic matter and, at the same time, the average size of the organic molecules was considerably reduced. Although the experimental conditions, which affect the rate of decomposition, such as bacterial populations, light, temperature, pH, etc. were not strictly controlled, the results obtained support the possibility of the occurrence of organically bound cobalt in natural waters for a certain period, due to the decomposition of dead organisms. Considering the favourable conditions for bacterial growth in the experiments, the rate of decomposition observed is likely to be much faster than that under natural conditions. Experimental proof of the occurrence of such compounds in natural waters and the identification of organic compounds involved and their kinetics are open for future studies.

5. Sorption on suspended matter and sediments

As already discussed in the foregoing, the major sink of radiocobalt in the freshwater or marine environment is bottom sediments. Although radiosilver may stay in solution for some time in the seawater medium, it should eventually be incorporated into suspended materials and deposited on the sediments. In this view the sorption kinetics and equilibria of these radionuclides with suspended matter and sediments play a substantial role in determining their fate in natural waters.

The sorption of various radionuclides including cobalt-60 on suspended particles and sediments was extensively studied by Duursma and Bosch [59] under laboratory conditions, although mechanisms of the sorption were not clearly defined. They found that the distribution coefficient of cobalt-60 between sediments and ambient water was 6×10^3 in oxygenated conditions and that, specifically for cobalt-60, the uptake by sediments was very much affected by the experimental methods used. The problem remained unsolved as to whether this observed difference in cobalt uptake by sediments between various experimental conditions is merely due to experimental techniques, which measure different phenomena, or represents quasi-equilibria where different chemical forms of cobalt are involved.

Murray and Murray [46] studied the adsorption of cobalt-60 and silver-110m by sediments in a freshwater medium and successive desorption of these radionuclides from the sediments when they were brought in contact with seawater. They found that more than 80% of both radionuclides were adsorbed on the sediments within 48 hours in a freshwater medium at pH 8.0; the adsorption of silver-110m was specially rapid and occurred within a few hours. As already discussed, the particulate formation of silver in a freshwater medium should substantially contribute to this apparent adsorption. Cobalt-60 and silver-110m behaved differently with regard to desorption. While about 30% of adsorbed cobalt-60 was desorbed from sediments in a seawater medium at pH 8.0 within 24 hours, more than 70% of the adsorbed silver-110m was desorbed. More time was required by silver to arrive at desorption equilibrium, around 48 hours, which is considerably longer than the 24 hours required to establish equilibrium in the case of cobalt-60. Again, the complex formation of silver with chloride ions in seawater seems to play a decisive role in the desorption process. One characteristic feature of these adsorption-desorption equilibria for cobalt is a strong dependence on the pH of the ambient water. The percentage adsorption of cobalt was approximately 20% at pH 7 and 80% at pH 8, while 75% and 30% was obtained at the same pH values using seawater. Under certain circumstances, a slight change in the pH of natural water may significantly alter the adsorption-desorption equilibria of cobalt with sediment particles. In the case of silver the pH dependency of sorption equilibria is much moderated.

Concerning the effects of soluble organic matter on the adsorption-desorption equilibria, data of Murray and Meinke [60] show that cobalt adsorption from a freshwater medium on sediment particles decreased with increasing concentrations of soluble organic matter, while the desorption in seawater of the adsorbed fraction of cobalt was not affected, irrespective of the concentration of soluble organic matter in the adsorption medium. The observations suggest that some fraction of cobalt was bound to non-adsorbable organic matter in the freshwater and the chemical form of adsorbed cobalt was not affected by the addition of soluble organic matter in the adsorption media. On the other hand, the reverse effect was observed for silver; the adsorption of silver was not affected by the addition of organic matter, while the desorption was decreased from 80 to 20% of adsorbed silver by the addition of a high concentration of organic matter in the adsorption media. It seems that the organic matter served as a bridge to bind silver more tightly to the sediments.

Reviewing both the field data and the results of laboratory experiments obtained to date, it is noted that the importance of the sorption of radiocobalt and radiosilver by suspended particles and sediments has been well demonstrated, while the mechanisms involved in the sorption processes are not yet clearly understood. Since the conditions encountered in situ represent a complicated combination of various mechanisms, a reasonable understanding of the mechanisms is essential to apply these data to new study sites. Further studies on the sorption equilibria of radionuclides by suspended matter and sediments are required to make the data obtained useful for predicting the fate of these radionuclides in the natural aquatic environment.

GEOCHEMICAL BEHAVIOUR IN THE HYDROSPHERE

The chemical speciation patterns of radiocobalt and radiosilver discussed above should also reflect the behaviour of stable cobalt and silver in natural waters. The behaviour of stable cobalt and silver in the Neuse River, North Carolina, was studied by Turekian et al.[61] and that of Long Island Sound by Schutz and Turekian [62]. On the basis of these data and those obtained on Connecticut streams Turekian [63] discussed the geochemical behaviour of stable cobalt and silver in the hydrosphere, especially in stream waters and estuarine conditions. He concluded that although cationic species of cobalt and silver are effectively adsorbed by suspended matter in stream water, they are partially released when the suspended matter comes into contact with the estuarine water; a short distance from the point of injection these redissolved metals are removed from the dissolved state by planktonic organisms, which disintegrate after death, mostly on the bottom in shallow areas; these metals are retained in reducing sediments, especially in the form of insoluble sulphides or reduced oxides; thus little or none of the cobalt or silver load supplied by streams leaves the estuary but is trapped in the bottom sediments. Although the behaviour of these metals in natural waters, especially in estuarine conditions, is hard to generalize to various different conditions, the overall picture of the behaviour of cobalt and silver in natural waters deduced by Turekian [63] agrees in general with the foregoing.

The seasonal variation in stable cobalt and silver in natural waters has not yet been thoroughly studied. Figures 6 and 7 present, as examples,

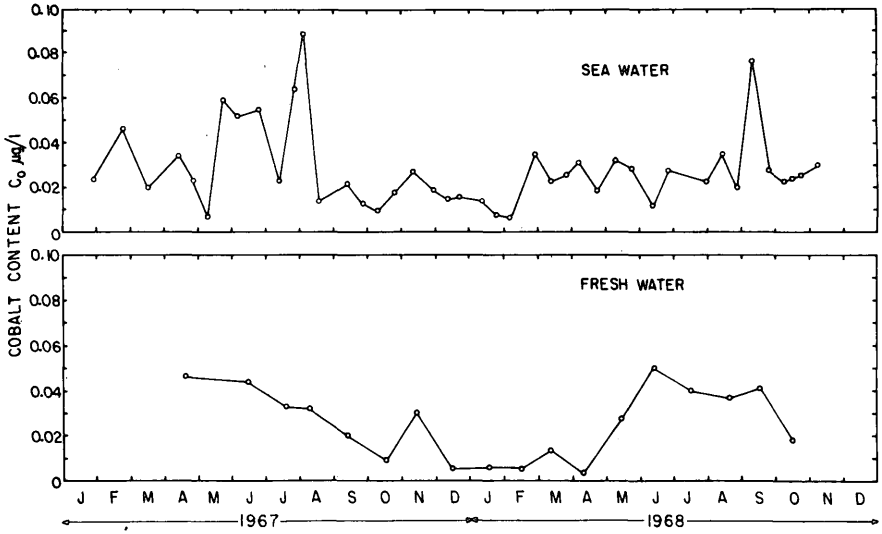

FIG. 6. Seasonal variation of cobalt in the seawater around Monaco and in the Var River.

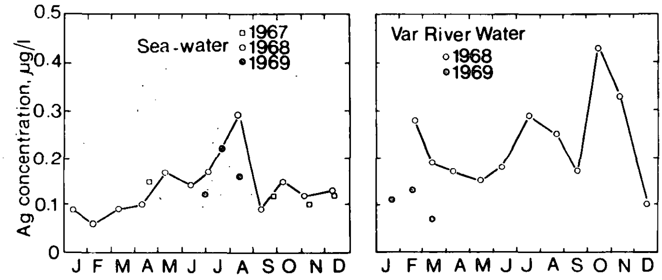

FIG. 7. Seasonal variation of silver in the seawater around Monaco and in the Var River [64].

the seasonal variations of stable cobalt (Fig.6) and silver (Fig.7) measured in the Var river and at a coastal station near Monaco. The cobalt data are original values obtained by the present authors and those for silver were taken from published data by Fukai and Huynh-Ngoc [64]. Since biological activities in the freshwater and coastal seawater in this area are not considered as a major factor in regulating the concentration of these elements, the variations should represent the reflection of combination of the chemical mechanisms discussed in the foregoing section. Attempts were made to correlate the variation in silver in the coastal area with that in the river, taking the expected desorption equilibria of silver in the estuarine condition into account [46, 65]. Although the results were not conclusive, they indicate that the silver desorption from the suspended particles transported by the Var River into the sea may affect the variation in silver in the estuary and its neighbourhood.

For a better understanding of the geochemical behaviour of cobalt and silver in the hydrosphere it seems important in future studies to quantify the contribution of various mechanisms to the overall fate of these elements under natural conditions. Studies on the radiologically labelled environment may give occasion to accumulate such information under various natural conditions.

CONCENTRATION FACTOR OF COBALT

It is not the purpose of the present paper to cover the interaction of radiocobalt and radiosilver with aquatic organisms, though this is another significant factor in controlling radionuclide concentrations in the aquatic environment and is especially important for hazard assessments of radiologically contaminated environments. The uptake and loss of radiocobalt and the related literature have been well covered by recent papers by various workers [66-69]. However, since, as has been pointed out by Robertson [70], most of the cobalt concentrations in seawater reported prior to 1968 are much too high, due perhaps to sample contamination, cobalt concentration factors for marine organisms computed on the bases of the former high data of stable cobalt for seawater are seriously in error. The cobalt data presented above by the present authors for fresh water and seawater, as well as those of Morris [71], Topping [72] and Robertson [70] for seawater and those of Merlini et al. [73] for Lake Maggiore water, are around 0.05 μg Co/litre or less, except under special circumstances. From this point of view the concentration factors for cobalt presented, for example, by Ichikawa [74] or Bryan et al. [75] for marine organisms should be reconsidered.

Based on the results of determinations of cobalt in various marine organisms given by Fukai [76, 77] and the seawater concentration of 0.05 μg Co/litre, approximate concentration factors of cobalt for several groups of marine organisms were computed and presented in Table II. The order of magnitude of the concentration factor of cobalt for fish

TABLE II. CONCENTRATION FACTORS OF COBALT FOR VARIOUS GROUPS OF MARINE ORGANISMS

Group of organism	Standard abundance of cobalt (μg/kg fresh)	Concentration factor
Algae	100	2000
Crustacea		
Copepoda (whole animal)	200	4000
Decapoda (soft parts)	40	800
Mollusca		
Lamellibranchia and Gastropoda (soft parts)	100	2000
Fish (flesh)	3	60

presented in Table II agrees well with those reported by Ophel and Fraser [40] for young fish, which were based on measurements of radiocobalt in water and fish collected from the radiologically contaminated Perch Lake, Canada. Since a number of determinations of extremely low concentrations of stable cobalt and silver in natural waters are not an easy task, the measurements of concentration factors in the radiologically labelled environment may offer simpler means of the study.

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DISCUSSION

A. ORTINS DE BETTENCOURT: In two series of similar experiments we have found 20-25% differences in the accumulation factor and I should therefore like to ask you whether, under experimental conditions that can be regarded as identical, you find variations in the ionic form of the cobalt present in the water.

R. FUKAI: If the experimental conditions really are identical, the variation should not, in my opinion, exceed ±5%. However, it may be that the water used in the experiment is not really identical and in such cases the ionic cobalt may vary by as much as 50%, as shown by Lowman in Ref.[56] of the paper.

N. T. MITCHELL: Mr. Fukai's paper is of considerable interest to us, not least because he has highlighted some of the problems we have ourselves been investigating, particularly the environmental behaviour of silver. I would, in this connection, be interested to hear his evidence for assuming that silver is in the form of the chloride complex; in particular, has this species been identified in estuarine seawater?

What is especially notable in the case of silver is the extreme contrast between its behaviour in the water mass, which can be described as conservative, and its behaviour in biota (we have found that it reconcentrates in oysters to a very high degree indeed). In this contrast between sediment and biota the behaviour of silver is not unique, and we have observed a similar situation with technetium-99, though in the case of this nuclide the extremely high reconcentration occurs with algae rather than shellfish.

R. FUKAI: The evidence for the formation of chloride complexes of silver in estuarine water is, strictly speaking, only circumstantial. However, as is discussed in the paper, the fact that increasing concentration of chloride affects the sorption characteristics or precipitation of silver in laboratory experiments can only be interpreted quantitatively assuming complex formation with the chloride.

The conservative behaviour of silver in water masses and its high concentration factor in oysters are not, I think, mutually contradictory, provided one postulates a specific mechanism for the uptake of silver by oysters, while in the case of ^{99}Tc the uptake by algae may be considered to be due to surface adsorption.

FLOOD PLAIN TRANSFER AND ACCUMULATION OF ^{137}Cs FROM A REACTOR EFFLUENT STREAM

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Presented by P.G. Mayer

Abstract

FLOOD PLAIN TRANSFER AND ACCUMULATION OF ^{137}Cs FROM A REACTOR EFFLUENT STREAM.

Analysis of the flood plain swamp forest ecosystem of a reactor effluent stream in South Carolina revealed significant lateral transport of ^{137}Cs from the stream channel to the flood plain. Six flood plain transects, representing 13 miles of the stream, were extended from the stream bank into the swamp forests. Soil, root, litter, branch and leaf samples were collected at 100-ft intervals up to a distance of 300 ft along each transect. Upstream burdens of ^{137}Cs reached mean values of 225 pCi/g-d (g dry wt.) in soil and 100 pCi/g-d in vegetation, while downstream burdens were found to be 100 pCi/g-d in soil and 15 pCi/g-d in vegetation. The upstream-downstream differences indicate that significant biological cycling of radiocaesium occurs upstream, while downstream there is accumulation through immobilization in the soil. Levels of ^{137}Cs in the flood plain were variable with soils, litter and vegetation ranging from near-background to 100 pCi/g-d and roots ranging up to 340 pCi/g-d. Flood plain distribution of radiocaesium correlated with topographic relief resulting in non-uniform levels of ^{137}Cs where flood plain depressions allowed for physical accumulation of radioactivity.

These results lead to a rejection of the commonly assumed hypothesis of stream containment of radionuclides for streams in coastal plain areas. A new model of the fate of reactor effluents discharged to a coastal plain stream must account for discreet flooding over low banks, physical movement of suspended and soluble forms of radionuclides in terrestrial habitats, biological cycling and accumulation in the flood plain, redistribution of biologically incorporated radionuclides back to the stream, and long-term erosional movement of sedimentary radionuclear materials due to stream bed movements.

INTRODUCTION

The release of radioactive substances into a stream pre-supposes they will be diluted by the stream water volume and dispersed downstream with the possibility of particulate and sediment sorption of the radioactivity [1,2]. Such a model also tacitly assumes total containment of the radioactivity within the stream channel. Cesium-137 is one of the environmentally important radionuclides which has received considerable attention because of its occurrence in fallout and as a by-product of applied nuclear technology, its long half-life, and its beta-gamma emissions. Large scale environmental surveys of radiocesium have documented its overland sedimentary flow [3,4]; its uptake, accumulation, and elimination in aquatic ecosystems [5,6,7,8]; its localization in sediments [9]; and its behavior in various substrate systems [10,11,12]. Smaller scale studies have utilized aquatic microcosms to focus on the mechanisms of ^{137}Cs kinetics under various imposed conditions [1,13,14,15,16]. The results of these and numerous other works [2,17,18] support the model of downstream dispersion and dilution of aquatically released ^{137}Cs.

This study reports on the subsequent fate of an accidental release of ^{137}Cs to a black water creek of the Atlantic coastal plain, South Carolina, U.S.A.

RESEARCH AREA

Lower Three Runs Creek (LTRC) is located on the Savannah River Plant property, USAEC, near Aiken, South Carolina (Fig. 1). This area is in the upper coastal plain, approximately 64 kilometers due south of the fall line marking the inland extent of the Atlantic and Gulf coastal plain. The watershed of LTRC occupies 466 km^2 and the creek, prior to 1958, extended 40 km to the Savannah River. Par Pond Dam was constructed in 1958 blocking off the upper 8 km of the creek and an associated 70 km^2 of watershed.

The stream bed slope of LTRC decreases continually from .0009 m/m at the dam to .00035 m/m near its confluence with the Savannah River. Travel time between the dam and the Savannah River is estimated as 29 hours under the assumption of a uniform channel width. However, this assumption is not valid since the stream channel width is variable from 3 to 12 m in those areas of the creek having a defined channel. At certain points in the stream length, between Donora and Boiling Springs, the channel is not defined and the flow diffuses into extensive swampy flood plains.

The banks along LTRC are easily categorized into low and high banks, usually opposing, with the low banks just containing the flow under expected rainfall conditions in mid-April. High banks are 0.6-3 m above the water surface at this

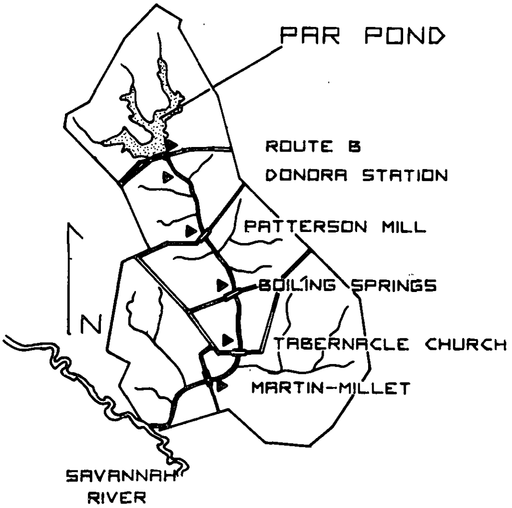

FIG. 1. Lower Three Runs Creek watershed and sampling stations.

period. The stream channel is open during the winter, but is much reduced in size during the spring and summer due to the growth of Polygonum sp., the dominant species of the emergent vegetation zone. A narrow strip of stream bank forest along the banks is characterized by black willow (Salix nigra) and associated tree species such as red maple (Acer rubrum), sycamore (Platanus occidentalis), and river birch (Betula nigra). The flood plains may extend to 244 m beyond the banks and swamp forests or oak-hickory forests commonly occur in the flood plains.

The low bank flood plain is characterized as a swamp forest. Red maple, ash (Fraxinus sp.), black and water gum (Nyssa sylvatica and N. aquatica), and bald cypress (Taxodium distichum) are the principal tree species. During the winter much of the flood plain is under water. The water table is level with, or slightly above, the forest floor during early spring.

The flood plains tend to be either flat or concave and in the latter case the water table remains near the forest floor surface for longer periods of time at the low point in the flood plain. Numerous intermittant streams and rivulets dissect the flood plain running both parallel and transverse to the stream channel.

METHODOLOGY

A 20 km segment of LTRC was sampled at six stations during April, 1972 (Fig.1). A control station was selected on Upper Three Runs Creek (UTRC), a separate but nearby tributary of the Savannah River which has not received reactor effluent. Flood plain transects, each consisting of four locations spaced 30 m apart, were established at the UTRC control and six LTRC stations. Substrate and vegetation samples were collected in triplicate at each transect location. Red maple trees were selected at each location and leaves and five year old stems were sampled. A 20 x 20 cm quadrat was placed near each selected tree for obtaining litter and humus samples. A 23 cm soil core was obtained from within each quadrat. Roots were then removed from the quadrat to a depth of 10 cm.

All samples were handled separately with attention to sample preservation and preparation in the laboratory. ^{137}Cs levels were determined using a Nuclear Chicago Autogamma system with a 3 x 3 inch Na(Tl) crystal and a 100 kev window. Preliminary screening by Ge(Li) gamma spectroscopy indicated that other nuclides were not contributing to the ^{137}Cs count. The minimum detectable activity was about 7 pCi and the exact detectable levels were determined with each count set. Samples whose count did not exceed the 3 sigma error of the average background during the count were rejected. Samples were corrected for sample volume geometry before calculating activity density on a pCi/g dry weight (g -d) basis.

Statistical comparisons of means of similar magnitude were made with analysis of variance tests on untransformed data. For certain parameters the presence of non-homogeneous variances necessitated the use of square root transformations. Both transformed and untransformed data satisfied the statistical requirement of linearity.

RESULTS

Mean ^{137}Cs burdens of the soil and vegetation of Upper Three Runs Creek (control area) ranged from 0.99 to 12.90 pCi/g (Table I). The soil burdens were fairly similar to 60 m and then dropped by 50% at the 90 m location. The mean radiocesium burden for bank roots was 9.84 pCi/g, about twice as great as the 30-90 m stations. Litter values ranged from 6 to 13 pCi/g over the 90 m transect. Red maple leaves averaged 7.6 pCi/g across the flood plain as compared to 3.6 pCi/g for branches.

Significant differences ($p<0.05$) existed among soil and among root mean activity densities across the floodplain. However, the means calculated over all flood plain locations were used as the naturally occurring background levels for flood plain components.

The ^{137}Cs burdens at Route B (Rt.B) varied from 1 to 43 pCi/g (Fig.2). Leaves, roots, and soil burdens were similar at all flood plain intervals with higher ^{137}Cs activities along the stream bank. Radiocesium levels decreased from 40 pCi/g at the bank to background levels along the flood plain. Litter and branch samples were near background at all transect stations.

TABLE I. ^{137}Cs BURDEN OF UPPER THREE RUNS CREEK FLOOD PLAIN COMPONENTS

	Bank	30 m	60 m	90 m	Average
0-7.5 cm Soil	2.18	1.75	3.68	0.99	*2.13
Roots	9.84	4.63	4.10	3.48	*5.51
Litter	8.17	6.27	12.90	6.42	5.69
Red Maple Leaves	7.04	6.88	6.80	9.66	7.60
Red Maple Branches	2.60	3.06	6.20	2.50	3.59

*Indicates significant differences were present at the 0.05 level.

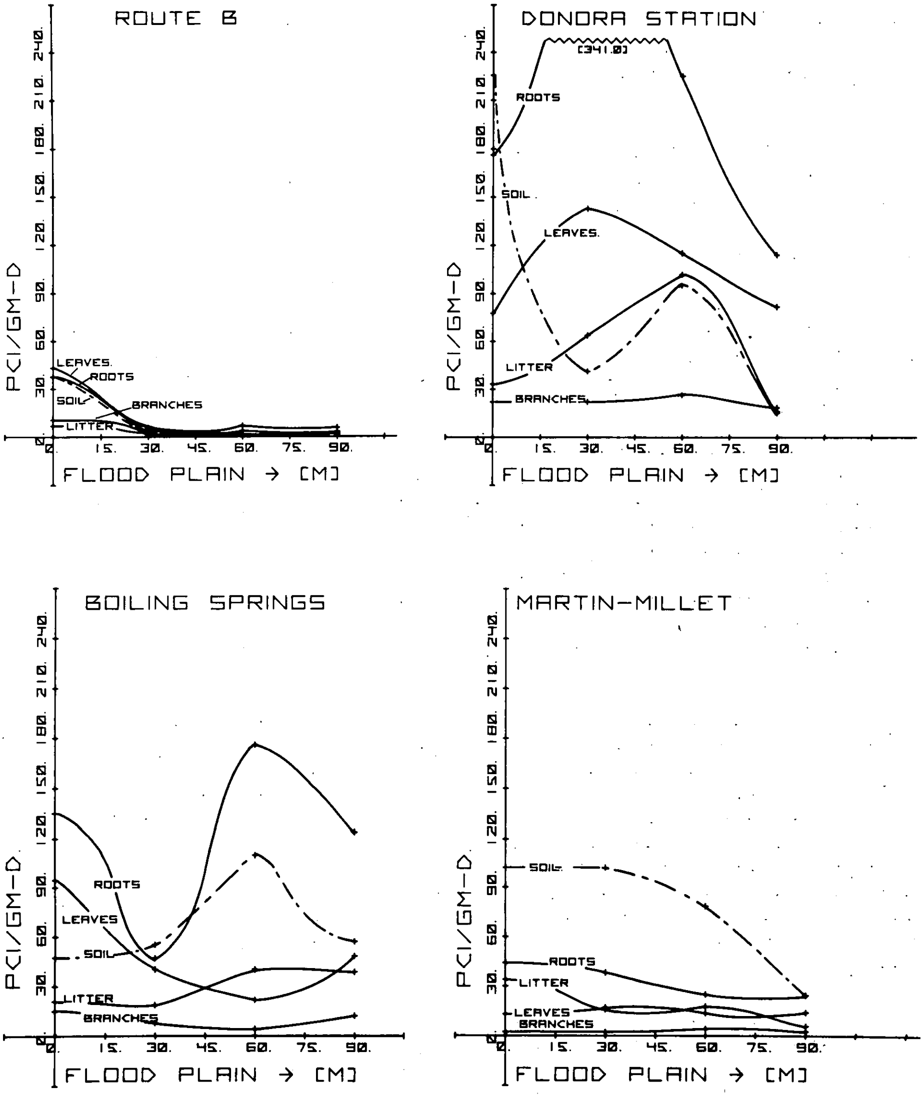

FIG.2. ^{137}Cs concentration in major ecosystem components at four locations along Lower Three Runs Creek (1972). The data are expressed as pCi/g dry weight.

At Donora Station (Fig. 2), the ^{137}Cs levels ranged from 18 to 341 pCi/g Leaves varied from 77 to 143 pCi/g with a maximum value at the 30 m location. Roots had the highest ^{137}Cs burden with a minimum of 113 pCi/g at 90 m and a maximum of 341 pCi/g at 30 m. Litter values were highest (100 pCi/g) at the 60 m location. The soil ^{137}Cs burden dropped from 225 pCi/gm at the bank to 14 pCi/g by the 90 m location.

The ^{137}Cs levels (4-176 pCi/g) at Boiling Springs (Fig. 2), were lower than those of Donora Station. Root activity densities were bimodal with peaks at the bank and 60 m locations. Branch activities were low and similar across the flood plain. Red maple leaves had a maximum of 95 pCi/g at the stream bank and decreased to an average of 36 pCi/g over the flood plain. Soil ^{137}Cs levels peaked at 60 m (109 pCi/g), with lower, similar values for the other locations. ^{137}Cs litter burdens were low at the bank and 30 m stations and increased to 40 pCi/g at more distant locations.

The maximum radiocesium level (102 pCi/g) at Martin-Millet (Fig. 2), was lower than both Donora and Boiling Springs. Soil ^{137}Cs levels remained well above the other ecosystem components up to the 90 m location. The root component was relatively constant up to 30 m, decreasing from 44 to 25 pCi/g by 90 m. Leaf burdens were similar across the flood plain. Branches averaged 2.8 pCi/g over all locations. For litter, a maximum burden of 34 pCi/g occurred at the bank and this decreased to 6 pCi/g at the 90 m station.

The greatest contrast among these four stations is the lack of significant radiocesium levels in the Rt. B flood plain. ^{137}Cs burdens at the Rt. B bank are comparable to those downstream. The lack of flood plain activity at Rt.B is explained by the presence of a high bank which is 1.8 to 2.4 m above the early spring water level. Since the Rt. B bank is labeled with ^{137}Cs it must be concluded that the released radiocesium was contained by the channel in this region. Another high bank station was sampled at Patterson Mill and the radioactivity levels were lower than those on the corresponding low bank at Patterson Mill.

Donora, Boiling Springs, and Martin-Millet all had low banks and the flood plains of each station have accumulated ^{137}Cs levels significantly higher than control and high bank stations. Comparison of the maximum levels of laterally transported ^{137}Cs between Donora and Martin-Millet shows a 6 to 8 fold decrease in the radioactivity of most components and only a 2 fold decrease in the ^{137}Cs soil burden. Statistical comparison of these levels indicated a significant downstream decrease in ^{137}Cs levels ($p<0.05$).

The patterns of ^{137}Cs distribution across the flood plain are not uniform for ecosystem components. At Donora and Boiling Springs there is a peak activity in the soil component at 60 m. At Martin-Millet, the greatest soil burdens occurred in the first 30 m. The litter and soil patterns are well correlated at each of the three stations.

The highest litter burdens occurred 30 to 60 m away from the highest leaf burdens at Donora and Boiling Springs. Statistical analyses indicate mean differences ($p<0.05$) in ^{137}Cs levels for these components over the flood plain.

At Donora and Boiling Springs the lowest point in the flood plain occurred at 45 to 60 m from the bank while the low point at Martin-Millet was at 30 m. These topographical lows were correlated with the maximum substrate burdens of ^{137}Cs. The 30 to 60 m difference between litter and leaf activities suggests that the leaves are deposited or transported overland to the low point of the flood plain.

The vertical distribution of ^{137}Cs in soils (Table II) shows that in the upstream areas most of the 137Cs is in the upper 7.5 cm of soil. Downstream, the radiocesium burden is relatively constant down to 15 cm. The increased depth of the ^{137}Cs burden at Martin-Millet suggests either downward mobility of the ^{137}Cs or possible sedimentary deposition on the Martin-Millet flood plain.

The question of ^{137}Cs cycling in these flood plain habitats can be examined by comparing the vegetation to substrate radioactivity burdens (Table III). The highest leaf/substrate ratios occurred at Donora station and at Martin-Millet they had decreased to fractional values. A similar pattern was present for root substrate ratios. The small root ratios, representing young actively growing tissue,

TABLE II. VERTICAL DISTRIBUTION OF RADIOCESIUM BURDENS IN SOIL

Station	Soil Depth[cm]	Bank	30 m	60 m	90 m
Donora	0-7.5	225	41	95	15
	7.5-15	74	7	9*	4
	15-22.5	12	10	-	2
Boiling Springs	0-7.5	48	55	109	57
	7.5-15	51	11	11	9
	15-22.5	9	3	4	3
Martin-Millet	0-7.5	102	102	79	25
	7.5-15	74	60	79	12
	15-22.5	12	-	43*	8

*One observation

TABLE III. VEGETATION TO SUBSTRATE RATIOS OF ^{137}Cs

Vegetation Component	Floodplain Location	Donora	Boiling Springs	Martin-Millett
Roots	30 m	8.3	0.85	0.3
	60 m	2.3	1.6	0.3
	90 m	7.6	2.1	0.9
Leaves	30 m	3.47	0.73	0.16
	60 m	1.20	0.20	0.17
	90 m	5.45	2.1	0.57
Young roots	30 m	15.42	1.43	0.64
	60 m	4.43	2.29	0.61
	90 m	13.12	4.03	1.65

ranged even greater from 4 to 15 at Donora and decreased to .6 at Martin-Millet. These vegetation to substrate ratios indicate a consistant pattern of bio-accumulation in the upstream locations, near equilibrium values at intermediate locations, and apparent downstream soil immobilization of the ^{137}Cs.

DISCUSSION

The question of stream channel containment of water is usually not raised in considering radiochemical releases to streams. The available data have not indicated a need for this consideration. In the case of LTRC and similar coastal plain streams, the containment of stream water within the channel is a significant problem. The exact mechanisms leading to the lateral displacement of radioactivity into the LTRC flood plain for this particular situation are unknown. If the accidental release of nuclides to the stream occurred during a period of heavy rainfall or during the winter or early spring when stream flow exceeded the low banks the adjacent areas would have been flooded at the time of nuclide release. However, if the release occurred during a period of relatively low water level the lateral displacement would have been delayed until subsequent flooding. The significant point is that flood plain contamination did occur, thereby clearly establishing the potential for lateral transfer of radioactivity from the stream channel.

A second consideration for coastal plain streams is the tendency for diffusion of the stream into wide swampy areas. For example, such diffusion occurs on LTRC mid-stream between Donora and Boiling Springs. Savannah River Plant Health Physics personnel have collected substantial ^{137}Cs data in the mid-stream swamp of LTRC and the activity levels are comparable to those at Donora station [19]. Even if the possibility of lateral dispersion over low banks was disregarded, the swampy sections of these creeks above the delta will almost certainly receive waterborne radionuclear materials. Thus, the dispersion of radionuclear releases in coastal plain streams will be both downstream and laterally into surrounding flood plains.

The fate of ^{137}Cs laterally dispersed into the flood plains is a function of the downstream location of the flood plain. Cesium activity at Donora Station has been accumulated in the vegetative tissue above levels found in the substrate. Downstream, the ^{137}Cs has accumulated in the soil and does not appear to be actively cycled. Recent work on ^{137}Cs cycling in the upper and lower coastal plains has documented increased cycling of ^{137}Cs in sandy substrates such as occurs at Donora Station [20]. It may be hypothesized that downstream stations contain greater clay fractions as a result of run-off from upland agricultural areas and consequently more ^{137}Cs is bound onto clay colloids at the downstream stations. There also may be greater availability of potassium ions at the downstream stations. Another alternative is that significant amounts of the downstream ^{137}Cs burden in soil has resulted from erosional deposition onto the Martin-Millet flood plain. Hence, particulate bound ^{137}Cs could have arrived at Martin-Millet. Any or all of these conditions would lead to the immobilization of the radiocesium burden at Martin-Millet.

The contrast between upstream ^{137}Cs cycling and accumulation in the biota and apparent downstream immobilization in the substrate raises the question of the long-term fate of this ^{137}Cs. The overland transport of particulate bound ^{137}Cs to the lower flood plains elevations will occur by annual leaf and branch fall and through uplifting of particulates and other litter with flooding conditions. The litter burden at the creek bank will be deposited in LTRC and move downstream subject to various physical, chemical, and biological transformations which determine its state as it moves downstream, into other flood plains, into the midstream swamps, or into the delta swamps. Thus, ecological reconcentration of the previously "diluted" radiocesium may occur through biotic mechanisms such as vegetation uptake and subsequent trophic exchanges and through abiotic mechanisms such as overland transport within flood plains and downstream sediment depositions onto flood plains.

The results of this study document the need for a new model of the fate of radionuclides released to coastal plain streams similar to LTRC. This model must account for discrete flooding of the low banks, physical movement of suspended and soluble forms of radionuclides, biological cycling and accumulation in the swamp forest flood plain, redistribution of both biologically and physically accumulated radionuclides back into the stream, and long term erosional movement of sedimentary radionuclear materials.

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DISCUSSION

I. L. BRISBIN, Jr.: You mentioned in your presentation that certain parameters in your study failed to show homogeneity of variance. We have also encountered this phenomenon in our own studies of radiocaesium in these same swamp ecosystems.

What patterns have you found in your sampling variance as you move to sampling stations located progressively further downstream? Also, what trend towards increasing or decreasing sampling variance is revealed as one moves laterally away from the stream onto the flood plain?

P. G. MAYER: In general, the sampling variance diminished in the downstream direction but increased in the transverse direction. Active biocycling occurred predominantly in the flood plains of the upper regions, whereas soil immobilization was primarily a factor in the downstream regions of the Lower Three Runs Creek.

MECANISMES DE TRANSFERT DANS LES SOLS CULTIVES DES RADIONUCLEIDES REJETES PAR LES CENTRALES ELECTRO-NUCLEAIRES DANS LE SYSTEME FLEUVE–SOL IRRIGUE–NAPPE

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Abstract–Résumé

MECHANISMS FOR THE TRANSFER TO CULTIVATED SOILS OF RADIONUCLIDES DISCHARGED BY NUCLEAR POWER STATIONS INTO THE SYSTEM: RIVER–IRRIGATED SOIL–GROUNDWATER.

The siting of nuclear power stations on the banks of rivers whose water is used for irrigation and industrial and domestic consumption must be preceded by a study in depth of the mechanisms of transfer in the system comprising the river, irrigated soil and groundwater. The mechanisms of transfer to cultivated soils are considered basically from three viewpoints: the influence of the quality of the river water, irrigation channel water and ground water on radionuclide mobility in soils; the influence of the type of soil (four types have been considered: brown acid soil, brown calcic soil, brown calcareous soil and alluvial calcareous soil) and the development and distribution of radionuclides in the soil (water-soluble forms liable to contaminate the groundwater and the more labile forms available to plants). The study described relates to the following radionuclides: ^{22}Na, ^{137}Cs, ^{85}Sr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{124}Sb, ^{141}Ce and ^{131}I. Knowledge of the transfer mechanisms in cultivated soils permits an evaluation of the risks of contamination of the food chain and the groundwater. This study also demonstrates new aspects of the behaviour of radionuclides as a function of the organomineral pollution by industrial and domestic waste of the water into which they are discharged. This pollution has a considerable effect on the formation of stable complexes transferred by the river to irrigated soils. The quality of the water determines the distribution of the radionuclides in the profile. The water-soluble complexes persist in the soil and migrate towards the groundwater if they are not biodegradable or are only slightly so. The stability of these complexes as a function of the pH of the soil and its physico-chemical characteristics and the radionuclides concerned can be used as a basis for drawing up a new balance of the radiological capacity of soils and for formulating new proposals for regulating nuclear power station siting.

MECANISMES DE TRANSFERT DANS LES SOLS CULTIVES DES RADIONUCLEIDES REJETES PAR LES CENTRALES ELECTRO-NUCLEAIRES DANS LE SYSTEME FLEUVE–SOL IRRIGUE–NAPPE.

L'implantation des centrales électro-nucléaires au bord des fleuves dont l'eau est utilisée pour l'irrigation et la consommation industrielle et domestique nécessite une étude approfondie des mécanismes de transfert dans le système fleuve–sol irrigué–nappe. Ces mécanismes de transfert dans les sols cultivés sont envisagés sous trois aspects principaux: l'influence de la qualité des eaux du fleuve, des canaux d'irrigation et de la nappe sur la mobilité des radionucléides dans les sols; l'influence du type de sol (quatre types de sol ont été retenus: sol brun acide, sol brun calcique, sol brun calcaire, sol alluvial calcaire); l'évolution et la répartition des radionucléides dans le sol: formes hydrosolubles pouvant contaminer la nappe, formes plus labiles mises à la disposition des plantes. Cette étude porte sur les radionucléides suivants: ^{22}Na, ^{137}Cs, ^{85}Sr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{124}Sb, ^{141}Ce, ^{131}I. Les mécanismes de transfert dans les sols cultivés permettent d'évaluer les risques de contamination de la chaîne alimentaire et des eaux de la nappe phréatique. Cette étude montre également de nouvelles perspectives de comportement des radionucléides en fonction de la pollution organo-minérale d'origine industrielle et domestique des cours d'eau dans lesquels ils sont rejetés. Cette pollution influe largement sur la formation de complexes stables véhiculés par le fleuve jusqu'aux sols irrigués. La qualité de l'eau détermine la répartition des radionucléides dans le profil. Les complexes hydrosolubles persistent dans le sol et migrent vers la nappe si ces produits sont peu ou pas biodégradables. La stabilité de ces formes en fonction du pH du sol, de ses caractéristiques physico-chimiques et des radionucléides considérés permet de dresser un nouveau bilan de la capacité radiologique des sols et de formuler de nouvelles propositions en matière de contrôle de site nucléaire.

INTRODUCTION

Cette étude vise à déterminer les différents mécanismes de transfert des principaux radionucléides rejetés dans les cours d'eau, dans les sols irrigués et dans la nappe.

Nous avons envisagé, dans une première hypothèse de travail, le rejet des radionucléides dans les eaux brutes d'un fleuve puis l'utilisation de cette eau à des fins agricoles au niveau des diverses prises d'eaux. La deuxième hypothèse de travail est fondée sur la contamination des sols par voie atmosphérique, suivie d'une dissémination par les eaux d'irrigation. La troisième hypothèse de travail repose sur une contamination de la nappe à partir des eaux du fleuve et des eaux d'infiltration du sol et l'utilisation de cette eau à des fins agricoles, industrielles et comme eau de boisson.

Ce rapport résume le comportement des différents radionucléides dans le milieu et les conséquences de ce rejet pour le milieu. L'étude des mécanismes est précédée d'un inventaire rapide des voies de transfert.

1. CHOIX DES TYPES DE SOL ET PARAMETRES D'ETUDE

Nous avons choisi pour présenter notre système quatre types de sols susceptibles de se rencontrer dans la plupart des sites: sol alluvial calcaire, sol brun calcaire, sol brun calcique, sol brun acide. L'hydrographie sera constituée, d'une part, par un fleuve dans lequel sont effectués les rejets de radionucléides, d'autre part, par des rivières secondaires et des canaux d'irrigation avec prise sur le fleuve.

Nous estimons que la surface irriguée recouvre un périmètre de 5 à 10 000 ha et que l'irrigation est effectuée par moitié par les canaux et par moitié par la nappe phréatique alimentée partiellement par le fleuve et les rivières secondaires. Nous admettrons une hauteur d'eau hebdomadaire variable selon les sols et les cultures de 25 à 50 mm, soit 250 à 500 m^3/ha. La profondeur de la nappe fluctue dans notre système entre 1 et 2 m. Elle sert, outre l'irrigation, de réserve d'eau pour les activités industrielles et pour la consommation de la population.

De ce fait, les mécanismes de transfert dans les sols cultivés des radionucléides rejetés dans le fleuve et ceux apportés par voie atmosphérique permettront d'évaluer les risques de contamination du milieu et de la chaîne alimentaire.

2. LES VOIES DE TRANSFERT

En fonction des hypothèses de travail retenues, l'utilisation de l'eau et les cultures irriguées constitueront les paramètres de base des voies de transfert critiques. Dans cet ordre d'idée nous retiendrons à la fois les utilisations d'eaux de surface et les utilisations d'eaux de la nappe phréatique.

2.1. Transfert par les eaux de surface

Les irrigations à l'aide des eaux de surface issues du fleuve sont susceptibles d'apporter les radionucléides sous forme colloïdale qui sont filtrés par le sol et sous forme dissoute (organique ou minérale). Dans notre schéma de transfert le débit global des canaux sera de 20 m^3/s. Selon la distance du point d'irrigation par rapport au point de rejet, les sols ne reçoivent que des formes dissoutes. Du point de vue du transfert il s'agit de déterminer la part absorbée par le sol et la part susceptible de migrer vers la nappe. Dans la première fraction il convient également d'évaluer la partie disponible pour les plantes.

2.2. Transfert par les eaux de nappe

L'origine des radionucléides des eaux de la nappe peut être soit le fleuve directement, soit les eaux d'infiltration du sol. Ces eaux ne renferment généralement que des formes dissoutes. Cependant, la charge saline de ces eaux est variable dans l'espace, et dans certains cas sa composition chimique est liée étroitement à des infiltrations de polluants divers: nitrate provenant de la fertilisation, composés organiques provenant des traitements agricoles, etc. Par ailleurs les utilisations des eaux de nappe peuvent être qualifiées de «sauvages» car aucun contrôle, ni sur la qualité de l'eau, ni sur la quantité d'eau puisée, n'est possible. Une infiltration accidentelle de radionucléides dans la nappe constitue de ce fait un des risques les plus importants de contamination du milieu en raison de la multiplicité d'utilisation de l'eau à des fins agricoles, industrielles ou domestiques.

2.3. Transfert par la chaîne alimentaire

L'utilisation des systèmes d'irrigation en bordure des fleuves fait évoluer dans une grande mesure le mode de culture. Les cultures traditionnelles et de type extensif sont remplacées par des cultures industrielles: maïs, céréales, etc., et à fort rendement. L'utilisation des engrais chimiques est de nature à faire évoluer le complexe absorbant du sol et les absorptions de radionucléides par voie racinaire. L'irrigation par aspersion représente pour de nombreux radionucléides une nouvelle voie de transfert non négligeable. La présence, dans les eaux d'irrigation et les solutions de sol, d'agents complexants organiques augmente la mobilité de ces éléments dans le sol et les possibilités d'assimilation par les végétaux. Les rejets accidentels, soit de radionucléides, soit de polluants organiques, constituent un risque sérieux de contamination de la chaîne alimentaire.

2.4. Paramètres de transfert liés à la physico-chimie des sols

Les paramètres de transfert des radionucléides au niveau des sols sont fonction de la surface occupée par chaque type de sol, de la perméabilité, de la rétention en eau, du complexe absorbant, de la teneur en matière organique et en argile.

2.4.1. Surface occupée

La surface occupée par les différents groupes pédologiques de sol doit être calculée en fonction de la contamination soit par irrigation, soit par dépôts atmosphériques, soit par les deux simultanément. Pour les dépôts atmosphériques on tiendra compte de la rose des vents: fréquence directionnelle et vitesse. Ces diverses considérations permettent de faire une classification par type de sol et mode de contamination.

2.4.2. Perméabilité

Parmi les facteurs physiques du sol intervenant dans la contamination, la perméabilité constitue un paramètre important dans la répartition des radionucléides. A l'exception de quelques sols argileux et limoneux, l'ensemble des sols alluviaux est assez perméable; la vitesse de circulation de l'eau fluctue généralement entre $2 \cdot 10^{-3}$ et $2,5 \cdot 10^{-2}$ cm/s. Cette excellente perméabilité favorise d'abord le transfert des radionucléides au niveau du système racinaire des plantes, ensuite la migration dans la roche-mère (cailloutis sableux) puis dans la nappe phréatique qui circule dans les bancs sableux.

2.4.3. Rétention en eau

A côté de la perméabilité, la rétention en eau (ou réserve hydrique) représente un paramètre qui conditionne le transfert vers les végétaux et la chaîne alimentaire ainsi que vers la nappe phréatique. Cette réserve hydrique est la quantité d'eau réellement disponible pour les plantes. Sa valeur fluctue pour les types de sols considérés entre 1 et 20%; les valeurs moyennes pour les sols alluviaux sont de 6 à 10% et pour les sols bruns de 8 à 16%. Pour de nombreux sols alluviaux sableux cette réserve d'eau est donc assez faible et les risques de lessivage sont certains, d'autant plus que l'irrigation à des doses de 10 à 50 mm de hauteur d'eau est surtout pratiquée sur sols peu profonds et caillouteux.

2.4.4. Nature du complexe absorbant

Dans les sols considérés, le complexe absorbant est généralement saturé en bases à l'exception des sols bruns acides. Le calcium et le magnésium sont les deux éléments prépondérants. La capacité d'échange de ces sols varie essentiellement en fonction de la teneur en argile car sur sols cultivés la teneur en matière organique oscille en général autour de 2,5%. Les valeurs du complexe absorbant des sols utilisés fluctuent entre 8 et 25 méq/100 g. Il est à signaler que ces valeurs tombent à 2 ou 3 méq/100 g dans les horizons sableux. L'efficacité de la rétention des radionucléides par le complexe absorbant est partiellement annihilée par la forte perméabilité et la faible capacité de rétention en eau.

2.4.5. Teneur en matière organique et en argile

Comme nous l'avons signalé précédemment, la teneur en matière organique des sols cultivés oscille autour de 2,5%. Par contre, la teneur en argile varie dans de plus grandes proportions. Dans la catégorie des sols bruns calciques, par exemple, on peut distinguer trois classes en fonction du taux d'argile: inférieur à 15%; compris entre 15 et 25%; supérieur à 25%.

Dans le groupe des sols alluviaux, deux classes peuvent être distinguées: les sols dont le taux d'argile est inférieur à 10%, les sols dont le taux d'argile est supérieur à 10%.

L'abondance d'argile conditionne la capacité de rétention en eau et la capacité d'échange; de ce fait, la valeur de 20% d'argile peut être retenue sur le plan de la rétention radioactive pour classer les sols:

- rétention bonne à moyenne si le taux est supérieur à 20%,
- rétention moyenne à médiocre si le taux est inférieur à 20%.

3. LES MECANISMES DE TRANSFERT

Les mécanismes de transfert des radionucléides du point de rejet jusqu'à la nappe en passant par les sols sont multiples. Nous essaierons dans cette étude de cerner les plus importants.

Le mécanisme de base est constitué par la réaction de l'eau du fleuve sur l'effluent. Le second est constitué par l'influence de la qualité de l'eau sur la migration des radionucléides dans les sols. Le troisième englobe l'interaction des différents types de sols sur la répartition des radionucléides dans le sol. Le quatrième enfin concerne l'ensemble des formes chimiques susceptibles de contaminer la nappe phréatique, donc l'évolution des radionucléides dans les sols.

3.1. Réaction de l'eau du fleuve et des eaux d'irrigation avec l'effluent

Les mécanismes de transfert dans les eaux ont été étudiés à l'aide de techniques originales qui consistent à voir sur des supports variés la réaction des eaux et des radioéléments. Les supports utilisés sont de trois types: le papier, le gel de silice, le gel de silice imprégné de résines échangeuses d'ions. Les solvants employés sont constitués par les différentes eaux susceptibles de transporter les radionucléides dans les sols: eaux brutes des fleuves, eaux d'irrigation, nappe et canaux.

Sur papier, par exemple, nous pouvons constater que la teneur en sels minéraux et en composés organiques est responsable du déplacement plus ou moins accentué des radionucléides.

La figure 1 montre l'effet obtenu sur un ensemble de cinq radionucléides: ^{137}Cs, ^{22}Na, ^{65}Zn, ^{59}Fe, ^{141}Ce et sept eaux de diverses origines: eaux brutes des fleuves, canaux d'irrigation, eau de la nappe phréatique.

La figure 2 montre l'effet obtenu sur gel de silice avec des eaux de composition organo-minérale variée.

Nous constatons sans équivoque que la qualité actuelle des eaux brutes des fleuves constitue un facteur primordial dans la réaction des radioéléments. Selon sa charge organo-minérale, cette eau est susceptible de mobiliser la presque totalité des radioéléments. Sur gel de silice la différence entre les diverses eaux est encore plus nette que sur papier. En présence d'une imprégnation de résine, par exemple, nous observons indubitablement la présence de complexes qui se retrouveront dans l'eau d'irrigation et joueront au niveau du sol un rôle important, pour la mise à la disposition des plantes des radionucléides rejetés dans un tel système comme pour la migration de ces mêmes composés vers la nappe phréatique.

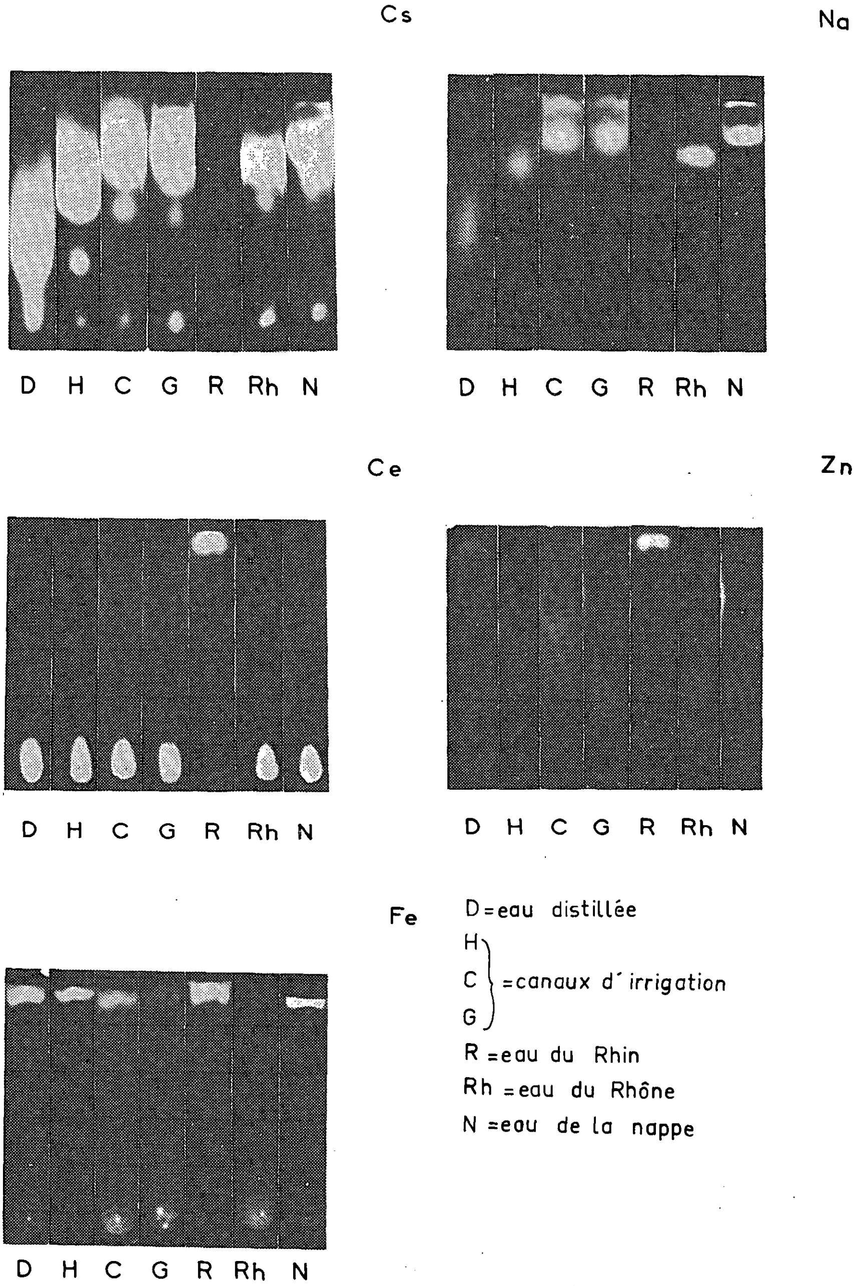

FIG. 1. Chromatographie sur papier des différents radionucléides avec les eaux comme solvant.

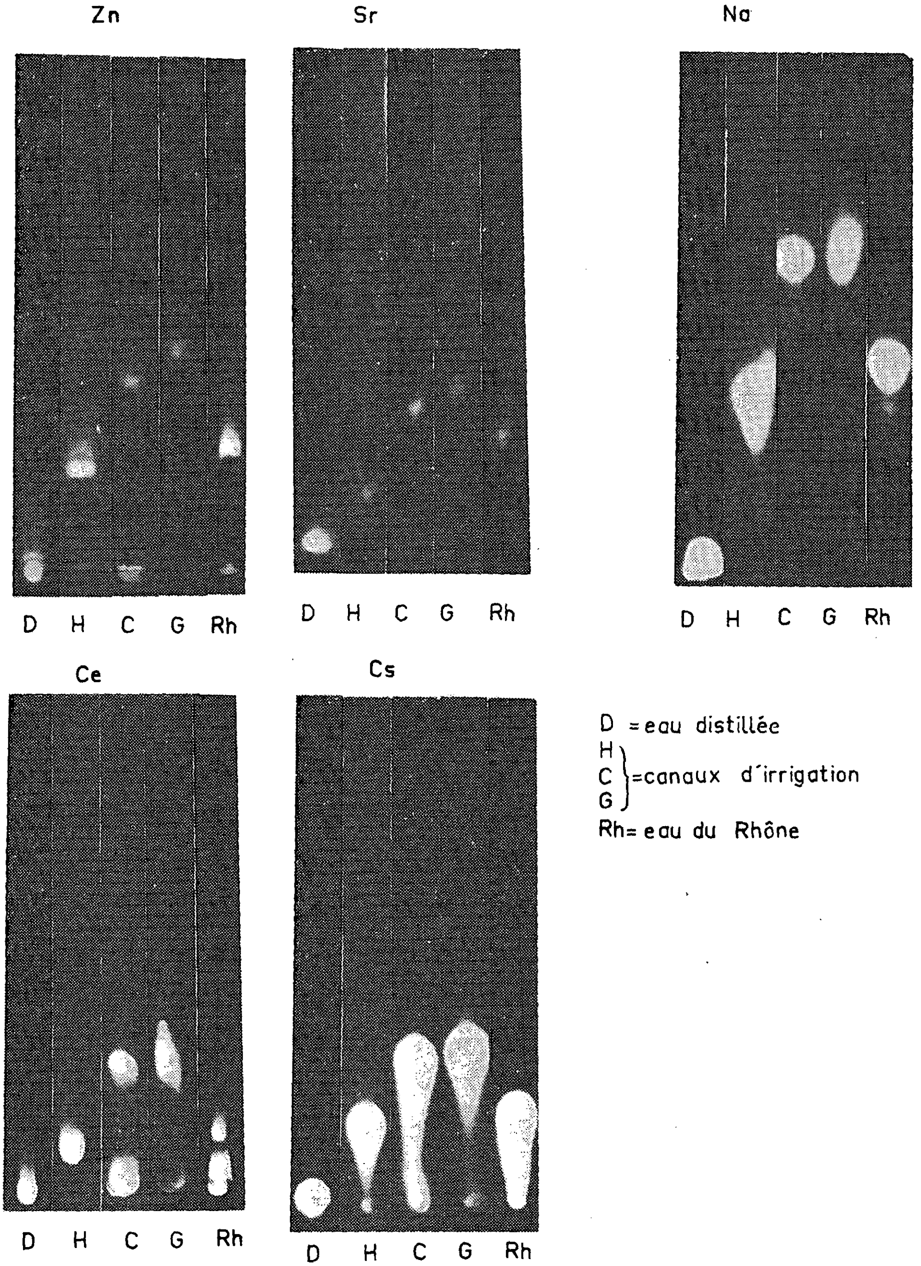

FIG. 2. Chromatographie sur gel de silice des différents radionucléides avec les eaux comme solvant.

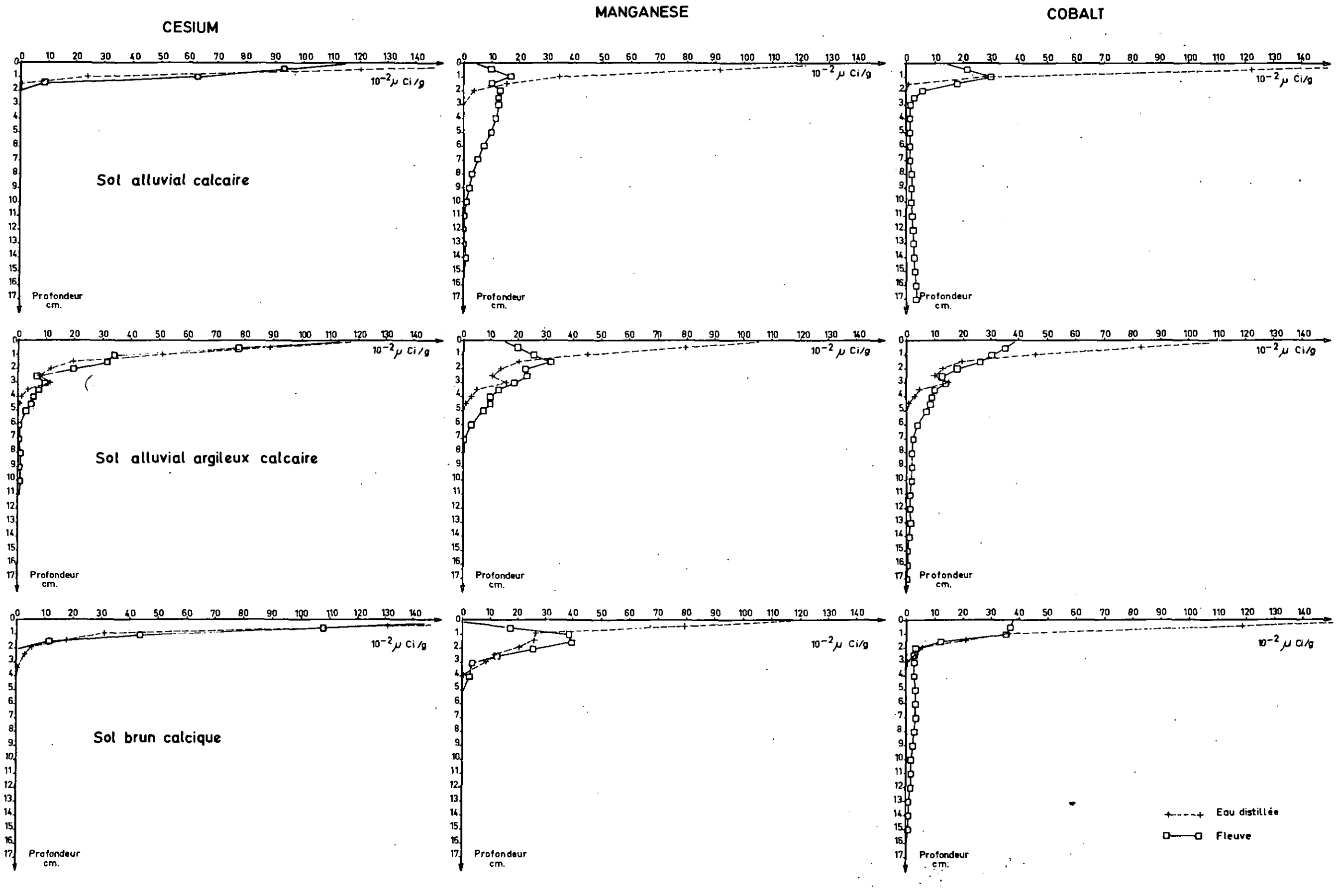

FIG. 3. Répartition du césium, du manganèse et du cobalt sous l'effet de différentes eaux sur trois types de sol.

3.2. Influence de la qualité de l'eau sur la migration des radionucléides dans les sols

Au niveau des sols, l'interaction radionucléides-eau se retrouve et nous remarquons, sur la figure 3, que la pénétration de ceux-ci est guidée par la qualité de l'eau. La différence de migration entre l'eau distillée et l'eau brute du fleuve est importante pour les éléments qui peuvent former des complexes avec les composés organiques. A cet égard, il faut faire remarquer que l'ensemble des produits d'activation est concerné par la présence d'une pollution organique dans l'eau brute des fleuves. Il apparaît que les mécanismes de transfert des radionucléides dans les sols dépendent, pour une grande part, de la qualité et de la composition organo-minérale des eaux qui véhiculent les radionucléides.

3.3. Evolution des radionucléides dans les sols

Le transfert des radionucléides dans les sols est lié à la présence des formes hydrosolubles; apportés par les eaux d'irrigation, une forte proportion en reste hydrosoluble et est capable de migrer en profondeur. A cet égard, le type de sol, et en particulier le pH de celui-ci, joue un rôle essentiel. Les complexes sont d'autant plus solubles et plus stables que le pH du sol est plus acide et que celui-ci ne renferme pas de calcaire. Ces complexes stables sont mobiles même sous l'action de l'eau de pluie. Les figures 4 et 5 illustrent les possibilités de migration en fonction du type de sol. Si nous examinons de plus près cette répartition de formes hydrosolubles dans le profil (fig. 6) nous remarquons qu'il subsiste plusieurs types de complexes stables. Ces composés peuvent atteindre rapidement la nappe phréatique et être repris lors de l'utilisation de l'eau de la nappe.

3.4. Conséquences pour le milieu

Les conséquences du rejet de radionucléides dans les environs immédiats d'une centrale nucléaire seront les suivantes:

— Risque de contamination de la surface du sol par irrigation et dépôts atmosphériques. La partie la plus touchée sera la zone où les deux modes de contamination se superposent.

— Contamination probable des eaux de la nappe phréatique par suite des interactions fleuve-nappe et des possibilités de migration rapide des radionucléides apportés par les diverses eaux d'irrigation sur les sols environnants.

— Risque de contamination des végétaux par voie foliaire (aspersion), et par voie racinaire (irrigation du sol avec migration en profondeur). Un cycle biologique complet risque de s'établir entre le sol, la nappe et les résidus végétaux.

— Contamination progressive et par vagues des eaux du fleuve sous l'influence des rejets industriels situés en amont. Il s'ensuivra une dissémination importante et des incidences pour l'aval du fait de la mobilité exceptionnelle des radionucléides sous l'influence de ces mêmes rejets industriels, et cela pose le problème du traitement de l'eau en amont pour éviter cette mobilité et en aval pour arrêter ces complexes au niveau des stations de traitement des eaux de boisson et des eaux industrielles.

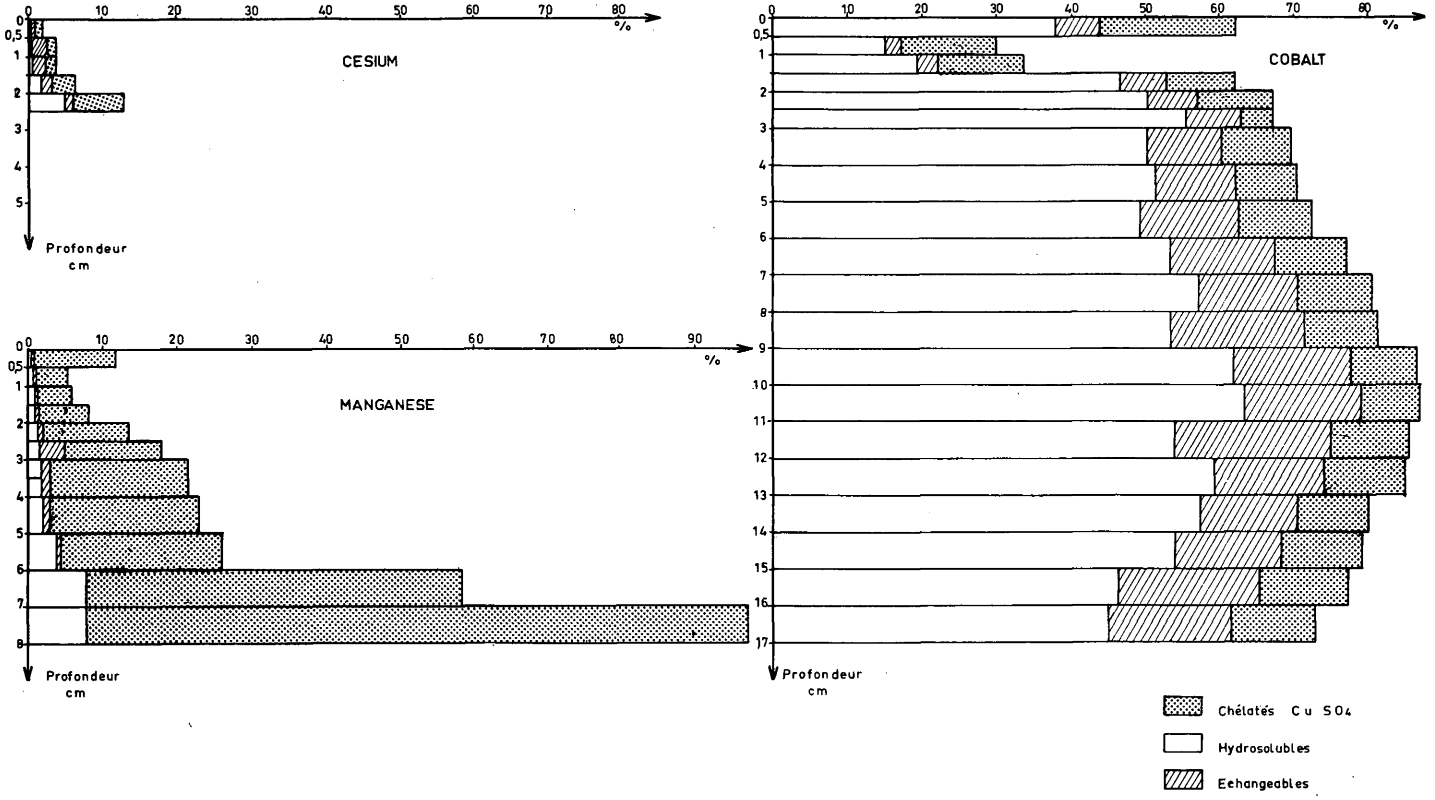

FIG. 4. Différentes formes chimiques du césium, du manganèse et du cobalt dans un sol brun calcique (eau du fleuve).

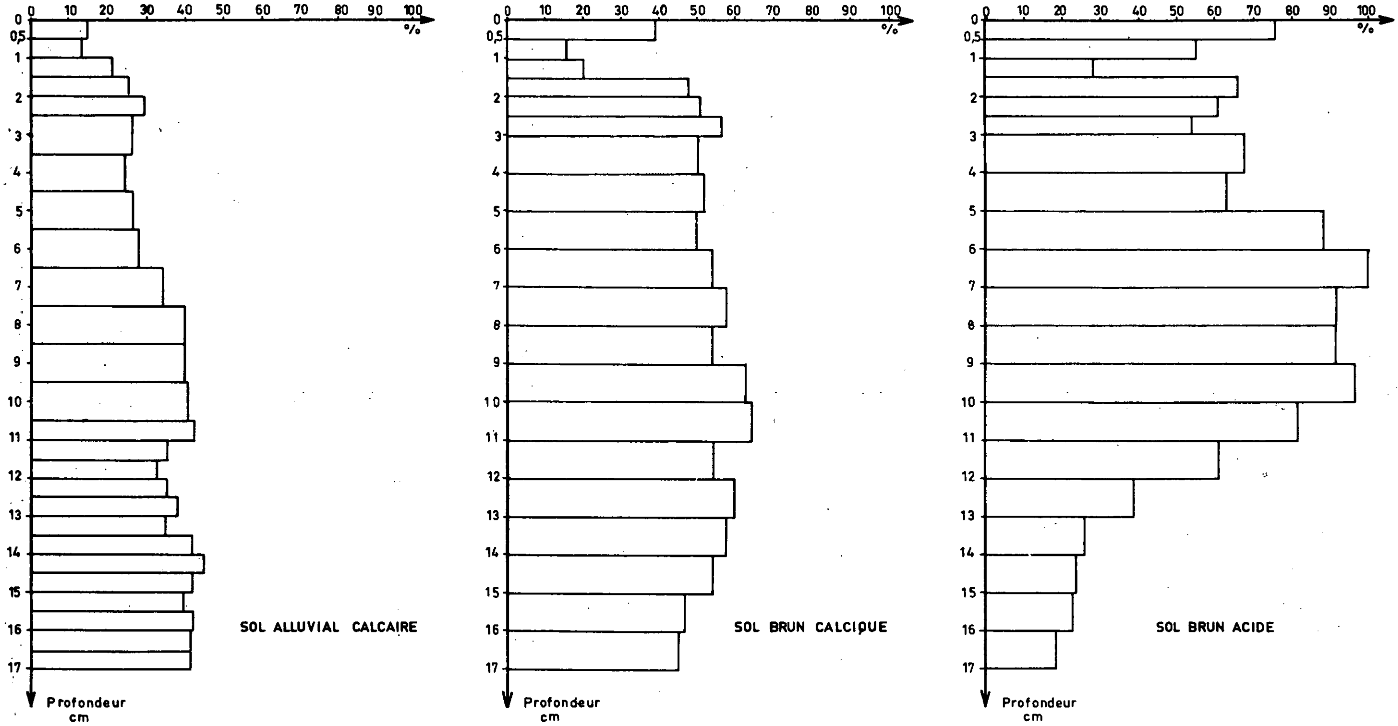

FIG. 5. Variation du pourcentage de formes hydrosolubles du cobalt dans trois sols typiques.

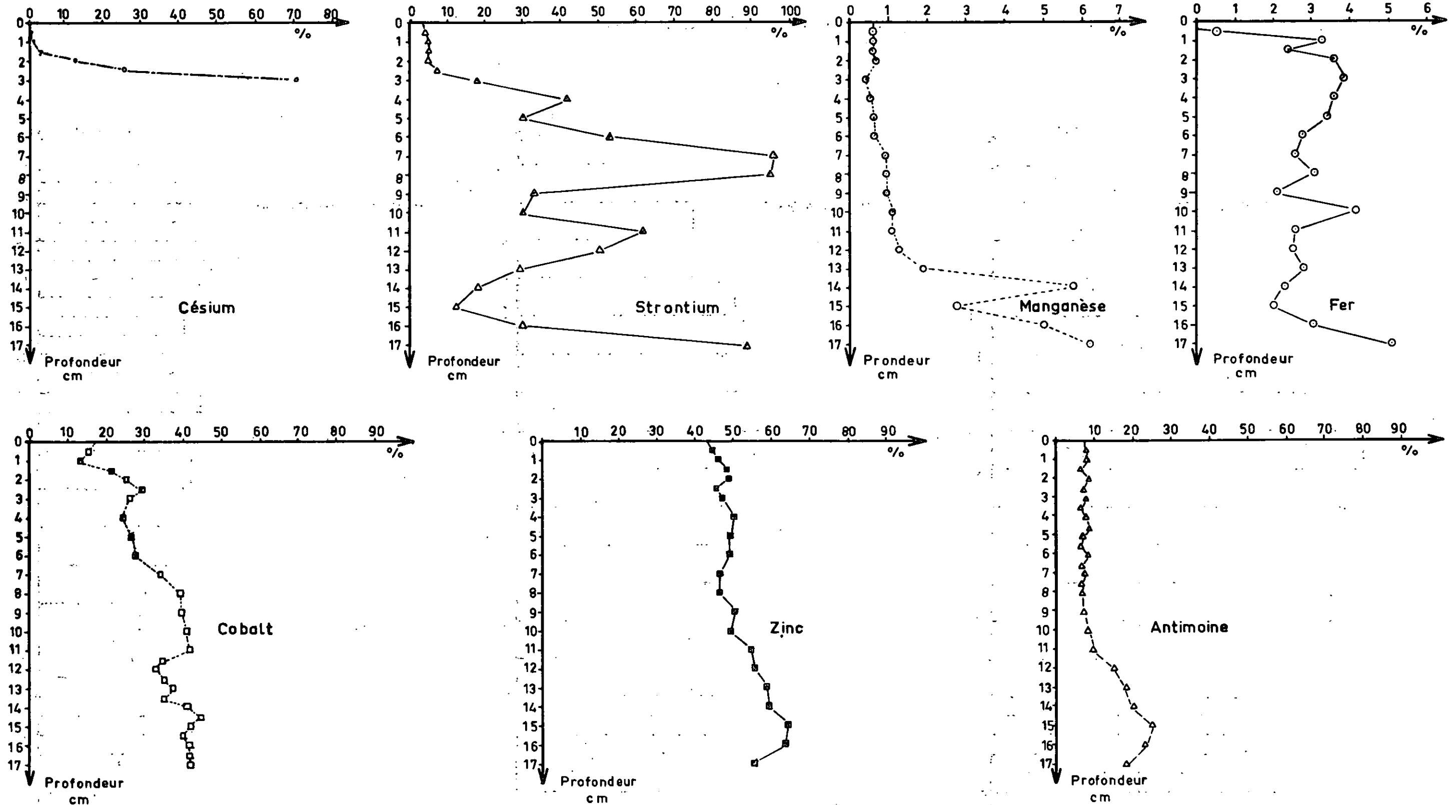

FIG.6. Formes hydrosolubles sur sol alluvial calcaire après migration avec l'eau du fleuve.

CONCLUSIONS

L'étude des mécanismes de transfert des radionucléides dans le système fleuve—sol irrigué—nappe permet de mettre en évidence les principales incidences des rejets radioactifs sur le milieu. L'évolution de la contamination dans le milieu environnant est liée à trois causes principales:

- la pollution importante des eaux des fleuves et en particulier en composés organiques
- la présence non loin des centrales nucléaires de points de rejets industriels où sont rejetés des composés peu biodégradables
- les prises d'eau pour l'irrigation situées en aval du point de rejet.

Sous l'effet de ces diverses causes, les techniques expérimentales utilisées [1, 2] font ressortir que la contamination des sols et de la nappe est fonction de la qualité des eaux car celle-ci intervient pour une large part dans la mobilité des radionucléides.

L'ensemble des études de mécanismes de transfert fait également ressortir la notion primordiale de site et montre que l'évolution des radionucléides dans le milieu est conditionnée par les facteurs spécifiques: pollution organo-minérale du fleuve, types de sols et types de cultures.

Pour les risques radioactifs, le facteur critique d'un site qui doit conditionner les rejets d'une centrale, et peut-être limitera son extension, est constitué dans le temps par la charge polluante des eaux du fleuve dans lequel sont effectués les rejets. Cette charge polluante favorise la mobilité des radionucléides vers les consommateurs d'eau situés en aval, accélère le transfert de la contamination dans les sols et, par voie de conséquence, augmente la diffusion vers les cultures et la nappe [3].

Le risque de pollution thermique, en dehors de ses effets propres sur le milieu aquatique, se superposera au risque radioactif en modifiant l'amplitude des phénomènes de radiopollution, à savoir surtout la mobilité et le métabolisme des radionucléides.

REMERCIEMENTS

Les auteurs désirent remercier ici Madame M. Favre et Messieurs P. Gille, J.M. Hetier et J.J. Wolter de la collaboration qu'ils leur ont apportée au cours de cette étude.

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DISCUSSION

K. B. MISTRY: Were there any differences in the predominant clay mineral type between the soils used in your experiments? I ask this question in relation to your observations on the behaviour of radiocaesium.

A. SAAS: We have in our soils several types of clay and various mixing proportions. In the calcic brown soil, for example, we have an illite-chlorite mixture.

P. G. MAYER: Were the soils saturated with water for purposes of the experiments?

A. SAAS: In our experimental procedure the soils are moistened to capacity in the field for 24 hours and then allowed to revert to the moisture content required for the experiment. We worked with several different moisture content values. This parameter affects in particular the distribution of the radionuclides but has little influence on the chemical forms.

R. FUKAI: With reference to your use of the term "hydrosoluble", have you any idea as to what chemical forms of elements are present in "hydrosoluble" form? Are they mainly water-soluble organic compounds or ions of the elements concerned?

A. SAAS: By "hydrosoluble" we mean the totality of the organic and mineral components of a soil which are soluble in water. The water/soil ratio is 2/1, the composition of the solution varying from one soil to another. The principal mineral components are K^+, N^+, Ca^{++}, Cl^-, $SO_4^=$ and HCO_3^-. The organic compounds present are molecules of low molecular weight; the carbon concentrations vary from 5 to 10 mg/litre.

H. WIJKER: Do the enhanced metabolism and mobility as a consequence of higher temperature always constitute an extra risk, or can they be beneficial in some circumstances? If the latter is the case, it might be preferable to speak of thermal 'changes' instead of thermal 'pollution'.

A. SAAS: The action of temperature may be described as follows:

(1) Increased solubilization of certain mineral and organic compounds; this has a tendency to increase leaching into the ground water;
(2) An effect on the oxidation-reduction system of the soil; this brings about a more active biodegradation of the complexes and some degree of immobilization of the minerals.

L. EDOUARD: You say in your conclusion that the nature of the receiving medium for the liquid effluents might be such as to require the abandonment of plans for expansion of a nuclear power station. This prompts me to put two questions, albeit slightly outside the field of our present discussions:

(1) In view of the regulations governing releases from nuclear power stations, is abandonment of such plans for expansion really conceivable?
(2) Likewise in view of the release regulations, and in the public interest, would it not be preferable to devote available financial resources to purifying domestic and industrial wastes, rather than try to chop off a few more milli- or microcuries from radioactive wastes?

A. SAAS: The purpose of our study was basically to define the transfer mechanisms of radionuclides in soils. We have no authority to take decisions regarding extensions – or lack of extensions – to power stations. However, in the context of this study the pollution load of rivers emerges as a limiting factor as far as radioactive releases are concerned.

Consequently, we make the point in our paper that purification of the water upstream can enhance the safety of radioactive disposals. In the case of certain nuclides, such as ^{22}Na and ^{131}I, the quality of the water into which the releases are effected and the purification of industrial and domestic wastes should be taken into account on both the ecological and the economic levels.

R. J. ROUX: You have described the significance of pollution loads deriving from sources outside the nuclear power station. However, there will be organic components present in the station's own wastes (overall wastes, that is, not just radioactive effluents), and perhaps you could indicate to us, on the basis of your researches, what the most important of these are.

A. SAAS: Among the organic components capable of forming complexes I would say that carboxylic compounds are the most important, followed by phenolic compounds and amines.

BEHAVIOUR OF RADIONUCLIDES IN TERRESTRIAL SYSTEMS

(Session IV)

Chairman:

E. KUNZ (CSSR)

AGRICULTURAL ASPECTS OF RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT BY THE NUCLEAR POWER INDUSTRY

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Abstract

AGRICULTURAL ASPECTS OF RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT BY THE NUCLEAR POWER INDUSTRY.

The significance of contamination of agricultural produce by radioactivity released into the environment by nuclear industry is considered. Hitherto, industrial uses of nuclear energy have exposed the population to only very small additional doses of radiation and the contribution from agricultural produce has been trivial. Under normal conditions this situation will continue, provided that the present stringent controls are maintained. However, despite all precautions the possibility of accidental releases of radioactivity cannot be entirely eliminated. Assessments of the agricultural implications of such releases from current installations have identified ^{131}I as the nuclide liable to cause the highest radiation doses because of its abundance and volatility, its rapid transfer to milk, and accumulation in the small thyroid of infants. ^{137}Cs, ^{90}Sr and ^{89}Sr also merit consideration but would usually signify lower radiation doses. The relative importance of these nuclides may vary with new types of reactors but there is no evidence that other nuclides will become of comparable significance.

Relationships between the deposition of these radionuclides on agricultural land and the contamination of diet are reviewed. Particular attention is given to the transfer of radioactivity to milk by cattle grazing contaminated pastures as this pathway can lead to the highest radiation doses. Where milk is not produced or when cattle are not at pasture the doses for a given release would be much lower. Most of the total dietary contamination will occur relatively soon after deposition. The short half-lives of ^{131}I and ^{89}Sr preclude long-term contamination, but even with ^{90}Sr and ^{137}Cs analysis of survey data shows that most of the dose commitment will be incurred within the first 2 years. Moreover, it is most unlikely that the amount of ^{90}Sr in the soil would ever be such that agricultural produce was severely affected.

In planning safeguards for the population after a reactor accident, only ^{131}I warrants consideration as a dietary contaminant. Even for this nuclide, inhalation is likely to be the dominant mode of exposure for people living close to the source and this will become relatively more important if reactors are sited close to towns. Remedial measures must take both modes of exposure into account.

INTRODUCTION

In the early years of the nuclear industry it was general policy to locate nuclear reactors and other installations remote from centres of population, and hence, usually in agricultural areas. It was natural, therefore, that considerable attention should be given to the possible contamination of foodstuffs produced near these sites, especially in a period when the contamination of diet with world-wide fallout from nuclear tests was a matter of general concern. The significance of environmental contamination with radioactivity from the nuclear power industry, however, lies in the total additional doses of radiation which the population receives both from the consumption of contaminated foods and from external sources. These doses have been reviewed recently [1, 2] in relation to natural background radiation and other sources of ionizing radiation to which man is exposed and

TABLE I. AVERAGE ANNUAL DOSES FROM NATURAL BACKGROUND AND WORLD-WIDE AVERAGE DOSE COMMITMENTS FROM MAN-MADE ENVIRONMENTAL RADIATION ESTIMATED BY UNSCEAR [1]

	Annual doses from natural background ($mrad \cdot y^{-1}$)	Dose commitments (mrad)		
		Atmospheric tests before 1971	Per year of electrical power production	
			1970	2000
Gonads				
External	72	84	0.00045	0.05
Internal	21	35	0.00047	0.16
Total	93	120	0.0009	0.2
Bone marrow				
External	72	84	0.00045	0.05
Internal	17	76	0.00047	0.16
Total	89	160	0.0009	0.2

it is clear that, under normal operating conditions, the nuclear industry is responsible for only a very small increment in the dose received by the world population (Table I). In the United States it was estimated [2] that in 1970 the average whole-body dose to the population from the nuclear power programme represented less than 0.002% of the total from all sources, natural background and medical exposure being responsible for about 96%; in the future, the developing nuclear power industry was expected to contribute a population dose of less than 1% of natural background.

In assessing the possible effects on agriculture of the nuclear power industry, two situations require consideration.

1. Normal operating conditions when discharges of radioactivity are subject to control.
2. Emergency conditions after an accident in which large quantities of radioactivity may be released.

For either situation, widespread contamination will occur only after discharges to atmosphere. Iodine-131, caesium-137, strontium-89 and strontium-90 have long been recognized as the nuclides potentially able to cause the highest radiation doses to man through contamination of diet and, although new information on the biological behaviour and radiation dosimetry of these nuclides has modified views on their relative importance as environmental contaminants [1], there is no evidence that other nuclides are likely to become of comparable significance.

THE SITUATION UNDER NORMAL CONDITIONS

Reference has already been made to the small contribution which the normal operations of the nuclear power industry make to the total radiation doses received by the world population. It should be emphasized, moreover, that this exposure to radiation is due almost entirely to external sources or to internal doses attributable to discharges into the aqueous environment; no instance was cited of significant doses to the population arising from the consumption of agricultural produce.

This is illustrated by the situation in the United Kingdom where analysis of critical pathways has shown that the radiation exposure of the population from discharges of radioactivity by nuclear establishments is due primarily either to external sources or to consumption of aquatic foods [3]. It has been the general experience that the concentrations of strontium-90 and caesium-137 in milk produced in the neighbourhood of nuclear installations lie within the range to be expected from world-wide fallout [4, 5]. Under normal conditions no iodine-131 is detected in milk produced near nuclear reactors. Even at sites where fuel is reprocessed and the nuclide is known to be released, iodine-131 is only occasionally detected in milk and the average concentrations are low. At Windscale the concentration was less than 15 pCi litre^{-1} in all samples analysed in 1970 [6], while at the Dounreay Experimental Reactor Establishment iodine-131 was present above the detectable level of 20 pCi litre^{-1} in only 6 out of 200 samples analysed in 1971 and the average concentration during the year was less than 20.5 pCi litre^{-1} [5].

Provided that the present stringent controls are maintained it would seem unlikely that normal operations would lead to increased contamination of agricultural land in the future. In a review of the significance of releases of caesium-137 and iodine-131 from the Advanced Gas Cooled Reactors, Beattie [7] concludes that under normal conditions, the quantities of either nuclide discharged would be most unlikely to be of serious concern as gaseous effluent. He also points out that the external gamma dose from deposited caesium-137 places a more restrictive limit on the continuous discharge of caesium-137 than does the contamination of cows' milk.

Expansion of the nuclear industry will lead to the production of increased amounts of the long-lived iodine-129 (half-life = 1.7×10^7 years) which, in contrast to iodine-131, is not reduced in quantity by radioactive decay when irradiated fuel is 'cooled' before processing. However, with present purification plant, the proportion discharged to atmosphere can be reduced to 0.1% and an assessment [8] based on specific activity in the environment indicates that iodine-129 will not be a serious problem for a chemical plant serving a power programme up to 100,000 MW(e). Indeed, Bryant [9] has recently estimated that the dose rate to the thyroid glands of the critical group living near Windscale was only about 10^{-6} rad year^{-1} in 1972 and might rise to about 4×10^{-5} rad year^{-1} in the year 2000 as a result of discharges of iodine-129 to atmosphere. If fuel from fast-reactors were reprocessed after shorter periods of cooling than is now customary it would be necessary to improve the decontamination factor for short-lived nuclides of iodine and the proportion of iodine-129 released would also be lowered [10].

ACCIDENTAL RELEASES OF RADIOACTIVITY TO ATMOSPHERE

Although the design and construction of reactors provide a high degree of intrinsic safety, the possibility of accidents occurring cannot be entirely eliminated. Only major accidents in which there was a substantial release of radioactivity to atmosphere would be of agricultural significance. In minor accidents radioactivity would often be contained and there would be no contamination of farmland. Operational errors or malfunctions causing releases to waterways would seldom seriously affect agricultural produce and any contamination of drinking water for animals or of crops by irrigation could readily be controlled.

TABLE II. RELATIVE QUANTITIES OF VOLATILE OR SLIGHTLY VOLATILE FISSION PRODUCTS IN A POTENTIAL RELEASE FROM A NUCLEAR POWER REACTOR

Fuel rating, 20 MW·tonne^{-1}; irradiation time 1000 days.

Nuclide	Ci·MW^{-1}	Nuclide	Ci·MW^{-1}
^{131}I	3.0×10^4	^{137}Cs	3.3×10^3
^{132}I	3.8×10^4	^{90}Sr*	2.4×10
^{133}I	5.6×10^4	^{89}Sr*	2.4×10^2
^{134}I	6.7×10^4	^{106}Ru	1.6×10^4
^{135}I	5.2×10^4	^{103}Ru	4.1×10^4
^{132}Te	3.8×10^4		

*1% of inventory. (From Beattie and Bryant, [11])

Critical Nuclides

The fission products likely to be released from current types of reactors in an accident have been discussed by Beattie and Bryant [11] and the composition of a possible release is illustrated in Table II. Of the radioisotopes of iodine, only iodine-131 is likely to require consideration as a contaminant of diet. Other isotopes of iodine, though they could make a major contribution to the radiation dose due to inhalation, are not of importance in agriculture owing to their short half-lives. Caesium-137 is also relatively volatile and substantial quantities may be released. The isotopes of strontium, however, are of low volatility under most conditions and would tend to be retained. Other fission products with appreciable half-lives, such as the isotopes of ruthenium and the rare earths, do not readily pass through food chains and are of minor interest as dietary contaminants [12].

The relative amounts of fission products released may vary according to the nature of the accident and the type of reactor. For instance, it is possible that rapid depressurization of a High Temperature Reactor could lead to caesium-137 being released but no iodine-131 [7], and in sodium-cooled reactors iodine-131 would be efficiently retained by the liquid sodium [13]. The nature of the accident may also influence the physical or chemical form of the radionuclides released; discussion in the following sections relates to finely divided, soluble material but if it were in large particles or of low solubility its transfer to diet would be greatly reduced.

Contamination of Agricultural Produce Soon After an Accident

In discussing the agricultural effects of an accidental release of radioactivity from a nuclear reactor both the short- and long-term contamination of produce will be considered. In the acute situation soon after the accident, direct deposition on pastures and crops will lead to the highest levels of contamination, but for relatively short periods. At this stage uptake from the soil is of little importance but subsequently it is possible that the longer-lived nuclides may be absorbed by plants and cause lower levels of contamination over prolonged periods.

The potential radiation dose to the population from the consumption of contaminated produce soon after an accident will depend greatly on the predominant type of production in the area and the season of year. In countries where milk is an important item of diet, the pathway from pasture to cow to milk is most likely to be the critical one. Iodine, strontium and caesium are all rapidly transferred to milk which is usually consumed within a few days of production. Milk is also an important component of the diet of infants who are the critical group in the population and may depend entirely on local supplies. The contamination of milk would be greatest in summer when cattle are grazing or receiving fresh forage. If cattle were kept indoors and being provided with stored foodstuffs, the potential radiation doses to the population would be much reduced.

In areas where no milk is produced, green vegetables would probably be the main source of dietary contamination but the daily intake of radioactivity would be much smaller than that from milk produced on pastures receiving the same deposit per unit area. For example, the daily intake of radioactivity on leafy vegetables in the United Kingdom has been estimated to be equivalent to the deposition on only 10 cm^2 of ground [11]. Moreover, delay between deposition and harvest, and the normal processes of domestic preparation, would lead to considerable reductions in the level of contamination.

Information on the time-course of contamination of milk after an accidental release assists in predicting the potential exposure to radiation from dietary sources and provides a basis for deciding whether any countermeasures are needed. Calculations using data on retention of radioactivity by pastures and on the metabolism of the nuclides by cattle make it possible to predict the pattern of contamination in the early period after an accidental release [14]. Typical assumptions are that:

1. The deposit is in a freely available form
2. 25% is retained initially on the edible tissue of plants
3. In addition to radioactive decay, field loss causes the level of contamination to decrease by one-half every 14 days
4. The cows obtain all their diet by grazing.

These assumptions are likely to lead to a pessimistic estimate of the levels of contamination in milk since, for instance, even in the summer months, productive herds are given supplementary feeding stuffs which would be uncontaminated.

Iodine-131

A typical curve showing the variation in the contamination of milk with time after a single deposition of iodine-131 on pasture is given in Figure 1. Iodine-131 will be present in milk within a few hours of the pastures being contaminated but maximum concentrations will not be reached for 2-3 days. Thereafter the concentration decreases with an effective half-time of about 5 days owing to the combined effects of radioactive decay and field loss. The area beneath the curve provides an estimate of the total contamination

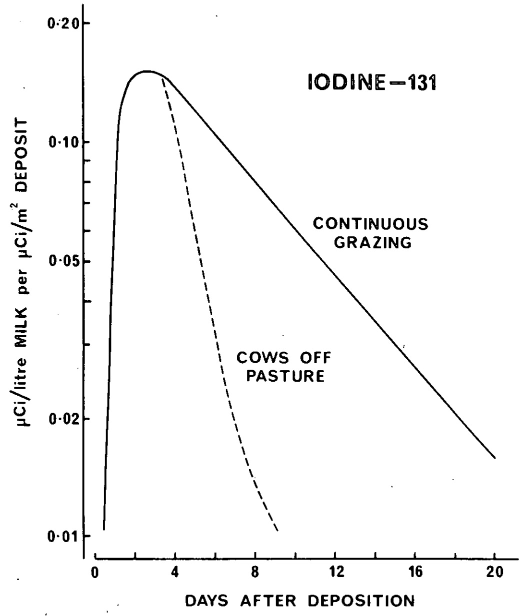

FIG. 1. Typical relationship between the contamination of milk with ^{131}I and time after the deposition of 1 μCi ^{131}I · m^{-2} on pasture (from Ref. [14]): (a) under conditions of continuous grazing; (b) if cows are taken off pasture after 4 days and given uncontaminated food.

of milk with iodine-131 and for planning purposes a value of 1.4 μCi days litre^{-1} per μCi m^{-2} deposit (Table III) is taken as representative for conditions in the United Kingdom. If cattle were moved to uncontaminated pasture or provided with stored food the concentration in milk would fall rapidly and the total contamination would be reduced.

The exact relationship between the quantity of radioactivity per unit area and the concentration in milk may vary widely depending on the quality of the pasture and the fraction of iodine transferred to milk [15, 16]. However, the shape of the curve tends to be more uniform and in an accident situation when the initial intake of iodine-131 by cattle may not be known at all precisely, the projected total contamination can be related to the peak level [17]. Lengemann [18-20] has extended this approach to allow predictions to be made from a measurement of the concentration at any interval after the contamination of pasture. Such relationships also make it possible to estimate the extent to which the radiation dose to the population might be reduced either by removing cattle from contaminated pastures and giving them uncontaminated food, or by controlling the use of milk from the contaminated area. For example, if cattle are taken off pastures 4 days after contamination has occurred, the total intake of iodine-131 with continued use of fresh milk would be about 30% of that if no protective action were taken compared with about 20% if the use of fresh milk were discontinued. It is evident that if the first alternative is practicable, a considerable reduction in the radiation dose to the population can be achieved with much less disruption of normal supplies.

TABLE III. THE INTEGRATED CONTAMINATION OF MILK FROM COWS GRAZING PASTURES CONTAMINATED IN THE SECOND HALF OF THE YEAR

Initial deposit 1 μCi·m^{-2}. For other assumptions see text.

	μCi·days·litre^{-1} per μCi·m^{-2}			
	Iodine-131	Caesium-137	Strontium-89	Strontium-90
Year of accident	1.4	10	0.4	0.5
Year after accident	-	2	-	0.6
Long-term from soil	-		-	0.4
Total	1.4	12	0.4	1.5

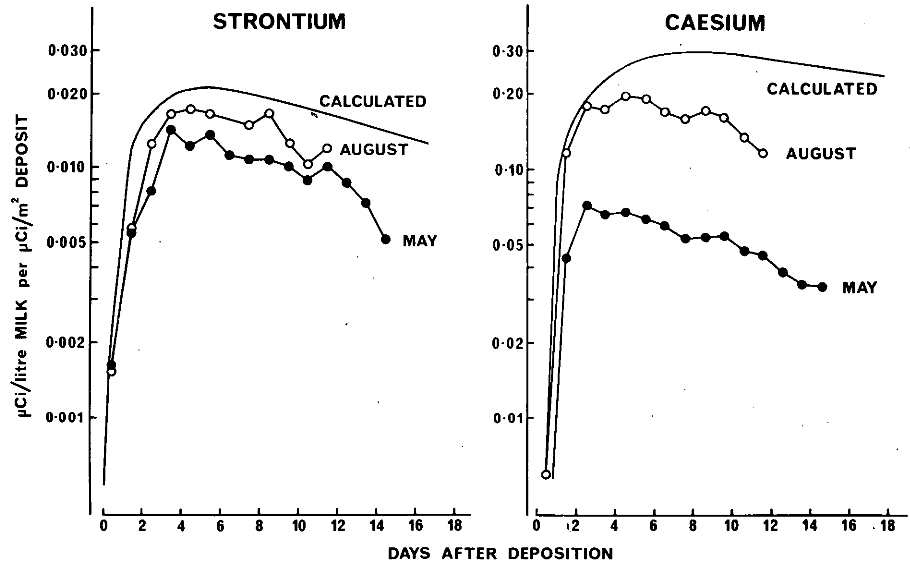

FIG.2. The contamination of milk with radioactive strontium and caesium at various intervals after the deposition of 1 μCi · m^{-2} on pastures. A comparison of results from an experiment in which ^{85}Sr and ^{134}Cs were sprayed onto pastures at two times of year [21] with calculated curves [14].

Strontium-89, strontium-90 and caesium-137

The contamination of milk with the isotopes of strontium and caesium-137 in the early period after an accident can be calculated in a manner comparable to that for iodine-131. The maximum concentrations after a single deposition on pasture would be reached in about 5-8 days and thereafter the rate of decrease would be determined largely by field-loss and the rate of biological turnover. In Figure 2 calculated curves showing the variation in concentration of strontium-90 and caesium-137 with time are compared with experimental results obtained by Van den Hoek et al [21] who allowed cattle to graze on pasture which had been contaminated by spraying with solutions containing radioactive strontium and caesium. The general forms of the observed and calculated curves agree quite well. However, while the experimental curves for caesium were similar in shape to the calculated curve, marked seasonal differences were observed, the concentration in milk in early summer for unit deposition on pasture being only about one-half that later in the year. For both strontium and caesium the observed levels were below those suggested by calculation which, as has been pointed out earlier, is based on assumptions likely to lead to conservative estimates. A recent analysis of American and British data [22] has shown that, despite considerable variations in the transfer coefficients of caesium-137 to milk in different animals, the form of the curves relating the concentration of caesium-137 in milk to time differed very little. Consequently the total acute contamination can be predicted with some confidence from measurements on milk in the early stages.

Estimates of the integrated contamination in milk in the early period after deposition of 1 μCi m^{-2} of the different nuclides, calculated on the basis of the assumptions listed earlier, are summarized in Table III.

Long-term Contamination of Diet

The short half-lives of iodine-131 and strontium-89 preclude any prolonged contamination of diet due to uptake by plants from the soil or to the use of stored animal feeding stuffs which were exposed to radioactivity before harvest. However, the contribution from these sources must be taken into account in assessing the total radiation dose commitments from strontium-90 or caesium-137. In the absence of direct experience of the consequences of a single heavy contamination of farmland, analysis of data from surveys of world-wide fallout provides the best basis for estimating the long-term consequences for these nuclides.

Strontium-90 in milk

The development of relationships between the deposition of strontium-90 and the contamination of milk in the United Kingdom has been reviewed recently by Bartlett, Russell and Jenkins [23]. It was recognized at an early stage [24] that direct contamination of pastures by recently deposited strontium-90 made an important contribution to the concentration in milk, especially when the rate of fallout was high, and that only part was attributable to indirect contamination due to entry of the nuclide into plants from the soil. Later it was found that, under United Kingdom conditions, the deposit in the latter half of the previous year also had a major influence on the contamination of milk, the estimated contribution from the cumulative deposit in soil being reduced still further [25]. However, up to 1966 the dominant effect of the recent fallout made it impossible to assess quantitatively the extent to which the availability to plants of the cumulative deposit in the soil changed with time. The subsequent relatively low annual deposition enabled an estimate of this to be made on the assumption that the availability decreases exponentially with time [26]. The equation used was of the form:

$$C_n = p_1F_n + p_2F_{n-1,b} + p_3[F_{n-1,a}\exp(-1.25k) + F_{n-2}\exp(-2k) + F_{n-3}\exp(-3k) + \ldots]$$

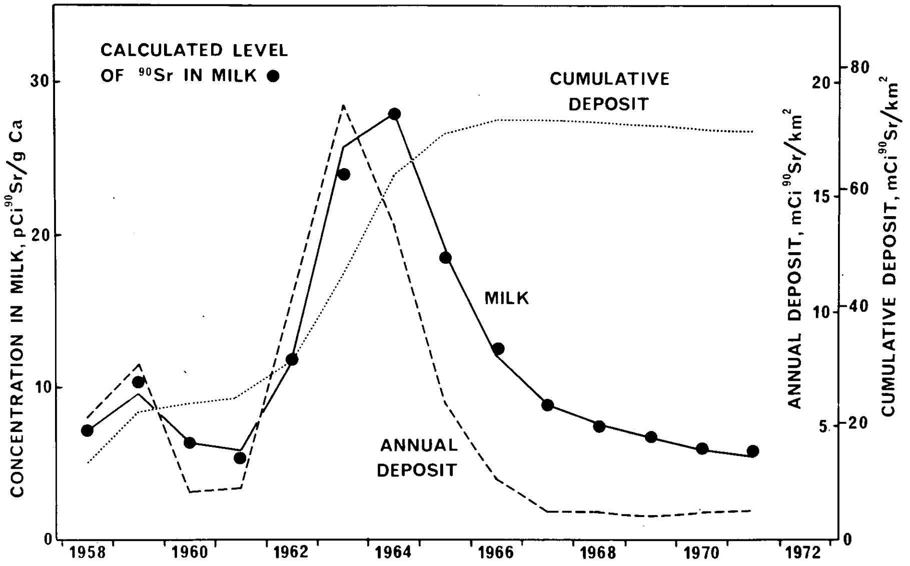

FIG.3. A comparison of observed and calculated average ratios of ^{90}Sr to calcium in milk in the United Kingdom, 1958-1971. The results of fall-out measurements used in the calculation are also shown. From Ref. [23].

where C_n is the annual ratio ^{90}Sr/Ca in milk (pCi ^{90}Sr (g Ca)$^{-1}$) in year n,

F_n, F_{n-1}, F_{n-2}, ... are the deposits of strontium-90 in year n and each previous year, corrected for decay to mid-point of year n (mCi km^{-2}),

subscripts a and b denote the first and second halves of year n-1,

p_1, p_2 and p_3 are proportionality factors,

exp(-k) gives the annual decrease in the availability to plants of strontium-90 in the soil, after allowance for radioactive decay.

Analysis of average survey results for the whole of the United Kingdom up to 1969 gave the following values for the constants in the equation: $p_1 = 0.70$, $p_2 = 1.41$, $p_3 = 0.20$ and $\exp(-k) = 0.86$. Figure 3 shows that there is close agreement between the observed and calculated ratios of strontium-90 to calcium in milk, not only over the period for which results were used in the calculation, but also in 1970 and 1971. Between 1958 and 1971 the maximum discrepancy was 7%. Values of the proportionality factors will vary for different localities but it is unlikely that even in poor agricultural conditions they will exceed the average by a factor of more than about 4-5 [27, 28]. The value for exp(-k) indicates an annual decrease of 14% in the availability of the strontium-90, in addition to the loss by radioactive decay (about 2%). This differs little from the estimate by van der Stricht et al [29] of 13% decrease annually in the environs of Ispra and an estimate of 11% in New York State [30].

From the equation it can be calculated that the total contamination of milk after fallout deposition of 1 mCi ^{90}Sr km^{-2} is 2.3 pCi ^{90}Sr y (g Ca)$^{-1}$ which is less than one-half that estimated from the original two-term equation. The reduction is due mainly to the lower estimate of the contribution from the cumulative deposit in the soil which has decreased from 84% of the total to only 47%.

The equation makes it possible to estimate the contamination of milk with strontium-90 in the years following an accidental release from a nuclear reactor. If it is assumed that deposition of 1 μCi ^{90}Sr m^{-2} occurs in the second half of the year, the average concentration in milk in the following year would be about 1.4 nCi ^{90}Sr (g Ca)$^{-1}$. This is equivalent to about 0.6 μCi days litre^{-1} during the year, a value comparable to the estimate of 0.5 μCi days litre^{-1} in the period soon after deposition occurred (Table III). In the second year after deposition the average contamination, due to uptake from the soil, would be only about one-tenth of this and decline subsequently by 16% each year, the integrated total from the soil amounting to about 0.4 μCi days litre^{-1}.

The overall total of 1.5 μCi days litre^{-1} (Table III) is likely to be an over-estimate of the actual contamination incurred since it was assumed that in the early phase the cattle obtained all their diet by grazing, and the use of the 'lag-rate' term in the equation implies that deposition over a limited area from an accidental release would have the same effect as widespread contamination from world-wide fallout. In fact, the use of uncontaminated feeding-stuffs from unaffected areas would lead to lower concentrations of strontium-90 in milk. Nevertheless it may be concluded that the highest doses from strontium-90 would be incurred within the first two years, and that subsequent contamination would be at much lower levels decreasing with a relatively short half-time of about four years.

Caesium-137 in milk

A calculation comparable to that in the previous section but based on the equation of Bartlett and Russell [25] for caesium-137 indicates that if 1 μCi ^{137}Cs m^{-2} were deposited in the second half of the year, the average concentration of caesium-137 in milk in the following year would not exceed 6 nCi litre^{-1}. This is equivalent to a total during the year of about 2 μCi days litre^{-1} compared to 10 μCi days litre^{-1} in the period immediately following the accident (Table III). Owing to the dominant influence of recent fallout on the contamination of milk with caesium-137, estimates of the small soil proportionality factor have failed to reach significance. However an average value of less than 0.1 appears probable [25, 28], so that the average concentration due to uptake from the soil would be less than 0.1 nCi litre^{-1}. In some areas, usually of low productivity, where the soil has a high content of organic matter and little clay, the contribution from the soil may be greater. However, even after heavy deposition on pastures, the period for which milk would be seriously contaminated is limited in most situations.

Other foodstuffs

Although there is less information on relationships between the deposition of fallout and the contamination of other foods than for milk, present estimates of uptake from the soil by different crops [28, 31] are adequate to establish that, even if the entire diet were produced locally, the overall ratio of strontium-90 to calcium in it would differ little from that in milk. If, as is much more probable, a high proportion of the diet were obtained from unaffected areas the ratio in the mixed diet would be considerably lower. For example, if locally produced vegetables were consumed but all other foods were uncontaminated, the ratio of strontium-90 to calcium in the diet (assumed to be the average for the United Kingdom) would be less than one-fifth of that in locally produced milk.

The small uptake of caesium-137 by plants from most soils precludes any prolonged contamination of diet. Meat from animals grazing contaminated pasture would be a possible source of caesium-137 in the first year after deposition, when the concentration in milk from the same area would provide a good guide to that in meat as the two are related [32, 33]. The concentration in meat could, however, be considerably reduced by feeding concentrates to the animals or by transferring them to uncontaminated pasture for a few weeks before slaughter.

Significance of Accidental Releases

The limited period for which deposition of radioactivity on farmland would cause dietary contamination is illustrated in Table III which summarizes the estimates of the total contamination of milk which could result from the deposition of 1 μCi m^{-2} of each of the nuclides. The entire intake of iodine-131 and strontium-89, and about 80% of that of caesium-137, would occur in the early period after an accident. About three-quarters of the total contamination of milk with strontium-90 would occur during the first two years and the remainder at much lower concentrations in subsequent years. No estimate is included for the contamination of milk with caesium-137 after the second year because, as explained earlier, there is as yet insufficient information to evaluate the long-term behaviour of the nuclide in the soil. However, the total contamination from this source would be small relative to that in the first two years and, moreover, external radiation from caesium-137 in the soil could expose the local population to higher radiation doses.

The figures in Table III can be used to estimate the radiation doses which might be received after the consumption of contaminated milk. As an example, doses which could arise from the deposition of 1 μCi m^{-2} have been calculated for an infant, six months old at the time of the accident, who is given milk from the contaminated area (Table IV). It has been assumed that

TABLE IV. DOSE COMMITMENTS TO AN INFANT CONSUMING MILK FROM COWS GRAZING CONTAMINATED PASTURE
For assumptions used in the calculation, see text.

Nuclide	Critical organ	Dose commitment (rads)	
		Initial deposit on pasture 1 μCi·m^{-2}	Nuclides deposited in same proportion as in assumed release (Table II) and dose from ^{131}I = 25 rads
Iodine-131	Thyroid	16	25
Caesium-137	Whole body	0.53	0.09
Strontium-89	Bone marrow	0.053	0.0007
Strontium-90	Bone marrow	1.5	0.0019

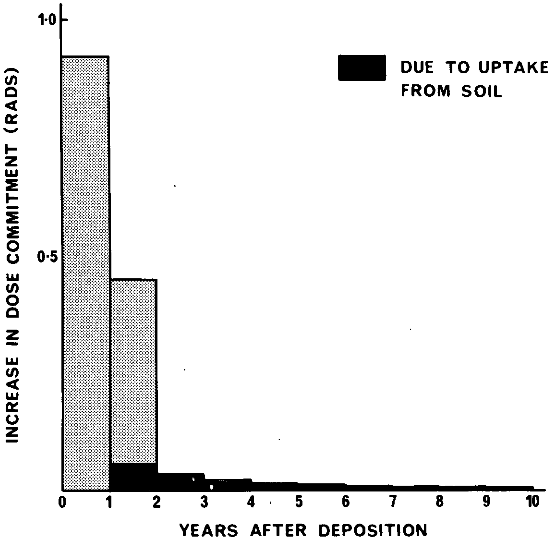

FIG.4. Estimated dose commitments from ^{90}Sr to the bone marrow of an infant, 6 months old at the time of an accident, who is given milk from an area where 1 μCi ^{90}Sr · m^{-2} is deposited on pasture. (See text for assumptions used in the calculation.)

the consumption of milk is 0.7 litre day^{-1} in the first year of life [34] and 0.5 litre day^{-1} thereafter [35]. The doses per μCi ingested have been taken as:- Iodine-131, 16 rad to the thyroid gland [34]; caesium-137, 0.066 rad to the whole body [34]; strontium-89, 0.19 rad to bone marrow [36]; strontium-90, 2.5 rad to bone marrow at age 6 months decreasing to about 0.5 rad in the adult [36]. The estimated dose commitments from strontium-90 to bone marrow in the years following an accident are shown in Figure 4. The high initial concentrations of strontium-90 in milk together with the greater radiation dose per μCi ingested in the first year of life lead to at least 60% of the total commitment being incurred in the first year. For reasons given earlier, the estimated dose commitment for the year following the accident is probably too high and it is more likely to lie between that shown by the shaded column and that due to the soil alone. The contribution attributable to uptake from the soil in subsequent years is relatively small.

Comparison of these estimates of dose per unit deposition with the relative quantities of the nuclides which may be deposited after an accident enables the relative significance of the nuclides as dietary contaminants to be assessed. Table IV shows the doses which could be caused by consumption of milk if the nuclides were deposited in the same ratio as that postulated by Beattie and Bryant from a long-irradiated fuel (Table II), the deposition of iodine-131 being such that it would result in a dose to the thyroid gland of 25 rad (the Emergency Reference Level currently in use in the United Kingdom [34]). It is evident that, compared with the dose from iodine-131, the doses from caesium-137 and the isotopes of strontium to the appropriate critical organs would be very small. The relative significance of the long-lived nuclides would be even lower for releases from a fuel which had been irradiated for a shorter period.

Thus, after a release of mixed fission products, iodine-131 is the nuclide which is most likely to require attention as a contaminant of diet. For other nuclides to reach levels in diet which would cause significant doses of radiation to the population, there would have to be a major accident involving large quantities of fuel and in these circumstances exposure of the population in other ways, particularly by inhalation, would be of dominant concern.

CONCLUSION

Provided that the present standards are maintained in the nuclear power industry the contamination of agricultural produce, under normal operating conditions, will make a negligible contribution to the radiation exposure of the population. Accidents to nuclear reactors are unlikely to cause major agricultural problems, and it is difficult to envisage a situation in which production would be severely affected for prolonged periods.

In planning safeguards for the population after an accident iodine-131 is the nuclide of principal concern as a dietary contaminant. Small accidents could give rise to local contamination of milk with iodine-131 for a relatively short period but widespread contamination of milk would occur only after a major release. Inhalation of radioactivity by the population living near the source would then be a more urgent problem and this will be of greater importance if reactors are sited close to towns. As the doses to the thyroid gland from inhaled and ingested iodine-131 are additive, any remedial measures to mitigate the effects of an accidental release must take both modes of exposure into account.

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DISCUSSION

F.O. HOFFMAN: In addition to caesium, which you have shown us, can you give us any idea about the variability of ^{131}I in milk under accidental release conditions?

R.S. BRUCE: After an accidental release of ^{131}I, quite wide variations are to be expected in the concentration in milk relative to the contamination per unit area of pasture, depending both on the type of pasture and the proportion of the ingested iodine which is transferred to milk. Some workers (but not all) have observed seasonal variation in the transfer factors, and the average factors found in experiments in the United States of America fairly consistently reached twice the level of those recorded in the United Kingdom, where Garner reported up to a tenfold variation between animals. I would not like to give a very definite estimate of the variability which might occur after an accident, but a difference of an order of magnitude may well be involved. Moreover, for a given air concentration, the deposition on pasture could vary appreciably according to the chemical form of the iodine, methyl iodide for instance having a much lower deposition velocity than elemental iodine.

K.B. MISTRY: Could you elaborate on the agronomic and soil factors that contribute to the estimated 14% annual decrease in the availability of ^{90}Sr as computed from the relationships between $^{90}Sr/Ca$ levels in the deposit and in milk?

R.S. BRUCE: First I would like to point out that the equation, which has been developed by Bartlett, represents an empirical approach and that the assumption of a single exponential change in availability is almost certainly an over-simplification of the actual situation. Further modifications to the equation will probably become necessary when we have information for a longer period in which the rate of deposition is low. The principal changes contributing to the reduction in availability are probably movement of the strontium-90 down the soil profile away from the main rooting zone of the pasture plants, and mixing of the upper soil layers when pastures are ploughed and re-seeded. Removal of strontium by crops and possibly slow changes in physico-chemical availability may also play some part. It is interesting that estimates of the rate of change in availability in different parts of the world have not differed very greatly, despite quite widely varying agricultural conditions.

K.-J. VOGT: With reference to Fig. 2 of the paper, have you an explanation for the fact that the ratio between the contamination of milk by ^{134}Cs and the contamination of the grass per unit area was different in May and August? Could this variation be caused by the difference in the grass density in the months concerned?

R.S. BRUCE: The experimental data that I presented were drawn from the work of Dr. Van den Hoek and his colleagues (Ref. [21] of the paper) as an illustration of the type of variation to be expected between deposition per unit area and contamination of milk. The authors attributed the seasonal changes partly to differences in the pasture quality and partly to variation in the transfer coefficient of caesium from diet to milk. The fraction transferred has been found by some workers to be related to the volume of milk produced and may also be affected by the nature of the fodder which the cows consume.

INCIDENCE DES PARAMETRES PHYSICO-CHIMIQUES SUR LA CONTAMINATION RADIOACTIVE DES ECOSYSTEMES IRRIGUES

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Abstract–Résumé

EFFECT OF PHYSICO-CHEMICAL PARAMETERS ON THE RADIOACTIVE CONTAMINATION OF IRRIGATED ECOSYSTEMS.

The paper is intended to assist in the selection of the values to be adopted in each particular situation for the parameters of radionuclide transfer from water to irrigated plants and the evaluation of the radiological capacity of irrigation water. The basis of the work is the experimental data recently derived by various laboratories in collaboration with the EURATOM-CEA Association. The radionuclides considered are the radioisotopes of ruthenium, cobalt, zinc and chromium. The behaviour of the pollutants in the various components of the ecosystem (water, soil, plants) has been studied. It depends principally on the physico-chemical form of the radionuclides discharged, the nature and concentration of the ions and compounds already present, and the kinetics of the complex equilibria existing in the environment.

INCIDENCE DES PARAMETRES PHYSICO-CHIMIQUES SUR LA CONTAMINATION RADIOACTIVE DES ECOSYSTEMES IRRIGUES.

L'objet de cette communication est d'apporter une contribution au choix des valeurs à adopter, dans chaque situation, pour les paramètres de transfert des radionucléides de l'eau aux végétaux irrigués et à l'évaluation de la capacité radiologique des eaux utilisées pour l'irrigation. On se base sur des données expérimentales récentes obtenues dans différents laboratoires en collaboration avec l'Association EURATOM-CEA. Les radionucléides envisagés sont des radioisotopes du ruthénium, du cobalt, du zinc et du chrome. Le comportement des polluants dans les différentes parties des écosystèmes (eau, sol, végétaux) est étudié. Il dépend notamment de la forme physico-chimique des radionucléides rejetés, de la nature et de la concentration des ions et composés déjà présents, enfin, des cinétiques des équilibres complexes existant dans le milieu.

INTRODUCTION

Parmi les processus liés à la mise en valeur du milieu, les pratiques de l'irrigation semblent avoir une incidence particulièrement importante sur la contamination interne de certains groupes de population. L'irrigation peut être réalisée par des méthodes très différentes; et, suivant qu'on utilise l'irrigation souterraine, l'irrigation au sol, l'irrigation par submersion, l'irrigation par aspersion, suivant les cultures et leur besoin en eau, il peut en résulter, dans le cas où les eaux sont contaminées, des apports polluants plus ou moins importants aux végétaux et, par suite, une pollution plus ou moins grave des chaînes alimentaires de l'homme.

Des études antérieures [1-3] ont montré qu'outre les radionucléides classiques à vie longue, ^{90}Sr et ^{137}Cs, il était utile d'envisager le cas des radioruthéniums et de produits métalliques d'activation à vie moyenne, en particulier celui des radiocobalts (^{58}Co et ^{60}Co), du zinc-65, du chrome-51, qu'on peut rencontrer dans de nombreux effluents liquides. Il s'agit d'isotopes d'éléments dont la physico-chimie est complexe et dont le comportement dans le milieu dépend de nombreux facteurs. L'objet de la présente communication est précisément de discuter et d'évaluer l'incidence de quelques facteurs physico-chimiques sur la contamination d'écosystèmes irrigués.

1. RAPPEL DE DONNEES PHYSICO-CHIMIQUES D'ORDRE GENERAL

Ruthénium

C'est un élément de la deuxième triade de la colonne VIII du tableau périodique. Il existe dans tous les états de valence. Les dérivés les plus importants, du point de vue nucléaire, sont ceux du nitrosylruthénium, groupement qui se comporte comme un ion trivalent $\mathrm{Ru\,NO^{3+}}$ susceptible de former de très nombreux complexes minéraux et organiques.

Cobalt

C'est un élément de la première triade de la colonne VIII du tableau périodique. Il existe sous deux valences fondamentales: 2 et 3. Le potentiel d'oxydoréduction du couple $\mathrm{Co^{2+}}$ - $\mathrm{Co^{3+}}$ est particulièrement élevé. Sous les deux valences, mais surtout sous la valence 3, le cobalt donne des complexes très stables.

Zinc

La physico-chimie du zinc est beaucoup plus simple. Il est essentiellement bivalent et donne des complexes moyennement stables.

Chrome

C'est un élément de transition du sous-groupe VI-a. Les valences normales vont de 2 à 6. Les sels chromiques ($\mathrm{Cr^{(III)}}$) sont très hydrolysables, les complexes correspondants sont stables. Le chrome VI existe sous forme anionique de chromates et de bichromates.

Il est actuellement difficile de préciser la forme physico-chimique exacte de ces éléments dans les effluents liquides. Il est vraisemblable que le ruthénium s'y trouve à l'état de nitrosylruthénium associé surtout à des groupements nitrites (nitronitrosylruthéniums), nitrates nitratonitrosylruthéniums, moins stables que les précédents), de chlorure et d'hydroxyde de nitrosylruthénium. Le zinc et le cobalt, et éventuellement le chrome III s'y trouvent en partie à l'état ionique simple, en partie à l'état complexé, notamment par les composés utilisés lors de la décontamination des surfaces métalliques irradiées (EDTA, détergents). Une partie du chrome doit se trouver à l'état de chromates. Enfin, une fraction plus ou moins grande de ces éléments existe, dans les effluents, à l'état d'ions polymères, de formes colloïdales et de formes particulaires.

Dès leur dispersion dans le système aquifère récepteur, les divers radionucléides, sous leurs différentes formes physico-chimiques, subissent l'action des facteurs intrinsèques du milieu: pH, anions précipitants, oxydoréduction, complexation, sorption sur des colloïdes argileux ou humiques, adsorption et absorption par les organismes irrigués, et cette action se poursuit évidemment dans les écosystèmes irrigués.

2. INCIDENCE DES FACTEURS PHYSICO-CHIMIQUES SUR LA CONTAMINATION DES ECOSYSTEMES IRRIGUES – EXEMPLES EXPERIMENTAUX

2.1. Ecosystèmes irrigués par submersion (études réalisées en collaboration avec la Division de biologie d'EURATOM à Ispra [1-3]

On a suivi la contamination des rizières et du riz irrigués par submersion. Le riz était cultivé soit en pots de Mitscherlich, soit dans des modèles de rizières expérimentales constituées par des séries de bacs remplis d'un sol prélevé dans la région rizicole de la Plaine du Pô (Italie) et irriguées avec de l'eau du Lac Majeur (Ca: 20 ppm; Co: 10^{-4} ppm; Zn: 0,04 ppm; traces de Cr). Les radionucléides envisagés étaient les radiocobalts, les radioruthéniums, le chrome-51, apportés par l'eau d'irrigation.

Cobalt

On a étudié l'incidence de l'état physico-chimique du radionucléide introduit dans l'eau: cobalt ionique et complexe cobalt-EDTA. Le cobalt ionique est capté par l'horizon le plus superficiel du sol et l'eau d'irrigation circulant sur le sol tend à s'appauvrir en cobalt; le cobalt complexé migre davantage en profondeur dans le sol et vers l'aval avec l'eau d'irrigation. Le cobalt est peu transféré de l'eau aux plantes et migre peu vers le grain, ce transfert et cette migration étant moindres dans le cas du cobalt complexé par l'EDTA que dans celui du cobalt introduit à l'état ionique. Le facteur de transfert

$$F = \frac{\text{Co dans le grain}}{\text{Co dans l'eau}}$$

est voisin de 0,6 pour Co^{2+} et de 0,1 pour Co-EDTA.

Ruthénium

On a testé l'incidence de l'état physico-chimique du radionucléide introduit dans l'eau. Les essais ont été réalisés en vases de Mitscherlich en utilisant diverses formes synthétiques du nitrosylruthénium préparées à cette occasion par Ugine Kuhlmann [3]: chlorure de nitrosylruthénium, tétranitronitrosylruthénium et hydroxyde de nitrosylruthénium. Dans les trois cas, l'horizon superficiel du sol retient une grande partie du ruthénium, le facteur de transfert

$$F = \frac{\text{Ru dans le grain}}{\text{Ru dans l'eau}}$$

est faible; cependant, il est cent fois plus grand dans le cas de l'hydroxyde de nitrosylruthénium que dans le cas des complexes nitrites et nitrates (respectivement, F = 0,03 et 0,0003).

Chrome

On a étudié l'influence de la teneur de l'eau d'irrigation en chromates stables sur la contamination du riz en chrome-51 introduit à l'état de chromate [4]. Les divers isotopes sont retenus avec la même intensité par le sol des rizières et l'eau d'irrigation s'épure au cours de sa circulation, parallèlement, en chrome stable et en radiochrome. Seule la surface du sol est contaminée. La contamination du grain en chrome-51 est extrêmement faible. Celle du chaume est d'autant plus forte que la concentration du chrome stable dans l'eau d'irrigation est plus grande:

$$F = \frac{^{51}Cr \text{ dans le chaume}}{^{51}Cr \text{ dans l'eau}}$$

est respectivement égal à 7, 10 et 20 pour des concentrations en chrome stable respectivement égales à 0,005 ppm, 0,05 ppm et 0,5 ppm, alors qu'au contraire les facteurs de transfert du chrome stable de l'eau vers les chaumes diminue quand la concentration de l'eau en chrome stable augmente. Du reste, le chrome stable s'accumule, en valeur relative par rapport à l'eau, beaucoup plus que le chrome-51, bien que, en principe, les isotopes soient introduits sous une même forme physico-chimique (facteurs de transfert eau → grain pour le chrome stable respectivement égaux à 90,5 et 0,5 pour des concentrations dans l'eau de 0,005 ppm et 0,5 ppm).

2.2. Ecosystèmes irrigués par aspersion

Les études expérimentales, réalisées en collaboration avec la Division de biologie d'EURATOM à Ispra et le Laboratoire de radio-écologie continentale du Département de protection du CEA à Cadarache portent surtout sur l'incidence de la forme physico-chimique du radionucléide introduit dans l'eau d'irrigation.

Ruthénium

On constate que seuls les organes directement exposés à l'eau contaminée sont pollués par le radioruthénium [5]. Les organes, comme les fruits, qui se sont développés après la fin des irrigations ne sont que très faiblement contaminés, et ceci, quelle que soit la forme physico-chimique du ruthénium. En ce qui concerne les organes directement exposés, comme les feuilles (salades par exemple), les facteurs de transfert eau → plantes sont de l'ordre de quelques dixièmes pour tous les composés du nitrosylruthénium envisagés. Par contre, l'élimination, par l'eau non contaminée, du ruthénium déposé sur des organes dépend de la forme physico-chimique: elle est plus intense pour le tétranitronitrosylruthénium, complexe non hydrolysable et très soluble, que pour le chlorure de ruthénium, le chlorure de nitrosylruthénium et l'hydroxyde de nitrosylruthénium, formes moins solubles ou plus instables en présence d'eau.

TABLEAU I. INCIDENCE DE L'ETAT PHYSICO-CHIMIQUE DU RADIOCOBALT ET DE LA QUALITE DE L'EAU D'IRRIGATION
Valeurs des facteurs de transfert eau → organes consommables par l'homme

	Eau déminéralisée		Eau du Rhône	
	Co salin	Co-EDTA	Co salin	Co-EDTA
Haricots (gousses)	2,75 ± 0,25	1,57 ± 0,25	0,77 ± 0,25	0,31 ± 0,25
Salades (feuilles)	4,93 ± 0,13	1,65 ± 0,13	1,57 ± 0,13	0,81 ± 0,13

Cobalt [6]

On a réalisé à Cadarache, sur des salades et des haricots placés en caisson phytotron, trois aspersions contaminantes (une aspersion chaque semaine à raison de cinquante litres d'eau par mètre carré en cinq heures). Les paramètres étudiés ont été, d'une part, l'état physico-chimique du cobalt, le cobalt étant introduit soit à l'état ionique, soit à l'état de complexe avec l'EDTA, d'autre part, la nature de l'eau d'irrigation, l'eau utilisée étant soit de l'eau déminéralisée, soit une eau du Rhône (Ca: 70 ppm; Fe total: 550 ppb; Fe soluble: 20 ppb; Mn soluble: 54 ppb; Pb soluble: 8 ppb; Cu soluble: 8 ppb; Zn soluble: 22 ppb; Co soluble: inférieur à 2 ppb). On a déterminé les facteurs de transfert du cobalt de l'eau aux organes consommés par l'homme (tableau I). On a constaté que, toutes choses égales par ailleurs, les facteurs de transfert étaient plus faibles pour le cobalt introduit à l'état de complexe que pour le cobalt introduit à l'état salin, qu'ils étaient plus faibles lorsqu'on utilisait une eau fortement minéralisée (Rhône) que lorsqu'on employait l'eau déminéralisée (tableau I).

Zinc

Des études expérimentales relatives au zinc-65 sont actuellement entreprises. En l'absence de résultats expérimentaux obtenus par nos soins, on s'est provisoirement référé à un certain nombre de données obtenues ailleurs. Il s'avère que de nombreux composés organiques présents dans les effluents et les systèmes hydrobiologiques sont susceptibles de complexer le zinc et de modifier son comportement vis-à-vis des sédiments et des sols, d'une part, des végétaux, d'autre part, les complexes anioniques ou neutres étant, bien entendu, moins prélevés à partir de l'eau que le zinc ionique et les complexes cationiques [7-11].

Chrome

L'expérience réalisée par la Division de biologie d'EURATOM à Ispra montre que le chrome III est plus retenu par les feuilles de salade (laitue romaine, non pommée) que le chrome VI des chromates, et que le lavage ultérieur par une eau non contaminée élimine le chrome plus intensément sous sa forme soluble que sous sa forme III très hydrolysable. Des expérimentations préliminaires effectuées jusqu'à présent, on peut, en admettant une additivité des dépôts de chrome lors de chaque aspersion, proposer les valeurs suivantes pour les facteurs de transfert eau → salades dans les cas où les plantes recevraient plusieurs arrosages:

$Cr^{(III)}$(ou Cr^{3+}) F : 2 à 5
$Cr^{(VI)}$(ou CrO_4^{2-}) F : 1

Cette dernière valeur serait à diminuer d'un facteur 2 environ pour les feuilles de salades lavées à l'eau (F : 0, 5).

3. DISCUSSION

Les résultats précédents seront discutés suivant différents aspects:

- incidence de l'irrigation sur certains paramètres physico-chimiques du milieu,
- incidence des équilibres fondamentaux existant dans le milieu, en particulier dans les eaux d'irrigation,
- incidence des conceptions précédentes pour la prévision et l'évaluation des conséquences d'une contamination radioactive des eaux d'irrigation.

3.1. Incidence de l'irrigation sur certains paramètres physico-chimiques du milieu

L'irrigation a toujours pour résultat d'accroître l'humidité du sol, ce qui peut avoir pour effet de modifier un certain nombre des caractéristiques des écosystèmes irrigués. En collaboration avec la Division de biologie d'EURATOM à Ispra, on a suivi, grâce à des mesures in situ et en lysimètres, l'incidence de l'immersion sur quelques paramètres importants de la contamination du riz [12].

On a constaté que la submersion du sol s'accompagnait d'une brusque diminution du potentiel d'oxydoréduction, d'une augmentation du pH de l'horizon superficiel du sol. On a noté, simultanément, une augmentation du manganèse échangeable du sol, du cobalt échangeable, et une augmentation plus tardive du fer mobile du sol. L'assèchement du sol, avant la récolte, conduit aux variations inverses. En ce qui concerne le manganèse, l'isotope ^{54}Mn et l'élément stable à l'état échangeable se comportent de la même façon.

On a constaté enfin que, dans le cas de rizières irriguées mais maintenues à la «capacité au champ», le prélèvement des radionucléides est inférieur à ce qu'il est dans les rizières submergées. En modifiant certaines caractéristiques fondamentales du sol, l'irrigation a donc, dans ce cas, créé des conditions favorables à une pollution plus grande des végétaux. Ces résultats pouvaient, dans leurs grandes lignes, être prévus d'après les valeurs des potentiels d'oxydoréduction [13] (tableau II).

D'autre part, en augmentant la mobilité chimique des isotopes stables des radionucléides apportés par l'eau, l'irrigation peut modifier, en général diminuer, l'activité spécifique de l'élément dans l'écosystème, ce qui devrait diminuer l'activité spécifique de l'élément dans la plante.

TABLEAU II. POTENTIELS D'OXYDOREDUCTION (E_7^0) (en volts)

$O_2 + 4H^+ + 4e$	$\rightleftarrows$	$2H_2O$	+ 0,83
$NO_3^- + H_2O + 2e$	$\rightleftarrows$	$NO_2^- + 2OH^-$	+ 0,43
$MnO_2 + 4H^+ + 2e^-$	$\rightleftarrows$	$Mn^{2+} + 2H_2O$	+ 0,41
$Fe(OH)_3 + e^-$	$\rightleftarrows$	$Fe^{2+} + 3OH^-$	- 0,13
$2H^+ + 2e$	$\rightleftarrows$	H_2	- 0,42
$Co^{3+} + e^-$	$\rightleftarrows$	Co^{2+}	+ 1,84 (NO_3H $3\underline{N}$)
$Co(OH)_3 + e^-$	$\rightleftarrows$	$Co(OH)_2 + OH^-$	+ 0,6

3.2. Incidence des équilibres existant dans le milieu

Il est clair qu'en raison de leur masse négligeable les radionucléides apportés subissent les équilibres existant normalement dans les écosystèmes irrigués. Il doit en résulter des comportements identiques pour tous les isotopes d'un même élément à condition, bien entendu, qu'ils se trouvent sous le même état physico-chimique. Or, il est peu fréquent que les radionucléides présents dans les effluents se trouvent dans le même état physico-chimique que leurs isotopes présents dans le milieu. Dans ce cas, la réalisation d'un équilibre demande un certain temps. Les délais sont très courts lorsque les réactions n'impliquent pas de remaniements moléculaires (par exemple, dans le cas de transferts d'électrons), mais ils sont plus longs dans les autres éventualités (équilibre entre Cr^{3+} et CrO_4^{2-} par exemple). Ces conceptions expliquent peut-être pourquoi isotopes instables et isotopes stables d'un même élément n'ont pas, même après un temps relativement long, un comportement identique.

Quoi qu'il en soit, les équilibres sont régis par des lois du type loi d'action de masse: c'est dire qu'il faut prendre en considération, et les concentrations de tous les éléments et composés susceptibles de réagir les uns sur les autres, et les constantes thermodynamiques définissant les cinétiques de chacun des équilibres possibles [10]. Cette conception explique en particulier l'incidence de la composition ionique de l'eau d'irrigation sur la contamination des espèces irriguées. Ainsi, la contamination en radiocobalt des végétaux irrigués par aspersion est moindre si le vecteur de la contamination est une eau très minéralisée que si l'eau d'irrigation est peu minéralisée. Tout se passe comme si des cations stables présents dans l'eau concurrençaient le cobalt.

Des expérimentations effectuées à Ispra conduisent à des conclusions de même signification: on a irrigué par aspersion des plants de tomates en utilisant deux types d'eaux: une eau déminéralisée et une eau du Lac Majeur auxquelles on a ajouté des quantités variées d'EDTA de façon à obtenir des concentrations échelonnées de 0 à 10 ppm, puis une quantité constante de radiocobalt sans extraîneur. Dans tous les cas, on a apporté dix litres d'eau d'irrigation par mètre carré en dix minutes. On a constaté (tableau III) que l'apport d'EDTA à l'eau du Lac Majeur favorisait la rétention du radiocobalt sur les parties aériennes de la plante, alors qu'un apport identique à l'eau déminéralisée était presque sans effet

TABLEAU III. FACTEURS DE TRANSFERT EAU → TOMATE (FEUILLES) EN FONCTION DE LA NATURE DE L'EAU D'IRRIGATION ET DE L'APPORT D'EDTA

Concentrations en EDTA (ppm)	Eau du Lac Majeur	Eau déminéralisée
0	1	4 à 5
1	2 à 3	4
10	2 à 3	3 à 4

TABLEAU IV. STABILITE DES COMPLEXES DE L'EDTA

Valeurs de log k_1 pour des solutions diluées ($k_1 = \frac{(\text{Me. EDTA})}{(\text{Me}).(\text{EDTA})}$

Eléments	log k_1	Eléments	log k_1
Ca^{2+}	10,7	Co^{3+}	≈36
Fe^{2+}	14,3	Zn^{2+}	16,3
Fe^{3+}	25,1	Cu^{2+}	18,8
Co^{2+}	16,2	Mn^{2+}	13,6

(effet légèrement dépressif). Pour les faibles additions d'EDTA la contamination est plus forte avec l'eau déminéralisée qu'avec l'eau du lac. Pour les plus fortes additions d'EDTA, les écarts constatés entre les résultats obtenus avec les deux types d'eau s'amenuisent.

La même expérimentation répétée sur des haricots n'indique aucune action de l'EDTA. Il est à remarquer que, tous autres facteurs étant égaux, la contamination de la feuille de haricot est moindre que celle de la tomate. Il existe donc, en ce qui concerne cet aspect, des différences spécifiques assez nettes, en rapport peut-être avec des minéralisations différentes des tissus foliaires.

Il est clair que, dans l'expérience précédente, l'EDTA introduit avant le radiocobalt dans l'eau du lac répartit son action complexante sur les divers ions présents dans l'eau en fonction de leurs concentrations respectives et des constantes de stabilité des complexes [14] (tableau IV): les traces de radiocobalt ont peu de chance d'être complexées, l'EDTA étant accaparé par les éléments stables. Suivant ce concept, il est logique de penser que l'addition d'EDTA peut inhiber par complexation l'action concurrentielle du ou des éléments antagonistes du cobalt. Par contre, on s'explique mal jusqu'à présent pourquoi l'EDTA introduit dans l'eau déminéralisée ne modifie que très peu la rétention du radiocobalt.

Quoi qu'il en soit, on comprend l'incidence possible d'une pollution des eaux par des produits chimiques complexants en amont des sites de rejets d'effluents radioactifs.

TABLEAU V. FACTEURS DE TRANSFERT F EAU → VEGETAUX IRRIGUES

Végétaux	Conditions d'irrigation	Radionucléides	Valeurs de F
Riz (grain)	Submersion, eau du Lac Majeur (Ca: 20 ppm)	Co^{2+}	0, 6
		Co^{2+} – EDTA	0, 1
		Hydroxyde de nitrosylruthénium	0, 03
		Autres formes de ruthénium	0, 0003
Salades non pommées	Aspersion, eau déminéralisée	Co^{2+}	5
		Co – EDTA	1, 5
	Aspersion, eau du Rhône (Ca: 70 ppm)	Co^{2+}	1, 5
		Co – EDTA	0, 8
	Aspersion, eau du Lac Majeur (Ca: 20 ppm)	Hydroxyde de nitrosylruthénium	0, 5
		Autres formes de ruthénium	0, 15
		Cr^{3+}	5
		$Cr^{(VI)}$	1
Tomates (fruits)	Aspersion, eau du Lac Majeur (Ca: 20 ppm)	Toutes formes physico-chimiques du ruthénium	0, 05
Haricots (gousses)	Aspersion, eau déminéralisée	Co^{2+}	3
		Co – EDTA	1, 5
	Aspersion, eau du Rhône (Ca: 70 ppm)	Co^{2+}	0, 8
		Co – EDTA	0, 3
Toutes productions irriguées	Aspersion	Zinc-65 dans la Columbia River	5

3.3. Incidence des conceptions précédentes sur la prévision et l'évaluation des conséquences d'une contamination radioactive des eaux d'irrigation

Il résulte des études précédentes que la prévision des conséquences de pollutions radioactives des eaux destinées à l'irrigation nécessite l'examen de chacune des situations possibles. Chaque cas concret doit être traité séparément. Dans la pratique, si on doit prévoir les conséquences d'un rejet d'effluents radioactifs dans un système hydrologique déterminé, on devra se reporter aux cas concrets déjà étudiés qui paraissent se rapprocher le plus du cas à traiter. A titre indicatif, le tableau V donne une idée de la variabilité expérimentale des facteurs de transfert F eau → végétaux irrigués en fonction de la forme physico-chimique. Entre les valeurs extrêmes relatives à un même radionucléide, une variabilité d'un facteur de l'ordre de 10 est souvent constatée. Dans le cas où on considère des organes végétaux consommés par l'homme, la dose délivrée à l'organe critique de l'homme du fait de l'ingestion de tels aliments est soumise à la même variabilité. D'une manière générale, les facteurs de transfert eau → végétaux irrigués sont pour un même nucléide moindres pour les formes complexées que pour les formes ioniques simples. Mais les zones intéressées par les pollutions par les formes complexes sont généralement plus étendues. Si les risques sont moins aigus, les surfaces concernées sont plus grandes et les problèmes de protection perdent, de ce fait, leur intérêt local, pour prendre parfois leur acuité maximale plus en aval. D'autre part, l'apport, parfois énorme, d'eau peut entraîner une contamination relativement grande des sols irrigués, facteur dont il faut tenir compte pour la protection du personnel chargé des travaux culturaux, d'une part, et pour l'évaluation des arrière-effets de la contamination des sols sur les cultures ultérieures dans le programme d'assolement utilisé.

CONCLUSION

En se basant sur des données expérimentales, on a étudié les variabilités des conditions de contamination d'écosystèmes irrigués, notamment en ce qui concerne le ruthénium et divers produits d'activation. Les paramètres envisagés ont été surtout la forme physico-chimique du radionucléide lors de sa dispersion dans l'eau et la nature physico-chimique de cette eau. L'accent doit être mis sur plusieurs aspects:

— Les formes physico-chimiques introduites à l'état de complexes sont chimiquement plus mobiles que les formes cationiques simples et moins prélevées que ces dernières par les organismes irrigués.

— Les différents ions et composés présents dans les écosystèmes agissent par leurs concentrations relatives et leurs affinités réciproques sur les équilibres cinétiques; il en résulte une modification plus ou moins rapide des conditions initiales réalisées au voisinage du point de rejet.

— Il peut exister une interdépendance nette entre les pollutions radioactives, d'une part, les pollutions chimiques, d'autre part, comme le montrent par exemple l'incidence d'une pollution des eaux par l'EDTA préalablement à une pollution par le radiocobalt, ou l'incidence du niveau du chrome stable dans les eaux sur la contamination des végétaux irrigués par le chrome-51.

— En ce qui concerne la protection du consommateur des végétaux irrigués, l'attention doit être attirée sur les cas où les facteurs de transferts eau → végétaux sont nettement supérieurs à l'unité, c'est-à-dire, pour le cobalt ionique simple sur le cas de certains légumes comme les salades et les haricots verts irrigués avec de l'eau peu minéralisée, pour le chrome III, sur celui de légumes dont les parties consommables sont celles qui interceptent directement l'eau d'arrosage (salade); le cas du zinc doit être étudié expérimentalement d'une façon approfondie.

— Ces études fournissent en principe des bases de départ pour des études systématiques relatives aux pollutions des écosystèmes irrigués par les métaux lourds. Mais l'expérience a montré que les isotopes d'un même élément peuvent se comporter d'une façon assez différente, du fait des ordres de grandeur très différents de leurs concentrations dans le milieu et de la réalisation incomplète d'équilibres physico-chimiques.

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DISCUSSION

R. FUKAI: Regarding the Co-EDTA used in the experiments, how did you prove that the cobalt added is in complex form? Is there any excess of EDTA present in the solution added?

R. BITTEL: The Co-EDTA complex solution was prepared before the experiment by adding excess EDTA to the cobalt. This solution was then introduced into the irrigation water. Some of the cobalt may subsequently be de-complexed through the effect of the cations present in high concentration in the water.

R. FUKAI: I understand that you oxidized chromium-51 with a carrier before use. Our experience has shown that commercial chromium-51 is always a mixture of Cr^{3+} and Cr^{6+}, and whenever we have wanted to be specific about the chemical form of the chromium we have had to oxidize or reduce to obtain a uniform chemical form.

R. BITTEL: The ^{51}Cr used is obtained from commercial chromate solutions claimed to be carrier-free. However, in these solutions the stable $Cr/^{51}Cr$ ratio is very high, and it is therefore possible that the ^{51}Cr is in a physico-chemical form which is different from the chromate form of the stable chromium, and that it evolves differently. At all events this is a working hypothesis which one might try to verify.

I quite agree with your reference to the importance of ensuring that the ^{51}Cr and the stable Cr are in a uniform physico-chemical state.

P. REINIGER: May I add a comment on the physico-chemical form of $Na_2{}^{51}CrO_4$. The hexavalent $^{51}CrO_4$ undergoes a reduction process depending on the storage time. On delivery, about 96% of the chromium is in hexavalent form, but this percentage drops to about 80 after six weeks' storage.

R. BITTEL: Your observation confirms the lability of oxidation-reduction equilibrium states with respect to chromium and is of particular interest in connection with Mr. Fukai's remarks.

R.J. ROUX: Over what sort of period did you measure the transfer factor?

R. BITTEL: Over a complete growth cycle of the plant.

R.J. ROUX: What are the concentrations of pollutants in your solutions in relation to the concentrations of Co, Ru or other substances normally present in the plant, and in relation to the MPC in water? Also, do these concentrations in your solutions modify the concentrations in the plant?

R. BITTEL: The radioisotopes are of course used in tracer amounts. The concentrations of stable elements in the irrigation water will be found in the text of the paper. The stable nuclides (except chromium) have not been determined in the plants.

INFLUENCE OF AGRONOMIC PRACTICES ON UPTAKE OF FISSION PRODUCTS BY CROPS FROM SOILS OF REGIONS ADJOINING NUCLEAR INSTALLATIONS IN INDIA

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Abstract

INFLUENCE OF AGRONOMIC PRACTICES ON UPTAKE OF FISSION PRODUCTS BY CROPS FROM SOILS OF REGIONS ADJOINING NUCLEAR INSTALLATIONS IN INDIA.

The influence of a variety of agronomic practices on the transfer of ^{90}Sr, ^{137}Cs, ^{106}Ru and ^{144}Ce to two major crops, maize and rice, from black soils was examined. These soils are typical of the regions where river and ground or well waters, to which low-level effluent discharges and accidental releases from nuclear installations, namely the Rajasthan and Tarapur Atomic Power Stations and Fuel Reprocessing Plant at Trombay, could have access, are used for irrigation. Parallel data on radionuclide uptake by plants from a typical laterite were also obtained.

Studies on radiostrontium indicated that the addition of compost results in reduced uptake from both black and laterite soils. Complexing action of the polysaccharide fraction of compost on radiostrontium markedly lowers its availability to plants. Beneficial effects of liming accrue in soils with low exchangeable calcium status. A reduction in the radiostrontium content of plants was obtained with nitrogen and potassium fertilizers; phosphate fertilization had no consistent effects. No significant differences were observed between various nitrogen or potassium carriers. Greater downward movement of radiostrontium in soil profiles was observed with moderately high calcium irrigation water than with either moderately high sodium irrigation water or simulated rain water. Radiostrontium was less mobile in black soil than in laterite. A flooded regime did not significantly influence the radiostrontium content of rice grown on both soils.

Studies on gamma emitters indicated that, over comparable durations, ^{137}Cs uptake was markedly greater than that of ^{106}Ru and ^{144}Ce in both soil types. While ^{106}Ru uptake was higher than that of ^{144}Ce in the black soil, in the laterite ^{144}Ce uptake far exceeded that of ^{106}Ru. Compost application reduced ^{137}Cs uptake in both soils, ^{106}Ru uptake was not affected, while variable effects were observed on ^{144}Ce uptake. Flooding resulted in marked increase in ^{137}Cs, ^{106}Ru and ^{144}Ce accumulated by rice in the laterite. In the black soil, however, a similar effect was obtained only on ^{137}Cs uptake.

INTRODUCTION

As part of our continuing program of investigations into the behaviour of long-lived fission products and naturally occurring radionuclides in the soil-plant continuum [1-9] we have undertaken studies on the influence of a variety of agronomic practices on the transfer of strontium-90, caesium-137, ruthenium-106 and cerium-144 to crop plants from typical black soils (pellusterts, chromusterts and pelluderts) and laterite soils (plinthudults and oxisols), which represent two of the principal soil groups of India [10]. The black-soil zone includes regions adjoining the major nuclear installations, namely the Tarapur Atomic Power Station, the CIRUS Reactor and the Trombay Fuel Reprocessing Plant in Maharashtra State and the Rajasthan Atomic Power Station in Rajasthan State (Table I).

TABLE I. NUCLEAR INSTALLATIONS COVERED IN THE PRESENT INVESTIGATION

Nuclear installation	Location	Power output	Reactor type	Fuel	Moderator/Coolant	Soil type of adjoining regions	Year of commissioning
CIRUS Reactor	Trombay, Maharashtra	40 MW	NRX	Natural uranium	Heavy water/ Light water	Black	1961
Tarapur Atomic Power Station	Tarapur, Maharashtra	2 × 210 MW(e)	BWR	Enriched uranium	Light water	Black	1969
Rajasthan Atomic Power Station	Ranapratapsagar, Rajasthan	2 × 215 MW(e)	CANDU	Natural uranium	Heavy water	Medium black	1972
Fuel Reprocessing Plant [a]	Trombay, Maharashtra					Black	1964

[a] Separation of plutonium and fission products from irradiated uranium from CIRUS reactor.

Our program on long-lived fission products has been planned primarily to investigate the fate of these radionuclides, which are deposited on soils from global fall-out, in contrasting soil-plant regimes of India typical of the tropics and sub-tropics for which, unlike the situation for northern temperate regions, hardly any quantitative data exist [11]. In addition, the study aims at obtaining information on factors governing the transfer to crops of these nuclides, which could be present in accidental releases or controlled low-level discharges into ground and well waters from the nuclear installations at Trombay and Tarapur [12, 13] and into the Chambal river waters from the Rajasthan Atomic Power Station [14], all of which are used for irrigation.

The present paper reports our findings on the effects of soil amendments, namely, lime and organic matter, and nitrogen, phosphorus and potassium fertilizers on radionuclide uptake by maize, a widely cultivated cereal crop of the black and laterite zones. The influence of a flooded water regime, normal for rice cultivation in the black and laterite soils, on the uptake of radionuclides by rice was examined. The effects of leaching with simulated rain, irrigation waters of various qualities and a chelating agent, EDTA, on the downward movement of radiostrontium in the two soil types are also reported. The agronomic treatments included in the present investigation represent the fertility and irrigation practices normally adopted for cereal crop cultivation in the black and laterite soil zones.

MATERIALS AND METHODS

The physico-chemical characteristics of black and medium black clay loams from Maharashtra and Rajasthan States and laterites from Maharashtra State are reported in Table II. These soils are representative of their respective soil types. In view of the similar characteristics of soils of each soil type, one black soil and one laterite were used for each experiment.

For the plant growth experiments 5-10 kg lots of surface soils (0-20 cm depth) passed through a 2 mm sieve were filled in porcelain or Polythene pots. The specific agronomic treatments including the dose rates employed for the experiments on effects of liming, organic matter addition and NPK fertilizer application are described in the tables reporting data from these experiments under Results and Discussion. After treatment, the soils were brought to field capacity moisture status for a period of 8 days and 5 μCi of radionuclides in a suitable volume of distilled water were then applied to the surface of soils to simulate conditions of contamination through irrigation waters and/or deposition of soluble global fall-out. ^{89}Sr was used as a tracer for ^{90}Sr. Each treatment was replicated five times. One week after contamination 12 seeds of maize (Zea mays L. var. NP 65 K) or rice (Oryza sativa L. var. D 622) were shown. The plants were thinned to two per pot one week after germination. For maize the pots were maintained at field capacity moisture throughout the duration of growth, while 5 cm of standing water was maintained in pots after 15 days growth in the flooding treatment for rice. The experiments were carried out in a greenhouse or in a growth room where the temperature was maintained at 23 ± 2°C, relative humidity 65 ± 5% and a 12-h photoperiod with light intensity of 800 foot-candles. The plants were harvested at flowering, dried at 90°C, weighed and taken up for radioassay.

TABLE II. SOIL CHARACTERISTICS

Soil type and location	Clay mineral type	pH (1: 2.5)	Total soluble salts (mmho/cm)	Cation exchange capacity (meq%)	Ex. Ca (meq%)	Ex. K (meq%)	Org. carbon (%)	Available nutrient rating		
								N	P_2O_5	K_2O
Black clay loam, Maharashtra	2: 1[a]	8.0	0.52	40.5	22.1	0.09	0.78	High	High	Low
Black clay loam, Maharashtra	2: 1	8.1	0.45	40.5	27.7		0.67	Medium	Medium	Medium
Medium black clay, loam, Rajasthan	2: 1	7.4		27.0			0.28	Low		
Medium black clay loam, Rajasthan	2: 1	8.5		29.7		0.9	0.68	Medium		
Laterite, Maharashtra	1: 1[b]	6.4	1.11	19.0	9.1	0.15	1.62	High	High	High
Laterite, Maharashtra	1: 1	5.5	0.10	19.0	3.0		0.90	High	Low	Low

[a] Montmorillonite/illite.

[b] Kaolinite.

For ^{89}Sr assay the dry plant material was wet-ashed with concentrated nitric acid and aliquots of clear extracts were pipetted on to 2.5 cm stainless steel discs, dried and counted in a low background GM counter. The gamma emitters were assayed through gamma-spectrometry of dry plant material packed in 15-mm diameter test tubes using a 3×3-in. NaI(Tl) crystal integral line assembly and a 512-channel pulse-height analyser attached to an oscilloscope and computer read-out typewriter. The determination of individual radionuclides was carried out using the photopeaks of 513 keV for ^{106}Ru, 81 keV for ^{144}Ce and 662 keV for ^{137}Cs. Since each plant tissue sample contained only one radionuclide, no interference from other nuclides occurred during radioassay. The radioassay data were corrected for background.

For the incubation studies carried out concurrently with the plant growth experiments on the effects of lime application, 100 g lots of uncontaminated soils in glass jars were mixed with $CaCO_3$ at the rates employed in the plant growth experiments. Four replicates were maintained for each treatment and the soils were incubated at field capacity moisture level and room temperature for the duration of the plant growth experiments, after which the soils were air dried and pH and exchangeable calcium were determined.

For the water culture experiment undertaken to examine the effects of polysaccharide fraction of organic matter on plant uptake of radiostrontium, the polysaccharide fraction, extracted from compost used in the soil experiments by the method of De Datta et al. [15], was dissolved in 900 ml of distilled water and 0.5 μCi carrier-free ^{89}Sr was added to the solution. Three-week old maize plants grown for 10 days on a full-strength Hewitt culture solution were transferred to Polythene jars containing a ^{89}Sr-labelled polysaccharide solution. Parallel jars containing ^{89}Sr added to distilled water served as controls. Two plants were kept in each jar and 5 replicate jars were maintained per treatment. The duration of radionuclide treatment was 12 days, after which the plants were sacrificed and separated into shoots and roots. The roots were treated for 1 hour in 0.01$\underline{M}$ $CaCl_2$ to remove non-specifically absorbed ^{89}Sr. Methods of subsequent processing and assay of the plant tissues were similar to those employed in the soil experiments.

For the leaching experiment a sufficient quantity of each soil was weighed and transferred to a glass leaching container so that after compaction it provided a depth of 15 cm. The soils were packed with a packing block similar to that described for measurement of hydraulic conductivity [16]. The 15-cm layer of untreated soil in the container was covered with 1.25-cm layer of soil labelled with 1.0 μCi ^{89}Sr. The labelled layer was covered with a 1.25-cm layer of untreated soil and packed with a wooden block to the same bulk density as the 15-cm layer. The leaching treatments consisted of various amounts of deionized water (representing rain water), 0.005$\underline{N}$ $CaCl_2$ solution (representing moderately high calcium irrigation water), 0.005$\underline{N}$ NaCl (representing moderately high sodium irrigation water) and 0.01$\underline{M}$ EDTA. One column of each soil type was also maintained at field capacity moisture level to serve as control. No breakthrough of the nuclide occurred during the leaching treatments, which extended up to 5 months in some cases. The soil columns were separated upon completion of the leaching treatment into layers of 2.5-cm thickness, the individual segments were air dried, weighed and assayed for ^{89}Sr.

RESULTS AND DISCUSSION

1. Studies on radiostrontium

1.1. Effects of lime

Data on the uptake of ^{89}Sr by maize under different levels of liming, per cent reduction in radionuclide uptake and incubation data on changes in exchangeable calcium levels and pH for the black and laterite soils are presented in Tables III and IV respectively.

TABLE III. EFFECT OF CALCIUM APPLICATION ON UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN ON BLACK SOIL

Calcium treatment[a] (meq/100 g soil)	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr (%)	Incubation data	
			pH	Exchangeable calcium (meq%)
Control, no Ca	0.75		8.0	23.50
4.48	0.69	8.1	8.0	24.88
11.20	0.68	9.0	8.0	27.46
22.40	0.68	9.2	8.0	28.16
Least sig. deviation (P = 0.05)	N.S.[b]			1.21

[a] $CaCO_3$

Basal dressing — N: P_2O_5: K_2O = 60: 40: 60 kg/ha.

[b] N.S. not significant.

TABLE IV. EFFECT OF CALCIUM APPLICATION ON UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN ON LATERITE SOIL

Calcium treatment[a] (meq/100 g soil)	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr uptake (%)	Incubation data	
			pH	Exchangeable calcium (meq%)
Control, no Ca	1.51		6.4	9.40
4.48	1.17	22.5	7.1	10.22
11.20	1.10	27.2	7.2	11.60
22.40	1.02	32.5	7.3	13.62
Least sig. deviation (P = 0.05)	0.25			0.33

[a] $CaCO_3$

Basal dressing — N: P_2O_5: K_2O = 60: 40: 60 kg/ha.

Our results indicate a highly significant reduction in the uptake of radiostrontium by plants with application of $CaCO_3$ in the laterite, which has low natural exchangeable calcium status. Data from incubation studies reveal that in this soil lime addition resulted in marked enhancement in exchangeable calcium, which was accompanied by a substantial increase in soil pH, indicating progressive replacement of exchangeable H^+ by Ca^{++}. On the other hand, liming treatments had no influence on the ^{89}Sr uptake from the black soil; pH and exchangeable calcium levels were also unaffected by lime application.

It is well established from nutrient culture experiments that the extent of uptake and upward transport of strontium is controlled primarily by the calcium level in the external medium [17]. Further, the absorption of strontium and calcium by plants grown in soil is reported to be largely dependent on the relative concentrations of the two ions in the soil solution [17]. These reports are compatible with our observations that plant uptake of ^{89}Sr is significantly reduced only in soils where $CaCO_3$ application resulted in a concommitant increase in the exchangeable calcium level, which is a fair index of the concentration of calcium ions in the soil solution [18].

1.2. Effects of organic matter

Data on the effects of organic matter addition on plant uptake of ^{89}Sr in the black and laterite soils are reported in Table V.

Addition of organic matter resulted in significant reduction in the uptake of ^{89}Sr by maize on both soil types. Earlier studies [15] have shown that radiostrontium added to soil and soil extracts is rapidly complexed by the polysaccharide fraction of soil organic matter. The complexing action of free functional groups, namely carboxylic and phenolic, of soil organic matter on a number of fission products including radiostrontium has been

TABLE V. EFFECT OF ORGANIC MATTER APPLICATION ON UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN ON BLACK AND LATERITE SOILS

Organic matter treatment[a] (g/kg soil)	Black soil		Laterite soil	
	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr uptake (%)	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr uptake (%)
Control, no o.m.	2.01		4.57	
11.2	1.28	36.6	3.96	13.3
22.4	1.08	46.3	3.67	19.5
Least significant deviation (P = 0.05)	0.79		0.69	

[a] Vegetable compost — C/N = 9.7
Basal dressing — $N:P_2O_5:K_2O$ = 60: 40: 60 kg/ha.

TABLE VI. EFFECT OF POLYSACCHARIDE FRACTION OF ORGANIC MATTER ON UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN IN WATER CULTURE

Treatment	Shoots		Roots	
	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr uptake (%)	^{89}Sr uptake (nCi/g dry wt.)	Reduction in ^{89}Sr uptake (%)
Distilled water (Control)	30.89	-	40.84	-
Distilled water + polysaccharide fraction	16.11	47.9	27.31	33.6
Least significant deviation (P = 0.05)	8.99		9.86	

also demonstrated [19, 20]. These reports, together with the data from our water culture experiments (Table VI), suggest that complexing effects of the polysaccharide fraction of organic matter on radiostrontium results in a markedly reduced availability of the radionuclide for absorption by plants. While the compost used in our experiments contained small amounts of exchangeable calcium, the quantities of calcium introduced into the soil (0.045-0.09 meq%) are not likely to contribute significantly to the reduction in ^{89}Sr uptake.

1.3. Effects of NPK fertilizers

Data on the effects of nitrogen, phosphorus and potassium fertilization on the uptake of ^{89}Sr by maize from black and laterite soils are shown in Table VII.

It is evident from Table VII that the radiostrontium content in plants grown in both soil types is significantly reduced by the application of nitrogen and potassium fertilizers at the rates normally recommended for a variety of crop plants. Phosphate addition did not, however, produce consistent effects on radionuclide uptake.

Since no significant yield response was obtained with either N or K fertilization on the two soils, which have a high fertiliy rating (Table II), the observed reduction in ^{89}Sr concentration in plants cannot be attributed to dilution with increased production of dry matter. Further, data in Table VIII indicate that similar effects were obtained with four different nitrogen carriers, namely $(NH_4)_2SO_4$, NH_4Cl, $NaNO_3$ and $CO(NH_2)_2$, or with the two potassium carriers, namely K_2SO_4 and KCl. Our present findings do not support the view [21] that reduction in ^{89}Sr uptake with $(NH_4)_2SO_4$ or K_2SO_4 addition could be due to formation of $SrSO_4$ precipitate in soil, resulting in a lowering of the available Sr^{++} ions for absorption by plants. It is likely that the effects observed are, in the main, due to antagonistic

TABLE VII. EFFECT OF NITROGEN, PHOSPHATIC AND POTASSIUM FERTILIZATION ON YIELD AND UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN ON BLACK AND LATERITE SOILS

Treatment (kg/ha)		Black soil		Laterite soil	
		Dry matter (g/pot)	^{89}Sr uptake (nCi/g dry wt.)	Dry matter (g/pot)	^{89}Sr uptake (nCi/g dry wt.)
N[a]	0	6.4	0.86	14.2	2.47
	60	7.1	0.72	12.4	2.00
	150	4.3	0.40	13.1	1.82
Least sig. deviation ($P = 0.05$)		N.S.	0.14	N.S.	0.09
P_2O_5[b]	0	7.2	0.86	13.3	2.23
	40	6.9	0.69	12.2	2.07
	100	7.5	0.76	12.6	2.39
Least sig. deviation ($P = 0.05$)		N.S.	N.S.	N.S.	N.S.
K_2O[c]	0	6.4	0.86	14.2	2.47
	60	6.9	0.54	12.3	2.22
	150	7.6	0.47	12.8	2.07
Least sig. deviation ($P = 0.05$)		N.S.[d]	0.09	N.S.	0.26

[a] $(NH_4)_2SO_4$.
[b] KH_2PO_4.
[c] K_2SO_4.
[d] N.S. not significant.

TABLE VIII. EFFECT OF DIFFERENT NITROGEN AND POTASSIUM CARRIERS ON THE UPTAKE OF ^{89}Sr BY MAIZE PLANTS GROWN ON BLACK AND LATERITE SOILS

Treatment[a]	Black soil ^{89}Sr uptake (nCi/g dry wt.)	Laterite soil ^{89}Sr uptake (nCi/g dry wt.)
Ammonium sulphate	0.24	2.19
Ammonium chloride	0.24	2.33
Sodium nitrate	0.16	
Urea	0.22	
Least significant deviation ($P = 0.05$)	N.S.	N.S.
Potassium sulphate	0.23	2.49
Potassium chloride	0.26	3.00
Least significant deviation ($P = 0.05$)	N.S.[b]	N.S.

[a] N at 150 kg/ha; K_2O at 150 kg/ha.
[b] N.S. not significant.

TABLE IX. MOVEMENT OF ^{89}Sr THROUGH BLACK SOIL BY SIMULATED RAIN, IRRIGATION WATERS AND EDTA TREATMENT

Depth of soil column (cm)	Distribution of ^{89}Sr in soil colum (% of total)						
	Field capacity H_2O	50 cm H_2O	50 cm 0.005$\underline{N}$ $CaCl_2$	250 cm H_2O	250 cm 0.005$\underline{N}$ $CaCl_2$	250 cm 0.005$\underline{N}$ NaCl	50 cm 0.01$\underline{M}$ EDTA
0 - 2.5[a]	72.6	70.5	73.9	64.0	35.3	62.4	54.2
2.5 - 5.0	26.6	28.9	26.1	35.6	50.0	37.1	44.8
5.0 - 7.5	0.8			0.4	12.8	0.6	1.1
7.5 - 10.0					1.8		
10.0 - 12.5					0.1		
12.5 - 15.0							

[a] Contaminated layer.

effects of increased concentrations of NH_4^+ or K^+ ions in the soil solution on the absorption of ^{89}Sr by plant roots and its subsequent transport to aerial tissues [22, 23].

1.4. Effects of rain, irrigation water and EDTA on leaching of radiostrontium

Tables IX and X report the data on the leaching of ^{89}Sr through black and laterite soils, respectively, by simulated rain and variable amounts of moderately high calcium and sodium irrigation waters and EDTA treatment.

Considerably greater downward movement of the radionuclide was observed with moderately high calcium irrigation water as compared to that with either moderately high sodium irrigation water or simulated rain. These findings are in close agreement with those of Miller and Reitemeier [24] and Koloskov et al. [25], whose studies have demonstrated that of a number of cations examined calcium was the most effective for replacing strontium from soils. With the possible exception of Li^+, Na^+ has been reported to be poorest of the cations for displacing adsorbed Sr^{++}.

The greater movement of ^{89}Sr with EDTA can possibly be ascribed to the formation of stable chelates with strontium. Nishita and Essington [26] have also observed that leaching with Na-EDTA increased the movement of ^{89}Sr as compared with leaching with deionized water in contrasting soils.

The greater mobility of radiostrontium in the laterite profiles as compared with that in the black soil profiles could possibly be explained in terms of differences in the physico-chemical properties, such as cation exchange capacity, organic matter content, composition of exchangeable cations, the clay content and the predominant type of clay minerals between the two soil types.

1.5. Effects of flooding

The data presented in Table XI indicate no significant effects of flooding treatment on ^{89}Sr concentration in aerial tissues of rice plants grown in black and laterite soils. Data are available in the literature [27] to show that flooding causes increases in the concentration of the lighter alkaline earth ions Ca^{++} and Mg^{++} in the soil solution as a result of the solvent action of CO_2 and cation exchange reactions. While similar qualitative effects on the solubility of Sr^{++} ions may occur, our present data, as also those of Verfaillie et al. [28] on rice grown on the sandy alluvium of the Po valley in Italy, show no evidence of enhanced uptake of strontium by rice plants under submerged conditions. It is also evident that direct entry into the shoot base, which has been shown to be largely responsible for enhanced uptake of ^{137}Cs in aerial tissues of rice under the flooded regime [29], does not contribute significantly to radiostrontium uptake.

2. Studies on gamma emitters

2.1. Uptake from contrasting soils

Data on the accumulation of gamma-emitting fission products by maize and rice plants from the two contrasting soil types are presented in Table XII.

TABLE X. MOVEMENT OF ^{89}Sr THROUGH A LATERITE SOIL BY SIMULATED RAIN, IRRIGATION WATERS AND EDTA TREATMENT

Depth of soil column (cm)	Distribution of ^{89}Sr in soil column (% of total)						
	Field capacity H_2O	50 cm H_2O	50 cm 0.005$\underline{N}$ $CaCl_2$	100 cm H_2O	100 cm 0.005$\underline{N}$ $CaCl_2$	100 cm 0.005$\underline{N}$ NaCl	100 cm 0.01$\underline{M}$ EDTA
0 - 2.5[a]	51.9	53.8	52.3	50.5	15.7	41.7	21.3
2.5 - 5.0	34.6	43.1	47.2	34.4	40.7	50.2	20.4
5.0 - 7.5	11.8	3.1	0.5	12.9	37.6	7.7	27.1
7.5 - 10.0	1.7			2.2	5.7	0.46	22.6
10.0 - 12.5					0.3		7.2
12.5 - 15.0							1.4
15.0 - 17.5							

[a] Contaminated layer.

TABLE XI. EFFECT OF FLOODING ON THE UPTAKE OF ^{89}Sr BY RICE PLANTS GROWN ON BLACK AND LATERITE SOILS

Treatment	Black soil ^{89}Sr uptake (nCi/g dry wt.)	Laterite soil ^{89}Sr uptake (nCi/g dry wt.)
Control[a]	0.17	1.72
Flooding (5 cm)	0.15	1.58
Least significant deviation ($P = 0.05$)	N.S.[b]	N.S.

[a] Field capacity moisture status.
[b] N.S. not significant.

TABLE XII. UPTAKE OF ^{106}Ru, ^{144}Ce AND ^{137}Cs BY MAIZE AND RICE PLANTS GROWN ON BLACK AND LATERITE SOILS
Values are concentration factors × 10^{-3}

Radionuclide	Maize		Rice	
	Black	Laterite	Black	Laterite
^{106}Ru	73.4	29.7	24.9	26.1
^{144}Ce	3.4	74.5	5.1	45.0
^{137}Cs	231.0	2274.0	241.9	626.0
Least sig. deviation ($P = 0.05$)	76.6	688.0	51.2	128.4
Least sig. deviation ($P = 0.05$) excluding ^{137}Cs	6.5	27.3	19.4	7.2

$$\text{Concentration factor} = \frac{\text{Radionuclide content / g plant shoot}}{\text{Radionuclide content / g soil}}$$

Data show that ^{137}Cs accumulation by both plant species was several-fold greater than that of the other gamma emitters in both the black soil and the laterite. Furthermore, the uptake of ^{137}Cs by plants growing in the laterite soil was considerably greater than in the black soil.

Considerable evidence exists to suggest that the availability of caesium to plants is markedly influenced by the type and content of clay mineral present in the soil [30-32]. Micaceous minerals of the 2:1 layer type such as illite, vermiculite and montmorillonite have been shown to fix caesium particularly effectively; by comparison, the 1:1 layer type minerals, namely, kaolinite and halloysite do so to a much smaller extent [30, 32]. It is well established that montmorillonite and illite are the predominant clay minerals in the black soils of India, while kaolinite is the principal alumino-silicate form in the laterite soils [10]. The differences in ^{137}Cs uptake from the black and laterite soils observed in the present study could, therefore, be attributed largely to greater fixation of the nuclide in the black soil, thereby rendering it relatively unavailable to the plant.

The data on relative uptake of the other gamma emitters (Table XII) show that in the laterite ^{144}Ce accumulation by maize and rice far exceeded that of ^{106}Ru. In the black soil, however, the uptake of ^{106}Ru was significantly higher than that of ^{144}Ce. The present findings on the enhanced ^{144}Ce uptake in the laterite are compatible with the report of Nishita et al. [33] that plant absorption of this radionuclide from acidic soils is greater than from soils with a high pH. Furthermore, it has been demonstrated in laboratory experiments that ^{144}Ce retention on 2:1 layer type minerals such as bentonite is considerably more than on kaolinite, which is a 1:1 layer type mineral [34].

Our findings indicate a greater availability of ruthenium to plants in the black soil than in the laterite. While previous work has indicated that ^{106}Ru absorption by plants varies widely between soil types, its uptake has been shown to be relatively greater from soils of high pH and exchangeable calcium [34, 35]. However, unlike other radionuclides examined in the present study, ^{106}Ru can be present in the anionic forms [36] and hardly any information is available on its chemical state in soils [34, 37].

2.2. Effects of organic matter

Data on the effects of organic matter addition on plant uptake of ^{106}Ru, ^{144}Ce and ^{137}Cs in the black and laterite soil are reported in Table XIII. Additions of organic matter did not significantly affect the plant uptake of ^{106}Ru and ^{144}Ce in the two soils. However, in the case of ^{137}Cs there was a reduction in the uptake in both soils, being highly significant in the laterite soil.

It is well documented that the exchangeable potassium level in soil markedly affects plant uptake of ^{137}Cs [38-40] as a result of competition between the two ions in the absorption and transport processes. It is suggested, therefore, that the observed reduction in ^{137}Cs uptake with application of organic matter may be, at least in part, due to the additional amounts of exchangeable potassium introduced with the added organic matter, which contained 6.41 meq K/100 g. Recent work of Evans and Dekker [41] indicates that in some mineral soils of Canada the addition of organic matter resulted in a lower ^{137}Cs uptake by oats and lettuce. The complexing action of organic matter constituents on radiocaesium [20] mey be yet another factor contributing to the reduced availability of this radionuclide for absorption by plants; similar effects on plant uptake of radiostrontium have been described in section 1.2. Our observations pertain to the effects of agronomic doses of organic matter applied to soils and are not directly comparable with those reported for soils in Sweden [42] and the United Kingdom [43], which show that the ^{137}Cs uptake by plants increased with increasing soil organic matter content.

2.3. Effects of flooding

Data on the effect of flooding treatment on the plant uptake of ^{106}Ru, ^{144}Ce and ^{137}Cs by rice grown in black and laterite soils are reported in Table XIV. Our data indicate that flooding treatment resulted in a

TABLE XIII. EFFECT OF ORGANIC MATTER APPLICATION ON THE UPTAKE OF ^{106}Ru, ^{144}Ce AND ^{137}Cs BY MAIZE PLANTS GROWN ON BLACK AND LATERITE SOILS

Values are concentration factors × 10^{-3}

Organic matter treatment[a] (g/kg soil)	Black			Laterite		
	^{106}Ru	^{144}Ce	^{137}Cs	^{106}Ru	^{144}Ce	^{137}Cs
0	73.4	3.4	231	29.7	74.5	2274
11.2	59.9	5.4	173	30.5	51.3	1112
22.4	69.6	5.1	174	21.9	44.1	637
Least sig. deviation (P = 0.05)	N.S.	N.S.	N.S.	N.S.	N.S.	748

[a] Vegetable compost — C/N = 9.7
Basal dressing — N: P_2O_5: K_2O = 60: 40: 60 kg/ha.

TABLE XIV. EFFECT OF FLOODING ON THE UPTAKE OF ^{106}Ru, ^{144}Ce AND ^{137}Cs BY RICE PLANTS GROWN ON BLACK AND LATERITE SOILS

Values are concentration factors × 10^{-3}

Treatment	Black			Laterite		
	^{106}Ru	^{144}Ce	^{137}Cs	^{106}Ru	^{144}Ce	^{137}Cs
Control[a]	26.9	7.2	95.8	8.8	27.3	321.3
Flooding (5 cm)	24.9	5.1	241.9	26.1	45.0	626.0
Least sig. deviation (P = 0.05)	N.S.[b]	N.S.	62.2	7.7	5.4	149.1

[a] Field capacity moisture status.
[b] N.S. not significant.

considerable increase in ^{137}Cs concentration in aerial tissues of the rice plant, amounting to 95 and 152% of the control in the laterite and black soils, respectively. These results confirm the findings of Myttenaere et al. [28, 29, 44], who have suggested that entry through the shoot base of ^{137}Cs present in the column of standing water is, in the main, responsible for the enhanced accumulation of this nuclide in rice shoots under the flooded regime.

While an increased ^{144}Ce content in shoots of rice grown under flooded regime in an alluvial sandy loam of the Po Valley in Italy has been reported [45], our data (Table XIV) on ^{144}Ce and ^{106}Ru, which show variable effects of flooding in the tropical black and laterite soils, suggest that altered redox potentials and other changes in the soil chemical regime brought about under submerged conditions [27] could differentially influence the solubility of these nuclides and their uptake by the rice plant in contrasting soil types.

In conclusion, it is evident that some of the soil amendment, fertilizer and irrigation treatments normal to agricultural practices in the black and laterite soil zones can have very marked effects on the quantitative transfer of long-lived fission products examined here to crop plants grown on these soil types. The findings reported in the present paper, as are those from related studies, are made available to the authorities responsible for release and surveillance of radioactivity levels in the environs of nuclear installations in the country. Such information could help to provide a realistic basis for determining permissible discharge limits for controlled releases of radioactive effluents into sources of waters used for agricultural operations. These data could also be of value in formulating plans for action in an emergency situation following accidental releases, as well as in more normal situations where controlled releases of effluents over a period of years could result in a build-up that approaches unacceptable levels of contamination. Our future work will include similar studies on other radionuclides, namely ^{3}H, ^{129}I and ^{85}Kr, which are of significance in the context of the quantities released from nuclear power stations or fuel-reprocessing plants.

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DISCUSSION

R.M. BARKHUDAROV: You referred, I understand, to the penetration of activity into well water. On what basis do you postulate the possibility of such contamination occurring?

K.B. MISTRY: The possibility is considered only in the context of an accidental large-scale release from the nuclear installation.

P. REINIGER: In the paper you cite work done by Myttenaere on the uptake of ^{137}Cs by rice. Now, his figures exceed yours by a factor up to 100, if I remember correctly, and I should be interested to hear your comments on this difference.

K.B. MISTRY: We referred to the work of Myttenaere in the context of significantly enhanced accumulation of ^{137}Cs in rice shoots under flooded conditions, as compared to accumulation under field capacity moisture status. I believe the high concentration factors that you mention pertain to plants grown in nutrient solution. In any event, the shoot concentration factors of ^{137}Cs for plants grown in soil would depend on the type of soil and on its physico-chemical characteristics, such as form and content of clay mineral, organic matter content and exchangeable potassium level. These are factors which are known profoundly to influence the uptake of ^{137}Cs by plants.

R.G. GUENNELON: I have two questions on this paper. First, do you consider that the organic matter figures used and mentioned in Table V represent the amounts normally employed as amendments, or do you think that much larger quantities (of the order of 100 t/ha) would be necessary in order, for example, to reduce the amount of Sr absorbed from 10 to 1?

Secondly, do the radiometric analyses of cultivated plants relate to the whole plant or to the edible parts thereof (as regards the effect of various forms of fertilizer). I am referring in particular to Table VIII.

K. B. MISTRY: The doses of organic matter indicated in Table V are those normally employed in agronomic practice in the regions to which the study relates. The question of using much higher doses of organic matter in order further to reduce radiostrontium uptake by crop plants would have to be carefully considered in the light of the economics of the matter and of the possible effects of the process on soil fertility. We have here confined our attention to the agricultural practices currently adopted in the regions concerned.

In reply to your second question, the data presented in the paper relate to the assay of the entire above-ground portion of the plants at the flowering stage. However, we have in other studies determined the distribution of the radionuclides in different plant organs, and one could extrapolate the present results to any specific aerial tissue.

C.A. MAWSON: With reference to the matter of concentration factors, I suggest that you determine these in the field, where crops have for many years past been irrigated with water containing fission products, in the form of fall-out.

K. B. MISTRY: Thank you for your suggestion. We shall certainly bear it in mind in our future work.

R.S. BRUCE: In your experiments on the effects of fertilizers on the uptake of radionuclides, were the fertilizers mixed into the soil or applied to the surface? If they are applied to the surface it is possible to envisage a situation in which plant roots proliferate in the surface zone, and this might lead to an increase in uptake of superficial contamination.

K. B. MISTRY: The fertilizers were incorporated in the soil in a manner that simulated the fertilizer placement procedure recommended for maize in black and laterite soil zones by the agricultural advisory services. In the case of both nitrogen and phosphorus this involves placement at a certain depth, and not application on the surface.

W.K.G. KÜHN: You mention briefly at the end of the paper that you intend to undertake similar studies on ^{3}H. Can you give any details of these plans?

K.B. MISTRY: No, I am afraid not. All I can do is repeat that, since ^{3}H is important in terms of the quantities released by nuclear power stations and fuel reprocessing plants, we may have to examine its behaviour in soil-plant regimes typical for India.

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ETUDES EXPERIMENTALES SUR LE TRANSFERT DANS LES CULTURES DE QUELQUES RADIONUCLEIDES PRESENTS DANS LES EFFLUENTS DES CENTRALES ELECTRO-NUCLEAIRES

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Abstract

EXPERIMENTAL STUDIES ON THE TRANSFER TO CROPS OF CERTAIN RADIONUCLIDES PRESENT IN NUCLEAR POWER STATION EFFLUENTS.

The use in crop irrigation of water contaminated by radioactive residues contained in nuclear power stations effluents entails a risk of the transfer of various radionuclides to man. In connection with such transfer of radioactive pollutants by water and plants to the human diet, the authors have attempted to define the relationship between the contamination rate of the water concerned and that of the crops harvested. To arrive at estimates that can be used by health experts, experimental models closely approximating actual conditions were utilized. A variety of fruit trees and various vegetables were contaminated by radionuclides (Cs, Co, Mn, Zn, Na) during genuine irrigation of the soil or of aerial parts of the plants. Cultivation was effected in large vats, in which the yields are comparable to those obtained in fields. An air-conditioned compartment was used for spraying. A distinction was maintained between direct and indirect contamination so that the respective hazards of the two pathways could be evaluated. Taking into account the various parameters of the environment, notably the quality of the water and the type of soil, it is intended on the basis of this work to assess the quantities of radioactive pollutants transferred through the water-soil-plant-diet chain in order to evaluate the consequences of any discharges into rivers.

ETUDES EXPERIMENTALES SUR LE TRANSFERT DANS LES CULTURES DE QUELQUES RADIONUCLEIDES PRESENTS DANS LES EFFLUENTS DES CENTRALES ELECTRO-NUCLEAIRES.

L'utilisation, pour l'irrigation des cultures, d'eau radiopolluée par les résidus d'effluents de centrales électro-nucléaires crée un risque de transfert de divers radionucléides jusqu'à l'homme. Dans le cadre de ces transferts de radiopolluants par l'eau et les plantes jusqu'à la ration alimentaire, les auteurs ont tenté de préciser les relations entre le taux de contamination des eaux et celui des denrées agricoles. Afin d'obtenir des estimations utilisables par les hygiénistes des modèles expérimentaux proches de la pratique ont été mis en œuvre. Une espèce fruitière et différents légumes ont été contaminés par des radionucléides (Cs, Co, Mn, Zn, Na) au cours de véritables irrigations appliquées au sol ou sur les organes aériens. Les cultures ont été conduites en bacs de grandes dimensions où les productions sont comparables à celles de plein champ. Un caisson climatisé a été utilisé pour distribuer les aspersions. Les contaminations, directe et indirecte, ont été dissociées afin d'évaluer les risques respectifs de ces deux modes d'apport. A partir de ces travaux, en tenant compte de divers paramètres du milieu, qualité des eaux et type de sol notamment, une estimation des quantités de radiopolluants transférés à travers le système eau-sol-plante-ration alimentaire est proposée pour l'évaluation des conséquences de rejets éventuels en rivière.

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* Les auteurs ont rédigé ce mémoire en collaboration avec H. KERNISAN, S. MARCHAND, C. TEMPIER et R. ESCARIOT (CEN Cadarache).

INTRODUCTION

L'accroissement du nombre de réacteurs de puissance produisant de l'électricité augmente les risques de pollution des eaux de rivière utilisées pour l'irrigation des cultures.

L'évaluation des niveaux de la contamination alimentaire qui peut en résulter est un des objectifs fondamentaux de la radioécologie.

Notre objectif est d'effectuer cette évaluation avec la double préoccupation de

- simuler au mieux les conditions de la pratique pour obtenir des valeurs représentatives des taux de transfert des polluants dans le milieu
- prévoir l'évolution de la contamination à long terme dans l'hypothèse d'un apport continu de polluant.

Nos précédentes expérimentations ont mis en évidence l'importance de l'irrigation par aspersion dans le transfert des radiopolluants [1, 2]. Ces essais, au cours desquels ont été utilisés des effluents de réacteurs (types à eau sous pression et à neutrons rapides) et d'une usine de traitement, ont montré l'existence d'un risque à court terme en ce qui concerne ^{60}Co, ^{54}Mn, ^{144}Ce, ^{131}I, ^{22}Na et ^{137}Cs par contact direct de ces radionucléides avec les denrées récoltées, et d'un risque à long terme à la suite d'une accumulation continue dans les sols; ces risques ont été précisés en ce qui concerne ^{90}Sr et ^{137}Cs [1, 2].

Dans le prolongement de ces travaux les nouveaux essais présentés dans cette communication ont pour objectif de chiffrer, dans des conditions proches de la pratique, les taux de transfert de ^{137}Cs, ^{54}Mn, ^{60}Co, ^{65}Zn et ^{22}Na, de l'eau d'irrigation aux parties consommables de trois légumes: la salade, le haricot vert et la carotte, ainsi que d'un arbre fruitier: le pommier.

Deux aspects sont envisagés séparément: la contamination directe et la contamination indirecte.

TABLEAU I. CARACTERISTIQUES DES ARROSAGES ET DE L'EAU

Arrosages	Eau		
	Caractéristiques physico-chimiques	Activité spécifique (μCi/l)	
Dose: 50 mm	pH: 7,7	^{60}CoCl$_2$	0,297 ± 0,041
Densité: 10 mm/h	Résidu sec: 0,320 g/l		
	Résistivité: 2200		
Nombre:	Ca: 70 mg/l	^{60}Co EDTA	0,260 ± 0,002
– légumes, 3	Mg: 6 mg/l		
– pommiers, 8	Na: 11 mg/l	^{54}Mn	0,154 ± 0,04
	K: 2,3 mg/l		
Espacement entre aspersions: 15 j	Bicarbonate: 16,8 mg/l	^{65}Zn	0,405 ± 0,096
	Chlorure: 25,1 mg/l		
	Sulfate: 59,4 mg/l	^{22}Na	0,342 ± 0,031
	Nitrate: 4,9 mg/l		
		^{137}Cs	0,29 ± 0,033

1. CONTAMINATION DIRECTE

1.1. Méthodes

L'objectif de ces essais est de préciser les taux de transfert des radiopolluants cités plus haut, de l'eau aux denrées consommables, qui résultent du contact direct des eaux polluées avec les organes aériens des plantes.

On a cherché à déterminer le niveau possible de cette contamination ainsi que ses variations sous l'effet de quelques-uns des paramètres principaux.

Pour cette évaluation, des aspersions qui simulent l'essentiel des conditions de la pratique ont été appliquées en caisson-phytotron sur des pommiers, des salades, des haricots verts et des carottes se développant en bacs d'un mètre carré.

Ces dispositifs ont été décrits dans d'autres communications [1, 2]. A la pratique ont été empruntées la qualité de l'eau et la dose, la durée, la densité d'arrosage; une feuille de polyvinyle isolait le sol de l'aspersion (sauf dans le cas des carottes) (tableau I).

1.2. Taux de transfert

Le tableau II présente les taux de transfert a pour les denrées à la récolte, après les arrosages: a = (pCi/kg frais)/(pCi/litre d'eau). Un kg de produit frais fixe une quantité de radioélément équivalente à celle contenue dans a litre d'eau.

Les moyennes des taux de transfert varient de 0, 2 à 0, 7 pour les différents radionucléides utilisés. Un kg de denrée serait contaminé approximativement comme 0, 6 litre d'eau d'arrosage.

TABLEAU II. TAUX DE TRANSFERT DE ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{22}Na, ^{60}Co DE L'EAU DANS LES PARTIES COMESTIBLES DE TROIS LEGUMES ET UN FRUIT
(Contamination directe)
(pCi/kg frais)/(pCi/litre d'eau)

	Volume d'eau distribué (litre/m^2)	^{137}Cs	^{54}Mn	^{65}Zn	^{22}Na	^{60}Co
Haricot (gousses)	150	0,73	0,04	0,12	0,07	0,72
Salade	150	1,5	0,75	1,6	0,9	1,57
Carotte	150	0,4	0,0	0,5	1,7	0,4
Pomme	400	0,15	0,05	0,15	0,07	0,08
Moyenne approchée		0,7	0,2	0,6	0,7	0,7

TABLEAU III. TAUX DE TRANSFERT DU ^{60}Co COMPLEXE AVEC EDTA ET SOUS FORME DE $CoCl_2$ DE L'EAU DANS LE HARICOT ET LA SALADE
(Contamination directe)
(pCi/kg frais)/(pCi/litre d'eau)

	Eau déminéralisée		Eau de rivière	
	$^{60}CoCl_2$	^{60}Co EDTA	$^{60}CoCl_2$	^{60}Co EDTA
Salade	4,9	1,65	1,57	0,8
Haricot (feuilles)	10,0	5,4	3,5	0,6

Ces taux diffèrent avec les espèces; la salade, légume-feuille, est la plus contaminée. C'est le cas des feuilles des quatre espèces étudiées. Les carottes, pour lesquelles aucune protection du sol n'avait été réalisée, sont également assez contaminées.

Les taux de transfert sont sensiblement équivalents pour ^{137}Cs, ^{65}Zn, ^{22}Na et ^{60}Co ($CoCl_2$). ^{54}Mn est le moins bien fixé par les plantes.

Ces taux de transfert sont conditionnés entre autres par la nature et la qualité de l'eau ainsi que par la forme physico-chimique du radionucléide [3]. Une fraction importante des radionucléides dans le milieu peut en effet se trouver à l'état complexé à la suite de l'utilisation d'agents de décontamination dans les installations nucléaires et de produits complexants présents dans l'eau. Les taux de fixation par des salades de ^{60}Co, sous les deux formes physico-chimique ($CoCl_2$) et complexe (à l'EDTA), dans l'eau déminéralisée et l'eau de rivière, ont été recherchés (tableau III).

Le cobalt complexé avant son introduction dans l'eau est plus mobile et moins bien fixé par les végétaux que le chlorure de cobalt. Cette technique de décontamination des installations joue par conséquent un rôle favorable dans ce mode de transfert. Les éléments dissous dans l'eau concurrencent les deux formes de cobalt; ils contribuent également à faire baisser les taux de fixation du ^{60}Co sur les végétaux.

Ces valeurs devraient être augmentées dans les cas d'arrosages plus fréquents. Par exemple, des cultures de tomate et de pomme de terre en région méditerranéenne nécessitent respectivement des quantités d'eau d'appoint de 8500 et 7000 m^3/ha. En effet nous avons constaté que les activités spécifiques des feuilles de pommier augmentent proportionnellement au nombre d'arrosages; on peut donc s'attendre a priori, avec des aspersions 3 à 4 fois plus abondantes (500 l/m^2), à des taux de fixation 3 à 4 fois plus élevés.

Par contre, une fois lavées les feuilles de pommier perdent à peu près 40% de leur activité.

Aussi, en tenant compte d'une quantité d'eau d'appoint moyenne de 500 mm, on peut admettre un taux de transfert moyen des radionucléides de l'eau aux denrées de 1 comme plausible pour le type d'eau considéré.

2. CONTAMINATION INDIRECTE

2.1. Méthode de travail

L'arrosage répété des cultures avec une eau radiopolluée crée un risque d'accumulation des polluants dans les sols. Pour la prévision des taux de contamination des denrées à la limite de cette accumulation, Barbier et coll. [4] ont préconisé un modèle mathématique que nous utiliserons pour la suite de notre exposé:

$$r = \frac{c\ d}{\lambda + \varphi}$$

r signifie que 1 kg de la denrée considérée contient autant de radioélément que r litre d'eau

d est la hauteur d'eau annuelle d'irrigation en litres par m^2 et par an

λ est le coefficient de décroissance radioactive, en an^{-1}

φ est la fraction de radionucléide perdue annuellement par infiltration

c coefficient de prélèvement des plantes, est le rapport entre la quantité de radioélément par kg de produit récolté et la quantité de radioélément par m^2.

Dans ces nouvelles études nous nous sommes préoccupés d'évaluer le taux de contamination possible à long terme pour la salade, le haricot, la carotte, la pomme par ^{137}Cs, ^{60}Co, ^{54}Mn, ^{65}Zn, ^{22}Na. Cette évaluation a été recherchée en deux temps: d'abord sur un sol pris comme référence, puis un essai de généralisation a été tenté à partir d'observations sur six types de sols. La nécessité, pour une bonne simulation, de travailler sur un modèle expérimental très voisin de la réalité pour la détermination des variables c et φ nous a conduits à l'emploi de bacs de végétation de 1 m^2 de surface contenant une couche de sol de 50 cm permettant la mise en œuvre de techniques proches de la pratique.

Les chlorures de ^{137}Cs, ^{22}Na, ^{65}Zn, ^{54}Mn, ^{60}Co ont été appliqués au sol, en une seule fois, dans des bacs contenant un sol limoneux; ils ont été mélangés au sol, avant les essais, sur une épaisseur de 20 cm, avec un instrument aratoire.

Les arrosages ont été déclenchés pour une humidité du sol à 70% de la capacité au champ. Après la récolte des plantes, 400 litres d'eau déminéralisée ont été appliqués afin de simuler les chutes de pluies d'hiver; les contaminations au cobalt ont été faites séparément. Les recherches sur le facteur «sol» ont été effectuées sur des plants de salade cultivés en pots de 1 kg (quatre répétitions). Les caractéristiques des sols figurent au tableau IV.

2.2. Essais sur sol de référence

2.2.1. Contamination des parties comestibles

Sur le tableau V sont présentés des taux de contamination des parties comestibles prélevées sur les végétaux cultivés sur le sol de référence

TABLEAU IV. CARACTERISTIQUES DES SOLS

	Sol de Référence Limon 1	Limon 2	Brun de Lehm	Brut de sable dunaire	Brun	Hydromorphe Calcimorphe
Argile (%)	17	9	8	4	12	20
pH eau	8,1	8,7	7,9	8,2	7,6	8,2
Calcaire total (%)	23	43	trace	15	7	50
Calcaire actif (%)	1,8	17	-	3	0	19
P (ppm)	16	26	125	198	690	125
Ca (meq/100 g)	59,6	53,8	24,6	45,1	36,3	63,6
K (meq/100 g)	0,52	0,24	0,69	0,27	0,76	0,70
Na (meq/100 g)	0,18	0,17	0,15	0,15	0,20	0,17
N nitrique (ppm)	99	19,5	38,5	21,5	100	76,5
C (%)	0,78	0,91	0,95	1,28	3,01	3,54
N (%)	0,09	0,09	0,09	0,15	0,26	0,31
Mn BITTEL (ppm)	99	23	64	19	70	17
Co extrait acide acétique 0,5N (ppm)	0,79	1,74	0,25	0,67	0,03	0,47
Zn extrait HCl 0,1N (ppm)	0,36	0,96	18	16	84	3,6

(sol 1) au cours de la première année d'observation. Ces taux sont très différents selon les denrées considérées et les radionucléides.

Aucune activité importante n'a été décelée dans les pommes au cours de cette première année. Par contre on constate dans les trois légumes des taux de contamination relativement élevés pour ^{22}Na, assez élevés pour ^{65}Zn, faibles pour ^{60}Co et ^{137}Cs, non décelables pour ^{54}Mn. Le ^{65}Zn s'accumule davantage dans les parties supérieures. L'écart est très grand entre les taux de contamination pour Na: minimal pour les gousses de haricot et maximal pour les salades. Ces différences semblent pouvoir être reliées aux teneurs en Na et K des diverses parties des végétaux (tableau VI).

2.2.2. Pertes par infiltration

Les arrosages ont été distribués de façon à humidifier jusqu'à la capacité au champ la couche de sol utilisée. Cette technique d'arrosage, courante dans la pratique, a peu fait migrer ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co. Par contre ^{22}Na a migré de façon plus importante.

Sous l'action des arrosages à l'eau déminéralisée, simulant les pluies d'automne et d'hiver, aucune quantité significative supplémentaire de ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co n'a été entraînée en dehors de la zone d'incorporation.

Les pertes par infiltration de ^{22}Na sont très élevées. Il est intéressant de relever cependant que 400 mm d'eau ne sont pas totalement éliminés de la zone d'apport: 17% y sont restés fixés.

2.2.3. Taux de contamination des parties comestibles sur le sol de référence

Ces résultats appliqués au modèle mathématique permettent de faire une première estimation du taux de contamination moyen des trois légumes à la limite d'accumulation des radionucléides dans le sol de référence (tableau VII). Un kg de légume frais contiendrait autant de radionucléide que r litre d'eau.

On peut considérer ces valeurs comme plausibles bien qu'elles aient été obtenues après une première année d'essais.

En particulier, la répartition dans le sol est suffisante pour ne pas provoquer une sous-estimation des niveaux de contamination; d'une part, dans la couche de sol où les radionucléides ont été apportés, les bonnes conditions d'aération, d'alimentation en eau et en éléments nutritifs attirent le maximum de racines; d'autre part, les radionucléides récemment incorporés sont restés à la surface des mottes et de ce fait sont plus absorbés qu'après dissémination dans toute la masse du sol et diffusion dans les particules.

En ce qui concerne φ, l'hypothèse d'une perte nulle par infiltration conduirait à majorer r pour Co, Cs, Mn, Zn, respectivement de 16%, 20%, 9%, 5%, les valeurs moyennes obtenues. Ces majorations sont petites par rapport aux variations des coefficients de prélèvement des plantes en fonction des espèces. Aussi, pour une première estimation moyenne, peut-on les négliger. La fraction de ^{22}Na non éliminée peut sembler élevée; Na est connu comme très lixiviable et, si l'on admet qu'il peut être totalement éliminé, nous commettons une erreur par excès de 16%. Nous négligeons également cette valeur pour la même raison que pour Co, Cs, Mn, Na, d'autant qu'une élimination totale du ^{22}Na par les pluies n'est pas assurée.

2.3. Influence du type de sol

Pour tester la représentativité de ces valeurs, nous avons recherché l'amplitude de variation du taux de contamination de la salade sur six types de sols différents (tableau VIII).

Le type de sol apparaît comme un facteur important de variation du taux de transfert des radionucléides à la plante. Les amplitudes de variation du taux de transfert sont comprises entre

1 et 6 pour Cs
1 et 2 pour Co, Zn et Na
ϵ, 1 et 3 pour Mn.

TABLEAU V. CONTAMINATION INDIRECTE ET CARACTERISTIQUES DES PARTIES COMESTIBLES DE TROIS LEGUMES ET UN FRUIT

	Taux de contamination $c_1 \cdot 10^3 = \frac{\text{pCi/kg frais}}{\text{pCi/m}^2 \text{ de sol}}\ 10^3$					Caractéristiques chimiques de la matière sèche des parties comestibles					
	^{137}Cs	^{54}Mn	^{65}Zn	^{22}Na	^{60}Co	Ca (%)	K (%)	Na (%)	Co (ppm)	Zn (ppm)	Mn (ppm)
Haricot vert	0,06	ND	8,9	0,3	0,9	0,77	2,5	0,01	46	46	24
Salade	0,10	ND	14	170	1,2	2,34	6,11	0,82	0,79	21	71
Carotte	0,02	ND	2	130	1,6	0,54	3,51	0,60	12,2	20	20
Moyenne	0,06	-	8,3	100	1,3						
Pomme	0,01	ND	ND	0,1	0,02	0,05	1,18	0,01	0,34	3	6

ND = non décelable.

TABLEAU VI. TENEURS EN Na ET K ET CONTAMINATION EN ^{22}Na

	Haricot		Salade	Carotte		Pommier	
	gousses	feuilles	feuilles	racines	feuilles	fruits	feuilles
$^{22}\mathrm{Na}\ \left(\frac{\mathrm{pCi/kg\ frais}}{\mathrm{pCi/m^2\ de\ sol}}\ 10^3\right)$	0,3	2	170	130	84	0,1	ND
Na (% matière sèche)	0,01	0,03	0,82	0,6	0,49	0,01	0,02
K (% matière sèche)	2,50	2,28	6,11	3,51	3,85	1,18	1,34

TABLEAU VII. CONTAMINATION DES PARTIES COMESTIBLES A LA LIMITE D'ACCUMULATION DES RADIONUCLEIDES DANS LE SOL DE REFERENCE

		^{137}Cs	^{54}Mn	^{65}Zn	^{22}Na	^{60}Co
Pertes par infiltration.	Après irrigation, 230 mm	0,005	0,05	0,03	0,21	0,02
	Après pluies simulées, 400 mm	0,005	0,05	0,03	0,83	0,02
Taux de transfert $\frac{\mathrm{pCi/kg\ frais}}{\mathrm{pCi/litre\ d'eau}}$	Haricot vert	1,1	-	6,2	0,2	3,2
	Salade	2	-	10,5	80	4,3
	Carotte	0,4	-	1,51	61	5,7
	Moyenne	1,1	-	6	47	4,4

Certaines de ces variations peuvent être reliées à des facteurs particuliers. C'est ainsi que la contamination en césium baisse quand la teneur en argile croît. Sur sol pauvre en Ca total, actif et échangeable, la contamination en ^{22}Na augmente.

L'absence de contamination en ^{54}Mn sur le sol limoneux 1 est une exception qui peut être expliquée par un passage de Mn sous forme d'hydroxyde de manganèse. En fait la contamination de la salade par Mn est du même ordre de grandeur que la contamination par Zn pour l'ensemble des sols.

En ce qui concerne φ, dans le cas d'une évaluation moyenne du taux de prélèvement des plantes il n'est pas absolument nécessaire de rechercher sa variation en fonction des différents types de sols. En effet c et φ varient dans le même sens en fonction de leur principale cause de variation, la capacité d'échange des sols. La connaissance de φ peut être, par contre, très intéressante dans certains cas particuliers, notamment dans celui d'une pollution possible des nappes. Aussi, pour une meilleure représentativité de r vis-à-vis de cet échantillonnage de sol, certaines corrections peuvent être apportées aux valeurs précédentes.

TABLEAU VIII. CONTAMINATION DES SALADES PAR ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{22}Na, ^{60}Co SUR SIX SOLS DIFFERENTS

	$c_2 = \frac{(\text{pCi/kg frais})}{(\text{pCi/kg de sol})}$				
	^{137}Cs	^{54}Mn	^{65}Zn	^{22}Na	^{60}Co ($^{60}CoCl_2$)
Limon 1	0,00024	0	0,038	0,53	0,0026
Limon 2	0,00018	0,052	0,031	0,4	0,0022
Brun de Lehm	0,00090	0,018	0,026	0,63	0,0031
Brut de sable dunaire	0,00110	0,026	0,017	0,5	0,0036
Brun	0,0007	0,030	0,025	0,66	0,0047
Hydromorphe calcimorphe	0,00020	0,037	0,030	0,36	0,0034
c_2 moyen	0,00055	0,027	0,028	0,51	0,0032
$\frac{c_2 \text{ moyen}}{c_2 \text{ sol de référence}}$	$\frac{2,3}{1}$	$\frac{0,27}{\epsilon}$	$\frac{0,73}{1}$	$\frac{1}{1}$	$\frac{1,23}{1}$
Amplitude	1-6,1	ε-1-2,9	1-2,2	1-1,8	1-1,8

TABLEAU IX. TAUX DE TRANSFERT A LONG TERME DES RADIONUCLEIDES DE L'EAU DANS LES PARTIES COMESTIBLES DES LEGUMES obtenus à partir de six sols différents et de trois espèces (haricot vert, salade et carotte) pour ^{137}Cs, ^{65}Zn, ^{60}Co, ^{22}Na, et une espèce (la salade) pour ^{54}Mn
(hauteur d'eau annuelle d'irrigation = 500 mm)
(pCi/kg frais)/(pCi/litre d'eau)

	^{137}Cs	(^{54}Mn) ^{65}Zn	^{60}Co	^{22}Na
Contamination indirecte	2,5	5,3	5,4	47
Contamination directe	1			
Total	3,5	6,3	6,4	48

CONCLUSION

Le taux de transfert moyen des radionucléides étudiés de l'eau dans les denrées comestibles végétales, évalué sur trois légumes et six sols différents, est compris entre 3, 5 et 6, 4 pour ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co; il est de 48 pour ^{22}Na (tableau IX).

Ces taux, nous l'avons vu, peuvent être largement dépassés dans certains cas, en fonction de la nature du végétal et des paramètres du milieu.

Par exemple sur le limon de référence 1 kg de salade peut contenir, à plus ou moins long terme, autant de radionucléides que 80 litres d'eau ou autant de Zn que 10 litres d'eau. Lorsque le pH du sol baisse, les plantes peuvent accumuler des quantités de ^{54}Mn plus importantes [5, 6]. Sur des sols acides, Zn peut évoluer vers des formes plus disponibles [7].

Par ailleurs la forme du radionucléide, avant son rejet dans le milieu, et la qualité de l'eau peuvent modifier le taux de transfert. La complexation du cobalt et du zinc à l'EDTA diminue leur fixation sur les sols et facilite leur absorption sur les végétaux [8]. Cependant ces produits complexés sont également plus mobiles. Les pertes par infiltration sont alors plus importantes et modèrent l'élévation du taux de transfert à long terme. Saas a mis en évidence la mobilité de ces radionucléides véhiculés par les eaux de rivière chargées en matière complexante. Pour l'instant ces taux de transfert peuvent être considérés comme plausibles.

Il ressort de ces observations que les plantes concentrent dans leurs tissus, en particulier dans les parties consommables, les radiopolluants qui leur sont apportés par les eaux d'arrosage. Au cours des aspersions, deux modes de contamination se superposent: la contamination directe, la contamination indirecte.

Les taux de contamination des denrées, qui résultent de leur contact direct avec l'eau d'arrosage, ne sont pas en moyenne très sensiblement supérieurs aux taux de contamination des eaux.

Le risque principal provient de l'accumulation dans les sols des radionucléides, à la suite d'arrosages répétés pendant plusieurs années. Les cinq radionucléides étudiés peuvent être groupés dans deux classes selon le niveau des taux de transfert:

- taux de transfert moyen compris entre 3, 5 et 6, 5: ^{137}Cs, ^{65}Zn, ^{60}Co et ^{54}Mn
- taux de transfert moyen de 48: ^{22}Na.

Le risque maximal pour la production maraîchère, et sans doute pour l'ensemble des cultures irriguées, peut donc résulter de la dispersion de ^{22}Na dans le milieu. Une surveillance sévère doit être exercée sur les végétaux des sites nucléaires où l'irrigation des cultures est pratiquée. Cette surveillance s'avère particulièrement nécessaire en ce qui concerne ^{22}Na, dans l'éventualité où les réacteurs rapides pourraient être à l'origine d'une dispersion de ce radionucléide dans le milieu.

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DISCUSSION

R. KIRCHMANN: On the subject of direct contamination, have you any experimental data on the percentage of the radionuclides studied that is retained by vegetables such as lettuces and French beans after washing for purposes of culinary preparation?

J. DELMAS: Speaking of the radionuclides covered by this study, about 40% of the amount retained on lettuces and beans irrigated by sprinkling are removed by washing. Our laboratory is at present running a program of tests to determine the effects of these operations and also the effects of food processing on the contamination levels of these commodities. It appears that processes such as bleaching and sterilization bring about a reduction in contamination levels. Test programs of this type represent essential supplementary data for estimating the quantities of radiopollutants that reach the consumer.

R.M. BARKHUDAROV: Have you devoted any attention to the accumulation of isotopes in milk, which, thanks to its capacity for isotope concentration, can constitute a 'critical product'?

J. DELMAS: Our laboratory is situated in a predominantly vegetable and fruit-growing area, and hence we were concerned primarily with these products. I agree that the chain water-fodder-milk can constitute a critical pathway, and similarly, for example, the large volume of water required in maize growing may be the cause of high transfer coefficients. At all events we have observed this with maize in the case of ^{90}Sr and ^{137}Cs, but for more details on the subject I think we should turn to Mr. Kirchmann.

R. KIRCHMANN: The experiments simulating direct (rain-water) contamination of pastureland carried out at the Belgian CEN's experimental farm did not in fact cover irrigation by sprinkling, which is only now beginning to establish itself as a technique in the northern countries. However, coefficients for grass-milk transfer, after deposition of radionuclides in solution, have been obtained in respect of ^{131}I, ^{134}Cs, ^{85}Sr, ^{103}Ru and ^{3}H (THO). The results have been published in the literature.

A. SPANG: Your results reveal a specific activity of about 10^{-4} μCi/ml in the irrigation water. This is much more than commercial nuclear power reactors release with their liquid waste discharges into a river system. Do you not fear that extrapolation to lower or higher specific activities may be attended by gross errors?

J. DELMAS: Like all models, the experimental models used here are only an approximation of the real situation. It is therefore necessary to compare the experimental results with field observations. In this connection our laboratory is performing radioactivity measurements on samples of water, soil, and plants obtained in the Rhone basin where a number of nuclear facilities are situated. A comparison of the two sets of results is no easy matter because the activities of the field samples are very low, but there is nevertheless evidence that, on average, the methods of simulation we are discussing do give a good approximation of transfer coefficients.

COMPORTEMENT DANS LES MILIEUX SOUTERRAINS DE L'URANIUM ET DU THORIUM REJETES PAR L'INDUSTRIE NUCLEAIRE

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Abstract–Résumé

THE BEHAVIOUR IN UNDERGROUND ENVIRONMENTS OF URANIUM AND THORIUM DISCHARGED BY THE NUCLEAR INDUSTRY.

Following various studies on the radioactive contamination of soil by fission products from a variety of effluents, it has been found necessary also to establish in what way long-lived heavy radionuclides could be diffused in underground environments. A proposal is made for the study of the behaviour in the soil of uranium and thorium originating from outside, as compared with the uranium and thorium existing as natural constituents of the soil. Use was made of various types of soil (acid and alkaline) and of various minerals (argillaceous minerals, calcite and quartz). Uranium in solution, or uranyl ion to be more precise, has a certain degree of mobility in soils. The coefficient for the distribution of uranyl ion between the solution and the mineral was measured at various concentrations. The influence of the pH of the soil-water solution is very considerable; the retention of uranium by the soil shows a marked increase within certain narrow pH ranges. Thorium is subject to high retention by argillaceous and calcareous soils, some of the latter strongly fixing the whole amount. The influence of various parameters (pH, concentration, soil composition) is also substantial, as retention can be very much reduced under certain extreme conditions. The risk of the spread of thorium in dilute solution over long distances is negligible in most soils. On the other hand, a certain degree of uranium diffusion under the influence of ground-water movements is to be anticipated; diffusion evaluations are presented.

COMPORTEMENT DANS LES MILIEUX SOUTERRAINS DE L'URANIUM ET DU THORIUM REJETES PAR L'INDUSTRIE NUCLEAIRE.

Après des études variées sur la contamination radioactive du sol par les produits de fission provenant d'effluents divers, il est apparu nécessaire de savoir aussi comment pourraient se propager dans les milieux souterrains les radioéléments lourds à vie longue. On propose une étude du comportement dans le sol de l'uranium et du thorium provenant d'un apport extérieur, par opposition au comportement de l'uranium et du thorium considérés en tant que constituants naturels du sol. Divers types de sols (acides, alcalins) ainsi que divers minéraux (minéraux argileux, calcite, quartz) ont été utilisés. L'uranium en solution, ou plus exactement l'ion uranyle, présente une certaine mobilité dans les sols. Le coefficient de distribution de l'ion uranyle entre la solution et le minéral a été mesuré à diverses concentrations. L'influence du pH de la solution sol-eau est très importante, la rétention de l'uranium par le sol augmente fortement dans certaines bandes étroites de pH. Le thorium est très fortement retenu par les sols argileux et calcaires, certains sols pouvant le fixer en totalité de façon très énergique. L'influence de divers paramètres (pH, concentration, composition du sol) est aussi très importante car la rétention peut devenir très réduite dans certaines conditions extrêmes. Le risque de propagation du thorium en solution diluée sur une longue distance est négligeable dans la plupart des terrains, par contre on peut prévoir une certaine diffusion de l'uranium sous l'influence des mouvements d'eaux souterraines; des évaluations de cette diffusion sont données.

1. INTRODUCTION

L'uranium et le thorium sont disséminés à l'état de traces dans diverses roches éruptives et sédimentaires avec des teneurs de quelques ppm; le rapport Th/U est variable mais, en moyenne, l'uranium est moins abondant [1] [2].

L'importance de ces deux éléments a justifié d'abondantes études géochimiques qui ont fait ressortir leurs comportements différents dans les sols sous l'effet du lessivage par les eaux souterraines : [3] [4] [5] [6].

Le but de cette étude est différent. On a voulu étudier, non pas le comportement de ces éléments en équilibre séculaire dans leurs états naturels, mais le comportement de l'uranium et du thorium rejetés par l'industrie nucléaire à des concentrations et sous des états différents de leurs formes géochimiques habituelles.

La contamination du sol par l'uranium et par le thorium peut avoir plusieurs origines : les mines, les usines de traitement ou de transformation des minerais, les installations d'élaboration des métaux ou de fabrication des combustibles.

Enfin, dans bien des cas, les déchets contiennent encore des proportions non négligeables de ces radioéléments et leur stockage peut entraîner une contamination du sol.

Il a paru intéressant d'étudier les mécanismes de rétention de ces radioéléments sur les sols et les minéraux afin d'évaluer les risques d'extension de cette pollution aux nappes aquifères. Plusieurs facteurs (nature du sol, nature de l'effluent ...) interviennent évidemment dans la migration de ces ions dans les milieux poreux souterrains.

2. LES SOLS ETUDIES

Les sols sédimentaires meubles communs sont essentiellement constitués de silice, de calcite, de minéraux argileux et de matière organique en proportions variables, les autres constituants (oxydes, silicates, divers) étant accessoires.

On a choisi quatre types de sols :

sol 1 ensemble silice-argile-calcaire-matière organique : alluvion de l'Essonne (Bassin parisien);

sol 2 tourbe (très riche en matière organique) : Essonne;

sol 3 ensemble silice-argile-calcaire (sans matière organique) : sédiment vindobonien de Cadarache (Provence);

sol 4 ensemble silice-argile (sans calcaire ni matière organique) : schiste altéré de La Hague (Cap de La Hague).

Les proportions relatives de ces différents constituants sont indiquées dans le tableau I.

3. LE COEFFICIENT DE DISTRIBUTION DES RADIOELEMENTS ENTRE L'EAU ET LA PHASE SOLIDE

Pour évaluer la rétention plus ou moins forte des radioéléments par les divers matériaux, on mesure le coefficient de distribution (kd) du radioélément entre la solution et la phase solide [7].

TABLEAU I. COMPOSITION DES SOLS ETUDIES (% EN POIDS DE SOL SEC)

SOL	ORIGINE	SiO_2	Argile	Calcium	M.O.
1	Alluvion de l'Essonne	23	20	45	12
2	Tourbe	5	12	23	60
3	Sédiment de Cadarache	45	30	25	< 1
4	Schiste altéré de La Hague	40	60	0	0

Des mesures ont aussi été faites sur des matériaux purs :

Sable de Fontainebleau : silice pure
Calcaire blanc de Meudon : $CaCO_3$ pur
et sur une argile pure : l'illite.

Lorsqu'une solution contenant un corps dissous à la concentration Co est mise en contact avec un matériau, un équilibre s'établit et la concentration devient alors $C \leqslant Co$ dans la solution et q dans la phase solide; par définition $Kd = \frac{q}{C}$.

La quantité d'élément qui a quitté le volume V de solution est passée dans la masse m solide, donc $V\,(Co - C) = mq$

ou $Kd = \frac{q}{C} = \left(\frac{Co}{C} - 1\right)\frac{V}{m}$ formule utilisée pour mesurer le Kd.

Le Kd ne fait pas intervenir le mode de rétention qui peut être dû à un phénomène d'échange d'ion, à une adsorption électrostatique ou aussi au passage de l'élément en phase solide par précipitation.

Comme le Kd varie avec la nature de la solution, on a, dans le cas des sols naturels, opéré avec l'eau naturelle du site sur lequel ils ont été prélevés (eau de la rivière Essonne pour les sols 1 et 2, eau de nappe pour les sols 3 et 4).

La dimension du Kd est exprimée en cm3/g.

4. COMPORTEMENT DE L'URANIUM

4.1. Quelques rappels de la chimie de l'uranium

L'uranium se rencontre le plus souvent à sa valence 6^+. Les cations uraneux U^{4+} sont rares, en solution ils sont instables ou insolubles. U^{6+} n'existe pas en solution mais forme l'ion uranyle très stable $U^{6+} + 2\,H_2O \longrightarrow UO_2^{2} + 4\,H^+$.

TABLEAU II. Kd DE L'URANIUM ENTRE L'EAU ET DIVERS MATERIAUX SOLIDES

SOL OU MINERAL	Kd en cm^3/g			Kd moyen
	1° essai	2^e essai	3^e essai	
Alluvion de l'Essonne	40	39	37	39
Tourbe	29	35	34	33
Sol de Cadarache	15	13	19	16
Schiste altéré de La Hague	269	267	(388)	270
Silice pure	Co = C	Co = C	Co = C	0
Calcaire pur	7	7	8	7
Illite	137	144	136	139

Les solutions diluées de sels d'U sont acides et le pH augmente avec la dilution [8].

La chimie des sels d'uranyle en solution est assez complexe, car ils existent sous différentes formes hydratées ou condensées selon la dilution ou le pH de la solution [9] [10].

Enfin les sels d'U se complexent facilement et en particulier en présence d'ions carbonates.

Pour les expériences on a utilisé le nitrate d'uranyle $(NO_3)_2UO_2$ bien soluble aux pH < 7, une précipitation partielle (hydroxydes) se produit aux pH basiques.

4.2. Le dosage de l'uranium

Les dosages ont été effectués par fluorimétrie, chaque fois 3 mesures sont réalisées; les valeurs trouvées sont en général assez voisines à ± 10 % (voir tableau II), quand les résultats sont trop dispersés, la mesure est recommencée.

La sensibilité de la méthode permet de doser des teneurs de l'ordre de 10^{-6} g/litre en U.

4.3. La rétention de l'uranium par les différents sols et minéraux étudiés

Pour une teneur initiale en uranium de l'eau de 10 mg/litre, on a obtenu les valeurs du Kd de l'U entre l'eau et la phase solide indiquées dans le tableau II.

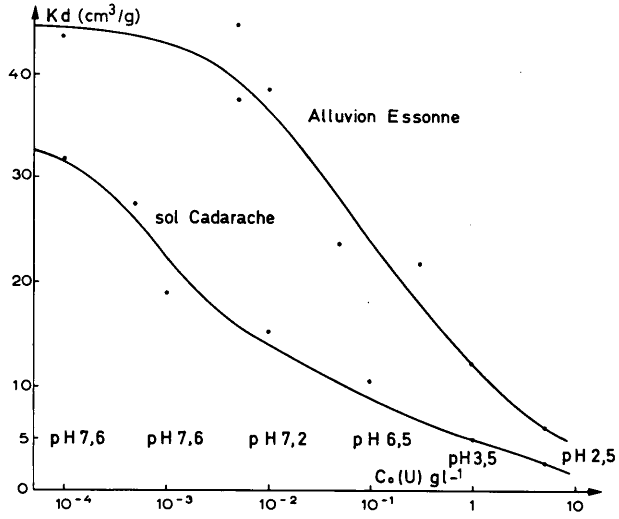

FIG. 1. Variation du Kd de l'uranium en fonction de sa concentration initiale.

Un premier examen montre donc que la rétention de l'uranium par les différents sols étudiés est assez importante.

Cette rétention est du même ordre de grandeur que celle du strontium 90 (exemple Kd Sr entre sol et eau de Cadarache = 20).

4.4. Influence de la concentration initiale en U de l'eau

Cette influence importante est montrée sur la figure 1.

Les courbes Kd = f (Co) montrent que le Kd est à peu près constant aux concentrations inférieures à 10^{-3} g/litre. Par contre, le Kd diminue quand la concentration augmente et il tend vers zéro quand la concentration initiale dépasse 10 g/litre.

Cet effet est commun à tous les phénomènes de rétention, en effet, à partir d'une certaine concentration, tous les sites adsorbants des minéraux sont occupés par l'élément adsorbé. Dans le cas présent, pour des teneurs supérieures à 10 g/l, il y a saturation en U des minéraux du sol qui ne peuvent plus le fixer, les ions restent donc en solution.

Ainsi une contamination du sol par un effluent concentré en sels d'uranium entraînerait un risque de saturation d'une certaine épaisseur de sol et, par conséquent, une dispersion de la contamination.

4.5. Influence du pH de la solution sur la rétention de l'uranium

Pour une concentration initiale donnée, le pH de la solution a une influence très importante sur le Kd comme le montre la figure 2. Cet effet a été étudié avec 2 sols différents :

— avec le sol 1 très calcaire, on ne peut pas faire varier le pH vers ses valeurs acides;

— avec le sol 4 , on a pu explorer toute la gamme des pH.

Les courbes Kd = f (pH) ne sont pas monotones, on y voit 3 pics : un pic aux alentours de pH 5,5, un pic très prononcé à pH 10 et un pic aux alentours de pH 12.

Parmi les explications de ce phénomène, on peut retenir celle donnée par ROJKOVA et Coll. [11] : le potentiel électrocinétique de certaines substances sorbantes varie avec le pH de la solution et pour certains matériaux (lignite, oxyde de fer, kaolinite, montmorillonite ...) passe par un maximum variable selon les corps. Dans notre cas, la présence de divers matériaux dans les sols étudiés explique la présence de plusieurs pics. Ceci permet de supposer une adsorption électrostatique de l'ion uranyle.

D'autres phénomènes peuvent se superposer à celui-ci : au-dessus de pH 7, il peut se produire une précipitation partielle d'hydroxyde d'uranyle qui expliquerait la rapide augmentation du Kd jusqu'à pH 10.

La remise en solution de l'UO_2 peut être due à des complexations; en effet, aux pH basiques, les acides humiques passent en solution et peuvent complexer UO_2^{2+}; d'autre part, entre pH 7 et 12 en solution avec des ions carbonates UO_2^{2+} donne les complexes $UO_2(CO_3)^{2-}$ et $UO_2(CO_3)^{3-}$; cette explication n'est pas valable pour le schiste altéré de La Hague exempt de calcaire et de matière organique [10] [12].

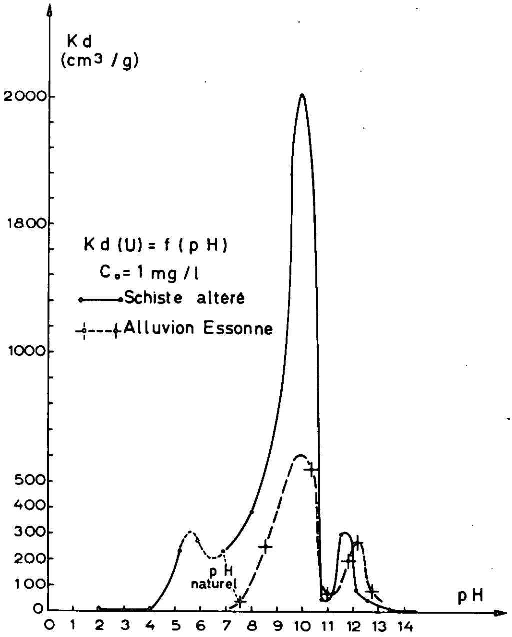

FIG.2. Variation du Kd de l'uranium en fonction du pH de la solution.

Enfin, on a vu que l'ion uranyle donne en solution de nombreux produits d'hydrolyse [10].

exemples : $2\ UO_2 + H_2O \longrightarrow U_2O_5 + 2\ H^+$

$UO_2 + U_2O_5 + H_2O \longrightarrow U_3O_8^{2+} + 2\ H^+$

$U_3O_8 + H_2O \longrightarrow U_3O_8\ (OH)^+ + H^+$

$U_3O_8\ (OH^+) + H_2O \longrightarrow U_3O_8\ (OH)_2 + H^+$

Ces réactions sont fonction du pH et les produits d'hydrolyse peuvent avoir un comportement différent avec les matériaux sorbants. Des études physico-chimiques plus approfondies seraient nécessaires pour expliquer ou prouver l'influence de ces phénomène sur le Kd.

4.6. Rôle des principaux minéraux dans la rétention de l'uranium

On a vérifié (tableau II) que la silice était inerte et on a vu que le calcaire retenait assez faiblement l'uranium en solution.

La rétention est donc surtout due aux minéraux argileux et aussi à la matière organique.

En ce qui concerne la tourbe, il faut préciser que nous avons étudié une tourbe calcique dont le pH se situe entre 7 et 8; les tourbes acides étudiées par SZALAY [12] retiennent beaucoup plus fortement l'uranium en solution diluée.

4.7. Les risques de diffusion de l'uranium dans le sol

Les risques de migrations de l'uranium dans le sol seront importants en cas de rejets d'effluents concentrés ou dans des conditions extrêmes de pH, mais, comme de telles conditions ne pourraient être qu'exceptionnelles, il faut surtout considérer le risque plus plausible de contamination du sol par des solutions diluées dans des conditions de pH voisines de celui du sol. Dans ce cas, la rétention de l'uranium est importante mais non totale; il se constitue un équilibre et l'activité reprise par l'eau est d'autant plus importante que le Kd est plus faible.

Dans une colonne de sol de dimensions finies, on peut évaluer les vitesses relatives de l'eau et de l'ion par la formule :

$\frac{v_{eau}}{v_{ion}} = 1 + Kd\ \frac{d}{f}$ (d poids spécifique et f porosité efficace du sol).

Cette formule appliquée aux sols saturés est approximative car, entre autre, elle ne tient pas compte de la dispersion, mais elle permet de donner une évaluation du risque de migration du radioélément étudié et aussi de comparer les risques relatifs des divers radioéléments.

5. COMPORTEMENT DU THORIUM

5.1. Quelques rappels de la chimie du thorium en solution

Le thorium ne forme qu'une série de sels correspondant à la valence 4+. Les sels de thorium solubles (nitrate, halogénures, sulfate,

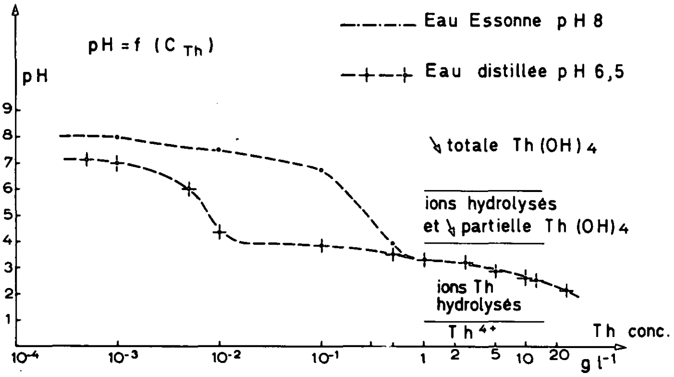

FIG.3. Variation du pH de l'eau en fonction de la concentration en Th.

acétate) sont acides en solution, le pH des solutions diminuant avec la concentration en thorium, comme le montre la figure 3, établie avec du $(NO_3)_4Th$.

L'état des ions en solution est variable selon le pH :

pour pH 1, l'existence de Th^{4+} est bien établie;

entre pH 1 et 4, il se forme divers ions hydrolysés [13];

entre pH 4 et 6, il commence à se former l'hydroxyde insoluble $Th(OH)_4$;

pour pH 6, la précipitation de $Th(OH)_4$ est complète et il ne reste plus que des traces de Th en solution.

Ces divers effets sont exposés sur la figure 3.

Au-dessous d'une certaine concentration variable selon la composition initiale de l'eau, on ne peut pas conserver le Th en solution car $Th(OH)_4$ précipite. Dans l'eau de l'Essonne (pH initial 8) le Th reste en solution seulement pour les concentrations supérieures à 1 g/litre (0,01 g/l pour l'eau de La Hague initialement plus acide).

Par conséquent, le risque de diffusion du thorium dans le sol n'existerait que pour des solutions suffisamment concentrées pour conserver le pH acide; encore faut-il que le sol ne contienne pas de $CaCO_3$ qui réagit avec les ions H^+ et tend à neutraliser la solution.

Ainsi une solution concentrée de Th mélangée aux alluvions de l'Essonne réagit avec le $CaCO_3$: il y a dégagement de CO_2, le pH se stabilise autour de 7 et $Th(OH)_4$ précipite.

5.2. Dosage du thorium en solution

Il existe un certain nombre de méthodes pour doser le thorium [6]. On a choisi l'analyse par activation neutronique qui a donné entière satisfaction [14].

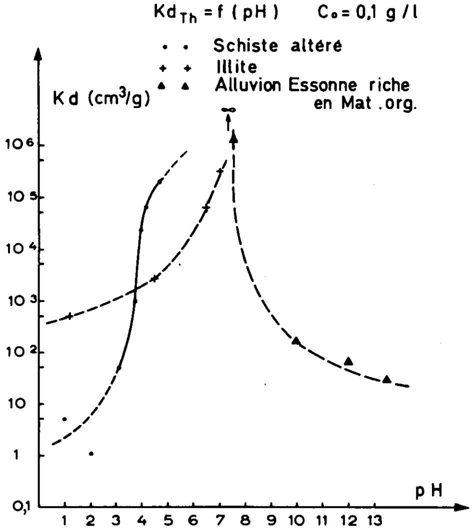

FIG.4. Variation du Kd du thorium en fonction du pH de la solution.

Le schéma d'activation est le suivant :

$$^{232}_{90}\mathrm{Th} \xrightarrow{n,\gamma} {}^{233}_{90}\mathrm{Th}\ (22,1\ \mathrm{min}) \searrow {}^{233}_{91}\mathrm{Pa}\ (27\ \mathrm{j})$$

Les comptages sont effectués sur le protactinium 233 dont la période de 27 jours est suffisamment longue pour donner assez de souplesse à l'expérimentation.

Les mesures ont été faites par spectrométrie gamma sur jonction Ge-Li; la discrimination des pics du ^{233}Pa et des pics des autres produits d'activation est excellente, aussi aucune séparation chimique préalable n'a été nécessaire.

La sensibilité est de 10^{-7} g en ^{232}Th.

5.3. Variations de la rétention du Th en fonction du pH

L'influence du pH est essentielle : figure 4.

Dans le cas des sols argileux exempts de calcaire et de matière organique, on a mesuré les Kd (schistes altérés, sol 4) (illite, argile pure) en faisant varier le pH de 1 à 14. Le Kd augmente très rapidement avec le pH.

Au-dessus de pH 6, la décontamination de l'eau est quasi totale. Il faut aussi noter qu'en solution très acide (pH 2) la rétention du Th par le schiste altéré est très faible.

Dans le cas des sols riches en matière organique (alluvion de l'Essonne sol 1) en raison de la teneur en $CaCO_3$, on n'a pu faire varier le pH qu'au-dessus de 7. On constate l'inverse du cas précédent; le Kd diminue quand le pH augmente. A pH 7,6, le Kd est infini (C_{Th} dans l'eau = 0); quand le pH augmente, $C_{(Th)}$ dans la solution, augmente (Kd = 28 pour pH 13,6).

Les acides humiques sont solubles en milieu basique (la solution de sol devient de plus en plus foncée quand le pH augmente). Le thorium est complexé par les acides humiques et par conséquent, il repasse en solution, la concentration en phase solide diminuant.

Cet effet a aussi été observé dans les milieux naturels [4]; on note une diminution de la teneur moyenne en Th des sols dans les horizons superficiels riches en matière organique.

Dans la pratique, le rejet d'un effluent basique sur un sol riche en matière organique augmentera le risque de migration du Th.

5.4. Influence de la concentration initiale en Th

Dans le cas des sols calcaires éventuellement riches en matières organiques (alluvions de l'Essonne sol 1) mis en contact avec les solutions concentrées - (au-dessus de 0,1 g/l) acides, il y a dégagement de CO_2 et le thorium précipite sous forme de $Th(OH)_4$. Les mesures effectuées sont exposées sur le tableau III.

TABLEAU III. VARIATION DU Kd ENTRE L'EAU ET UN SOL CALCAIRE (SOL 1) EN FONCTION DE LA CONCENTRATION INITIALE EN Th

Concentration en Th (g/l)	pH de la solution	pH après contact avec le sol	Kd cm^3/g
0,01	7,5	7,6	$> 10^6$
0,1	6,7	7,6	$> 10^6$
0,5	3,9	7,1	$\simeq 10^5$
1	3,3	7,1	$\simeq 5.10^5$
5	3,2	6,7	$\simeq 2.10^5$
10	2,6	5,9	$\simeq 2.10^4$

On peut donc considérer que, dans les sols calcaires, le risque de migration du Th est très faible, même s'ils sont riches en matière organique pourvu que le pH ne solubilise pas les acides humiques.

Dans le cas des sols sans calcaire, étudiés sur le schiste altéré (sol 4), la concentration Co en Th ayant une action directe sur le pH du mélange sol-eau, la rétention est influencée à la fois par le pH et par cette concentration (effet de saturation vu en 4.4).

Ainsi, par exemple, avec le schiste altéré :

pour Co (Th) = 1 g.l^{-1}, on a le pH 3,2 et on mesure Kd = 8

pour Co (Th) = 0,1 g.l^{-1}, acidifié à pH 3,2, on a mesuré Kd = 60 (fig. 4)

Autre exemple avec l'illite :

pour Co (Th) = 1 g.l^{-1}, on a le pH 3,2, on mesure Kd = 120

pour Co (Th) = 0,1 g.l^{-1}, acidifié à pH 3,2, on a mesuré Kd = 1000 (fig. 4).

Donc, à pH égal, la rétention du thorium diminue avec la concentration en raison de la saturation des positions adsorbantes des minéraux.

La figure 5 montre l'évolution du Kd (Th) entre le schiste altéré et l'eau de La Hague en fonction de la concentration Co (Th).

En solution diluée (< 0,1 g/l), le risque de migration et de dispersion du thorium dans les eaux souterraines est négligeable.

5.5. Le comportement du thorium rejeté dans le sol

Il est assez difficile de comparer la rétention du thorium par les divers matériaux étant donné la grande amplitude des variations du Kd sous l'effet de divers facteurs et en particulier sous l'effet combiné de la dilution et du pH.

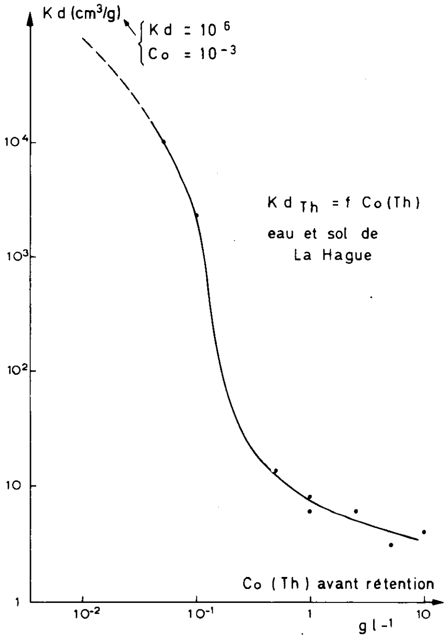

FIG. 5. Variation du Kd du thorium entre eau et schiste altéré en fonction de la teneur en thorium.

TABLEAU IV. Kd (Th) ENTRE LA SOLUTION ET DIVERS MATERIAUX

	Nature du sol ou minéral	Co = 1 g/l		Co = 0,1 g/l		Observations
		Kd	pH	Kd	pH	
Sol 1	Alluvion de l'Essonne	$5\ 10^5$	7,0	10^4	7,3	Il ne reste pratiquement plus de Th en solution
Sol 2	Tourbe	$8\ 10^4$	6,7	$1{,}5\ 10^4$	7,4	Dans 2, la diminution du Kd est due à l'augmentation du pH
Sol 3	Sédiment de Cadarache	$1{,}5\ 10^5$	7,0	$> 10^6$	7,4	
	Calcaire de Meudon ($CaCO_3$ pur)	$5\ 10^4$	6,8	10^5	7,3	
	Sable de Fontainebleau (SiO_2) pure	0	3,3	0	3,8	Rétention nulle $C = Co$
Sol 4	Schiste altéré de La Hague	8	3,2	$\simeq 10^5$	4,8	Grande importance de la dilution qui influe sur le pH.
	Illite	120	3,7	$\simeq 4\ 10^5$	7,0	
	Kaolinite	600	3,0	$\simeq 2\ 10^5$	6,2	
	Montmorillonite	2 000	5,5	$\simeq 2\ 10^5$	7,9	

Les valeurs comparées du Kd (Th) entre l'eau et les solides étudiés est donnée dans le tableau IV pour deux valeurs de la concentration initiale en thorium.

On peut ainsi définir 3 grandes catégories de sols :

— Dans les sols calcaires sans matière organique : le risque de migration du thorium est très faible, quelles que soient les conditions initiales de pH ou de concentration.

— Dans les sols acides sans matière organique : en solutions diluées, neutres ou basiques, les Kd sont très grands et le risque de migration très faible. Mais en cas de rejet de Th dans un effluent concentré ou très acide, le risque de migration devient important; au-dessus de quelques g/l ou en dessous de pH2 la rétention deviendrait négligeable et le thorium serait entraîné par l'eau.

— Dans les sols contenant de la matière organique : le thorium se comportera comme dans les sols précédents, sauf en cas de rejet d'un effluent basique qui dissout les acides humiques; une partie du thorium est alors complexée par les acides humiques et le risque de migration augmenté. La tourbe calcique retient très fortement le thorium quand le pH ne dépasse pas 7 - SZALAY [12] a montré qu'il en était de même pour les tourbes acides.

Remarque :

Il conviendrait aussi d'étudier les interférences possibles de l'uranium ou du thorium sur la rétention de l'un ou de l'autre, ou l'influence de divers autres éléments ou composés qui pourraient aussi induire des phénomènes de complexation, co-précipitation ou saturation des sites adsorbants.

6. CONCLUSION

Le comportement de l'uranium et du thorium dans les sols est différent : il faut noter en particulier, dans le cas du thorium, le rôle essentiel du pH et de la concentration et, dans le cas de l'uranium, les variations irrégulières de la rétention en fonction du pH.

D'une façon générale, en cas de rejet de solutions diluées, l'uranium présenterait une faible mobilité dans le sol alors que le risque de migration du thorium serait négligeable, sauf dans certains cas particuliers comme la complexation du thorium par les acides humiques en milieux basiques.

Il convient de noter que le risque plus important de migration, après rejets de solutions concentrées ou dans des conditions extrêmes de pH, ne pourrait provenir que de circonstances exceptionnelles et serait, de ce fait, limité en importance et en durée.

Une pollution continue du sol peut, par contre, provenir du lessivage par les eaux de pluies ou d'infiltrations de déchets stockés; dans ce cas, les concentrations en uranium ou thorium seraient assez faibles pour que les risques de migration sur de longues distances restent peu importants.

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DISCUSSION

K.B. MISTRY: It is gratifying to see such a detailed study as that performed by Mr. Rançon on the soil chemistry of thorium and uranium. However, I would like to point out that among the radionuclides of the uranium and thorium series the radium isotopes ^{226}Ra and ^{228}Ra are known to be transferred to plants in appreciable amounts. Again, we should bear in mind that the discrimination offered by the gastro-intestinal tract against uranium and thorium is considerably greater than against radium.

D. RANÇON: Thank you for your comment. We are in fact carrying out a study on radium at present.

TRANSPORT OF ^{131}I, ^{137}Cs, ^{106}Ru, ^{144}Ce AND ^{54}Mn IN AN UNDISTURBED SOIL UNDER NATURAL ENVIRONMENT CONDITIONS

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Abstract

TRANSPORT OF ^{131}I, ^{137}Cs, ^{106}Ru, ^{144}Ce AND ^{54}Mn IN AN UNDISTURBED SOIL UNDER NATURAL ENVIRONMENT CONDITIONS.

An unusually large, undisturbed and overgrown soil sample with a diameter of 1.9 m and a height of 1.5 m was taken from a site assigned for a reactor. Over a long period of time the translocation of a mixture of fission products, containing ^{131}I, ^{137}Cs, ^{106}Ru, ^{144}Ce and ^{54}Mn was investigated in the soil sample under natural atmospheric conditions. The surface activity corresponded to an accident situation that might occur when activity is released through the chimney. The penetration of the radionuclides into the soil was determined by γ-spectroscopy and autoradiography.

For 10 months after the contamination the fission products, mainly cationic, could only be found in the superficial soil layers. Additional investigations with ^{131}I and inactive iodine, determined by activation analysis, showed that anionic iodine penetrates deeper into the soil within a shorter period of time than do cations. This offers the possibility of determining in advance the extent of a casual iodine contamination by investigations with inactive iodine in a specific soil.

1. INTRODUCTION

In the coming decades nuclear power plants will meet a considerable portion of the increasing demand for energy. By the year 2000 about 50% of the world's energy will be supplied in the form of electricity. About 85% of that is likely to be supplied by nuclear energy [1]. In addition, the development of nuclear techniques has inspired a remarkable increase in the industrial, medical and scientific use of radionuclides.

In view of this development an urgent demand has arisen to keep the surroundings of a nuclear power plant or of any institution handling radionuclides free from radioactivity above the permissible limits. The construction and operation of a nuclear power plant are certainly planned with this aspect in mind. Other nuclear technical institutions, too, have to meet the recommendations of the ICRP. Maximum credible accidents (MCA) will not be considered here as they are technically hardly possible [2].

Regardless of the care taken in following the safety rules, one has to consider the possibility of fission products or other radionuclides escaping into the surrounding soil. Infiltration and transport of radioactive substances by rain may cause contamination of the groundwater and possibly drinking water too.

In the special case of a nuclear power plant the decision on the proper location is therefore also dependent upon the question of whether such expected or unexpected sources of danger can be limited by the sorption properties of the specific soil. If the proposed location of a nuclear power plant is inside or at the edge of a drinking-water source area, the question may be important for the operation permission of if and to what extent any contamination of the deeper soil layers down to the groundwater table is possible in this area. In other words, can contamination by radionuclides be reduced extensively in the soil itself by sorption and ion exchange. If the retention of the soil is high and the groundwater table is deep enough, there is no objection, under the premises of a low population density, to weak and semi-strong radioactive waste waters from nuclear plants being released to the subsoil. This is possible mainly because of the ion exchange within the soil, which can slow down the movement of a radioactive solution and consequently restrict the resulting contamination.

The first investigations on mineral exchanges in soils were described more than a century ago [3,4]. They referred to cations (without defining them as such) applied to the soil with fertilizers. These are retained by the soil and remain available for plants over long periods of time. Today the ion-exchanging properties of natural soils are attributed mainly to clay fractions [5]. Important information on this phenomenon has been obtained from laboratory experiments into the composition of the adsorbed elements, their chemical compounds and concentrations, times of contact and pH values. A number of basic reports on this have been published [6,7].

In a natural soil, of course, several cations have to be considered in the case of a real soil contamination. The speed of penetration of each type of ion and its sorption is dependent not only upon the affinity of the exchange centres for the specific cations but also upon their composition and the soil components. Consequently, predictions on the retaining factors and decontamination of radionuclides are hardly possible without experimental investigations. Therefore, quite a number of institutions are engaged in studies on the sorption properties of soils. Today, this is mainly a question of routine investigations, the methods of which have been described repeatedly [8-18].

Model tests of this kind and experiments with more or less disturbed soils have shown that all cationic radioactive components are retained within the first centimetres of the soil. Anions, like nitrate ions, iodine and the ammonic complexes, outstrip the cations and can be detected at greater depths after a relatively short time. This is why they can generally be used as indicators of spreading contamination, by fission products, for example. The advance of the actual, long half-life activity frontier can normally be expected one or two years later. This mainly depends on the questions of how fast the anions penetrate into the soil and to what extent are the cations really retained in the upper soil layer. These are imponderables, which cannot be determined by model experiments as the latter provide only an approximate simulation of a natural soil. Therefore, recent papers on infiltration and exchange processes emphasize the difference between disturbed and undisturbed soil samples.

Experiments concerning the decontamination of drain water as well as those dealing with the movement of water and certain substances in soils are occasionally carried out with undisturbed soil samples [19-21], i.e. soil columns extracted with special equipment and kept undisturbed during the experiment. In this way greater attention is paid to the mechanical structure and to the accidentally occurring properties of the natural soil than can be the case with an artificially, homogeneously filled soil column. However, the natural precipitation (rain, snow, melted snow and ice) and its connection with the groundwater must also be considered with regard to their influence on decontamination in undisturbed soils. Consequently, the results from model experiments can only give an idea of the properties being examined. Nevertheless, they are extremely important as they are the only way to get a general picture of the relative decontamination properties of soils.

The experiments described below deal with translocations in an undisturbed large soil profile under natural climatic conditions to get a true and detailed picture of the real situation in the soil. Another specific purpose is to find out whether or not the translocation and the retention of anionic fission products (esp. ^{131}I) in the presence of cationic components can be considered characteristic for the decontamination properties of a soil under natural conditions.

2. TRANSLOCATION OF ^{137}Cs, ^{106}Ru, ^{144}Ce AND ^{54}Mn

An undisturbed soil column, 1.9 m in diameter and 1.5 m in height, was taken from a site at the River Danube, officially selected as the location for a nuclear power plant. A steel cyclinder was placed on the completely flat ground and driven into the ground by a 2-ton drop hammer. During the lowering process the soil around the cylinder was taken away to reduce the friction on the cylinder walls. Afterwards a small dredging machine excavated a large quarry around the cylinder to enable a bottom plate to be attached to the container. After removing the container the soil surface (sward) was covered with two layers of plastic foil to prevent the upper soil layers drying out. Before depositing the cylinder on four concrete blocks inside a steel tank at the Technical University of Hanover, several holes

FIG. 1. Driving the steel cylinder into the ground.

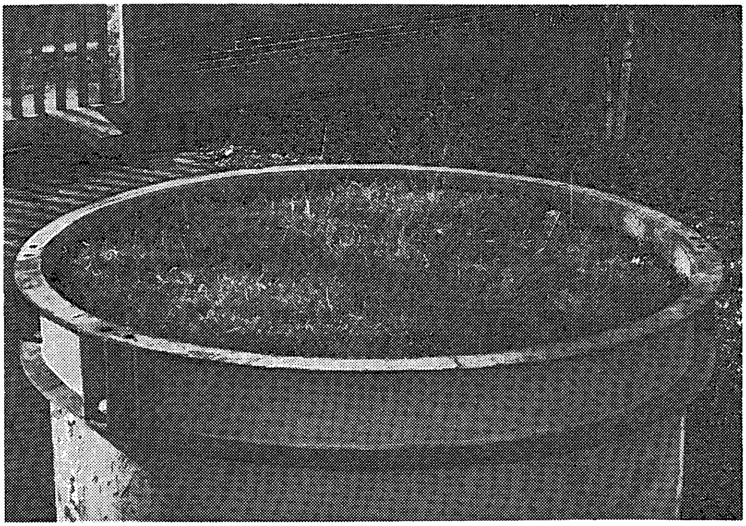

FIG. 2. Soil column after installation.

were drilled into the bottom plate to enable an artifical groundwater table to be applied. Control of the height of the soil column showed that no measurable compression of the undisturbed soil had occurred during transport. Figures 1 and 2 show stages of the extraction of the soil column.

A first coarse inspection of the soil showed the following stratification:

(1) Humus layer with dense sward down to 0.25 m
(2) Native soil, very fine sand and fine sand down to 0.6 m
(3) Fine sand down to 1.0 m
(4) Below that: gravel with some coarse sand.

The analysis of the soil yielded the results given in Table I.

An area of 50 × 50 cm^2 of the grass-covered soil surface was contaminated with the carrier-free fission products ^{144}Ce, ^{137}Cs, ^{131}I, ^{106}Ru and ^{54}Mn. The water applied corresponded to an amount of precipitation of 4 mm. ^{54}Mn is produced in the neutron field of the reactor by the reaction $^{55}Mn(n, 2n)\,^{54}Mn$ and occurs mainly in the cooling water of pressurized water reactors.

The contamination was carried out in the fall by applying 200 μCi of each of the elements mentioned. The whole specific surface activity was $4 \times 10^{-1} \mu Ci/cm^2$. The elements Ce, Ru, Cs and Mn were applied as chlorides and iodine as NaI in the following molar concentrations:

CsCl: 10^{-8} $\underline{M}$
$CeCl_3$: 10^{-9} $\underline{M}$
$MnCl_3$: 10^{-10} $\underline{M}$
$RuCl_3$: 10^{-10} $\underline{M}$
NaI: 10^{-11} $\underline{M}$

TABLE I. COMPOSITION OF THE SOIL COLUMN

	Analysis	
Upper soil layers		(%)
(down to 20 cm below surface)	SiO_2	53.6
	Al_2O_3	11.0
Organic substances 2.3%	Fe_2O_3	2.75
Clay fraction 22.6%	MnO	0.077
	CaO	4.93
Sand		
grain size 2- 63 μm 39.1%	MgO	5.64
63- 200 μm 32.3%	Na_2O	0.54
200- 630 μm 5.5%	K_2O	1.20
630-2000 μm 0.5%	P_2O_5	0.12
	SO_4	0.05
	Cl	0.06
Lower soil layers		
	SiO_2	53.8
(20 to 100 cm deep)	Al_2O_3	9.9
Slightly sandy loam	Fe_2O_3	2.18
Organic substances 0.9%	MnO	0.068
Clay fraction 12.4%	CaO	6.43
	MgO	5.64
grain size 2- 63 μm 31.5%	Na_2O	0.52
63- 200 μm 40.4%	K_2O	1.06
200- 630 μm 15.3%	P_2O_5	0.09
630-2000 μm 0.4%	SO_4	0.05
	Cl	0.04

The translocation of these radionuclides could be studied with respect to location, time and depth under natural weather conditions in this natural soil. There were no wall effects whatsoever, which usually falsify the results from common model experiments and from set-ups with smaller undisturbed columns of soil. Furthermore, the existence of vegetation is very important as the retention in the rooting zone of plants may be different from that in bare soils. Moreover, plants allow the study of the uptake of certain fission products from the soil.

At certain time intervals samples were taken from the soil column by a cutting tube, 1 m in length and 2 cm in diameter. A sample can only be taken once at a specific spot, as the soil must be considered as disturbed at this point, although the hole was always refilled with the proper soil. Soil samples were taken over a period of 10 months:

(a) at times of poor precipitation, usually once a week
(b) additionally after heavy rain and snow
(c) after the snow had melted and the soil had thawed.

This was done to find out any change in retention after heavy precipitation or after snow break. Each sample was divided into 5-cm pieces, which were put into polyethylene capsules.

Except for ^{131}I, the fission products applied were cationic, long-lived gamma sources, which were measured in the usual way in a borehole crystal and analysed by gamma-spectroscopy. As in all cases of a mixture of fission products, quantitative analysis is only possible by deploying the complex spectrum. This was done by the computer program "alpha" (Fortran 63), which was developed at Oak Ridge to determine radionuclides of low concentrations from gamma spectra. It also considers several possible ways of correcting typical errors arising in the pulse-height analysis [22-24].

The translocation of radionuclides could also be determined by autoradiography of the soil samples. Although this method yields only qualitative results, it gives a good idea of the retention in the upper soil layers, which can vary extraordinarily in undisturbed soils, as was apparent in this case. Spectroscopy and autoradiography of the soil 5-cm samples as functions of vertical and horizontal position give a picture of the distribution of the fission products in the soil profile.

Figure 3 shows the translocation of the entire activity of the fission products after 10 months. The activities at 36 drilling positions with 12 samples each have been related to the activity of the first 5-cm sample. Activities from the same depth have been averaged so that the diagram shows the mean distribution of the whole activity below the contaminated surface area down to 100 cm.

Figure 3 gives a sufficiently representative picture of the infiltration process. It shows both the extremely slow motion of the fission products and the translocation. After 10 months and even much longer about 99% of the total activity applied is retained in the first 35 cm of soil. Between a depth of 20 and 30 cm a slight concentration of radionuclides is noticeable.

The soil samples taken 7 days after contamination showed the same activity distribution as found after one year. Therefore, it is possible to describe the percolation as the mean value of quantities measured at different times. The period of one year obviously does not play any role as far as the penetration process of cationic fission products is concerned. Therefore, different times of sampling have no influence on the result. The amount of precipitation by months for the entire measuring period is shown in Fig.4. The total precipitation amounted to about 700 mm. A depth of 35 cm, just barely penetrated by an activity of one thousandth of the surface activity, corresponds approximately to the range of precipitation. The translocation of fission products in this 35-cm range below the surface is therefore caused by the entire precipitation during the measuring time, minus the portion that evaporated or was taken up by the vegetation. As the infiltration depth of the precipitation and the activity is about the same,

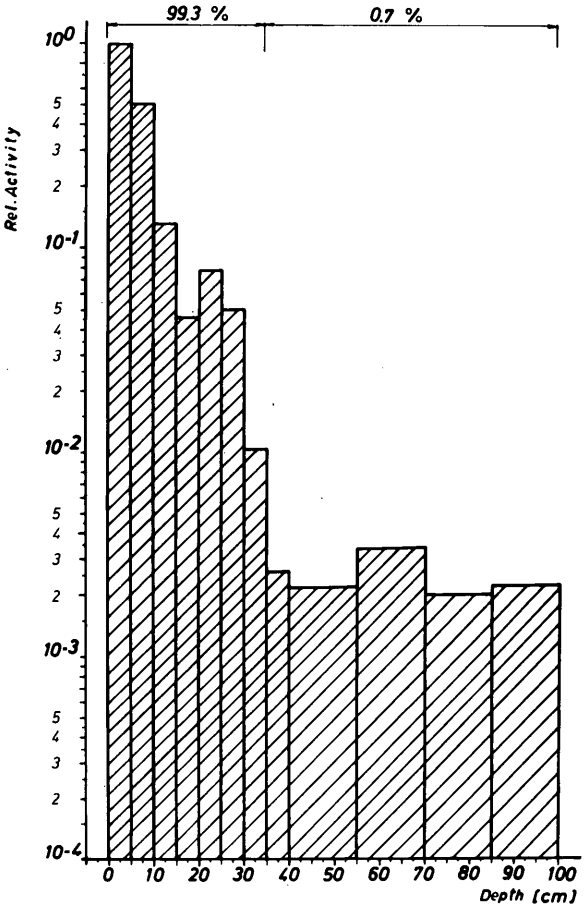

FIG. 3. Distribution of the whole activity after 10 months.

it can be assumed that the translocation would be negligibly small in the case of no precipitation. Obviously the ions are transported by mass flux in water, as far as they are not retained by surface or interspace adsorption at minerals. Experiments with sandy loam of various soils carried out by Squire and Middleton [25], Thornthwaite et al. [26] and Schuffelen [27] yielded similar results.

Frederikson et al. [28] showed that 12 months after contamination with ^{106}Ru, ^{137}Cs and ^{144}Ce at least 80% of the contamination was found in a layer 2.5 cm below the surface. Peirson and Salmon [29] pointed out that 80% of the total world-wide fall-out can be found in a layer of 7 cm below the surface.

This appears to be true also for the soil column under consideration, which contains 2.3% of organic substances and 22.6% of clay in the upper soil layer supporting the adsorption.

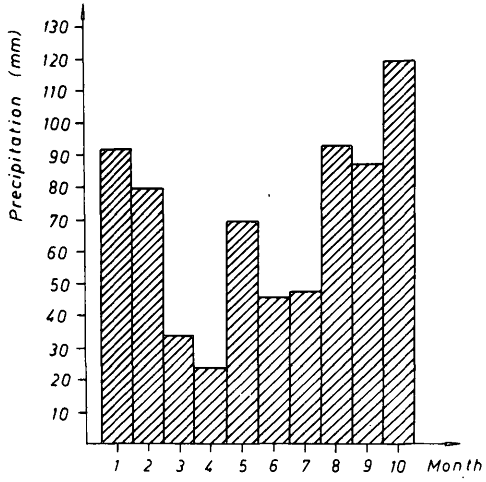

FIG. 4. Precipitation during period of measurement.

In connection with fall-out investigations R. Scott Russell [30] and Klechkovsky and Gulyvakin [31] have discussed the incorporation of fission products by plants above the ground. Therefore, it was assumed that retention by the sward would play a role, which would be indicated by a concentration of radionuclides in the grass. Surprisingly enough, we found that of the applied fission products ^{54}Mn had been favouredly incorporated into the grass. Four weeks after contamination the activity of the grass was already about 200 pCi/g fresh weight with an original area activity of 8×10^{-2} μCi/cm^2 of that radionuclide. The other radionuclides were detectable only as trace amounts or not at all.

As a result of atom-bomb tests ^{54}Mn was found to be one of the most frequent radionuclides in plants, and its gamma activity in fall-out is comparable to that of ^{137}Cs [32, 33]. The concentration of ^{54}Mn by grass-grown soils should therefore be emphasized. In the case of surface contamination it should receive the same special attention as the other radionuclides within the food chain. As a result of its half-life of 303 days and its favoured incorporation into grass, it can be of influence even at low activities. If ^{54}Mn enters the food chain via grass as animal food, it can add to the radiation burden from the K_α radiation of the decay product ^{54}Cr. The biological effects of soft and ultra-soft X-rays are discussed more frequently in the literature nowadays [34]. This energy transfer must be estimated as being higher than that of gamma radiation (0.84 MeV).

During this investigation we also examined whether or not the various fission products act according to the cationic affinity sequence:

Cs^+	Ba^{++}	Sr^{++}	La^{3+}	Ce^{4+}	Y^{3+}	$Mn^{2/4+}$	Ru^{4+}	
1.69	1.35	1.13	1.15	1.01	0.93	0.9	0.65	ångströms

In general, decreasing ion radius and increasing valence means increasing affinity, a rule with numerous exceptions. This means that it might be

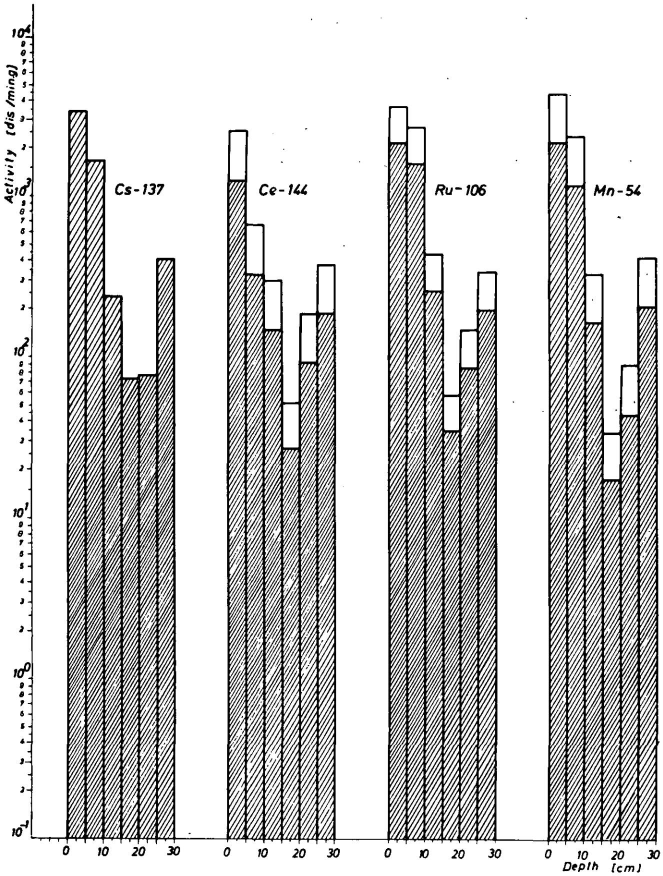

FIG. 5. Mean values of the activity distribution of ^{137}Cs, ^{144}Ce, ^{106}Ru and ^{54}Mn from 18 soil core positions.

possible to find one or other fission product outside the activity front. To check this, the activity of each fission product measured at a certain depth (5-cm steps as before) was averaged over 18 soil positions. The evaluation yielded the activity distributions of each element, showing that about 99% of the activity can be found within the first 30 cm below the surface. Figure 5 shows that none of the cationic elements overtook the others subsequently. This again indicates a connection between translocation of activities and precipitation. The cationic affinity seems to be a secondary effect in the case of an undisturbed soil.

3. TRANSLOCATION OF FISSION IODINE

The dangerousness of fission iodine in the food chain and in drinking water is well known [35]. Its translocation in relation to the pH value and to the clay fractions has been investigated [10, 36].

Occasionally the question has arisen of whether or not there is a mutual influence on fission products by the formation of complexes, for example, which may result in an unreasonable translocation of iodine. It should be taken into consideration that iodine can form compounds with all other elements except the inert gases [36]. For this reason, experiments were carried out to investigate the translocation of iodine, especially locally limited infiltration differences and effects from precipitation shortly after the contamination.

Because only the centre part of the soil surface had been contaminated, it was possible to study the percolation of iodine together with the other fission products and simultaneously without them outside the centre part under the same conditions of soil, vegetation and climate.

3.1. Low iodine concentrations

Figure 6 shows the results of three experimental series (a, b, c) of three sample positions each. The samples were taken at different times within 1 to 14 days after contamination. The molar concentration of ^{131}I amounted to 10^{-11} with an area activity of 3.3×10^{-1} μCi/cm^2, similarly to that of the mixture of fission products.

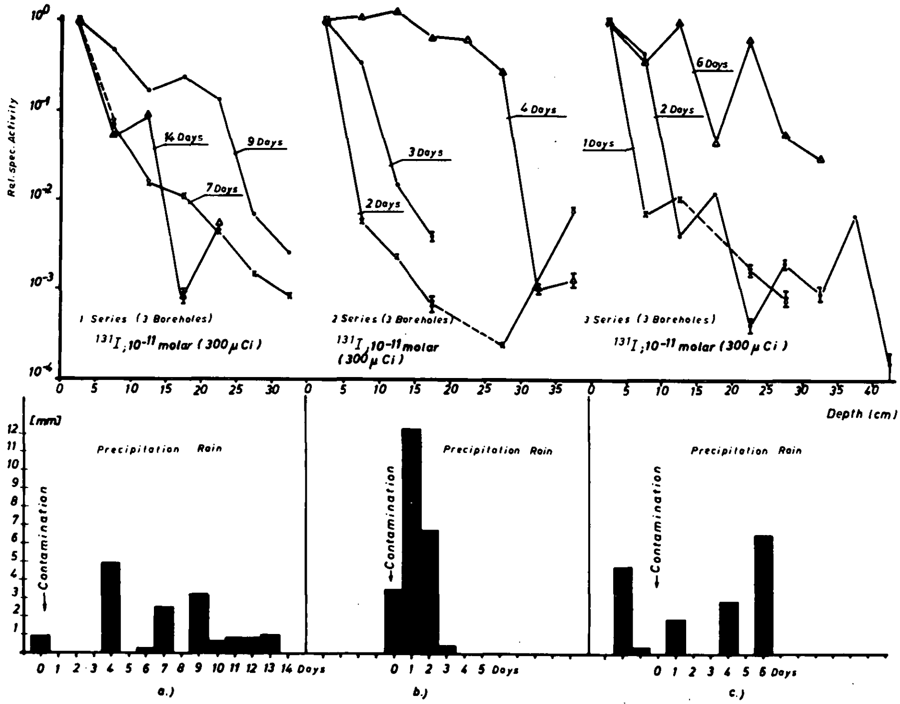

FIG. 6. Translocation of ^{131}I (10^{-11} molar) in the soil column and its distribution in relation to position, depth and time after contamination and the amount of precipitation; three series of experiments (a-c).

Series a: amount of precipitation 14.7 mm

In the case of relatively little amounts of precipitation during the experiments iodine could be detected down to 40 cm below the surface. The change in activity went over several powers of ten at different times and positions. For example, the concentration 9 days after contamination 15 to 20 cm below the surface is 20 times higher than 7 days after contamination without any considerable rain in between. Similar results were found at other depths.

The iodine distribution is probably strongly dependent upon the locally varying soil structure. Obviously small distances strongly affect the percolation due to structural differences. This explains why the concentration 14 days after contamination was partly much lower than in samples taken earlier at other positions.

Series b: amount of precipitation 22.7 mm

During this experiment more rain transported the iodine to deeper soil layers within a short period of time. The activities measured at the same depth vary clearly from one day to the next. The quantities of iodine found on the fourth day after contamination were 2 to 3 orders of magnitude larger than the preceding samples. The fact that the activity of the third sample between 10 and 15 cm below the surface was higher than that at the surface is indeed remarkable. Two days after contamination the activity had already reached a depth of 40 cm. This seems to express the influence of a higher precipitation rate and a larger total amount on the translocation of iodine.

Again, the soil structure plays a part: on the second day after contamination the iodine content at a depth of 40 cm was already higher than on the fourth day, and three days after contamination in the second sample there was no iodine below 20 cm. This again shows the great differences of the retention properties of the soil at different sample positions.

Series c: amount of precipitation 16.6 mm

The soil samples were taken on the first, second and sixth day after contamination. On the second day activity was detected at depths of between 40 and 45 cm. High concentrations were found in the third sample taken on the sixth day after the relatively heaviest rainfall. This and the second sample show that iodine is retained preferentially at certain depths. The first two samples indicate a related infiltration tendency. The precipitation during this time interval was low. So again the soil structure must be held responsible for this behaviour.

Summing up, the three experimental series yield the following results: 1 to 2 days after the contamination iodine can already be found down to 40 cm. As the first sample was taken 24 hours after the contamination, an earlier reaching of this depth is probable. The depletion can range over several orders of magnitude. The varying concentration is a result of both the soil structure and the precipitation. The reason for this is the fine structure of the soil, causing a very different infiltration process from position to position. It can be called fine structure as the distance between the samples was only 15 to 20 cm.

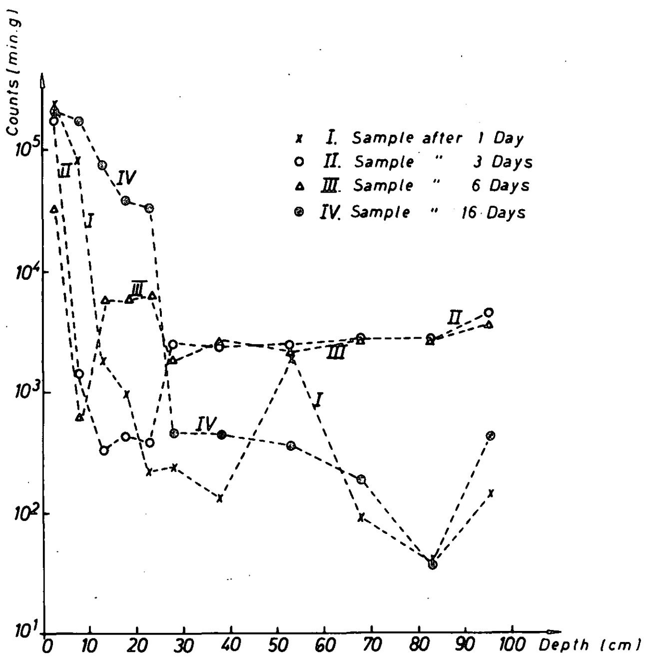

FIG.7. Translocation of iodine in a 1.2 molar solution and its distribution in relation to position, depth and time after the contamination. Detection of the iodine by activation analysis with the reaction $^{127}I(n,\gamma)^{128}I$. (Amount of precipitation 18 mm.)

Precipitation undoubtedly increases the activity at the depth already reached. However, there is no increase in depth of infiltration after heavy rain. The depletion ranges in all cases over several orders of magnitude down to 35 cm below the surface.

3.2. High concentrations

Besides these investigations with low surface concentrations translocation experiments were carried out with high concentrations to find out whether in this case iodine penetrates to greater depths. We used 1.2 molar inactive iodine solutions. The detection of the iodine was done by indicator activation analysis [37-39] with the reaction $^{127}I(n,\gamma)^{128}I$. The total amount of iodine corresponded to an activity of about 10^7 Ci. The results are shown in Fig.7. The amount of precipitation was about 18 mm. The contamination solution added another 3 mm.

The figure clearly shows that the percolation is not only dependent upon time, but also very much upon position. Also, it can be detected at greater depths.

At very high concentrations, too, as in this case, the fine structure of natural soil again plays an important part. But in all cases the depletion ranges over several decades down to 30 cm below the surface.

3.3. Fission iodine in the presence of other fission products

Finally, it is important to know the process of percolation of fission iodine in the presence of long-lived fission products in this soil. As against the long-lived radionuclides to which ^{131}I had been added in low concentration from the beginning, the iodine is present for only a relatively short period of time because of its short half-life.

Its percolation in company with the other fission products is demonstrated in Fig. 8. The decay of iodine has been corrected to indicate the real translocation. Four soil samples were taken within 26 days, on the 5th, 12th, 19th and 26th days after contamination. The figure shows also the precipitation during that time. The total amount was 98 mm, which is a lot more than during the preceding iodine experiments.

The figure also shows the sum of all the other fission products 110 days after the contamination when there was no iodine left. One can see that the iodine has penetrated down to 55 to 60 cm in the first and last samples. This again demonstrates the effect of the position of the sample, as the time interval between these samples was rather long and there was varying precipitation during that time. Below 30 cm the iodine concentration is much higher than that of the mixture of fission products, proving clearly a deeper translocation of the iodine. Above 30 cm the behaviour of iodine and the other fission products is very similar, although position has an effect.

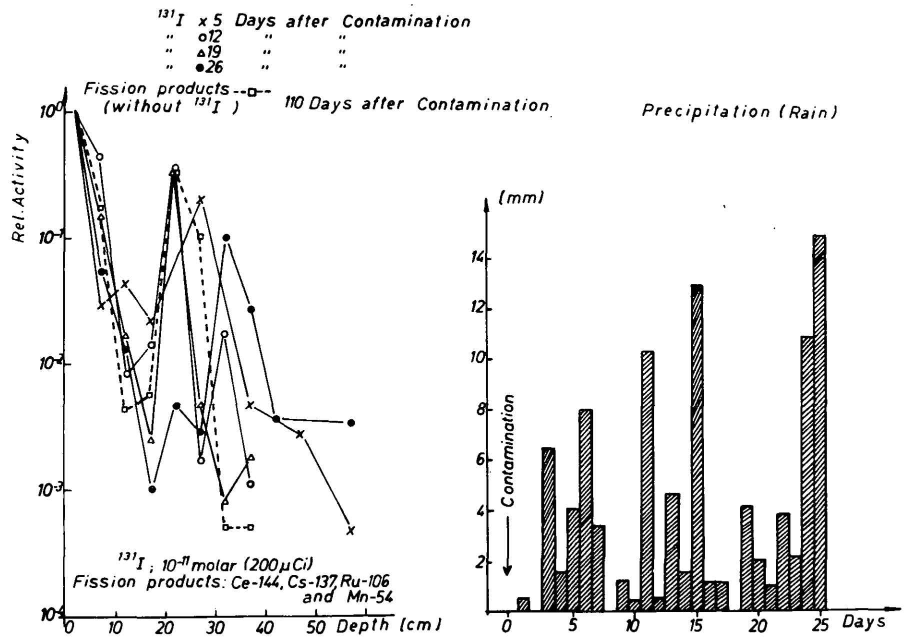

FIG. 8. Translocation of ^{131}I in the mixture of fission products consisting of ^{144}Ce, ^{137}Cs, ^{107}Ru and ^{54}Mn and the amount of precipitation.

The maximum between 20 and 30 cm is caused by the structure of the soil column. This layer delays the iodine penetration. However, the tendency to penetrate further than the other fission products remains, as the maximum reaches deeper layers.

According to these results iodine is undoubtedly suited in a specific way to determine the decontamination properties of a soil in translocation experiments. As an anionic fission product it obviously penetrates deeper into the natural soil than the other fission products. As the investigations with an undisturbed soil under natural climatic conditions show, whether in low or high concentrations it penetrates beyond the main activity front.

The other fission products show hardly any influence. This makes it possible to estimate the extent of a possible hazard situation when using radioiodine as a control substance for the maximum advance of radioactive contamination of a soil, especially, as radioiodine presents one of the greatest hazards.

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DISCUSSION

O.J.A. TIAINEN: In the Introduction to the paper you state that the sorption properties of the soil may affect the selection of nuclear plant sites. Is there any case in the Federal Republic of Germany in which the soil properties and the associated danger of groundwater contamination have indeed influenced the choice of site?

W.K.G. KÜHN: No, there have been no specific examples of this, but in the special field I mentioned the Government authorities and the operating company were interested in obtaining the relevant data from an undisturbed soil sample.

C.A. MAWSON: Perhaps I may mention an experiment on undisturbed soil that we have been conducting over the past 20 years at Chalk River. We have put millions of gallons of water containing a total of over 30 000 Ci of mixed fission products directly into the ground in our waste management area. The soil is sandy and the water table is about 3 metres below the surface. The ground water discharges into a lake about 0.5 km away. Over the 20 years during which the 30 000 Ci were discharged, a total of 13.3 Ci passed into the lake, nearly all in the form of ^{90}Sr.

O. L. van der BORGHT: If I understand correctly, you consider ruthenium to be a cationic fission product. However, the results of the experiments we have performed in our laboratory indicate that most of the Ru is in anionic form. Is this your experience also?

W.K.G. KÜHN: Yes, ruthenium forms anionic complexes and these may be transported like iodides or other anions. However, we did not find this in the undisturbed soils studied.

BIOLOGICAL TRANSFER OF RADIONUCLIDES

(Session V)

Chairman:

K.B. MISTRY (India)

ФАКТОРЫ, ОПРЕДЕЛЯЮЩИЕ ПОВЫШЕННУЮ МИГРАЦИЮ ЦЕЗИЯ-137 ИЗ ОКРУЖАЮЩЕЙ СРЕДЫ В ОРГАНИЗМ ЧЕЛОВЕКА

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Abstract–Аннотация

FACTORS DETERMINING THE INCREASED MIGRATION OF CAESIUM-137 FROM THE ENVIRONMENT INTO THE HUMAN BODY.

The authors present results of observations carried out in various regions of the Soviet Union over a period of five years. In these regions the uptake by the rural population of ^{137}Cs from all sources with the diet- and consequently its concentration in the body - is comparatively high. Investigations have shown that the high internal radiation doses received by the population are due to the characteristics of ^{137}Cs migration within the soil-plant chain. Increased ^{137}Cs uptake from the soil by plants was observed in alluvial lowlands with various kinds of marshy ground and turfy-podsolic sandstone. It has been established that the main reason for the high migration of ^{137}Cs from such soils into plants is the absence (or low concentration) of argillaceous minerals, particularly illites, in them. The low concentration of mobile potassium in the soil and the high water content also favour migration. The coefficients of ^{137}Cs migration into plants from such soils are several factors of 10 higher than the average values for the country as a whole, with the result that the concentration of ^{137}Cs in locally produced foodstuffs - especially milk - is 10-100 times higher. Determinations of the concentration of ^{137}Cs in the bodies of a large number of local inhabitants revealed differences by age and sex. Distribution curves for the probability of different ^{137}Cs concentrations in the body were obtained and exhibit a log-normal character. The maximum concentration of ^{137}Cs in the body is 6×10^{-7} Ci, with average values of $(5\text{-}10) \times 10^{-8}$ Ci. A risk assessment based on calculated population doses shows that the risk of leukaemia with the existing radioactive contamination levels is low (1.7×10^{-3}%). On the basis of the results, the authors have formulated prophylactic measures for regions of the kind investigated. However, such measures would need to be taken only if there were a further substantial increase in environmental contamination.

ФАКТОРЫ, ОПРЕДЕЛЯЮЩИЕ ПОВЫШЕННУЮ МИГРАЦИЮ ЦЕЗИЯ-137 ИЗ ОКРУЖАЮЩЕЙ СРЕДЫ В ОРГАНИЗМ ЧЕЛОВЕКА.

В докладе представлены результаты пятилетних наблюдений, проведенных в некоторых районах Советского Союза. Эти районы характеризуются сравнительно высокими поступлениями цезия-137 глобального происхождения с рационом и повышенным содержанием его в организме сельских жителей. Исследования показали, что повышенные дозы внутреннего облучения населения обусловлены особенностями миграции цезия-137 по цепи: почва-растение. Повышенное поступление цезия-137 из почвы в растения наблюдается в районах полесских низин, где почвенный покров представлен некоторыми разновидностями болотных и дерново-подзолистых песчаных почв. Установлено, что основной причиной повышенной миграции цезия-137 в растения из данных почв является отсутствие (или малое содержание) в них глинистых минералов, в частности, гидрослюд. Факторами, способствующими миграции, являются ограниченное содержание подвижного калия и высокая обводненность почв. Коэффициент перехода цезия-137 из почвы в растения на этих почвах в десятки раз выше средних показателей по стране. Результатом является повышенное в 10-100 раз содержание этого изотопа в пищевых продуктах местного производства, из которых критическим является молоко. Определение содержания цезия-137 в организме большого числа местных жителей позволило показать возрастные и половые отличия в накоплении изотопа. Получены кривые распределения вероятности различных уровней содержания изотопа в организме. Это распределение носит логарифмически нормальный характер. Максимальное содержание цезия-137 в организме достигает $6 \cdot 10^{-7}$ Ки при средних показателях $5\text{-}10 \cdot 10^{-8}$ Ки. Оценка степени риска, проведенная на основе рассчитанных популяционных доз, показывает, что величина риска заболевания лейкемией при существующих уровнях радиоактивного загрязнения низка и составляет $1{,}7 \cdot 10^{-3}$%. Материалы исследований позволили разработать для подобных районов ряд профилактических мероприятий, необходимость применения которых, однако, может возникнуть лишь при существенном увеличении загрязненности внешней среды.

В процессе текущего контроля за радиационной обстановкой, обусловленной глобальными выпадениями, начиная с 1965-66 г.г., на территории некоторых районов Советского Союза стали обнаруживаться повышенные концентрации цезия-137 в пищевых продуктах: молоке, мясе, овощах. Причем концентрации цезия-137 в почве указанных районов были типичными для средней полосы Советского Союза. Это обстоятельство, а также своебразие природных условий указанных районов позволили предположить, что основной причиной повышенного содержания цезия-137 в пищевых продуктах местного производства является интенсивная миграция его из почвы по корневым путям, что в значительной мере определяет его поступление в организм человека. Из работ, выполненных в условиях лабораторных и небольших полевых экспериментов, известно, что повышенная миграция цезия-137 из почвы в растения может иметь место при низком содержании в ней калия, глинистых минералов и т.д. [1,2]. Однако в натурных условиях факты столь интенсивной миграции цезия-137 из почвы в организм человека, которые наблюдались в некоторых районах Советского Союза, нам ранее не были известны. Изучение этого вопроса и явилось одной из основных задач настоящего исследования.

Как было установлено путем многолетних натурных наблюдений и исследований (1967-1972 г.г.), явления повышенной миграции цезия-137 из почв в пищевые продукты наблюдаются преимущественно на территории полесских низин, расположенных вдоль южной границы таежной (лесной) зоны от Польши до Урала. Особенно это явление выражено в пределах Белорусско-Украинского полесья.

Почвенный покров этих провинций представлен разновидностями дерново-подзолистых (лесных) и торфяно-болотных почв различной степени заболоченности и оглеености. Для них характерны высокая кислотность и низкое содержание обменных форм фосфата, кальция, калия, натрия, что обусловлено их значительным увлажнением, а в ряде мест интенсивным промыванием почвенного покрова с выносом обменных оснований в нижележащие грунты.

В результате исследований установлено, что максимальная миграция цезия-137 в растения имеет место на целинных участках, используемых в ряде случаев под пастбища. Примером могут служить данные по Белорусско-Украинскому и Подмосковному (Мещера) полесьям, приведенные в табл.I.

Как видно из табл.I, наиболее высокие показатели миграции цезия-137 в растения наблюдаются из торфяно-болотных и лесных (дерново-подзолистых песчаных) почв, причем эти явления особенно выражены на территории Белорусско-Украинского полесья. На супесчаных и суглинистых почвах показатель миграции в 6-10 раз ниже.

Необычно высокие коэффициенты накопления цезия-137 пастбищной растительностью объясняются особенностями механического и минералогического состава почв, низким содержанием доступного растениям

ТАБЛИЦА I. ПОКАЗАТЕЛИ ПЕРЕХОДА ЦЕЗИЯ-137 ИЗ ПОЧВЫ В ПАСТБИЩНУЮ РАСТИТЕЛЬНОСТЬ

Тип почв	Белорусско-Украинское полесье		Мещерская низменность	
	содержание цезия-137 в растениях, пКи/кг	$K_н$	содержание цезия-137 в растениях, пКи/кг	$K_н$
Разновидности торфяно-болотных и подзолистых песчаных	2370 ± 900	4,5 (1,6-1,1)	950 ± 760	1,2 (3-0,7)
Разновидности дерново-подзолистых супесчаных и суглинистых	450 ± 330	0,5 (1,5-0,2)	90 ± 20	0,2

Примечание: в скобках указан разброс величин.

калия и высокой степенью влажности в зоне активных корней. По механическому составу легкие почвы данных районов можно отнести к группе связанных и рыхлых песков, более тяжелые почвы представлены супесями и легкими суглинками. Основными почвообразующими минералами являются кварц и полевые шпаты. Величина илистой фракции (< 0,001 мм) незначительна и составляет 0,5-0,6% в почвах Белорусско-Украинского полесья и 2,3-7,8% в почвах Мещерской низменности. Основную часть этой фракции составляет органическое вещество – в почвах Белорусско-Украинского полесья – и глинистые минералы (монтмориллонит и гидрослюды), иногда в смеси с органическим веществом – в почвах Мещерской низменности.

Исследования показали, что ответственной за поступление цезия-137 в растительность изучаемой зоны является именно минеральная часть почвы, в частности, наличие и качественный состав глинистых минералов, из которых доминирующую роль в миграции цезия-137 играют гидрослюды.

Другим важным фактором, влияющим на поступление цезия-137 в растительность, является содержание калия в почве, особенно его подвижных форм. Для почв данной местности характерно невысокое содержание калия – 0,97-0,22% [3,4]. Содержание подвижных форм калия, включающих в себя обменную и кислоторастворимую форму, представлены в табл.II.

Сопоставление данных, характеризующих уровни поступления цезия-137 в растительность, и содержание в почвах подвижного калия (табл.I и II), указывают на наличие между ними обратной зависимости, что хорошо согласуется с литературными данными [1, 2]. Для почв изучаемых районов, фиксирующая способность которых крайне мала, существенное влияние на увеличение миграции цезия-137 в растительность

ТАБЛИЦА II. СОДЕРЖАНИЕ ПОДВИЖНЫХ ФОРМ КАЛИЯ В ПОЧВАХ БЕЛОРУССКО-УКРАИНСКОГО ПОЛЕСЬЯ И МЕЩЕРСКОЙ НИЗМЕННОСТИ (мг/кг)

Тип почв	Белорусско-Украинское полесье	Мещерская низменность
Разновидность торфяно-болотных и дерново-подзолистых песчаных	63 ± 8	109 ± 35
Разновидность дерново-подзолистых супесчаных и суглинистых	119 ± 39	178 ± 59

ТАБЛИЦА III. СОДЕРЖАНИЕ ЦЕЗИЯ-137 В ПАСТБИЩНОЙ РАСТИТЕЛЬНОСТИ НА ОДНОТИПНЫХ ПОЧВАХ С РАЗНЫМ УРОВНЕМ ГРУНТОВЫХ ВОД

Тип почв	Уровень грунтовых вод от поверхн. земли, м	Содержание цезия-137 в траве, пКи/кг	Содержание калия, мг/кг	Содержание углерода органического вещества, %
Перегнойно-торфяно-глеевые песчаные	0,5 и меньше	4800	22,2	4,5
	1,0 и больше	1300	26,6	1,7
Дерново-подзолисто-глеевые песчаные	0,5 и меньше	3600	37,0	3,5
	1,0 и больше	300	48,0	4,5

оказывает избыточное увлажнение почвы в зоне активных корней. Под воздействием этого фактора, при прочих равных условиях, фиксация цезия-137 почвами уменьшается, а скорость миграции катионов возрастает [5,6]. Это положение хорошо согласуется с полученными нами данными, характеризующими уровни поступления цезия-137 в растительность из однотипных почв на пастбищах до и после их осушения (табл.III).

Как видно из табл.III, при понижении горизонта грунтовых вод более чем на 0,5 метра содержание цезия-137 в растительности уменьшается в 3-10 раз.

Аналогичная картина в изучаемом районе наблюдалась и при естественном понижении уровня грунтовых вод, что произошло, например, в 1972 году в результате очень сухого и жаркого лета. Из-за понижения горизонта грунтовых вод на 1 м концентрация цезия-137 в растительности снизилась по сравнению с предшествующим годом с 2370 ± 900 пКи/кг до 1100 ± 650 пКи/кг, а коэффициент накопления ($К_н$) – с 4,5 до 1,4.

ТАБЛИЦА IV. СОДЕРЖАНИЕ ЦЕЗИЯ-137 В ОВОЩНЫХ КУЛЬТУРАХ

Тип почв	Содержание цезия-137, пКи/кг	$К_н$
Разновидности окультуренных торфяно-болотных и дерново-подзолисто-песчаных	110 ± 100	0,29
Дерново-подзолистые песчаные	75 ± 65	0,20
Разновидности дерново-подзолистых супесчаных и суглинистых	34 ± 22	0,07

Не только мелиорация, но также последующая распашка и окультуривание заболоченных и лесных целинных почв способствуют уменьшению миграции цезия-137 из них в растения. Это является следствием воздействия таких факторов как механическое разбавление за счет перемешивания наиболее загрязненного поверхностного слоя с массой земли на глубину 20-30 см (глубина вспашки), дальнейшее снижение водонасыщенности почвы в результате ее разрыхления, а также обогащение почвы подвижными соединениями калия, вносимыми с удобрениями. Поэтому содержание цезия-137 в сельскохозяйственных культурах, выращенных на пахотных почвах, значительно ниже, чем в растениях с целинных участков, имеющих аналогичный тип почв (табл.IV и I).

Таким образом, установлено, что в условиях полесских низменностей повышенная миграция цезия-137 из почвы в растения обусловлена главным образом наличием легких по механическому составу почв, малым количеством, а иногда и полным отсутствием в них глинистых минералов, особенно гидрослюд, низким содержанием подвижных форм калия и значительной обводненностью почв. При воздействии всех этих факторов содержание цезия-137 в сельскохозяйственных культурах и пастбищных травах данной местности в 10-100 раз превышает средние по стране значения. Поскольку миграция цезия-137 по пищевым цепям существенно не отражается на его содержании в последующих звеньях, эти же соотношения характерны для таких пищевых продуктов, как молоко и мясо рогатого скота.

Одинаковая направленность сельского хозяйства в районе Белорусского полесья (молочное животноводство) и однотипные условия землепользования позволили на основании собранных обширных материалов установить зависимость между разными типами почв и содержанием цезия-137 в молоке. Однако в других районах страны из-за иных условий землепользования, а также меньших (относительных и абсолютных) размеров площадей с почвами, обладающими повышенной миграцией цезия-137, подобная зависимость отсутствует. Достаточно отметить, что в Мещере и Лесном Поволжье под выпасы молочного скота используются угодья с преобладанием суглинистых почв, откуда миграция цезия-137 происходит очень слабо. Поэтому содержание цезия-137 в молоке и мясе коров здесь приближается к средним значениям по стране.

Существенное влияние на поступление цезия-137 в организм человека оказывают также социальные факторы, в частности структура питания. Применительно к условиям Белорусского полесья критическим пищевым продуктом является молоко, с которым поступает до 70%, а детям – до 80%, цезия-137 (от содержания его в рационе). Это обусловлено повышенным содержанием в нем изотопа (от 100 до 2000 пКи/л), а также широким употреблением всеми возрастными группами сельского населения в относительно больших количествах (в среднем 1 л/сут). Вторoе место по вкладу цезия-137 в рацион (около 20%) принадлежит картофелю. В лесных районах Полесья существенный вклад цезия-137 могут давать грибы, произрастающие на песчаных почвах и накапливающие в себе изотопа до 5000 пКи/кг. Мясо крупного рогатого скота содержит цезия-137, примерно, в 4 раза больше, чем молоко. Однако как источник цезия-137, оно здесь не играет существенного значения, т.к. употребляется эпизодически. В свинине, которая является постоянной частью рациона, цезия-137 содержится мало.

Результаты непосредственного измерения цезия-137 в организме более чем 1000 сельских жителей показали, что максимальное абсолютное содержание его достигает 600 нКи/организм, а в среднем $\sim 10^{-7}$ нКи/организм, что существенно выше среднесоюзных величин. При сравнительно однотипной структуре питания сельского населения полесья картина территориального распределения уровней содержания цезия-137 в организме та же, что и для молока. Максимальное содержание цезия-137 наблюдалось у мужчин. Если не говорить об удельной активности, являющейся эквивалентом поглощенной дозы, то у детей и взрослых мужчин они практически равны (табл.V).

Сравнительно высокие дозы, получаемые детскими контингентами, обусловлены постоянным употреблением относительно больших количеств молока (свыше 1 л/сут).

Анализ полученных материалов показал, что распределение вероятности различных уровней содержания изотопа в организме носит логнормальный характер.

Таким образом, основными факторами, определяющими повышенное содержание цезия-137 в организме сельских жителей ряда районов Белорусско-Украинского полесья, являются: высокий переход изотопа

ТАБЛИЦА V. СРЕДНЕГОДОВЫЕ ДОЗЫ ВНУТРЕННЕГО ОБЛУЧЕНИЯ ЖИТЕЛЕЙ РЯДА НАСЕЛЕННЫХ ПУНКТОВ БЕЛОРУССКОГО ПОЛЕСЬЯ (мбэр/год)

Контингент, лет	Средняя	Максимальная
Мужчины	19	43
Женщины	11	29
Дети 4-6	16	30
7-8	15	39
9-10	16	62
11-16	12	43

из некоторых типов почв в растения, широкое использование этих целинных почв под выпасы и сравнительно большое потребление молока населением.

В отличие от сельских жителей, в рационе которых преобладает молоко и другие пищевые продукты местного производства (кроме хлеба), население городов получает эти продукты из многих пунктов. В итоге происходит нивелирование концентраций цезия-137 в рационе за счет разбавления (усреднения). Поэтому в организме городских жителей полесья содержание цезия-137 существенно ниже, чем у сельских. Однако дозы, обусловленные инкорпорированным цезием-137, даже у сельских жителей не требуют проведения специальных профилактических мер. Вместе с тем выявление районов, где в силу природных и других условий имеет место повышенная миграция радиоактивных веществ из почвы в организм человека, а также установление основных факторов, способствующих формированию доз внутреннего облучения, позволяют, при необходимости, заблаговременно планировать, применительно к конкретным условиям, соответствующие профилактические мероприятия.

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DISCUSSION

R.J. ROUX: In view of the more or less uniform contamination of the soil over wide areas of the country, can you explain the increased accumulation of ^{137}Cs in cow's milk in certain regions of the USSR?

R.M. BARKHUDAROV: In some areas the soil-grass transfer coefficient is 10-20 times greater than in others. It is also the case that many of these same areas have particularly rich pastures, which can be used for feeding dairy cattle practically all the year round. All this naturally adds up to an especially high ^{137}Cs content in the milk in the regions concerned.

R. BITTEL: Similar situations to those described by Mr. Barkhudarov also arise in France, particularly in the Massif Central, Britanny and the Landes, which are regions with a damp, cool climate, a high water-table and largely decalcified and hydromorphic soils. It appears that a substantial fraction of the ^{137}Cs passes directly from the soil water to the grass and abnormally high ^{137}Cs concentrations in milk have been recorded in these areas.

R.M. BARKHUDAROV: Yes, I am sure the situation is very similar. Furthermore, in such soils there are practically no clay minerals, which would otherwise fix a large part of the caesium.

G.H. PALMER: Did you, as a part of this work, derive a coefficient for the transfer of ^{137}Cs from soil to grass?

R.M. BARKHUDAROV: The accumulation factor in grass is 4.5 on average, with a maximum of about 18.0.

G.H. PALMER: And could you now extend this to give a factor for the transfer from grass to milk?

R.M. BARKHUDAROV: We have found that a litre of milk contains about 1% of the ^{137}Cs ingested with the daily fodder ration; this is in good agreement with the findings of other workers.

J.P. MORONI: The dose equivalents represented by contamination of the environment through ^{137}Cs, brought about by particularly high transfer factors in certain areas, are appreciable, although still low in absolute terms. I should like to stress that this contamination is caused exclusively by radioactive fall-out, and not by the operation of nuclear power stations.

ACCUMULATION AND REDISTRIBUTION OF RADIOCAESIUM BY MIGRATORY WATERFOWL INHABITING A REACTOR COOLING RESERVOIR

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Abstract

ACCUMULATION AND REDISTRIBUTION OF RADIOCAESIUM BY MIGRATORY WATERFOWL INHABITING A REACTOR COOLING RESERVOIR.

Changes in whole-body burdens of radiocaesium were studied in monthly samples of American coots (Fulica americana) and other species of migratory waterfowl inhabiting a 2800-acre reactor cooling reservoir on the AEC Savannah River Plant near Aiken, South Carolina, USA. In October newly arrived coots averaged between 4-8 pCi radiocaesium/g live weight and approximated a 50-50 sex ratio. Between October and January the sex ratio of the coots rose steadily to a maximum of 87% males as the females continued to move on to more southerly wintering grounds. In the predominantly male population remaining on the reservoir, radiocaesium body burdens continued to rise at a rate of approximately 2-3 pCi/g per bird per month, to a maximum of between 15-20 pCi/g.

From February through April the predominantly male population of coots began to leave for northern breeding grounds and were apparently replaced by populations that had wintered further to the south and had a higher percentage of females. As a result, the sex ratio began to decline until it once again approximated 50-50 in the late spring. Since these spring transient birds had not wintered on the reservoir, they had lower radiocaesium contents. Thus, the average body burden of the coot population began to decline to a level closely approximating that of the first birds to arrive in fall. Simply sampling the first and last coots seen on the reservoir would have failed to detect the radioisotope build-up shown by the winter resident birds.

In any given month, the radiocaesium content of the coots was generally higher than that of any of nine other species of waterfowl and aquatic birds sampled. The higher body burdens of the coots may be related to their tendency to rely heavily upon submerged aquatic plants as a food resource. Migratory waterfowl may remove up to 3.75×10^{-5} Ci of radiocaesium from the reservoir each year and redistribute it elsewhere along their migratory pathways. The radiocaesium levels found in these birds do not indicate any present health hazard to the general public who may use them for food.

The disposal of most radioactive waste materials occurs in areas of restricted public access and therefore little or no direct human contact with these materials is usually likely. However, the possibility often exists that certain species of wild game, which may occasionally be shot and eaten by man as food, may gain access to such areas of high radionuclide contamination and subsequently move out and thereby serve as vectors of contamination to the food chain of man. Because of their ability to fly, game birds are particularly important in this regard since their ability to move rapidly over long distances makes it possible for them to enter areas of human habitation and hunting long before natural processes of biological turnover have begun to eliminate significant amounts of the radionuclide body burden which they accumulated in the contaminated area. Because the majority of radionuclide wastes are water soluble and often have an opportunity to enter natural aquatic systems, waterfowl may assume a particularly important role as agents for the transfer of radionuclide contamination to the food chain of man.

The present study was designed to describe the accumulation and redistribution of radionuclides by migratory waterfowl populations inhabiting a reactor cooling reservoir on the Savannah River Plant of the United States Atomic Energy Commission, located near Aiken, South Carolina, USA. The Savannah River Plant, which is closed to public access, consists of approximately 750 km^2 of largely undisturbed habitats, several of which have been contaminated with production reactor effluents [1]. The present study dealt with Par Pond, a 1130 ha reservoir on the Savannah River Plant, which acts as part of a closed-loop cooling system for one or more production reactors. A detailed history and habitat description of the Par Pond reservoir system is given by Parker et al. [2]. This reservoir system serves as a wintering refuge area for between 5,000 and 10,000 migratory waterfowl of approximately 20 species, most of which arrive in October-November and depart the following spring for their more northerly breeding grounds [3,4]. In the present study, radiocesium body burdens were determined for monthly samples of these waterfowl populations as they wintered on the reservoir.

Attention was focused on radiocesium because of the particular abundance and biological significance of this radionuclide in game populations of the southeastern United States [5,6]. Furthermore, the majority of this effort was confined to the population of American coots, Fulica americana, inhabiting the reservoir. This species was chosen for particular emphasis because of: (1) its abundance on the reservoir, relative to other species, (2) the relative ease with which it could be systematically sampled in large numbers at particular locations on the reservoir, and (3) because the relatively weak flying ability and limited mobility of the coots, as compared to most other species of waterfowl, made them a preferred species for the likely indication of particular local areas of radionuclide contamination. Whenever possible, however, other species of waterfowl were also sampled in limited numbers in addition to the coots.

MATERIALS AND METHODS

The Par Pond reservoir, which was created in 1959 by damming a natural stream watercourse, may be considered to be comprised of three major extensions, the Hot, North and West Arms (Fig. 1). Both the Hot Arm and North Arm areas received direct inputs of reactor effluents from the time of creation of the reservoir. In the summer of 1964, however, the direct introduction of reactor effluents into the North Arm ceased, and that area has remained undisturbed since that time, while reactor effluents continued to be introduced into the Hot Arm. The West Arm, on the other hand, has remained free of any direct reactor effluent introduction, and since the construction of the reservoir, this area has served as the source of cooling water which is recycled through the reactor whose effluent is returned to the Hot Arm. Because there is some evidence that there may be differences in the extent of radiocesium contamination of the three arms [7], presumably due to differences in their proximity to the site of reactor effluent entrance, an attempt was made to collect 10 coots from each of the three arms during each month that coots were present on the reservoir. However, because of the scarcity of coots at the times of their earliest arrivals and latest departures from the area, the samples for October and May consisted of only 8/7/6 and 0/7/7 individuals for the Hot/North/West arms, respectively. Sample sizes were 10/10/10 coots for all other months. Birds were collected by shooting, between October 1971 and May 1972. Whenever possible, attempts were made to collect all birds of a monthly sample on the same day--preferably a day near the middle of the month. An attempt was also made to collect all birds from as near the extremities of the reservoir arms as possible.

In addition to the 215 coots sampled in this study, radiocesium body burdens were also determined for 46 other individuals representing nine additional species of waterfowl and aquatic birds as follows: 3 common gallinules, Gallinula chloropus; 9 pied-billed grebes, Podilymbus podiceps; 6 horned grebes, Podiceps auritus; 7 ruddy ducks, Oxyura jamaicensis; 5 bufflehead ducks, Bucephala albeola; 6 lesser scaup ducks, Aythya affinis; 3 ring-necked ducks, Aythya collaris; one black tern, Chlidonias niger, and 6 common terns, Sterna hirundo. With the exception of the last named, all of these species have been recorded by Norris [3] as occurring on the Savannah River Plant. This latter species has been recorded on several occasions, however, in the immediate vicinity of the Savannah River Plant [8].

Immediately after collection, birds were weighed to the nearest 0.01 g and the chord of the wing, measured from the proximal end of the carpometacarpus to the tip of the longest primary, was recorded to the nearest mm. Birds were sexed

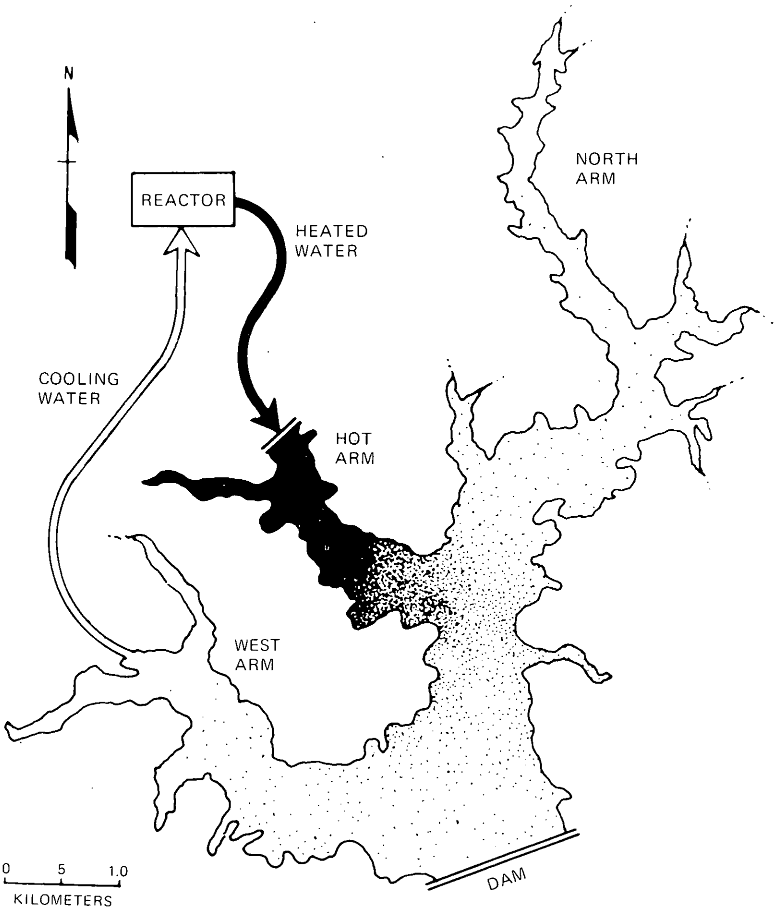

FIG.1. Map of the Par Pond cooling reservoir of the USAEC Savannah River Plant. Intensity of stippling is roughly proportional to the degree of influence of the influx of heated reactor effluents which enter the reservoir in the Hot Arm. The North Arm area has been free of any direct reactor input since 1964. The locations of the reactor and thermal canals are presented diagrammatically.

by internal examination of the gonads and coots were aged by the method of Gullion [9] . Briefly, this method classifies coots, on the basis of tarsal color, into three age categories: (1) immatures (birds of the year) with blue or gray-green tarsi, (2) 2-3 year old adults with yellow-green tarsi, and (3) adults of three or more years of age with yellow tarsi.

Whole-body radiocesium determinations were made on two Packard Model 446 Armac liquid scintillation detectors equipped with Packard tri-carb scintillation gamma spectrometers. Cross-calibrations of the two counters indicated agreement within an average of 0.26 pCi radiocesium/g. This value was less than the maximum sensitivity of either machine which was 1.13 pCi/g for the smallest individual counted in this study. To determine radiocesium concentrations, same-day counts were made of background and aqueous phantoms of known radiocesium content which approximated the size and shape of the bird being counted. Counting periods were either 20 or 50 min, depending on the body size of the bird and were selected to insure a sensitivity of less than 1.50 pCi/g. Radiocesium concentrations which were greater than twice the sensitivity calculated for the smallest individual used in this study were considered to be significantly distinguishable from background.

Although radiocesium content was quantified by counting the combined gamma emissions of both ^{134}Cs and ^{137}Cs, it is unlikely that the former isotope contributed significantly to the counts observed. Marter [1] has shown the ratio of ^{137}Cs to ^{134}Cs to be approximately 20:1 in waterfowl from the Par Pond area. Other gamma-emitting isotopes were present in only relatively small amounts in other similarly contaminated areas of the Savannah River Plant [1] , and had energy spectra which were widely separated from the $^{134-137}$Cs energy peak and were, therefore, unlikely to contribute significant errors to the radiocesium analyses [10] .

RESULTS AND DISCUSSION

Bartlett's test [11] indicated that the data for the radiocesium body burdens of the coots did not show homogeneity of variance ($X^2 = 24.65$; df = 7; $P \leq 0.01$). Applications of several transformations to the data were unsuccessful in producing any reduction in the heterogeneity of variance distribution. Bias in the analysis was therefore minimized by eliminating the data for May because of the inequality of sample sizes during that month. Under the resulting conditions of nearly identical sample sizes in all categories of month and locality, any bias introduced by non-homogeneity of variance would be minimal [12] . This correction to maximize even distribution of sample sizes, together with the fact that many of the differences which were found were demonstrable at a highly significant level (P often $\leqq 0.001$) suggests that any bias due to non-homogeneity of variance would not be likely to significantly alter the results obtained. Under these conditions, a multiple analysis of variance was conducted to evaluate the effects of body weight, sex, age, location in the reservoir, month of collection and the interaction of month and location upon the radiocesium body burdens of the coots (Table I).

The simple effects of body weight and age, if not adjusted for the effects of the other variables, appeared to be significant (F = 11.98 and 4.34; df = 1/176 and 2/176; $P \leq 0.05$) for body weight, treated as a covariant, and age, respectively. However, when adjustment was made for the effects of the other variables, no significant effects of these factors could be demonstrated, as was also the case for sex (Table I). Only the variables of month, location and the interaction of month X location showed significant effects in the analysis (Table I). A Duncan's multiple-range test conducted at the 0.05 level of probability indicated that maximal values for coot radiocesium body burdens occurred during January and February. Coot body burdens during these two months did not differ significantly from one another. Although not as great as those of January and February, the average body burdens of coots during December, April and March were significantly greater than those of October and November. Within each of these two latter groups of months, there were no significant differences in average coot body burdens (Fig. 2).

A similar Duncan's multiple-range test, conducted at the 0.05 probability level for location, indicated that coots from the North Arm had significantly higher body burdens than those from either the Hot or West Arms. Birds from the Hot and West Arms, however, did not differ significantly from each other. The high body burdens of coots from the North Arm were probably due to the contamination of this area with about 150 Ci of radiocesium as the result of reactor operations between 1961 and 1964 [7] . There is no such evidence, however, of current radiocesium contamination occurring in the Hot Arm of the reservoir as a result of on-going reactor operations. This same spatial pattern of differential radiocesium contamination would be reflected in the vegetation and other populations of sessile or less mobile animals inhabiting the reservoir, although confirmatory data are not yet available.

TABLE I. SUMMARY OF A MULTIPLE ANALYSIS OF VARIANCE FOR RADIOCESIUM BODY BURDENS OF COOTS FROM THE PAR POND RESERVOIR OF THE AEC SAVANNAH RIVER PLANT *

Source	df	F	Probability of Greater F
Body Weight	1	2.83	0.09
Age	2	1.78	0.17
Sex	1	0.39	0.54
Location	2	7.72	0.0009
Month	6	26.27	0.0001
Month X Location	12	2.22	0.01
Error	176		

*Based on partial sums of squares which have been adjusted for the covariant effect of body weight. First, second and third order interactions involving sex and age were not calculated because of low or unequal subclass replication in some of these categories as a result of the absence or scarcity of certain sex or age classes of coots on the reservoir during certain months.

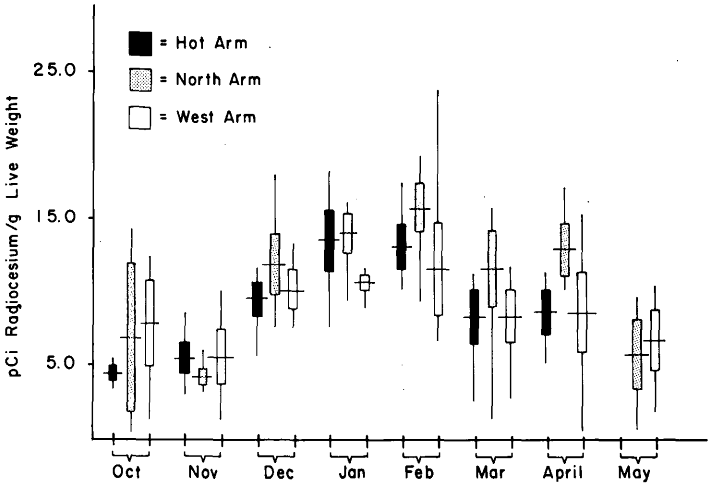

FIG. 2. Radiocaesium body burdens of American coots sampled from three different localities on the Par Pond cooling reservoir of the USAEC Savannah River Plant (Fig. 1). Birds were collected between October 1971 and May 1972. Horizontal lines represent means, rectangles represent plus or minus two standard errors and vertical lines represent the ranges. Means are based on sample sizes of ten birds each, except for those for October and May, which were 8/7/6 and 0/7/7 birds each, for the Hot Arm/North Arm/West Arm samples for these two months, respectively.

The interaction between the effects of month and location (Table I) is undoubtedly due to the fact that the differentially high body burden levels of coots from the North Arm are not apparent during the early fall months before the birds begin to show significant increases in their radiocesium levels (Fig. 2). A periodic shifting of the relative magnitudes of the average body burdens of coots from the Hot and West Arms throughout the year undoubtedly also contributes to the significance of the observed interaction between the variables of month and locality.

The failure of the age variable to show a significant effect upon coot radiocesium body burdens could be due to one of two factors. It is possible that few or no birds ever return to the Par Pond reservoir during successive years. The tendency of many such birds to show site faithfulness to specific wintering or nesting localities, however, would argue against this hypothesis. A more likely explanation would involve a sufficiently rapid turnover and elimination of radiocesium by the birds during their five month absence from the contaminated reservoir so that little if any residual contamination would remain in a bird returning to the area for the second or third year. Preliminary data suggests that the biological half-life of radiocesium is between 20 and 30 days in coots, thereby supporting this latter hypothesis.

Although coots are present on the Par Pond reservoir from October through May, several lines of evidence suggest that the same population contingent is probably not continuously present during all months. As pointed out by Rogers and Odum [13], changes in the ratio of body weight to wing length are often indicative of migratory status in a population of birds, high ratios indicating high fat deposition such as would usually occur immediately prior to migratory departure. The ratios of body weight to wing length showed homogeneity of variance ($X^2 = 11.91$; df = 7; $P > 0.05$) and a one-way analysis of variance indicated a significant effect of month upon this ratio for the coots ($F = 10.99$; df = 7,207; $P \leq 0.01$). There was an increase in this ratio throughout the early winter (Fig. 3), as might be expected for birds which had just completed their southward migration and were rebuilding body fat reserves prior to spring departure. The continuous decrease in this ratio from December through May, however, suggests that the birds present during that period were transients which were progressively nearer the end of their northward spring movements, which must have originated further to the south of the Savannah River Plant area. There was thus no evidence to indicate that these spring birds were simply winter residents which had not yet departed to the north.

Monthly changes in the sex ratios of the coots present on the reservoir (Fig. 3) suggest that coots, like many other species of waterfowl, tend to show differential migration patterns for the sexes, with males tending to remain in the more northerly parts of the wintering range (such as the Savannah River Plant area), while the females tend to move-on, wintering in more southerly parts of the range including southern Georgia and Florida [14,15]. Thus, the sex ratio of early arriving fall birds does not differ significantly from 50-50. Later into the fall, however, as the females continue to move south, the sex ratio begins to deviate significantly from a 50-50 ratio, with a maximum of 86.7% males being attained in February (Fig. 3). Later in the season, the sex ratio again falls as the predominately male population contingent apparently moves northward and is replaced, in turn, by the predominately female contingent which had been wintering further to the south. In testing for significant deviations in sex ratios, an initial contingency Chi Square test was conducted for all months ($X^2 = 20.46$; df = 8; $P \leq 0.01$), followed by Chi Square tests for individual months to which Yates' correction [16] was applied (Fig. 3).

The periodic change in population contingents of coots throughout the months, as produced by the differential migratory patterns described above, may help explain the seasonal variation in the radiocesium body burdens of these birds on the reservoir (Fig. 2). At the time of their fall arrival on the reservoir, most birds had low radiocesium levels due to the presumably low radionuclide contamination levels of their northern breeding grounds, relative to the Par Pond reservoir. During their winter stay on the reservoir, however, the predominately male wintering contingent increased steadily in

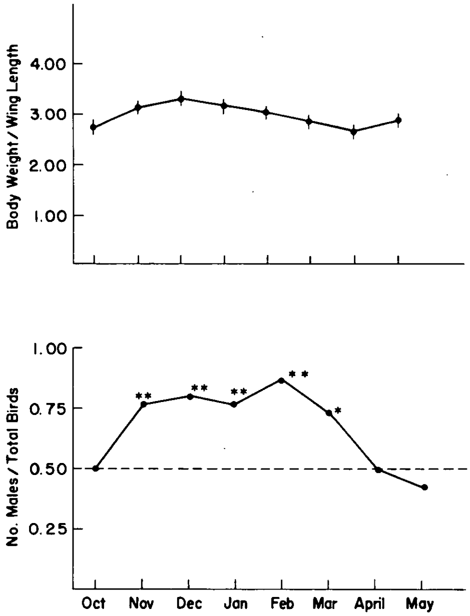

FIG.3. Changes in average body weight/wing length and sex ratios of monthly samples of American coots collected from the Par Pond cooling reservoir of the USAEC Savannah River Plant. Sample sizes were 30 birds for each month except October and May, which were 21 and 14 birds each, respectively. Data were calculated for all birds, pooled with respect to sampling locality within the reservoir. Sex ratios indicated with single and double asterisks differ from a 1:1 ratio at the 0.05 and 0.01 levels of probability, respectively. Vertical lines represent two standard errors above and below the mean.

radiocesium content until January or February at which time these birds apparently began moving north and were replaced, in turn, by increasing numbers of the predominately female population contingent of transient birds which had wintered further to the south in relatively uncontaminated habitats. These spring transient birds therefore had lower radiocesium levels and apparently began to have a diluting effect which continued to reduce, with increasing intensity, the average radiocesium level of the reservoir coot population throughout the late winter and spring months.

A linear regression analysis on body burden data pooled for all coots with respect to locality, sex and age indicated that between November and February, the winter resident birds increased their body burdens at an average rate of 2.69 ± 0.249 (SE) pCi/g live body weight/month ($r = 0.706$; df = 118; $P \leq 0.01$). This rate of body burden build-up compares favorably with information obtained from a single banded coot which was captured on the reservoir on December 10, 1971 and was released at the site of its capture on December 22, 1971 with a total radiocesium body burden of 1,911 pCi (4.04 pCi/g live weight). Ninety-four days later, this bird was shot while collecting the March coot sample and was found to have increased its total body burden during its period of release on the reservoir by 211%, or at an average rate of 1.80 pCi/g live weight/month. However, this rate or body burden increase could not be sustained indefinitely by the birds if they were to remain on the reservoir for an extended period of time. It is not yet possible to say, however, exactly how long would be required for this rate of body burden increase to diminish and begin to approach some as yet undetermined asymptotic level. A biological half-life of 20-30 days for radiocesium in coots would almost certainly suggest that most of the birds would begin to show decreases in the rate of body-burden build-up by the end of their 3-4 month stay on the reservoir. Pendleton and Hanson [17] have shown that waterfowl are somewhat slower than other inhabitants of aquatic communities, such as fish, to attain maximum radiocesium concentration levels. These authors suggest that maximum concentration factors appear in waterfowl flesh after 36-48 days of exposure, although no data are given for total body burdens, which presumably would require longer to approach asymptotic levels.

Seasonal trends were also apparent in the variability as well as the average coot radiocesium body burdens as indicated, for example, by the varying width of the 95% confidence interval about the means of the monthly samples (Fig. 4). When the variability of log-transformed data for coot body burdens was considered, minimal variability was found during the period of the late fall and early winter build-up. During this period, when the exchange of birds between the reservoir population and those in adjoining non-contaminated habitats is apparently minimal, the variability was only about 10% that of the late winter or spring periods when the dilution effect of incoming transient birds increases the population's total variability by mixing relatively non-contaminated birds with those which had accumulated relatively high body burdens during their winter stay.

The monthly fluctuations of coot body burdens on the reservoir was successfully simulated by the sine-wave equation used by Kolehmainen [18] to describe annual seasonal changes in the radiocesium contents of bluegill (Lepomis macrochirus) in a contaminated lake. The sine-wave transformation for this study was made in the form: Y = a + bX, where a = the annual mean body burden,

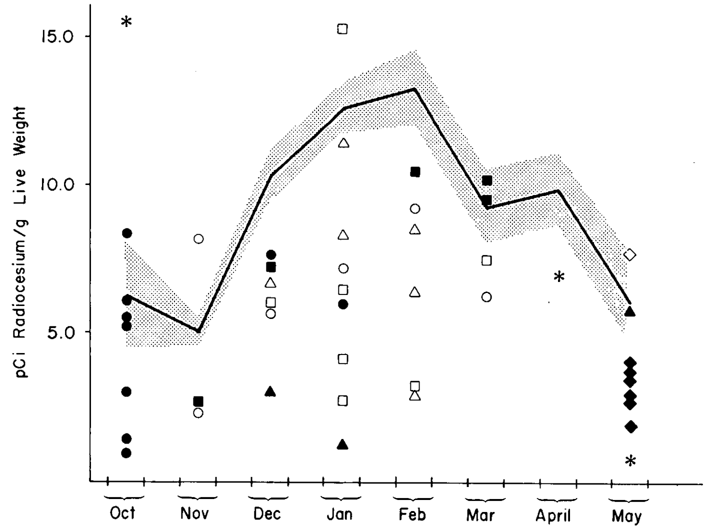

FIG.4. Radiocaesium body burdens of nine species of waterfowl and other aquatic birds collected from the Par Pond cooling reservoir of the USAEC Savannah River Plant at the same times as monthly samples of American coots. The solid line connects means and the shaded area represents the 95% confidence interval about the means for the monthly coot samples (Fig. 2). Asterisks represent common gallinules, solid circles pied-billed grebes, open circles horned grebes, solid triangles ring-necked ducks, open triangles lesser scaup ducks, solid squares bufflehead ducks, open squares ruddy ducks, solid diamonds common terns and the open diamond represents a black tern. Body burdens less than 3.00 pCi/g were generally considered to be indistinguishable from background (see text).

b = half of the amplitude and X is transformed into the form, $[\sin \frac{2\pi}{360}(t - t_m)]$, where t = the day of the year on which body burden is being determined, numbered consecutively from 1 January, and t_m = the day of the year upon which the sine-wave increases through the annual mean. In the present case, inspection of the data gave an initial estimate of 318 for t_m. A least-squares analysis gave a significant linear fit to the equation:

$$\text{pCi/g live weight} = 5.09 + 7.19\,[\sin \frac{2\pi}{360}(t - 318)]\ (r = 0.56;\ df = 213;\ P \geq 0.01)$$

Although the least-squares fit to this equation was statistically significant, it is possible that subsequent refinements of the estimate of t_m could give even better fits to the data. The sine-wave equation derived for the coots in the present study is in phase with the corresponding equation found by Kolehmainen [18], even though the factors producing the observed changes in body burdens were quite different in the two studies. Monthly changes in average coot body burdens were apparently due to changes in the individuals actually present in the population being sampled, while the fluctuations found by Kolehmainen for bluegill were attributable to seasonal changes in the feeding habits of the same individuals which were present throughout the year.

The coots seemed to show relatively higher levels of radiocesium accumulation than any of the nine other species studied (Fig. 4). Of the 46 individuals of other species, only 4 birds (8.7%) showed body burdens which exceeded the 95% confidence limits about the average coot body burdens for the same month, while 35 birds (76.1%) were lower than the coot confidence limits. The highest body burden of any individual other than a coot was shown by a common gallinule collected in October, at a time when this species is at the end of its period of summer residence on the reservoir. Other gallinules collected in the spring when they were newly arrived in the area were much lower in radiocesium content (Fig. 4). Watson et al. [19] point out that the feeding habits of the coot, in combination with the presence of shearing edges on its beak, result in its taking a high proportion of algae and other aquatic vegetation in its diet, with little dietary overlap between this species and ducks of either the diving or dabbling groups. Since algae is well known for its ability to accumulate radiocesium in relatively higher quantities than most other components of aquatic food webs [17,20], such dietary habits may help explain the higher body burdens of the coots relative to other species, most of which tend to incorporate a somewhat higher percentage of animal matter into their diets.

An estimate of the total impact of the Par Pond waterfowl community upon the redistribution of radiocesium from the reservoir may be made by assuming that approximately 5,000 coots and other waterfowl reside on the reservoir each winter [4], and that each bird, weighing on the average 500 g, departs with a maximum body burden of 15 pCi radiocesium/g live weight. Under such conditions, waterfowl should be expected to remove approximately 3.75×10^{-5} Ci from the reservoir annually and redistribute it elsewhere along their migratory pathways. Such an impact estimation is probably maximal, however, since many of the birds visiting the reservoir in the late spring months would not have an opportunity to accumulate body burdens as high as 15 pCi/g.

The general range of radiocesium body burdens showed by the coots and other waterfowl during January and February, including the maximal value for this study of 23.58 pCi/g shown by a coot in February, are well within the range of values reported by other workers for various species of wild game from the Savannah River Plant and southeastern United States Coastal Plain habitats in general [5,6]. Marter [1] reports a maximal value of 171 pCi radiocesium/g live weight for a green-winged teal (Anas carolinensis) collected from Par Pond. Even at this level, the radiocesium contents of the Par Pond waterfowl are well below the levels which would begin to suggest a health hazard to persons who may shoot and eat them as food [5]. Waterfowl collected from other contaminated habitats of the Savannah River Plant, however, have shown radiocesium body burdens as high as 923 pCi/g live weight [1].

Perhaps the most important aspect of the present study, however, is that it demonstrates the importance of the careful and detailed collection of background information concerning the basic ecology and biology of the birds being studied, as a prerequisite to the proper understanding and interpretation of radionuclide monitoring information. For example, simply monitoring the radiocesium body burdens of the first and last coots to arrive on the Par Pond reservoir would not have revealed any significant differences in the average body burdens of the birds. Failure to monitor monthly changes throughout the year or failure to understand the implications of the differential migratory patterns, feeding habits and other characteristics of the birds involved could have resulted in erroneous or misleading interpretations of the monitoring data obtained. While the fall-winter body burden build-up of the coots on the Par Pond reservoir is not now of any present health or safety significance, the proper understanding of the factors involved in this phenomenon could be of vital importance in the event of a subsequent radionuclide accident or spill which would greatly increase the degree of contamination of the wildlife of the area.

ACKNOWLEDGEMENTS

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DISCUSSION

B.K. BORISOV: Can you, on the basis of your studies, give separate estimates of the internal and the external radiation doses from radiocaesium suffered by birds living the whole year round on the reservoir investigated by you?

I.L. BRISBIN, Jr.: Basically, there are no waterfowl species that live the whole year round on the reservoir. The wood duck (Aix sponsa) is present throughout the year and breeds in adjacent swamp lands. Here again, however, we are not sure whether the same population contingent is present at all times of the year. We are now conducting intensive studies on this species and will also soon begin studies in which we will clip the wings of coots and thus force them to remain on the reservoir throughout the year.

To determine the internal and the external doses to such birds, we would have to obtain additional information on the radiocaesium concentrations of stomach contents, pond vegetation and other environmental components. We hope soon to have such information available for the wood duck study mentioned above.

S.V. KAYE: The results of your work are most interesting and you have conducted a comprehensive investigation of the natural history aspects of caesium transport by waterfowl. I should like to ask if concomitant measurements of caesium were made of water, algae and other aquatic plants each time waterfowl were sampled. If a relationship between waterfowl body burden and concentration in water or food could be worked out, this relationship could be incorporated into a predictive model. Has this extrapolation been done for your study at the Savannah River Plant?

I.L. BRISBIN, Jr.: The work which I have reported here is only a part of a larger team effort currently being undertaken at our laboratory. Other parts of this team study are investigating radiocaesium cycling as well as thermal effects upon other components of the biotic communities of this cooling reservoir and its adjoining swamp and reactor effluent streams. These other components now being investigated in this regard at our laboratory include the plankton community, aquatic vegetation and algae, macroinvertebrates, fish, turtles and even alligators. When the results of these investigations become available, particularly those concerning the vegetation and algae (which are the coot's principal food), and when we are able to conduct some laboratory studies on such subjects as coot food-intake rates, stomach content analyses etc., we will be able to provide the additional information that you have mentioned.

I entirely agree with you that the extension of this work to the point where it will eventually yield such predictive information, with direct relevance to the analysis of potential environmental impacts, represents a very important project, and we hope to undertake it in the near future.

F.O. HOFFMAN: What in your opinion is the feasibility of using the coot as a biological indicator of environmental radiocaesium originating from reactors using similar cooling reservoirs?

I.L. BRISBIN, Jr.: In principle, I am opposed to the concept of the 'indicator species' approach. All too often, I'm afraid this approach tends to encourage the feeling that there are cheap and easy solutions to problems of environmental monitoring (i.e. simply study one 'indicator' species and ignore the other system components). Such an attitude tends to divert interest and attention from the pressing need to conduct in-depth studies of all system components — which we now know to be the proper way to assess environmental impacts.

I realize, however, that frequently limitations of time and money force us to make environmental decisions on the basis of data available for only a few representative species. Under such conditions, and within all the limitations inherent in such an 'indicator species' approach, I think that the coot would serve as an excellent 'worst possible case' indicator of the radionuclide contamination of a waterfowl community. I could not say at the moment, however, how adequate an indicator the coot would be of contamination levels in other system components such as fish, vegetation or plankton. The answer to that question will have to await the completion of some of our other studies involving this reservoir system.

DISTRIBUTION ET INCORPORATION DU TRITIUM DANS LES ORGANES DE RUMINANTS

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Abstract–Résumé

DISTRIBUTION AND INCORPORATION OF TRITIUM IN THE ORGANS OF RUMINANTS.

A research program on the transfer of tritium in the food chain has been in progress for several years on the experimental farm of the Nuclear Energy Research Centre at Mol. Besides the contamination of grass and fodder by tritiated water and the incorporation of tritium into milk constituents, the authors have also investigated the distribution of tritium in the organs of ruminants contaminated in various ways. A cow was sacrificed 23 days after ingestion of 250 mCi of tritiated water in a single dose; three young male calves ingested 1 mCi of tritiated water daily for 25, 33 and 40 days respectively and were then sacrificed; three other calves ingested tritiated milk; a calf whose mother had received about 11 mCi THO per day for 40 days during gestation received mother's milk for 3½ months and was then sacrificed; finally, a pregnant goat was given water containing 46.6 μCi of ^{3}H/litre and the three kids were sacrificed at birth, at the age of one month and two months, respectively.

The various organs were removed and analysed for the content of ^{3}H in the tissue water and in the organic matter. Some DNA was prepared by the conventional method from the thymi, testicles and liver of young ruminants (calves and kids). The presence of ^{3}H was detected in the purified DNA. However, since there was a possibility of contamination of the DNA by traces of amino acids, the DNA was precipitated in a CsCl medium. Radioactivity was present in the peaks of DNA prepared from the thymi and testicles of a calf which had received 1 mCi THO/d since birth, and from the testicles of a calf which had ingested tritiated powdered milk daily (15 μCi/d). Deeper fractionation of the cell constituents of certain organs (thymi, testicles and liver) indicates the degree of incorporation of ^{3}H into lipids, RNA, proteins and DNA.

DISTRIBUTION ET INCORPORATION DU TRITIUM DANS LES ORGANES DE RUMINANTS.

Un programme de recherches sur le transfert du tritium dans la chaîne alimentaire est poursuivi depuis plusieurs années à la ferme expérimentale du Centre d'étude de l'énergie nucléaire à Mol. A côté des études sur la contamination des pâtures et des fourrages par l'eau tritiée ainsi que sur l'incorporation du tritium dans les constituants du lait, les auteurs ont aussi étudié la répartition du tritium dans les organes de ruminants contaminés de diverses façons. Une vache a été abattue 23 jours après une ingestion unique de 250 mCi d'eau tritiée; trois jeunes veaux mâles ont ingéré quotidiennement 1 mCi d'eau tritiée, respectivement pendant 25, 33 et 40 jours puis ont été sacrifiés; trois autres veaux ont ingéré du lait tritié; un veau dont la mère avait reçu environ 11 mCi THO par jour durant 40 jours au cours de la gestation a reçu le lait maternel pendant trois mois et demi puis a été sacrifié; enfin une chèvre en gestation a été abreuvée avec de l'eau contenant 46,6 μCi ^{3}H/l et les trois chevraux ont été sacrifiés respectivement à la naissance, à l'âge d'un mois et de deux mois.

Les divers organes prélevés ont été analysés en vue de déterminer la teneur en ^{3}H de l'eau des tissus, d'une part, et de la matière organique d'autre part. De l'ADN a été préparé, selon la méthode classique, à partir des thymus, testicules et foie de jeunes ruminants (veaux, chevreaux). Dans l'ADN purifié, la présence de ^{3}H a été observée. Cependant comme il existe une possibilité de contamination de l'ADN par des traces d'acides aminés, on a fait sédimenter cet ADN en milieu CsCl; de la radioactivité est présente au niveau des pics de l'ADN préparé à partir, d'une part de thymus et testicules de veau ayant reçu 1 mCi

THO/jour depuis la naissance, d'autre part de testicules de veau ayant ingéré journellement du lait tritié en poudre (15 μCi/j). Un fractionnement plus poussé des constituants cellulaires de certains organes (thymus, testicules, foie) indique le degré d'incorporation du ^3H dans l'acido-soluble, les lipides, l'ARN, les protéines et l'ADN.

1. INTRODUCTION

Un programme de recherches expérimentales sur le transfert du tritium dans la chaîne alimentaire est poursuivi depuis 1968 au Département de radiobiologie du Centre d'étude de l'énergie nucléaire à Mol.

Ces études trouvent leur justification dans le développement de l'industrie nucléaire qui a comme corollaire une contamination croissante du milieu par le tritium, rejeté essentiellement sous forme d'eau tritiée.

Après avoir étudié la contamination des pâtures et des fourrages par l'eau tritiée ainsi que l'incorporation du tritium dans les constituants du lait [1-5] notre effort s'est orienté vers l'étude de la répartition du tritium dans les organes de ruminants contaminés selon divers modes. Par ailleurs on connaît la radiotoxicité élevée de certaines molécules tritiées, il importe donc d'examiner si l'eau tritiée, au travers de certaines chaînes alimentaires, n'est pas susceptible d'entraîner la tritiation de molécules biologiquement importantes tels que les acides nucléiques [6, 7].

2. EXPERIENCES ET RESULTATS

2.1. Matériel et méthodes

2.1.1. Bétail et modes de contamination

a) Une vache de race ardennaise, en période de lactation, a été placée en étable pour expérimentation et a reçu une ration composée de foin, d'ensilage d'herbe et de pulpe séchée de betterave. L'eau d'abreuvage, distribuée par un abreuvoir automatique, a été contaminée au jour 0 de l'expérience par un pulse de 250 mCi de THO. Des échantillons de lait ont été récoltés les jours suivants pour étudier l'évolution de l'activité en tritium de l'eau du lait, de la matière grasse, de la caséine, du lactose et de la matière sèche totale.

L'abattage de l'animal a eu lieu 23 jours après l'ingestion unique de l'eau tritiée et certains organes ont été échantillonnés en vue de l'analyse radiochimique.

b) Six jeunes veaux mâles (numéros de code: I, II, III, IV, V, VII), de race ardennaise, d'âge variant entre 10 et 40 jours au début de l'expérience, ont été placés dans des cages spécialement conçues pour des études métaboliques. Un harnais pour la collecte des fèces a été placé à chacun des animaux, l'urine était récoltée en-dessous de la cage grâce à un entonnoir.

Trois des animaux [I, II, III], âgés de 10 jours au début de l'expérience, ont reçu matin et soir un volume connu de lait entier auquel était ajouté 10 ml d'eau tritiée dont la concentration était environ 50 μCi/ml. Les animaux ont été sacrifiés respectivement après 25, 33 et 40 jours d'ingestion continue de lait contaminé par l'eau tritiée, les activités ingérées étant respectivement de 25 630, 33 519 et 40 380 μCi de tritium.

Deux autres veaux [IV, V], âgés l'un de 18 jours et l'autre de 40 au jour 0 de l'expérience, ont reçu chacun, matin et soir, 125 g de poudre de lait tritié dissoute dans 1 litre d'eau déminéralisée. (La poudre a été fabriquée à partir du lait produit par une vache ayant ingéré au total 456 mCi d'eau tritiée en 40 jours.) La ration est complétée avec du lait «froid» de manière à atteindre le huitième du poids vif. L'activité moyenne de tritium lié organiquement ingérée journellement par chacun des veaux était d'environ 15 μCi. Les activités ingérées jusqu'au moment du sacrifice (à l'âge de 46 et 68 jours respectivement) ont été de 482,5 μCi pour chacun des veaux IV et V.

Enfin un veau [VII] âgé de 12 jours a consommé durant 25 jours du lait frais tritié provenant d'une vache recevant dans sa ration du foin de ray-grass contaminé (environ 950 μCi/j dont 750 μCi de ^{3}H lié organiquement); la teneur en ^{3}H du lait était, en moyenne, de 9 μCi/l dont le tiers lié à la matière organique.

Ensuite le jeune animal a ingéré, jusqu'à la date du sacrifice, du lait reconstitué à partir de la poudre produite par cette même vache nourrie avec du foin de ray-grass tritié. L'activité totale ingérée par le veau VII a été de 937 μCi dont 84 μCi sous forme de lait reconstitué.

c) Une chèvre [I] commune, en gestation depuis 45 jours, a reçu à partir de ce moment du tritium dans l'eau d'abreuvage à raison de 46,6 μCi/l. Sa ration était constituée de foin et d'ensilage de maïs à volonté, et de 800 g de tourteau par jour. L'ingestion totale de tritium jusqu'au moment de la mise bas de 3 chevreaux [I, II, III] intervenue 134 jours plus tard a été de 20 178 μCi. Le chevreau III a été sacrifié à la naissance. La mère a continué à recevoir de l'eau d'abreuvage tritiée durant le mois suivant la mise bas; les 2 chevreaux restants ont été nourris avec le lait contaminé de leur mère et la ration a été complétée avec une quantité connue de lait «froid» de vache.

Le chevreau II a alors (30 jours) été sacrifié, après ingestion de 570 μCi, et les organes ont été prélevés en vue de l'analyse radiochimique. Le chevreau I survivant n'a plus reçu que du lait «froid» en quantité connue afin de déterminer le taux de décontamination après un mois de ce régime; ces résultats ne figurent pas dans la présente communication.

2.1.2. Préparation des échantillons

Les organes des animaux, prélevés immédiatement après le sacrifice, sont placés en sacs plastiques immatriculés, pesés et conservés au surcongélateur en attendant la préparation en vue de l'analyse.

La préparation consiste en un dégel, toujours en sac plastique, suivi d'un broyage au moulin à viande. On prélève une aliquote pour la détermination de l'humidité (24 h à 110°C), une autre pour la mesure de l'eau tritiée et une troisième pour la mesure du tritium incorporé dans la matière sèche.

La séparation de l'eau tritiée des tissus s'effectue par distillation sous toluène, opérée dans un appareil Witeg pour le dosage de l'eau.

Le séchage des aliquotes d'organes est d'abord réalisé, dans une première étape, au moyen d'une étuve sous vide (trompe à eau), à une température de 80°C. La majeure partie de l'eau tritiée des tissus est ainsi évacuée, après dilution, dans le réseau d'égout «tiède». Le séchage final, à 110°C, est effectué dans une étuve placée dans une hotte bien ventilée. La préparation de l'ADN est décrite dans la section 2.4.

2.1.3. Mesure du tritium

Le tritium de l'eau des tissus est mesuré après dissolution de l'échantillon d'eau tritiée dans un mélange scintillant et application de la procédure habituelle utilisée pour les scintillateurs liquides.

Quant à la matière organique qui n'est pas susceptible de se dissoudre dans un mélange scintillant, il faut l'oxyder et recueillir l'eau de combustion qui est alors mesurée aisément. Cette oxydation est opérée soit dans un appareil à combustion (courant d'oxygène et piégeage de l'eau formée), soit dans un «Sample Oxidizer Packard 300» selon les cas. La première technique permet de connaître la quantité d'eau formée, ce qui conduit donc à exprimer les résultats en activité spécifique (Ci ^{3}H/g H).

2.2. Contamination des organes de ruminants ayant ingéré le tritium sous forme THO

2.2.1. Vache en lactation

Les résultats des mesures de l'activité en tritium de l'eau des organes et de la matière sèche sont indiqués dans le tableau I.

On constate que la répartition du tritium dans l'eau des organes est toujours assez homogène 23 jours après une ingestion unique. Par contre la contamination de la matière sèche varie d'un facteur 3 environ selon les

TABLEAU I. DISTRIBUTION DU TRITIUM DANS LES ORGANES D'UNE VACHE
23 jours après une ingestion unique de 250 mCi de THO

Organe	^{3}H sous forme d'eau		^{3}H lié organiquement		$\frac{\text{AS MS}}{\text{AS H}_2\text{O}}$
	nCi/ml H_2O	AS = nCi ^{3}H/g H	nCi/g matière sèche	AS = nCi ^{3}H/g H	
M. semitendinosus	57	515	11	155	0,30
Yeux	64	578	8	101	0,17
Pancréas	63	570	14	167	0,29
Cerveau	66	592	11	126	0,21
Intestin	62	560	9	106	0,19
Foie	65	588	25	368	0,63
Reins	65	582	19	255	0,44
Ovaires	65	588	20	281	0,48
Rate	66	595	20	276	0,46

AS, activité spécifique; MS, matière sèche.

organes considérés, le foie restant le plus contaminé. Le rapport des activités spécifiques (nCi ^3H/g H) de la matière sèche et de l'eau des tissus est relativement élevé dans certains organes (foie, ovaires, reins, rate).

Une estimation du taux de contamination résiduelle, après 21 jours, donne les valeurs ci-après: 6,72% de l'activité ingérée initialement serait encore présente dans l'eau des tissus, quant au tritium incorporé dans la matière sèche, il représenterait 0,56% de cette activité.

2.2.2. Veaux I, II, III

Le tritium présent sous forme d'eau dans les organes et tissus ci-après: reins, rate, cervelle, thymus, poumons, muscles divers, graisse, langue, foie, estomac, intestins et cœur, a été mesuré. Les valeurs extrêmes des activités spécifiques (nCi ^3H/g H) sont respectivement de 1013 (cœur) et 1123 (rate) chez le veau I ayant ingéré pendant 25 jours de l'eau tritiée; pour le veau III (33 jours d'ingestion) ces valeurs extrêmes sont de 948 (cœur) et 1092 (muscle), quant au veau I (40 ingestions) les valeurs sont respectivement 893 (graisse) et 980 (thymus).

TABLEAU II. REPARTITION DU TRITIUM DANS LES EXCRETATS ET ORGANES DE VEAU
(en ‰ de la quantité totale ingérée)

Répartition	Tritium administré sous forme			
	d'eau (veaux I, II, III)		de poudre de lait (veaux IV, V)	
	^3H lié organiquement	^3H forme THO	^3H lié organiquement	^3H forme THO
A. Excrétats				
Urine	584,8		498,0	
Fèces	0,3	6,6	4,7	3,7
B. Organes				
Reins	0,033	0,70	0,62	0,69
Rate	0,020	0,42	0,33	0,65
Cervelle	0,023	0,51	0,34	0,44
Poumons	0,051	1,15	0,78	1,90
Muscles	2,46	50,29	35,6	88,0
Langue	0,030	0,59	0,43	0,85
Cœur	0,046	0,72	0,90	1,02
Foie	0,14	2,55	3,44	3,50
Total organes	2,803	56,93	42,44	97,05

L'expérience s'étant déroulée en cage métabolique, les activités en tritium évacuées dans l'urine et les fèces ont pu être estimées. Cette estimation de la répartition (en ‰ de la quantité totale ingérée) du tritium est reprise dans le tableau II qui indique aussi les valeurs concernant le tritium lié organiquement (c'est-à-dire incorporé dans la matière sèche). On remarque que le tritium lié organiquement dans les organes ne représente que 5% du tritium présent sous forme d'eau dans ces organes et tissus.

2.2.3. Chevreau III

Les résultats de mesures du tritium dans les organes et tissus d'un chevreau dont la mère avait consommé, durant les 134 jours précédant la naissance, de l'eau contenant 46, 6 μCi ^{3}H/l sont mentionnés dans le tableau III.

On remarque d'une part que la concentration en tritium de l'eau des tissus est très uniforme et d'autre part que cette valeur n'est qu'environ les deux tiers de celle de l'eau d'abreuvage: il y a donc dilution par l'eau «froide» des aliments de la ration et l'eau résultant de leur oxydation. On constate aussi que l'incorporation du tritium dans la matière organique des organes de l'animal à sa naissance est très nette; le rapport des activités spécifiques (^{3}H organique/^{3}H eau) est d'environ 0, 5.

TABLEAU III. TENEURS EN TRITIUM DE L'EAU ET DE LA MATIERE SECHE D'ORGANES DU CHEVREAU III NE D'UNE MERE AYANT CONSOMME DE L'EAU TRITIEE (46, 6 μCi/l) DURANT LA GESTATION

Organes	^{3}H dans l'eau (nCi/ml H_2O)	^{3}H lié organiquement (nCi/g matière sèche)
Estomacs (parois)	30,85	10,95
Langue	31,14	9,68
Cœur	31,16	10,38
Petit intestin	30,79	9,70
Gros intestin	31,04	10,00
Muscle arrière	31,95	9,24
Muscle dos	31,80	10,09
Poumons	30,06	10,03
Foie	31,28	10,09
Cervelle	30,10	11,95
Reins	30,58	4,59

2.3. Contamination des organes de ruminants ayant ingéré le tritium sous forme organique (lait)

2.3.1. Veaux IV, V, VII

La concentration en tritium de l'eau des organes des veaux [IV et V] ayant reçu de la poudre de lait tritiée pendant 27 jours est assez constante d'un organe à l'autre: en effet les valeurs extrêmes des activités spécifiques varient de 14, 67 (cæcum) à 17, 13 (foie). Quant au tritium lié organiquement les concentrations (nCi/g matière sèche) dans les divers organes sont plus variables, les différences pouvant atteindre un facteur 3 entre le muscle (2, 06 nCi/g matière sèche) et le foie (5, 98 nCi/g matière sèche). Par ailleurs si l'on considère le taux d'incorporation du tritium dans la matière sèche (en ‰ de la quantité totale ingérée) on constate qu'environ 42 ‰ du ^{3}H ingéré est retrouvé dans les organes prélevés au moment du sacrifice (tableau II), ce qui représente 43% du tritium présent à ce moment, sous forme d'eau dans ces organes. Quant au veau VII, les résultats d'analyse de ses organes indiquent une distribution uniforme du tritium dans l'eau des tissus (valeurs variant de 45,18 à 47,73 nCi ^{3}H/g H).

2.3.2. Chevreau II

Les résultats de mesure des teneurs en tritium de l'eau des organes indiquent une répartition assez homogène (valeurs extrêmes, 126 nCi ^{3}H/g H à 157 nCi ^{3}H/g H).

Les valeurs des activités spécifiques de l'eau des tissus sont la moitié de celles observées chez le chevreau III (tableau III) sacrifié à la naissance mais la contamination du lait produit par la mère a aussi diminué de moitié durant cette période à la suite de l'introduction d'herbe fraîche dans la ration (40, 8 μCi/l lors de la mise bas, à 20, 1 μCi/l un mois plus tard).

2.4. Etude de l'incorporation du tritium dans l'ADN

2.4.1. Préparation de l'ADN

L'organe congelé est homogénéisé à l'Ultraturrax pendant environ 10 s dans une solution de saline-EDTA (NaCl 0, 15$\underline{M}$, EDTA 0, 1$\underline{M}$, pH 8,0). Pour 1 g de tissu nous employons 40 ml de saline-EDTA dans le cas du thymus, 20 ml pour les testicules, 10 ml pour le foie. A la suspension homogène on ajoute du sodium lauryl-sulfate (concentration finale 1%). L'action du détergent se manifeste par une augmentation immédiate de la viscosité.

On ajoute ensuite de la pronase (concentration finale 2 mg/ml); après agitation, la solution est incubée à 37°C pendant 2 h.

Les acides nucléiques sont ensuite précipités par addition d'alcool (2 volumes d'alcool pour 1 volume de solution). Le précipité fibreux est enroulé sur une tige de verre; il est ensuite remis en solution dans du SSC (saline sodium citrate) dilué (NaCl 0, 014$\underline{M}$, citrate de Na 0, 0014$\underline{M}$).

On ajoute ensuite de la ribonucléase (concentration finale 50 μg/ml) et la solution est incubée pendant 1 h à 37°C.

On ajoute de la pronase (concentration finale 2 mg/ml) et la solution est réincubée pendant 2 h à 37°C.

La solution est ensuite déprotéinisée par agitation pendant 15 min avec du chloroforme (vol. /vol.). Par centrifugation (30 min à 5000 tours/min) on sépare la solution aqueuse contenant l'ADN du chloroforme, les protéines formant une galette à l'interface. Le traitement par le chloroforme est répété jusqu'à ce que la solution aqueuse soit parfaitement claire. L'ADN est ensuite précipité du surnageant par addition de 2 volumes d'alcool. Les fibres sont récoltées sur tige de verre, l'alcool en est exprimé par pression sur les parois du tube. Le précipité est remis en solution dans du SSC dilué ou du NaCl 0,01M.

La quantité d'ADN est estimée par absorption dans l'ultraviolet à l'aide d'un spectrophotomètre enregistreur.

La radioactivité est estimée sur une aliquote par scintillation liquide.

Les échantillons sont ensuite analysés par ultracentrifugation en gradient de chlorure de césium. L'ADN obtenu par cette méthode est considéré comme «pur». La solution contient cependant quelques contaminants qui coprécipitent avec l'ADN en solution alcoolique: ce sont de l'ARN (environ 1%), des protéines ou acides aminés (2 à 3%) et d'autres composants comme le glycogène des polysaccharides dont il est difficile de se séparer si on veut préserver la structure macromoléculaire de l'ADN. Par cette méthode, le poids moléculaire de l'ADN peut être estimé à 8 à 10×10^6 daltons pour le thymus, de 6 à 8×10^6 daltons pour les testicules et de 1 à 3×10^6 daltons pour le foie.

Analyse par ultracentrifugation en gradient de chlorure de césium

A 4 ml de solution d'ADN on ajoute 5 g de chlorure de césium (Merck suprapur). On obtient ainsi une solution 5,651M CsCl dont la densité (1,699 g/cm^3) est très proche de la densité de l'ADN animal (1,700 g/cm^3). Après solubilisation 4,5 ml de solution contenant environ 100 µg d'ADN sont centrifugés dans des tubes en nitrate de cellulose.

Les tubes sont placés dans le rotor et sont centrifugés à 33 000 tours/min pendant 63 h à 25°C. Après centrifugation, chaque tube est placé successivement dans un appareil à fractionner. Le fond du tube est percé à l'aide d'une aiguille et environ 100 fractions de deux gouttes sont récoltées dans des fioles de comptage. L'élution a lieu sous une pression constante d'huile de paraffine et à une vitesse contrôlée à l'aide d'une seringue à vis micrométrique.

En général, 1 ml d'eau distillée est ajouté à chaque fraction paire et le spectre ultraviolet est enregistré à l'aide d'un spectrophotomètre enregistreur (Cary, modèle 14).

La radioactivité des fractions est estimée après addition de 75 ml d'Instagel Packard (ou d'un milieu scintillant équivalent de notre fabrication) à l'aide d'un scintillateur liquide (Packard 3380). En portant en ordonnée les valeurs de l'absorption ultraviolette et celles de la radioactivité et en abscisse le numéro des fractions, on obtient des distributions comme celles représentées dans les figures ci-après.

2.4.2. Résultats

Dans le tableau IV sont indiquées les activités spécifiques des ADN extraits des différents organes.

TABLEAU IV. ACTIVITE SPECIFIQUE (en dpm/μg ADN) DES DIFFERENTES PREPARATIONS D'ADN

		Thymus	Testicules	Foie
Veau	I	36 (8)	96 (2)	252
	II	28	44	84
	III	36	68	52
	IV	680	720 (4)	620 (148)
	V	580	212	36 (4)
	VII	220	204	52
	VIII	-	20[a]	
			16[b]	56
Chevreau	I	140	128	-
	II	162	32	-

Les valeurs entre parenthèses sont les valeurs corrigées, calculées sur les fractions après ultracentrifugation en gradient de CsCl.

[a] Activité spécifique de l'ADN extrait d'un testicule prélevé à la naissance.

[b] Activité spécifique de l'ADN extrait du second testicule prélevé au moment du sacrifice de l'animal.

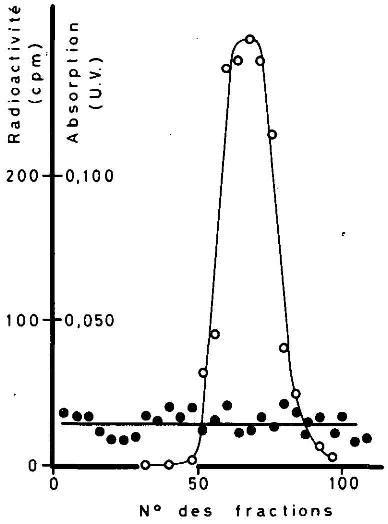

FIG.1. Ultracentrifugation en gradient de CsCl de l'ADN extrait du thymus du veau V.

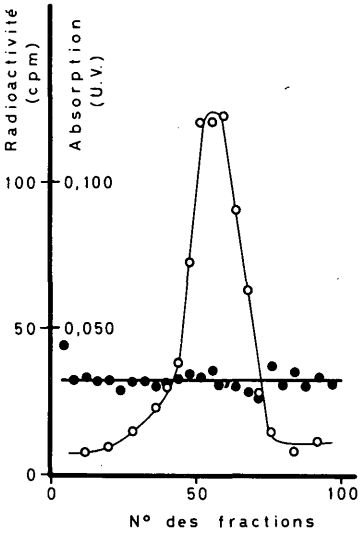

FIG. 2. Ultracentrifugation en gradient de CsCl de l'ADN extrait d'un testicule (naissance) du veau VIII.

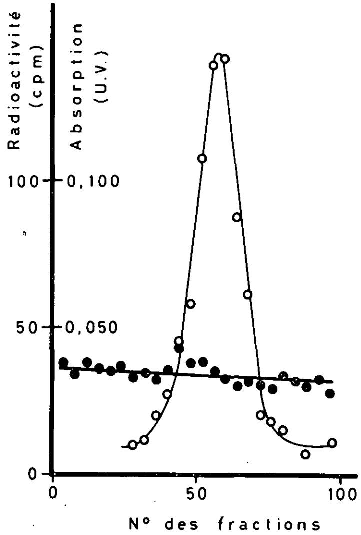

FIG. 3. Ultracentrifugation en gradient de CsCl de l'ADN extrait d'un testicule du veau VIII.

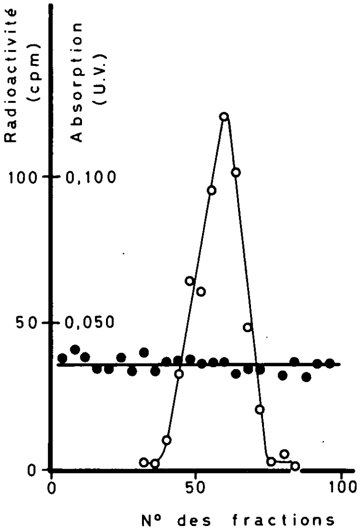

FIG.4. Ultracentrifugation en gradient de CsCl de l'ADN extrait des testicules du chevreau I.

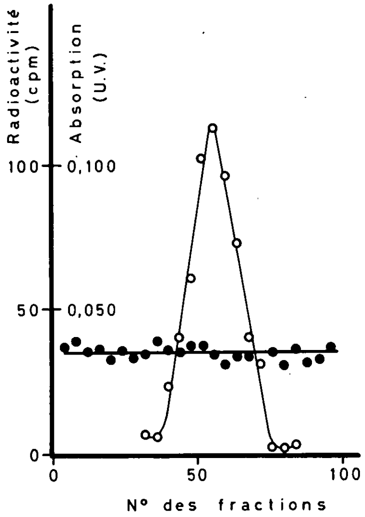

FIG.5. Ultracentrifugation en gradient de CsCl de l'ADN extrait des testicules du chevreau II.

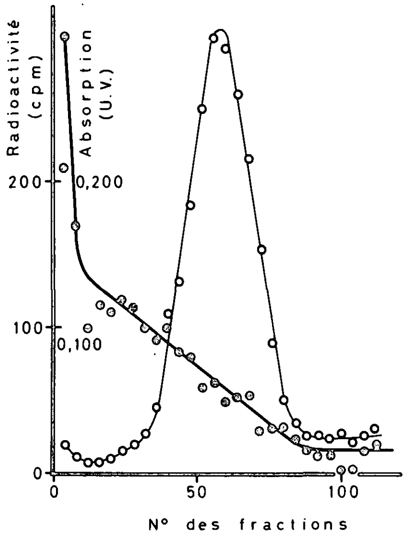

FIG. 6. Ultracentrifugation en gradient de CsCl de l'ADN extrait des testicules du veau II.

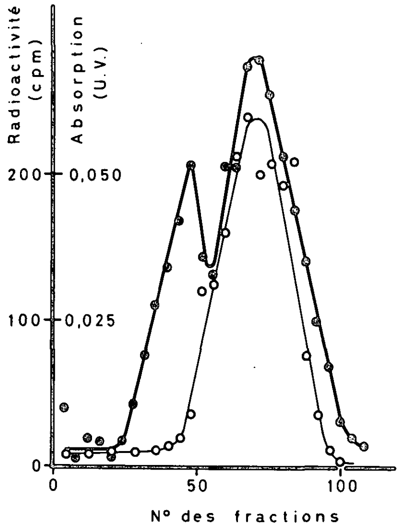

FIG. 7. Ultracentrifugation en gradient de CsCl de l'ADN extrait du thymus du veau I.

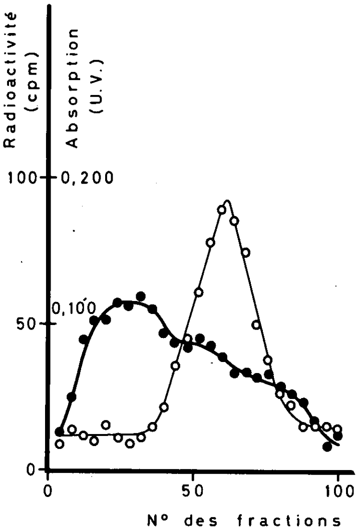

FIG. 8. Ultracentrifugation en gradient de CsCl de l'ADN extrait des testicules du veau I.

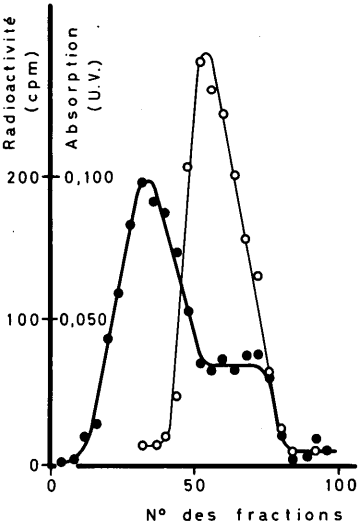

FIG. 9. Ultracentrifugation en gradient de CsCl de l'ADN extrait des testicules du veau IV.

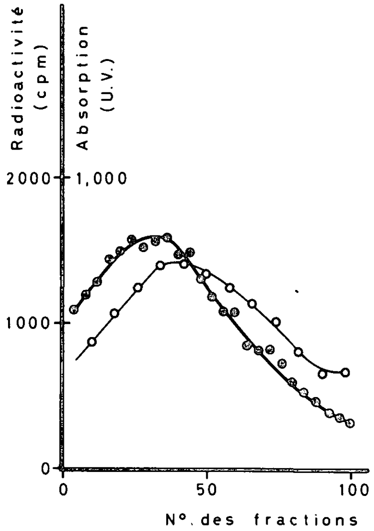

FIG.10. Ultracentrifugation en gradient du CsCl de l'ADN extrait du foie du veau IV.

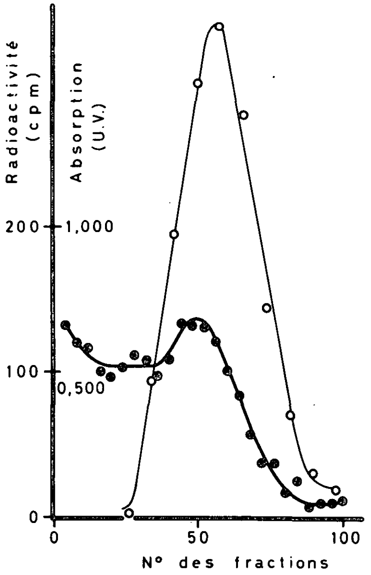

FIG.11. Ultracentrifugation en gradient de CsCl du DNA extrait du foie du veau V.

Les valeurs sont très différentes, non seulement quand on compare les organes entre eux mais également pour un même organe, sans qu'on puisse tirer de conclusions évidentes au sujet du traitement que les animaux ont reçu.

Après analyse des différents échantillons d'ADN par ultracentrifugation en gradient de CsCl, on observe trois types de résultats:

a) La radioactivité est étalée uniformément sur toutes les fractions du gradient. Cette radioactivité semble portée par des molécules qui coprécipitent avec l'ADN mais qui ne sont pas sédimentables en gradient de CsCl dans nos conditions expérimentales (fig. 1 à 5).

b) La radioactivité sédimente dans le gradient dans des positions différentes de celle de l'ADN caractérisé par son spectre ultraviolet.

Cette radioactivité peut être portée par des molécules qui sédimentent dans le fond du tube (ARN probablement, fig. 6) ou qui sédimentent dans une position proche de celle de l'ADN (fig. 7 à 11). Cette radioactivité est portée par des contaminants (glycogène, polysaccharides) qui coprécipitent avec l'ADN et dont la densité est voisine de celle de l'ADN.

c) Dans d'autres cas, les molécules radioactives ou du moins une partie de celles-ci sédimentent à une position qui correspond à celle de l'ADN (fig. 7 à 11).

3. DISCUSSION ET CONCLUSIONS

Une ingestion unique d'eau d'abreuvage tritiée provoque, chez la vache laitière, une contamination générale de l'eau des organes et tissus encore uniforme 23 jours après l'ingestion mais dont la concentration a baissé d'un facteur 10 environ. Le rapport des activités spécifiques de la matière sèche et de l'eau des tissus varie de 0,19 à 0,63 selon les organes, il reste donc toujours inférieur à l'unité. Ces valeurs sont à rapprocher de celle (0,40) observée dans le lait d'une vache 16 jours après une ingestion unique d'eau tritiée [2].

Selon la forme chimique du tritium ingéré (THO ou organique) par un jeune veau, la répartition du tritium dans l'eau ou la matière sèche des organes et fèces change. En effet dans le cas d'ingestion chronique d'eau tritiée on ne retrouve qu'environ 2,8‰ sous forme ^{3}H organique de la quantité totale ingérée alors que dans le cas d'ingestion de poudre de lait tritié dans la ration le taux d'incorporation dans les organes augmente d'un facteur 15 (42,4‰). On observe le même phénomène pour les fèces où le facteur 15 se vérifie (tableau II). Par contre l'excrétion urinaire du tritium est à peu près équivalente quelle que soit la forme chimique du précurseur tritié.

Dans le cas où l'animal consomme du lait produit par une vache consommant du foin tritié, lait contenant à la fois du tritium dans l'eau et dans la matière sèche (veau VII), le taux d'incorporation dans la matière sèche du foie augmente d'un facteur 40 environ par rapport au cas où le veau reçoit le tritium sous forme d'eau d'abreuvage, et dans le cas d'ingestion de poudre de lait tritié provenant d'une vache abreuvée avec de l'eau tritiée ce facteur est de 25.

A remarquer aussi que l'activité spécifique du foie de chacun des veaux I, II, III, abreuvés avec de l'eau tritiée et sacrifiés à une semaine d'intervalle, est restée inchangée du 25^e au 40^e jour.

Le chevreau, mis bas par une chèvre contaminée d'une façon chronique par l'eau d'abreuvage tritiée durant sa gestation, accuse une incorporation nette de tritium dans la matière sèche de ses organes (tableau III); cependant l'activité spécifique reste inférieure à celle de l'eau des tissus.

ADN

Par ultracentrifugation en gradient de CsCl, il a été possible de déterminer la nature exacte des molécules radioactives extraites par la méthode de préparation de l'ADN.

Sur les 25 échantillons analysés (tous radioactifs), dans 5 cas seulement on peut penser que la radioactivité ou une partie de celle-ci est portée par des fragments d'ADN: ADN de thymus du veau I (fig. 7), ADN de testicules des veaux I et IV (fig. 8 et 9), et ADN de foie des veaux IV et V (fig. 10 et 11).

Dans ces derniers cas, en analysant les fractions après ultracentrifugation, l'activité spécifique exacte de l'ADN a pu être estimée et elle est très différente de celle de l'extrait total. Cette radioactivité spécifique est en général peu élevée et ne semble pas en relation avec la vitesse de synthèse de l'ADN.

L'activité spécifique corrigée la plus élevée est observée pour l'ADN extrait du foie du veau IV et on sait que le foie est un tissu qui se divise très peu.

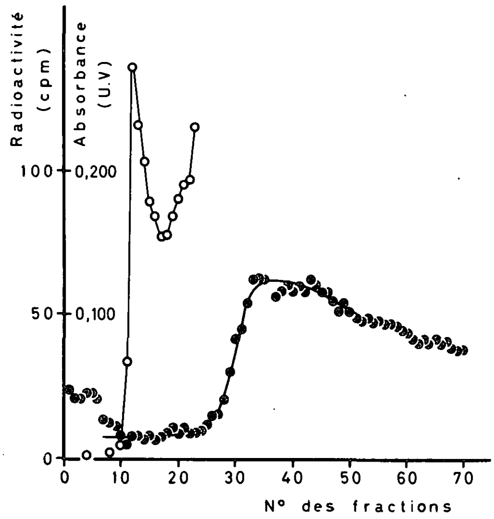

FIG. 12. Analyse par filtration sur gel d'agarose d'un homogénat de foie (veau IV).

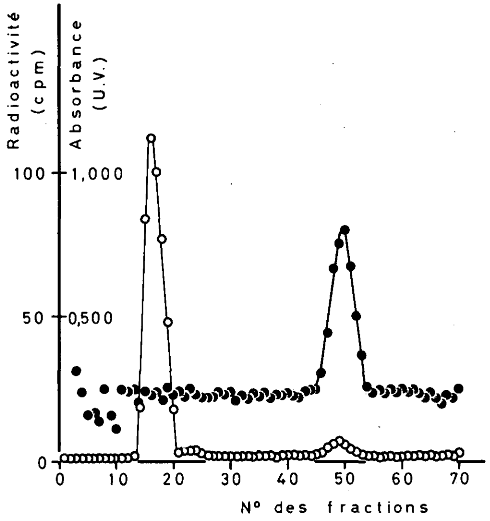

FIG.13. Analyse par filtration sur gel d'agarose de l'ADN extrait du thymus (veau I).

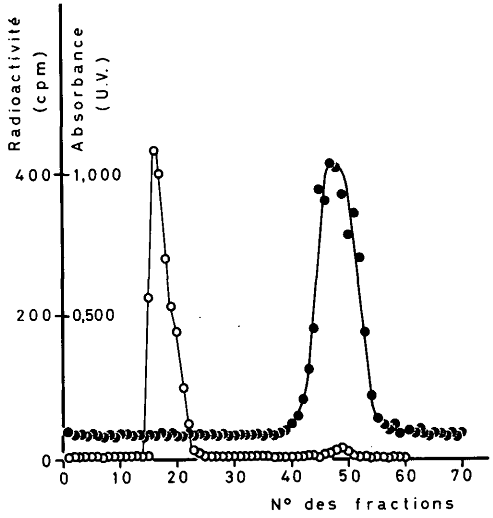

FIG.14. Analyse par filtration sur gel d'agarose de l'ADN extrait des testicules (veau IV).

Les échantillons où par analyse en gradient de chlorure de césium de la radioactivité était observée au niveau de l'ADN ont été analysés par filtration moléculaire sur gel de Sépharose 4B [8].

Les figures 12, 13, 14 montrent que pour les trois échantillons étudiés – ADN de foie (veau IV), ADN de thymus (veau I), ADN de testicules (veau IV) – par filtration moléculaire, la radioactivité est dissociée de l'ADN.

Il semble donc que le tritium n'est pas incorporé dans la molécule d'ADN. La radioactivité est portée par des molécules protéiques notamment très fortement liées à l'ADN (elles coprécipitent et cosédimentent avec la macromolécule) et seule la filtration moléculaire nous a permis de déterminer la nature des molécules radioactives.

REMERCIEMENTS

Nous avons vivement apprécié l'aide technique de MM. V. Adam, A. Bossus et E. Fagniart. Les analyses radiochimiques ont été effectuées par Mlle C.P.J. Oudenaarden et M. Verhoeven, que nous remercions également.

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ACCUMULATION OF TRITIUM IN VARIOUS SPECIES OF FISH REARED IN TRITIATED WATER

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Abstract

ACCUMULATION OF TRITIUM IN VARIOUS SPECIES OF FISH REARED IN TRITIATED WATER.

The release of tritium into aquatic ecosystems has resulted from nuclear industry operations. Because of the projected expansion of the nuclear power industry and associated fuel reprocessing plants, such releases can be expected to increase and to require further assessment of the environmental impact. Considerable information exists for the behaviour and fate of tritiated water in mammals; however, few experimental data are available on the incorporation of tritium from tritiated water into fresh water fishes. Of particular interest are fish that begin and end their lives in a tritiated environment.

In the present study trout and channel catfish eggs were hatched and the fish maintained in tritiated water for four months. In addition, two species of minnows native to the southwest United States of America were maintained in tritiated water and offspring from these fish were reared in tritiated water for five months. Some of these native fish were maintained for two months in large outdoor pools in which part of their food was grown naturally. The tritium concentrations in aquaria water were held constant during the experimental period. The results show that for these species of fish living in tritiated water the concentration factor for organic bound tritium is generally less than unity. The concentration factor is defined as the specific activity of tritium in dried body tissue divided by that in the water in which the fish were reared. Literature concerning the behaviour of tritium in aquatic food chains after release in nuclear industry effluents is reviewed.

1. INTRODUCTION

Substantial amounts of tritium have been released into the environment as a result of nuclear industry operations. Most of this tritium becomes incorporated into water molecules to form tritiated water (HTO) and can then be incorporated into the organic constituents of plants and animals which come in contact with the water [1, 2, 3].

Because of the projected expansion of the nuclear power industry and associated fuel reprocessing plants, the environmental releases of tritium can be expected to increase. Current design of fuel reprocessing plants is such that most of the tritium entering the plant in spent nuclear fuel is released to the environment as tritiated water [4, 5]. The capacity of the worldwide environment for diluting such tritium to negligible levels is not unlimited [6]. Furthermore, to the extent that such tritium is not rapidly diluted throughout the water of the world, the tritium concentrations in certain areas would be higher than estimated for rapid dilution in the entire hydrosphere. It has been reported that tritium released to the environment as tritiated water may be concentrated in some organisms through ecological food chains [7]. If these organisms are part of a food chain leading to man, the significance of environmental releases of tritium must be reexamined.

The accumulation of tritium in organisms through ecological food chains is being studied at the U.S. Environmental Protection Agency's

National Environmental Research Center at Las Vegas. Tritium accumulation in fish through aquatic food chains is one of the studies in progress. Considerable information exists for the behavior and fate of tritiated water in mammals [1, 2, 3, 8]; however, little experimental data are available on the incorporation of tritium from tritiated water into fresh water fishes. To determine the significance of tritium in the food chain of fish, tritium incorporation into fish from tritiated water alone must be evaluated. This report presents results for tritium incorporation into fish living in tritiated water and eating non-tritiated food. In an associated experiment, some fish were raised in tritiated water in which a substantial portion of their food was grown.

2. METHODS

2.1 Fish in aquaria

Small fish of two species native to the southwestern United States and suitable for rearing in small aquaria were collected from streams and placed in tritiated water. These species, Gamusia affinis (mosquito fish) and Poeciliopsis occidentalis (Gila topminnow), both of the family Poeciliidae, are live-bearing in that eggs are hatched within the mother and both present live young. Both species have a lifetime of about three years. Adult fish, one to two years old, were used and the species would not interbreed.

Fertilized trout (Salmo gairdnerii) eggs, obtained from a hatchery, were hatched in 380-liter (100 U.S. gal.) tanks with cooled and filtered circulating tritiated water. The trout were reared in this same tank to an age of 140 days and then placed in non-tritiated water for 72 days to determine the gross tritium loss rate from the fish.

Fertilized eggs of the channel catfish (Ictalurus lacustris), obtained from a catfish farm, were hatched in a tank identical to the trout tank, and fry were reared to adulthood in this tank. Trout and catfish were selected for study because they are used as food by man.

Young produced by mosquito fish and Gila topminnows were separated from the adults at birth and reared in different aquaria. Information was thus obtained on tritium incorporation into fish which start and end their lives in tritiated water. Some of these young fish were placed in non-tritiated water at an age of 200 days and periodic samples were collected to determine the rate of tritium loss from the fish.

The tritium concentration in all aquaria was maintained at a constant level throughout the study. Hatching of eggs and care of fish was performed as recommended in the literature [9, 10, 11]. Trout and catfish were fed a balanced commercial ration. The mosquito fish and Gila topminnows were fed a mixed diet of commercial foods for tropical fish. These commercial foods did not contain tritium incorporated in non-exchangeable positions [3].

2.2 Fish in outdoor pools

Six plastic-lined pools, constructed at the EPA Experimental Farm on the Nevada Test Site, were filled with 3700 liters (980 U.S. gal.) of tritiated water. Inorganic fertilizer, tritium and green algae (Chlorella pyrenoidosa) were introduced and the algae allowed to produce a full bloom. The tritiated water level in the pools was maintained by water level controllers connected to a reservoir of tritiated deionized water.

Windblown dirt, debris, and hay from the farm entered the pools, and a variety of insect life was observed in the pools before fish were introduced. While the algae and biota which spent a significant proportion of their lifetime in the pools would have considerable tritium metabolically incorporated into their tissues, the same would not be true for material which blew into the pools [2, 3]. Mosquito fish placed in the pools on several occasions resulted in complete mortality within a few days. Despite identical treatment, the pools developed individual characteristics after several weeks, and four pools then provided adequate conditions to maintain fish. Adult mosquite fish, one to two years old, were introduced into three pools, and young mosquito fish, four months old, into the fourth pool. The food supply from algae, insects, and windblown debris appeared to be adequate for these fish, and no additional food was provided. The pools were stocked in mid-July, and the fish harvested in mid-October when freezing temperatures threatened survival. Some of the adult fish were returned to tritiated laboratory aquaria and fed commercial fish food to determine the importance of the tritiated food in the environment.

2.3 Sampling and analysis

Water samples were collected from aquaria in the laboratory on a weekly basis, and farm pool water samples were collected about every two weeks on inspection visits. Tritium concentrations in aquaria and pools were maintained at 1 nCi/ml ±10 percent. This concentration was selected to provide easily measurable tritium levels and not yield unacceptably high radiation dose rates to fish. The fish received a dose of about 0.1 rad/yr [12].

Fish from pools and aquaria were collected periodically for analysis. Freeze-dried tissues of whole fish were oxidized in a Model 300 Packard Tricarb Sample Oxidizer and the water of oxidation analyzed for tritium.

All water samples were analyzed for tritium specific activity (μCi/gH or an equivalent unit) using a Beckman Model 100 Liquid Scintillation Counter. The scintillator solution contained 7g diphenyloxazole (PPO) and 1.5g p-bis-(o-methylstryl)-benzene (bis-MSB) dissolved in a mixture of p-xylene and Triton N-101 with a volume ratio of 2.75:1. Samples were analyzed by liquid scintillation techniques previously described [13, 14]. All samples were counted to at least ±2 percent at the 95 percent confidence interval.

3. RESULTS

3.1 Fraction of specific activity equilibrium

Results are presented to show the ratio between the tritium specific activity in freeze-dried fish tissue and that in water in which the fish were grown. A ratio, or fraction, greater than unity would indicate concentration of tritium within the organic constituents of fish relative to the water environment. No fraction greater than unity was measured in these experiments.

The results obtained for mosquito fish and Gila topminnows grown in tritiated water and fed a commercial diet are shown in Table I. The fraction of equilibrium attained is about 0.5 for both fish born in the tritiated

TABLE I. SMALL FISH GROWN IN AQUARIA AND FED COMMERCIAL FOOD: SPECIFIC ACTIVITY OF TRITIUM IN FREEZE-DRIED FISH TISSUE AS A FRACTION OF THAT IN WATER IN WHICH THE FISH WERE GROWN

SPECIES	AGE[a]	DAYS IN HTO	FRACTION[b]
Mosquito fish	180 d	21	0.42
"	1-2 yr	60	0.37
"	1-2 yr	66	0.31
"	1-2 yr	101	0.44
"	1-2 yr	106	0.47
"	1-2 yr	158	0.73[c]
"	1-2 yr	198	0.52
"	1-2 yr	203	0.52
Gila Topminnow	1-2 yr	79	0.56
Gila Topminnow	---	88	0.49
"	---	185	0.45

a. Age at time of introduction into tritiated water; dash (-) indicates fish were born in tritiated water.

b. Tritium specific activity in dry tissue divided by that in aquaria water.

c. These fish were inadvertently fed for three days with brine shrimp reared in tritiated water for a different experiment.

TABLE II. GAME FISH GROWN IN AQUARIA AND FED COMMERCIAL FOOD: SPECIFIC ACTIVITY OF TRITIUM IN FREEZE-DRIED FISH TISSUE AS A FRACTION OF THAT IN WATER IN WHICH THE FISH WERE GROWN

SPECIES	DAYS IN HTO[a]	FRACTION	COMMENT ON SAMPLE
Trout	1	0.27	Dead eggs and debris
	9[b]	0.34	Fry with yolk sac
	14	0.23	Dead fry and debris
	22[c]	0.22	Dead fry and debris
	32	0.39	Dead fry
	119	0.42	Fish
	140	0.42	Fish
Catfish	3[d]	0.51	Dead eggs
	49	0.43	Small fish
	53	0.45	Small fish
	62	0.58	Small fish
	133	0.41	Small fish

a. Fertilized eggs placed in tritiated water when received

b. All trout eggs hatched by day 9

c. Fry began eating well on day 22

d. All catfish eggs hatched by day 7

TABLE III. MOSQUITO FISH GROWN IN OUTDOOR POOLS: SPECIFIC ACTIVITY OF TRITIUM IN FREEZE-DRIED FISH TISSUE AS A FRACTION OF THAT IN WATER IN WHICH THE FISH WERE GROWN

AGE[a]	DAYS IN HTO	FRACTION
180 d	11	0.36
180 d	39	0.56
180 d	94	0.74
1-2 yr	20	0.54
1-2 yr	58	0.63
1-2 yr	93	0.80
1-2 yr	93	0.92
1-2 yr	93	0.88

a. Age at time of introduction into tritiated water.

water and fish introduced into tritiated water as adults. The data in Table II for trout and catfish indicate that a somewhat lower fraction of equilibrium is attained in these game fish grown in tritiated water and fed a commercial diet.

The data in Table III show that, for mosquito fish grown in outdoor pools in which a substantial fraction of their food was grown, the fraction of equilibrium attained is about 50 to 90 percent higher than for aquarium fish eating commercial food. The mosquito fish introduced into the pools as one- to two-year-old adults were full grown and did not increase in size during their sojourn in the pools. The fish introduced at 180 days of age, however, increased in body weight by a factor of three during the 94 days in the pool. As mentioned earlier (Section 2.2) the outdoor pools differed in the amount and type of biota present. The pool containing the young fish appeared to have very sparse populations of plant and insect life. Whether this is due to the fish keeping such populations sparse or to other unfavorable environmental conditions in the pool, it appears likely that the small fish ate a greater fraction of their diet from sources outside the pool than did the adult fish. Two of the three pools in which adult fish were kept contained lush growths of biota, and in the third pool populations of biota were intermediate between these two pools and that in which the young fish were raised. The fraction of tritium equilibrium attained in adult fish tissues in these pools (Table III) appears to be in proportion to the observed growth of biota in the pools. This fraction approached but did not exceed unity.

3.2 Tritium loss from fish tissue

After several months of growth in tritiated water while eating commercial fish foods, some of the trout and mosquito fish were transferred to non-tritiated water. Data on the loss of tritium from tissues of these fish are given in Table IV and Figure 1, curves C and D. The data for mosquito fish indicate that about 50 percent of the tritium in tissue is eliminated with a biological half-time of about five days, and the remainder is eliminated at a much slower rate. The two data points for trout (line D

TABLE IV. LOSS OF TRITIUM FROM TISSUES OF FISH GROWN IN TRITIATED WATER AND THEN TRANSFERRED TO NON-TRITIATED WATER

SPECIES	AGE[a]	DAYS IN H_2O	FRACTION[b]
Mosquito fish born in HTO	200 d	0	0.52
"	200 d	8	0.39
"	200 d	11	0.30
"	200 d	60	0.19
Trout hatched in HTO	119 d	0	0.42
"	119 d	72	0.18

a. Age at time introduced into non-tritiated water

b. Tritium specific activity in dry tissue divided by that in aquaria water during HTO exposure

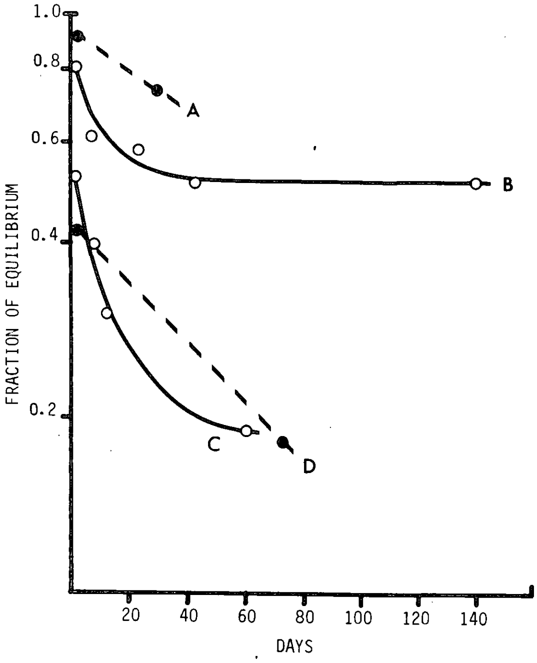

FIG. 1. Loss of tritium from fish after transfer to a less tritiated environment: A/B Mosquito fish from outdoor pools: to HTO; C Mosquito fish born in HTS : to H_2O; D Trout hatched in HTO : to H_2O.

TABLE V. LOSS OF TRITIUM FROM TISSUES OF FISH GROWN IN TRITIATED OUTDOOR POOLS AND THEN TRANSFERRED TO TRITIATED AQUARIA AND FED COMMERCIAL FOOD

SPECIES	AGE[a]	DAYS IN HTO[b]	FRACTION[c]
Mosquito fish adults	1-2 yr	0	0.92
"	1-2 yr	28	0.74
Mosquito fish adults	1-2 yr	0	0.80
"	1-2 yr	6	0.62
"	1-2 yr	22	0.58
"	1-2 yr	41	0.51
"	1-2 yr	139	0.52

a. Age at time transferred from outdoor pools to aquaria containing HTO
b. Days in aquaria containing same HTO concentration as outdoor pools
c. Tritium specific activity in dry tissue divided by that in aquaria water

in Figure 1) are sufficient only to indicate that not all the tritium in tissue is eliminated rapidly when the trout are placed in non-tritiated water. The two data points are connected by a dashed line to facilitate association of the data and not to represent an estimate of the tritium loss rate.

Adult mosquito fish from two of the farm pools were placed in tritiated water in laboratory aquaria. The aquaria water had the same tritium concentration as the farm pools; however, the fish food was changed from the food grown in the farm pools, plus some outside sources (Sections 2.2 and 3.1), to a commercial diet. The data for these fish are given in Table V and Figure 1, curves A and B. Only two samples were obtained for fish from the pool which had attained the highest fraction of equilibrium, 0.92. After 28 days in the aquarium the tritium concentration in these fish had decreased some 20 percent. These two data points are connected by a dashed line in the figure only to facilitate association of the data. The more interesting data are for the fish from the pool in which the fraction of equilibrium had reached 0.8 after 93 days in the pool. After 41 days in the aquarium the fraction of equilibrium in tissues of these fish had decreased to about that expected for fish grown in tritiated water and fed commercial food (Table I). The tissue concentration remained at this level after an additional 98 days in tritiated water.

4. DISCUSSION

4.1 Tritiated water exposure

The fraction of tritium specific activity equilibrium in tissue of small fish living in tritiated water (Table I) was found to be about 30 percent higher than that reported for tissue of mammals exposed to tritiated drinking water [1, 2, 3]. The reason for this difference is uncertain;

however, one explanation is that the small fish eat small biota which grow naturally even in clean aquaria. Intake of such organisms with tritium already incorporated into protein, fat, and carbohydrate molecules would tend to increase the tritium specific activity in tissues of the fish. This explanation is supported by the data for trout and catfish in Table II. The trout were raised in very clean and cold water (11°C) in which little biota grew. In addition, the trout ate large amounts of commercial food and grew rapidly to a size for which small aquatic biota would be expected to be a small portion of their total diet. The highest tritium specific activity in trout tissue was about 20 percent less than that for mosquito fish. The catfish grew very slowly during the first 60 days and possibly ate substantial amounts of biota growing in their warm (24°C) and relatively messy aquarium. By the time the catfish were 133 days old they were 7 to 8 cm in length, ate large quantities of commercial food, and kept their aquarium clean by devouring everything possible. From the very small fish at age 60 days to the much larger ones at age 133 days, the tritium specific activity in tissue decreased by approximately 30 percent to about the same level as for trout.

4.2 Influence of tritiated food

The consumption of food grown in their tritiated environment increases the tritium specific activity in tissue of mosquito fish to levels about 50 to 90 percent higher than in fish grown in tritiated water and fed commercial foods. Algae, which reproduce from carbon dioxide, water and inorganic materials in the water, incorporate tritium from tritiated water into cell components. The algae were introduced into pools to provide food for both fish and other aquatic organisms in the pools. The pools were filled initially with water from an irrigation reservoir which contained populations of unidentified aquatic organisms. Although food produced in this manner could have provided adequate food for the fish, other foods not produced in the tritiated environment (Sections 2.2 and 3.1) were probably eaten. The tritium specific activity in organic components of this material would not be in equilibrium with the aquatic environment and, therefore, would tend to lower the fraction of equilibrium in the fish. An example of this is the insect larvae developed from eggs in which all the nutrients are contained. If the adult insect producing the eggs lives and/or feeds in other than the tritiated pool environment, the larvae when hatched would contain little tritium bound to organic molecules. It should be noted that such introduction of food material from outside the tritiated water environment is a phenomenon which also occurs in natural waters.

4.3 Loss of tritium from fish tissue

Limited data are presently available on the loss of tritium from fish tissue after transfer to a less tritiated environment (Figure 1). It is clear, however, that a portion of tritium in tissue is not excreted rapidly. This finding is of significance in environmental monitoring in that the tritium concentration in fish tissue as a result of previous tritium exposures may be higher than in the ambient water. Furthermore, this elevated concentration may persist for longer than indicated by the data shown in Figure 1 because the fish are eating organisms from an entire food chain which has had a similar tritium exposure history. The organic tritium persistence in the food chain to the organism of interest is the major factor of importance, and limited data in the literature [15] indicate that such tritium persists in ecological cycles for several years.

5. CONCLUSIONS

5.1 Fish living in tritiated water

The experimental results indicate that for fish living in tritiated water and eating foods produced elsewhere, the tritium specific activity in tissue would reach about 50 percent of that in the water.

5.2 Fish living in a tritiated environment

The consumption by fish of foods grown in tritiated water where the fish are living increases the tritium specific activity in fish tissues significantly as compared to tissues of fish eating foods not grown in their tritiated environment. The fraction obtained by dividing the tritium specific activity in fish tissues by that in the water of their environment approached, but did not exceed, unity in the studies described.

5.3 Persistence of tritium in fish tissue

Tritium incorporated into fish tissue from tritiated water or from fish food grown in tritiated water is not excreted rapidly. Approximately 50 percent of this tritium appears to have a half-time of about five days, and the remainder has a much longer half-time in the mosquito fish studied.

Mention of commercial products used in connection with work reported in this article does not constitute an endorsement by the Environmental Protection Agency.

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DISCUSSION

O.L. van der BORGHT: For some heavy metals, such as ruthenium, we have observed high radioactive contamination localized in the gastro-intestinal tract of fish. We feel that this probably originated from the swallowing of tagged water, as the fishes were fasting during the experiments. Have you any indication regarding the quantity of water swallowed by the fish in your study?

R.G. PATZER: We did not have occasion to determine the amount of water taken into the gastro-intestinal tract by fish and I do not recall seeing anything on this subject in the literature. Our own observations on dissolved heavy metals, not documented in any reports, implicate fine particulate matter such as food particles, protozoa, algae, etc., as vehicles carrying adsorbed metal ions into fish.

R. KIRCHMANN: In the case of fish living in tritiated water but eating 'cold' food, the ratio of the specific activities (^{3}H tissue/^{3}H water) is about 0.5. What is the rate of incorporation of ^{3}H in the dry matter of the fish?

R.G. PATZER: All the results are presented in terms of relative specific activity of tritium, or activity per gram of hydrogen. Initially, the tritium appeared to be incorporated very rapidly into organic constituents up to a specific activity ratio of about 0.2 (tissue/fish). The tritium incorporation rate then appeared to decrease and to have an effective half-time of about 12 days.

G.N. KISTNER: What is the half-life of tritium in the second compartment (organic part) of the fish? Could it be in the region of 100 days?

R.G. PATZER: The data from these experiments are too limited for us to make a reliable estimate of second component half-life. This half-life would indeed seem to be between 60 and 120 days; but if excretion of tritium in fish is similar to that in mammals, then a third component of much longer half-life will be present. The data from the preliminary experiment described are not sufficiently comprehensive to permit identification of this anticipated third component. I am hesitant to assign a value to the second component because I suspect that the anticipated third component may exercise a strong influence on the data.

ETUDE DE LA RADIOCONTAMINATION DES POISSONS DE LA MEUSE

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Abstract–Résumé

STUDY OF RADIOCONTAMINATION OF FISH IN THE RIVER MEUSE

Since the construction of a nuclear power station at Chooz in 1967 the water, fauna and flora of the river Meuse have been sampled regularly in order to observe their radioactive contamination. The contents of ^{90}Sr, ^{137}Cs, ^{134}Cs, ^{60}Co and ^{54}Mn in particular have been investigated in the fish of this river, together with the distribution of these isotopes in the bone and muscle of bream. Since, generally speaking, the radioactivity of this species is low, various contamination experiments have been carried out in order to get a better idea of the radioactive contamination of fish. Minnows were chosen for the purpose of experiments. (a) 'Cold' fish were released downstream from the power station during a discharge period. After two weeks the above-mentioned gamma-emitters were investigated in the fish that had been exposed. (b) ^{60}Co and ^{54}Mn were studied more particularly, because they make a significant contribution to contamination of the water. Minnows contaminated artificially with 5 μCi ^{60}Co/litre or 5 μCi ^{54}Mn/litre for 7 days were transferred upstream from the power station. The values obtained after decontamination for 1 and 21 days were similar to those yielded by experiments performed entirely in the laboratory. (c) Under laboratory conditions the water of two aquaria was contaminated with a solution of effluents from the power station. The minnows took up especially ^{137}Cs, ^{134}Cs, ^{60}Co and ^{54}Mn in this experiment. ^{60}Co and ^{54}Mn were retained in a proportion close to that observed during contamination by radioisotopes added to the aquarium water in ionic form. The set of results obtained in the laboratory and under natural conditions is discussed.

ETUDE DE LA RADIOCONTAMINATION DES POISSONS DE LA MEUSE.

Suite à l'implantation d'une centrale nucléaire à Chooz en 1967, l'eau, la faune et la flore de la Meuse sont régulièrement échantillonnées pour suivre leur radiocontamination. Notamment, les teneurs en ^{90}Sr, ^{137}Cs, ^{134}Cs, ^{60}Co et ^{54}Mn ont été recherchées chez des poissons de cette rivière et la répartition de ces isotopes entre os et muscles a été étudiée chez des brèmes. D'une manière générale, la radioactivité de ces poissons étant faible, diverses expériences de contamination ont été effectuées pour mieux étudier la radiopollution des poissons; l'animal expérimental choisi est le véron. a) Des poissons ≪froids≫ ont été placés en aval de la centrale, durant une période de rejet. Après deux semaines, les émetteurs γ ci-dessus ont été recherchés dans les poissons exposés. b) Le ^{60}Co et le ^{54}Mn ont été plus spécialement étudiés, car leur contribution dans la pollution de l'eau est significative. Des vérons contaminés artificiellement soit par 5 μCi ^{60}Co/l soit par 5 μCi ^{54}Mn/l pendant 7 jours ont été transférés en amont de la centrale. Après 1 et 21 jours de décontamination, on obtient des valeurs semblables à celles provenant d'expériences entièrement menées en laboratoire. c) On a contaminé, en laboratoire, l'eau de deux aquariums par une solution d'effluents de la centrale. Les vérons incorporent notamment ^{137}Cs, ^{134}Cs, ^{60}Co et ^{54}Mn. Dans cet essai le ^{60}Co et le ^{54}Mn sont retenus dans une proportion voisine de celle observée lors d'une contamination par des radioisotopes ajoutés à l'eau des aquariums sous forme ionique. L'ensemble des résultats obtenus en laboratoire et en conditions naturelles est discuté.

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INTRODUCTION

La radiocontamination des poissons d'eau douce s'insère dans un problème très vaste: celui de la pollution de l'homme et de son environnement. L'un des volets les plus importants de la pollution est le rejet des déchets conventionnels et radioactifs. Malgré les efforts récents pour diminuer la quantité et la nocivité des rejets conventionnels, l'environnement a déjà subi les contrecoups de l'ignorance et de l'inconséquence de l'homme. La contamination radioactive de l'environnement, encore faible actuellement au regard de la contribution due au bruit de fond naturel, augmente d'année en année avec le développement de l'énergie nucléaire, d'où la nécessité de surveiller les sites des industries nucléaires et d'étudier les transferts de la radioactivité des rejets dans l'environnement.

En Belgique, la Meuse, qui représente une source d'eau potable pour la population, est le milieu récepteur des effluents liquides de basse activité de la centrale nucléaire de Chooz, située en France, à moins de 10 km de la frontière belgo-française. Le tableau I présente le niveau de contamination par certains radioisotopes des différents constituants de la Meuse, à la suite de la mise en service de la centrale nucléaire de Chooz en 1967. Si la radioactivité de l'eau reste relativement faible, en revanche la radioactivité des sédiments, de la flore et de la faune est plus élevée qu'avant le fonctionnement de la centrale. Sur ce site, les poissons accumulent une quantité suffisante de radioéléments pour présenter un intérêt particulier, étant donné leur rôle dans la chaîne alimentaire humaine.

1. ECHANTILLONNAGE DE POISSONS EN MEUSE

Régulièrement des poissons, ainsi que d'autres constituants de la Meuse, sont échantillonnés. Le tableau II reprend la concentration en radioisotopes de quelques poissons prélevés en aval de la centrale de Chooz. Les nucléides ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{54}Mn et ^{60}Co sont parfaitement détectables. Le rapport de concentration (tableau III) est de quelques centaines. Les valeurs maximales observées sont: 630 (^{90}Sr), 880 (^{134}Cs), 607 (^{137}Cs), 863 (^{54}Mn), 138 (^{60}Co), 24 (^{58}Co). En moyenne il est légèrement plus élevé pour le ^{90}Sr et les radiocésiums que pour le ^{54}Mn et les radiocobalts. Des valeurs analogues ont été observées par De Bortoli et coll. [1] dans le cas du ^{90}Sr et du ^{137}Cs provenant des retombées dans les lacs italiens. Cependant si l'on considère les concentrations de l'eau en ^{54}Mn et ^{60}Co au moment du prélèvement des poissons, les rapports de concentration pour ces nucléides doivent être réduits d'un facteur 150 pour le ^{54}Mn et 60 pour le ^{60}Co. Ceci est dû à la discontinuité et à la variabilité des rejets.

Les unités strontium (U.S.: pCi ^{90}Sr/g Ca, voir tableau IV) varient de 2 à 12 environ. Par ailleurs, la valeur du rapport ^{134}Cs/^{137}Cs de ces poissons est voisine de 1. Il en va de même pour l'eau de Meuse prélevée (0,76) et les effluents (0,72).

Ces résultats ont poussé à l'étude de la distribution de ces isotopes dans les organes. C'est ainsi que les os et les muscles d'un lot de brèmes, pêché pendant la même période que les poissons précédents, ont été mesurés séparément. Le tableau V reprend les résultats des analyses. Les radiocésiums contaminent fortement les muscles: ils sont aussi

TABLEAU I

VARIATION DE LA RADIO-ACTIVITE (pCi/kg poids humide ou/l) DE DIFFERENTS CONSTITUANTS DE LA MEUSE RECOLTES A HASTIERE DE PAR LA MISE EN SERVICE DE LA CENTRALE NUCLEAIRE DE CHOOZ (en avril 1967)

Constituants	Année de prélèvement	RADIO-ELEMENTS					
		^{54}Mn	^{60}Co	^{58}Co	^{134}Cs	^{137}Cs	^{90}Sr
Eau	1966	–	–	–	–	1,7	0,14
	1971	3	10	12,29	3,41	4,46	0,86
Boue sèche	1965	–	–	–	–	1700	140
	1971	30 000	30 000	50 000	15 500	60 000	92
Mousses	1965	–	–	–	–	60	250
	1971	89 000	38 000	84 500	17 700	9600	–
Animaux aquatiques (sauf poissons)	1965	7000	–	–	–	100	900
	1971	67 200	5800	9120	1100	1000	–
Poissons	1965	–	–	–	–	65	14
	1971	122	28	250	4850	8200	74

N.B.–Signifie : inférieure au seuil de détection ou non mesuré.

TABLEAU II

RADIO-ACTIVITE DES POISSONS EVISCERES DE LA MEUSE ECHANTILLONNES EN SEPTEMBRE 1971

(Résultats exprimés en pCi/kg frais)

ESPECES	RADIO-ELEMENTS					
	^{90}Sr	^{134}Cs	^{137}Cs	^{54}Mn	^{58}Co	^{60}Co
Goujons	30	610	400	900	non détecté	140
Goujons	38	2200	2100	1200	non détecté	210
Barbeaux	189	1200	1400	non détecté	non détecté	non détecté
Brèmes	51	1400	1300	200	non détecté	130
Rousses	120	1400	1500	200	non détecté	60
Brochets	17	3000	2700	600	300	non détecté

TABLEAU III

RAPPORT DE CONCENTRATION DES POISSONS EVISCERES DE LA MEUSE ECHANTILLONNES EN SEPTEMBRE 1971

(Calculs basés sur la moyenne annuelle de la concentration de l'eau)

ESPECES	RADIO-ELEMENTS					
	^{90}Sr	^{134}Cs	^{137}Cs	^{54}Mn	^{58}Co	^{60}Co
Goujons	100	178	90	647	non détecté	92
Goujons	127	645	471	863	non détecté	138
Barbeaux	630	352	314	non détecté	non détecté	non détecté
Brèmes	170	411	291	144	non détecté	86
Rousses	400	411	336	144	non détecté	39
Brochets	57	880	605	432	24	non détecté

TABLEAU IV

UNITES STRONTIUM ET RAPPORT $^{134}Cs/^{137}Cs$ DES POISSONS EVISCERES DE LA MEUSE, CAPTURES EN SEPTEMBRE 1971

ESPECES	^{90}Sr (pCi/g cendres)	Ca (mg/g cendres)	S.U. (pCi $^{90}Sr/g$ Ca)	$\frac{^{134}Cs}{^{137}Cs}$
Goujons	0,93	333	2,8	1,53
Goujons	1,3	289	4,6	1,05
Barbeaux	4,0	328	12,2	0,86
Brêmes	1,24	326	3,8	1,08
Rousses	2,61	314	8,3	0,93
Brochets	0,56	297	1,9	1,11

TABLEAU V

RADIO-ACTIVITE DES ORGANES DE BREMES ECHANTILLONNEES EN SEPTEMBRE 1971 EN MEUSE

RADIO-ELEMENTS	ORGANES	
pCi/kg frais ± écart quadratique	Muscles	Os
^{134}Cs	200 ± 20	420 ± 40
^{137}Cs	300 ± 6	220 ± 20
^{54}Mn	100 ± 30	230 ± 90
^{90}Sr	5 ± 3	70 ± 20
Ca (g/kg frais)	0,70	80
S.U. (pCi $^{90}Sr/g$ Ca)	6,5	0,9
$^{134}Cs/^{137}Cs$	0,65	1,9

accumulés (surtout le ^{134}Cs) par les os. Les os incorporent plus le ^{54}Mn que les muscles. Le ^{90}Sr est 10 fois plus concentré dans les os que dans les mucles; ceci est prévisible étant donné la forte quantité de Ca dans les os. L' unité strontium est voisine de 1 pour les os et de quelques unités pour les muscles. Le rapport $^{134}Cs/^{137}Cs$ est semblable pour les organes de brèmes et pour les poissons entiers.

Afin de faciliter l' interprétation des données obtenues, dans le cadre du programme de surveillance, on a été amené à réaliser des expériences tant en laboratoire qu' en milieu naturel. En effet, des échantillonnages sur

TABLEAU VI

COMPOSITION EN EMETTEURS γ DE L'EFFLUENT DE CHOOZ REJETE PENDANT L'EXPERIENCE DE CONTAMINATION EN MILIEU NATUREL

Résultats exprimés en nCi/l d'effluents ± écart quadratique de la moyenne

ISOTOPES	RADIO-ACTIVITE
^{144}Ce	≈ 11,5 ± 7,5
^{58}Co	⩽ 10
^{60}Co	4,75 ± 0,75
^{134}Cs	430 ± 4
^{137}Cs	490 ± 5
^{54}Mn	2,7 ± 1

Nota. Le facteur de dilution de l'effluent dans la Meuse est de 8×10^4 (facteur déterminé par calcul) pour le rejet en question.

le terrain ne conduisent qu'à des valeurs de rapports de concentration «apparents» étant donné l'irrégularité des activités rejetées au cours du temps, liée aux contraintes d'exploitation d'une centrale nucléaire. Le but visé est l'étude de la cinétique et du transfert d'un maillon à l'autre de la chaîne alimentaire de certains radioisotopes. L'attention a porté principalement sur le ^{54}Mn et le ^{60}Co, qui sont des radiocontaminants peu étudiés mais aisément décelables dans les effluents liquides radioactifs et dans les constituants de la Meuse (tableaux I et VI).

Pour les essais décrits ci-après, le véron (Phoxinus laevis, Cyprinidae) a été employé: c'est un omnivore de la Meuse et de ses effluents. En outre, étant de petite taille, il sert d'aliment aux poissons carnivores qui représentent le maillon de la chaîne trophique supérieure étudiée.

2. RADIOCONTAMINATION DES POISSONS INTRODUITS DANS LE MILIEU NATUREL

Un rejet a été effectué par la centrale nucléaire de Chooz dans la Meuse pendant près de 3 semaines (en automne 1972). La composition en émetteurs γ du rejet est reprise dans le tableau VI; le facteur de dilution en Meuse était d'environ 8×10^4.

Trois lots de 30 vérons chacun ont été constitués. L'un des lots a servi de témoin et est resté dans l'aquarium de stockage pendant toute la durée de l'essai. Les deux lots restants ont été transférés dans une cage fixée à une barque amarrée en Meuse à 8 km en aval de la centrale. La température de l'eau était de 15°C pendant la durée de l'expérience. L'un des deux lots a été relevé 4 jours et l'autre 2 semaines après l'immersion. Les 30 vérons de chaque lot ont été mesurés ensemble.

TABLEAU VII

COMPOSITION EN EMETTEURS γ DES VERONS INTRODUITS EN MEUSE PENDANT LA PERIODE DE REJET DE LA CENTRALE NUCLEAIRE DE CHOOZ

Résultats exprimés en pCi/kg de poids frais ± écart quadratique de la moyenne

Temps de séjour en Meuse	^{58}Co	^{60}Co	^{137}Cs	^{54}Mn
0 jour	< 270	400 ± 60	< 50	⩽ 321
4 jours	< 730	290 ± 40	< 145	4640 ± 240
14 jours	< 360	190 ± 20	< 98	1370 ± 160

TABLEAU VIII

COMPOSITION EN EMETTEURS γ DE LA SOLUTION D'EFFLUENTS REELS UTILISEE DANS LES ESSAIS EN LABORATOIRE

Radio-isotopes	quantité en nCi/l *
^{54}Mn	113 ± 10 *
^{60}Co	78 ± 20
^{137}Cs	82 ± 10
^{134}Cs	68 ± 8

*± $t_{0,05}^{n-1}$ déviation standard

TABLEAU IX

RADIO-ACTIVITE DES POISSONS A JEUN EN FONCTION DU TEMPS, SUITE A UNE CONTAMINATION PAR UN EFFLUENT REEL (PWR)

(Résultats exprimés en nCi/g poisson ± $t_{0,05}^{n-1}$ déviation standard)

Radio-isotopes	Jour après le début de la contamination		
	1	4	11
^{54}Mn	0,29 ± 0,55	0,091 ± 0,054	0,48 ± 0,54
^{60}Co	1,03 ± 2,18	0,220 ± 0,006	0,028 ± 0,014
^{137}Cs	0,021 ± 0,015	0,041 ± 0,018	0,092 ± 0,042
^{134}Cs	0,08 ± 0,17	0,026 ± 0,022	0,082 ± 0,065

Le tableau VII montre que seul le ^{54}Mn est accumulé de façon notable par ces poissons. Les valeurs sont plus élevées au jour 4 (4640 pCi/kg de matière fraîche) qu'au jour 14 (1370 pCi/kg de matière fraîche). Cela indique que, dans la nature, l'accumulation du ^{54}Mn n'est pas linéaire: elle présente un maximum au bout de quelques jours. En laboratoire, ce maximum est au jour 7. Peut-être aussi le ^{54}Mn se trouve-t-il en quantité moindre dans les effluents à la fin du rejet qu'à son début. Les autres radioisotopes recherchés ne se retrouvent pas en quantité suffisante dans les poissons pour être mesurables.

3. CONTAMINATION EN LABORATOIRE PAR UN EFFLUENT REEL

Les deux aquariums utilisés renfermaient 8 litres de liquide: 2 litres de la solution d'effluents et 6 litres d'eau de ville; la radioactivité dans ces aquariums est mentionnée dans le tableau VIII. Le pH final était de 5,5 à 6. L'eau et la solution radioactive étaient renouvelées tous les 4 jours. Huit vérons, ayant jeûné préalablement pendant une semaine, ont été introduits dans chaque aquarium au jour 0: ils sont restés ensuite à jeun pendant toute la durée de l'essai.

On constate que les poissons sont contaminés dès le jour 1 par les radioisotopes présents dans le mélange d'effluents employé (tableau IX, première et deuxième colonnes). La pénétration des isotopes dans les vérons n'est pas régulière au cours du temps, sauf peut-être celle du ^{137}Cs. Le rapport de concentration (tableau X) oscille entre 0,38 (^{134}Cs au jour 4) et 13,22 (^{60}Co au jour 1). Il est plus élevé pour le ^{54}Mn et le ^{60}Co que pour les deux radiocésiums.

Une comparaison a été établie entre, d'une part, les résultats obtenus en laboratoire à l'aide d'un mélange d'effluents et, d'autre part, ceux provenant d'essais dans les mêmes conditions avec du ^{60}Co et du ^{54}Mn sous forme ionique. L'examen des résultats (tableau X) indique que le ^{60}Co et le ^{54}Mn sont absorbés en proportions semblables dans les deux types d'expériences. Une fraction considérable des radioisotopes dans les effluents réels est probablement sous forme physico-chimique complexe.

TABLEAU X

RAPPORT DE CONCENTRATION DE LA CONTAMINATION EN LABORATOIRE PAR DES EFFLUENTS REELS ET PAR DES ISOTOPES SOUS FORME IONIQUE

Isotopes	Jour d'échantillonnage					
	Effluents réels			Isotopes sous forme ionique		
	1	4	11	1	4	11
^{54}Mn	2,58	0,81	4,24	0,2	3,3	12,9
^{60}Co	13,22	2,82	0,36	0,4	1,7	1,7
^{137}Cs	0,26	0,50	1,12			
^{134}Cs	1,22	0,38	1,21			

Nassogne [2] et Piro [3] constatent qu'un isotope sous forme complexe n'est pas absorbé par les êtres vivants. La variabilité dans les résultats rend les différences éventuelles difficiles à mettre en évidence.

4. DECONTAMINATION DANS LA NATURE DE VERONS CONTAMINES PAR DU ^{54}Mn ET DU ^{60}Co SOUS FORME IONIQUE

Etant donné les difficultés que présente l'emploi d'effluents réels comme contaminant (composition chimique, forme physico-chimique des radionucléides présents, etc.), il a paru préférable d'employer des émetteurs γ sous forme ionique pour l'étude de la décontamination.

Schéma expérimental. Six aquariums renfermant 10 litres d'eau + 10 vérons + 5 μCi ^{54}Mn/l, et six aquariums renfermant 10 litres d'eau + 10 vérons + 5 μCi ^{60}Co/l ont été mis simultanément en expérience, la température moyenne étant de 11°C. Les poissons étaient nourris pendant toute la durée de l'expérience. La période de contamination choisie est de 7 jours, car une expérience précédente a montré qu'il fallait 7 jours pour obtenir une contamination maximale des vérons en ^{54}Mn. L'eau et les radioisotopes des aquariums étaient renouvelés au jour 5 de la contamination. Un lot de 10 vérons était échantillonné aux jours 5 et 7, ceci pour chaque radioisotope.

Le jour 7, les vérons restants ont été transférés en amont de la centrale nucléaire de Chooz. Pour chacun des deux radioisotopes étudiés, trois lots de 13 vérons ont été introduits dans une cage et immergés dans la Meuse. Aussi bien pour le ^{54}Mn que pour le ^{60}Co, un lot de poissons a été relevé aux jours 1, 7 et 21 après le début de la décontamination.

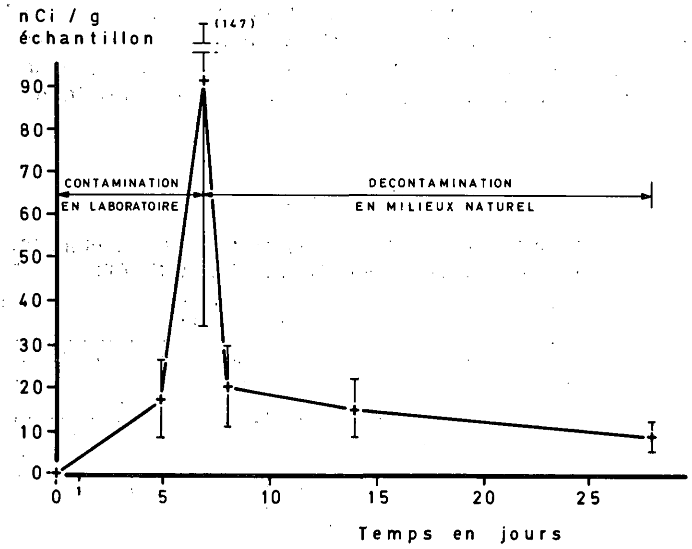

FIG. 1. ^{54}Mn: Décontamination en Meuse de poissons nourris et contaminés au laboratoire par 5 μCi ^{54}Mn ionique/litre d'eau.

TABLEAU XI

$$\left(\frac{^{54}\text{Mn ou }^{60}\text{Co du poisson entier nourris en nCi/g}}{^{54}\text{Mn ou }^{60}\text{Co de l'eau au jour 0 en nCi/ml}}\right),$$

SUITE A UNE CONTAMINATION EN LABORATOIRE ET LORS D'UNE DECONTAMINATION A HAM-SUR-MEUSE

Temps après la (dé)contamination	^{54}Mn	^{60}Co
1) Contamination		
5 jours	3,586	2,063
7 jours	19,108	5,281
2) Décontamination		
1 jour	4,195	1,096
1 semaine	3,107	0,381
3 semaines	1,787	0,056

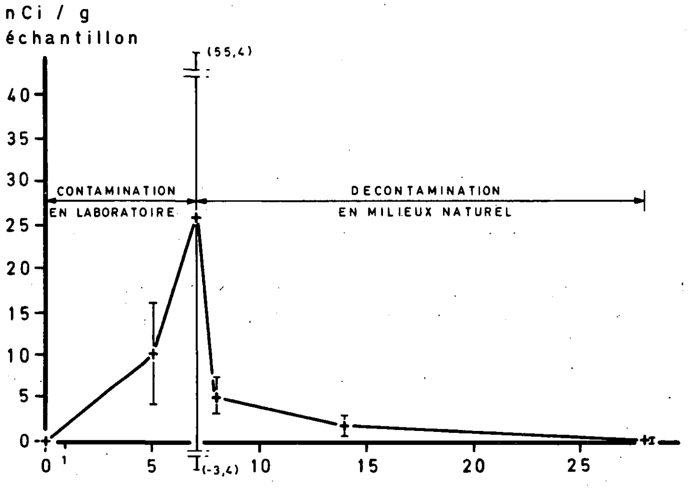

FIG. 2. ^{60}Co: Décontamination en Meuse de poissons nourris et contaminés au laboratoire par 5 μCi ^{60}Co ionique/litre d'eau.

La figure 1 (résultats exprimés en nCi/g de matière fraîche) montre l'accumulation rapide du ^{54}Mn pendant les 7 jours de contamination. A ce moment, le rapport de la radioactivité des poissons (en nCi/g de matière fraîche) à la radioactivité initiale de l'eau (en nCi/ml) est de 19 (tableau XI). Mais le rapport de concentration s'élève jusqu'à 32 (tableau non repris): ceci s'explique par la diminution enregistrée du ^{54}Mn dans l'eau.

Dès le début de la décontamination (pendant les premières 24 heures) en Meuse, la radioactivité diminue fortement (figure 1 et tableau XI).

TABLEAU XII

DONNEES PRINCIPALES CONCERNANT LA CONTAMINATION DES POISSONS ENTIERS NOURRIS (par 5 µCi/l de ^{60}Co ou de ^{54}Mn ioniques) EN LABORATOIRE ET LEUR DECONTAMINATION

	1ière expérience		2ième expérience	
	^{54}Mn	^{60}Co	^{54}Mn	^{60}Co
Jour de contamination maximale enregistrée	7ième	4ième	7ième	7ième
Radio-activité maximale enregistrée en nCi/g	70,7	52,6	92,0	26,0

	Temps (jours)	Décontamination			
		au laboratoire		à Ham-sur-Meuse	
		^{54}Mn	^{60}Co	^{54}Mn	^{60}Co
Radio-activité résiduelle (en % du maximum)	1ier	12,0	13,6	22,0	20,8
	17ième ou 21ième*	6,7	7,2	9,3	1,1

* 17ième jour pour la décontamination en laboratoire
21ième jour pour la décontamination à Ham-sur-Meuse.

Par la suite, la radioactivité ne baisse que très lentement. Cette courbe, semblable à celle obtenue lors d'expériences menées en laboratoire, montre que la décontamination de ces poissons se fait selon deux composantes au moins.

Quant à la contamination des vérons par le ^{60}Co (fig. 2), elle présente la même silhouette que celle relative au ^{54}Mn. La radioactivité enregistrée au jour 7 est plus faible pour le ^{60}Co que pour le ^{54}Mn: 26 nCi/g de matière fraîche au lieu de 92. Le rapport maximal du ^{60}Co dans le poisson (en nCi/g de matière fraîche) au ^{60}Co dans l'eau (en nCi/ml) (tableau XI) est donc plus faible pour le ^{60}Co que pour le ^{54}Mn: 5 au lieu de 19 au jour 7. Le rapport de concentration (voisin de 5, tableau non repris) montre que le ^{60}Co reste dans l'eau.

Pendant les premières 24 heures de décontamination, la vitesse de désorption en ^{60}Co est similaire à celle enregistrée pour le ^{54}Mn (figure 2 à comparer à la figure 1). Après cette période, la vitesse de décontamination faiblit: elle reste toutefois plus élevée dans le cas du ^{60}Co que dans le cas du ^{54}Mn. Il semble donc que, comme pour le ^{54}Mn, la décontamination des poissons renfermant du ^{60}Co s'effectue selon deux composantes au moins. Reed [4] tire une conclusion similaire à la suite d'expériences sur le poisson-chat.

Le tableau XII donne les résultats comparatifs de cette expérience et d'une autre qui s'est complètement déroulée en laboratoire. Le ^{54}Mn et le ^{60}Co accumulés par les poissons pendant la période de contamination est comparable pour les deux types d'expériences. Ceci montre que, malgré le taux élevé de variabilité observé, il existe une certaine reproductibilité dans ce type d'expérience.

La décontamination démarre plus lentement en milieu naturel qu'en milieu artificiel. Mais le ^{54}Mn résiduel est en quantité sensiblement semblable pour les deux essais: 7 et 9% respectivement pour la décontamination en laboratoire et celle en amont de la centrale de Chooz. Par contre, il reste plus de ^{60}Co dans les poissons décontaminés en laboratoire que dans ceux remis dans leur milieu naturel (7 et 1% respectivement).

CONCLUSIONS

L'investigation menée permet de dégager les points suivants:

1) La contamination des poissons de Meuse par les rejets d'effluents liquides est mesurable.

2) Cette absorption de radioisotopes par les poissons est relativement faible en milieu naturel et ne pose pas de problème sanitaire. Toutefois les valeurs maximales du rapport de concentration «apparent» poisson/eau observées sont de quelques centaines pour les radioisotopes enregistrés (^{90}Sr, ^{134}Cs, ^{137}Cs, ^{54}Mn et ^{60}Co). Etant donné l'irrégularité dans la concentration radioactive des rejets, ces valeurs ne peuvent pas être considérées comme une estimation valable des rapports de concentration «réels».

3) La contamination expérimentale des poissons par du ^{54}Mn et du ^{60}Co provenant d'effluents réels suit une courbe relativement semblable à celle de ces deux radioisotopes sous forme ionique. La décontamination de ^{54}Mn et de ^{60}Co ioniques de ces poissons a une allure similaire, qu'elle se fasse en laboratoire ou en Meuse. A noter que dans les expériences en aquarium, il y a lieu de tenir compte de la fixation au cours du temps du ^{60}Co et du ^{54}Mn par les parois des aquariums.

REMERCIEMENTS

Les auteurs remercient les collaborateurs ayant participé à l'échantillonnage et à la préparation des échantillons, principalement Mme E. Bonnijns-Van Gelder et M. E. Fagniart. Ils expriment aussi leurs remerciements aux membres des sections «Mesures bas-niveau» et «Spectrométrie γ» pour l'exécution des mesures de nombreux échantillons. Les données concernant les rejets de la centrale de la SENA proviennent entre autres de renseignements fournis par le SCPRI. Nous exprimons notre reconnaissance à M. le professeur Pellerin, directeur du SCPRI. Les administrations de la Meuse Namuroise (Ponts-et-chaussées) nous ont offert toutes facilités pour la réalisation de nos prélèvements.

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DISCUSSION

E. KUNZ: What is your explanation for the substantial difference in ^{90}Sr content in relation to calcium in bones and muscles, as shown in Table V? Under conditions of chronic and relatively constant exposure it seems to me that it should signify a strong discrimination process between bone and other tissues.

R. KIRCHMANN: The data in the literature on the distribution of stable Ca and Sr in the various parts of the fish organism do not in fact indicate an Sr/Ca discrimination.

The results given in Table V are derived from fish caught under natural conditions where the presence of ^{90}Sr is due to nuclear weapon fall-out (the long-term situation) and to effluents from the Chooz station. It is known that fish bone is an integrator of environmental ^{90}Sr, while the flesh rapidly reflects temporary changes in the ^{90}Sr concentration, such as occurred, for example, in August and September 1971.

J.P. MORONI: Have you any results on possible contamination of the persons eating these fish? Similar work done in France indicates that the population effect of consuming fish contaminated at such low levels is quite negligible; in man we find no significant contamination from ^{134}Cs or ^{60}Co, which are present only in effluents, and as for ^{137}Cs, which comes mainly from radioactive fall-out, this shows no appreciable variation with respect to the mean body burden.

R. KIRCHMANN: We have no data on any contamination of the populations consuming Meuse fish. In order to determine this contribution it would be necessary to undertake an enquiry on diet, but since fishing in the Meuse is to a large extent carried out for sport, it is most probable that the effect of this contamination pathway on the population is negligible.

R.L. BLANCHARD: It has been our experience that extreme care must be exercised to ensure that small quantities of bone are not included in the muscle fraction when separating bone from muscle. Because of the much greater concentration of Sr and Ca in bone relative to muscle, a very small mass of bone in the muscle fraction will increase its Sr concentration and invalidate the muscle to bone concentration ratios. What precautions did you take to ensure that the muscle sample was not contaminated with small quantities of bone?

Catherine M. MICHOLET-COTÉ: I dissected the fish as carefully as possible. I did not boil them because Sr could pass from the tissues into the water during that operation. At all events it can be said that one has two masses of tissue, one with a very high percentage of bone, and the other with a very high percentage of muscle. Incidentally, the contents of muscle and bone given in Table V are in good agreement with the values quoted in the literature for perch and pike.

G. CANTILLON: We had a double aim in carrying out the study described in the paper: on the one hand we wished to anticipate any nuclear 'opposition' by providing some definite values; on the other hand, particularly in view of the construction of the Tihange power station, we needed an estimate for the critical group that must be taken into account for safety evaluations. I quite agree with Dr. Moroni that the burden on individual members of the population is not significant. May I take this opportunity of thanking the S.C.P.R.I., which facilitated our work by granting us access to the Chooz station and quoting us figures from its reports.

J.-J. MARTIN: Regarding your Table I and the ^{90}Sr results, what conclusions do you draw from the activity values measured in 1965/1966 and in 1971, the ratio being variable depending on the material analysed? I feel one must consider this matter in the light of the fact that in 1971 the ^{90}Sr releases from the Chooz power station were less than the fall-out activity carried by the Meuse.

R. KIRCHMANN: Your question highlights the differences that may be observed between the values expected on the basis of dilution of the effluents by the Meuse water and the values actually measured. The latter values are governed by numerous factors (method of sampling, exchange phenomena, measurement techniques etc.). The variation in the ^{90}Sr ratio in water is also found in fish caught in 1965 and 1971, and fish reflect the mean level of contamination of the medium in which they live.

I have no knowledge of any specific measurement value for the contribution of fall-out to the ^{90}Sr content of Meuse water.

B.K. BORISOV: What is the discrimination coefficient for strontium relative to calcium upon incorporation in bone?

R. KIRCHMANN: Using the values for the ^{90}Sr and Ca contents of water sampled in 1971 and the strontium unit (S.U.) values for the bones of bream caught at the same period, the observed ratio (OR)

$$\mathrm{OR} = \frac{(\mathrm{pCi}\ {}^{90}\mathrm{Sr/g\ Ca})_{\mathrm{bone}}}{(\mathrm{pCi}\ {}^{90}\mathrm{Sr/g\ Ca})_{\mathrm{water}}}$$

However, this value must be treated with some reserve because equilibrium conditions were not fulfilled, mainly owing to releases from the power station in August and September 1971.

MODELS FOR PREDICTING RADIATION EXPOSURE TO POPULATION GROUPS

(Sessions VI and VII)

Chairmen:

Pamela M. BRYANT (United Kingdom)
W.O. SCHIKARSKI (Federal Republic of Germany

A STUDY OF THE POTENTIAL RADIOLOGICAL IMPACT OF AN EXPANDING NUCLEAR POWER INDUSTRY ON THE TENNESSEE VALLEY REGION

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Abstract

A STUDY OF THE POTENTIAL RADIOLOGICAL IMPACT OF AN EXPANDING NUCLEAR POWER INDUSTRY ON THE TENNESSEE VALLEY REGION.

The US Atomic Energy Commission initiated a study in January 1970 to develop a computer model (HERMES) and the necessary data to permit estimates to the year 2000 of the radiation potentially received by individuals and population groups in large regions as a result of various estimated radioactive release rates from reactors and fuel reprocessing plants. The initial phase of the study concerned development of the computer model and data for the analysis of the region comprising the Upper Mississippi River Basin (UMRB) of the United States of America.

The paper concerns the second phase of the study, an interagency study by the US Atomic Energy Commission and the Tennessee Valley Authority of the potential radiological impact of an expanding nuclear power industry in the Tennessee Valley Region (TVR) to the year 2000. The Oak Ridge National Laboratory, the Hanford Engineering Development Laboratories and the Atomispheric Turbulence and Diffusion Laboratory at Oak Ridge, Tennessee are also participating in the study.

The TVR region is subdivided into about 140 populations centres and the dose is calculated for the average individual in each centre for each of four age groups (baby, child, teenager and adult). Contributions to dose by isotope and pathway for each of seven body components (total body, liver, lungs, gastro-intestinal tract, thyroid, bone and skin) are determined for each of the age groups. Sensitivity analyses are also done to evaluate the effects of uncertainties in the parameters believed to be most significant in their effects on dose. The procedures, scope and HERMES model modifications established for the TVR study were guided by the results of the study of the UMRB. Those results of the UMRB that had a major effect on the planning of the TVR study are discussed. An overview of the TVR study is also provided. Details on site selection, radwaste treatment systems, radionuclide transport and dose modeling for the TVR study are given in following papers.

INTRODUCTION

In January 1970 the US Atomic Energy Commission initiated a study to develop a computer model (HERMES) and necessary data to permit estimates of population dose rates from radiological releases from the normal operation of a large nuclear power industry in large regions of the United States of America. This was essentially the first significant departure in the USA from the plant-by-plant approach in studying the radiological effect of a large nuclear power industry. It was also decided that the study should provide an understanding of the contributions to the population dose rate of specific radionuclides and their associated pathways to man. This in-depth understanding was believed to be necessary to help validate the study and enable it to assist in guiding the development of improved radwaste treatment systems, design criteria and regulatory standards related to radioactive emissions from nuclear power facilities. Further, it was felt that there were many members of the public and also the nuclear industry who had a need for an in-depth understanding of the full potential radiological implications of a large nuclear power industry.

The first phase of the study involved the development of the HERMES model and its application to determine the radiation potentially received by the population of the Upper Mississippi River Basin (UMRB) in the USA from the operation of nuclear facilities, with their assumed radwaste treatment systems, that might be located in this region in the year 2000. This phase of the study was conducted by the Hanford Engineering Development Laboratory (HEDL) under the direction of the USAEC and is reported in WASH-1209, The Potential Radiological Implications of Nuclear Facilities in the Upper Mississippi River Basin in the U.S. in the Year 2000.[1] The second phase of the study, which is the subject of this paper, is the application of the HERMES model to the Tennessee Valley Region. This is an interagency study conducted by the US Atomic Energy Commission (AEC) and the Tennessee Valley Authority (TVA) with the participation of the Oak Ridge National Laboratory (ORNL), HEDL and the National Oceanic Atmospheric Administration (NOAA).

This joint AEC/TVA study was undertaken because of the unique hydrological and topographical characteristics of the Tennessee Valley Region (TVR) and the anticipated large-scale use by the TVA of large nuclear power stations to help meet much of the region's continuing need for electricity. The physical characteristics of the TVR are very different from those found in the UMRB and will allow the HERMES model to be expanded and tested for these new conditions.

The study involves an appraisal of the generation, management and control of radioactive effluents from nuclear power plants and fuel reprocessing plants, transfer of radionuclides through air and water, and concentration in terrestrial and aquatic systems and the calculations of potential radiation dose commitments to man. Data on long-range power projections by sites, load distribution among different plants, and generating unit availability are used in the model along with steam flow and run-off data,

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[1] This report will be available from the Superintendent of Documents, US Government Printing Office, Washington, D.C. 20402.

sediment adsorption characteristics, and existing and projected uses of the TVR reservoir system for irrigation and recreation. The study was initiated October 1972 and a draft report is expected about the end of 1973.

This paper provides a survey of the TVR study with particular emphasis on power projections by sites. As the HERMES model modifications, procedures and scope established for the TVR study were guided by the results of the study of the UMRB, a summary of this study is provided. The features of the HERMES model will be explained in conjunction with the summary of the UMRB study as the basic features of the HERMES model are unchanged for the TVR study. This will also permit factoring in with the explanation of the model results obtained from its usage.

THE UMRB STUDY

As indicated in Fig. 1, the UMRB region includes an area of approximately 300 000 square miles and encompasses one of the major power supply/demand regions of the USA. By the year 2000 the population of the region is projected to reach 29 million and to account for about 10% of the total electric energy generation and consumption of the United States of America.

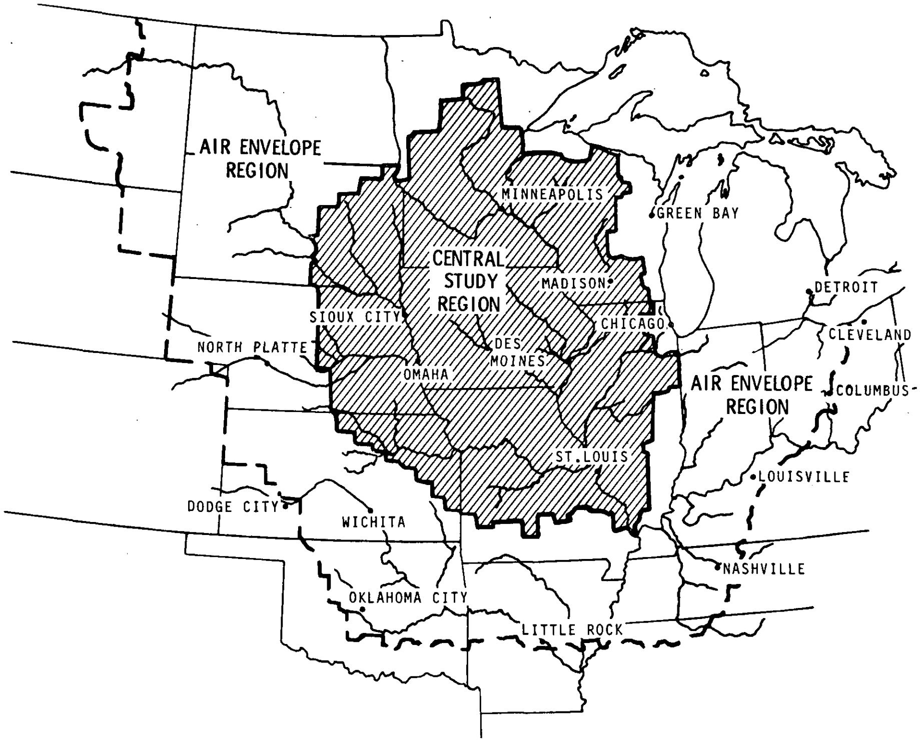

FIG. 1. Upper Mississippi River Basin region.

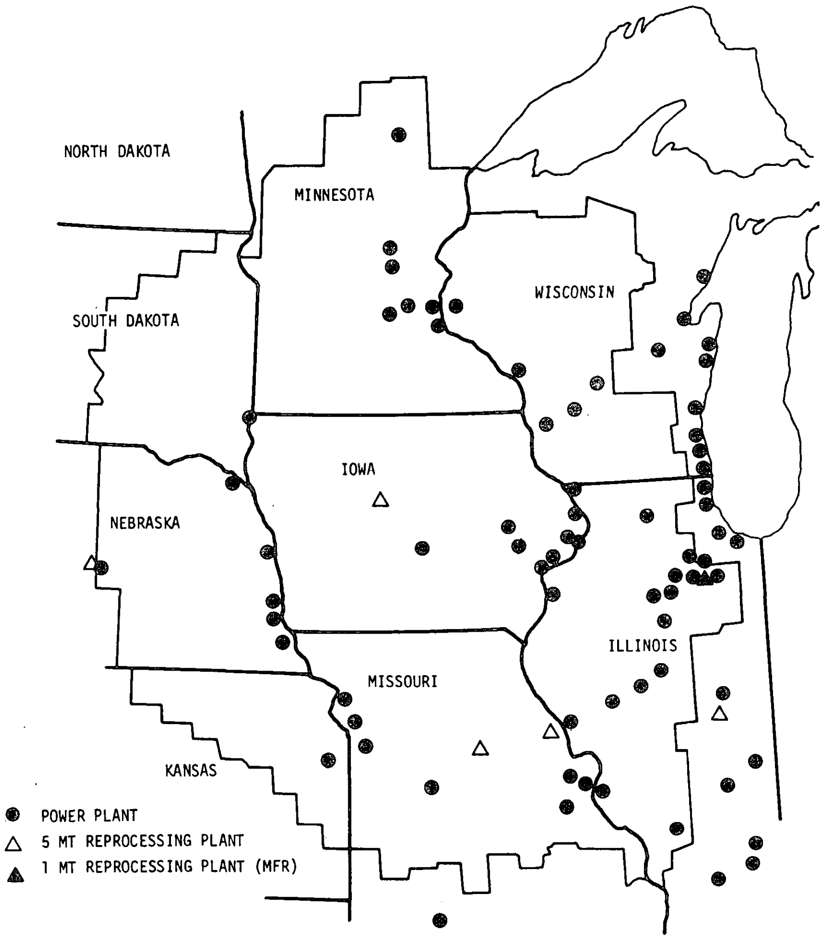

FIG. 2. Nuclear power and fuel reprocessing plants sites in the UMRB.

To allow for the contribution from the airborne radionuclides released from nuclear facilities in adjacent areas, a peripheral zone some 200 miles wide and surrounding the basic study area was defined and designated as the 'air envelope'. With the air envelope boundary region added, about 25% of the total US electric generation is represented. Waterborne radionuclides released in the air envelope area were not considered. Essentially no water flowed into the central study region from the air envelope. Much of the area consists of flat or undulating prairie land, interrupted by locally hilly areas and incised by the waterways of the region. The generally uniform topography simplifies the broad-scale air flow, thus avoiding undue complications in modelling meteorology. The major waterways are the Mississippi and Missouri Rivers.

Recent increases in nuclear energy generation in the region are expected to continue with nuclear expected to provide over 80% of the projected generation in the year 2000. It is projected that there will be 189 gigawatts (GW(e)) of LWR capacity and 172 GW(e) of LMFBR capacity at that time. To reprocess the fuel from these reactors, eight 1500-tonne per year and one 300-tonne per year reprocessing plants are required. These facilities are sited as shown in Fig. 2 on the basis of FPC projections to the year 1990 and informed guesses from the year 1990 to the year 2000.

The information flow in the HERMES model (see Fig. 1 of the companion paper IAEA-SM-172/40, these Proceedings) is as follows:

(1) Using the projected power plant and reprocessing plant siting patterns, a source map showing the location and quantity of radionuclide releases (as a function of time to the year 2000) is established.

(2) The air concentration and deposition at a central point in each of the 300 subregions (centroids) is determined by computing the air transport of the released radionuclides. The radionuclide deposition at the centroids due to water transport is also determined. In transporting the radionuclides through air and water, the effect of radioactive decay is included.

(3) The uptake of deposited radionuclides and their subsequent concentration in various food types was then estimated.

(4) Transport of food to meet subregional demand (by centroid) was simulated. It was assumed that demand for food was first met by food produced in the region and, if food had to be shipped in from outside the UMRB, it would be non-radioactive.

(5) Accounting for work and recreational habits, as well as living habits, the dose to each population group in each centroid was calculated. The calculations are based on direct exposure to airborne, deposited and dissolved radionuclides together with internal exposure from those ingested or inhaled. At present the code can handle up to 50 radionuclides, 200 release points and 300 receptor (centroid) locations.

The analyses of the UMRB involve 45 separate fission and activation products and their movement through ten separate pathways that included some 35 separate categories of foods. The potential population annual dose was calculated as whole-body dose and as dose to each of six body components — liver, lung, gastro-intestinal tract, thyroid, bone and skin. Dose commitments for 50 years from the year 2000 intake were also calculated. Individual annual doses at each centroid were calculated for average individuals and to individuals representing maximum exposure population groups in each of three age groups (child, teenager and adult). In addition, integrated doses (man-rems) were calculated for the study area and for each centroid. Finally, dose contributions by isotope and by pathway were calculated for each centroid. An indication of the complexity and detail associated with one regional set of dose calculations is the three hours of running time required on a UNIVAC 1108.

Because the various nuclide sources and receptors are treated individually, it is possible to determine for any of the many radionuclides considered and for any source the effect on dose to several specific body components in specified population groups. It is also possible to examine the ultimate effectiveness, i. e. the reduction in dose to the population, of various particular radwaste treatment systems including technological improvements.

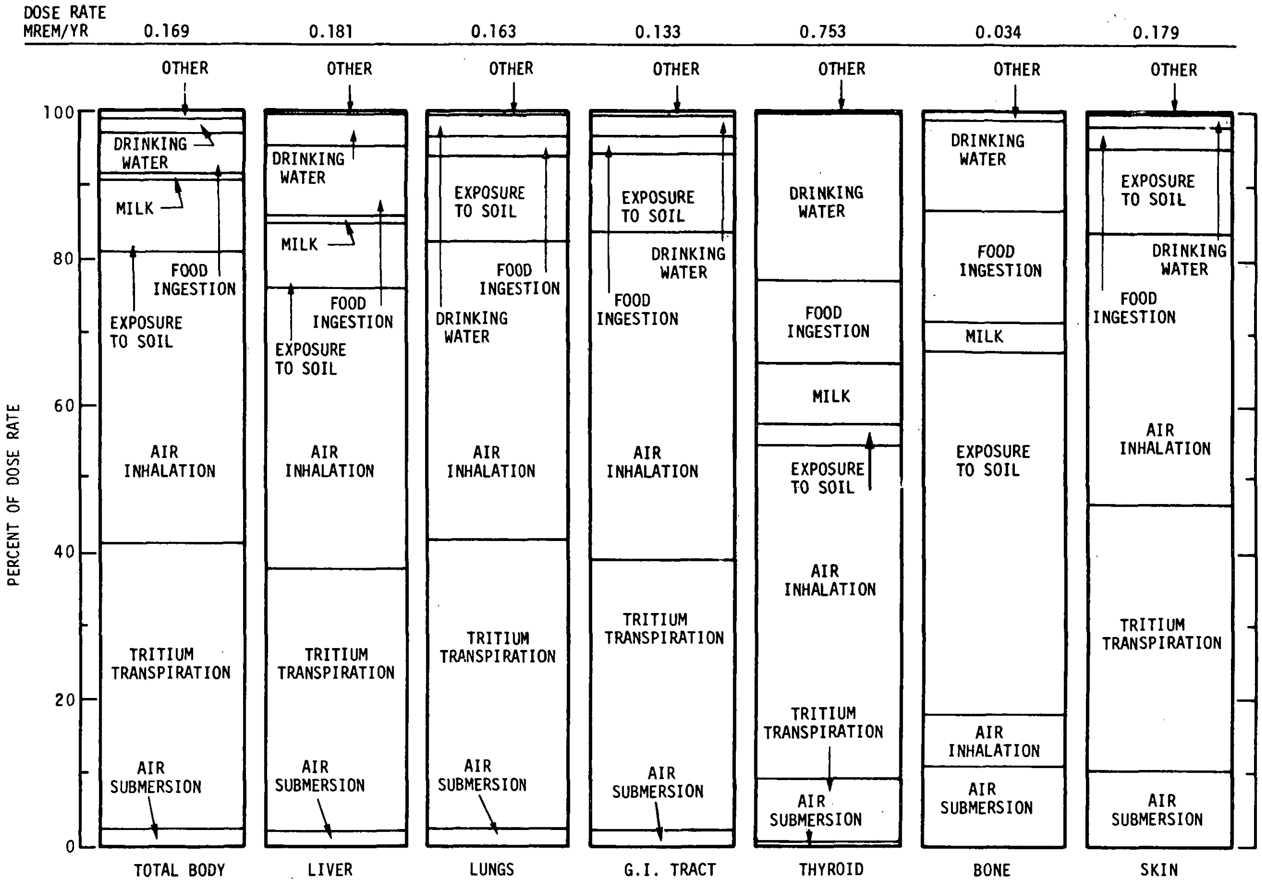

FIG. 3. Contribution by pathways to average adult dose rate in UMRB.

The radwaste treatment assumptions used in the UMRB study are:

(1) Light water reactor (LWR) radwaste systems were representative of those presently included in plant designs to meet the recent proposed USAEC guidelines for radionuclide release;

(2) Liquid metal fast breeder reactors (LMFBR) and fuel reprocessing plants, besides utilizing the radwaste treatment systems applied to LWRs, have a noble gas absorption system;

(3) No gaseous tritium radwaste treatment systems were included in standard designs of nuclear power or fuel reprocessing plants; and

(4) Tritium generated in reactor fuel was assumed to remain mostly in the fuel until released during reprocessing. Sensitivity studies were performed to determine the effects of the above assumptions.

The results of the study of the UMRB based on the selected radwaste treatment showed that, on the average throughout the region, the potential dose a representative individual could receive in the year 2000 would be increased by roughly 0.2 millirem per year because of the presence of nuclear facilities. Not included in this results was the contribution of transuranium nuclides or close-in dose associated with exposure near specific facility fencelines. However, subsequent analyses indicate that these refinements do not significantly change the results. They will be included in the TVR study to help ensure the completeness of the study.

As indicated in Fig. 3, the bulk of the population dose rate was attributed to four pathways: submersion in air, inhalation of air, transpiration of tritium and exposure to soil. The dispersion of radionuclides was governed primarily by air transport rather than by water transport or shipment of foodstuffs.

Air inhalation was the major contributor for most body components, especially the lungs. Air submersion and soil exposure were major sources of external exposure. Hence, they were major contributors to skin dose. Tritium transpiration is a relatively large factor for the whole body and all body components considered except bone. Generally, only about 10% of the dose rate was contributed by pathways other than the four major ones, although for some body components it was larger, reaching 25% for bone. Of the lesser pathways, ingestion of drinking water, milk and beef were the most important.

Of the 45 radionuclides considered in the study, Fig. 4 shows that tritium, iodine and caesium contributed about 95% of the whole-body dose rate and 80% of the dose rate to the body components, except for the skin where krypton and xenon contributions were important. Tritium was released primarily from reprocessing plants and was a major contributor for many of the body components and for the whole body. For example, it accounts for about 85% of the whole-body dose to the average adult. One reason why tritium was the dominant contributor was that it was the only gaseous radionuclide for which there was no waste stream removal.

Due to its affinity for the thyroid, iodine is a major contributor to dose rate to that organ. Major pathways for iodine are air inhalation, ingestion of drinking water, fresh milk, beef and lamb. Iodine also contributed substantially to dose to the gastro-intestinal (GI) tract, bone, lungs and skin.

TABLE I. RESULTS OF PERTURBATIONS OF THE BASE CASE

Perturbations	Results
No bottling systems for noble gases.	A 30% increase in regional population dose rate primarily from the additional ^{85}Kr released from reprocessing plants.
Delay of radwaste treatment system to meet new AEC guidelines.	A 35% increase in regional population dose rate. A substantial fraction of increase due to ^{85}Kr.
Tritium in LMFBR fuel leaks to coolant (90%) where it is cold trapped out.	The gaseous release from fuel reprocessing plant (handling combination of LWRs and LMFBRs) decreased by 15-25%.
Noble gases bottled on LWRs.	On a regional basis decreases skin doses by 20-30%.
No floor drain recycling for PWRs.	Increases drinking water contribution to dose by factor of three and increases integrated regional dose by about 6%.
Tritium removal systems on all facilities with a removal factor of 100.	Reduces the population total dose by a factor of 5.
Omission of 200 mile air envelope.	Decreases population total dose by about 18%.
Omission of close-in dose around power and fuel reprocessing plant.	Decreases population total dose by about 5%.
Delay in reproducing LMFBR fuels reduced from 150 days to 30 days.	Increases population total dose by about 40%. Increase due mostly to ^{131}I and ^{131}Xe.
No nuclear power plants.	Reduces the dose by about 25%; hence, reprocessing plants contribute about 75% of dose.

Caesium-134 and 137 contributed heavily to the dose rate in the bone, liver, total body and GI tract. Principal pathways were drinking water and exposure to soil. Cerium-144 contributed only to the GI tract.

Xenon-133 and krypton-85 were important in the skin dose rate; Kr accounted for about 6% and Xe about 40%. Xenon also contributed about 9% of the bone dose. Strontium-90, zirconium-95, niobium-95 and ruthenium-106 together contributed 3% of the dose to the GI tract and about 1% of the whole-body dose. Strontium-90 accounted for less than 1% of the annual dose rate to the bone and about 32% of the 50-year commitment to the bone.

The effect of the radwaste treatment and other assumptions were examined on the basis of the sensitivity studies given in Table I. Of particular interest in the sensitivity studies were:

(1) 75% of the population dose is from the reprocessing plants
(2) Tritium contributes about 80% of the population dose
(3) The air envelope contributes about 18% of the population dose
(4) A 30-day delay instead of 150 days in the reprocessing of LMFBR results in a 40% increase in population dose
(5) The omission of close-in dose around nuclear facilities results in only a 5% decrease in integrated population dose
(6) Noble gas bottling has only a moderate effect on the total population dose.

THE TVR STUDY

The area of the central study region for the TVR study is shown in Fig. 2 of the companion paper IAEA-SM-172/40 (these Proceedings). It is defined by the drainage basin of the Tennessee and Cumberland Rivers and their tributaries and it encompasses an area of over 60 000 square miles. The population of the area is presently about 4.6 million and the TVA system which serves it has presently an electric generating capacity of about 20 000 MW(e). Additional capacity now under construction or on order will add another 14 000 MW(e) of new capacity to the TVA system by the end of this decade. About 65% of the new capacity will be nuclear and it is expected that nuclear will continue to be a major source of new capacity throughout this century.

An air envelope surrounding the study region has been defined, as in the UMRB study, to include the radiological dose contributions due to air transport of radioactive releases into the study region from plants with this envelope. The results of the UMRB study indicated that the air envelope contributed about 18% of the population dose and that an air envelope width of about 50 miles would provide more than 90% of its contribution. Accordingly, the air envelope for the TVR study extends 50-80 miles from the central study region's boundaries and is indicated by the dashed line in Fig. 5.

In the year 2000 it is estimated that the central study region will have a population of about 7 million and about 54% of the people will be urban dwellers, 36% will live in rural areas and not be farmers and 10% will be farmers. The TVR is subdivided into about 140 population centres and the dose is calculated for the average individual in each centre for each of four age groups (baby, child, teenager and adult). The TVA system which will serve the central study region and portions of the air envelope is estimated to have 42 935 MW(e) of nuclear generating capacity at that time. In the air envelope an additional 101 000 MW(e) of nuclear generating capacity is estimated to be available in addition to TVA's capacity. The types of plants hypothesized for the air envelope region are similar to those used in the central study region and include pressurized light water reactors (PWRs), boiling light water reactors (BWRs), high temperature gas-cooled reactors (HTGRs) and liquid metal fast breeder reactors (LMFBRs). While HTGRs were not included in the UMRB study, their introduction in this study is not expected to significantly change the results.

The selection of the number and types of reactors and their sites both in the central study region and air envelope region was a matter of

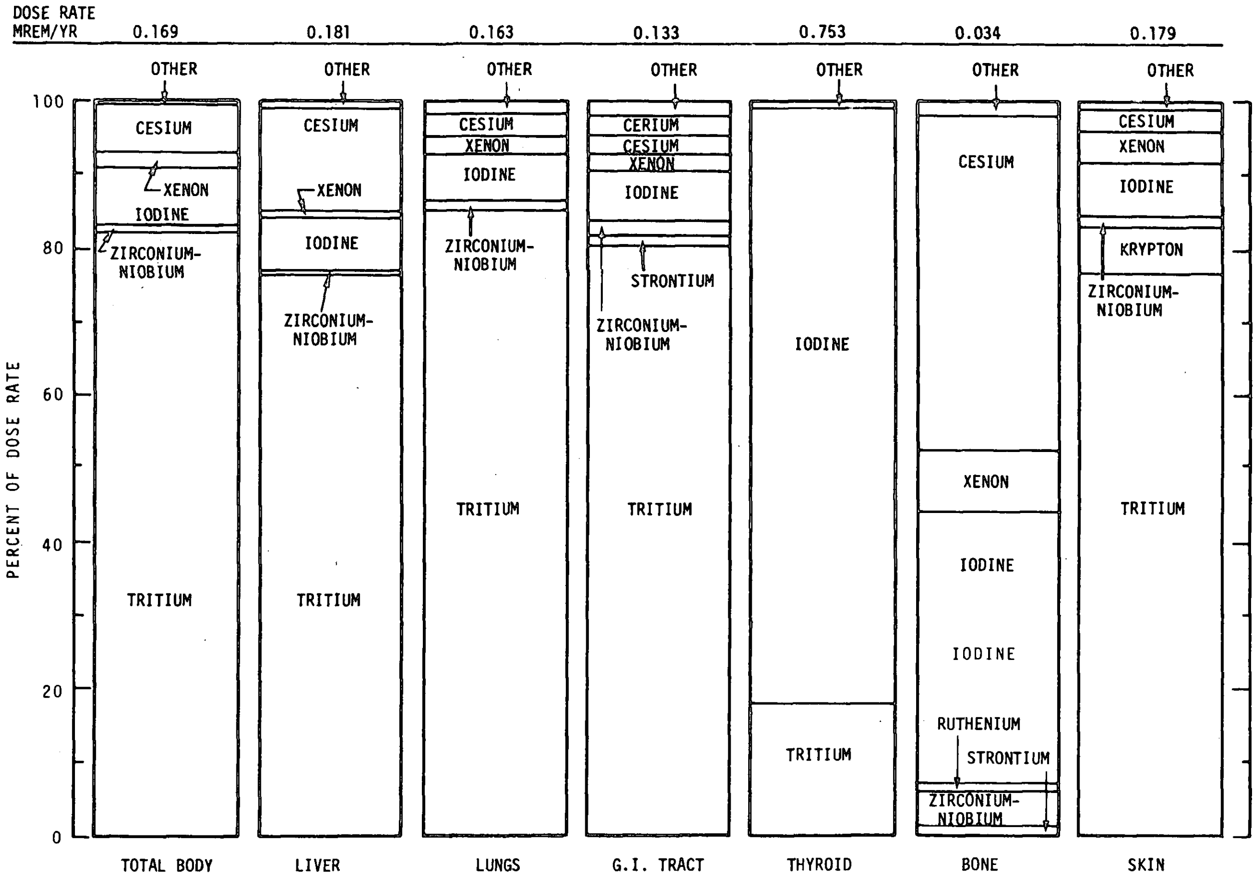

FIG. 4. Contribution by radionuclides to average adult dose rate in UMRB.

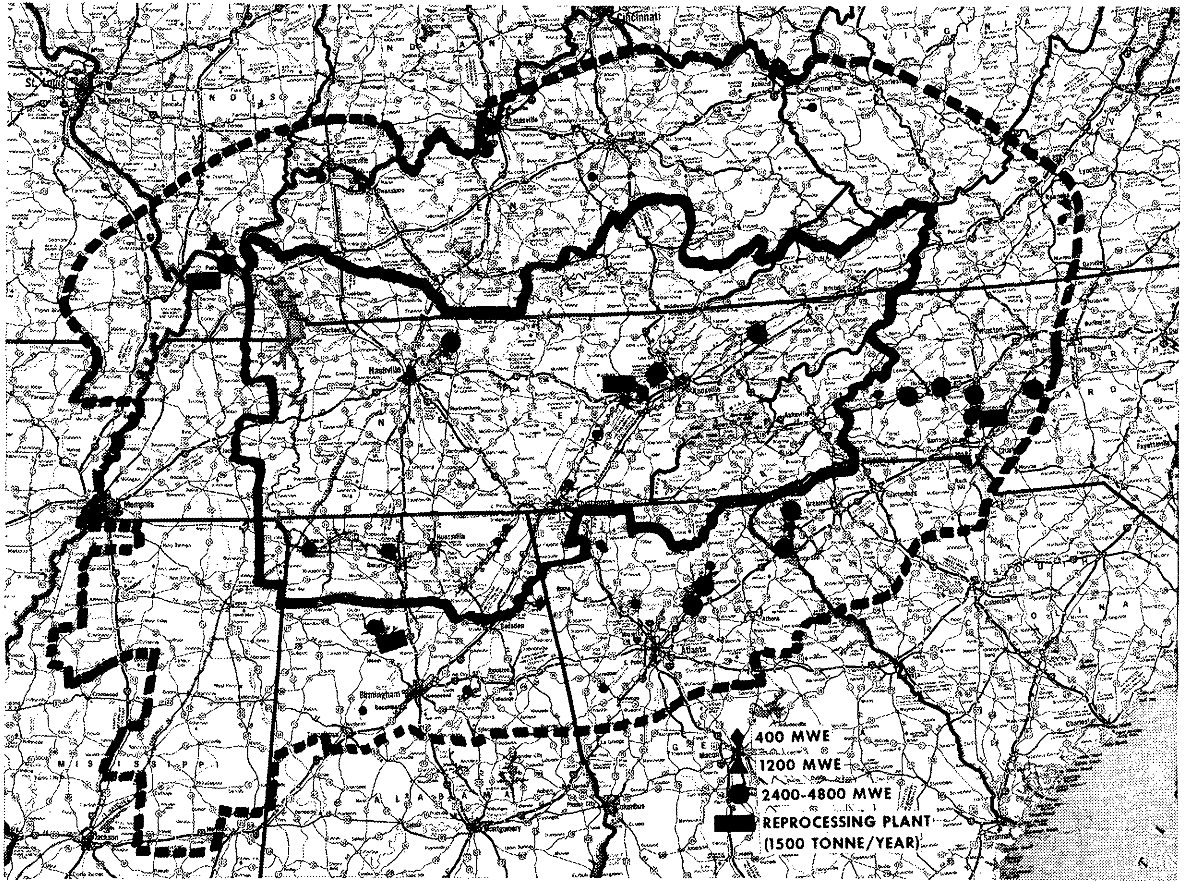

FIG. 5. Nuclear power and reprocessing plant sites - TVA year 2000 study.

TABLE II. RADIONUCLIDES ANALYSED IN THE TVR STUDY

Radionuclides	LWR	HTGR	LMFBR	Fuel reprocessing plant
^{3}H	X	X	X	X
^{14}C	-	-	-	X
^{22}Na	-	-	X	-
^{58}Co	X	-	-	-
^{60}Co	X	-	-	-
^{85m}Kr	X	-	-	-
^{85}Kr	X	X	X	X
^{87}Kr	X	X	-	-
^{88}Kr	X	X	-	-
^{89}Kr	X	-	-	-
^{89}Sr	X	X	-	X
^{90}Sr	X	X	-	X
^{90}Y	X	X	-	X
^{91}Y	X	X	-	X
^{95}Zr	X	-	-	X
^{95}Nb	X	X	-	X
^{103}Ru	-	-	-	X
^{106}Ru	-	-	-	X
^{123m}Sn	-	-	-	X
^{125m}Te	-	X	-	-
^{127m}Te	-	X	-	-
^{129m}Te	X	X	-	-
^{132}Te	X	-	-	-
^{131}I	X	X	-	X
^{133}I	X	X	-	-
^{135}I	X	-	-	-
^{133}Xe	X	X	-	X
^{135m}Xe	X	-	-	-
^{135}Xe	X	X	-	-
^{137}Xe	X	-	-	-
^{138}Xe	X	-	-	-
^{134}Cs	X	X	X	X
^{137}Cs	X	X	X	X
^{140}Ba	X	-	-	-
^{140}La	X	-	-	-
^{141}Ce	-	-	-	X
^{144}Ce	-	-	-	X

Table II (cont.)

^{154}Eu	-	-	-	X
^{238}Pu	-	-	-	X
^{239}Pu	-	-	-	X
^{240}Pu	-	-	-	X
^{241}Am	-	-	-	X
^{242}Cm	-	-	-	X
^{244}Cm	-	-	-	X

personal judgement by a few members of the TVA staff. Their judgement was guided primarily by Federal Power Commission projections and their own understanding of the TVA and adjacent systems. There was no attempt to obtain official validation of these selections by the utility systems involved and the reader is cautioned to remember that they are at best informed guesses for the year 2000. In instances where the TVA staff had knowledge of specific sites for nuclear plants they were excluded and nearby sites with similar topography were chosen. This technique of utilizing staff of the concerned utilities for power plant projections and siting is believed to be superior to the computational and judgement techniques used in the UMRB study.

The circles and triangles in Fig. 5 indicate the location and size of the nuclear power plant sites in the year 2000. In the central study region it is projected that there will be 29 units of which 13 will be PWRs, 9 BWRs, 2 HTGRs and 5 LMFBRs (included is a 400-MW(e) LMFBR demonstration plant near Oak Ridge, Tennessee) on 11 sites. In the air envelope region it is projected that there will be 93 units of which 38 will be PWRs, 13 BWRs, 20 HTGRs and 22 LMFBRs on 35 sites.

It is assumed that all the fuels from the nuclear power plants in both the central study and air envelope regions will be reprocessed in local plants. The fuel throughput in the year 2000 requires the equivalent of four fuel reprocessing plants that have the capability of handling 1500 tonnes per year of LWR fuels (LMFBR fuel can be handled in multipurpose fuel reprocessing plants at one-half the rate of LWR fuel). Two of the fuel reprocessing plants are multipurpose plants that are capable of processing LWR, HTGR and LMFBR fuels. The LWRs will recycle the plutonium they generate until there are LMFBRs available. The siting of the fuel reprocessing plants was also done by the people who sited the power plants. They were sited on the basis of location of the load, availability of services such as cooling water and access to transportation and conformance of AEC siting criteria. The location of the fuel reprocessing plants are designated in Fig. 5 by the rectangles.

Analysis of releases for all reactor and reprocessing plant types suggested a list of over 100 radionuclides for initial consideration. These radionuclides were screened on the basis of the results of the UMRB study, recent studies at ORNL, HEDL and Battelle Northwest Laboratories on LWR nuclear power plant radionuclide releases and preliminary sensitivity calculations. Table II indicates the radionuclides that will be included

TENNESSEE VALLEY STUDY

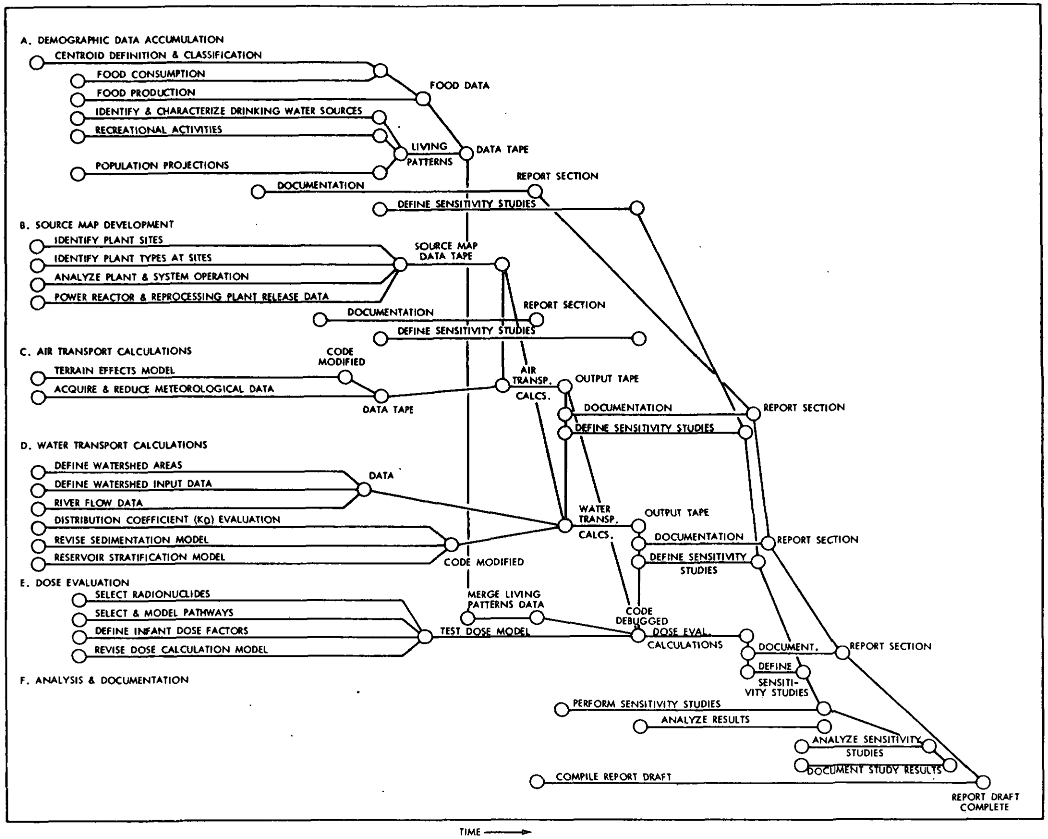

FIG. 6. Information flow in TVR study.

in the plant release analysis. It has been determined that these 44 radionuclides will provide over 99% of any body component dose. Sixteen of the 45 radionuclides analysed in the UMRB study are not involved in the TVR study. Of the fifteen new radionuclides on the TVR list the addition of transuranics, plutonium-238, 239 and 240, americium-241 and curium-242 and 244 are possibly of the most interest. The transuranics, although they were evaluated in supplemental calculations, were not included in all phases of the UMRB study because of anticipated computer limitations.

The HERMES model simulates generation of fission products in reactor fuel, as also activation products formed in the coolant and reactor. It then estimates the diffusion of fission products from the fuel into the coolant, the transfer of radionuclides to the radiation waste treatment system and the treatment of radionuclides as they pass through each component of the treatment systems. Finally it calculates the rates of release of radionuclides through gaseous and liquid discharge streams. The reprocessing plants are treated in an analogous manner with appropriate consideration of plant feed rate and fuel type. The choice of the basic radwaste treatment system for the individual plant types for the base case in the TVR study was essentially the same as used in the UMRB study with decontamination coefficients modified to reflect the most recent information. The release to the region environment are the results of releases associated with the normal operation of nuclear power plants and nuclear fuel reprocessing plants. Other potential releases are not considered in this study.

The submodel in the HERMES model that considers water transport (WTRAN) of radionuclides has been modified to account for the large dams and reservoirs of the TVA system. Time delays associated with reservoir storage and stratification and techniques for describing recently measured and historical patterns of deposition of sediments in reservoirs and non-impounded reaches have been added to WTRAN. The major waterways in the region are the Tennessee and Cumberland Rivers; with their tributaries there is about 2000 miles of rivers in the central study region.

The submodel concerned with air transport of radionuclides (ARTRAN) has also been modified to account for the very diverse topography of the TVR. It may be necessary to track the plant releases from sites that are located in sharply defined valleys. Meteorological conditions in these valleys may result in the radionuclides piling up and moving down the valley slowly.

The data necessary to compile the living patterns and demographic information for the TVA year 2000 are being assembled and compiled in essentially the same manner as the UMRB year 2000 study. Details on radionuclide transport and dose modelling for the TVR study are given at this Symposium in the papers, "Modelling the regional transport of radionuclides in a major United States river basin" (IAEA-SM-172/40) and "Methods of estimating dose to man from regional growth of nuclear power" (IAEA-SM-172/41).

Figure 6 indicates the scheduling of the major subcomponents of the study. Most of the input data are provided by ORNL and TVA and all of the major parties are involved with HERMES model modifications for the TVR study. HEDL has the prime responsibility for effecting the HERMES model modifications and performing the required calculations. All the major parties will participate in establishing the set of calculations, analysing them and preparing the study report.

As the study is essentially the product of four major parties and requires input from other sources, it requires close and effective management. This management is provided by an executive committee that is composed of one representative from each of the major parties (AEC, TVA, ORNL and HEDL). Judiciously applying the specific capabilities and resources available within the co-operating agencies will result in the study being conducted in an optimum manner. This aspect has been recognized by all parties and the study is being conducted with a high degree of enthusiasm and co-operation.

DISCUSSION

A. BAYER: I understand that your model is for a central food supply, covering a large area. Have you also considered a self food supply model in your program, i.e. a model relating to one farm?

S. STRAUCH: For the Tennessee Valley Region study it is being assumed that food transported into the Region will be subjected to essentially the same radioactivity as food in the Region itself. Hence the model used is similar to a self food supply model.

R.S. BRUCE: I was interested in your estimates of doses from iodine (Fig. 5 of the paper). Could you comment on the high proportion (40%) of the dose to bone which is attributed to iodine?

J.F. FLETCHER: The iodine contribution to bone dose was in the form of external gamma radiation from iodine deposited on the ground.

H.F. MacDONALD: Do you make any allowance for accidental releases in your current HERMES studies?

S. STRAUCH: Not in the studies discussed in the paper. However, the model can be utilized for accidental releases and it is anticipated that it will be so used.

H.F. MacDONALD: In the light of the relatively low population doses due to normal operation, as shown in your Upper Mississippi River Basin results, the dose commitment resulting from a single accident might easily be highly significant.

S. STRAUCH: This is strictly a function of the type of accident. As I have just mentioned, we will probably examine possible accidents with HERMES.

A. MARTIN: Does the value of 0.2 mrem/yr by the year 2000 include the contribution from world-wide distribution of ^{85}Kr?

S. STRAUCH: The Mississippi study does not include the world-wide contribution of ^{85}Kr. However, it will be included in the Tennessee study.

A. MARTIN: I would also like to make a comment connected with tritium in drinking water. Work which my company has done for the Nuclear Installations Directorate in the United Kingdom suggests that the contribution to dose from tritium in drinking water may be of the same order as that due to its transpiration.

J.F. FLETCHER: Tritium in drinking water appeared to contribute only to a small extent to dose. Except for light water reactors, all tritium releases were to air. Only minor portions were subsequently transferred to watercourses. Also the large rivers in the area had a considerable diluting effect.

A. MARTIN: Finally, may I say that this is a most impressive study and I wonder whether you could indicate the scale of the effort involved.

S. STRAUCH: It took about two years to develop the HERMES model and to conduct the study of the Upper Mississippi River Basin. This portion of the work was done at the Hanford Engineering Development Laboratory under the direction of the USAEC.

H. WIJKER: From how many meteorological stations were the data gathered for the calculations and was there a marked regional spread in the meteorological conditions?

J.F. FLETCHER: Data for the Upper Mississippi River Basin were obtained from 44 meteorological stations operated by the US Weather Service in the region under study. This region is fairly uniform topologically, and hence the changes in meteorological conditions across the area are gradual rather than abrupt.

S. STRAUCH: Meteorologically speaking, we shall have greater problems in the Tennessee Region, where the valley structures will make it very difficult to model the air movements.

MODELLING THE REGIONAL TRANSPORT OF RADIONUCLIDES IN A MAJOR UNITED STATES RIVER BASIN*

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Abstract

MODELLING THE REGIONAL TRANSPORT OF RADIONUCLIDES IN A MAJOR UNITED STATES RIVER BASIN.

A comprehensive study of the radiological implications of large-scale use of nuclear power generation, addressed to the Tennessee-Cumberland River Basins in the year 2000, is being undertaken. In the course of this study computer modelling techniques have been developed to evaluate the movement of radionuclides, by water and air transport, from points of release at a large number of plant sites and to calculate patterns of radionuclide concentration throughout the region of study.

Air transport calculations utilize a modification of the bivariate normal equation, with the vertical parameter σ_z characterized as a function of distance from the source, stability and mixing layer depth. Local variations in air flow caused by major terrain features are modelled. Depletion of the airborne radionuclides by wet and dry precipitation processes is accounted for and the concentrations of deposited radionuclides are calculated. The model sums contributions from all sources to provide resultant patterns of concentration of airborne and deposited radionuclides, including contributions from radionuclides deposited in prior years. The water transport model calculates stream concentrations of radionuclides in solution and absorbed on sediments. Effects of flow stratification and sediment trapping in reservoirs are considered. The solution of air-deposited radionuclides and their subsequent transport in ground and surface waters are modelled. Calculational routines are provided for simulating radionuclide transport in each stream in the basin and for calculating the resultant concentrations in solution and associated with suspended and deposited sediments.

Together, the air and water transport models are designed to calculate regional patterns of radionuclide concentrations in the environment that result from the operation of nuclear facilities in a region. These concentration patterns can then be applied to the evaluation of radiological dose to man.

A comprehensive study of the radiological implications of large-scale use of nuclear power generation, addressed to the Tennessee-Cumberland river basins in the Year 2000, is being undertaken jointly by the United States Atomic Energy Commission and the Tennessee Valley Authority, with participation by the Hanford Engineering Development Laboratory (HEDL), the Oak Ridge National Laboratory (ORNL), and the Atmospheric Turbulence and Diffusion Laboratory (ATDL). This study builds upon a previous study of the Upper Mississippi River Basin[1] which was performed by HEDL under AEC sponsorship. During that study a computer model, HERMES,[2] was developed to permit evaluation over a large region, of the relationships involving the release of radionuclides from operating nuclear facilities, the transport of these radionuclides into the environment, and the resulting dose to the population of the region. The

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* Work performed for the US Atomic Energy Commission under contract AT(45-1)-2170.

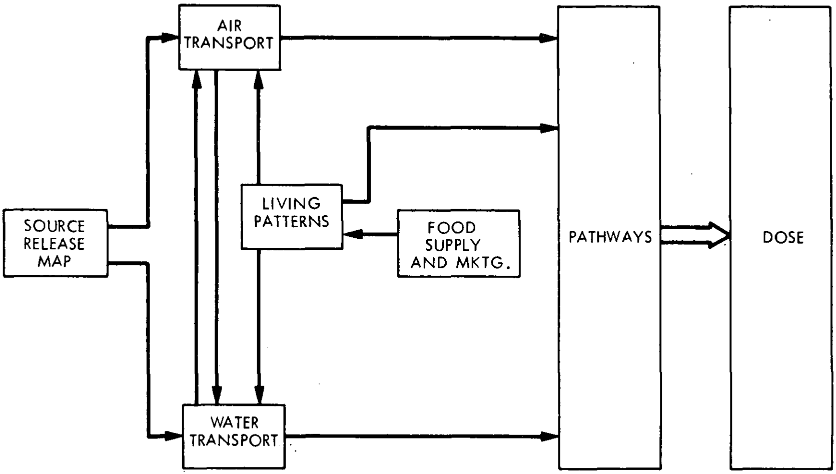

FIG.1. HERMES model information flow structure.

air and water transport codes ARTRAN and WTRAN, used in the HERMES model, are designed to simulate the movement of radionuclides from their points of release at a large number of plant sites, and to evaluate the resulting patterns of radionuclide concentrations in the air, on the ground, and in the surface and ground waters and sediments. During the present study the ARTRAN and WTRAN codes have been modified for use with the more diverse topography of the Tennessee Valley Region, and the consequent complexities in meteorological and streamflow conditions.

The relationship of the ARTRAN and WTRAN codes to other elements of the HERMES model is indicated in Figure 1.

The HERMES model, and particularly the transport codes ARTRAN and WTRAN, are designed to handle massive amounts of data and to provide results of accuracy consistent with that required for regional evaluations three decades in the future. Auxiliary codes are available to provide more detailed results close by specific sites.

Calculations in the HERMES model are referenced to the concept of the "centroid." The region under study is subdivided into smaller areas--for convenience, the political boundaries of counties are used (Figure 2). Within each area a single point is designated as the centroid of that area. All calculations leading to estimates of radiation dose are referenced to these centroids; thus, the regional study may be carried out using a reasonable number of receptor locations.

AIR TRANSPORT CALCULATIONS

The air transport code ARTRAN (Figure 3) is designed to calculate the atmospheric radionuclide concentrations at each of a large number of "receptor" locations which result from airborne releases at each nuclear facility under consideration. Depletion of the airborne radionuclides by

FIG.2. Tennessee-Cumberland Valley study area.

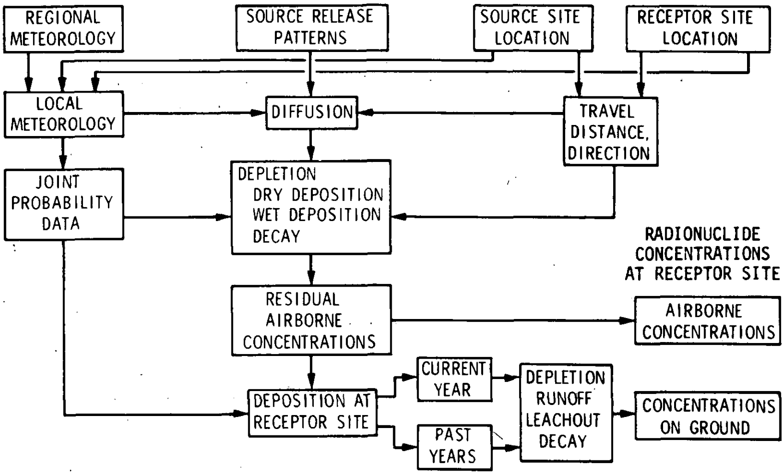

FIG.3. ARTRAN code structural diagram.

wet and dry precipitation processes is considered, and the resulting concentrations of ground-deposited materials at each receptor location are calculated. In the same manner, depletion of radionuclides by radioactive decay is considered during transit and after surface deposition, as is the buildup of daughter nuclides. Finally, the residual concentrations of nuclides resulting from deposition in prior years are estimated. Using the present structure of the code, these calculations may be made for up to 50 radionuclides, 200 release points, and 300 receptor locations.

Plume Diffusion

The diffusion of radionuclides downwind from their point of release is calculated using modified forms of the Gaussian diffusion equation. During early phases of dispersion of the effluent plume, average standard concentrations are calculated from equation (1):

$$\bar{\chi}_x = \sqrt{\frac{2}{\pi}} \frac{f\dot{Q}}{\bar{u}\theta\, x\, \sigma_z} \exp - \left(\frac{(h_s + \Delta h - h_t)^2}{2\, \sigma_z^2} \right) \qquad (1)$$

where:

$\bar{\chi}$ = mean air concentration, pCi/m^3
x = downwind distance
f = percent frequency of occurrence of wind direction
$\dot{Q}$ = rate of release of radionuclide, pCi/sec
$\bar{u}$ = mean wind velocity, m/sec
θ = angular width of azimuthal sector being considered, radians (usually taken as $\frac{\pi}{8}$ for a 16-sector azimuth)
σ_z = standard deviation of vertical distribution of airborne contaminant, m
h_s = stack height, m
Δh = thermal plume rise, m
h_t = terrain height (at distance x) above stack base, m.

For great distances, the vertical distribution of the airborne contaminants is uniform rather than Gaussian normal. In the code, Gaussian distribution is assumed out to the distance where $2.15\,\sigma_z = Z_m$[3] (Z_m is the height of the mixing layer) or to a distance of 70 km, whichever is less. Beyond this transition point, the radionuclides are assumed to be uniformly distributed vertically within the mixing layer. Concentrations are calculated as:

$$\bar{\chi}_{x,Z_m} = \frac{f\dot{Q}}{\bar{u}\theta\, x\, Z_m} \qquad (2)$$

The meteorological data required for equations (1) and (2) are obtained by computer analysis of standard records of observations routinely made by the U. S. National Weather Service. This analysis is made using a modification of a code attributed to Turner.[4] This code provides statistical joint probabilities of occurrence of the Pasquill stability classes "A" through "F", [5] of wind direction (grouped by azimuth sector), wind-speed (grouped into six speed categories), the probability of occurrence of precipitation, and precipitation rate.

Values of the vertical standard deviation parameter, σ_z, are calculated by three different methods in ARTRAN. For distances less than 2 km, estimates of σ_z are obtained from the curves usually attributed to Pasquill,[5] as shown in Figure 4. For distances greater than 2 km but

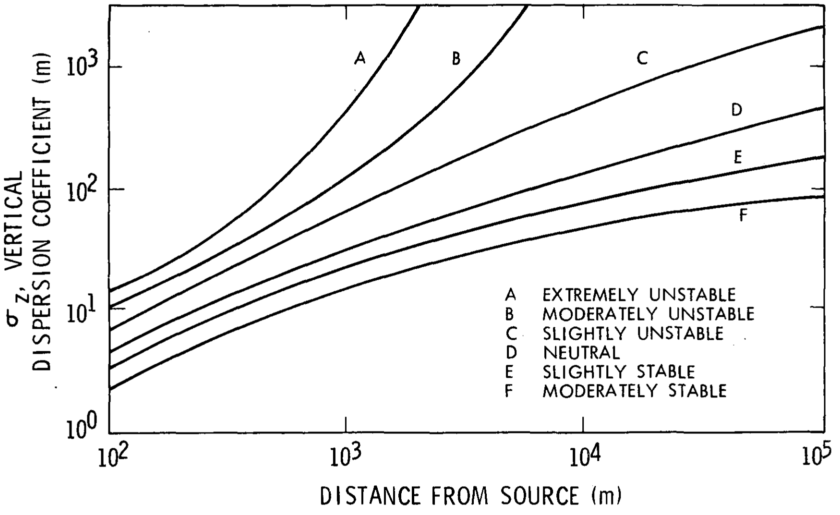

FIG.4. Correlations for vertical diffusion parameter, σ_z.

TABLE I. VALUES OF FACTORS "a" AND "b" FOR EVALUATING

Stability Class	A	B	C	D	E	F
a	0.45	0.11	0.061	0.033	0.023	0.015
b	2.1	1.1	0.92	0.60	0.51	0.45

within the region where equation (1) is used, σ_z is calculated as a function of distance, $\sigma_z = 1000\ a\ x^b$, with x expressed in kilometers. The factors a and b are listed in Table I. At greater distances where equation (2) is used, σ_z is assumed to be identical to the mixing layer depth, Z_m. Mixing depth data are provided in accordance with the work of Holzworth. [6]

The expression for effective height of radionuclide release as given in equation (1) includes factors for stack height (h_s), plume rise (Δh) and a "terrain factor" (h_t). Stack height is normally assumed to be 100 meters in regional evaluations, but may be separately specified for each nuclear site if desired. Plume rise due to thermal conditions or exit velocity is usually negligible for nuclear facility effluents, but the factor Δh may be calculated by the method given by Briggs[7] as a code option. The terrain factor, provided to account for differences in elevation in the area surrounding a nuclear site, is normally significant only in the highly stable atmospheric conditions specified by Pasquill classes E and F.

Deposition and Plume Depletion

Radionuclide concentrations in the plume may be depleted during transit by dry deposition and by precipitation-induced deposition, as well as by radioactive decay. In evaluating these depletion processes, the radionuclides considered in ARTRAN (Table II) are grouped according to their physical states and chemical natures, and coded as follows:

GS - soluble or chemically reactive gas
GN - noble gas
PH - particulate halogen isotope
PF - other particulate isotope (usually assumed to be an oxide)

The depletion of particulate matter in the plume due to dry deposition processes during upwind travel is derived from the relationship given by I. Van der Hoven,[5] as expressed in equation (3):

$$\frac{\dot{Q}_x}{\dot{Q}_o} = \left[\exp \int_o^x \frac{dx}{\sigma_z \exp\left(h^2/2\sigma_z^2\right)}\right]^{-\left(\frac{2}{\pi}\right)^{1/2} V_d/\bar{u}} \qquad (3)$$

Equation (3) is analyzed external to the ARTRAN code to provide a family of curves as a function of travel distance and stability class, normalized to values of V_d = 0.01 m/sec and u = 1.0 m/sec. These data are represented in ARTRAN as a set of depletion-ratio values, $\dot{Q}_x/\dot{Q}_o$, compiled as a function of downwind distance. Intermediate values of the depletion ratio are derived in the code by interpolation. Translation of these ratio to those specified by different values of V_d and u is accomplished as suggested by Van der Hoven by

$$\left(\frac{\dot{Q}_x}{\dot{Q}_o}\right)_2 = \left(\frac{\dot{Q}_x}{\dot{Q}_o}\right)_1^{\bar{u}_1 V_{d2}/\bar{u}_2 V_{d1}} \qquad (4)$$

where the subscript 1 refers to the curve value and the subscript 2 refers to the desired value.

Plume depletion factors for precipitation processes during transit are derived for soluble gases and particulate matter (the insoluble noble gases are not depleted). Engelmann[5] gives a set of curves that depict the ratios of washout coefficients to the diffusivities for soluble gases as a function of precipitation rate. The Kelkar-Hanford curve (Figure 5), which appears to be very nearly the mean of the other curves, was chosen for use in ARTRAN. A least-squares best-fit equation for this curve was derived as a function of the molecular diffusivity of the soluble gas constituents of the plume and precipitation rate. The washout coefficients for these gases may then be calculated. Very small particles, groups "PF" and "PH" in Table II, may also be regarded as gases if they are considered to be unaffected by electrical charges and impaction. Inasmuch as there are no definitive data available from which washout coefficients could be reliably deduced for particulates, this equation was also chosen to calculate values for particulates with the diffusivity factor set to 1. Because of the uncertainties that exist in defining rainout mechanisms and the additional difficulties in specifying the drop size spectra from monthly mean climatology, ARTRAN uses the estimate attributed to Byutner and Gisina [5] of

TABLE II. RADIONUCLIDES CURRENTLY INCLUDED IN HERMES MODEL CALCULATIONS

Nuclide	Key†	Nuclide	Key†	Nuclide	Key†
^{3}H	GS	^{95}Nb	PF	^{138}Xe	GN
^{14}C	GS	^{103}Ru	PF	^{134}Cs	PF
^{22}Na	PF	^{106}Ru	PF	^{137}Cs	PF
^{58}Co	PF	^{123}Sn	PF	^{140}Ba	PF
^{60}Co	PF	^{125}Te	PF	^{140}La	PF
^{85m}Kr	GN	^{127m}Te	PF	^{141}Ce	PF
^{85}Kr	GN	^{129m}Te	PF	^{144}Ce	PF
^{87}Kr	GN	^{132}Te	PF	^{154}Eu	PF
^{88}Kr	GN	^{131}I	PH	^{238}Pu	PF
^{89}Kr	GN	^{133}I	PH	^{239}Pu	PF
^{89}Sr	PF	^{135}I	PH	^{240}Pu	PF
^{90}Sr	PF	^{133}Xe	GN	^{241}Am	PF
^{90}Y	PF	^{135m}Xe	GN	^{242}Cm	PF
^{91}Y	PF	^{135}Xe	GN	^{244}Cm	PF
^{95}Zr	PF	^{137}Xe	GN		

†Key indicator symbols (used in ARTRAN code)

GN - Noble gas

GS - Soluble (or reactive) gas

PH - Particulate halogen

PF - Other particulate

3×10^{-5} sec^{-1} for the rainout coefficient. Thus, the plume depletions for washout and rainout are estimated, respectively, from

$$\chi = \chi_o \; e^{-\Lambda t} \tag{5}$$

and

$$\chi = \chi_o \; e^{-\psi t} \tag{6}$$

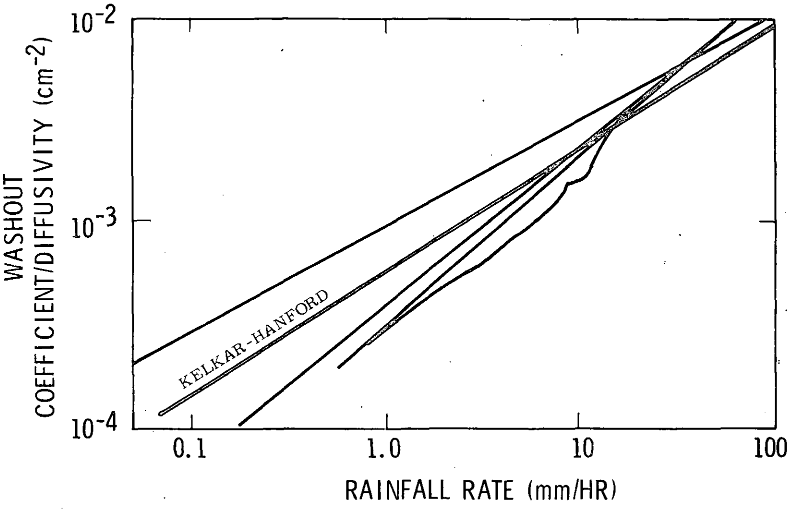

FIG.5. Ratios of washout coefficients to diffusivities for soluble gases.

During the winter season the general precipitation character changes from rain to snow in the northern latitudes. It has been noted by Engelmann[5] that snow apparently has about one to three times the effectiveness of rain at the same precipitation rate, for both washout and rainout of particles. As a conservative estimate, the precipitation character is taken to be snow for months of December, January and February, north of 34° latitude, and the washout and rainout coefficients are increased by a factor of two for this period.

Plume depletion due to radioactive decay, and enhancement from radioactive daughter products, are calculated respectively from

$$\chi = \chi_o \; e^{-\lambda t} \tag{7}$$

and $$\chi = \chi_o \; \frac{\lambda_1}{\lambda_2 - \lambda_1} \left[e^{-\lambda_1 t} - e^{-\lambda_2 t} \right] \tag{8}$$

The residual plume concentrations, after depletion by deposition and decay, represent the instantaneous concentrations at the receptor location. Deposition processes, of course, also operate at that location. Estimates of the rates of surface accumulation, δ, are obtained as the product of the air concentration, χ, and the appropriate deposition coefficient (V_d, Λ, ψ). The concentrations χ are calculated in ARTRAN as monthly average contributions from each source to the concentrations at the receptor location. However, deposition at the receptor location of radionuclides from a single source is not generally continuous. Hence, a factor τ must be introduced, representing the effective time each month in which a given deposition process is active. Thus, the monthly accumulation of radionuclides from a given source is:

$$(\delta\tau) = \bar{\chi}\ (V_d\tau_d + \Lambda\ \tau_w + \psi\tau_r) \qquad (9)$$

where the subscripts of τ represent effective times for dry deposition, washout, and rainout respectively.

The contributions $(\delta\tau)$ from each source are summed to obtain the total monthly deposition. Then, assuming that the summation $(\delta\tau)$ represents continuous deposition at reduced rates during the month, the monthly resultant concentration of each isotope, after decay, at the receptor location is calculated as:

$$C_m = \frac{\sum(\delta\tau)}{\lambda t}\left(1 - e^{-\lambda t}\right) \qquad (10)$$

where t is the total available time (e.g., in one month).

Long-Term Accumulation

In addition to its calculations of radionuclide deposition during the year of study, ARTRAN estimates the residual concentrations of radionuclides accumulated from deposition in prior years. In making these estimates, the monthly meteorological conditions affecting transport and deposition in each prior year are assumed to be identical to those of the year of study. Data on the year of startup and the assumed operating history of each nuclear facility are available from other portions of the HERMES model. Thus, the contribution of a plant to deposition in a prior year may be calculated by multiplying each month's contributions to deposition in the study year by a factor representing the plant's operating characteristics in the prior year.

For each increment of deposition thus calculated, estimates are made of subsequent depletion through removal by runoff water, leaching into subsoil regions, and radioactive decay, to provide estimates of residual concentrations in the year of study. Runoff and leaching calculations are made using rainfall data for the receptor location, and chemical and solubility characteristics of the elements involved.

In the modeling of large regions, use of the ARTRAN code can involve on the order of 10^{12} calculations in the iterative loops. To avoid excessive computer time requirements, therefore, options are provided which permit the omission of calculations which will have insignificant effect on the results -- for example, the calculation of contributions from a nuclear site at significantly greater distance from a receptor location than several closer, similar sites. Judicious use of these options permits significant savings in computer time without appreciable effect on results.

WATER TRANSPORT MODELING

The transport of radionuclides by water is modeled by means of the HERMES code WTRAN. This code is designed to calculate concentrations, in streams, of radionuclides both in solution and adsorbed on sediments.

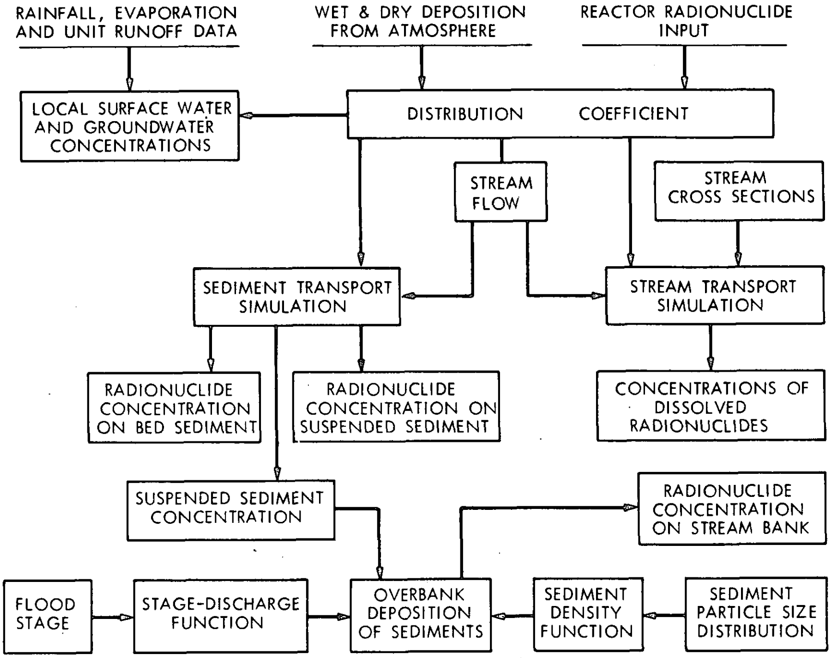

FIG.6. WTRAN code functional diagram.

In addition, concentrations are calculated for lakes fed by local runoff, for shallow groundwater recharged locally, and for artesian groundwater recharged from distant sources. A data flowchart for WTRAN is shown in Figure 6.

Chemical elements can be transported in a stream in two general ways: in solution, either ionic or molecular, and in suspension, through association with particulates ranging in size from colloidal material to particles several millimeters in diameter.[8,9] In the WTRAN model, ion exchange is assumed to be the determining factor in the transport of radionuclides through the hydrologic system. The chemical and physical composition of a stream can be an important factor in determining the fates of the various radionuclides that enter a stream. Consequently, the proportion of a given radionuclide entering a stream or the groundwater system that is transported in solution is determined by a distribution coefficient, K_D, which is the ratio of the fraction of ions sorbed per unit weight of sediment to the fraction of ions remaining in solution per unit volume of solution.

The distribution coefficient represents an equilibrium condition for trace quantities of chemical elements in the presence of relatively high homologous salt concentrations. It may be found experimentally for a watershed using representative samples of sediment and stream water. For rivers in the Tennessee and Cumberland Basins, K_D values for selected radionuclides are based on experimental data obtained at Oak Ridge National Laboratory.

Streamflow Calculations

The first step in the evaluation of transport of radionuclides in streams is the simulation of the river systems under study and the derivation of their streamflow characteristics. The basic structure of the river system is defined through use of two-letter symbols. The first letter corresponds to the designation of the trunk stream of one of the 14 major river basins of the United States, as defined by the U. S. Geological Survey. [10] The second letter of the symbol defines tributary streams, in upstream-to-downstream order, which enter the main stem. Distances along each river are defined in distance upstream from the mouth (river miles or river km). The confluences of streams are given in terms of river mile of the main stem (mile 0 of the minor stream is defined at that point). Centroidal areas which are included in the drainage basin of each stream are identified. The Tennessee-Cumberland river system is shown in Figure 7.

For computational purposes, the rivers are subdivided into reaches (or reservoirs). Detailed dimensions of river cross sections are modified for computer use by transforming them into equivalent trapezoidal sections where the surface and cross-sectional areas are conserved and the depth is taken as the average depth (Figure 8). A reach is specified in the code in terms of its length, the river-mile location of its upstream end, and the dimensions of the trapezoidal sections at each end of the reach.

Streamflow data are read in as mean flows for each month of the year. These data are not restricted to average conditions; median or low flows may also be of interest. The streamflow records, the basis for input information, are usually obtained from U. S. Geological Survey Water Supply Papers. [10] Frequently, the U. S. Army Corps of Engineers has

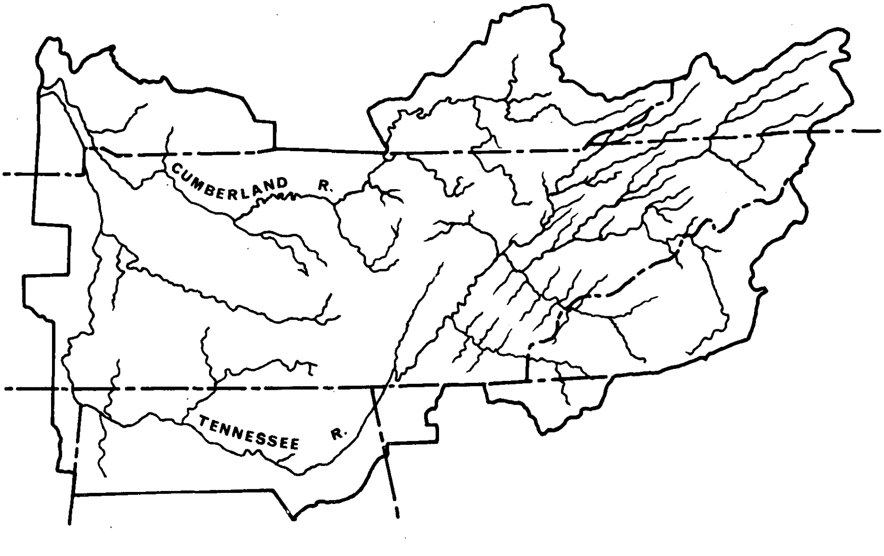

FIG.7. Tennessee-Cumberland River system.

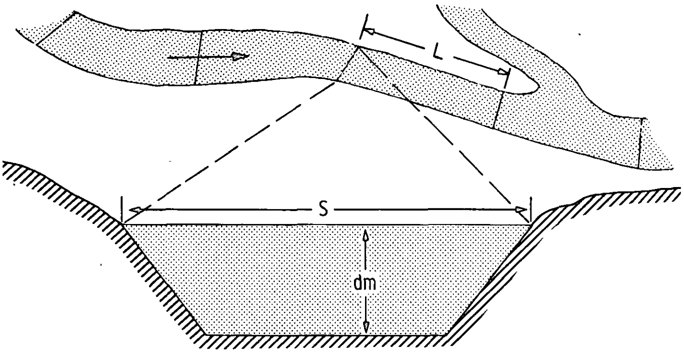

FIG. 8. Method of modeling river sections.

additional flow information in unpublished form; thus, statistical flow-duration curves or tables are often available. In the Tennessee Valley the Tennessee Valley Authority, which has detailed information of the area's hydrology, is providing the input data for the study.

Normally, recorded flow data are available only for a few points along a given stream. The flow data are interpolated by WTRAN to assign monthly flow values for each reach of river. In this process consideration is taken both of the discrete additions as tributaries enter into the main stream, and of the additions from surface water runoff in each centroidal area in the drainage basin. Using the assigned monthly flow values and dimensions of each river reach, flow transit times are calculated.

WTRAN is also capable of modeling the effects of transient conditions in flow or in radionuclide additions, and of density currents; normally these capabilities are not used in the present studies.

Solution Transport

The river basin streamflow model is used to calculate the transport, dilution, and radioactive decay of radionuclides introduced into the system. The model considers both the direct introduction of nuclides through the effluent streams of nuclear facilities and the additions of surface runoff water carrying air-deposited radionuclides. It was found convenient in the computer calculations to integrate the air-deposited radionuclides for a given area and assume that their entry into the stream occurs at a single point, rather than to utilize a more complex overland hydrologic routing procedure. For this a lumped-parameter approach is used in which a river location in the drainage network is specified at which the load from a given drainage is released to a stream. Releases from power plants or reprocessing plants are calculated as direct additions to streamflow at the plant locations.

Concentrations of radionuclides in solution in a given stream are calculated by the algorithm:

$$C_{x,t} = \frac{1}{Q_{x,t}} \left[Q_{(x-\Delta x),\ (t-\Delta t)}\ C_{(x-\Delta x),\ (t-\Delta t)}\ e^{-\lambda \Delta t} + \sum_{1}^{n} Q_i C_i \right] \quad (11)$$

where $C_{x,t}$ = concentration in reach x, at time t.
$Q_{x,t}$ = streamflow in reach x, at time t.
λ = radioactive disintegration constant.
Q_iC_i = fraction added in reach x due to tributary flow, cooling water from power plants, etc.

In cases where a tributary stream is large enough to be considered the trunk river of a subbasin, a concentration is calculated at its mouth for each time period simulated (normally corresponding to monthly steady-state conditions) and stored with its associated flow for input to the main stem. The calculations above are stepped through a stream system, from upstream to downstream, and are repeated for each nuclide for each month.

In the case of the Tennessee-Cumberland Basin and also other basins with large stratified reservoirs, a delay factor between the month of entry of radionuclides in solution into a reservoir and the month of exit from the reservoir is necessary to account for attenuation resulting from storage in stratified zones and from given patterns of withdrawal. This is accomplished in the code with an inflow-outflow monthly array for each reservoir in the system.

Sediment Transport

Sediment transport is determined by a set of logarithmic rating equations that describe the sediment load characteristics of a given reach as a function of streamflow:

$$S = a\,Q^b \tag{12}$$

where

S = sediment concentration or flow rate
Q = streamflow
a,b = constants

These equations are empirically determined at scattered locations throughout a given river basin. Since they generally would not be available at all points required for simulation purposes, it is frequently necessary to translate the equation to a desired location.

The coefficient "a" is a function of the drainage area involved. This can be visualized by assuming a steady-state discharge; if the drainage area doubles, Q would double; therefore, to maintain a steady-state sediment load the coefficient "a" would have to decrease. The decrease in "a" would be a function of "b". Therefore the correction for "a" is:

$$\hat{a} = a\,\frac{A}{A \pm \Delta A} \tag{13}$$

where

$\hat{a}$ = coefficient "a" translated to new drainage area
A = drainage area at sediment measuring station
$A \pm \Delta A$ = drainage area at site where equation is to be applied.

If streamflow regulation is present, coefficient $\hat{a}$ becomes further modified as follows:

$$\hat{\hat{a}} = \hat{a} \ \frac{A_L}{A \pm \Delta A} + \frac{A_1}{A \pm \Delta A} \ (1-E_1) + \frac{A_2}{A \pm \Delta A} \ (1-E_2) + \ . \ . \ .$$

$$. \ . \ . + \frac{A_\eta}{A \pm \Delta A} \ (1-E_\eta) \tag{14}$$

where

$\hat{\hat{a}}$ = translated "a" adjusted for regulation
A_L = local area unaffected by regulation
A_η = drainage area at dam site
E_η = sediment trap efficiency of dam

At steady state conditions, the trap efficiency of a dam would reduce the load at a downstream site. Since all of the contributions are linear, they can be absorbed into the coefficient, $\hat{\hat{a}}$ which can replace $\hat{a}$ of the previous equation to provide the sediment load at a site with upstream regulation.

Deposition or scour in a given reach, of sediments with sorbed radionuclides, is applied uniformly unless input data specify a special distribution. A mechanism is available so that deposition and scour are not necessarily mirror images of each other. Provision is also made to permit the rates and patterns of deposition in reservoirs to follow historical data, when these data are available.

Overbank Deposition

The deposition of radionuclide-bearing sediments on the flood plains of streams can contribute to radiation dose in some segments of the population. The periodic silting of flood plains is modeled by WTRAN as an extension of the sediment-transport calculations. These calculations require input data defining stage-discharge functions of a stream (or, alternately, the specification of flood stage) and the composition of sediments transported in the stream. Thicknesses and radionuclide content of sediment layers deposited during each period of flood are calculated.

Groundwater Transport

As infiltrating water containing air-deposited radionuclides percolates into the soil, ion exchange occurs between the soil and radionuclides in solution and/or suspension, gradually removing the active ions from the water. The exchanged ions are left behind in the soil phase, whereas the remaining solution percolates down to the water table where it enters the groundwater system. Groundwater, if not intercepted by wells, normally travels very slowly down the hydraulic gradient to a point where the water table intersects the land surface. Many streams, especially in humid regions, represent line sources along which groundwater effluent emerges to provide a relatively steady base flow. Groundwater may also enter aquifers which become confined between relatively impermeable strata. This confined water may be developed as a water supply several tens or hundreds of miles from the source area. Other percolating waters may be intercepted by impervious strata to form perched zones of saturation, which are above the normal water table and which may contribute slightly delayed storm runoff to streams. Percolating waters may also enter zones of cavernous limestone in which flow velocities may be measured in miles per day in contrast to velocities in granular material which are normally less than 15 miles per year. Hardened lava sometimes exhibits transmissive characteristics similar to cavernous limestone.

Because of the complexities involved in defining groundwater transport, WTRAN uses a simplified input to calculate concentrations, which requires the monthly runoff and radionuclide deposition rates for each centroid being studied. In addition, the centroid where the water is used as a water supply must be correlated with the source centroid and travel times specified. The input data may be secured by a cursory or detailed study as required. For example, in the case of confined aquifers which are extensively developed, it may be necessary to describe both potentiometric patterns and transmissive characteristics, or even carry out a special simulation, before the necessary input data could be developed. On the other hand, water supply obtained from shallow aquifers, recharged locally, would require minimal input data. Concentrations in both shallow groundwater and pothole lakes are calculated from the ARTRAN deposition data, since both have similar characteristics.

Together, the ARTRAN and WTRAN codes provide patterns of radionuclide concentrations in the air, on the ground, and in surface and ground waters at each centroidal location in the study area. In addition, concentrations of dissolved and sediment-borne radionuclides are provided at locations of interest -- e.g., drinking water and irrigation supply intake locations, sites of hunting, fishing, water sports, or other recreational activities -- throughout the study area. These patterns of radionuclide concentration provide the basis for the calculation of population dose in other parts of the HERMES model.

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DISCUSSION

W.O. SCHIKARSKI: Are you planning to introduce into your air transport model other processes such as photochemistry, catalytic chemistry, evaporation, condensation, etc., which might contribute considerably to the removal of nuclides from the atmosphere?

Secondly, what would you say is the accuracy of your results? For example, is it better than one order of magnitude?

B. LACHET: May I echo the question about accuracy. I think it would be a good idea to evaluate the prediction ranges of your estimates on the basis of the many empirical parameters or measured variables involved. I should like to hear an order of magnitude for the coefficient of variation of your estimates.

J.F. FLETCHER: At present we are considering as removal processes only dry deposition, in-cloud rainout, and below-cloud washout processes. However, we are looking at other processes as well, and if their effects appear important they will be included.

In answer to your question about accuracy, I should first point out that accuracy in a large-scale regional study such as this is very hard to establish. However, similar regional and national studies by other workers have resulted in estimates of 0.2 - 0.24 mrem/yr as an average dose, compared to our 0.17. Also, sample sub-regions of the study area have been investigated in detail by HEDL and ORNL, using different techniques. These analyses agree with the regional study to within about 10 - 15%.

R.G.D. OSMOND: Have you any figures available, or have any been published, for the fractions of different radionuclides transferred into streams after deposition on land catchment areas?

J.F. FLETCHER: Sensitivity studies were carried out under the Upper Mississippi River Basin project to evaluate the effects of transfer of air-deposited radionuclides into water. It appeared that, at least for some nuclides, appreciable amounts were in fact transferred into watercourses.

S. PRÊTRE: In paper IAEA-SM-172/26 it was shown that ^{32}P can make an important contribution to dose. However, I observe that ^{32}P does not appear in your list. Could you comment on this point, please?

J.F. FLETCHER: ^{32}P was one of the radionuclides initially considered in the study, but it was screened out because it did not appear to contribute appreciably to regional dose. The radwaste systems assumed for nuclear facilities in the study featured hold-up times long enough to ensure that only minute amounts of ^{32}P would be released.

K.-J. VOGT: While believing that HERMES may be considered as the most advanced model for calculating environmental exposures on a regional base, I feel that two points concerning the diffusion equation need discussion. First, I do not believe that terrain height can be taken into account by the correction factor h_t in the exponent of Eq. (1). As the pollution follows the stream lines over hills, it might be better to neglect terrain height.

Secondly, the application of the standard deviations recommended by Pasquill should be re-examined, because these values have been taken from ground-level, short-term-release diffusion experiments over flat and relatively smooth terrain. Studies on releases of longer duration from stacks, with diffusion over terrain of greater roughness, reported in the literature (e.g. the Brookhaven and the St. Louis investigations and the experiments performed at Jülich), yielded considerably greater standard deviations, which may be more realistic.

J. F. FLETCHER: I agree substantially with both your objections. The terrain factor is now being investigated in the context of modifications to our model. We are developing a modified factor which better takes into account the following of stream lines by air masses. This work, however, is not yet complete.

We use the Pasquill standard deviation only for the first two kilometres from the point of release. Because of the deviations you note, we use an empirical correlation for greater distances. The correlations shown were chosen in accordance with the best available supporting data.

IAEA-SM-172/41

METHODS OF ESTIMATING DOSE TO MAN FROM REGIONAL GROWTH OF NUCLEAR POWER*

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Abstract

METHODS OF ESTIMATING DOSE TO MAN FROM REGIONAL GROWTH OF NUCLEAR POWER.

In co-operation with the United States Atomic Energy Commission (USAEC), the Oak Ridge National Laboratory (ORNL), the Tennessee Valley Authority (TVA), the Atmospheric Turbulence and Diffusion Laboratory-National Oceanographic and Atmospheric Administration (ATDL-NOAA), and the Hanford Engineering Development Laboratory (HEDL) have undertaken a joint study of the potential radiological impact of an expanding nuclear power economy on the Tennessee Valley Region (TVR). The TVR study involves an appraisal of the generation, management, and control of radioactive effluents from nuclear facilities in the region to the year 2000. Consideration is given to nuclear power requirements to the year 2000 and sites for nuclear facilities, to radionuclides that may be released and their transport in air and water and concentration in terrestrial and aquatic systems, and to the estimation and interpretation of the potential dose to man.

This is the second such regional study initiated by the USAEC in the continental United States of America, the first considering the region defined as the Upper Mississippi River Basin (UMRB). A computer model known as HERMES (Hanford Engineering Regional Model for Environmental Studies) was developed by HEDL and applied in the UMRB study. The model permits estimates, on a regional basis, of the potential radiation dose that may be received by individuals and population groups as a result of radioactive materials estimated to be released from reactors and fuel reprocessing plants. Modifications in Hermes are discussed within the context of the uniqueness of the TVR and additions or improvements based on the initial application of HERMES. The paper, one of three companion reports, includes information on the data requirements and the components of HERMES concerned with the estimation of dose to man from internal and external modes of exposure.

INTRODUCTION

Current plans call for approximately 7800 MW(e) of nuclear power in the Tennessee Valley Region (TVR) by 1978. An additional 7200 MW(e) of nuclear units have been ordered and are tentatively scheduled for operation in the early 1980's. Further expansion of nuclear power in the TVR is expected; and, this expansion will contribute to the nuclear industry in the southeast United States, estimated to be about 150 GW(e) by year 2000 [1].

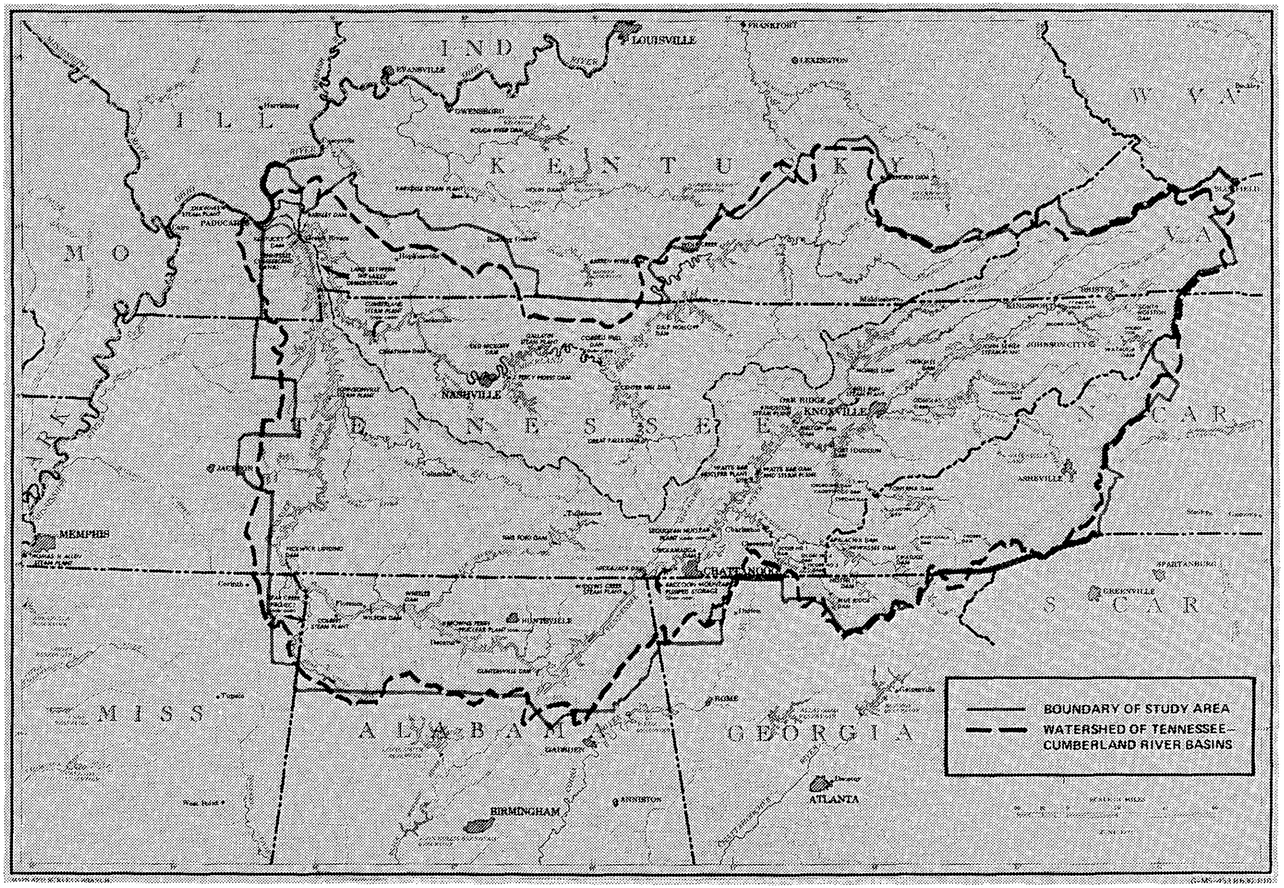

FIG. 1. Outline of Tennessee Valley Region study area.

Regional planning for such power growth patterns becomes essential not only to satisfy the lead time for commitment of power plant facilities but also to select the most effective locations for the new plants. One facet of regional planning in an expanding nuclear power economy is the need to assess the potential accumulative radiological impact of an increasing number of nuclear power plants. This need is recognized in the Tennessee Valley Region Study, and organization of the study team reflects the advantages of interagency cooperation that make use of special talents and resources within the cooperating groups (USAEC, ORNL, TVA, ATDL-NOAA, and HEDL).

Study Region

Figure 1 depicts the study region, an area of about 155,000 sq. km. The region is effectively the Tennessee-Cumberland River Basins and includes a total of 140 counties located in seven states (Alabama, Mississippi, Georgia, Kentucky, North Carolina, Virginia, and Tennessee). An air envelope of approximately 160 kilometers extends outward from the study region to include the effects on the region of the airborne transport of radionuclides released from plants located outside the region. In this paper we limit our consideration to that part of the regional model concerned with the estimation of dose to man and the data requirements to make such estimates. Information is included in two companion papers on those parts of the regional model concerned with power projections, radionuclide release from power reactors and fuel reprocessing plants, and the movement of radionuclides by water and air transport [2,3].

DESCRIPTION OF REGIONAL MODEL

A computer model known as HERMES (Hanford Engineering Regional Model for Environmental Studies) was developed by HEDL in the first regional analysis of potential radiological impact initiated by the USAEC [4]. The model is modular in form and may be used to forecast regional growth of the nuclear power industry and to consider releases of radionuclides from routine operation of nuclear facilities, the subsequent sojourn of these materials in the environment, and the pathways resulting in radiation exposure to man [5]. Details of the entire model and the results of its application in the region comprising the Upper Mississippi River Basin (UMRB) of the United States can be found in earlier publications [4,6].

Living Patterns Submodel

The physical habits and characteristics of an individual all contribute to a person's potential exposure to radiation. A Living Patterns submodel is used to generate some of the quantitative information needed to estimate the individual and population dose. Essentially the submodel is a data bank containing organized data on a county level--the lowest level at which data are available in sufficient detail in the United States.

Information is provided on the location, population, food production and food consumption for 73 types of foods, sources of drinking water, and recreational activities of each county in the study region. Each county is first classified as urban, rural-nonfarm, or rural-farm, depending upon the degree of urbanization, and appropriate coordinates are determined to identify either the largest population center (for an urban county), or geographic center (for rural-nonfarm and rural-farm counties). Food production includes homegrown food, commercial farming and irrigation practices, and food consumption considers the amount of each food consumed. The data are collated to establish the food balance of the area and, consequently, the source of food consumed. Drinking water is specified as to source (surface or groundwater) and to type of treatment. Recreational activities include swimming, boating and water skiing, fishing, and hunting. Examples of the format, sources, and limitations of these data are provided in a subsequent section on Data Requirements for TVR.

Radiation Dose Submodel

The last in the series of submodels is used to calculate dose to man and makes use of data processed from all preceding codes. The model includes a set of data tables that contain information on demography, diet, recreational activities, and dose factors to enable calculation of dose to several organs of individuals of various age due to different exposure modes [7].

At present consideration can be given to the radionuclide contribution to total body dose and the dose to six specific organs--the GI tract, bone, liver, skin, thyroid, and lungs. Three age groups, each including an average and a maximum individual, were considered in the UMRB study; i.e., children

TABLE 1. PRINCIPAL FACTORS IN ESTIMATING RADIONUCLIDE INTAKE BY INGESTION OF FOOD AND WATER

A. Concentration in Terrestrial Food
  1. Radionuclide intake by cow, cattle, etc.
    a. Feed ingestion rate
    b. Crop yield
    c. Foliar deposition – air and irrigation water
    d. Retention of foliar deposition
    e. Soil accumulation – air and irrigation water
    f. Concentration factor – soil to crop
    g. Translocation to edible parts of plant
    h. Time between deposition and harvest and between harvest and consumption
  2. Fraction of radionuclide intake by cow, cattle, etc., appearing in food product.

B. Concentration in Aquatic Food
  1. Concentration factor – water to fish or to waterfowl

C. Radionuclides Remaining in Processed Food

D. Concentration in Drinking Water
  1. Distribution coefficient
  2. Efficiency of water treatment plant

E. Consumption of Food and Drinking Water

(0-11 years), teenagers (12-17 years), and adults (>18 years). The four-year-old, the fourteen-year-old, and the Standard Man of ICRP were selected to represent the average child, average teen, and average adult, respectively. The maximum individuals are persons living at population centers whose dietary and living habits tend to maximize the radiation doses they may receive.

The modes of exposure considered are: (1) external exposure by immersion in air and water containing radionuclides; (2) external exposure from a surface containing radionuclides (ground surface, river banks, etc.); (3) inhalation of radionuclides in air; and (4) ingestion of radionuclides in food and water. The ingestion pathway incorporates a number of specific foodchain pathways and considers the effects of radionuclide deposition on plant surfaces and on soil, incorporation of radionuclides into plant and animal tissues, and concentration effects that occur in specific foodchains. The principal factors that are incorporated in the calculation of radionuclide ingestion are listed in Table 1. The interested reader is referred to Reference 4 in which the relationships of these factors are described and expressed in equation form.

Dose factors for external exposure were all derived on the assumption that the contaminated media (air, water, or ground surface) were of infinite extent and of uniform contamination [8,9]. Appropriate geometries were assumed for immersion in contaminated air and water and for exposure to contaminated surfaces. Ground roughness was assumed to attenuate ground surface contamination by a factor of 2 and dose factors were calculated for a distance of three feet above the surface. Expressions used to calculate internal dose due to inhalation and ingestion of radioactive materials were derived from those given by ICRP [10]. Standard Man values were used in the absence of better information for most age-dependent, metabolic parameters. These values include fractional uptake by the organ of interest of radionuclides taken into the body and the effective half time of these materials in the organ. Effective absorbed energy of all radiation emissions except gamma and x-ray photons were assumed to be independent of the individual's age. Absorbed fractions of photons within organs were adjusted in proportion to the one-third power of the ratio of the organ mass for a given age group to the mass of the respective organ for Standard Man.

DATA REQUIREMENTS FOR TENNESSEE VALLEY REGION

The data requirements of HERMES are considerable and not all of the data are readily available in the literature. Each region has its own peculiar physical, chemical, biological, and demographic characteristics which restrict direct use of some of the data developed in earlier studies and in other areas of the country. Several examples are used in this section to illustrate both the requirements and some of the limitations in the development of data for regional studies.

County Classification and Demography

The entire population of each county was assumed to belong to one of the three population types: urban, rural-nonfarm, or rural-farm. The 140 counties were first ranked and listed according to population density. This list was then divided into four density groups with different classification criteria applying to each group. For example, as listed in Table 2, a county in density group I was classified urban if 40% or more of its 1970 population was urban. A county with a density low enough to be placed into density group II, on the other hand, would require an urban population greater than 50% in order to be classified as urban. The numerical

TABLE 2. COUNTY CLASSIFICATION CRITERIA

Group	County Density (Persons/Square Mile)	County Classification
I	D>100	1. If urban population is greater than 40%, classify as urban. 2. If urban population is less than or equal to 40%, classify as rural-nonfarm.
II	100>D>50	1. If urban population is greater than 50%, classify as urban. 2. If urban population is less than or equal to 50%, classify as rural-nonfarm.
III	50>D>36	1. If rural-farm population is less than 29.5% classify as rural-nonfarm. 2. If rural-farm population is greater than 29.5%, classify as rural-farm.
IV	D<36	1. If rural-farm population is less than 16%, classify as rural-nonfarm. 2. If rural-farm population is greater than 16%, classify as rural-farm.

criteria used to classify counties were chosen to make the total assigned urban, rural-nonfarm, and rural-farm populations of the study region closely approximate the populations recorded by the 1970 census. These populations were distributed as follows: urban - 46%; rural-nonfarm - 42%; and rural-farm - 12%.

The total population of each county was assumed to be concentrated at a single point called the centroid. As in the UMRB Study, centroids of urban counties were chosen as the location of the most populous city within the county, while the location of the city or town nearest the geographical center was chosen for rural counties. Centroids chosen in this manner were within a few miles of the center of population mass calculated for each county from 1970 census data [11].

Projections of population to the Year 2000 for the counties within the study region have been made by others [12]. Table 3 illustrates the type of information developed on county identification and population. In addition, the total projected populations were divided into four age groups: less than 1; 1-11; 12-18; and greater than 18. The category of less-than-one-year-old was chosen in order to give particular attention to the infant. Since projection techniques to provide accurate age detail for small areas to the Year 2000 are not available, the percentage of the population within each age group was assumed to be the same for each county. The percentages chosen were consistent with state-wide projections of the Year 2000 population based on Series C fertility rate.

Food Production and Consumption

Consideration is given to 73 types of food comprising 126 items of fresh and processed materials. The latter permits distinction in radionuclide loss during processing; but information is so limited that an

TABLE 3. EXAMPLE OF COUNTY IDENTIFICATION AND DEMOGRAPHIC INFORMATION IN TENNESSEE VALLEY REGION

County		State	Centroid			Classification	Population		
							1970 Census	Density	Year 2000 Projection
Number	Name		Latitude	Longitude	Name		(In Thousands)	(No. per Sq. Mi.)	(In Thousands)
1	Davidson	Tennessee	36°08′	86°48′	Nashville	Urban	448	882	677
2	Knox	Tennessee	35°58′	83°55′	Knoxville	Urban	276	544	371
3	Hamilton	Tennessee	35°02′	85°16′	Chattanooga	Urban	254	462	388
4	Sullivan	Tennessee	36°33′	82°33′	Kingsport	Urban	127	308	249
5	Hamblen	Tennessee	36°13′	83°17′	Morristown	Urban	39	245	69
6	Madison	Alabama	34°44′	86°36′	Huntsville	Urban	187	233	331

TABLE 4. QUANTITY OF FOODS ENTERING HOUSEHOLDS IN THE TENNESSEE VALLEY REGION (1965 Data)[a]

Category Number		Food Category[b]	(Lbs/Week Entering Household/Person) Urban	Rural Farm	Rural Non-Farm
1	F	Berries	0.045	0.150	0.087
2	P	Berries	0.015	0.005	0.006
3	F	Tree Fruit	0.667	0.985	0.723
4	P	Tree Fruit	1.054	0.499	0.955
5	F	Melons	0.704	1.232	0.890
6	F	Tropical Fruit	0.427	0.351	0.389
7	P	Tropical Fruit	0.085	0.037	0.041
8	F	Citrus Fruit	0.803	0.469	0.652
9	P	Citrus Fruit	3.559	2.426	2.747
10	F	Potatoes	1.420	2.010	1.750
11	P	Potatoes	0.141	0.061	0.116
12	F	Root Vegetables	0.473	0.422	0.462
13	P	Root Vegetables	0.079	0.018	0.047
14	F	Leafy Vegetables	0.712	0.743	0.739
15	P	Leafy Vegetables	0.072	0.018	0.047
16	F	OAG Vegetables	1.139	2.150	1.558
17	P	OAG Vegetables	1.238	0.766	1.089
18		Misc. Grain	0.494	1.142	0.844
19		Rice	0.177	0.414	0.235
20		Wheat	2.982	3.350	3.335
21	F	Milk[c]	4.710	5.140	4.620
22		Milk Products	2.207	2.015	2.362
23		Butter	0.063	0.077	0.058
24		Eggs[d]	0.801	0.747	0.777
25		Beef & Lamb	1.671	1.204	1.378
26	F	Pork	0.446	0.513	0.485
27	P	Pork	1.007	1.096	1.033
28		Poultry	0.941	1.006	0.925
29		Game Birds	0.011	0.010	0.016
30	P	Ocean Fish	0.123	0.099	0.096
31	F	Ocean Fish	0.205	0.083	0.062
32		Shellfish	0.065	0.022	0.041
33		Sports Fish	0.000	0.179	0.167
34		Waterfowl[e]	0.002	0.013	0.009
35		Secondary Water	9.033	6.899	8.192

[a] Taken from References 14 and 15.

[b] F = Fresh; P = Processed; OAG = Other Above Ground.

[c] Quarts.

[d] Dozens.

[e] Taken from Reference 4, Table III-17 and corrected for Number of Persons per Household.

average loss factor for each type of crop for all radionuclides is assumed [13]. Loss factors range from 0.15 for wheat to 1.0 for milk. The food types are combined into 35 categories as a convenience in data handling. Food production by counties is based on statistics developed by the U.S. Department of Commerce [14]. Conversion of production data to common units (e.g., the processed weight of cattle) makes use of information published by the U.S. Department of Agriculture [15].

Transportation of food between centroids is constrained by (1) imports to satisfy a deficit must be supplied by excess production from the nearest centroid, and (2) exports outside of the region are permitted only after deficits are satisfied. An extensive market survey would be necessary to decide if local practices of distribution have an appreciable effect on these assumptions.

Food consumption data for the Tennessee Valley Region is summarized in Table 4 . The amount of food entering each household per week per person is listed by food category and by county classification [16,17]. These data coupled with information on the fraction of food entering the household that is consumed, the number of persons of each age group in the household, and the diets by age group permit calculation of the average food consumption by individuals of various ages [17-20]. Data are also developed for the infant (considered to be a six-month-old child) as another age category to be included in this study. Information on maximum diets in the study region is essentially non-existent and necessitates use of data developed in the earlier UMRB Study. It is necessary to use information on current agricultural practices and diet statistics in projecting the intake of food by residents in the area at Year 2000.

Irrigation Practice

The major source of data relating to the quantity of irrigation water used came from the 1969 U.S. Census of Agriculture [14]. This information was supplemented by field surveys conducted by the Tennessee Valley Authority. Table 5 provides a summary of available data, listing the annual quantity of irrigation water used by food crop in the seven states of the region. The sources of water are various ponds and streams; and these sources and the acreage irrigated are a part of the computer code specifications. Also, 10% of the home gardens on approximately 170,000 farms are estimated to be irrigated with an additional 1735 acre-feet of water per year.

Recreational Activities

Recreational activities considered in the study include swimming, boating and water skiing, fishing and hunting. Table 6 illustrates the format in which data are provided for each county in the study area. The participation shown as hours per man year is the per capita rate which also includes those persons who do not engage in these activities. Factors internal to the HERMES code are used to adjust the per capita data to obtain estimates for the average and maximum participant within each age group.

Information on swimming activities is derived from data for the age group of six and older [21]. Approximately two-thirds of those swimming are within the six to eighteen age range, with the remainder in the group over 18. Swimming activity is assumed to occur primarily in the centroid of residence. The hours spent swimming are distributed over the period May through September; off season swimming activity is believed to be insignificant.

Similar information is developed for boating, water skiing and fishing. Boating and water skiing activities are also derived from data for ages six and older with most of the participation due to persons in the 15-45 year

TABLE 5. FOOD CATEGORIES AND IRRIGATION WATER USED IN 140 SELECTED COUNTIES IN THE SEVEN TENNESSEE VALLEY STATES[a]
(Acre-Feet per Year)

Category Number	Food Category[b]	Ala.	Ga.	Ky.	Miss.	N.C.	Tenn.	Va.
1	Fresh Berries					0.5	12.0	0.5
2	Processed Berries							
3	Fresh Other Tree Fruit	3.0		62.5		8.0	62.0	45.0
4	Processed Other Tree Fruit							
5	Fresh Melons	6.0						
6	Fresh Tropical Fruit							
7	Processed Tropical Fruit							
8	Fresh Citrus Fruit							
9	Processed Citrus Fruit							
10	Fresh Potatoes			1.0			112.0	0.5
11	Processed Potatoes							
12	Fresh Root Vegetables							
13	Processed Root Vegetables							
14	Fresh Green Leafy					71.0	275.0	
15	Processed Green Leafy						208.5	
16	Fresh Other Above Ground Vegetables	70.0	6.0	26.0	2.0	191.0	311.0	
17	Processed Other Above Ground Vegetables	45.0					455.0	
18	Grain & Grain Products						116.0	
19	Rice & Rice Products							
20	Wheat & Wheat Products						165.0	6.0
	Total	124.0	6.0	89.5	2.0	270.5	1716.5	52.0

[a] Primarily from Reference 14.
[b] Information not available on Categories 21 through 35.

age range. Fishing activities are based on data for the age group of 12 and older. These activities are more dispersed than swimming and boating, and this is so indicated in Table 6 . In general, destinations for fishing are assigned within a 50-mile radius of the centroid of residence. Data on hunting activities are now being developed.

MODIFICATIONS IN REGIONAL MODEL

Several changes and additions are being made in the radiation dose submodel of HERMES prior to its application in the Tennessee Valley Region; changes in other components are explained elsewhere [2,3]. These modifications reflect in part the initial application of HERMES in the UMRB and the regional characteristics of the TVR.

Contaminated Ground Surface

Beta radiation from various distances above the surface of contaminated ground will be included in the calculation of dose to the skin. The calculation assumes that the beta emitters are associated with an infinite plain source of negligible thickness. Development of the dose-rate equations and the computer alogrithms is explained in Hine and Brownell, and Turner, respectively [22,23]. Table 7 contains several examples of dose factors

TABLE 6. EXAMPLE OF RECREATIONAL ACTIVITIES IN TENNESSEE VALLEY REGION

County		Swimming			Boating and Water Skiing			Fishing		
No.	Name	Activity Location	River	Hours Person Year	Activity Location	River	Hours Person Year	Activity Location	River	Hours Person Year
1	Davidson	Davidson	Cumberland RM[a] 190	6.35	Davidson	Cumberland RM 190	34.8	Davidson	Cumberland RM 170	63.6
								Sumner	Cumberland RM 230	5.3
								Wilson	Cumberland RM 250	6.9
								Rutherford	Stoney RM 40	5.3
								DeKalb	Caney Fk. RM 40	2.0
								Houston	Tennessee RM 80	2.0
								Stewart	Cumberland RM 90	2.0
									Total	87.1

[a] RM = river mile.

TABLE 7. EXAMPLES OF DOSE FACTORS TO SKIN WHILE STANDING ON CONTAMINATED GROUND
[(mrem/hr)/(pci/m^2)] × 10^{10}

Nuclide	Total Dose[a]			Gamma Dose[a] 1 Meter
	1.0 Meter	0.75 Meter	0.5 Meter	
^{90}Sr	41	130	420	
^{95}Zr	140	160	230	130
^{131}I	110	200	470	73
^{134}Cs	330	440	740	280
^{137}Cs	170	250	510	110

[a] EXREM code was run for a point 108 cm above the surface, equivalent to approximately 100 cm of air plus 9 mg/cm^2 of tissue. [23] The beta dose correction factors obtained by comparing the 108 cm dose to that 100 cm in air were used also for the other distances.

with skin as the reference organ. The importance of beta radiation as a contributor to skin exposure is reflected by the increase in total dose with a decrease in distance above the contaminated surface. The gamma component of the total dose at 1 meter is included for comparative purposes.

Distances of about 0.5 meter seem reasonable for exposure of the child and will also be included in the study.

Calculation of Potential Dose

The annual exposure in Year 2000 is a summation of twelve monthly values of external and internal dose calculated with HERMES. External dose is based on concentrations estimated to be in the environment each month. Internal dose includes the contribution from radionuclides taken into the body each month and from the residual material remaining in the body, integrated over a total period of twelve months. This procedure may overestimate the internal dose in Year 2000. The overestimate may be as large as a factor of 2 for uniform intake of a radionuclide whose effective half life in the body is greater than one year; as the effective half life approaches 0.01 years, the factor approaches 1.

Dose commitment is calculated for a fifty-year period only for the radioactive material inhaled or ingested during Year 2000. This will be supplemented to include dose commitment calculations beginning with the first year of the study and progressing to Year 2000 and beyond.

External Dose to Organs from Immersion in Air

Radionuclides present in air may irradiate internal organs as an external source in addition to the radiation received following inhalation or ingestion. A capability will be added during the TVR study to consider

TABLE 8. TOTAL DOSE RATE TO ORGANS OF THE BODY FROM ^{85}Kr
(rads/yr)/(μCi/m^3)

Organ	Dose Rate
Skin	1.8
Adipose tissue	1.5×10^{-2}
Lungs	3.2×10^{-2}
Red bone marrow	1.8×10^{-2}
Skeleton	3.65×10^{-2}
Ovaries	6.2×10^{-3}
Testes	1.6×10^{-2}
Lenses of eyes	1.8×10^{-2}

this external exposure mode. It is likely to be of particular interest for the noble gases because of their rapid elimination from the body and the resultant low dose delivered from the internal sources.

The methods and equations to be used in the development of dose factors are explained by Poston and Snyder [24], Dillman [25], Snyder et al. [26], and Snyder [27]. The source is assumed to be a semi-infinite cloud of uniform contamination. Radiation considered in the calculation of dose includes photons, beta rays, and bremsstrahlung produced in the cloud and bremsstrahlung produced as beta particles and electrons pass through the skin of the body. Also considered for the noble gases is the dose associated with the absorption of the gas in the tissue of the body and present in the airways of the lungs.

Table 8 contains an example of the dose factors that will be developed. Listed is the total dose rate to various organs of the body due to a unit concentration of krypton 85 in a semi-infinite volume of air.

CONCLUDING REMARKS

Only now do we approach the time for computer calculations in the Tennessee Valley Region Study. Many data have been collected and some remains to be developed. Modifications have been made in some of the submodels of HERMES based in part on its first application in the Upper Mississippi River Basin, and on the differences in the environmental characteristics of the Tennessee Valley Region. Additional changes in HERMES are likely as it continues to be used and as information becomes available from environmental monitoring programs and controlled experimentation by which to test its predictive capabilities.

Further enhancement of the radiation dose submodel is possible. This could include the ability to consider such factors as future changes in land use (i.e., agricultural practice, industrial and residential development, etc.) and in demographic characteristics (i.e., distribution of individuals by age and sex, changes in their dietary habits, etc.), the resuspension of radionuclides deposited on ground surfaces, the possible buildup and global circulation of radionuclides in the atmosphere and the potential dose to aquatic and terrestrial organisms other than man.

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DISUCSSION

K.-J. VOGT: As I understand it, the HERMES model has been established for calculation of the doses to different age groups of the population. I wonder whether the model can be used for the assessment of critical population groups on a regional basis, taking into account the effects of all installations contributing to the environmental exposure and thus extending the single-source approach to critical-group analyses.

K.E. COWSER: Included in the HERMES model are many parameters concerned with the habits and characteristics of people. We will be able to estimate dose for a variety of conditions at each centroid as a consequence of a number of sources; thus we can search out and identify the age group receiving the largest dose due to multiple sources as well as its centroid of residence.

R. LE QUINIO: Under conditions of 'normal' operation, is account taken of incidents that do not halt the operation of the facilities? In particular, has the model been checked with reference to actual releases of a previous year?

K.E. COWSER: Routine operation of nuclear power plants is predicated on releasing some radioactive materials into the environment under the constraints imposed by the operating licence issued by the US Atomic Energy

Commission. Waste management systems are designed to concentrate and contain these materials as far as practicable and to minimize releases to the environment. Reactors now in service are found to operate well within the authorized release units. Reactors for which licences are pending will release even less radioactive material because of modified operating procedures and equipment. Operational flexibility is to be required under the terms of the operating licences of these new facilities. Since the release rates to be used in the Tennessee Valley Region study are based on newer designs, we can only check our predictions of release rates when the plants become operational.

R.M. BARKHUDAROV: In establishing your forecasts, do you take account of technological progress, which should result in a reduction in radioactive releases to the environment?

K.E. COWSER: Yes, improvements in designs of waste management systems are taken into account in developing the source terms. For example, for light water reactors, consideration is given to charcoal absorbers in the gas system, treatment of building ventilation exhaust, and handling and treatment of steam generator blowdown.

F.O. HOFFMAN: Where do you get the data required for evaluating the age-dependence of food consumption and water intake? Are the US Census data comprehensive and accurate enough?

K.E. COWSER: Information on food consumption is available from surveys conducted by several organizations, including the US Department of Agriculture and the US Department of Commerce (Refs [14 - 19]). Information on population statistics for large areas is available from the US Bureau of Census and the US Environmental Protection Agency (Refs [11, 12]) and is sufficiently accurate for our present purposes.

APPLICATIONS OF A GENERAL COMPUTATIONAL MODEL FOR COMPOSITE ENVIRONMENTAL RADIATION DOSES*

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Abstract

APPLICATIONS OF A GENERAL COMPUTATIONAL MODEL FOR COMPOSITE ENVIRONMENTAL RADIATION DOSES.

A mathematical model for calculation on a large general-purpose digital computer of regional radiation doses resulting from large-scale use of nuclear energy was previously developed and reported. This general model has now been sub-divided to permit rapid calculations for the several exposure pathway groupings in an interactive mode using the BASIC computer language. The sub-programs are completely flexible as to the nuclides, body organs, and pathways for which radiation doses are to be calculated, but include at the present time: (1) approximately 150 radionuclides, including transuranics; (2) doses to whole body, skin, bone, lungs, thyroid and gastro-intestinal tract; and (3) sub-programs for cloud submersion, inhalation of nuclides other than radioiodines (resuspension of deposited nuclides is not included), ingestion of water and aquatic foodstuffs along with external dose from water and sediments, and ingestion of irrigated crops. Thyroid dose from inhalation and ingestion and dose to aquatic biota can also be calculated.

Dose factors in the programs for the various media-nuclide-organ combinations have been calculated using ICRP methods. For radionuclides with long effective half-lives the sub-programs calculate either total dose commitment for a single year's intake or the dose rate at the end of a specified period of years at constant annual intake. Transfer factors between trophic levels have been taken in most part from summaries published by others, and are updated as newer data become available. The major application to date of the compartment model has been the calculation for the US Atomic Energy Commission of environmental impact statements. Use is also being made of the same model for evaluation of potential radiological impacts associated with the ultimate fate of radioactive wastes for various long-term waste disposal concepts.

INTRODUCTION

Proper evaluation of radiation dose to people from environmental radiation sources requires the summation of dose contributions from all nuclides and exposure pathways of consequence. The relative importance of a particular pathway or nuclide frequently cannot be judged without calculation of the incremental radiation doses and the total dose.

A mathematical model has been developed at the Pacific Northwest Laboratory (operated for the USAEC by Battelle Memorial Institute) to calculate annual radiation doses and dose commitments, to the total body and selected organs of individuals and population groups, from both internal and external sources of radiation. The model includes all exposure pathways thought to be significant and for which a reasonable amount of basic data were available; radionuclides may be added or deleted as required. The dose calculation scheme was originally designed as the

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* Based on work performed under US Atomic Energy Commission contract AT(45-1)-1830.

final portion of a complex mathematical model developed to provide estimates of radiation doses resulting from a nuclear power economy. As such, it was programmed to be solved on a large digital computer (UNIVAC-1108). The model has since been subdivided into several exposure pathway groupings for rapid calculations, in an interactive mode using BASIC computer language, for application to less complex environmental situations.

This paper briefly reviews the original development of the mathematical model and the further modifications and applications for calculating radiation doses for 1) the environmental statements required by the USAEC licensing procedure for nuclear facilities and 2) hypothetical releases of nuclear wastes after extremely long periods of storage.

MODEL DEVELOPMENT

The original dose calculation scheme[1] was devised as part of the computer program HERMES[2] designed to (a) define the nuclear power economy potentially in place in the year 2000, (b) site the required nuclear facilities, (c) estimate the release and dispersion of their radioactive effluents, (d) calculate resulting radionuclide concentrations in air and water, and on soil and sediment, (e) calculate concentrations in foodstuffs, and (f) calculate radiation doses from intakes of air, water and foods, and from external exposure to air, water and ground. The output from item (d) above (in FORTRAN binary format) is used directly by the computer as input to items (e) and (f).

Data describing the following parameters were stored in tabular form in the program: 1) food consumption, and location and time of year of production, 2) hours per year and location of each of several types of recreation, and 3) factors for converting external exposure to radionuclides in air, water and on soil to skin and total-body doses. Diet, occupancy factors and dose factors are supplied in sets that are detailed by population type, age of person, time of year, and/or organ.

A schematic drawing of the computational scheme is given in Figure 1. The program contains a subroutine for calculating concentrations in foodstuffs at time of harvest from the concentrations in air, water and soil, and applies corrections for subsequent losses from radioactive decay, and home and commercial food preparation prior to consumption. This subroutine is represented by the lower half of the column labeled pathways in Figure 1 and the general equations involved are illustrated in Figure 2. The numerous parameters required for the application of the equations are defined in Table I.

Equations for calculating internal dose factors were derived from those given by the ICRP[3] and have previously been published by Soldat.[1,2] Effective decay energies for the radionuclides were calculated from the ICRP model, which assumes all of the radionuclide is in the center of a spherical organ with an appropriate effective radius. Where data were lacking, metabolic parameters for the standard man were used for other ages as well. Provision was made to allow addition of the dose contributions from certain nuclides which are essentially uniformly distributed in the body, such as ^{3}H, ^{14}C, ^{22}Na and ^{24}Na, to the individual organ doses.

Since the geographical region for which doses were originally calculated was large, it was subdivided in about 300 population centers. The doses to average and maximum individuals were calculated for each population center separately. An age-weighted, man-rem dose was calculated for each population center and then summed to get the total man-rem dose in the region. The reports printed by the program include total doses to various organ-pathway-age combinations and percent contribution to dose by either pathway or nuclide. The numerical results for the conditions postulated to exist in the upper Mississippi River valley region of the U.S. in the year 2000 are in press at this time as an AEC report.[4]

APPLICATION TO USAEC ENVIRONMENTAL IMPACT STATEMENTS

Currently, before any nuclear power reactor can be licensed in the United States, the USAEC prepares an Environmental Statement delineating the potential impacts on the environment from construction and operation of the facility. Battelle-Northwest, as an AEC contractor, has been assisting in preparation of the Environmental Statements. Included are evaluations of the radiological impacts on man and other biota from routine operation of the facility and from transportation of radioactive materials (clean fuel, irradiated fuel and wastes).

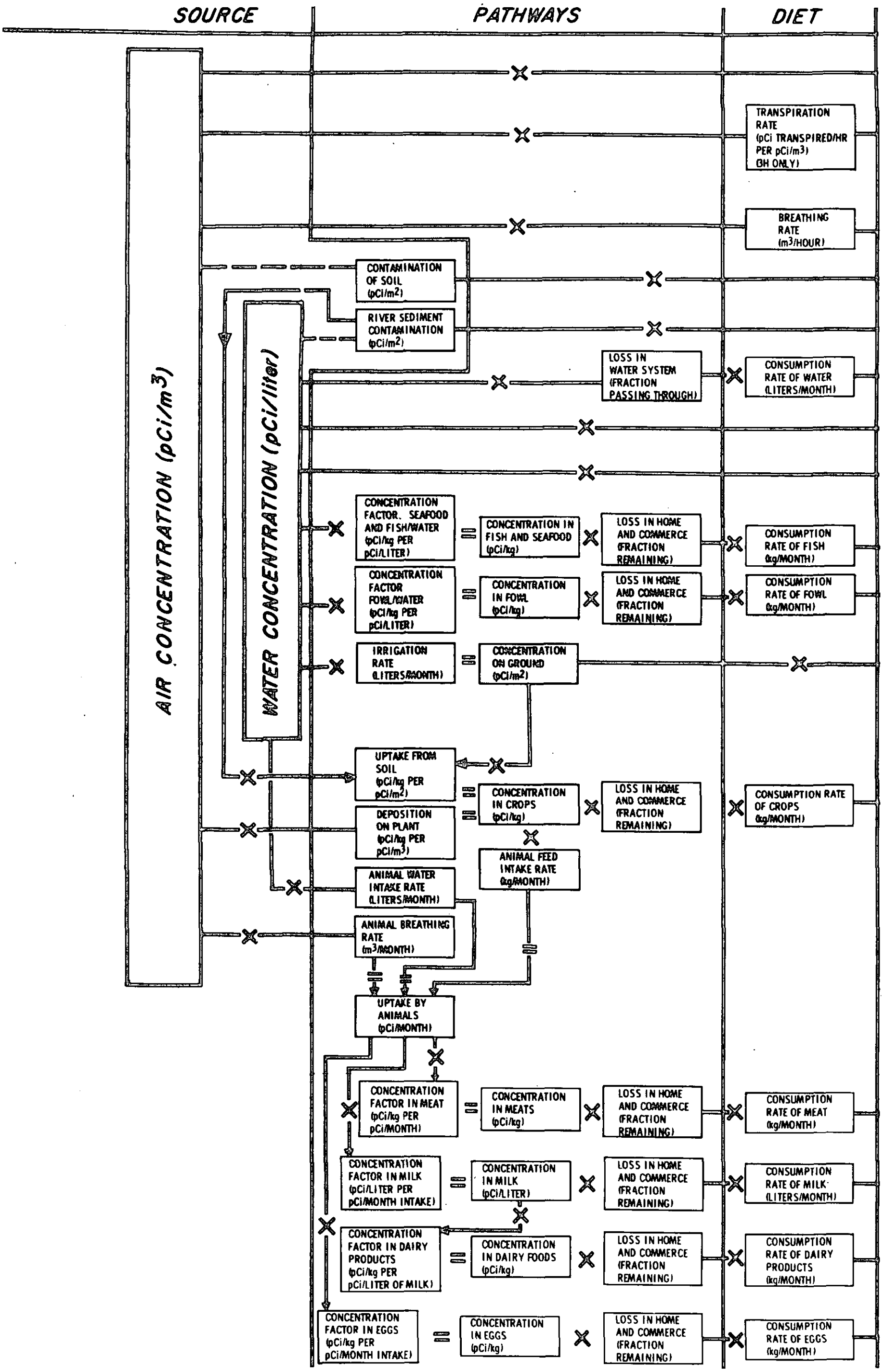
The radiological dose model developed as part of HERMES was selected as an appropriate basis for evaluating the radiological impact for these statements. The calculations for an Environmental Statement usually involved only one site with one set of meteorological data, and one population distribution. Because many of the input parameters would be the same for each case, it was convenient to reduce the complexity of the HERMES dose model and to reprogram the equations into an interactive computer language. The particular language available to us was an improved version of the BASIC computer language adapted by the Computer Sciences Corporation for its time-sharing system.

The dose calculation scheme was divided into two large segments -- ARRRG for radiation doses from aqueous effluents (Table II) and GRONK for radiation doses from gaseous effluents (Table III). Certain auxiliary equations were programmed to permit additional calculations. These programs are also described in Tables II and III.

Information specific to each reactor site was entered into files established for each reactor; for example, radionuclide release rates (source terms), meteorological data (joint frequency distributions) and population distribution (by direction and distance out to 50 miles). Data which varied by radionuclide, such as decay constants, bioaccumulation factors and dose conversion factors, were also entered into other permanent files. All of these files are described in Table IV.

Certain parameters which were highly variable, such as diets assumed for the particular locality, reactor coolant flow rate, decay time between radionuclide release and intake of contaminated food, were entered directly at the time each calculation run was made. This was conveniently accomplished through the interactive computer mode in which the local teletype terminal prints out a statement or question to be answered by the operator, such as "cooling flow in cfs =?"

The auxiliary program THY was originally designed to calculate thyroid dose to adults and children and the air concentration of specific radioiodine nuclides at the location of interest. It has since been incorporated into an expanded version of GRONK and now utilizes the release rates and atmospheric dispersions directly from the main program to calculate thyroid doses from seven iodine isotopes, I-129 through I-135.


SOURCE
PATHWAYS
DIET
AIR CONCENTRATION (pCi/m3)
WATER CONCENTRATION (pCi/liter)
TRANSPIRATION RATE (pCi TRANSPIRED/HR PER pCi/m3) (3H ONLY)
BREATHING RATE (m3/HOUR)
CONTAMINATION OF SOIL (pCi/m2)
RIVER SEDIMENT CONTAMINATION (pCi/m2)
LOSS IN WATER SYSTEM (FRACTION PASSING THROUGH)
CONSUMPTION RATE OF WATER (LITERS/MONTH)
CONCENTRATION FACTOR, SEAFOOD AND FISH/WATER (pCi/kg PER pCi/LITER)
CONCENTRATION IN FISH AND SEAFOOD (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF FISH (kg/MONTH)
CONCENTRATION FACTOR FOWL/WATER (pCi/kg PER pCi/LITER)
CONCENTRATION IN FOWL (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF FOWL (kg/MONTH)
IRRIGATION RATE (LITERS/MONTH)
CONCENTRATION ON GROUND (pCi/m2)
UPTAKE FROM SOIL (pCi/kg PER pCi/m2)
DEPOSITION ON PLANT (pCi/kg PER pCi/m3)
CONCENTRATION IN CROPS (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF CROPS (kg/MONTH)
ANIMAL FEED INTAKE RATE (kg/MONTH)
ANIMAL WATER INTAKE RATE (LITERS/MONTH)
ANIMAL BREATHING RATE (m3/MONTH)
UPTAKE BY ANIMALS (pCi/MONTH)
CONCENTRATION FACTOR IN MEAT (pCi/kg PER pCi/MONTH)
CONCENTRATION IN MEATS (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF MEAT (kg/MONTH)
CONCENTRATION FACTOR IN MILK (pCi/LITER PER pCi/MONTH INTAKE)
CONCENTRATION IN MILK (pCi/LITER)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF MILK (LITERS/MONTH)
CONCENTRATION FACTOR IN DAIRY PRODUCTS (pCi/kg PER pCi/LITER OF MILK)
CONCENTRATION IN DAIRY FOODS (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF DAIRY PRODUCTS (kg/MONTH)
CONCENTRATION FACTOR IN EGGS (pCi/kg PER pCi/MONTH INTAKE)
CONCENTRATION IN EGGS (pCi/kg)
LOSS IN HOME AND COMMERCE (FRACTION REMAINING)
CONSUMPTION RATE OF EGGS (kg/MONTH)

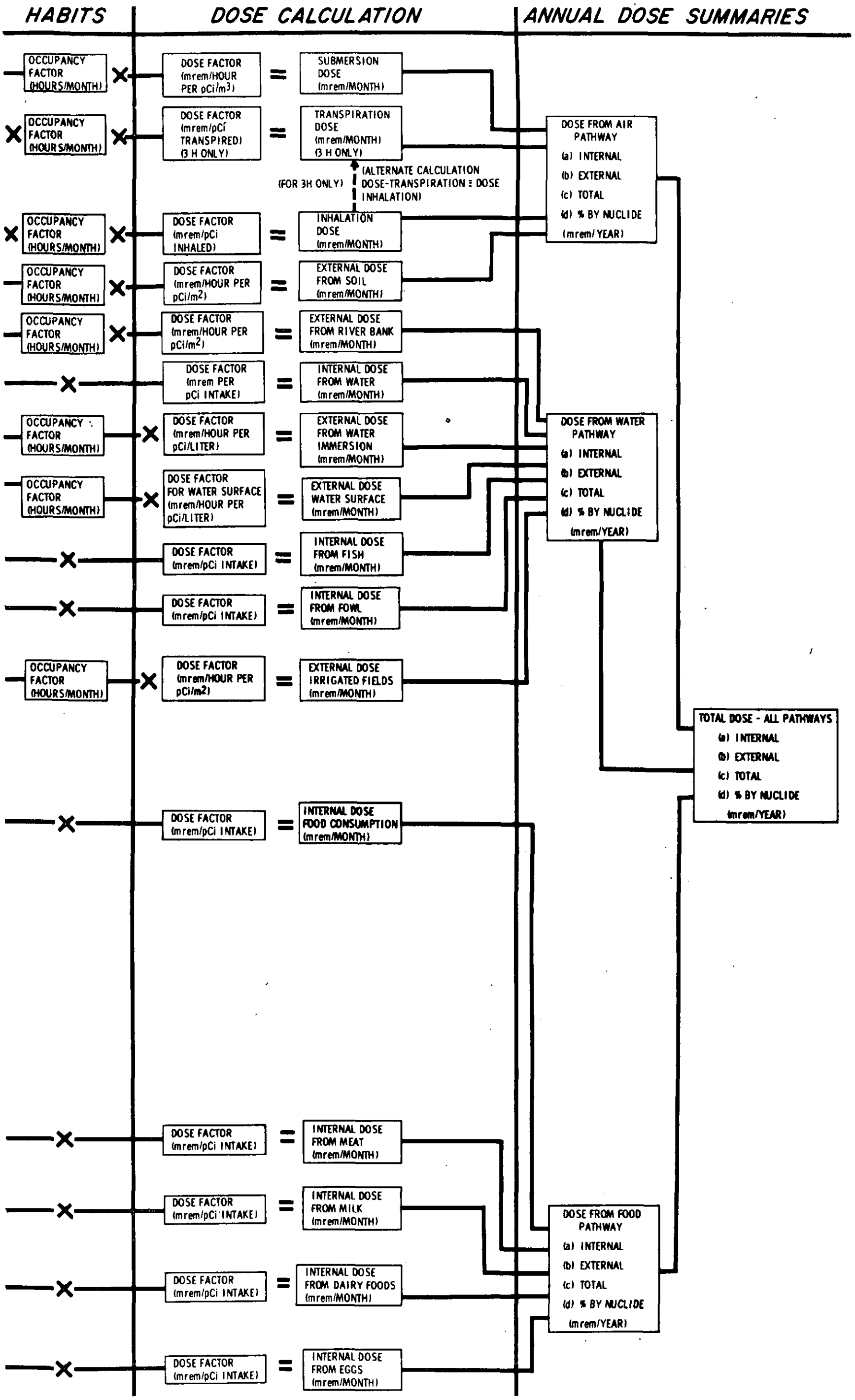


FIG. 1. Dose calculation model.

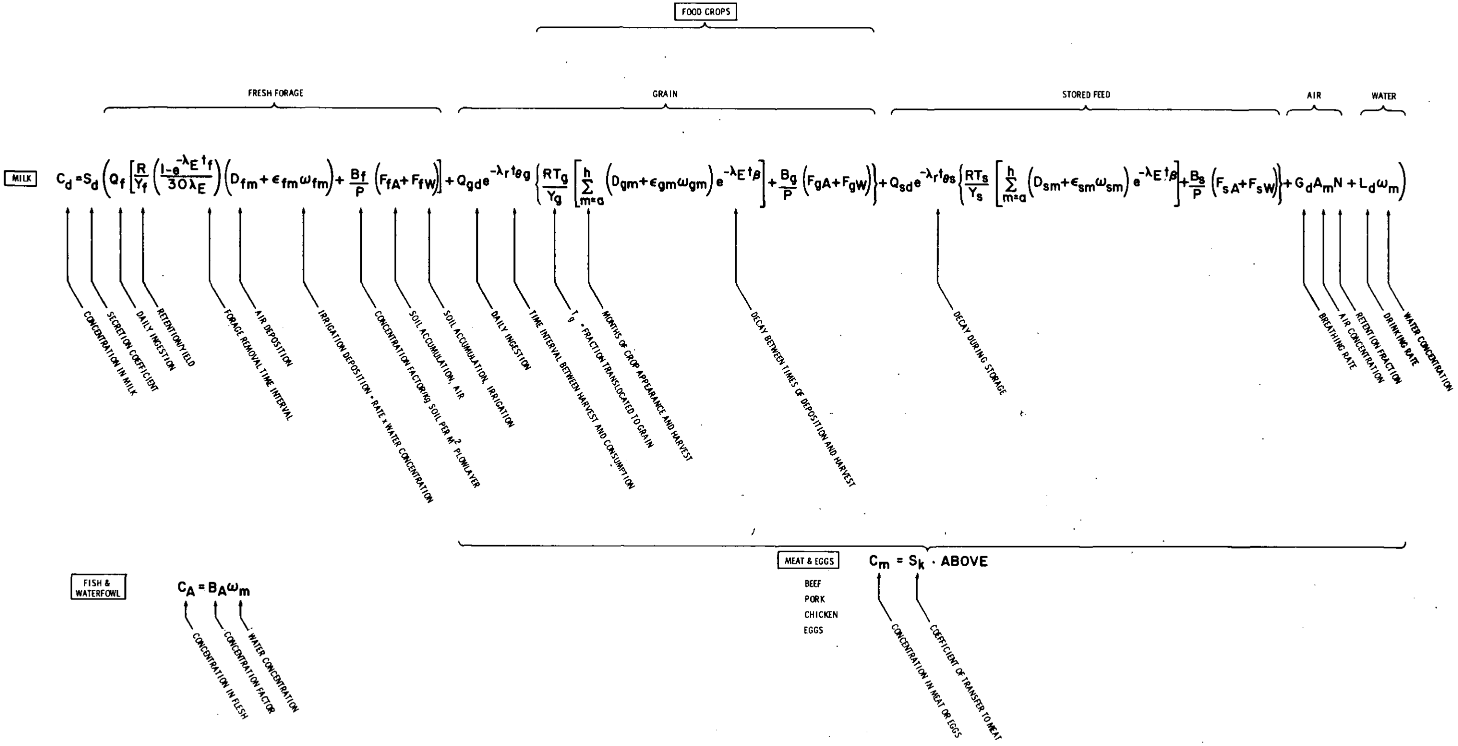


FIG. 2. Food pathways.

The mathematical model used to calculate the doses from air submersion is given in the equations that follow.

$$(D)_{x,\theta,d} = 8766 \sum_{i=1}^{I} \overline{\chi}_i \, (DF)_i \qquad (1)$$

Where

$(D)_{x,\theta,d}$ = The annual dose to total body (or skin) of a person located at a point x meters from the source in a direction d, averaged over a sector width of θ radians.

8766 = Hours per year

I = Number of isotopes

$(DF)_i$ = Dose factor for isotope (i) in units of mrem/hr per pCi/m^3 based on a half-infinite cloud geometry and corrected for the fractional penetration of beta and gamma radiations to the appropriate tissue depth (7×10^{-3} cm for skin and 5 cm for total body). (Reference 1)

$$\overline{\chi}_i = \sum_{J=1}^{N} \left(\frac{2}{\pi}\right)^{1/2} \frac{0.01\, f_J 10^{12} Q'_i}{(\sigma_z)_J (\bar{u})_J (\theta x)} \left[\exp\left(-\frac{h^2}{2(\sigma_z)_J^2}\right)\right] \left[\exp\left(-\frac{\lambda_i x}{\bar{u}_J}\right)\right] \qquad (2)$$

Where

$\overline{\chi}_i$ = Annual average concentration (pCi/m^3 of isotope (i) at point (x,θ,d). (Reference 5, pp. 113).

f_J = Percent of time wind blows in direction d under meteorological conditions J.

10^{12} = Picocuries per curie.

Q'_i = Release rate of isotope (i) in curies per second.

θ = Sector width in radians = ($2\pi/n$) where n is the number of sectors, normally 16.

x = Downwind distance in meters.

$\bar{u}_J$ = Average wind speed for meteorological condition (J) in meters per second.

$\frac{x}{\bar{u}_J}$ = Travel time of released material to point (x,θ,d) under meteorological conditions (J) in seconds.

λ_i = Radioactive decay constant for isotope (i).

h = Height of release in meters.

$(\sigma_z)_J$ = Standard deviation of vertical dispersion under meteorological condition (J)

N = Number of meteorological conditions.

The vertical dispersion may be derived for the Hanford (Fuquay-Simpson) four stability class format from equations given on pp. 141 and 405 of Reference 5, or from tables of (σ_z) versus Pasquill stability category taken from pp. 409 of Reference 5.

TABLE I. DEFINITION OF SYMBOLS FOR FOOD PATHWAYS EQUATIONS

A_m	Radionuclide conc. in air during month "m" (pCi/ℓ)
a	First month of crop contamination
B_A	Conc. factor for fish (pCi/kg per pCi/ℓ)
B_c	Food crop conc. factor (pCi/kg per pCi/kg soil)
B_f	Forage crop conc. factor (pCi/kg per pCi/kg soil)
B_g	Grain conc. factor (pCi/kg per pCi/kg soil)
B_s	Stored feed conc. factor (pCi/kg per pCi/kg soil)
C_A	Radionuclide conc. in fresh fish flesh (pCi/kg)
C_c	Radionuclide conc. in food crop (pCi/kg wet weight)
C_d	Radionuclide conc. in milk (pCi/ℓ)
C_{kb}	Radionuclide conc. in beef (pCi/kg)
C_{kc}	Radionuclide conc. in poultry muscle (pCi/kg)
$C_{k\alpha}$	Radionuclide conc. in egg (pCi/egg)
D_{cm}	Deposition on food crop during month "m" (pCi/m^2)
D_{fm}	Deposition on forage crop during month "m" (pCi/m^2)
D_{gm}	Deposition on grain crop during month "m" (pCi/m^2)
D_{sm}	Deposition on stored feed crop during month "m" (pCi/m^2)
F_{CA}	Radionuclide conc. in plowlayer of food crop from air deposition (pCi/m^2)
F_{CW}	Radionuclide conc. in plowlayer of food crop from water deposition (pCi/m^2)
F_{fA}	Radionuclide conc. in plowlayer of forage crop from air deposition (pCi/m^2)
F_{fW}	Radionuclide conc. in plowlayer of forage crop from water deposition (pCi/m^2)
F_{gA}	Radionuclide conc. in plowlayer of grain crop from air deposition (pCi/m^2)
F_{gW}	Radionuclide conc. in plowlayer of grain crop from water deposition (pCi/m^2)
F_{sA}	Radionuclide conc. in plowlayer of stored feed from air deposition (pCi/m^2)
F_{sW}	Radionuclide conc. in plowlayer of stored feed from water deposition (pCi/m^2)

TABLE I. (cont.)

G_b	Breathing rate of beef cow (m^3/day)
G_d	Breathing rate of dairy cow (m^3/day)
G_c	Breathing rate of poultry (m^3/day)
h	Month of crop harvest
I_b	Daily ingestion of radionuclide by beef cow (pCi)
I_c	Daily ingestion of radionuclide by poultry (pCi)
I_d	Daily ingestion of radionuclide by dairy cow (pCi)
L_b	Daily ingestion of drinking water by beef cow (ℓ)
L_c	Daily ingestion of drinking water by poultry (ℓ)
L_d	Daily ingestion of drinking water by dairy cow (ℓ)
m	Month
N	Radionuclide inhalation retention factor
P	2.24×10^2 kg soil/m^2 plowlayer
Q_f	Daily fresh forage ingestion by dairy cow (kg)
Q_{gb}	Daily grain ingestion by beef cow (kg)
Q_{gc}	Daily grain ingestion by poultry (kg)
Q_{gd}	Daily grain ingestion by dairy cow (kg)
Q_{sb}	Daily stored feed intake by beef cow (kg)
Q_{sc}	Daily stored feed intake by poultry (kg)
Q_{sd}	Daily stored feed intake by dairy cow (kg)
R	Deposition retention factor
S_d	Coefficient of transfer of radionuclide from diet to milk (pCi/ℓ per pCi/day)
S_{kb}	Coefficient of transfer of radionuclide from diet to meat of beef cow (pCi/kg per pCi/day)
S_{kc}	Coefficient of transfer of radionuclide from diet to meat of poultry (pCi/kg per pCi/day)
$S_{k\alpha}$	Coefficient of transfer of radionuclide from diet to egg (pCi/egg per pCi/day)
T_c	Food crop translocation factor
T_g	Grain crop translocation factor
T_s	Stored feed translocation factor
t_f	Time between successive removals of forage crop (days)
t_β	Time between deposition and harvest (h-m)(days)

TABLE I. (cont.)

$t_{\theta g}$	Time between harvest and consumption of grain (days)
$t_{\theta s}$	Time between harvest and consumption of stored feed (days)
Y_c	Food crop yield (kg/m^2)
Y_f	Forage crop yield (kg/m^2)
Y_g	Grain crop yield (kg/m^2)
Y_s	Stored feed crop yield (kg/m^2)
ε_{cm}	Irrigation rate for food crop during month "m" (ℓ/m^2)
ε_{fm}	Irrigation rate for forage crop during month "m" (ℓ/m^2)
ε_{gm}	Irrigation rate for grain crop during month "m" (ℓ/m^2)
ε_{sm}	Irrigation rate for stored feed during month "m" (ℓ/m^2)
ω_{cm}	Radionuclide conc. in irrigation water during month "m" (pCi/ℓ)
ω_{fm}	Radionuclide conc. in irrigation water during month "m" (pCi/ℓ)
ω_{gm}	Radionuclide conc. in irrigation water during month "m" (pCi/ℓ)
ω_{sm}	Radionuclide conc. in irrigation water during month "m" (pCi/ℓ)
λ_E	Effective decay constant
λ_r	Radioactive decay constant

TABLE II. PROGRAMS FOR CALCULATING RADIATION DOSES FROM LIQUID EFFLUENTS

Program	Data Files Used	Doses Calculated
ARRRG	ARGIN REL	Individual and population internal doses from ingested water and seafood plus individual and population external doses from water and sediment exposure.
FOOD	ARGIN REL FOOIN	Individual internal doses from various garden foods irrigated with water containing reactor effluent. Milk and egg doses from animals grazed on irrigated pasture and grain.
CRITR	ARGIN REL CRITEN	Internal doses to aquatic biota such as fish, crustacea, molluscs and plants from surrounding water; and to their predators such as ducks, muskrats, raccoons and herons.

TABLE III. PROGRAMS FOR CALCULATING RADIATION DOSES FROM GASEOUS EFFLUENTS

Program	Data Files Used	Items Calculated
GRONK	GIN TONIC GRONK REACTOR DATA FILE	Annual average atmospheric dilution factors (Ci/m^3 per Ci/sec released) versus distance and direction from the source ($\overline{\chi}/Q'$). Individual skin and total-body doses from exposure to 1/2 infinite cloud of effluents versus distance and direction from source and integrated man-rem (total body) doses to total population within 50 miles. Thyroid dose to child and adult from inhalation, and consumption of milk and leafy vegetables.
THY*	None	Air concentration of radioiodines from $\overline{\chi}/Q'$ and release rate. Thyroid doses to child and adult from inhalation and milk and leafy vegetable consumption.

* May be run independently of GRONK to calculate thyroid doses only.

Both the Hanford and the Pasquill formats are programmed into GRONK since raw meteorology data may be given in either format, depending upon the particular system employed by the nuclear power utility.

Equation 1 yields the yearly external dose to a person located at point (x,θ,d). The man-rem/yr is determined by multiplying the dose from Equation 1 by the population located within the sector of concern. Values of the dose at the point (x,θ,d) are assumed to be applicable to all individuals located in that sector from a distance of $x-\Delta x$ to $x+\Delta x$.

Dispersion factors of radioactive liquid effluents in rivers and lakes or oceans around the site are normally defined by the hydrologists concerned with thermal effects, and are entered as fixed ratios into ARRRG for each of the points of public exposure (fishing, swimming, drinking water withdrawal, etc.). The ARRRG program can calculate the increased environmental concentration resulting from recirculation of released effluent back into the water intake structure.

One important exposure pathway associated with liquid effluents is the external exposure to contaminated sediments. The factors for converting surface contamination in pCi/m^2 to gamma dose at one meter above a uniformly contaminated plane have been described in References 1 and 2.

TABLE IV. DATA FILES UTILIZED BY THE DOSE CALCULATION PROGRAMS

File	Content
ARGIN	Decay constants and internal dose factors for total-body, GI-LLI, thyroid and bone, plus external dose factors for skin and total body for exposure to sediment and to water for 136 radionuclides. Bioaccumulation factors for fish, crustacea, molluscs and algae for both fresh and salt water.
REL	Radionuclides released with liquid effluents (Ci/yr) for each plant under study.
CRITEN	List of absorbed energies in muscle by selected effective radii from 1.4 cm to 30 cm for 136 radionuclides.
GIN	Decay constants, external skin and total-body dose factors for air submersion for 136 radionuclides, child and adult thyroid dose factors for inhalation, ingestion and leafy vegetables.
TONIC	Constants used for the Pasquill meteorology calculation.
GRONK DATA FILE	Files for a particular plant containing radionuclides released with gaseous effluents (Ci/yr), population distribution out to 50 miles in 16 sectors and up to 10 annular rings, and meteorological data in tables of joint frequency of wind speed, direction and stability class in either Hanford or Pasquill format.
FOOIN	Plant uptake factors, egg transfer factors and milk transfer factors.

The calculation of sediment load, transport and concentrations of radionuclides associated with suspended and deposited materials is a complex problem. One method of addressing this problem was discussed in an earlier paper at this symposium by Fletcher, et al.[6] A simplified scheme for obtaining an order of magnitude estimate of the concentration of shoreline sediments was required for the dose calculation program ARRRG. The following equations were postulated as a possible method of quickly estimating such contamination levels.

$$C_{s,i} = K_C \left[\frac{C_{w,i}(1-e^{-\lambda_i t})}{\lambda_i} \right] \quad (3)$$

Where

$C_{s,i}$ = the concentration of nuclide (i) in sediment (pCi/kg)

$C_{w,i}$ = the concentration of the nuclide in water adjacent to the sediment

λ_i = the decay constant of the nuclide*

K_C = an assumed constant in units of liters/kg-day

t = the length of time the sediment is exposed to the contaminated water.

In the original evaluation of the equation, λ_i was chosen to be the radiological decay constant although the true value should include an unknown "environmental" removal constant. The relationship was tested and the value of K_C derived for several radionuclides by utilizing data on water and sediment samples collected over a period of several years in the Columbia River between Richland, Washington and the river mouth and in Tillamook Bay, Oregon, 75 km south of the river mouth.[7,8] The nuclides for which data were available, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{65}Zn, ^{95}Zr-Nb, ^{103,106}Ru, included those present from fallout and from operation of the Hanford reactors. Specific values of K_C for each nuclide averaged over all locations where shoreline samples had been collected[7] ranged from about 2 to 6 (liters/kg-day). Values of K_C for sediments collected in mid-stream[8] were about an order of magnitude lower.

Since the primary use of the equation is to facilitate estimates of the exposure rate from gamma emitters one meter above the sediment, an effective surface contamination was required. This was taken to be equivalent to having all of the nuclides contained within the top 2.5 cm of sediment** located at the surface. The dose contribution from the radionuclides below 2.5 cm in depth was ignored. The resulting equation is given below:

$$S_i = 10^2 T_i (1-e^{-\lambda_i t})(C_{w,i}) \qquad (4)$$

Where

S_i = the "effective" surface contamination (pCi/m^2) to be used in subsequent calculations, and

T_i = the radiological half-life of isotope (i) in days

A literature search is currently underway to find data for more locations and nuclides so that the validity of the equation can be tested. Preliminary indications are that most values of K_C derived for a wide variety of conditions seem to be within an order of magnitude of the average values found for the Columbia River.

APPLICATIONS TO LONG-RANGE WASTE MANAGEMENT STUDIES

Adaptation to long-range waste management studies has further demonstrated the flexibility of the original concept and of the conversational computer programs which implement it. For waste management studies,

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*If the presence of a radionuclide in water and sediment is controlled primarily by radioactive equilibrium with its parent nuclide, then the water concentration and half-life of the parent should be used in Equations 3 and 4.

**With a mass of 40 kg/m^2 of surface.

a very different group of radionuclides is involved than those released during normal operation of nuclear power reactors. Actinide elements and very long-lived fission and activation radionuclides are of much greater interest since the time span may be thousands or hundreds of thousands of years. Over 50 radionuclides in these categories have been included in the model to date. The importance of the actinide elements has dictated the addition of other body organs (lungs and liver) to those for which doses are calculated. The contributions of the inhalation pathway to organs other than the thyroid have also been incorporated into the model. For many of the transplutonium elements, valid data on biological transfer and accumulation factors are scarce; the gaps have been filled where necessary by chemical analogy with plutonium.

For studies of different waste management concepts, it is convenient to calculate population doses from potential releases to three different media - the ground, surface waters, and directly to the atmosphere. Studies in progress include the effects on calculated doses of different meteorological regimes, different geographical distributions of population, and varying waste storage periods. Outputs will include dose summaries by storage period, population groups and body organs, with listings of percentage contribution to dose by nuclide and by pathway. These listings will provide ready comparison of the effect on potential radiation doses of alternate separation processes for removal of specific elements from the wastes, as well as alternate storage or disposal concepts. Source terms and potential release mechanisms have not yet been completely delineated, but preliminary calculations with hypothetical source terms have demonstrated the utility of this version of the original HERMES computational model.

CONCLUSIONS

The generalized mathematical model described here for environmental dose calculations has now been applied to several problems of varying size and complexity, and its flexibility and adaptability demonstrated. As with any predictive model, its accuracy is dependent upon the accuracy of the input data and the pre-determined parameters. Advantages are the ease with which parameters can be changed as better data becomes available and the speed with which results can be obtained directly at a remote computer terminal. Future application to other problems of environmental dose estimation is planned.

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DISCUSSION

R. E. HOLMES: Can you give an estimate of the overall accuracy of the predictive programs taking into account their complexity and the paucity of input data in some areas?

R. F. FOSTER: The accuracy of the predictive programs is not easy to determine. Certainly it is no better than the accuracy of the assumptions fed into the program. For large populations exposed to average concentrations of ^{85}Kr and ^{3}H on a regional basis and over extended periods of time the accuracy is likely to be quite good. For close-in populations exposed to complex pathways I personally would consider accuracy within a factor of five to be very satisfactory.

Pamela M. BRYANT (Chairman): We have heard four papers this morning giving details of comprehensive studies of the potential radiological impact of nuclear power programs. I have also seen a draft environmental impact statement which shows the cost of saving population dose by means of additional waste treatment systems. All the countries with extensive nuclear power programs are involved in detailed calculations of potential doses to populations. As these potential doses are all very low, it might be interesing to build into computer codes the means of calculating the cost of calculating these doses!

S. STRAUCH: I feel that the converse might be more important: namely, determining the costs of not making the calculations and reporting on them in language the layman can understand. Perhaps the most recurring theme at this Symposium has been the difficulty of informing and convincing the public of the safety and low environmental hazard of nuclear power facilities.

Regarding the Upper Mississippi River Basin study, about 100 copies of the draft report were released to interested parties and government agencies and the response was most positive. One of the major reasons for conducting the Tennessee Valley Region study was the importance attached to it by the TVA as far as its public relations value was concerned.

P. J. BARRY: Taking the paper I am about to present myself as an anticipatory example, perhaps I may also offer some observations on the theme just broached by the Chairman. When working on the subject covered by my paper I was conscious of the cost of carrying out exhaustive environmental studies and of preparing input assessments. Thus my paper will show how I have attempted to estimate dose commitments due to a specific practice using the simplest possible model and whatever information was easily available to me. The purpose was, as I shall be explaining, to establish priorities concerning the need for further experimental work, and to assess the type of work that should be carried out with a view to optimizing the return on the resources committed. Unless ample resources are available, this preliminary survey should be undertaken before any large program of environmental dose assessment is started.

ESTIMATING DOSE COMMITMENTS TO POPULATIONS FROM RADIOACTIVE WASTE DISPOSALS INTO LARGE LAKES

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Abstract

ESTIMATING DOSE COMMITMENTS TO POPULATIONS FROM RADIOACTIVE WASTE DISPOSALS INTO LARGE LAKES.

A radiation dose commitment is a measure of the long-term consequences for a population of a practice leading to the irradiation of its members. An estimate of a dose commitment is useful for assessing the trend when long-lived radionuclides are released into the environment. In this case the per caput dose rate when equilibrium is eventually reached is numerically equal to the dose commitment per unit rate of release. An estimate of the dose commitment per unit released is therefore a necessary parameter for consideration at an early stage in planning the development of an area for the disposal of low-level radioactive waste. An important area in North America for this purpose is the Great Lakes basin. An abundance of cooling water and a large growing population combine to make the shore lines of the lakes attractive sites for nuclear power stations. It is reasonable therefore to expect that some low-level radioactive wastes will enter the lakes. Because of the importance of the lakes for other purposes, their use for waste disposal should be carefully planned. In the paper a preliminary attempt has been made to estimate the dose commitment following the release of radioactive wastes with particular reference to the release of ^{137}Cs to the lower lakes.

The dose commitment following a release of one curie of ^{137}Cs into Lake Huron is 0.4 man-rem. This estimate is very crude. The purposes for making it at this time were to (a) estimate the order of magnitude; (b) develop computation models; and (c) discover the sensitivity of the estimate to assumptions so as to define the most important variables for experimental investigation. Analyses to date suggest that the distribution of fish feeding and harvesting areas and the kinetics of exchange between sediments and water are particularly important areas for future study.

INTRODUCTION

The trend of the past few years has been toward lower prescribed radiation dose limits for the population from nuclear and related operations. Since, for the most part, actual population doses due to environmental releases of radioactivity from nuclear power stations have always been very small, prescribing lower maximum dose limits has had little effect on the actual doses received by people. The main effect of this trend has been to increase the difficulties and to direct efforts demonstrating that the doses from existing releases do not exceed arbitrarily low limits.

For example, it is relatively simple to demonstrate that a dose to a critical subpopulation does not exceed the maximum recommended for the group by ICRP because one can ignore many natural processes that would lower the actual doses received.

From the nature of the gross conservative assumptions that may be made one has good reason to suspect that actual doses will be lower by one, two and perhaps even three orders of magnitude compared with those calculated. However, to prove that the actual doses are lower by these orders requires a large increase in effort and the effort becomes disproportionately larger for each order of magnitude requested.

Before any such exercise is undertaken, therefore, its potential value should be appraised to ensure that its execution will serve worthy ends. This paper is an attempt to apply this principle. The special case chosen for illustrative purposes is that of estimating doses to populations due to releases of ^{137}Cs into Lake Huron.

ESTIMATING POPULATION DOSES DUE TO ^{137}Cs IN LAKE HURON

Population Doses from Distributed Sources

An initial amount (Q_o) of ^{137}Cs released into the lake will, by decay and elimination, decline with a rate constant k which is the sum of all the rate constants for decay, elimination and sedimentation assumed to be first order processes. The instantaneous dose rate to the population is proportional to the amount of ^{137}Cs present. Hence the total dose to which a population is committed following the release of Q_o Curies is:

$$\text{Dose} \ \alpha \int_{0}^{\infty} Q(t)\, dt = Q_o/k \qquad \text{..........(1)}$$

A rough estimate of k for ^{137}Cs in Lake Huron can be obtained by analysing the time course of ^{137}Cs concentrations measured at Kincardine and Port Elgin on the eastern side of the lake between 1963 and 1971 by the Department of Health and Welfare [1]. These are shown in Figure 1. The solid line was obtained assuming (1) all the ^{137}Cs came from fallout of weapon debris (2) the fallout rate is 1.5 times that of ^{90}Sr (3) the concentration in January 1963 was 0.46 pCi ℓ^{-1} and (4) the value of k is 0.06 month^{-1}.

A close fit to such scattered data would be difficult to obtain and hardly meaningful. The intention here was simply to obtain an order of magnitude estimate of k and this has been achieved. The effective half-life is a little less than a year.

Since the radioactive half-life of ^{137}Cs is 30 years and the half-residence time of water in Lake Huron is about 50 years the very short half-life of ^{137}Cs in the water must be due to rapid sedimentation. If the sedimentation rate in Lake Superior

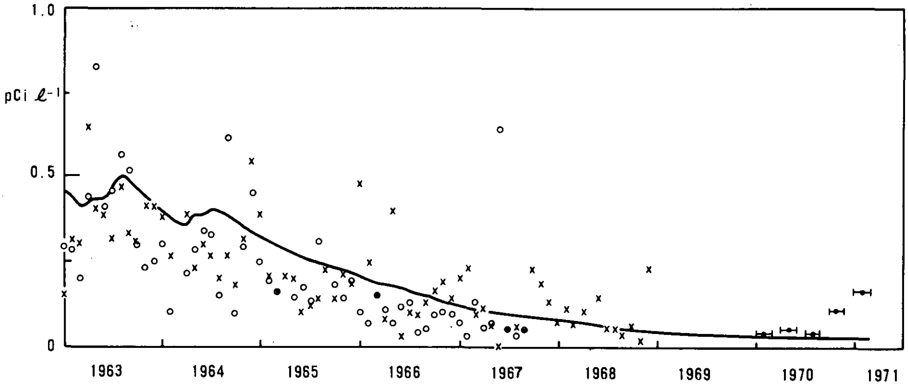

FIG. 1. Concentrations of ^{137}Cs in Lake Huron: Crosses – Kincardine; circles – Port Elgin, monthly samples; circles with bars – quarterly samples from Kincardine [1].

is similar very little ^{137}Cs is carried over from that lake to Lake Huron and presumably very little is also brought in by surface run-off so the simple assumption that the only source of the radionuclide is direct fallout on the lake surface is reasonable. The high sedimentation rate in Lake Huron also means that little of the ^{137}Cs in that lake will enter the lower lakes and the St. Lawrence River so the bulk of the transfer of ^{137}Cs to humans due to releases into Lake Huron will arise from the uses made of that lake alone.

The value of $k \approx 0.06$ month^{-1} gives an infinite time integral of ^{137}Cs equal to 1.4 Ci y for each Curie entering the lake. Assuming uniform mixing and a lake volume of 5×10^{18} cm^3 the time integral of the concentration is about 3×10^{-19} Ci y cm^{-3}. Taking an average drinking water intake to be 5×10^5 cm^3 y^{-1} and neglecting any beneficial effects of water purification the average intake through drinking water is 1.5×10^{-13} Ci per Curie entering the lake. The infinite time integral of the whole body dose rate following ingestion of one Curie of ^{137}Cs is 6.1×10^4 rem [2]. Hence the dose commitment per caput from drinking water is $6.1 \times 10^4 \times 1.5 \times 10^{-13}$ or about 9×10^{-9} rem per Curie. The size of population served with drinking water from Lake Huron is estimated to be 5×10^6 so the integrated population dose is 4.5×10^{-2} man-rem per Curie.

The population dose commitment due to consuming fish can be calculated from the annual fish yield and the concentration factor i.e. the ratio of the activity in one gram of fish to the activity per cm^3 of lake water.

Commercial yields of all fish taken from Lake Huron have declined during the last 30 to 40 years. In 1931 the total yield was 11 x 10^6 kg of which two thirds was from the United States side. In 1960 the total yield was about 5 x 10^6 kg of which again about two thirds was from the United States [3]. Canadian commercial landings from Lake Huron proper (excluding Georgian Bay and the North Channel) declined from 2 x 10^6 kg in 1961 to 0.6 x 10^6 kg in 1970. On the other hand recreational fishing catches on both sides of the border have increased and are now thought to be comparable to the commercial landings [4]. Thus the total annual consumption of fish is uncertain while predictions of future consumption are even more so. For present purposes the total commercial landings in the late 1950's 5 x 10^9 g y^{-1} will be taken and will almost certainly exceed the actual modern yields from both commercial and recreational fishing combined.

A concentration factor for ^{137}Cs in fish flesh depends on a number of variables and in view of the uncertainties and the absence of any direct measurements in Lake Huron a value of 4000 will be used as an average [5].

The total ^{137}Cs intake from fish consumption per Curie released is 3 x 10^{-19} x 4 x 10^3 x 5 x 10^9 Ci or 6 x 10^{-6} Ci corresponding to a population dose commitment of 60 x 10^{-7} x 6.1 x 10^4 equal to 0.4 man-rem.

This is about 9 times the corresponding dose due to drinking contaminated water. Since the average drinking water intake and concentration factor are constant the value of the ratio of the two dose commitments varies with the ratio of the number of people obtaining drinking water from the lake to the total amount of fish consumed. On this basis, the dose ratio will be smaller (6:1) only for Lake Ontario. For Lakes Erie and Superior it is about 60:1 and 120:1 respectively. In practice these ratios will be even larger because of removal of ^{137}Cs by water purification so for practical purposes the dose commitment due to drinking water can be neglected. However, this may not always be so if present trends of increasing human population and declining fish yields around the lakes continue.

Population Doses from Point Sources

In these calculations uniform mixing of the radionuclide in the body of the lake has been assumed. This is very nearly true for fallout from weapons tests. However, releases from nuclear power stations and other land-based sources are normally made from points into shallow water around the margin. This means that local concentrations will, for a fixed amount of ^{137}Cs, be higher than those calculated assuming uniform mixing. Since water intakes are close to shore and fish tend to feed in the shallow water, doses may be higher. Nevertheless for a fixed amount of radioactivity and given water depth, the mean concentration in a pool of contaminated water is inversely proportional to the area occupied while the fish yield from the pool is directly proportional to its area. Thus these two effects approximately cancel so that while the fish from the pool carry a greater body burden they make a proportionally smaller contribution to the total catch so the population dose should remain roughly the same as for the uniform mixing case. In fact, if the fish stay relatively close to shore the population dose may actually decline because after the ^{137}Cs has diffused beyond the limit of fish migration it is not available for uptake.

On the other hand for a given pool area near to shore the concentration is inversely proportional to the depth of water but fish yields will tend to be about the same irrespective of the depth. In this case the two effects do not cancel so that some dose enhancement over the uniform distribution case must be expected. However, to increase the dose by an order of magnitude the depth of the larger part of the productive zone would have to be less than one tenth the mean lake depth, i.e. less than 7.5 m, and this is not the case. If the average distance from the shoreline to the limit of migration of the fish to deeper water were known, the dose enhancement over that for uniform mixing could be estimated from the ratio of the mean depth of the lake to the mean depth of the productive zone.

The above arguments assume that no correlation exists between the spatial distributions of radionuclide, on the one hand, and the fish feeding and harvesting areas on the other. If a significant correlation between these variables did exist the dose per unit of radioactivity released would be greater. However, there is no known reason why such a correlation might exist except very close to the point of release. Here fish may be attracted to the higher temperatures and consequently greater food supplies. This would, in turn, tend to attract a greater number of fishermen. The temperature of the effluent will not be much more than 10°C above the unheated lake water and the water at this temperature would be confined to a small area because of the rapid losses due to mixing, latent heat of evaporation and heat transferred to the atmosphere.

A rise of temperature by 10°C will cause the productivity to roughly double. The rate of transfer of radionuclide from the lake to humans should thus increase by an amount equal to two multiplied by the ratio of the area of the water heated by 10°C to the total productive area of the lake. Since this ratio is extremely small for each source the overall increase in the population dose is negligible. Furthermore, during the warmer months of the year, the fish actually tend to leave the heated water zone [6] which effect will tend to lower the overall population dose.

Two other processes may result in higher doses. The ^{137}Cs held in the sediments may be slowly released back into the water or it may be taken up by bottom feeding organisms and subsequently enter higher food chains. The life of ^{137}Cs is limited ultimately by the radioactive decay rate so that the maximum dose increment these processes may cause is a factor of 30, and would almost certainly be a lot less in practice. Since water concentrations were used to estimate k any corrections required because of resolution of the nuclide from the bottom sediments are automatically included in the value selected. If the radionuclide deposited on the bottom subsequently enters food chains directly a separate correction would have to be made.

DISCUSSION

The estimate of 0.4 man-rem for the dose commitment due to eating fish after 1 Curie of ^{137}Cs is released uniformly into Lake Huron thus appears to be reasonable even for the case of releases occurring near the shore. There is a possible though unlikely chance that this estimate may, because of re-entry into the food chain of radionuclide deposited on the bottom sediments, be too low by up to an order of magnitude. There is also the possibility that because of the shallow water near shore the higher local concentration may lead to doses from shoreline releases of up to an order of magnitude greater than those for uniform releases across the lake. The population dose commitment from a release of 1 Ci of ^{137}Cs from a nuclear power plant could therefore conceivably be as high as 40 man-rem.

The practical question posed is whether or not this estimate of the upper limit for the dose commitment is high enough to justify the additional effort required to remove major sources of uncertainty attached to the dose commitment estimate which is one or two orders of magnitude lower. The population integrated dose is not very useful for estimating the injury that may be suffered. This purpose is better served by looking at the average doses to individuals. If we take the per caput average fish intake to be 20 g per day [7]* then 5 x 10^9 g of fish would be consumed by about 7 x 10^5 persons. Thus the average per caput dose corresponding to a population dose of 40 man-rem is 6 x 10^{-5} rem, which, bearing in mind the various time constants involved, would be delivered over 2 or 3 years.

During 1970 the release of ^{137}Cs from Douglas Point Nuclear Generating Station was about 25 Ci. The per caput dose commitment due to such a release would be 1.5 mrem. This is still very small and lower by a factor of at least 1000 compared to the lowest dose that has been shown to have caused harmful effects. However, if a large number of reactors and fuel reprocessing plants having similar release characteristics were to be installed

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* 15 g d^{-1} edible weight. Corrected for ratio gross weight/edible weight.

on the shores of Lake Huron then an assurance that the actual doses were at least an order of magnitude lower than the upper limit would be necessary. On this basis then it appears the required effort could perhaps be justified particularly if information gained is exploitable in other fields. In this case better knowledge of the migration distances and feeding habits of fish in the Great Lakes would be useful to the fishing industry.

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DISCUSSION

R.M. BARKHUDAROV: Is the monitoring of power station releases carried out in the cooling water in the pipe-line or in the lake at the point of release?

P.J. BARRY: The concentration referred to is that in the pipe-line discharging into the lake, and no 'credit' is taken for the dilution which occurs beyond the point of discharge.

R.M. BARKHUDAROV: Are the data on the ^{137}Cs concentration factors in fish your own, or are they taken from the literature?

P.J. BARRY: I have no measurements of my own about concentration factors in Lake Huron. The value of 4000 used is an average of measurements given in the literature.

P. BEAU: Have you made a similar evaluation of the dose received by the critical group, which is a traditional method of approaching these problems?

P. J. BARRY: The critical group in the traditional sense is not normally specifically identified. The present basis of licensing releases assumes that the 'critical group' is represented by a person deriving all his drinking water from the undiluted effluent or who eats 50 g per day of fish in equilibrium with the undiluted effluent. The critical group concept has no place in assessing population dose commitments if the maximum permissible dose commitment is orders of magnitude lower than the corresponding individual or critical group dose limit. For the population dose commitment assessment, doses lower by two or even three orders of magnitude than the critical group dose may contribute significantly and cannot be neglected without running the risk of seriously underestimating the dose commitment.

ETUDE RADIOECOLOGIQUE DU BASSIN RHODANIEN – STRATEGIE ET BILAN

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Abstract–Résumé

RADIOECOLOGICAL STUDY OF THE RHONE BASIN – STRATEGY AND BALANCE SHEET.

A special feature of the Rhone Basin is that its area of 120 000 km^2 includes all types of nuclear industrial activity. For about 15 years past some tens of thousands of persons have been handling and have been in a position to discharge radionuclides at several nuclear research centres and institutes, two fuel fabrication plants, a uranium enrichment plant, an irradiated fuel reprocessing plant, several nuclear power reactors and various other nuclear facilities. To this list must be added two power reactors under construction and several reactors in the planning stage. As regards power reactors, another original feature of the Basin is that along one and the same river we find three types of industrial reactor – graphite-gas, light-water and breeder – in operation or under construction.

In view of the foreseeable quantity of discharges, the authorities responsible for nuclear development initiated some 10 years ago a radioecological study, the strategy of which involved two-fold action: (a) to identify the pathways followed by the radionuclides discharged into the Basin from the whole nuclear complex and thereby to determine the critical points of the Basin, which could be used for guidance in future siting; (b) to verify the observations and estimates in the Basin itself at the critical points selected, and to modify the location of those points in the light of the results obtained and of new projects in the Basin as a whole (hydraulic works, new use of water). From numerous experiments and measurements carried out in situ the authors have found that the environment has in fact 'accepted' the discharges but in the context of this conclusion, which is optimistic as regards the past, they utter a warning for the future. Any new facility in the Rhone Basin must take account of the existing discharges. Discharge limits cannot be fixed merely by reference to the accepted standards. The modification of the profile of several watercourses in the Basin, especially the increase in the number of dams, indeed introduces new dynamics into the transfer of radionuclides. On the basis of this study of the Basin, the authors caution power station builders, during site selection, against the appearance of a new handicap that seems to assume greater importance with industrial development – the effect of synergism between radioactive, thermal and chemical pollutions.

ETUDE RADIOECOLOGIQUE DU BASSIN RHODANIEN – STRATEGIE ET BILAN.

Le Bassin rhodanien présente la particularité de rassembler sur ses 120 000 km^2 tous les types d'activités industrielles nucléaires. Depuis quinze ans environ plusieurs dizaines de milliers de personnes manipulent et peuvent rejeter des radionucléides dans plusieurs centres et instituts de recherches nucléaires, deux usines de fabrication des combustibles, une usine d'enrichissement de l'uranium, une usine de traitement de combustibles irradiés, plusieurs réacteurs électro-nucléaires ainsi que des installations nucléaires diverses. A cela s'ajouteront deux réacteurs de puissance en construction et plusieurs réacteurs en projet. Au niveau des réacteurs de puissance le Bassin présente une autre originalité qui réside dans le fait qu'on trouve, le long du même fleuve, en activité ou en construction, les trois types de filières au stade industriel: graphite-gaz, eau légère, surrégénérateur.

L'importance prévisible des rejets a conduit les responsables du développement nucléaire à lancer, il y a une dizaine d'années, une étude radioécologique dont la stratégie consiste en une double action: a) identifier les voies de transfert des radionucléides rejetés dans le Bassin par l'ensemble du complexe nucléaire et en déduire les points critiques du Bassin, ces points pouvant servir d'élément directeur pour les implantations futures; b) vérifier les observations et les prévisions dans le Bassin lui-même à travers les points critiques choisis, modifier le choix de ces points en fonction des résultats obtenus et des nouveaux aménagements réalisés dans l'ensemble du Bassin (aménagements hydrauliques, nouvelle utilisation de l'eau). Après dix ans un premier bilan peut être dégagé. A partir de nombreuses expériences et de mesures in situ les auteurs constatent que le milieu a bien «accepté» les rejets effectués, mais à travers cette conclusion,

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optimiste pour le passé, ils donnent en fait un avertissement pour l'avenir. Toute nouvelle installation implantée dans le Bassin devra tenir compte des rejets existants; il ne semble pas possible de fixer des limites de rejets par simple référence aux normes admises; en effet la modification du profil de plusieurs cours d'eau du Bassin, notamment la multiplication des barrages, provoque l'apparition d'une nouvelle dynamique dans le transfert des radionucléides. A partir de cette étude de Bassin les auteurs mettent en garde les constructeurs de centrales, lors du choix du site, contre l'apparition d'un nouvel handicap qui semble prendre une plus grande importance avec le développement industriel: l'effet de synergie entre pollution radioactive, pollution thermique et pollution chimique.

INTRODUCTION

Le Bassin rhodanien présente la particularité de rassembler sur ses 120 000 km^2 tous les types d'activités nucléaires: des centres et instituts de recherches nucléaires, deux usines de fabrication des combustibles, une usine de séparation isotopique de l'uranium, une usine de traitement des combustibles irradiés, plusieurs réacteurs électro-nucléaires ainsi que des installations nucléaires diverses, auxquels s'ajouteront, dans les années à venir, deux réacteurs de puissance en construction et plusieurs réacteurs en projet. Au niveau des réacteurs de puissance le Bassin présente une autre originalité, celle de regrouper, le long du même fleuve, en activité ou en construction et au stade industriel, les trois types de filières: graphite-gaz, eau légère et neutrons rapides.

L'importance prévisible des rejets a conduit les responsables du développement nucléaire à lancer, il y a une dizaine d'années, une étude radioécologique dont la stratégie consiste en une double action:

- identifier les mécanismes et les voies de transfert des radionucléides rejetés dans le Bassin par l'ensemble du complexe nucléaire et en déduire les cheminements préférentiels et les points «critiques» d'accumulation, en vue d'estimer les risques actuels ou futurs et de fournir les éléments directeurs pour les implantations futures;
- vérifier sur le terrain les hypothèses et les prévisions à travers les points critiques choisis dans la première phase, modifier éventuellement le choix de ces points en fonction d'une part des résultats obtenus et d'autre part des aménagements réalisés dans l'ensemble du Bassin et susceptibles d'avoir une influence sur les transferts (aménagements hydrauliques) ou sur les risques (nouvelle utilisation de l'eau).

1. LE BASSIN RHODANIEN

Cette implantation nucléaire a été réalisée et se développe dans une aire géographique qui compte actuellement 10 millions d'habitants. Cette région à forte expansion économique et démographique devrait atteindre, selon les estimations les plus plausibles, 19 millions d'habitants en l'an 2000.

L'activité économique de la région est à la fois industrielle et agricole. L'implantation industrielle la plus forte se situe suivant l'axe nord-sud, le long du fleuve lui-même. Elle est extrêmement variée, avec un développement marqué vers le pétrole, la chimie et la pétrochimie. Un point important est commun à toutes ces industries: la nécessité de prélever de l'eau du Rhône pour leur fonctionnement.

De son côté, l'agriculture a connu, dans ce Bassin, depuis 15 ans un développement assez important grâce à l'irrigation. Les surfaces irriguées, en constant accroissement, atteignent actuellement 300 000 hectares, ce qui représente le plus grand périmètre de l'Europe Occidentale.

L'activité agricole est constituée essentiellement par trois productions: viticulture, fruits, légumes, qui, à l'exception de la production viticole, sont en plein développement. La production fruitière de cette région représente 30% de l'ensemble de la production française et la production légumière 10%. Par ailleurs une production céréalière spéciale, le riz, mérite une mention particulière: toute la production rizicole française (85 000 tonnes en 1971) est concentrée dans le Bas-Rhône à l'aval de toutes les installations nucléaires. Il en est de même d'ailleurs pour une part importante des autres cultures citées précédemment, situées dans le périmètre d'irrigation de la Compagnie nationale du Bas-Rhône-Languedoc dont les ouvrages de prises d'eau dans le Rhône se situent à l'aval d'Arles, donc à l'aval de toutes les installations nucléaires actuelles.

Une autre activité économique est également concernée par les implantations nucléaires: le tourisme et les loisirs nautiques. Le fleuve lui-même est peu concerné, exception faite de la navigation de plaisance. La zone côtière par contre est utilisée d'une façon croissante pour la baignade et les sports nautiques. La portion du littoral influencée par les rejets, qui s'étend sur 80 km principalement vers l'ouest, possède actuellement une capacité d'hébergement de 20 000 lits.

Enfin un site protégé, la Camargue, reçoit à travers différents canaux une partie des eaux du Rhône. Cette réserve écologique méritait d'être étudiée en raison de sa richesse faunistique et floristique.

2. DEFINITION DE L'ETUDE

La mission principale qui nous était confiée consistait à identifier, par l'observation sur le terrain et l'expérimentation, les principales voies de transfert des radioéléments vers l'homme, de façon à fournir les éléments nécessaires pour la surveillance des sites actuels et le choix des implantations futures, l'objectif final étant d'estimer la capacité d'acceptation radiologique de ce Bassin en vue d'harmoniser le développement nucléaire et la sauvegarde de la santé et de l'environnement.

Les mécanismes de transfert mis en évidence, et les «situations critiques» identifiées, il convenait de vérifier in situ les prévisions déduites de la connaissance du site et de l'expérimentation, de manière à modifier éventuellement les conclusions en tenant compte, en particulier, des modifications pouvant résulter d'aménagements en cours de réalisation dans le Bassin.

Cette étude radioécologique se déroule depuis sept ans; ce délai s'explique à la fois par l'importance de la zone à étudier et par l'évolution continuelle de l'environnement (implantations nouvelles, travaux hydrauliques, évolution de l'agriculture).

Rapidement est apparu le rôle dominant de l'utilisation de l'eau dans l'estimation des risques. En effet la pollution atmosphérique ne dépasse guère le niveau des installations, alors que toutes les activités du Bassin, domestiques, agricoles, industrielles ou touristiques, sont directement concernées par le cycle de l'eau.

La consommation d'eau atteignait en 1969, pour l'ensemble des usagers, une valeur de 7 milliards de m^3 par an se répartissant comme suit:

usages domestiques	1 milliard de m^3
usages agricoles	3 milliards de m^3
usages industriels	3 milliards de m^3

L'étude radioécologique s'est donc développée à partir du thème central de l'utilisation de l'eau.

Les recherches effectuées in situ et les expérimentations de laboratoire ont été réalisées d'après le schéma suivant:

- étude purement hydrologique, mettant en évidence les mécanismes de diffusion des radionucléides dans l'eau, les interactions avec le débit solide dans le lit de la rivière, le régime de sédimentation et de dépôt en rivière et dans la zone côtière;
- étude de la radiocontamination du milieu vivant aquatique pour l'évaluation en laboratoire du facteur de concentration des principales espèces présentes dans le Bassin et la mesure de ces mêmes espèces prélevées dans le fleuve;
- étude des mécanismes de fixation et de diffusion des radionucléides apportés dans les sols par les eaux d'irrigation;
- étude du transfert des radionucléides, de l'eau d'irrigation, aux cultures et aux produits alimentaires transformés.

Les résultats obtenus au cours de ces expérimentations concernant les principaux compartiments du milieu ont permis d'établir un schéma prévisionnel du cheminement et du transfert des radionucléides jusqu'à la ration alimentaire.

3. LE BILAN

Le Bassin a reçu au cours des quinze dernières années plusieurs dizaines de milliers de curies à partir des différentes installations nucléaires. Que sont devenus ces rejets? Quel est la situation radioécologique actuelle du Bassin?

Un bilan peut être dressé en conclusion des travaux déjà effectués qui se décomposent en deux phases:

- étude des mécanismes et prévisions à partir des expérimentations réalisées en laboratoire (en particulier simulations effectuées sur des portions du milieu reconstituées dans des serres, des cases lysimétriques, des aquariums et dans une maquette hydrologique);
- constats.

3.1. Le fleuve

3.1.1. Etude des mécanismes et prévisions

Le Rhône, dont le débit moyen est de 1500 m^3/s, véhicule annuellement la masse de $49 \cdot 10^9$ m^3 d'eau. C'est dire l'importance du problème à traiter.

Tout d'abord a été recherchée une bonne connaissance des caractéristiques hydrologiques et physico-chimiques de l'eau et du débit solide.

Pour chaque radionucléide ont été déterminées les fractions solubles, fixées ou complexées ainsi que le rôle du débit solide dans la fixation, le transport et le dépôt.

Parmi les nombreux radioéléments étudiés, nous présentons ici les résultats obtenus pour les plus caractéristiques:

Strontium: fraction soluble 99, 5%
fraction fixée sur les suspensions 0, 5%
Césium: fraction soluble 75%
fraction fixée sur les argiles (inférieurs à 5 μm) 25%
Ruthénium: fraction soluble 80%
fraction associée 20%
Cérium: fraction soluble 2, 5%
fraction associée 97, 5%.

Les données recueillies ont permis de déterminer la dynamique globale des radioéléments.

En fonction du régime des eaux et des caractéristiques hydrologiques du fleuve nous avons projeté sur ce cours d'eau la distribution attendue des radionucléides.

L'absence de retenue sur le cours inférieur jusqu'à une date récente (1970) avait pour conséquence une faible sédimentation dans le lit principal et cette observation avait conduit à situer le risque au niveau des prises d'eau d'irrigation. La situation était d'ailleurs différente selon que l'eau brute était pompée dans le fleuve directement pour l'utilisation ou qu'au contraire elle transitait par les canaux, qui sont à la fois une zone de dépôts et une voie d'amenée d'eau aux sols et aux cultures. Les estuaires du Petit et du Grand-Rhône et la zone côtière, où la sédimentation est active, ont également attiré notre attention ainsi que toutes les solutions de continuité dans le régime ou la composition des eaux du fleuve (front de la langue salée, bras morts, etc.)

3.1.2. Constats

L'exploration systématique du lit du fleuve dans sa partie aval (180 km de fleuve) et la zone côtière (80 km) a fait appel à deux techniques:

- la mesure in situ avec une sonde gamma immergée sur les fonds et reliée à un sélecteur d'amplitude multicanaux;
- le prélèvement par carottage dans les zones détectées par la mesure radioactive en vue de l'analyse fine en laboratoire.

La figure 1 résume les résultats obtenus:

- au niveau de l'eau les teneurs moyennes en strontium-90 fluctuent entre 0, 8 et 1 pCi/l alors que la teneur en césium-137 reste inférieure à 0, 2 pCi/l;
- au niveau des dépôts nos estimations prévisionnelles ont été en partie confirmées: le lit du fleuve est peu concerné par les rejets (1700 à 6500 pCi/kg de sédiment en césium-137).

Comme prévu, les dépôts les plus importants se situent au niveau des ouvrages de la Compagnie nationale du Bas-Rhône-Languedoc (20 000 pCi de césium-137 par kg). Par contre les dépôts dans les estuaires et la zone côtière n'ont pas provoqué un taux de contamination notable (6000 pCi de césium-137 par kg). Les sédiments marins ne contiennent que des traces de césium non mesurables.

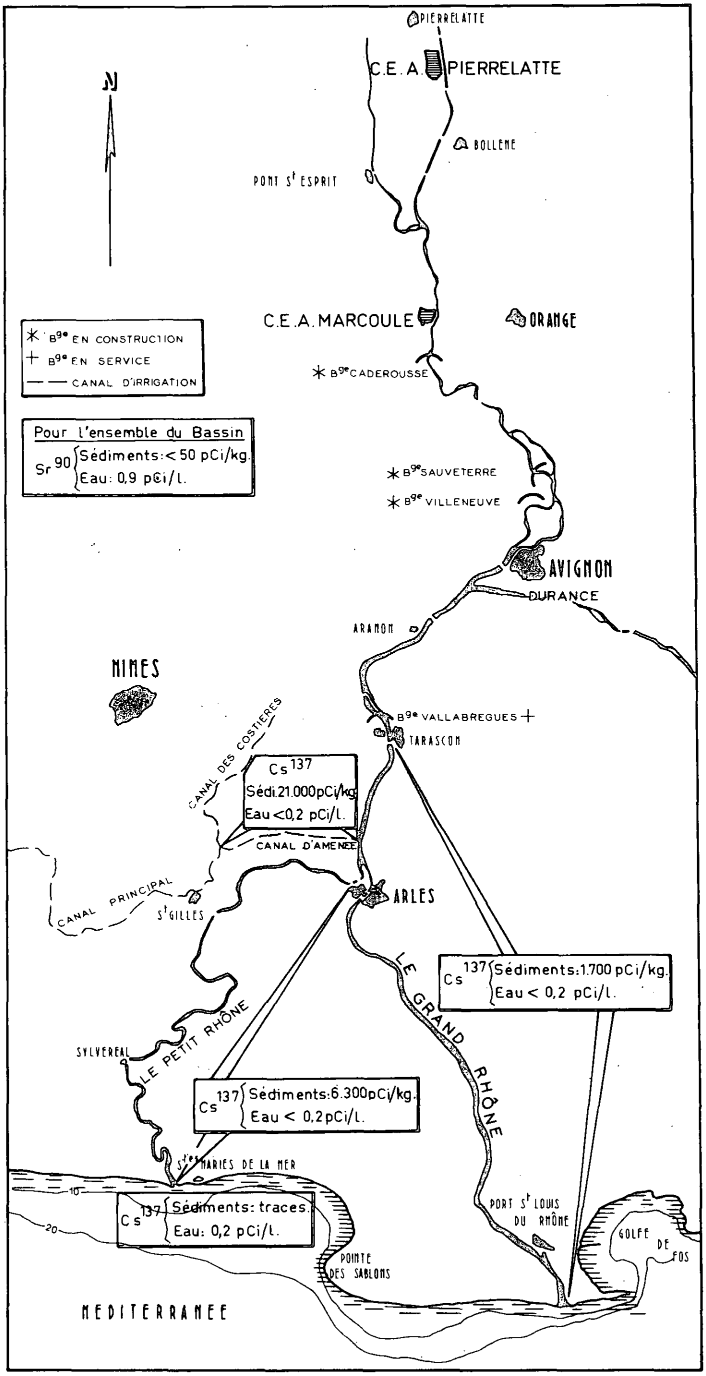

FIG. 1. Bassin du Rhône.

3.2. Le milieu vivant aquatique

3.2.1. Etude des mécanismes et prévisions

En l'absence de baignade ou d'alimentation directe du bétail par l'eau du Rhône, le milieu vivant aquatique est le premier maillon biologique concerné par les rejets radioactifs.

Un recensement des espèces aquatiques a été réalisé en tenant compte des travaux déjà effectués. Des essais de contamination en laboratoire sur plantes aquatiques, mollusques et poissons ont permis de déterminer leurs facteurs de concentration pour différents radionucléides tels que Sr, Cs, Co, Mn, Cr.

Les conditions hydrologiques, physico-chimiques et biologiques du Bassin du Rhône sont très variables, elles rendent difficiles une recherche et un contrôle biologiques. C'est pourquoi nous nous sommes attachés à choisir un indicateur représentatif sur le plan biologique et sur le plan sanitaire. L'espèce retenue a été l'anguille à cause de son caractère eurhyalin, de sa biomasse, de l'importance de sa pêche dans le Bassin rhodanien et de sa large consommation par l'homme.

Pour cette espèce, les études sur la contamination, la période biologique des différents éléments, l'effet des transformations culinaires sur la quantité de radioéléments ingérée par le consommateur, ont abouti à une estimation prévisionnelle des concentrations en radionucléides dans les poissons vivant dans le Rhône. Ces prévisions ont permis d'estimer que la concentration ne dépasserait pas quelques pCi/kg frais pour le strontium-90 et 50 à 100 pCi/kg frais pour le césium-137.

3.2.2. Constats

Le programme de prélèvements in situ mis en place a permis d'obtenir les résultats suivants: radioactivité globale moyenne 1500 à 5000 pCi/kg frais pour les anguilles pêchées en eau douce et 2500 pCi/kg frais pour celles pêchées en eau de mer.

A l'embouchure du Rhône, la concentration moyenne en strontium-90 des poissons est d'environ 0,30 pCi/kg frais, celle du césium-137, 150 pCi/kg frais.

En conclusion les résultats obtenus in situ induisent que la contamination est négligeable au niveau de la ration alimentaire même pour les gros consommateurs de poissons.

3.3. Les sols

3.3.1. Etude des mécanismes et prévisions

Les sols de la région peuvent se diviser en deux groupes:

- les sols alluviaux situés à proximité du fleuve, riches en éléments minéraux, exploités pour des cultures maraîchères et fruitières;
- les sols bruns calcaires situés en faible pente, souvent peu profonds, dont la mise en valeur est ancienne et où sont cultivés essentiellement arbres fruitiers et vigne.

Les mécanismes de transfert des radionucléides dans ces deux types de sol sont sous la dépendance de deux facteurs principaux: la présence de carbonate de calcium et le complexe absorbant.

Le carbonate de calcium insolubilise partiellement en surface un assez grand nombre de radionucléides et les rend peu disponibles pour la plante. Les formes échangeables représentent généralement 15 à 20% de l'activité présente en surface. Cette action du calcaire est plus particulièrement sensible pour le strontium (coprécipitation) et les lanthanides (formation d'hydroxydes): le ruthénium par contre est assez peu affecté dans son évolution.

Le complexe absorbant quant à lui fluctue entre 10 et 30 méq/100 g de sol. La teneur en argile a pour effet de retenir fortement le césium, tandis que la présence de matière organique favorise la rétention du strontium, du césium et de quelques formes de ruthénium; ce dernier élément se caractérise dès lors par une grande dilution dans l'ensemble du profil et il contribue pour une large part à la présence d'une activité notable en profondeur.

Des études en laboratoire permettent d'évaluer la répartition de la radioactivité de la zone superficielle comme suit:

- environ 40 à 50% de césium-137,
- 20 à 30% de strontium-90,
- le reste se répartit entre le ruthénium, l'antimoine, le zirconium-niobium et divers produits d'activation.

3.3.2. Constats

La connaissance de la répartition des radionucléides dans les profils pédologiques offre un intérêt tout particulier car elle permet de retracer «l'histoire» de la contamination du fleuve.

La mesure de la pollution radioactive de la zone étudiée a montré que les sols soumis à l'irrigation sont contaminés par l'eau à des degrés divers.

Les mesures globales d'indices bêta et gamma donnent des valeurs très dispersées. Le césium-137 et le strontium-90 sont pour l'instant les deux éléments rencontrés à des teneurs supérieures à celles résultant des retombées atmosphériques. Les mesures de spectrométrie gamma montrent que le césium-137 est présent dans presque toute la zone étudiée et que son taux se situe entre 2000 et 50 000 pCi/kg de sol sec, soit 40% de la radioactivité totale due aux produits artificiels. Le strontium, quant à lui, en représente 30%, le reste se répartissant entre le ruthénium, l'antimoine et le couple niobium-zirconium. Ces résultats confirment les prévisions.

La satisfaction, par l'irrigation, du besoin en eau de la culture apparaît comme le facteur primordial du niveau de contamination du sol, le type pédologique et la topographie du terrain ne jouant qu'un rôle secondaire.

Qualitativement, la répartition des radionucléides dans le sol dépend moins de leur nature que des façons culturales. Ainsi, il a été constaté que pour une rizière, la contamination est uniforme sur 20 cm de profondeur; pour une vigne, cette profondeur est de 10 cm et pour un verger de 30 cm.

On doit cependant noter que, pour l'ensemble des profils étudiés (150 environ), la fraction des nucléides disponible réellement pour la plante, représentée par les formes échangeables, n'est que de 15 à 20% de l'activité totale déposée.

L'état actuel de la pollution de la région a été schématisé sur des cartes où les terrains sont répartis en quatre catégories en fonction de leur contamination:

1) < 500 pCi/kg de sol sec
2) de 500 à 1 000 pCi/kg de sol sec
3) de 1 000 à 10 000 pCi/kg de sol sec
4) de 10 000 à 100 000 pCi/kg de sol sec.

D'après les premières observations, la contamination des zones étudiées semble liée à la proximité du fleuve, à la qualité de l'eau (décantée ou non), à l'intensité de l'irrigation (liée au besoin en eau des cultures), aux techniques culturales et aux modifications survenant dans les caractéristiques du fleuve: ralentissement du débit, différence de la composition des eaux (affluents, langue salée).

La contamination des sols est en fait assez réduite aussi bien en superficie (5000 hectares) qu'en intensité (faible radioactivité spécifique). De plus les transferts du sol vers la nappe ou la production végétale des différents radionucléides se font très lentement; toutes ces conditions tendent à minimiser les conséquences sanitaires de cette pollution.

3.4. Les productions agricoles

Seule la production végétale a été prise en considération, l'élevage pouvant être considéré comme négligeable.

3.4.1. Etude des mécanismes et prévisions

Une étude prévisionnelle en séries et sur parcelles expérimentales a permis d'établir les coefficients de transfert en fonction des principaux paramètres: sol, façons culturales, besoins en eau, espèces végétales.

La contamination directe des plantes a pu être réalisée grâce à un phytotron de grandes dimensions, dans lequel l'irrigation par aspersion a été simulée.

Il ressort des expériences ainsi menées que la contamination des espèces est directement liée à leurs besoins en eau et que les feuilles sont en général la partie de la plante la plus contaminée.

A travers les valeurs obtenues nous avons établi un ordre de risques de pollution encourus par les cultures:

- à court terme prédomine le transfert par la voie foliaire (irrigation par aspersion); le radioélément à prendre en considération est le césium-137;
- à long terme se surimpose la contamination indirecte par voie racinaire qui redonne toute son importance au strontium-90.

3.4.2. Constats

L'observation sur le terrain a débuté en 1969 en tenant compte des résultats déjà obtenus en hydrologie et pédologie et des estimations prévisionnelles (fig. 2). Des témoins ont été choisis sur des parcelles non irriguées par le Rhône. Trois espèces fruitières et six légumes ont été prélevés pendant trois ans.

Au niveau des mesures de radioactivité globale (β, γ) (tableau I) les valeurs ne sont pas sensiblement différentes de celles obtenues par ailleurs et qui peuvent être attribuées aux retombées. (En strontium-90 les contaminations consécutives aux retombées peuvent atteindre 30 pCi/kg frais.)

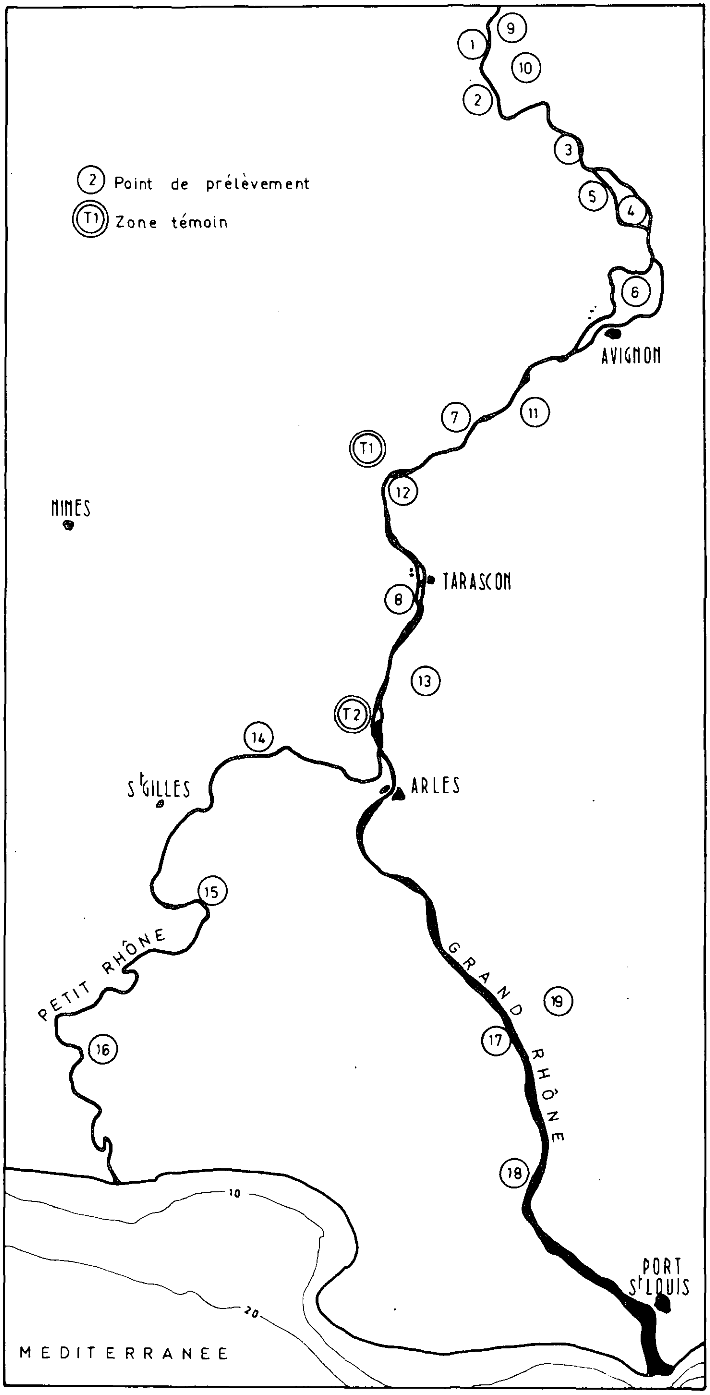

FIG. 2. Campagne du Rhône, prélèvements de végétaux.

TABLEAU I. NIVEAU D'ACTIVITE DES VEGETAUX
Activité bêta totale (pCi/kg frais)

N° du Prélèvement	Distance de Marcoule (km)	Pommes	Pêches	Raisins	Tomates	Salades	Choux	Epinards	Asperges
1	2	1100	4200	1900		2600			2200
2	6	1400	1400	1400	1900	4500			2100
3	10	990	1800	1400				5200	2100
4	15	1050	2200	1900				5500	
5	13	1250	2200	1400	2000	8500	3600	4300	2300
6	20	1000	2400	1200	2100	3600	3900	5600	2300
7	27	1250	2300	1000	2000	2300	3100	6300	2200
8	39	1000	2400	1200	2300		4500	5000	2200
9	0	1500	2100	1700	2600		6000	3300	2300
10	4	950		1800	2000		5200	4600	2200
11	27	1150	2100	1200	2300	2250	5300	5000	2600
12	32	1200	2250	1250	1900	2300	3600	10700	
13	44	1200	1920	1600		5900	3800	5000	
14	50	800	2200	1800			3000	3200	2200
15	60	800		1250	2200				2200
16	73	1100	1800	1850	2100				1950
17	67	1300	2600	1400	2500			5900	1900
18	76	1200	1700	1400	2100	6400			1300
19	65	1150	1300	1750	1800	5600	2000	8600	2050
T1		800	2300	1700			3400	6000	2200
T2		1000		1150	1500	5200	4000	3900	2200

Aucune différence significative entre les activités des différentes espèces ne peut être mise en évidence.

Par la méthode des rangs appliquée aux activités bêta totales de cinq espèces végétales bien représentées sur l'ensemble du site: pommiers, pêchers, vignes, tomates, asperges, on constate que les moyennes des rangs des activités des lieux de prélèvement ne sont pas significativement

TABLEAU II. NIVEAU D'ACTIVITE DES VEGETAUX
Césium-137 (pCi/kg frais)

N° du Prélèvement	Distance de Marcoule (km)	Pommes	Pêches	Raisins	Tomates	Salades	Choux	Epinards	Asperges
1	2		237			150			
2	6								
3	10	194	248	1100					
4	15		243						
5	13	54							
6	20	240	140						
7	27								
8	39	345		190					
9	0		64						
10	4	71			189				
11	27	69					83		
12	32							11	
13	44						145		
14	50	137	188		15794				
15	60								
16	73				68				
17	67	129	104		129				
18	76	2,333	85	103	136				
19	65	257	69		302				
T1									
T2								47	

différentes. Les niveaux des activités sont indépendants des situations géographiques; néanmoins aux abords immédiats d'une installation (usine de retraitement de Marcoule) l'activité des produits récoltés est légèrement plus élevée. A quelques kilomètres en aval, les activités redeviennent comparables à celles obtenues tout le long du Rhône.

La teneur en césium-137 (tableau II) d'une bonne partie des échantillons est non décelable; deux points cependant présentent une activité relativement élevée. Exception faite de ces deux points la moyenne des activités des échantillons contaminés est 140 pCi/kg frais de denrée non lavée. Cette activité, faible par rapport à la CMA de l'eau, est cependant plus élevée que celle que l'on peut noter dans les relevés de surveillance des retombées et peut être attribuée aux rejets de produits radioactifs dans le Rhône.

L'activité calculée de l'eau du Rhône au moment des rejets est au maximum de 7,3 pCi/l en césium-137. La contamination de 1 kg de denrée contaminée non lavée est donc équivalente à celle de 20 litres d'eau environ. Ces valeurs sont nettement plus élevées que celles calculées sur modèles expérimentaux.

Le tableau II montre que ce sont les espèces normalement irriguées par aspersion qui sont contaminées par le césium-137: pommes, pêches, tomates.

La répartition géographique peut donner une explication: les points apparemment les plus contaminés sont situés en Camargue, dans la partie aval du cours où la vitesse du courant est ralentie et où l'activité des sédiments en césium-137 est plus élevée; le pompage direct de l'eau sans l'intermède de sédimentation dans le canal d'irrigation a pour conséquence la mise à disposition d'une eau plus contaminée que la moyenne calculée.

En résumé, les activités sont faibles, très éloignées des CMA de l'eau. La contamination en strontium-90 n'est pas significativement différente de celle due aux retombées. Une très légère contamination en césium-137 est attribuable aux rejets radioactifs.

Il en résulte une charge polluante trop faible pour provoquer un risque de contamination du consommateur par la ration alimentaire.

4. COMMENTAIRES

Comme le laissaient prévoir les études expérimentales, le Bassin a bien «accepté» les rejets effectués par l'ensemble des installations nucléaires. Les phénomènes de dilution et de dispersion l'ont emporté sur les risques de reconcentration des radionucléides.

Des traces de reconcentration existent tout de même au niveau du milieu vivant aquatique, au niveau de zones à sédimentation active et sur certains sols, mais les valeurs obtenues n'ont jamais atteint et de loin les seuils critiques. De plus, en ce qui concerne la production végétale, le produit effectivement consommé correspond souvent à un des organes les moins contaminés de la plante (fruits).

La comparaison des QMA de strontium-90 et de césium-137 avec les concentrations obtenues par l'expérimentation ou mesurées in situ montre que pour un consommateur absorbant quotidiennement 1 kg de poisson et 1 kg de légumes et de fruits (provenant des zones influencées par les rejets) les rapports sont les suivants:

	QMA	Quantité ingérée
^{90}Sr	2 200 pCi/j	32 pCi/j
^{137}Cs	440 000 pCi/j	300 pCi/j

Ainsi, dans les conditions les plus critiques et difficilement réalisables, le facteur de sécurité reste de 100 pour le strontium et de 1 000 pour le césium.

Ce bilan optimiste concerne le passé; en effet, deux facteurs nouveaux vont modifier cette «acceptation» par le milieu des rejets de radionucléides:
- d'abord l'augmentation des activités nucléaires dans le Bassin;
- la construction, en cours ou projetée, de plusieurs centrales électronucléaires de type à eau sous pression et surrégénérateur;
- mais surtout le remodelage du fleuve par la création de plusieurs barrages, notamment dans le tiers aval de son parcours.

De ce fait les installations nucléaires actuelles et futures vont être amenées à effectuer leurs rejets dans des eaux tranquillisées, les phénomènes de dilution seront atténués et les échanges avec les sédiments constitutifs du lit seront plus importants. En conséquence, la portion la plus aval, l'estuaire et la mer, ne joueront plus qu'un faible rôle dans la dispersion des éléments radioactifs.

La notion de «capacité radiologique» prend ici toute sa valeur. Cette capacité peut évoluer avec le temps, dans un sens aussi bien que dans l'autre d'ailleurs.

Les données obtenues par l'expérimentation et les mesures et observations in situ permettent de déterminer les facteurs limitants d'un bassin. Le choix du site, le choix des programmes de surveillance devraient donc dans une certaine mesure s'appuyer sur l'étude de Bassin pour apporter aux populations riveraines du fleuve des garanties sur la qualité des eaux et des produits irrigués.

5. CONCLUSION

A la suite des travaux dont nous venons de parler, nous sommes persuadés que dans le domaine de la sauvegarde de l'environnement, il convient de ne pas se borner à un examen de la situation à un instant donné mais de suivre l'évolution du milieu avec le temps.

Cette recommandation trouve une justification supplémentaire à la suite d'observations faites récemment par notre équipe dans le Bassin rhodanien et sur d'autres cours d'eau: l'effet de synergie entre pollution radioactive, pollution thermique et pollution chimique. Les études expérimentales entreprises il y a près de dix ans concernaient un Bassin où la pollution chimique, bien que présente, était relativement faible et la pollution thermique inexistante, mais depuis la qualité des eaux s'est dégradée.

Concurremment à cette situation nouvelle nous avons observé au niveau du cours d'eau une modification du comportement des ions radioactifs, par exemple une plus grande mobilité ayant pour origine la formation de complexes à partir des rejets industriels effectués en amont. Cette mobilité provoque un transfert des radionucléides à longue distance dans le cours du fleuve, dans les canaux d'irrigation et dans les nappes. Certains éléments tels que le cobalt se déplacent dans le sol avec le front de l'eau. Le risque de pollution des nappes est à prévoir.

La pollution thermique favorise au contraire les phénomènes de sédimentation donc de dépôts dans le lit des fleuves et dans les barrages. Au niveau du milieu vivant, l'accroissement même faible de la température des eaux provoque une activité métabolique plus importante avec accroissement du taux de contamination des espèces aquatiques. Les cultures sont concernées à la fois par la pollution chimique qui favorise le transfert par l'eau d'irrigation et la pollution thermique qui accélère le processus métabolique. Cette forme de pollution fait l'objet de recherches au profit de l'agriculture, avec comme conséquence l'atténuation de la charge thermique des cours d'eau.

L'étude du Bassin rhodanien à travers quinze ans d'activité nucléaire nous a apporté de nombreuses informations sur les mécanismes de transfert des radionucléides.

La possibilité que nous avons eue d'associer des études expérimentales et des mesures in situ nous a permis d'observer comment un milieu hétérogène était concerné par les rejets et quelles étaient les voies préférentielles de contamination.

Grâce à une politique de rejet conforme aux prescriptions, le milieu a bien accepté ces rejets et si, en tant que radioécologistes, nous devons reconnaître que le Bassin est marqué par les rejets radioactifs au niveau de la plupart de ses constituants, les concentrations observées n'ont jamais atteint des seuils critiques.

Mais, à travers cette conclusion optimiste pour le passé, nous donnons en fait un avertissement pour l'avenir. Le Bassin est en voie de transformation, les implantations nucléaires vont augmenter, les pollutions chimiques et thermiques suivront vraisemblablement cet accroissement, c'est pourquoi l'observation du Bassin doit continuer et l'implantation de nouveaux sites nucléaires doit tenir compte de son évolution.

L'intégration des implantations actuelles dans l'environnement a été réussie, il serait regrettable pour l'avenir de ne pas tenir compte de l'expérience acquise et en particulier de concevoir une implantation nouvelle de site nucléaire sans l'intégrer radioécologiquement dans le Bassin. Les conclusions générales tirées de l'étude du Bassin rhodanien nous paraissent extrapolables aux autres bassins fluviaux.

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DISCUSSION

J. P. MORONI: As a representative of the French public health authorities, I should like to make two comments on the papers so far presented at the Symposium, particularly at the present session. In the first place, tribute should be paid to the high quality of the studies — both experimental and theoretical — which have been devoted to evaluating the impact of nuclear power stations on the environment; apart from some uncertainties on points of detail, the solidity of the resulting conclusions is beyond doubt.

Secondly, it should be noted that the contamination found — at very low levels and in limited areas — is confined to the environment and to certain links in the food chain: no significant effect on man has been detected.

E. KUNZ: What portion of the activity measured or evaluated in your study can be assigned to global radioactive fall-out?

A. GRAUBY: As regards plants, the ^{90}Sr due to fall-out masks any contamination from discharges by nuclear power facilities. In the case of Cs, the fraction represented by discharges is greater than that due to fallout. For each compartment of the environment (sediments, soils, etc.) we find similar phenomena in respect of each radionuclide, with the discharge fraction being more or less masked by the fall-out.

E. KUNZ: Are there any advantages of what one might term a 'non-radiological' nature likely to emerge from your most valuable study?

A. GRAUBY: Yes, I think one can say that such is in fact the case: some of our results have already been used for establishing the transfer mechanisms of cations in the mineral nutrition of plants.

B.K. BORISOV: What is the method of preparing the fish samples for analysis?

A. GRAUBY: After the sample has been taken the radioactivity of the whole fish or of individual organs thereof can be measured with the help of traditional methods of dissection. Each organ (or the whole fish) is weighed fresh. It is then oven-dried at 110°C for 24 hours and, after crushing, the gamma activity is measured in a well-type crystal.

In the case of elements for which it is necessary to work under thin-layer conditions, the organs are incinerated in a programmed furnace until white ash is obtained.

EVALUATION OF RADIOACTIVE AND NON-RADIOACTIVE TRACE CONSTITUENTS EMITTED FROM FOSSIL-FUEL AND NUCLEAR POWER PLANTS

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Abstract

EVALUATION OF RADIOACTIVE AND NON-RADIOACTIVE TRACE CONSTITUENTS EMITTED FROM FOSSIL-FUEL AND NUCLEAR POWER PLANTS.

On the basis of a simplified box model the relative body burden function S = K/MPC for the pollutants released in major quantities from fossil-fuel and nuclear power plants has been calculated and compared. Besides these main pollutants (SO_2, NO_2, ^{133}Xe, ^{85}Kr), other substances are released but in smaller quantities. Since some of these substances (for example: Ra, Hg, ^{3}H etc.) are toxicologically important, the environmental effects are not negligible.

Using a further developed box model that takes into consideration the vertical distribution of pollutants and the behaviour of the pollutants in the atmosphere body burden functions of high toxic trace constituents released from coal, oil and gas-fired and nuclear power plants are calculated. The trace constituents are: radium, fluor, cadmium, mercury from fossil-fuel power plants and ^{131m}Xe, ^{3}H and ^{131}I from nuclear power plants. The body burden functions are compared with those of the major pollutants. It is found that the environmental effects of radium from coal-fired power plants are small compared with the environmental effects of radioactive materials released from nuclear stations. Other trace pollutants, for example fluor compounds, provide a larger contribution to the integral body burden. The relative importance of the various trace constituents released from power stations is discussed.

1. INTRODUCTION

The production of electrical energy is always bound up with the emission of several pollutants into the environment. The most important burden to the environment from the operation of power plants is atmospheric pollution. In comparing the environmental burden from air pollutants of the various types of power plants up to now only the materials released in major quantities from fossil-fuel and nuclear power plants have been considered [1]. Besides these main pollutants, which are SO_2, NO_x, F and particles for fossil-fuel power plants and ^{133}Xe and ^{85}Kr for nuclear power plants, other substances are released in smaller quantities. The emission rate of these trace pollutants alone cannot be the basis for comparison, as has been shown in Ref. [1], since the body burden depends on the toxicity of the substance released. A number of these trace substances are relatively highly toxic.

In continuation of the studies on the comparison of air pollution from fossil-fuel and nuclear power plants an improved box model [2] has been used to calculate the relative body burden, S = K/MPC, where K is the average concentration of the pollutant and MPC the corresponding maximum permissible concentration for the public.

This paper deals with the more important trace pollutants released to the atmosphere from power plants. The significance of the various trace pollutants from coal, oil and nuclear power plants is discussed by means of the relative body burden S of these trace pollutants.

2. DESCRIPTION OF THE MODEL

To calculate the relative body.burden S the following model was used, which was first published in Ref. [1] and in its improved version in Ref. [2].

All pollutants released into the atmosphere have a certain residence time there. The mass balance follows Eq.(1):

$$dM = F dt - R dt \qquad (1)$$

where M is the amount, F the emission rate and R the removal rate of the pollutant considered. The average concentration K in a certain volume V can be calculated if we know F and R. It is

$$K = \frac{M}{V} \qquad (2)$$

The relative body burden S is then the ratio between K and the corresponding maximum permissible concentration MPC for the public, that is

$$S = \frac{K}{MPC} \qquad (3)$$

For purposes of comparison the F values (emission rate) were calculated on the basis that the total electrical energy produced in 1970 in the Federal Republic of Germany (FRG) of 228 TW · h was produced by the one power plant type considered alone, i.e. by coal-fired, oil-fired or nuclear-fuelled LWR power plants respectively. For the removal rate an exponential decay of the atmospheric pollutants was assumed according to

$$R = \lambda M \qquad (4)$$

where $\lambda = 1/\tau = \ln 2/T_H$ with τ = residence time and T_H = half-time of the pollutants. The volume V of the box consisted of the area of the FRG (250 000 km^2) and a height of 3 km. Since the vertical distribution of the various pollutants in the lower troposphere will not be the same, the relative concentrations of the pollutants in the first 100 m above the ground derived from the concentration profiles given in Eq.(2) were used.

If the dimensions of F are t/yr or Ci/yr and the dimensions of the corresponding MPC values are $\mu g/m^3$ or pCi/m^3, the relative body burden S can be calculated from Eq.(5):

$$S = \frac{F \cdot \tau \cdot \alpha}{MPC}\, 1.2 \times 10^{-4} \qquad (5)$$

where α gives the percentage of the amount M of the pollutant existing in the first 100 metres above the ground. In cases where the vertical distribution of the pollutants is not known, α stands for the homogeneously mixed box of limited height, as explained in detail in Section 3.

3. DATA

3.1. Trace pollutants from hard-coal-fired power plants

We restricted our calculations to the emission of trace substances from hard-coal-fired plants because the content of trace constituents in lignite (brown coal) is small compared with that of hard coal [3].

Most of the trace substances in coal and oil we are concerned with are metals or metal oxides. They are released from the stack in the form of particulate matter. In Germany the emission of particulate matter is restricted by law for newly built power plants. The mean emission rate of dust from German coal-fired power plants was estimated by Peters et al. [4] to be 500 mg/m^3 smoke-gas. The mean ash content of trace metals in German hard coals was determined by Otte [3]. We assume that the trace constituents are released from the stack in the same quantities and percentages as fly ash. The total emission rates for the FRG are calculated with the assumption that modern coal power plants release into the atmosphere 4 m^3 smoke gas per kW · h.

As reported by Eisenbud and Petrow [5] and Kellermann [6], decay products of the uranium-238 and thorium-232 series are found in coal.

TABLE I. BODY BURDEN OF TRACE SUBSTANCES RELEASED INTO THE ATMOSPHERE FROM HARD COAL-FIRED POWER PLANTS

Trace substance	Mean ash content	Residence time (d)	Emission rate	Standard	Body burden S
$^{226+228}$Ra	6.2 pCi/g	5	2.83 Ci/yr	1 pCi/m^3	3.5×10^{-4}
^{228}Th	2.6 pCi/g	5	1.2 Ci/yr	0.2 pCi/m^3	7.5×10^{-4}
F[a]	5×10^{-3}%[b]	5	1.2×10^{4} t/yr	10 μg/m^3	0.15
F[a]	5×10^{-3}%[b]	0.04	1.2×10^{4} t/yr	10 μg/m^3	6.0×10^{-4}
Pb	0.3%	5	1.4×10^{4} t/yr	2 μg/m^3	0.85
Zn	0.8%	5	3.7×10^{4} t/yr	500 μg/m^3	0.001
V	1.1%	5	5.0×10^{4} t/yr	5 μg/m^3	1.2
Ni[a]	1.6%	5	7.4×10^{4} t/yr	1 μg/m^3	9.1
Ni[a]	1.6%	5	7.4×10^{4} t/yr	0.1 μg/m^3	91
Mn	2.2%	5	10×10^{4} t/yr	250 μg/m^3	0.005
Cu	0.4%	5	1.8×10^{4} t/yr	5 μg/m^3	0.44
Cd	n.a.	5	n.a.	5 μg/m^3	-
Co	0.2%	5	0.9×10^{4} t/yr	25 μg/m^3	0.04
Sn	0.6%	5	2.8×10^{4} t/yr	100 μg/m^3	0.03
Hg	10^{-4}%[b]	5	72 t/yr	1 μg/m^3	0.010

[a] Explained in text.
[b] Content in coal.
n.a. not available.

Eisenbud determined the radium content of ash to be 6.2 pCi/g and that of thorium to be 2.6 pCi/g.

The amount of mercury in coal is not well known. Joensuu [7] analysed 36 American coals and estimated more conservatively the content of mercury in coal to be 1 ppm. The emission of fluor compounds was calculated from the values given by Brocke [8]. The mean ash content of different trace substances in coal is listed in column 2 of Table I. In 1970 228 TW · h electrical energy were produced in the FRG. In column 4 of Table I the total emission of trace substances per year by coal-fired power plants is listed under the assumption that the 228 TW · h are produced only by coal-fuelled power plants.

Pollutants are removed from the atmosphere by decay processes, chemical interaction and scavenging processes. For calculating the relative body burden the residence time in the atmosphere of the various trace substances has to be known. Most of the trace substances listed in column 1 of Table I are metals and emitted from power plants as fly ash. Residence times of natural aerosols in the troposphere have been observed by several authors and obtained from data on atmospheric radioactivity. The results of these measurements are discussed in detail by Martell [9]. The residence time of particulates up to a height of 5000 metres was reported by Martell to be 3 to 7 days. For our calculations we used a mean residence time of particulates in air of five days, as listed in column 3 of Table I.

The chemical behaviour of fluor in the atmosphere is not well known. Atmospheric fluor compounds are very reactive and might attach fast to aerosols. In this case they have a residence time similar to that of particulate matter. Fluor might also react with gaseous carbon hydrates or anorganic compounds to an inert compound. In this case the residence time of fluor would be very short, probably not longer than one hour. We calculated the body burden for both residence times. $^{226/228}$Ra and ^{228}Th containing fly ash will have the residence time of suspended particles in air.

Atmospheric trace substances are usually not uniformly distributed in the troposphere. As a result of meteorological conditions and relative short residence times through chemical or physical processes they have a nearly exponential profile with altitude with the highest concentration near the ground. We have already discussed profiles for different pollutants in a previous paper [2]. Because of the different stack height, we assumed a constant concentration of pollutants up to 100 metres. The portion of particulate matter in the first 100 metres is about 20% of the total emission. For gaseous fluor compounds (F^{++}) an atmospheric residence time is not known and we therefore assumed a homogeneous mixing in a box up to 1000 metres above the ground.

To compare the effect on body burden by the various trace substances from different power plants air quality criteria are used. Air quality standards for the main pollutants have been published for the FRG as basic standard concentrations (MIK values) [10]. Basic standards have not yet been established for most of the trace substances. The Maximum Workroom Concentrations (Maximale Arbeitsplatzkonzentration, MAK values) are well known [11]. Basic standard concentrations are usually one twentieth of the threshold limit values for workroom concentrations. We defined therefore the standards for trace substances as 1/20th of the MAK values. The basic standard for lead and its compounds is under discussion and recommended by Knelson and Bridbord [12] as 2 $\mu g/m^3$. Nickel is known to cause allergic reactions on the skin and to be carcinogenic to man. No threshold value for workroom atmospheres has yet been established. We therefore calculated the body burden for two standard values.

The standard value for mercury has been recommended by Foote [13].

For radioactive trace substances we used one tenth of the Maximum Permissible Concentrations (MPC) as published in the amendments to the German Atomic Law [14]. The standards used for our calculations are listed in column 5 of Table I.

3.2. Trace pollutants from oil-fired power plants

The content of metallic trace substances in oil varies considerably and depends on the origin of the oil. The origin of the heating oil used in the FRG is Africa and Arabia. Column 2 of Table II lists the mean content of trace substances in heavy heating oils.

For purposes of comparison of the emission rates the trace pollutants from the various power plants have again been calculated on the assumption that the total amount of electric energy consumed in the FRG is produced by oil-fired power plants. The emission rates are listed in column 4 of Table II.

The radioactivity in oil has been reported by Eisenbud and Petrow [5] and the cadmium content of heating oil by Lagerwerff and Specht [15].

Residence times, standards and distribution of the pollutants in the atmosphere have already been discussed in section 3.1. For the pollutants fluor and nickel in the atmosphere we made the same assumptions as mentioned there for coal-fired power plants.

TABLE II. BODY BURDEN OF TRACE SUBSTANCES RELEASED INTO THE ATMOSPHERE FROM OIL-FIRED POWER PLANTS

Trace substance	Mean content in fuel	Residence time (d)	Emission rate	Standard	Body burden S
$^{226+228}$Ra	3.5×10^{-4} pCi/g	5	0.018 Ci/yr	1 pCi/m^3	1.1×10^{-5}
^{228}Th	3.3×10^{-4} pCi/g	5	0.018 Ci/yr	0.2 pCi/m^3	5.5×10^{-5}
F[a]	18 ppm	5	1000 t/yr	10 μg/m^3	1.23×10^{-1}
F[a]	18 ppm	0.04	1000 t/yr	10 μg/m^3	5.0×10^{-5}
Pb	0.4 ppm	5	20 t/yr	2 μg/m^3	1.23×10^{-3}
Zn	0.2 ppm	5	10 t/yr	500 μg/m^3	2.4×10^{-6}
V	10 ppm	5	500 t/yr	5 μg/m^3	1.23×10^{-2}
Ni[a]	6 ppm	5	300 t/yr	1 μg/m^3	3.69×10^{-2}
Ni[a]	6 ppm	5	300 t/yr	0.1 μg/m^3	3.69×10^{-1}
Mn	2 ppm	5	100 t/yr	250 μg/m^3	4.92×10^{-4}
Cu	0.2 ppm	5	10 t/yr	5 μg/m^3	2.46×10^{-4}
Cd	0.5 ppm	5	25 t/yr	5 μg/m^3	6.16×10^{-4}

[a] Explained in text.

TABLE III. BODY BURDEN OF RADIOACTIVE TRACE SUBSTANCES RELEASED INTO THE ATMOSPHERE FROM NUCLEAR POWER STATIONS

Trace substance	Residence time (d)	Emission rate, Yankee type (Ci/yr)	Emission rate, Dresden type (Ci/yr)	Estimated emission rate, German LWR (Ci/yr)	1/10 MPC (pCi/m^3)	S Yankee type	S Dresden type	S German LWR
^{131m}Xe	17	n.a.	n.a.	3.6×10^4	4×10^{-5}	-	-	1.8×10^{-6}
^{135}Xe	0.57	3.7×10^1	2.9×10^6	n.a.	1×10^5	2.5×10^{-9}	2.0×10^{-4}	-
^3H	10	2.4×10^3	1.9×10^1	9.5×10^3	2×10^5	1.4×10^{-6}	1.1×10^{-8}	5.7×10^{-6}
$^{131}I^a$	11.6	5.6×10^{-2}	1.4	9.5×10^1	2×10^2	3.9×10^{-9}	1.0×10^{-7}	7.0×10^{-6}
$^{131}I^a$	5	5.6×10^{-2}	1.4	9.5×10^1	2×10^2	3.4×10^{-8}	8.0×10^{-7}	6.0×10^{-4}
^{90}Sr	5	3.7×10^{-2}	0.9×10^{-2}	n.a.	1×10^1	4.5×10^{-5}	1.1×10^{-5}	.

^{a} Explained in text.

n.a. not available.

3.3. Radioactive trace pollutants released from nuclear power plants

Besides the main pollutants ^{85}Kr and ^{133}Xe, nuclear power stations release to the atmosphere several radioactive gases and particulates. Some of these are listed in column 1 of Table III. The emission rates of the various power stations depend on the type of reactor used. For our calculation of the body burden we used the emission rates of the boiling water nuclear power reactor Dresden [16], those of the advanced pressurized water nuclear power reactor Yankee [17] and the estimated emission rates for German water reactors given by Schikarski [18]. Again we assume that the total consumed electric energy of 228 TW · h is produced alternatively by one of the three different reactor types.

The residence time of radioactive substances is related to the half-life time by Eq. (4).

Tritium is released from the stack mostly as HTO, otherwise HT would react very fast with oxygen to water. The residence time of radioactive tritium in the atmosphere is limited by the residence time of water in the troposphere. Junge [19] estimates the residence time of water in the air to be 10 days.

Iodine-131 is released from nuclear power plants both as methyliodide and as molecular iodine. The behaviour of the two types of iodine in the atmosphere is not well known. We calculated two alternatives: (a) iodine is uniformly mixed throughout the whole troposphere and its atmospheric residence time is limited by the radioactive half-life; (b) we assume a fast attachment of iodine to atmospheric aerosols and therefore a residence time of 5 days. It should be noted that an assumption of the grass-cow-milk pathway would increase the S-values for ^{131}I considerably, relative to the standards. In this respect ^{131}I would be a significant trace pollutant for atmospheric pollution from LWR power plants.

For the relatively long-lived nuclide ^{131m}Xe we assume uniform mixing throughout the troposphere.

The vertical distribution of water vapour is reported by Junge [19]. About 10% of the water vapour distributed up to a height of 3000 m is in the first 100 m. The radiological residence time of ^{135}Xe is so short that we assume a mixing height of 1000 m. The vertical distribution of aerosols has already been discussed in section 3.1.

We again defined the air quality standards for radioactive pollutants as one tenth of the MPC, as published in the amendments to the German Atomic Law [14].

TABLE IV. INTEGRAL S-VALUES

Plant	Sum of body burden S	
	Main pollutants	Trace substances
Coal	2.01	2.73
Oil	1.69	0.64
LWR	0.002	< 0.001

Since the half-lives of ^{131m}Xe, ^{135}Xe and ^{131}I are short, compared with the in-pile times and the time between deloading and reprocessing emissions of these nuclides from reprocessing plants are negligible. Of the long-lived radionuclides in the scope of this paper only ^{3}H is of interest. With the present technology the release of ^{3}H to the atmosphere from small reprocessing plants is of the order of 50 Ci ^{3}H per ton of reprocessed fuel (UO_2) [18]. Large advanced reprocessing plants, which will be necessary to reprocess the spent fuel corresponding to the production of 228 TW · h nuclear energy, will have the necessary hold-up system for ^{3}H.

4. RESULTS

For calculating the body burden we used the model described in section 2, assuming an exponential decay of the pollutants. The ratio of the calculated concentration and the basic standard we call 'body burden'. These body burden values are listed in column 6 of Tables I, II and III for the three different types of power plant.

These figures may be off considerably from reality for specific sites since our model does not take into account local conditions, weather, distribution of emitters etc. However, it is interesting to compare these figures for different trace substances and for different types of power stations.

Among the trace substances released from coal-fired power stations nickel, lead, copper, vanadium and fluor are of major importance for the total body burden. Because of the carcinogenic effect of nickel aerosols and the high nickel content of coal, the body burden of nickel is the highest, even if a relatively high standard is assumed. The values for lead, vanadium and copper are high and their contribution to the total body burden is only one order of magnitude lower than that of the main pollutants such as SO_2 etc. It should be mentioned that recent measurements in the USA [20] show that in particular lead, vanadium and, to a certain extent, also nickel are present in polluted atmospheres at high concentrations in predominantly submicron aerosols, which can deposit easily in the pulmonary compartment of the respiratory system. The radioactive emission from coal-fired power plants is negligible. Fluor only offers a major contribution to the total body burden for an assumed residence time of 5 days.

For all trace substances released from oil-fired power plants the emission rates and body burdens are at least one order of magnitude lower than those for coal-fired plants. Here also nickel has the highest S-value. Fluor emissions are of interest only when assuming a long residence time in the atmosphere. Iodine has the highest S-value of the listed radioactive trace substances released into the atmosphere from German nuclear power stations.

It is of great interest to compare the body burden values of the trace substances with the S-values of the major pollutants released from power plants. In Table IV the S-values for the pollutants SO_2, fly ash and NO_x, as reported by Jansen et al. [2], are summed up for the specific plant type in column 2. The sum of the body burden values of the trace substances according to Tables I to III without nickel are listed in column 3. Because of the high emission rate and toxicity of nickel it is necessary to establish a basic standard for this pollutant.

It should be noted that the simple addition of S-values may not be correct because the biological effects of several pollutants existing in a polluted area may be synergistic rather than simply additive.

From Table IV it can be concluded that especially for coal-fired power plants the biological body burden of trace substances released to the atmosphere is as important as that of the main pollutants. For oil-fired plants the trace substances have a smaller but not negligible body burden effect. Solely the body burden of trace substances released from nuclear power plants do not contribute to the total LWR S-value. Here ^{85}Kr and ^{133}Xe are the main pollutants.

SUMMARY

Simple box models have the advantage of making relatively rough predictions of air potentials for regional planning with only little data. Within the scope of this model air pollution potentials for the release of trace substance from coal, oil and nuclear-fuelled power stations have been calculated. The following conclusions can be drawn:

1. Trace substances released from fossil-fuelled power plants are not negligible compared to the main pollutants SO_2, NO_x and inert particulate matter.
2. The highest contribution of trace pollutants to the atmospheric burden comes from nickel, lead and vanadium.
3. In contrast to other publications, mercury, radium and thorium are of minor importance to the relative body burden.
4. Since the contribution of Ni, Pb and V to the atmospheric burden is not negligible compared to SO_2, the reduction of sulphur in fuel or in off-gas should be optimized, taking into account these trace pollutants.
5. Trace pollutants emitted from LWR nuclear power plants contribute much less to the atmospheric burden than do the main pollutants ^{85}Kr and ^{133}Xe. Only ^{131}I makes a certain contribution.
6. It is necessary to establish Maximum Permissible Concentrations for the public for several trace pollutants with high body burdens (for instance Ni, V, Pb) taking into account synergistic effects.
7. The behaviour and fate in the atmosphere of these pollutants should be investigated more thoroughly.

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DISCUSSION

C.A. MAWSON: I think M. Eisenbud has claimed that the radiation dose received from radium emitted by coal- and oil-fired plants is greater than that from emissions by nuclear plants. You have said that radium emissions from conventional plants are negligible: do you disagree with Eisenbud, or do you mean that the hazard from all the other components in conventional stack wastes outweighs the hazard from the radium?

W.O. SCHIKARSKI: Eisenbud made the comparison between coal and nuclear plants on the basis of emissions versus maximum permissible concentrations (E/MPC). However, this neglects the fact that the pollutants (i.e. ^{226}Ra etc. in the case of coal-fired stations and ^{85}Kr and ^{133}Xe in that of LWRs) have widely differing atmospheric residence times. Radium would be released as an aerosol (residence time ~ 5 days), whereas ^{85}Kr, as a noble gas, has a residence time of roughly 15 years. Taking into account this time-dependent factor of residence in the atmosphere, the ambient body burden of radioactivity from fossil-fuel plants is negligible compared to the chemical pollutants and is much smaller than that from an LWR plant of equivalent size.

R.J. GARNER: In making this kind of comparison, is it fair to neglect the contribution from nuclear fuel reprocessing plants?

W.O. SCHIKARSKI: The contribution of reprocessing plants is small in the context of a large nuclear energy system producing 228 TW·h/yr, which was the assumption in the model. Except for ^{3}H, all other trace constituents are negligible. However, the ^{3}H emissions of a large reprocessing plant to operate in the Federal Republic of Germany cannot be linearly extrapolated, since by the time the plant is needed (perhaps in 1985), the ^{3}H removal system then in use will have reduced ^{3}H emissions considerably. This is admittedly an optimistic viewpoint. At all events, the paper is focussed on trace constituents and not on ^{3}H.

R. J. ROUX: Among the components of smoke you have not mentioned uranium. Is this because the coal used in Germany contains none? Assuming that uranium is present, what is the percentage contained in the smoke in relation to that contained in the coal?

W.O. SCHIKARSKI: The contribution of U is relatively small compared with that of Ra. Since U and Ra are associated in a radioactive decay chain, it is easy to calculate the percentages of each. As I said, that of U is very small compared with Ra and Th.

STUDIES OF DOSE PATHWAYS FROM A NUCLEAR FUEL REPROCESSING PLANT

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Abstract

STUDIES OF DOSE PATHWAYS FROM A NUCLEAR FUEL REPROCESSING PLANT.

Nuclear Fuel Services operates the United States' first commercial nuclear fuel reprocessing plant in western New York State. Studies were carried out by the US Environmental Protection Agency to characterize the waste discharges from the plant and to determine the radiation doses to the population living in the vicinity of the plant. Included as a part of these studies was an evaluation of the radiation doses to the exposed population from ingestion of fish and deer during 1971. The report presents the procedures and data used in evaluating these dose pathways.

Dose estimates for the fish and deer pathways indicated that the integrated whole-body population dose from each of these pathways was about 0.1 man-rem during 1971. The maximum whole-body dose an individual could have received from ingestion of venison was 14 mrem. The maximum dose to an individual from the fish pathway was estimated to be 1.4 mrem whole-body and 7 mrem to bone. The data on the fish pathway indicated that the external whole-body doses received while fishing were greater than the doses received from ingestion of fish.

1. INTRODUCTION

Nuclear Fuel Services began operation of the first commercial nuclear fuel reprocessing plant in the United States of America in April 1966. This plant is located on a 13-km^2 acre site in western New York State. This site is located 48 km southeast of Buffalo, New York and about 6 km south of Springville, New York, a village with a population of about 4000. The plant employs the Purex process and the original design capacity was one metric ton of uranium per day. From start-up to December 1971 the plant processed 625 metric tons of uranium with an exposure of 4220 GW·d. The plant ceased processing fuel in December 1971 and is currently undergoing changes that will increase the capacity to 2.5 t/d.

The operation of this plant resulted in the discharge of some radioactive waste to the surrounding environment. Gaseous activity was discharged to the environment through a 60-m stack with the plant located 1.2 km from the nearest site boundary. Low-level liquid waste was discharged to a series of three lagoons, which provided a hold-up period of 30-60 days prior to discharge of the waste to the environment to allow for precipitation and sedimentation of radionuclides. The waste from the lagoons was then discharged to a series of small on-site creeks that empty into Cattaraugus Creek, an unrestricted stream with an average flowrate of 11 m^3/s. The Cattaraugus Creek eventually empties into Lake Erie approximately 64 km from the plant site. Figure 1 is a diagram showing the lagoon and stream system into which the low-level waste was discharged.

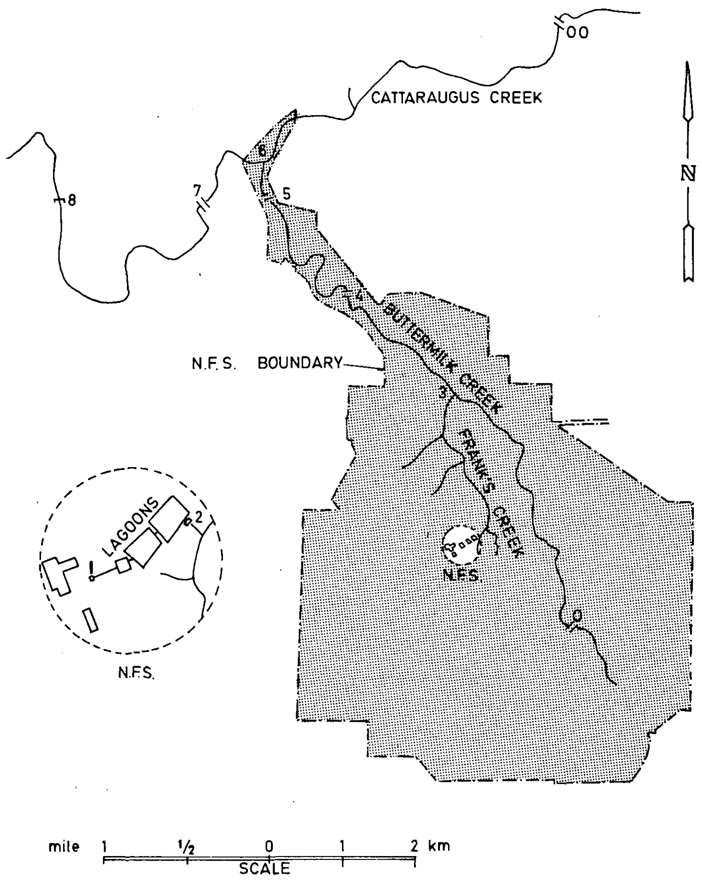

FIG. 1. Stream system into which NFS waste was discharged.

The US Environmental Protection Agency has carried out studies to characterize the discharge of radioactivity from this plant and to estimate the radiation doses to the population living in the vicinity of the plant [1-5]. Preliminary estimates indicated that the ingestion of fish and deer represented two of the more significant dose pathways resulting from the discharge of radioactive waste from the plant. To better define the radiation exposures to the population from these pathways, the US Environmental Protection Agency, in co-operation with New York State, conducted a detailed survey during 1971 to determine fish and venison intakes by the population living in the vicinity of Nuclear Fuel Services. This report presents:

(a) A description of the survey procedures used to determine fish and venison intakes
(b) The results obtained from the survey

(c) Dose estimates for the maximum individual and a man-rem dose for the local population from these pathways

(d) Data on radionuclide discharges from the plant that have resulted in these doses.

2. RADIONUCLIDE INTAKES AND DOSE ESTIMATES FOR THE POPULATION FISHING THE CATTARAUGUS CREEK

During the spring and summer of 1971 the US Environmental Protection Agency, in co-operation with the New York State Department of Environmental Conservation, conducted a fishing survey to determine intake of fish by the population fishing the regions of the Cattaraugus Creek near the NFS plant into which the liquid waste is discharged. During this same period samples of fish were collected from these regions and analysed for their radionuclide content.

2.1. Survey procedure

Each month from April to August 1971 the fishing areas between Bigelow Bridge and Otto Road were surveyed for seven consecutive days. Samples of fish previously collected from these regions of the Cattaraugus Creek had shown detectable concentrations of radionuclides, which could be identified as resulting from the NFS waste discharges. Radionuclide concentrations in fish collected from other regions of the Creek were lower than in these regions nearest the plant and could not readily be associated with the NFS discharges. For this reason, this study was limited to the areas indicated. Six significant fishing areas had been identified along the approximately 24 km of stream involved. These fishing areas had been identified by two aerial inspections to ensure that all possible fishing areas would be investigated.

Each of the identified fishing areas was visited at least five times each day between the hours of 6.00 a.m. and 9.00 p.m. Observations of the number of fishing trips per day were made and interviews were conducted with as many of the people fishing as possible. Information was obtained on the frequency of fishing, length of fishing trip, number of meals of fish eaten and species eaten. The number, size and species of fish caught at the time of the interview were also recorded. The survey data were extrapolated to cover the full five-month period of April to August 1971. Table I presents a summary of the survey data. Further details on the survey procedures and the methods of extrapolation are presented elsewhere [6].

The results of the survey show that the maximum number of fish meals eaten by an individual was 24 meals of trout per year. Three individuals interviewed indicated intakes of 16-20 meals of trout per year. One individual reported an intake of 12 meals per year of combined sucker and trout.

Using a conservative assumption of 200 grams of fish flesh per meal, the maximum intake of trout flesh per person was 4.8 kg/yr and the maximum intake of sucker flesh per person was 2.4 kg/yr.

TABLE I. SUMMARY OF DATA OBTAINED DURING FISHING SURVEY OF CATTARAUGUS CREEK APRIL - AUGUST 1971[a]

Total fishing trips for period	2000
Average length of fishing trip	3.7 h
Number of trips by individuals eating fish	1400
Number of trips by individuals eating trout only	1100
Number of trips by individuals eating trout or sucker[b]	300
Average catch of trout per trip by trout eaters	1.5
Average catch of sucker per trip by sucker eaters	5
Average measured edible weight of trout flesh[c]	100 g
Average measured edible weight of sucker flesh	45 g
Maximum intake of trout by individual	4.8 kg
Maximum intake of sucker by individual	2.4 kg
Total intake of trout flesh	210 kg
Total intake of sucker flesh	70 kg

[a] All data approximate because of the limitations resulting from assumption used in extrapolating survey data (see Ref. [6]).

[b] 95% of fish caught were trout or sucker.

[c] Average weight of edible flesh per fish.

TABLE II. AVERAGE CONCENTRATION OF RADIONUCLIDES IN FISH FROM SURVEY AREA OF CATTARAUGUS CREEK - 1971

Type	Weighted Average Concentration[a] (μCi/kg)			
	^{137}Cs	^{134}Cs	^{90}Sr	^{65}Zn
Trout	1.05×10^{-3}	1.9×10^{-4}	6.5×10^{-5}	7.8×10^{-4}
Sucker	1.14×10^{-3}	1.9×10^{-4}	3.6×10^{-4}	2.0×10^{-3}

[a] These averages were weighted according to the edible weight of flesh in each fish sample.

2.2. Radionuclide concentrations in fish from Cattaraugus Creek

Data on the concentrations of caesium-137, caesium-134, strontium-90 and zinc-65 in fish samples collected during this study from the area of the Cattaraugus Creek covered by the survey are presented in Table II. These concentrations are weighted averages according to the amount of edible flesh in each sample.

Data on the concentrations of radionuclides in fish from the Cattaraugus Creek have been reported annually by the New York State Department of

Environmental Conservation and by Nuclear Fuel Services [7, 8]. The data presented in Table II are in general agreement with these data, although the strontium-90 concentrations in fish flesh report by New York State for 1971 appear somewhat lower than had been previously reported.

From a comparison of the levels of radionuclides in fish 24 km upstream of the survey area [8] with the measured concentrations downstream of the entry of the waste discharge it appears that most of the activity present in the fish samples collected from the survey area resulted from waste discharged from Nuclear Fuel Services. The presence of caesium-134 in fish at concentrations about 20% of the caesium-137 concentrations (the same ratio as present in the liquid waste discharge) supports the above observation that the radionuclides in fish resulted from the discharge of waste from Nuclear Fuel Services.

2.3. Radionuclide intakes from ingestion of fish

Table III presents the radionuclide intakes from ingestion of fish by the population fishing the Cattaraugus Creek near the NFS plant. As indicated previously, essentially all these intakes resulted from the discharge of waste from the NFS plant. The population intakes were calculated from the data presented in Tables I and II.

TABLE III. ESTIMATED RADIONUCLIDE INTAKES FROM CONSUMPTION OF FISH FLESH FROM CATTARAUGUS CREEK

Radionuclide	Intake Maximum by individual (μCi)	 Total by population (μCi)
^{137}Cs	5.0×10^{-3}	3.0×10^{-1}
^{134}Cs	9×10^{-4}	5.3×10^{-2}
^{65}Zn	4.8×10^{-3}	3.0×10^{-1}
^{90}Sr	9×10^{-4}	3.9×10^{-2}

TABLE IV. ESTIMATED DOSES TO POPULATION FISHING THE CATTARAUGUS CREEK NEAR NFS PLANT DURING 1971

Organ	Mode	Dose Maximum individual (mrem)	 Population (man-rem)
Whole-body	Ingestion [a]	0.4	0.02
Whole-body	External	1	0.04
Bone	Ingestion [b]	7	0.3

[a] Includes contribution from ^{137}Cs, ^{134}Cs and ^{65}Zn.

[b] Dose from ^{90}Sr only.

2.4. Dose estimates

Table IV presents a summary of doses due to NFS discharges to the population fishing the Cattaraugus Creek. Estimates are presented for the maximum individual and for the total population fishing the survey regions. These estimates include both internal exposures from ingestion of fish and external exposures received while fishing from radionuclides deposited along the streams.

The whole-body ingestion doses include contributions from caesium-137, caesium-134 and zinc-65. The bone doses are from strontium-90 only. The estimates presented are the total doses delivered over the lifetime of the individual (50-year period) from the ingestion of radionuclides during 1971. These doses were calculated following the procedures utilized by Shleien [9]. The dose from caesium is essentially (> 90%) all delivered in the first year after intake. For strontium-90 about 7% of the dose is delivered during the first year after intake and for zinc-65 about 70% of the dose is delivered in the first year.

The estimates of the external doses received by the population fishing regions of the Cattaraugus Creek near the NFS plant were made using the occupancy data obtained during this survey and dose measurement data obtained with a pressurized ionization chamber by the New York State Department of Environmental Conservation [8].

The results of the fishing survey had shown that the maximum individual had spent 200 hours and that the total population had spent 7500 man-hours fishing these regions of the Cattaraugus Creek. The results of the dose-rate measurements indicated that the liquid discharges from NFS had resulted in an average dose rate of 5 μR/h above background for the region of the Cattaraugus Creek used in the fishing survey.

The maximum whole-body external dose to an individual fishing the Cattaraugus Creek during 1971 from NFS discharges is estimated to be approximately 1 millirem. This exposure was about 2.5 times higher than the estimated whole-body doses from the ingestion of radionuclides in fish flesh. The maximum dose commitment to bone of an individual was estimated to be 7 millirems.

The whole-body doses from NFS discharges to the population fishing the Cattaraugus Creek was estimated to be 0.04 man-rem from external radiation and 0.02 man-rem from ingestion of radionuclides in fish. The dose commitment to bone for the total population was 0.3 organ-rem.

These dose estimates are considerably smaller than those reported by Shleien for 1968 [1]. The differences result mostly from lower estimates of intake based on the fishing survey, but are also partly due to lower concentrations of radionuclides in fish flesh in 1971 than were used by Shleien for 1968.

3. RADIONUCLIDE INTAKES AND ESTIMATES OF INGESTION DOSE TO POPULATION FROM VENISON CONSUMPTION

To obtain information on the size of the population involved in deer hunting and the intake of deer meat for this population, the US Environmental Protection Agency, in co-operation with the New York State

Department of Environmental Conservation, conducted a survey of hunters taking a deer within a 32-km radius of the NFS during the 1970 hunting season.

Presented below are: (a) a description of the survey procedures, (b) a summary of the survey results, (c) maximum and average concentrations of radionuclides in venison, and (d) dose estimates for venison intake from deer kills near NFS.

3.1. Survey procedures

The New York State Department of Environmental Conservation, Division of Fish and Wildlife requires that each deer killed within the state be reported to them by means of a card furnished to the deer hunter when a licence is issued.

During the deer hunting season of 1970 approximately 50 000 hunters reported deer kills for the entire State of New York. A tedious search of the 50 000 cards produced the names of 2531 deer hunters who had reported a kill within a 32-km radius of the plant. By means of supplemental checks of hunters for several years (roadside and in the field checks), the Division of Fish and Wildlife has determined that 80% of the hunters actually officially report their kills. Correcting the reported kills for this factor, the deer kill for the area of interest as determined by the survey would have been about 3200. The estimated deer population in the area within a 32-km radius of the plant is estimated by the New York State Department of Environmental Conservation to be about 20 000 deer.

Of the 2531 reported deer kills only 2100 reports were sufficiently legible to provide adequate information to be included in the survey. A questionnaire and cover letter were mailed to these 2100 deer hunters with a postage-paid, return envelope furnished. Fifteen hundred and fifty-eight completed forms were received, representing a slightly more than 75% return. Forty-five questionnaires were returned undelivered. Of the completed questionnaires 1455 provided sufficient information to be useful in the data evaluation. From these questionnaires information was obtained on: (a) the number of deer taken by a hunter; (b) the dressed and edible weights of the deer; (c) the number and ages of individuals consuming the venison; (d) the amount of venison given away; and (e)

TABLE V. SUMMARY OF SURVEY DATA ON VENISON INTAKES FROM DEER TAKEN WITHIN A 32-km RADIUS OF NFS DURING 1970

Deer hunters surveyed	1455
Individuals consuming venison in hunter's household	6100
Average venison consumption	2.7 kg
Maximum venison consumption by an individual	44.8 kg
Total venison consumption within surveyed households from own deer	16.7×10^3 kg
Total venison consumption outside surveyed households (given away by hunters)	6.5×10^3 kg
Total venison consumption inside and outside surveyed households	2.3×10^4 kg

amounts of venison received from other sources. From this information an estimate was made of the total and average consumption of venison by the population at risk.

This report will present only a brief summary of the venison intake data obtained during the survey and these data are presented in Table V. A much more detailed evaluation of these survey data are presented elsewhere [6]. The survey data showed that there were 6100 individuals consuming venison in the households of the 1455 deer hunters from whom usable survey data were obtained. The average consumption for this population was 2.7 kg per person. The maximum intake reported by an individual was 44.9 kg. The total consumption of venison from deer taken within a 32-km radius of the NFS by the families of the 1455 deer hunters surveyed was 16 700 kg. A total of 6500 kg of venison was given away by the surveyed hunters, making a total of 23 200 kg of venison consumed from deer taken within the 32-km radius of the NFS.

The determination of consumed venison was based upon individual deer hunter estimates of the dressed and edible weights. The various weights are defined as follows:

W_d dressed weight = live weight (W_l) less entrails
W_e edible weight = dressed weight less skin, head, hooves but including bone
W_c consumed weight = edible weight less bone and fat (essentially muscle only).

Based upon approximate relations determined by the Division of Fish and Wildlife of the New York State Department of Environmental Conservation, the following conversion factors were used:

$$W_d = 0.78\ W_l$$
$$W_e = 0.75\ W_d$$
$$W_c = (0.30\text{-}0.35)\ W_d = 0.33\ W_d$$
$$W_c = 0.44\ W_e$$

To extrapolate the data from the households surveyed to the total population consuming venison from legally taken deer in the area of interest the survey data were corrected for the return of usable questionnaire and for the number of deer taken that are not reported. This correction factor was calculated as follows:

$$\frac{1455\ \text{(Usable questionnaires)}}{2531\ \text{(Reported deer kill)}} \times 0.80\ \text{(Fraction of deer kill reported)} = 0.46$$

TABLE VI. EXTRAPOLATED VENISON INTAKES FROM LEGALLY TAKEN DEER WITHIN A 32-km RADIUS OF NFS DURING 1970

Persons consuming venison in hunter's household	13 260
Venison consumed in hunter's household	3.6×10^4 kg
Venison consumed outside hunter's household (given away)	1.4×10^4 kg
Total weight of venison consumed	5.0×10^4 kg

Table VI represents a summary of the extrapolated data for the consumption of venison by the total population involved in deer hunting in the area of interest. The total weight of venison consumed from deer taken legally within a 32-km radius of the NFS for 1970 was estimated to be 5.0×10^4 kg.

3.2. Radionuclide concentrations in venison from the vicinity of NFS during 1970

Data on the maximum and average concentrations of caesium-137, caesium-134, strontium-90 and zinc-65 in venison samples obtained from deer kills in the vicinity of the NFS during 1970 are presented in Table VII. The concentrations of ^{137}Cs, ^{134}Cs and ^{65}Zn present in on-site deer were significantly higher than in deer taken off-site or in the background deer. It appears that most of the ^{137}Cs, ^{134}Cs and ^{65}Zn present in the on-site deer resulted from waste discharged from the NFS. The presence of ^{134}Cs at concentrations 10-20% of the caesium-137 concentrations verifies the origin of the caesium radionuclides as coming from the NFS operations.

The picture for strontium-90 is not clear. The contribution of the NFS operations to the strontium-90 concentrations in venison cannot be determined from the available data. The strontium-90 concentrations in on-site deer do not differ significantly from those in the deer taken off-site, although the strontium-90 concentrations in both the on-site and off-site deer appear to be higher than in the deer taken in Albany. Additional data are necessary to determine if the deer grazing on the NFS site accumulated strontium-90 due to the NFS operations.

Caesium-134 and zinc-65 were not detected in deer taken off-site and this indicates that these deer were probably not influenced by the NFS waste discharges. The radionuclide concentrations in these samples are probably more representative of background levels in the area than are the samples collected at Albany.

3.3. Radionuclide intakes

Table VIII presents the radionuclide intakes from the consumption of venison obtained from deer kills in the vicinity of NFS during 1970.

The radionuclide intakes for the maximum individual were calculated using the maximum venison intake reported by an individual during the survey and the maximum concentrations of radionuclides measured in a deer collected on-site. For a reliable estimate of the intake by the total population from the deer pathway from the operation of the NFS much more data are needed than at present available on the concentrations of radionuclides in the deer population potentially influenced by the waste discharge. A large sample of the deer population would be necessary to obtain this information and more detailed information on background levels for the region would also have to be obtained, and even then the data may be inclusive. The data presented in Table VIII are therefore only a gross estimate of the population intake determined using the following approach.

Deer freely enter and leave the NFS site. They accumulate radionuclides only while grazing on-site. Approximately 125 deer constitute the on-site deer population at any given time, assuming the deer density to

TABLE VII. RADIONUCLIDE CONCENTRATIONS IN VENISON OBTAINED FROM VICINITY OF NFS DURING 1970

Collection location	Concentrations (μCi/kg)								
	Maximum				Average				
	^{137}Cs	^{134}Cs	^{65}Zn	^{90}Sr	No. of samples	^{137}Cs	^{134}Cs	^{65}Zn	^{90}Sr
On 3300-acre NFS site	4.1×10^{-3}	7.0×10^{-4}	-	2.8×10^{-5}	22	1.2×10^{-3}	1.9×10^{-4}	6.3×10^{-5}	6.0×10^{-6}
Off-site within 16-km radius	4.8×10^{-4}	$< 1.5 \times 10^{-5}$	$< 2.5 \times 10^{-5}$	2.0×10^{-5}	10	2.8×10^{-4}	$< 1.5 \times 10^{-5}$	$< 3.0 \times 10^{-5}$	1.0×10^{-5}
Albany, N.Y., control	1.4×10^{-4}	$< 1.5 \times 10^{-5}$	-	$< 2.0 \times 10^{-6}$	4	8.8×10^{-5}	$< 1.5 \times 10^{-5}$	-	$< 2.0 \times 10^{-6}$

TABLE VIII. ESTIMATED RADIONUCLIDE INTAKES FROM CONSUMPTION OF VENISON OBTAINED FROM DEER KILLS IN THE VICINITY OF NFS - 1970

Radionuclide	Intake Maximum by an individual (μCi)	Total by population (μCi)
^{137}Cs	1.8×10^{-1}	1.4
^{134}Cs	3.2×10^{-2}	2.5×10^{-1}
^{65}Zn	-	2.2×10^{-3}
^{90}Sr	1.3×10^{-3}	9.8×10^{-3}

be the same as in the surrounding area. The maximum concentrations of radionuclides measured in a deer collected on-site constitute the amounts that would be accumulated by a deer grazing on-site for a full year. By applying the maximum concentrations to all 125 deer, we can approximate the amounts of activity that would be accumulated by the entire deer population grazing on-site during the year (the equivalent of 125 grazing years). Of course the actual accumulation would be distributed among many more deer involved in grazing for shorter periods of time. Based on the deer kill data for the area, 22 deer from the population grazing on the NFS site would be killed by hunters. The venison intake from these deer would represent ~350 kg, using a value of 16 kg of consumed venison per deer. The radionuclide intake from these 22 deer would then represent an estimate of the radionuclide intake for the total population.

3.4. Dose estimates

Table IX presents a summary of the doses to the population consuming venison from deer killed in the vicinity of NFS. Estimates are presented for the maximum individual and for the total population. The maximum whole-body dose an individual could have received was 14 mrem. The

TABLE IX. ESTIMATED DOSES TO POPULATION FROM CONSUMPTION OF VENISON OBTAINED FROM DEER KILLS IN THE VICINITY OF NFS - 1970

Organ	Dose Maximum individual (mrem)	Population (man-rem)
Whole-body [a]	14	0.1
Bone [b]	11	0.1

[a] Includes contribution from ^{137}Cs, ^{134}Cs and ^{65}Zn.

[b] Dose from ^{90}Sr only and not directly attributed to NFS.

maximum bone dose to an individual from strontium-90 was 11 mrem with only 0.7 mrem being delivered during the first year. These doses are the maximum doses an individual could have received. Most probably the dose that an individual actually did receive was considerably smaller than the doses listed in Table IX since the probability is extremely small that the individual with the maximum intake would consume venison from two deer with the maximum concentrations of radionuclides.

The whole-body integrated population dose from the deer pathway was estimated to be about 0.1 man-rem.

4. WASTE DISCHARGES FROM NFS AND RADIONUCLIDE CONCENTRATIONS IN THE CATTARAUGUS CREEK

A summary of the radioactive waste discharges from NFS during the period of the plant operation from April 1966 to December 1971 is presented in Table X. The data presented were obtained from NFS reports [7] or were estimated from EPA studies [2-5]. The principal radionuclides discharged in the liquid effluent were tritium, ruthenium-106, caesium-137, caesium-134 and strontium-90. The principal radionuclides discharged in the gaseous effluent were krypton-85, tritium and iodine-129. The particulate beta activity in the stack effluent was $< 0.1\%$ of the beta actively discharged in the liquid effluent.

Approximately 40 curies each of strontium-90 and caesium-137 were discharged in the liquid effluent during the period of operation of the plant. These radionuclides were discharged from the lagoon system at a concentration of about 10^{-4} μCi/ml and at a flow rate of 3-6 litres/s.

Table XI presents data on the concentrations of strontium-90 and caesium-137 in the Cattaraugus Creek at Springville Dam, a location 4 km downstream of the point of entry of the waste. Also presented in Table XI are the yearly discharges of strontium-90 in the liquid waste.

TABLE X. RADIOACTIVE WASTE DISCHARGES FROM NFS APRIL 1966 - DECEMBER 1971

Liquid effluent		Gaseous effluent	
Radionuclide	Curies released	Radionuclide	Curies released
Gross α	8×10^{-1}	Gross β	2
Gross β	3.9×10^{2}	^{85}Kr	1×10^{6}
^{3}H	2.2×10^{4}	^{3}H [a]	2×10^{3}
^{129}I	9×10^{-1}	^{129}I [a]	1
^{106}Ru [a]	2.5×10^{2}	^{131}I	< 0.36
^{134} + ^{137}Cs [a]	5.0×10^{1}		
^{90}Sr	4.1×10^{1}		
Others [a]	1.5×10^{1}		

[a] Estimate based on EPA Studies [2-5].

TABLE XI. RADIONUCLIDE CONCENTRATIONS OF ^{90}SR AND ^{137}CS IN CATTARAUGUS CREEK - 1966-1971

Year	Discharges (Ci)	Water (pCi/litre)		Sediment (pCi/g)	
	^{90}Sr[a]	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
1966	-	62	-	-	-
1967	4	24	<20	2	37
1968	5	25	20	1	12
1969	10	47	<20	10	35
1970	14	69	25	2	38
1971	7	37	<20	-	-

[a] ^{137}Cs discharges are estimated to be approximately equal to ^{90}Sr discharges.

The water concentrations presented are average values of weekly composite samples. The sediment samples are average values for a series of samples collected during the month of June.

The average yearly strontium-90 concentration in water ranged from 24-69 pCi/ litre and essentially all this strontium-90 was presented in the dissolved fraction of the water. The average yearly caesium-137 concentration was ~ 20 pCi/litre with essentially all the caesium associated with the suspended solids in the sample. For a more detailed discussion of the variables affecting the caesium-137 concentration in Cattaraugus Creek see Ref. [2].

The caesium-137 concentration in sediment from the Cattaraugus Creek was an order of magnitude greater than the strontium-90 concentration. There was no apparent build-up of the concentration of either of these radionuclides in sediment with time. It is believed that most of the sediment from the Cattaraugus Creek is washed out into Lake Erie. The concentrations of these radionuclides in sediment at the mouth of the Creek, 64 km downstream from the plant site, are about the same as the concentrations at Springville Dam.

Strontium-90 is transported through the creek system to Lake Erie primarily by the movement of water, while the caesium-137 is transported through the creek system to Lake Erie primarily by the movement of sediments.

5. SUMMARY AND CONCLUSIONS

Estimates of the doses resulting from NFS waste discharges to the population hunting and fishing in the vicinity of the plant during 1971 are presented. These estimates indicate that the whole-body man-rem doses to the population from fishing and hunting were about the same magnitude, i.e. 0.1 man-rem for the year. The external dose received from radionuclides deposited along the Cattaraugus Creek was twice the internal exposure received from ingestion of radionuclides in fish, indicating that this pathway should receive more consideration in evaluating exposure from released radionuclides. The dose commitment to bone for the population fishing in Cattaraugus Creek was estimated to be 0.3 organ-rem. Data did not permit an estimate of the population bone dose from ingestion of venison but it was determined that this dose would be < 0.1 organ-rem.

The maximum whole-body dose to an individual fishing the Cattaraugus Creek during 1971 was estimated to be about 1.4 mrem: 1 mrem from external exposure and 0.4 mrem from ingestion of radionuclides in fish. The 50-year dose commitment to bone of an individual from the 1971 intake was estimated to be 7 mrem. These are dose estimates for a real, identified individual.

The maximum whole-body dose an individual could have received from ingestion of venison from deer kills in the vicinity of NFS was estimated to be 14 mrem. This is based on a hypothetical radionuclide intake that has a low probability of occurrence since it requires that the individual with the maximum venison intake kill deer containing the maximum concentrations of radionuclides. The maximum dose to an individual therefore was most probably considerably < 14 mrem.

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DISCUSSION

I. L. BRISBIN, Jr.: Maximum exposure risk through the ingestion of venison frequently occurs in the case of poachers, who may often rely on game as food staples. Since poachers usually kill their deer out of season, or take deer or fish above the legal limit, they naturally do not report their activities to the appropriate authorities. How large do you think the poaching element might be in the area of your study?

P. J. MAGNO: New York State Department of Environmental Conservation personnel with whom I have spoken consider that about 80% of the deer kill is reported.

F. O. HOFFMAN: I see that you list venison eaters and fish eaters separately. Did you consider the possibility that, in sporting circles, a hunter and a fisherman may be one and the same person?

P. J. MAGNO: Yes, I considered the possibility, but in spite of having gone through all the questionnaires and interview sheets, I was unable to identify any individual who both reported a deer kill and was recorded as a fisherman.

E. KUNZ: In literature previously published on exposure evaluations connected with this same plant a specific pathway was mentioned, namely deer coming through a hole in the fence of the high exclusion area in order to drink water directly from the lagoons. Was this pathway soon interrupted?

P. J. MAGNO: Entry by the pathway you mention may have occurred in the early days of plant operation, but precautions have now been taken to prevent its recurrence.

W. O. SCHIKARSKI (Chairman): Have you examined your stack release values for particulates containing plutonium?

P. J. MAGNO: We have studied particulate stack releases in a report entitled: "An Investigation of Airborne Radioactive Effluent from a Nuclear Fuel Reprocessing Plant" (BRH-NERHL 70-3 (1970)).

MODELLING RADIATION EXPOSURES TO POPULATIONS FROM RADIOACTIVITY RELEASED TO THE ENVIRONMENT*

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Abstract

MODELLING RADIATION EXPOSURES TO POPULATIONS FROM RADIOACTIVITY RELEASED TO THE ENVIRONMENT.

Two approaches are used to estimate the potential population dosages (man-rem) per curie release of caesium-137 in the United States of America from a hypothetical release of a fraction of the projected fission product inventory in 2000. One method involves a parametric analysis of relationships between fall-out rates and caesium-137 in the various components of the diet. The other method is a pathways analysis using dose-to-man models of radionuclide transport in the environment and uptake in man. Both methods are applied to an input-output model of radionuclide flux through environmental compartments and man. Population characteristics are developed using census data and dietary habits. Man-rem/Ci estimates are developed for several food-chains and external exposures. External exposure from soil burdens contributes well over half of the man-rem, followed by terrestrial food-chains, with much lower contribution coming from aquatic food-chains. Submersion and inhalation exposures from either the initial cloud or from resuspension are insignificant contributions. Calculations by the two methods are in good agreement. The man-rem/Ci estimates are used to order the importance of the various routes of exposure and to investigate the significance of contamination in various parts of the environment.

INTRODUCTION

By the year 2000, 500 to 1000 nuclear power reactors will be in operation in the United States, according to present projections. Accompanying this growth in nuclear power plants will be a large inventory of radioactive wastes to be managed; for example, it is projected that by the year 2000, the accumulated inventory of cesium-137 will be approximately 15 billion curies [1]. Even with strict controls and containment, releases will likely occur. It is pertinent to evaluate the impact of a hypothetical release of a portion of the fission product inventory. One step in evaluating the biological cost of such releases is to estimate the relationship between a curie of release and the resulting dosage in the population.

Assessments have been made of population dosages which would occur from projected nuclear applications involving release of tritium [2] and krypton-85 [3], and Russell has evaluated the dose commitment to the population from a massive deposition of strontium-90 [4].

The present assessment is an estimate of the man.rem per curie release of cesium-137 in the United States population in the year 2000. Computations are carried out on an input-output model of radionuclide flow through environmental pathways which include man (see Fig. 1). One evaluation is a parametric analysis of dose-to-man based on data and models available from analysis of worldwide fallout in the biosphere. A second evaluation is carried out using existing dose-to-man pathway models developed in the LLL. Some data and modeling steps are common to both evaluations, so that we have not developed two totally independent evaluations.

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* Work performed under the auspices of the US Atomic Energy Commission.

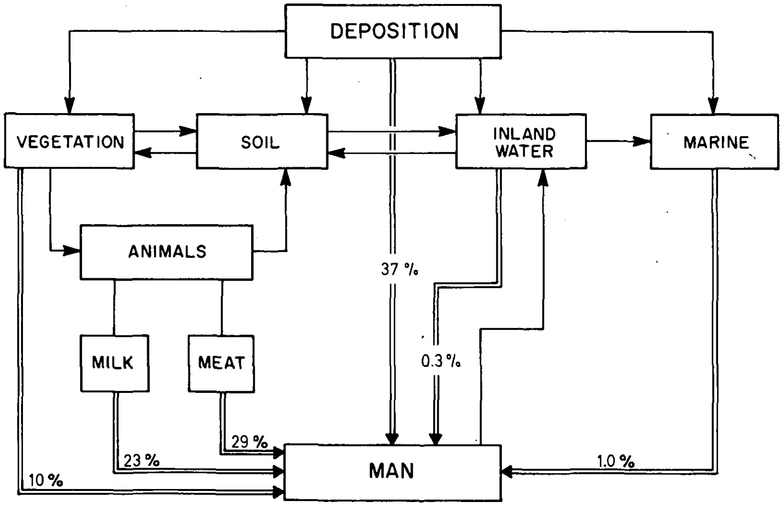

FIG. 1. Pathway model for incorporation of caesium-137 in man after deposition showing the per cent man-rem from each pathway.

The model used in this study is a special case of a more general model for determining population dosages. The general model demands accountability for all nuclear decay occurring in man, and thus is global in concept. Such a concept is particularly necessary for evaluation of radionuclides and pathways for which dispersion and cycling are rapid, for instance in the case of tritium and the noble gases. For radionuclides which move slowly in the environment, population dosages tend to be confined to a population in or near the region of initial contamination. Importation-exportation of foods and materials tend to level such partitions. In the present case, the evaluation of man•rem is for the United States population only, and from a uniform hypothetical deposition on the continental United States.

Such evaluations are important for gaining perspective, and emphasis should be placed upon the generalizations which can be derived from them. Thus it is important to remember the assumptions, to recognize the uncertainties, and to carry out sensitivity analysis and verification.

In the present evaluation, the release to the environment is arbitrarily assumed to be 1.5×10^6 Ci, which is 10^{-2}% of projected year 2000 inventory of cesium-137. This release is assumed to occur uniformly in time over one year over the United States. No attempt is made to characterize deposition, transport or fate of radioactivity on a regional basis. The study then, assumes a homogeneous, evenly distributed environmental system and population distribution. This assumption is implicit in the parametric analysis (dealing with averages) and explicit in the detailed pathway analysis.

The models are used to ascertain:

1) Estimates of total man•rem per curie and the contribution of the various pathways.

2) Comparison of results obtained from simple pathway analysis with those from an empirical parametric analysis of worldwide fallout.

3) Sensitivity of man•rem dosage prediction to assumptions in the various models of transport and fate.

A Man·Rem Model from Parametric Analysis of Worldwide Fallout

A. Prediction of Concentrations in Foods and Total Activities Ingested

1. Terrestrial Foods

Empirical models have been developed to predict cesium-137 in foodstuffs as a function of the fallout rate and the accumulated deposit in soil. Such analyses have been extensively carried out for milk. Cesium-137 in milk is often described as a function of the current rate of deposition, and the accumulated deposit. If F_r is the current rate of deposition and F_d, the cumulative deposit,

$$\text{cesium-137 in milk [pCi/liter]} = p_r F_r + p_d F_d \quad . \qquad (1)$$

Data from various locations including the United States [5], Denmark [6], Italy [7], Australia [8], and the United Kingdom [9], indicate p_r values vary between 2.2 and 37.8 when F_r is expressed in mCi/km²/yr and F_d is expressed mCi/km². The p_d values, which will be discussed later, also vary over a wide range. We shall assume that the deposition F(0) [mCi/km²] is uniformly deposited over a period of a year, serving to balance out seasonal influences and to simplify the calculations. Equation 1 can now be used to derive an expression for the total integrated intake of activity via milk:

$$\text{Total cesium-137 ingested via milk } [\mu\text{Ci}] = (A + B)F(0) \quad . \qquad (2)$$

Coefficient A is a measure of the intake during the first year and coefficient B is a measure of the intake during subsequent years. If milk is consumed at the rate of 1 liter/day, the p_r values lead to A values in the range 8×10^{-4} to 1.4×10^{-2}. Simple food-chain models [10,11] also lead to values of A lying within this range for the situation in which a discrete or continuous deposition takes place within a year's time. The coefficients p_r and A of Eqs. [1] and [2] derived for milk from the various worldwide locations vary by more than an order of magnitude.

Wilson's assessment of the concentration of cesium-137 in milk from various U.S. milksheds [5] suggests that the average p_r value for the U.S. would be near the low end of the range. We use a value of $p_r = 5.5$ which reflects these data. This gives an A value of 2.0×10^{-3} when milk is consumed at the rate of 1 liter/day.

The long-term availability of land-deposited cesium-137 to the diet has been difficult to assess [12]. Soil factors p_d derived for the assessment in milk have varied over a wide range depending on the pattern assumed for the disappearance of the activity in soil [6,7,8,9]. One can estimate a soil factor p_d for milk by attributing the cesium-137 in milk that is not associated with the deposition rate to the cumulative deposit in soil. On the basis of Wilson's assessment of cesium-137 in U.S. milksheds [5] and data on cumulative deposits [13], we have estimated a range of p_d values that would be expected to include the national average. A range of 0.14 to 0.20 can be derived if it is assumed that the activity in soil is effectively diminished by radioactive decay alone [6]; a range of 0.65 to 0.9 can be derived if it is assumed that only 60% of the previous year's deposit is available in a given year [7]. These values produce an extreme range of 4.7×10^{-4} to 3.1×10^{-3} for the B factor in the expression for the total intake of cesium-137 via milk. When the estimated values $A = 2.0 \times 10^{-3}$ and $B = 3.0 \times 10^{-3}$ are substituted into Eq. (2), the expression for the total cesium-137 ingested via milk consumption at the rate of 1 liter/day becomes

$$\text{Total intake via milk } [\mu\text{Ci}] = 5.0 \times 10^{-3} F(0) \quad . \qquad (3)$$

Federal milk marketing order statistics indicate an average per capita consumption of fluid milk in 67 U.S. cities of 0.3 liter/day [14]. Per capita consumption of milk and cream in Denmark [15] is reported as 0.4 liter/day; cheese, butter, and margarine contribute an additional 100 g of dairy products to the daily diet. Brar and Nelson assume an average milk consumption rate of 0.6 liter/day in their studies of cesium-137 in the Chicago diet [16]. We assume an intake of dairy products equivalent to 0.6 liter/day of fluid milk (see Table II).

Dairy products, grain products and meat have been the major contributors of cesium-137 in the diet. In the decade 1958-1968, milk products contributed 13 to 23% of the cesium-137 in the Danish diet [6] and 25 to 40% in the U.S. diet [17]. The lower percentages were observed during periods of low fallout-rate. These considerations can be used to derive an expression for the total intake of cesium-137 from the terrestrial diet from the expression previously derived to predict the total cesium-137 ingested via milk. The expression takes the form of Eq. (2). We have chosen a value of 3.0×10^{-3} for the A factor and have adjusted the B factor for milk assuming milk contributes 25% of the dietary cesium. The resulting expression is

$$\text{Total cesium-}^{137}\text{ ingested via terrestrial foods } [\mu\text{Ci}] = 1.0 \times 10^{-2}\, F(0)[\text{mCi/km}^2] \;. \quad (4)$$

Aarkrog has derived equations for predicting mean levels of cesium-137 in the Danish diet from the fallout rate and cumulative deposition [6]. These equations predict an integrated intake of 9.1 to 9.8×10^{-3} μCi of cesium-137 from a deposition of 1 mCi/km^2. Approximately the same total ingestion of cesium-137 has been estimated in Table II for terrestrial foods from the U.S. and Denmark. Cesium-137 in foods and in the population closely follow the rate of fallout [12]. In the period 1960-1968 average deposition rates and cumulative deposits of cesium-137 in the U.S. and Denmark were comparable, but average body burdens and concentrations in food in Denmark were higher [6,17]. The soil in Denmark has a high clay content [6], and only a small fraction of the estimated intake of cesium-137 in Denmark is attributable to long-term uptake from the soil. On the other hand, a substantial contribution from long-term uptake from soil is implicit in our estimate for the U.S. intake of cesium-137. More reliable data of soil factors and long-term availability of cesium-137 from soil remain to be established.

2. Freshwater Foods

During the period 1962-1968 concentrations of cesium-137 in freshwater fish from Finland [18], Chicago [19], the Red Lakes, Minnesota [20] and Sweden [21] were measured at levels up to several nCi/kg. The concentrations were 1 to 2 orders of magnitude greater than those in marine fish. Gustafson [20] derived an expression to represent the concentration of cesium-137 in fish taken from the Red Lakes, Minnesota, 1954-1965. The expression included contributions by terms containing the deposition rate F_r and the total deposition F_d in the same manner shown previously for the assessment of milk contamination [see Eq. (1)].

$$\text{cesium-137 in fish [pCi/kg]} = p_r F_r\ [\text{mCi/km}^2/\text{yr}] + p_d F_d\ [\text{mCi/km}^2] \;. \quad (5)$$

Application of Gustafson's model for the Red Lakes to predict the concentration of cesium-137 in freshwater fish and the total intake of cesium-137 via freshwater foodstuffs leads to:

$$\text{Total cesium-137 ingested via freshwater foods } [\mu\text{Ci}] = (A + B)F(0)[\text{mCi/km}^2] \;. \quad (6)$$

Values of $p_r = 110$ and $p_d = 20$ were adopted for Eq. (5) on the basis of the highest concentrations measured in the large carnivores representing the highest trophic levels. If freshwater fish are consumed at the rate of 1 kg/yr, the resulting coefficients for Eq. (5) are $A = 1.1 \times 10^{-4}$ and $B = 8.7 \times 10^{-4}$. The contribution from the term involving A is associated with the intake over the first year, that from the term involving B is associated with intake over the succeeding years. Coefficient B was evaluated assuming an effective half-life in the environment equal to the radioactive half-life. The 1 kg/yr average consumption rate for freshwater fish implicit in the calculations is the rate assumed by Brar and Nelson in their studies of cesium-137 in the Chicago diet [16]. This rate is less than that in Sweden, where freshwater fish is consumed at an average rate of 2.5 kg/yr [21]. Substituting for A and B in Eq. 6 leads to:

$$\text{Total cesium-137 ingested via freshwater fish } [\mu\text{Ci}] = 9.8 \times 10^{-4} F(0)[\text{mCi/km}^2] \quad (7)$$

3. Marine Foods

The marine ecosystem is assumed to be bounded by the 8000-km U.S. coastline, extending a distance of 75 km, and having an average depth of 130 m (see Table I). The width and depth selected represent average values for the continental shelf [22]. The ecosystem receives runoff from the continent, exchanges water with the deep ocean beyond the continental slope and has bottom sediments extending halfway across the width [22].

A simple model was developed to predict cesium-137 in marine fish as a function of time and the total cesium-137 ingested via marine foods. Cesium-137 in the water, fish, and sediments are in equilibrium. Concentration factors reported for stable cesium and cesium-137 in marine fish range from about 20 to 120 [23,24,25,26] and are comparable to those in marine invertebrates [27,28], Concentration factors upwards of 10^2 have been measured between sediments and water in Chesapeake Bay [29], Humboldt Bay [30] and the Hudson River estuary [31]. We have assumed concentration factors of 10^2 in marine fish and 10^3 in sediments. On the basis of data from Humboldt Bay [30] we have assumed that the activity in sediments is contained within a layer 3.6 cm in depth. Residence time of the water is assumed to be 3 yr, a value intermediate between the residence time of about 10 yr for the water in the mixed surface layer of the deep ocean [32] and the shorter residence time of about 1 yr for the water in an estuary such as Chesapeake Bay [29].

TABLE I. PROPERTIES OF THE MARINE ECOSYSTEM

Length (U.S. Coastline excluding Alaska, Hawaii)	8000 km
Width (ave. width continental shelf)	75 km
Depth (ave. depth continental shelf)	130 m
Width of sediment layer	40 km
Depth of sediment layer	3.6 cm
Bulk density of sediments	1.5 g/cm^3
Residence time of water	3 yr
Concentration factor of cesium-137 in fish	100
Concentration factor of cesium-137 in sediments	1000
Annual runoff of cesium-137 (fraction of cumulative deposition)	10^{-3}/yr
Land area (U.S. excluding Alaska)	7.8×10^6 km^2

Bowen [33] has estimated that oceans receive about 12% of the yearly strontium-40 deposited on land or about 0.4% per year of accumulated deposition. Runoff could be expected to contribute even smaller amounts of cesium-137 to the oceans, since cesium is more tightly bound to the clay minerals present in soil. The average annual cesium-137 to strontium-90 ratio in New York City tap water 1965-1971 varied between 0.08 and 0.16. During 1968-1970, cesium-137 in New York City tap water and in freshwater from streams from Pennsylvania to Georgia [29] was for the most part below 0.1 pCi/liter. Assuming that the annual runoff to the oceans is 9 inches [34] and that the mean cesium-137 in runoff from the entire United States did not exceed 0.1 pCi/liter during this period, the yearly runoff of cesium-137 would be about 1.5% of the yearly deposition or about 0.025% of the cumulative deposit. We adopt the value of 10^{-3} for the fraction of the cumulative land-deposited cesium-137 that is delivered annually to the oceans via runoff.

Let us consider this ecosystem from the standpoint of the annual marine-food requirements of the population. Per capita consumption of fish in the United States has been estimated to be upwards of 5 kg/yr [35]. If the average consumption of seafood is 7 kg/yr, a value suggested by Brar and Nelson in their studies of cesium-137 in the Chicago diet [16], 2.1×10^6 metric tons of seafood are needed annually to feed the U.S. Since about two-thirds of the seafood harvest is utilized as human food [36], the ecosystem would have to provide an annual seafood harvest of 3.1×10^6 metric tons in order to accommodate the total population. This situation can be described in terms of an annual production of 3.1×10^6 metric tons/6×10^5 km^2 or approximately 5 metric tons/km^2. The 1962 catch from the northwest Atlantic area is about 1 metric ton/km^2 [37]. If we assume that the marine-food harvest landed in California ports is derived from a 75-km region adjacent to the Pacific coast of the United States, the 1969 catch [38] was about 2 metric tons/km^2. In an analysis of the marine catch obtained from California coastal waters, Vaughan and Strand [39] considered the surface in block areas as small as 100 square miles. An average catch of 3.5 metric tons/km^2 can be derived from this study.

Evaluation of the marine-ecosystems model leads to expressions that predict cesium-137 in seafood as a function of time. These are integrated to yield the total intake of cesium-137 resulting from a given rate of consumption of seafood. When the coefficients are evaluated, the expression for the total intake is

$$\text{Total cesium-137 ingested via marine food } [\mu\text{Ci}] = A\ F(0)\ [\text{mCi/km}^2] \ . \qquad (8)$$

This assessment assumes seafood consumption at a rate of 7 kg/yr. Three situations are contrasted (see Table II). The "limiting" case, which is associated with the maximum intake of activity, is the hypothetical situation in which (1) sediments are absent, (2) the land-deposited activity is instantaneously transported to the marine environment in runoff, and (3) water does not exchange with waters of the deep ocean beyond the continental slope. A more realistic case is the situation in which (1) sediments would be present, (2) runoff would deliver the land-deposited activity to the marine environment at the rate of 10^{-3}/yr or less, and (3) there is exchange with the deep ocean. An "intermediate" case is also of interest, namely the case in which sediments are present and runoff delivers land-deposited activity to the marine environment at the rate of 10^{-3}/yr, but in which there is no exchange of water with the deep ocean. This case examines a situation in which the activity is at all times accessible to the U.S. population. As shown in Table II, the total intake in the "intermediate" case differs from those in the other cases by an order of magnitude. The estimated intake of cesium-137 in the presence of sediments is slightly less than that in their absence and the contribution of normal runoff to cesium-137 ingestion is insignificant.

B. Assessment of Population Dosage from Foods

1. Activity Ingested via Terrestrial and Aquatic Foods

Table II summarizes the calculations for the total ingestion via terrestrial and aquatic food chains. A comparison of terrestrial and aquatic foods indicates that when cesium-137 is released to the environment and deposited on both water and land surfaces, the average individual ingests a far greater quantity of cesium-137 via milk and the terrestrial diet than from freshwater or marine foods. However, if all the activity released is directed to inland waters, the ingestion of cesium-137 is comparable to that via the terrestrial diet. For the marine ecosystem, only the "limiting" case (where the total activity released to the environment is concentrated and retained within the marine ecosystem) approaches the significance of the milk and the terrestrial diet. The "representative" case is 2 orders of magnitude less than this "limiting" case. Marine foods would not be expected to contribute more than 1% of the cesium-137 in the average individual's diet.

Intakes of cesium-137 are listed at 30-yr intakes and for infinity (Table II). For the "limiting" and "representative" cases, 50% or more of the intake-to-infinity is achieved in 30 years. In the "intermediate" case a substantial fraction of the infinite-time integrated intake is achieved in 30 years. Thus, in realistic situations, a major fraction of the dosages associated with the integrated intake of cesium-137 would be delivered within the lifetime of a generation.

2. Man·Rem Dosage

Table III lists the total activities ingested and the whole-body dosages to the average individual and to the population from the release of 1.5×10^6 Ci of cesium-137. In the calculations relating to the terrestrial and freshwater pathways, the deposition F(0) is based on a combined land and inland-water surface area of 7.8×10^6 km^2, with F(0) = 190 mCi/km^2. In the calculations relating to the marine pathway, F(0) is based on a combined land and water surface area of 8.4×10^6 km^2, with F(0) = 180 mCi/km^2. Population intakes of cesium-137 via milk and terrestrial diet, and as indicated earlier, that via seafood, are based on a population of 3×10^8 people. The population intake via freshwater fish is based on a smaller population, 5×10^7, since the yearly commercial harvest of freshwater fish, which we estimate to be about 50,000 metric tons of consumable food [38], can supply only a sixth of the population at the consumption rate of 1 kg/yr.

Results allow a comparison of the maximum intakes via the terrestrial and aquatic food-chains. For example, even the "limiting" case for the marine-foods pathway is shown to contribute less activity to the diet than terrestrial foods. On the other hand, in the special situation in which all of the cesium-137 released to the environment is initially deposited in inland waters, the total population intake would be comparable to that from terrestrial foods. Since the surface area of inland waters is about 2% of the total area [40], the concentration of cesium-137 in freshwater fish and the total intake of cesium-137 via freshwater fish would then be 50 times greater than in the situation in which the activity released in uniformly deposited over the total surface of the land and inland waters.

The assessment of freshwater foods is based on the cesium-137 concentrations measured in carnivorous fish representing the highest trophic level [20]. The concentration averaged over the entire harvest from all freshwater ecosystems could be expected to be lower. In contrast to the intake of cesium-137 from the terrestrial diet, the intake via freshwater fish has a strong dependence

TABLE II. INGESTION OF CESIUM-137 VIA TERRESTRIAL AND AQUATIC FOOD CHAINS FOLLOWING A DEPOSITION $F(0) = 1$ mCi/km^2.

Pathway	Rate of Consumption	Intake Integrated to 30 yr (μCi)	Intake Integrated to Infinity (μCi)
1. Milk	0.6 liter/day	2.1×10^{-3}	3.0×10^{-3}
2. Terrestrial Diet			
(a) Estimated from milk		6.6×10^{-3}	1.0×10^{-2}
(b) Based on Danish data [6]		$9.1 - 9.6 \times 10^{-3}$	$9.1 - 9.8 \times 10^{-3}$
3. Freshwater Fish	1 kg/yr		
(a) Activity uniformly deposited on land and water		5.5×10^{-4}	9.8×10^{-4}
(b) Total activity transported directly to inland waters, i.e., deposition = 50 F(0)		2.7×10^{-2}	4.9×10^{-2}
4. Marine Fish	7 kg/yr		
(a) "Limiting" case: sediments absent, runoff instantaneous, no exchange		1.5×10^{-3}	3.0×10^{-3}
(b) "Intermediate" case: sediments and normal runoff but no exchange		1.2×10^{-4}	3.1×10^{-4}
(c) "Representative" case: sediments, normal runoff, exchange		2.0×10^{-4}	2.5×10^{-5}

TABLE III. INFINITE-TIME TOTAL INGESTION OF CESIUM-137 AND WHOLE-BODY DOSAGE TO THE POPULATION VIA TERRESTRIAL AND AQUATIC FOOD CHAINS RESULTING FROM THE RELEASE OF 1.5×10^6 Ci OF CESIUM-137

	To Average Individual		To Population	
Pathway	Intake (μCi)	Dosage (millirem)	Intake (Ci)	Dosage Man·Rem
1. Milk, 0.6 liter/day	0.57	35	170	1.1×10^7
2. Terrestrial Diet:				
(a) Derived from evaluation of milk	1.9	120	570	3.5×10^7
(b) Based on Danish data [6]	1.8	110	540	3.3×10^7
3. Freshwater Fish, 1 kg/yr				
(a) Uniform deposition on land and water	0.19	12	9.3	6×10^5
(b) Total activity transported to inland waters	9.3	570	470	2.9×10^7
4. Marine Fish, 7 kg/yr*				
(a) "Limiting" case	0.54	33	160	9.9×10^6
(b) "Intermediate" case	0.056	3.4	17	1.0×10^6
(c) "Representative" case	0.0045	0.28	1.4	8.4×10^4

[*] As explained in the text and noted in Table II, the "limiting" case is characterized by absence of sediments and exchange and by instantaneous runoff. The "intermediate" case is characterized by absence of exchange, but presence of sediments and normal runoff. The "representative" case is the expected situation, characterized by presence of sediments and occurrence of normal runoff and exchange.

on the long contributions from the cumulative deposit, which accounts for almost 90% of the total intake. The long-term contribution for the case where all the activity is delivered to inland waters is undoubtedly an overestimate, since we have assumed that the effective decay of the deposit is given by the radioactive half-life. A significant fraction of the aquatic activity would be transported to the ocean in runoff and be immobilized in sediments. Thus the terrestrial pathway is shown to be the dominant food-chain pathway.

The total activity that will reach man via food chains is computed by summing the appropriate population intakes: the intake from the terrestrial diet, the intake from the freshwater foods following uniform deposition on land and water surfaces, and the intake from marine foods in the "intermediate" situation where there is no exchange of water with waters of the deep ocean. In this scheme, the activity released is at all times accessible to the population. The total population intake is computed to be

$$570 + 9 + 17 \cong 600 \text{ Ci}$$

TABLE IV. TOTAL POPULATION INTAKE AND WHOLE-BODY DOSAGE FROM THE RELEASE OF 1.5×10^6 Ci OF CESIUM-137

	Intake (Ci)	Dosage Man·Rem
1. Terrestrial foods	570	3.5×10^7
2. Freshwater foods	9.3	5.7×10^5
3. Marine foods	17	1×10^6
TOTAL	600	3.7×10^7
TOTAL per Ci released	4.0×10^{-4}	25

Expressed as a fraction, the total population intake is about 4×10^{-4} of the 1.5×10^6 Ci released. The corresponding population dose, computed using the conversion factor 6.1×10^{-2} rem/μCi, is 3.8×10^7 man·rem. When expressed as a fraction of the curies released, the population dose via food chains is 25 man·rem per curie released. Results are summarized in Table IV.

Analysis of Man·Rem by Pathway Models

A. Description of Pathway Models

We have developed models for food-chain pathways for estimating dose to man [10]. These deterministic models are designed for general applications. As needed, we supply further detail on environmental conditions, agricultural practices, seasonal variations, etc., to refine the models.

A brief description of the model which is applicable to the forage-cow-milk, the pasture-meat-man and lichen-caribou-man pathways is presented below. All the food chain models are designed with a similar approach and only specific detail pertinent to each pathway change.

The basic model to describe the dynamics involved in a food chain are similar; parameters involved such as retention time for nuclides in vegetation, soil and water, elimination rates in man, intake rates for man, etc., change depending upon the pathway. A description of the continuous-deposition model for the forage-cow-milk pathway will serve as an example. Letting R be the deposition rate of a nuclide, the equilibrium deposition on pasture $F_{eq.}$ is

$$F_{eq.} = R/\lambda_P$$

where

R = deposition rate, $\mu Ci/m^2 \cdot day$

λ_P = effective elimination constant for plant, day^{-1}

$F_{eq.}$ = deposition at equilibrium, $\mu Ci/m^2$.

The concentration of the nuclide in milk is determined from the expression

$$\frac{dC_M}{dt} = F(t)\ (UAF)\ f - \lambda_M C_M$$

where

C_M = Concentration of the radionuclide in milk, μCi/kg

$F(t)$ = deposition, $\mu Ci/m^2$

UAF = utilized area factor, the effective area grazed by the cow in m^2/day. If K is the kg/day of forage needed by the cow and W is the plant density (kg/m^2) of the agricultural area; then (UAF) = K/W in m^2/day

f = the fraction of isotope ingested reaching milk, kg^{-1}

λ_M = effective elimination constant for milk, day^{-1}

t = time, days.

The equilibrium concentration in milk is given by

$$C_M = \frac{R(UAF)f}{\lambda_P\lambda_M} = \frac{R(UAF)f_M}{\lambda_P}$$

where

$f_M \equiv f/\lambda_M$ is the transfer coefficient to milk, that is, the fraction of nuclide ingested daily that is secreted in milk per liter or per kg (in day/liter or day/kg).

The radionuclide concentration in man is obtained from

$$\frac{dC_B}{dt} = \frac{C_M\ I\ f_B}{m} - \lambda_B C_B$$

where

C_B = concentration of nuclide in organ of reference, $\frac{\mu Ci}{g}$

I = average daily intake of milk, kg/day or liter/day

f_B = fraction of isotope reaching organ of reference

m = mass of organ, g

λ_B = effective elimination constant in man day^{-1}.

The concentration of the nuclide in man at equilibrium is

$$C_B = \frac{R(UAF)f_M\ I\ f_B}{\lambda_P\ m\ \lambda_B}\ .$$

The dose rate is given by

$$D_R = 1.85 \times 10^4\ E\ C_B$$

where

D_R = dose-rate, rem/yr

E = effective disintegration energy in MeV.

The coefficient relates nuclide concentration in μCi/g to dose-rate in rem/yr.

Similar procedures are used to model other pathways. In general, these models do not consider second order variables such as length of time between milking, slaughter, or fish catch and consumption of the product by man. Delay in consumption varies considerably between countries, regions of a country, and pathway and can, for the shorter lived isotopes, affect the radioactivity ingested by man. We take these basic models and perform such refinements as dictated by the specific situation we are called upon to consider. We also include refinements depending on the season of the year. Seasonal-variation affects such parameters as the amount of fresh forage and the amount of stored feed consumed by animals; depending on the type of storage the stored feed may be completely uncontaminated.

Food-chain models used for evaluation of the problem (see Fig. 1) include: soil-root-vegetation, vegetation-man, forage-cow-milk, forage-beef, and aquatic food chains. Space does not permit further discussion of details on these models; however, they have been described elsewhere [10,11,41,42].

B. Man•Rem Dosages

Dosages are calculated for each pathway and man·rem estimates are made using the same set of assumptions and population characteristics used in the previous parametric analysis. Highest man•rem dosage occurs through the forage-cow-milk and the forage-beef pathways (see Table V). The plant-man pathway, including leafy vegetables and grain products, contributes 14% of the total man·rem. The soil-root pathway, contributes only 2% to the total which would appear in all foods of the terrestrial diet. Aquatic pathways contribute insignificantly to the total man∘rem.

TABLE V. COMPARISON OF THREE ESTIMATES OF THE Man · Rem per Ci FOR CESIUM-137

Method	Man·Rem/Ci		
Parametric analysis of worldwide fallout data	25		
Calculations from total diet and fallout	9		
Food-Chain Pathways	Man·Rem	$\frac{\text{Man·Rem}}{\text{Ci}}$	Percent of Total Food Chain
Cow-milk-man	16×10^6	10.7	37
Pasture-meat-man	20×10^6	13.3	46
Plant-man	6.0×10^6	4.0	14
Sea-food-man	0.78×10^6	0.52	1.8
Freshwater-man	0.2×10^6	0.15	0.5
Soil-root-man	0.9×10^6	0.60	2.1
Σ All pathways	44×10^6	29	

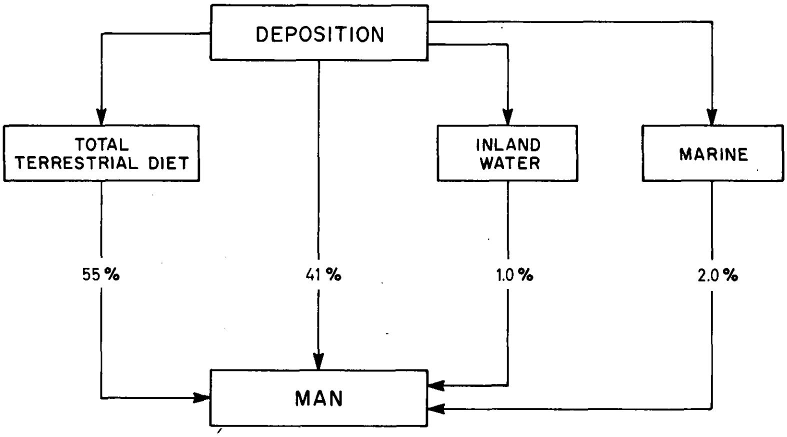

FIG. 2. Man-rem from parametric model showing per cent man-rem from each pathway.

RESULTS AND CONCLUSION

Results of the two calculational methods for determining man·rem are similar (see Table V). Of the 25 and 29 man·rem computed by the two methods, nearly all man·rem are accounted for in the terrestrial diet.

Good agreement was also obtained by comparison with mrem/mCi estimates made by Gustafson from fallout and diet data [17], where he determined that an annual deposition of 1 mCi of cesium-137/km^2 results in an internal dose of 0.24 mrem/yr from fallout occurring in the growing season. Using again a land mass of 7.8×10^6 km^2 the direct curie input to the food-chain is 7.8×10^3 Ci for 0.24 mrem average dose commitment. For the 3×10^8 people in the U.S. in 2000, one estimates 9 man·rem/Ci by this method.*

Total man·rem estimates include external exposures as well as food-chain pathways. The dose received by direct exposure to cesium-137 deposited on soil can be estimated by

$$\frac{1.44 \text{ rad}\cdot\text{km}^2}{\text{Ci}} \times \frac{3 \times 10^8 \text{ persons}}{7.8 \times 10^6 \text{ km}^2},$$

giving 54 man·rem/Ci. The $\frac{1.44 \text{ rad km}^2}{\text{Ci}}$ conversion factor does not consider shielding but does include a depth distribution of cesium-137 in soil [43]. If the shielding factor of 0.32 recommended by UNSCEAR is used, which includes shielding by buildings, time spent indoors and conversion from air dose to tissue dose, then the man·rem per Ci is 17. By comparison, if the shielding factor is included, external exposure contributes about the same man·rem as the food-chain pathways (see Figs. 1 and 2 and Table VI). Total $\frac{\text{man}\cdot\text{rem}}{\text{Ci}}$ estimates range from 26 to 46, or approximately 7×10^7 man·rem, which is about 2 times the man·rem from natural background in the United States.

Inhalation, either during the initial input of cesium-137 or from resuspension, can be shown to result in small contributions to the total man·rem. For the hypothetical uniform release over one year, average air concentrations would be approximately 5 pCi/m^3, confining the cesium-137 to an air space 100

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*The 25 man·rem calculated from analysis of fallout data purposely includes a substantial long-term contribution from soil.

TABLE VI. Man•Rem AND Man Rem per Curie: EXTERNAL GAMMA PLUS FOOD CHAINS

	External Gamma	Parametric Analysis of Fallout	Food Chain Pathway Models	Measurements
Man•rem	8.1×10^7	3.7×10^7	4.4×10^7	--
$\frac{\text{Man}\cdot\text{rem}}{\text{Ci}}$	17	25	29	9
Total man•rem per Curie (external + food chain)	--	42	46	26

meters over the United States. Calculations show this cesium-137 concentration in air to result in approximately 0.3 man•rem/curie, which is about 0.4% of the total man•rem. Submersion dosages are two orders of magnitude lower. Resuspension dosages can be estimated from measurements of fallout at Livermore, Ca. [44] by assuming that current air concentration (2×10^{-3} pCi/m^3) are derived from resuspension. This tends to overestimate the importance of resuspension, since part of the atmospheric burden is from current fallout. The top centimeter of soil contains approximately 15% of the soil burden of some 4×10^4 pCi/m^2. These data produce a resuspension factor of 2×10^{-7}/m. Upper limit resuspension air levels would be approximately 0.04 pCi/m^3, giving 0.002 man•rem/Ci. The calculated resuspension factor, we emphasize, constitutes an upper limit estimate.

In reality, actual releases of fission products would not result in a uniform contamination of the environment. There are 4 extreme sub-cases to be analyzed, each case involving an evaluation of $\frac{\text{man}\cdot\text{rem}}{\text{Ci}}$ for the independent system. Releases in urban areas would result in the highest man•rem because of the high population density coupled with the relative importance of external exposures. Man•rem/Ci is directly proportional to the contaminated land area x the population density of the area. For perspective, the $\frac{\text{man}\cdot\text{rem}}{\text{Ci}}$ from external exposure can be estimated for two geographical locations, distinctly different in population density, but of comparable land areas. The land area of Kansas is comparable to that of the projected Atlantic coast megalopolis. By 2000, one might project a population of 130 million in this population corridor, and one of about 5 million in Kansas. The man•rem/Ci attributable to external exposure from a uniform deposition on those areas would be 20-30 times higher in the megopolis than in a low-population density area such as Kansas.

For food-chains, the man•rem depends on transfer rates to the foods, and the unit-area food production rate. For any food-chain, man•rem from any release will be proportional to the amount of surface area which is contaminated, assuming a uniform rate of food production. For the aquatic systems, release to the freshwater system is estimated to result in 19 man•rem/Ci. The comparable result for the marine ecosystem is 7 man•rem/Ci. Contamination of the agricultural environment would result in approximately 45 man•rem/Ci from foods.

These results can be used to examine the alternatives available in power plant siting and waste management from the standpoint of potential dose to man. The trade-off involved can be weighed systematically in terms of costs per man·rem potential versus actual costs incurred in optimizing site selection. It should be stated, however, that cesium-137 would likely be accompanied by other radionuclides in any release, certainly strontium-90 and fissionable materials, and possibly radioiodines. Assessments of these radionuclides are being carried out.

In terms of planned releases as part of licensed waste management, it is clear that releases to aqueous systems are preferable to releases through the atmosphere from a man·rem point-of-view. Point-source releases in real systems are subject to similar evaluations. In such cases, regional and local considerations can be made, and man·rem models developed for decision-making purposes regarding point-of-emission source strength content and measurements necessary to verify compliance with regulations.

The fission product inventory in 2000 will be managed effectively to insure containment and minimal population exposure to radiation. The role of such models and evaluations is to provide guidance so that techniques and policies can be developed in a timely manner, contributing to an optimization of waste management.

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ESTIMATION OF ENVIRONMENTAL CAPACITY

(Sessions VIII and IX)

Chairmen:

V.D. TUROVSKIJ (USSR)
R. ICHIKAWA (Japan)

Invited paper

RADIOLOGICAL LIMITS AND ENVIRONMENTAL CAPACITIES

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Abstract

RADIOLOGICAL LIMITS AND ENVIRONMENTAL CAPACITIES.

In the last three decades weapons testing and nuclear power programs have added artificially produced radioactive materials to the natural inventory in the environment. Figures for the total activity released are unhelpful in deciding to what extent the environment has been polluted. The most realistic and useful definitions of man-made pollution refer only to those changes that man makes in his environment that are in some way detrimental. The most satisfactory measure of any detriment due to environmental radioactivity is the radiation dose to living tissue.

The expression 'environmental capacity' is used to mean that if a defined mixture of radionuclides is introduced into a particular sector of the environment over a prolonged period an equilibrium situation will eventually be reached. Practical experience has shown that with rare exceptions the radiation dose to man is the over-riding consideration and the capacity is said to be the rate of release that will result in a dose rate to man equal to some appropriate recommended limit. These dose limits imply a certain degree of risk and the assumed risks inherent in the current limits are examined in the context of the natural radiation background and the natural incidence of cancer and gentically transmitted disease. If the concept of environmental capacity is to be useful quantitatively it must be unambiguously related to dose. The 'limiting capacity' is based entirely on the ICRP Dose Limits; the 'stipulated capacity', which describes the total of all releases actually approved, includes other factors, social, economic and political, which may have the effect of restricting releases to a level below that of the limiting capacity.

1. INTRODUCTION

The management of artificially produced radioactive materials on a large scale began about three decades ago and since that time radionuclides have been released to the environment in effluents from installations in the nuclear fuel cycle and the many processes which involve applications of radioisotopes; there have also been releases as a result of nuclear weapons testing. These releases represent a man-made addition to the global inventory of natural radioactivity. In the oceans alone it has been estimated that the natural activity content is 500,000 MCi, mainly due to ^{40}K, and that weapons testing and nuclear power programmes have so far added some hundreds of megacuries and a few hundred kilocuries respectively. Such figures are unhelpful in deciding to what extent the environment has been polluted.

The most realistic and useful definitions of man-made pollution refer not to all changes which man makes in his environment but to those changes which are in some way detrimental. The detriment may be to man's health, to his food and other resources, to opportunities to pursue his activities or to the aesthetic qualities of his surroundings. The most satisfactory measure of any detriment due to environmental radioactivity is the radiation dose to living tissue, expressed in rads, or dose-equivalent expressed in rems.

The expression "environmental capacity" in the context of radioactive waste disposal has become widely used in the last three or four years only; its meaning is not self-evident and there is at present no internationally agreed definition. It is most often used to mean that if a defined mixture of radionuclides is introduced into a particular sector of the environment over a prolonged period an equilibrium situation will eventually be reached. Under these conditions the concentrations of radioactivity in the different components of the environment are supposed to be sensibly constant and the radiation dose-rate to living organisms, including man, can be estimated. Practical experience of waste disposal operations have shown that with rare exceptions the radiation dose to man is the over-riding consideration and the capacity of the sector of the environment to receive radioactive waste is said to be the rate of release which will result in a dose-rate to man equal to some appropriate national or international recommended limit (see Sect.2). This general description of the common meaning of environmental capacity could not serve as a formal comprehensive definition; such a definition would have to make provision for other factors, such as the identification of a representative exposed individual or population group and the meaning of the expression when applied to very long-lived nuclides which will accumulate in the environment for as long as they are released. The methodology underlying the concept of environmental capacity is not new; it has been applied systematically in the United Kingdom since the studies in the early 1950's of the liquid radioactive waste discharges into the Irish Sea from Windscale. The terminology in the UK has been different and the expression "Derived Working Limit" (DWL) has been used extensively /1/ to mean a rate of release of radionuclides which will lead to doses to critical groups of the population equal to the Dose Limits for members of the public recommended by ICRP.

The setting of dose limits is fundamental to the estimation of environmental capacity. These limits may be either internationally agreed limits such as those of ICRP which are intended to apply to radiation doses received from all sources other than the natural background and medical procedures or national limits which are set specifically for application to radioactive waste disposal. There are some circumstances, however, in which a useful alternative measure of environmental capacity is a comparison of additional doses due to radioactive waste disposal with doses due to natural background radiation, or to the natural variations in background, rather than with dose limits.

2. THE RELATIONSHIP BETWEEN DOSE AND RISK

The criteria by which any changes in the levels of radioactivity or radiation in the environment can be judged have recently been reviewed by Marley /2/ and the following paragraphs include summaries of material and conclusions from that review.

2.1 ICRP Dose Limits for Individuals and Local Populations

In its recommendations for the control of doses to the public the ICRP has distinguished between individual persons and whole populations or large sections of populations. The Commission has stressed that it is necessary to consider not only the magnitude of individual risks but the number of persons exposed since the total burden of somatic and genetic dose in any population under consideration may justify the effort required to achieve further limitation of exposure. The ICRP Dose Limit for individuals is set at 0.5 rem/y for exposure of the gonads, blood-forming organs or the whole body for individuals; for local populations it is recommended that the dose should be calculated by identifying an appropriate critical group, small enough to be homogeneous with respect to those factors such as age, diet and those aspects of behaviour that affect the doses received. The group should be representative of those individuals in the population expected to receive the highest dose from each particular source and the Commission's Dose Limit for individuals should be applied to the mean dose to this group. This procedure is used in many countries as a method of evaluating the significance of the dose to limited groups of people exposed to radiation as a result of the discharge of radioactive effluents.

2.2 ICRP Dose Limits for Large Populations

The ICRP has recommended that the genetic dose to the population, that is the average annual gonad dose of the whole population multiplied by the mean age of child-bearing, should be kept to the minimum amount consistent with necessity and should certainly not exceed 5 rems per generation (i.e. 30 years) from all sources other than natural background and medical procedures. The equivalent annual dose-rate of 170 mrem/y is often a feature of discussions on radiation exposure standards. The Commission points out the importance of ensuring that no single type of exposure should take up a disproportionate share of the total but leaves it to national authorities to decide the apportionment between different sources of exposure. In the United Kingdom 1 rem/generation has been fixed as the upper limit for exposure due to the disposal of radioactive wastes.

2.3 Doses due to Natural Background

The most recent estimates of the dose-rate to man due to the natural background in "normal areas" have been provided by UNSCEAR /̲3̲7̲ and are given in Table I. The dose-rates to the gonads, bone-lining cells and bone-marrow are about 90 mrad/y and these tissues are thought to be the most significant in terms of the risk of biological effects from ionising radiation. Local variations in background dose-rate are commonly of the order of 20-30 mrad/y and regional variations may be a factor of three (and in a few areas ten) higher than the 90 mrad/y quoted here as typical.

2.4 Risk Estimates

UNSCEAR has recently re-estimated the risk of malignant disease following large doses of radiation /̲3̲7̲. These estimates, summarised in Table II, were based in part on observations on the survivors of Hiroshima and Nagasaki and included findings up to the year 1970 reported by Jablon and Kato /̲4̲7̲. The figures represent the excess incidence of malignancy per million man-rads for individuals exposed as adults and include all cases identified in the period 1945-1970. They are quoted as ranges because the incidence varies with the level of dose received. The figures for "other cancers" include some 50 cases of thyroid and breast cancer which are not necessarily associated with short life expectancy. Marley has used these figures to estimate the risk over a whole life-time expressed in terms of tissue rather than air dose and has also adjusted them to allow for the greater effectiveness of neutron doses in relation to gamma doses. There is no direct evidence of the likely future

TABLE I. DOSE-RATES DUE TO NATURAL SOURCES IN 'NORMAL' AREAS

Source of irradiation	Dose-rates (mrad y^{-1})		
	Gonads	Bone-lining cells	Bone marrow
External irradiation			
Cosmic rays	28	28	28
Terrestrial radiation	44	44	44
Internal irradiation	21	20	17
TOTAL	93	92	89

Ref: United Nations Scientific Committee on the Effects of Atomic Radiation, 1972.

TABLE II. JAPANESE ATOMIC BOMB SURVIVORS: EXCESS INCIDENCE OF MALIGNANT DISEASE 1945-1970

Type of Malignancy	Number of Cases per 10^6 man-rads (air dose)
Leukaemia	15 - 40
Other cancers	90 - 130
Total	105 - 170

Derived from: United Nations Scientific Committee on the Effects of Atomic Radiation 1972.

incidence of cancers other than leukaemia in the Japanese survivors and Marley thought it reasonable to double the figures found in the first 25 years. The incidence of leukaemia started to fall after an average latency period of the order of ten years and he increased these cases by a smaller factor. His conclusion is that the life-time risk of all malignancies may be about 250 cases per million man rads including 40-50 cases of leukaemia. These figures are based upon data for the exposure of adults but after reviewing the data for children he concluded that present evidence does not warrant a different figure for the overall incidence of malignancy.

Exposure to background radiation and environmental radioactivity from waste disposal generally occurs chronically in the range 0.1-0.5 rad/y whereas the doses to the Japanese survivors were acute and amounted to at least a few tens of rads. UNSCEAR drew attention to the evidence from animal experiments that the yield of tumours per unit dose should be lower when the doses are low and protracted. Marley concluded that an overall risk of 100 fatal malignancies per million-rads was a reasonable figure for the assessment of environmental problems such as those due to radioactive waste disposal and it represents an upper limit to the incidence which would be expected in practice.

Estimates of the incidence of genetic damage following irradiation are derived from animal experiments, particularly experiments with mice because human data are not available, but UNSCEAR considered that the results of these experiments can now be used with some confidence to calculate effects in human populations. The natural incidence of genetically transmitted disease in humans is about 30,000 cases per million (Table III) live-births. UNSCEAR concluded that this incidence would be increased by about 1% i.e. 300 cases per million live births for every rad received by the parental generation under conditions of chronic exposure. Only about 20 of these 300 additional cases will occur in the immediate descendants of the irradiated parents; the rest will be spread over many generations.

These risk coefficients for genetic and somatic injury can be used to estimate the upper limit of real biological damage associated with the low doses of chronic radiation such as those due to background and radioactive waste disposal (Table IV). In the United Kingdom deaths from all forms of cancer occur at the rate of 2,300 per million per year. Marley's estimate of 100 cases per million man-rem suggest that a background dose-rate of the order of 0.1 rad/y could account at most for only ten of these 2300 cases i.e. less than 0.5%. The birth-rate in the United Kingdom is about 15000 per million and in this figure are about 500 cases of severe genetically transmitted disease. The UNSCEAR risk coefficient of 1% per rad per generation suggests that background radiation is responsible for not more than 15 (i.e. 3%) of these cases. The conclusion from this kind of analysis seems to be that natural background radiation is responsible for a very small proportion of disease in man and that exposure of populations to levels of man-made radiation which are, on average, not very much higher than background cannot result in marked increases in the incidence of either somatic or genetic injury in those populations.

TABLE III. INCIDENCE OF GENETICALLY INDUCED SEVERELY HANDICAPPING DISEASES

Natural incidence	30,000 per million live births
Excess incidence per rad per generation	< 300 " " " " (at equilibrium)

Derived from: United Nations Scientific Committee on the Effects of Atomic Radiation 1972

TABLE IV. ESTIMATED EFFECT OF BACKGROUND RADIATION IN UNITED KINGDOM

All deaths from malignant neoplasms	2300 per million/year
Incidence due to background radiation (0.1 rad/y)	<10 " " "
All cases of genetically induced severely handicapping disease	500 per million/year
Incidence due to background radiation (0.1 rad/y)	<15 " " "

Derived from ref. /2/

At the ICRP Dose Limit of 0.5 rad/y these risk coefficients imply a statistical additional risk of cancer of 1 in 20,000 for every year of exposure and an additional risk of 1 in 100,000 for first generation genetic damage in the children of the exposed individual; this application of the coefficients to estimate risks to individuals is a tenuous extrapolation of the available scientific evidence but clearly indicates that the risks are small compared with the ordinary risks of life.

3. "LIMITING" AND "STIPULATED" ENVIRONMENTAL CAPACITIES

The present convention of assuming a positive relationship between dose and risk, irrespective of dose-rate or level of dose, implies that even when a sector of the environment is uniquely defined there can be no rate of introduction of radioactivity which can be guaranteed to produce zero injury in man. It follows that if the concept of environmental capacity is to be useful quantitatively it must be unambiguously related to dose; if different dose criteria are applied to different situations the term environmental capacity must be qualified appropriately. A panel under the chairmanship of Polvani convened by the IAEA in 1970 suggested a consistent terminology /5/. Although this panel was concerned with the principles for limiting the introduction of radioactive waste into the sea the terminology is more generally applicable. The term "limiting capacity" of a sector of the environment was used to mean the rate of introduction of radionuclides which would result in doses equal to ICRP Dose Limits. The calculation of limiting capacity is based entirely on scientific factors, e.g. critical path methodology, and no safety factors are deliberately introduced into the calculation. The panel recognised that there are other factors, some being scientific (e.g. ecological) but others being social, economic and political, which may have the effect of restricting releases of radioactive wastes to a level below that of the limiting capacity. The term "stipulated capacity" was used as a term to describe the total of all the releases actually approved for release into a sector of the environment. The stipulated capacity cannot exceed the limiting capacity without the risk of exposing individuals or critical groups to doses exceeding ICRP Dose Limits. The term "provisional environmental capacity" has been used /6/ to describe estimates of limiting capacity in which safety factors have been introduced because assumptions or uncertainties in data leave some doubt about the validity of the calculations.

This separation of scientific and socio-political considerations was considered to be the approach least likely to cause confusion to both scientists and laymen. The Panel gave as an example those countries in which the establishment of allowable rates of release to local environments based upon the observed habits of the local population might not be favoured. The argument for this view might be that the growth of nuclear power will eventually result in a proliferation of sources of radiation exposure; drinking water and foodstuffs consumed by any population group might then contain radioactivity from several sources. If the concentrations of radionuclides in all water and food are kept below safe limits then the general population is protected independently of the proliferation of sources of waste or the dietary habits of particular population groups. In these circumstances no credit is taken for the fact that a particular critical foodstuff may constitute only a small fraction of the total dietary intake. This is a more restrictive approach than the use of the critical pathway - critical group concept in which there is a systematic evaluation of all routes by which radionuclides are returned to man and the relative contribution of each route to total dose. The Panel argued that although the more restrictive approach might well be appropriate for the estimation of stipulated capacity it should not be used for the estimation of actual doses received or for establishing limiting capacity.

4. DOSE ASSESSMENT

Techniques for estimating the relationship between release rates of radioactive effluents and dose to man are described comprehensively at this symposium and elsewhere /7,8/; only a few general observations are appropriate in this paper.

Information about the initial composition of the waste may be of great importance. A knowledge of the identity of the nuclides which are present in the wastes is essential since different radionuclides may have widely differing toxicities and exhibit profoundly different behaviour in the environment. This behaviour may also be influenced by the physical and chemical form of wastes at the time of release. The mode of release may have a considerable effect on doses to local populations. Limiting environmental capacity is most often determined by the dose to a local critical group and stack height, in the case of discharges to atmosphere, or the positioning of a liquid effluent outfall may be of great importance. The transport and dilution of wastes after release may usually be calculated from numerical models suited to the scale of the environment under consideration.

Critical pathway and critical group methodology depend upon the identification of a critical group which may be real and defined by means of a population habit survey; alternatively a hypothetical critical group may be postulated with extreme habits in which case the estimated doses are likely to exceed those actually received. If a real and large local exposed population is involved it may not be easy to calculate or judge the "mean dose" to the critical group if no distinct sub-group with exceptional habits can be clearly identified. When the exposed group is very small assessments may have to be based on the most highly exposed individuals. It must always be kept in mind that both effluent composition and population habits may change with time and review procedures are essential.

An alternative to critical path analysis for dose assessment is the concept of "maximum specific activity" which was considered by the Polvani panel /5/. The starting point is the ICRP recommended limit for the organ or body burden of a particular radionuclide. The concentration of the stable analogue in the same organ is determined and the ratio of the two concentrations used to calculate the limiting specific activity in the medium into which waste is released and then, on the basis of a suitable environmental model, to the limiting specific activity in the waste itself. The method is attractive for isotopes of those elements which may be under homeostatic control in the body so that radionuclide retention is more readily related to the rate of intake of the stable analogue than to the rate of intake of the radionuclide itself. The method has frequently been applied to the tritium content of water and with some modification to ^{90}Sr, using stable calcium as an analogue. The Polvani panel, however, pointed out that the validity of the method depends upon the rapid establishment of equilibrium between the released radionuclide and the various forms of the same element in the environment. In the context of marine disposal the panel also concluded that the approach assumes a knowledge of trace element concentrations and biological availabilities in the marine reservoir which does not currently exist for the majority of elements of interest in waste disposal; it is also inapplicable to external dose problems and the assessment of dose to the gastro-intestinal tract.

5. ENVIRONMENTAL SCALE

Description of environmental capacity in terms of release rates associated with ICRP or equivalent national dose limits and critical path analysis seems well suited to the consideration of individual releases which lead to doses to small population groups within national boundaries. Within countries a consistent system of national dose standards can be expected. In the majority of cases somatic doses to local populations will be the limiting factor. Even if the exposed population is relatively large and the doses which it receives have a significant genetic component, the contribution to the total population genetic dose of the country concerned can be assessed and if necessary regulated in accordance with the policy of the national competent authority. Further, it is within the jurisdiction of national competent authorities to ensure that the total of all stipulated capacities, in the sense described in Section 3, do not exceed the limiting capacity of a particular sector of the national environment. These tasks are much simplified when the environment into which releases are made are bounded by topographical or geological features as is the case when discharges are made into rivers, lakes and into the ground. International rivers and lakes are a special case which involves both national and regional interests.

When releases are made into the atmosphere the environmental scale to be considered may be determined by the radioactive half-lives of the nuclides involved.

Atmospheric transport processes are rapid, and although nuclides with half-lives of the order of a few hours may be considered in a local or national context only, nuclides with half-lives of the orders of days and years, if released in significant quantity, must also be assessed in terms of regional and global consideration.

The dispersion of radionuclides in the sea is heavily dependent upon chemical identity and physico-chemical form. Releases from coastal sites may be of regional interest in the case of the longer-lived mobile nuclides while disposal into the deep ocean has long been recognised as having international significance. The mobility of tritium in the atmosphere and the marine environment and the importance of this nuclide in fission and possible future fusion power programmes has already resulted in much study at the international level.

The definition and application of the concept of environmental capacity as environmental scale increases are matters which are still to be resolved. The limitation and apportionment of genetically significant dose to regional and global populations cannot be agreed on a national basis. Many scientists involved in studies of environmental radioactivity on such a scale compare both the genetic and somatic doses which they derive with natural background radiation levels and with present nationally and internationally recommended dose limits as the best method presently available for illustrating their importance. In the context of somatic dose, the ICRP has already envisaged the possibility that a "somatically significant population dose limit", which will presumably be set at a lower level than the corresponding Dose Limits for individuals, might be a useful concept when there is sufficient knowledge and experience to determine such a figure [9].

It will be clear from Section 2 that detriment, or strictly potential detriment, from environmental radioactivity is best expressed in terms of risk to individuals and populations. In the case of populations the risk is conveniently and conventionally expressed in terms of the possible incidence of disease or death per unit of integrated population dose (i.e. man-rads). A review paper of this nature would not have been complete without a reference to the role which risk expressed in that form might play as the concept of environmental capacity is further developed.

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DEVELOPPEMENT ET APPLICATION DU CONCEPT DE LA CAPACITE RADIOLOGIQUE EN RADIOPROTECTION

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Abstract

DEVELOPMENT AND APPLICATION OF THE RADIOLOGICAL CAPACITY CONCEPT IN RADIATION PROTECTION.

The proliferation of nuclear facilities involves an ever larger output of radioactive wastes, a small part of which is discharged to the environment in accordance with regulations laid down by the authorities responsible for the protection of public health and the environment. The presence in these wastes of long-lived radionuclides, and the fact that this contamination extends over an area frequently going beyond national frontiers, have led to the need for strict calculation of the resulting radiation doses to man, particularly in the international context (international rivers and the seas and oceans). The radiological capacity of a hydrobiological basin is a fundamental concept which should be developed and applied to the problem of radiation protection. This makes it necessary to take account of all the processes of transfer and distribution by which the radioactivity reaches man. Examples of the application of the concept confirm its interest for radiological protection.

DEVELOPPEMENT ET APPLICATION DU CONCEPT DE LA CAPACITE RADIOLOGIQUE EN RADIOPROTECTION.

La multiplication des installations nucléaires entraîne une production de plus en plus importante de déchets radioactifs dont une faible partie est rejetée dans l'environnement suivant les prescriptions établies par les responsables de la protection de la santé publique et de la sauvegarde de l'environnement. La présence, dans ces rejets, de radionucléides à longue période, et l'influence de cette contamination, qui se manifeste dans un espace dépassant souvent les frontières nationales, conduisent à la nécessité d'un calcul rigoureux des doses de rayonnement qui en résultent pour l'homme, notamment si l'on se réfère à l'échelle internationale (fleuves internationaux, mers, océans). La capacité radiologique d'un bassin hydrologique est une notion fondamentale qu'il convient de développer et d'appliquer pour ce problème de radioprotection. Ceci nécessite la prise en considération de tous les processus de transfert et de distribution de la radioactivité dans son cheminement vers l'homme. Des exemples d'application confirment l'intérêt de cette procédure en radioprotection.

1. INTRODUCTION

La multiplication du nombre des installations nucléaires pose pour les pays à relativement forte densité de population des problèmes de plus en plus nombreux en ce qui concerne la gestion des déchets radioactifs solides, liquides et gazeux.

Deux grands principes sont appliqués pour le traitement de ces déchets:

- leur rétention sous une forme appropriée en un lieu adéquat,
- leur dispersion dans un milieu susceptible de les accepter sans qu'il en résulte des risques dépassant les prescriptions établies par les responsables de la Santé publique et de la sauvegarde de l'environnement.

Ces deux principes sont d'ailleurs quelque peu complémentaires en ce sens que le premier s'applique surtout aux déchets fortement et moyennement radioactifs et le second concerne les déchets faiblement radioactifs qui peuvent être rejetés (souvent néanmoins après un traitement de décontamination) dans l'atmosphère ou dans un milieu hydrobiologique.

Les prescriptions qui régissent les rejets sont établies à partir des recommandations ou directives formulées par des organismes internationaux (Commission internationale de protection radiologique, Euratom, etc.) auxquelles se réfèrent généralement les réglementations nationales. Ces normes d'irradiation permettent d'établir pour les radionucléides des concentrations maximales admissibles dans l'air et dans l'eau de boisson. Les formules de rejet, qui en découlent, ne prennent généralement en considération, à part la radiotoxicité évidemment, que la dilution atmosphérique ou hydraulique des effluents dans le milieu récepteur pour la détermination des quantités de substances radioactives à rejeter.

Or, dans un milieu naturel beaucoup de phénomènes autres que la dilution (dispersion ou diffusion) peuvent modifier de façon très significative la pollution de ce milieu, à la fois dans le temps et dans l'espace, comme par exemple l'adsorption sur des matériaux ou la concentration dans des organismes: il est donc indispensable d'en tenir compte si l'on veut évaluer objectivement les conséquences pour la santé publique d'un déversement de substances radioactives dans l'environnement. Ceci doit permettre de déterminer sur des bases scientifiques les quantités de radioactivité dont le rejet pourrait être autorisé sans qu'il y ait de conséquences néfastes d'ordre sanitaire et écologique.

La connaissance et la prise en considération de tous les processus de modification de la répartition de la radioactivité dans un milieu récepteur doit permettre une meilleure évaluation des risques d'irradiation de l'homme. Cette évaluation pourrait être basée sur l'établissement d'une quantité maximale de radioactivité admissible dans un réseau hydrobiologique sans que les niveaux d'irradiation qui en résulteraient pour l'homme puissent être supérieurs aux normes adoptées. Cette notion peut être définie comme la «capacité radiologique limite d'un milieu».

Ce concept avait été présenté, discuté et accepté au cours de réunions d'experts des six Pays Membres, organisées en 1965 et 1966 par la Direction Protection sanitaire de la Commission des Communautés européennes. Un document récapitulatif a été publié à ce sujet en 1969 [1].

Par suite une série de travaux ont permis de préciser cette notion et d'en promouvoir l'application dans différents domaines [2-5].

Le présent document donne d'abord un aperçu des caractères généraux de la capacité radiologique, puis présente trois exemples significatifs et enfin propose une analyse critique de la notion de capacité radiologique limite.

2. CARACTERES GENERAUX DU CONCEPT DE LA CAPACITE RADIOLOGIQUE

La détermination de la capacité radiologique limite d'un milieu est basée sur la prise en considération d'un certain nombre de paramètres importants:

- la qualité, la quantité et la fréquence des rejets de substances radioactives;
- les atteintes de l'homme par les éléments du milieu récepteur, l'irradiation de l'homme peut se faire par ingestion de substances radioactives ou par exposition externe aux rayonnements;
- les conséquences écologiques sur le milieu qui peuvent retentir indirectement sur l'homme, par exemple l'altération d'un écosystème.

De plus, il importe de déterminer les deux séries de processus suivants pour une évaluation des risques pour la santé publique:

- processus régissant les lois de la contamination des derniers maillons de la chaîne alimentaire de l'homme;
- processus concernant le transfert de cette contamination à l'homme.

L'évaluation de la première série de processus nécessite une étude écologique détaillée des mécanismes de transfert de la radioactivité déversée (dépositions, accumulations, élutions) dans les cycles biologiques naturels.

En ce qui concerne la seconde série de processus, son évaluation requiert les études suivantes en s'inspirant notamment des recommandations de la publication 7 du Comité 4 de la CIPR:

- établissement des données concernant le métabolisme de chaque radionucléide ingéré par l'homme, ce qui permet de définir sa radiotoxicité et, en considérant les normes de base relatives à ce radionucléide, conduit à la détermination du radionucléide critique; il est possible que dans certains cas on puisse caractériser d'ailleurs plusieurs radionucléides critiques;
- évaluation des processus relatifs à l'apport à l'homme de substances radioactives à partir d'un milieu contaminé: par exemple en ce qui concerne les habitudes alimentaires (quantité consommée, préparation des aliments); ceci permet d'établir la voie critique (ou les voies critiques) par laquelle (ou lesquelles) un radionucléide parvient à l'homme;
- détermination des groupes d'individus d'une population dont l'exposition aux rayonnements ionisants est sensiblement homogène et représentative de celle des individus les plus exposés dans cette population, c'est le groupe critique.

La juste appréciation de tous ces processus est difficile, mais nécessaire pour une évaluation objective des risques d'irradiation pour l'homme. En particulier la détermination des éléments de caractère économique, social et psychologique peut réclamer des enquêtes minutieuses sur les habitudes alimentaires[1] et une surveillance attentive des centres de production, des réseaux de distribution et des lieux de consommation.

Il faut souligner que le terme «critique» emprunté à la publication 7 du Comité 4 de la CIPR vise à donner aux éléments du milieu une qualification permettant la prise de décisions sur le plan de la protection sanitaire. Cette acceptation du terme «critique» ne comporte en elle-même aucune notion de danger. En particulier, on entend par groupe critique un groupe d'individus dont l'exposition aux rayonnements ionisants est sensiblement homogène et représentative de celle des individus les plus exposés d'une population. De même, on entend par voie critique la voie par laquelle le groupe critique peut être exposé aux rayonnements ionisants. Enfin la zone critique est l'aire géographique qui est le siège des processus de transport des radionucléides par la voie ou les voies critiques.

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[1] Dans quelques cas particuliers on peut même parler de régimes alimentaires critiques.

3. DETERMINATION DE LA CAPACITE RADIOLOGIQUE

Par suite de la variété et de la complexité des interactions entre les radionucléides et le milieu récepteur, la détermination de la capacité radiologique d'un site ne peut être effectuée sans se référer à des modèles mathématiques capables de fournir une description quantitative des multiples phénomènes en jeu.

En outre, il est nécessaire que les modèles choisis possèdent certaines caractéristiques, afin que le processus qu'ils décrivent ne reste pas un exercice académique élégant, mais présente un caractère éminemment pratique.

En particulier, il convient d'accorder l'exigence d'une représentation suffisamment fidèle de la réalité avec la nécessité d'éviter le recours à des artifices de calcul trop sophistiqués.

Dans de précédents travaux [2, 5], les auteurs du présent document, s'inspirant des critères ci-dessus, ont développé un modèle mathématique adapté à la définition de la capacité radiologique d'un certain milieu hydrobiologique lié aux eaux superficielles.

Ce modèle sera ici brièvement évoqué, avec l'exposition du processus suivi pour permettre la détermination de la capacité radiologique. Ensuite, on décrira le mode suivant lequel le même problème peut être affronté et résolu, aussi dans le cas de rejet de substances radioactives dans l'atmosphère, et dans le sol.

Il faut préciser que les considérations qui suivent ne tiennent pas compte des modalités de rejet des substances radioactives dans les divers milieux récepteurs étudiés. Le but des auteurs est en fait de développer une méthodologie applicable, soit aux décharges volontaires, soit au rejet involontaire de matériaux radioactifs.

Dans le premier cas, la méthodologie peut permettre par exemple de calculer les formules de rejet pour les effluents liquides et gazeux ou la capacité d'un site à être utilisé comme stockage de déchets solides. Dans le second cas, elle représente un moyen de définir et de mettre en œuvre les programmes de contrôle et de surveillance en matière de radioprotection.

3.1. Cas d'un rejet dans les eaux douces de surface

Le modèle proposé dans ce cas est reproduit à la figure 1.

Comme on peut le voir, le milieu hydrobiologique est divisé en trois systèmes partiels, correspondant respectivement

- au milieu récepteur primaire (milieu aquatique)
- au milieu récepteur secondaire végétal
- au milieu récepteur secondaire animal.

Chacun de ceux-ci comprend à son tour un certain nombre de compartiments.

Le passage d'une substance radioactive d'un compartiment à un autre est représenté à l'aide de procédés utilisés dans le calcul des systèmes à compartiments linéaires, ce qui, avec une bonne approximation, peut être appliqué aux compartiments que nous avons considérés.

Une approche semblable permet en principe de faire correspondre à chaque compartiment une équation différentielle du premier ordre qui décrit tous les transferts de substances radioactives d'un compartiment à l'autre.

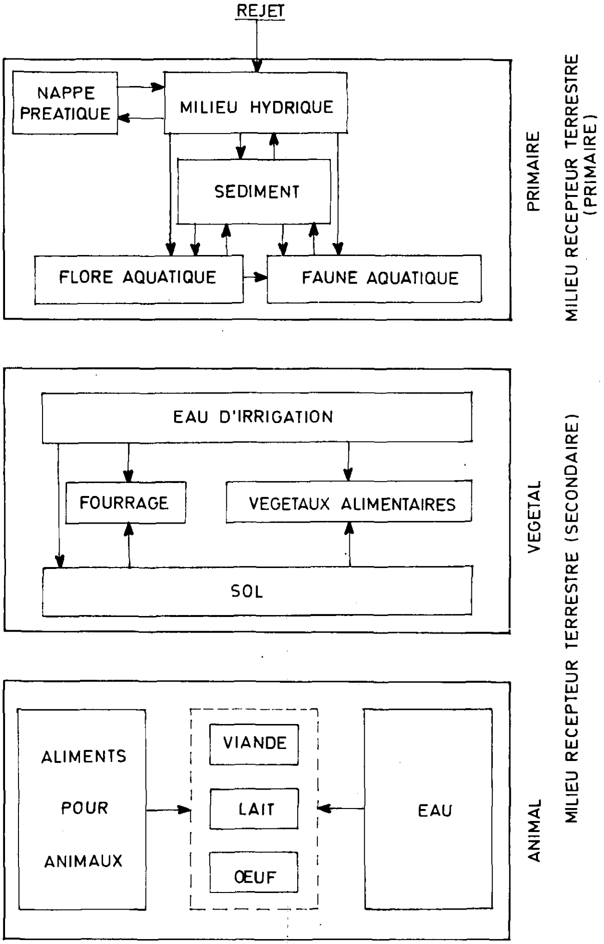

FIG. 1. Modèle de milieu hydrobiologique récepteur (eaux douces superficielles).

Evidemment, la résolution numérique du système d'équations différentielles est possible si l'on connaît les valeurs à attribuer aux diverses constantes de transfert, ou d'élimination, des substances radioactives relatives au compartiment considéré.

Or de telles valeurs ne sont pas toujours connues; dans ce cas il est possible de décrire le comportement des substances radioactives à l'intérieur d'un compartiment ou d'un groupe de compartiments au moyen de fonctions empiriques déterminées expérimentalement ou à l'aide d'observations effectuées sur des milieux analogues à celui qui est étudié.

Le système d'équations différentielles une fois résolu, on obtient les valeurs des concentrations à un instant donné dans les divers compartiments pour un certain radionucléide introduit d'une façon ou d'une autre dans le milieu considéré. Sur la base de ces informations, il est possible de caractériser, comme nous le préciserons par la suite, les éléments critiques du milieu (groupe critique, voie critique, zone critique ...).

Pour pouvoir caractériser les éléments critiques, une fois établies les concentrations d'un certain radionucléide dans chaque compartiment du milieu récepteur, il convient d'effectuer dans ce domaine des examens et des enquêtes pour obtenir une connaissance analytique complète soit des divers composants de chaque écosystème intéressé, soit de la composition des populations et de leurs habitudes de vie (alimentation par exemple).

En pratique on observe que, dans la majeure partie des cas[2], la voie critique des expositions aux risques des rayonnements ionisants est constituée par la chaîne alimentaire. De ce fait une attention spéciale doit lui être consacrée.

Dans une telle optique, il n'est pas seulement nécessaire de caractériser les principales espèces végétales et animales du milieu, mais il est aussi indispensable d'effectuer des déterminations quantitatives sur chaque espèce (par exemple production) en distinguant la partie destinée à la consommation locale et celle destinée à un marché plus vaste. De ceci on peut déduire la zone de distribution qui, si elle est très limitée, entraîne l'augmentation du risque individuel pour cette population soumise à une éventuelle contamination, tout en diminuant le nombre des personnes exposées aux risques de la contamination.

En déterminant qualitativement et quantitativement ainsi les consommations de denrées alimentaires, les modes de vie des groupes particuliers de population liés d'une manière quelconque au milieu récepteur, et en connaissant les valeurs des concentrations dues aux rejets unitaires de chaque radionucléide, on peut calculer les expositions qui dérivent de ces rejets unitaires pour de tels groupes de population.

On pourra qualifier de critique pour chaque radionucléide le groupe qui correspond à l'exposition la plus élevée calculée de cette sorte. Il faut dire que ce n'est pas toujours que l'on obtient une valeur nettement supérieure à toutes les autres. Il est aussi possible que, pour plusieurs groupes, nous obtenions des valeurs également élevées d'exposition ou d'irradiation. Dans de tels cas on parlera de plusieurs groupes critiques (pour un radionucléide particulier). En fait il est illusoire d'établir une différence entre des valeurs qui ne sont pas nettement distinctes entre elles (distinctes d'au moins un ordre de grandeur), étant donné l'incertitude associée à l'évaluation de plusieurs paramètres utilisés pour calculer ces valeurs.

Une fois le groupe critique (ou les groupes critiques) établi(s), on pourra déterminer sans équivoque la voie critique correspondante.[3] La zone critique sera enfin la zone géographique dans laquelle est située chaque voie critique mentionnée ci-dessus.

Ces définitions de voie et de zone critiques sont évidemment très importantes pour, d'une part, établir la capacité radiologique et, d'autre part, définir et mettre en œuvre le programme de surveillance et de contrôle.

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[2] Il existe néanmoins certains problèmes d'irradiation externe par les rejets de gaz rares radioactifs.

[3] Pour ceci les mêmes considérations pourraient être développées que pour le groupe critique: par la même manière on peut arriver à caractériser éventuellement pour un même radionucléide plusieurs voies critiques.

En fait ce contrôle et cette surveillance peuvent être effectués sur une seule partie du milieu récepteur, partie qui est le siège des phénomènes les plus importants de propagation des radionucléides.

Tout ce qui précède se réfère, comme il a été dit, à un radionucléide particulier. Evidemment en cas de rejets de plusieurs radionucléides, il convient de répéter pour chacun d'eux le processus entier déjà décrit. Il est possible que le résultat de cette opération se traduise par l'établissement de groupes critiques distincts en totalité ou en partie.

Si dans le milieu aquatique il n'existe ni faune ni flore comestible, et s'il n'y a aucune utilisation pour l'irrigation de l'eau contaminée en aval du point de rejet des substances radioactives, il n'est pas nécessaire d'effectuer de vastes enquêtes écologiques, et la concentration maximale admissible pour l'eau potable peut être considérée comme facteur limitatif.

Une fois individualisé le groupe critique, on peut calculer la quantité maximale de chaque radionucléide particulier qui peut être introduit dans l'environnement sans que les individus de ce groupe soient soumis à des valeurs de dose d'irradiation supérieures aux normes de radioprotection (rejet annuel admissible ou dose limite annuelle). En fait nous aurons:

$$Q_{max} = \frac{\text{rejet maximal admissible annuel}}{\text{rejet correspondant à l'émission unitaire}}$$

$$Q_{max} = \frac{\text{dose limite annuelle}}{\text{dose correspondant à l'émission unitaire}}$$

La valeur Q_{max} est la capacité radiologique pour le radionucléide considéré.

3.2. Cas d'un rejet dans l'atmosphère

Quand les substances radioactives sont rejetées dans l'air, elles sont dispersées dans des volumes plus ou moins grands en fonction de la capacité de dilution de l'atmosphère, dépendant essentiellement des conditions météorologiques locales. L'étude de la distribution des radionucléides dans l'atmosphère peut être effectuée à partir de la formule de Sutton [6] et de Pasquill [7], et suivant des variantes plus ou moins élaborées de ces méthodes [8, 9].

Ensuite une détermination simple, basée sur la vitesse de retombée et sur les facteurs de «wash out», permet de passer au calcul de dépôt au sol de la contamination corpusculaire.

Il est alors possible de déterminer la répartition dans le temps et dans l'espace de la concentration de substances radioactives soit dans l'atmosphère, soit sur le sol.

Ceci permet, en dernière analyse, d'évaluer quantitativement et qualitativement la pénétration des contaminants particuliers dans les divers compartiments reliés directement à l'atmosphère: eaux superficielles, terrains, végétaux comestibles, fourrages, animaux.

A partir de ce moment, le problème ne se différencie pas de celui étudié dans le paragraphe 3.1 et il est possible de la résoudre de la même manière, avec l'unique différence que, cette fois, les rejets ne sont plus effectués en un point précis d'un milieu hydrique par exemple, mais ils intéressent directement plusieurs compartiments envisagés à la figure 1.

3.3. Cas d'un rejet dans le sol

Dans le cas d'un rejet dans le sol de substances radioactives, on peut se trouver devant l'une des deux situations suivantes:

- les substances radioactives passent dans la circulation hydrique souterraine,
- les substances radioactives restent hors de contact avec la nappe phréatique.

Dans le second cas, le terrain constitue un compartiment fermé (cas de formations géologiques particulières: dépôts salins ou couches argileuses) pour lequel le problème de l'évaluation de la capacité radiologique se transforme en celui de la détermination de la capacité physique, dépendant de divers paramètres en fonction des objectifs considérés (volume utile, dissipation de la chaleur, etc.).

Dans le premier cas, au contraire, il existe en principe la possibilité que les radionucléides atteignent la nappe phréatique, et qu'ils se dispersent dans le milieu extérieur. Etant donné, malgré tout, que le mouvement des radionucléides peut, dans des conditions déterminées, être extrêmement lent, le compartiment «nappe phréatique» doit être étudié en détail dans son caractère interne. Ce qui le différencie des cas examinés dans les précédents paragraphes, c'est l'intervalle de temps pour atteindre un état d'équilibre. Ici le temps est souvent très long, largement supérieur à plusieurs années.

La nécessité de prévoir les mouvements de la contamination au sein de la nappe phréatique a conduit à l'élaboration de théories et de modèles plus ou moins sophistiqués et capables de donner dans quelques cas particuliers des informations intéressantes [10-13].

Sans entrer ici dans une discussion sur la valeur de tels processus, on peut observer, par suite de la grandeur des temps de transfert, que la nappe phréatique est soit un compartiment fermé soit, pour quelques cas précis, un compartiment ouvert en communication avec le milieu extérieur.

Dans la première hypothèse, il est possible d'utiliser les mêmes considérations que celles déjà faites à propos des formations géologiques absolument imperméables. Dans la seconde hypothèse, le problème se ramène à celui du paragraphe 3.1.

4. CONSIDERATIONS CRITIQUES SUR LE CONCEPT DE LA CAPACITE RADIOLOGIQUE

Le choix d'un site d'implantation d'installations nucléaires est motivé par des considérations politiques, économiques, sociales, sanitaires et écologiques. Il est a priori difficile d'établir une gradation entre ces considérations. Il est généralement de mise de réaliser un certain compromis entre tous ces paramètres. Pourtant il est impératif d'accorder une importance particulière aux considérations de caractère sanitaire.

Dans le problème particulier des rejets de substances radioactives, l'impératif sanitaire est de ne pas entraîner un risque supérieur à celui accepté par les réglementations sur les débits de doses admissibles pour la population. Encore faut-il que l'évaluation du risque couru soit faite d'une façon aussi objective que possible. Ainsi le fondement de la politique sanitaire, en ce qui concerne la pollution radioactive comme en ce qui concerne tous les autres types de pollution, doit porter sur l'évaluation objective des risques, et sur la connaissance scientifique des phénomènes.

La détermination scientifique de la capacité radiologique limite d'un milieu récepteur est une recherche essentielle pour cette évaluation objective des risques auxquels l'homme et son milieu pourraient être soumis. Cette approche de nature socio-écologique se différencie de tous éléments subjectifs tels que peuvent l'être notamment des considérations d'ordre économique (qui dépendent de l'importance des ressources d'un pays, d'une région ou d'une collectivité) ou des contraintes d'ordre technologique (fonction du développement industriel).

De tout ceci il ressort que les autorités responsables de la radioprotection doivent rechercher, établir et imposer une méthodologie scientifiquement acceptable qui conduise au respect des normes sanitaires imposées. Cette méthodologie peut être celle de l'établissement de la capacité radiologique quant à ses principes et ses objectifs.

L'entité à considérer est le site récepteur (hydrobiologique ou atmosphérique) et non l'installation nucléaire particulière. Le principe reconnu par tous de l'intérêt de la protection de la santé publique associé à celui de la préservation du patrimoine naturel nous oblige à la prise en considération d'une manière fondamentalement prioritaire de la capacité d'un site à recevoir des effluents radioactifs. Comme dans toutes les décisions concernant le monde moderne, il y a en adoptant ceci un risque à courir, mais les normes de radioprotection en ont précisé l'importance et en ont fixé la limite supérieure. Mais si cette dernière ne doit pas être dépassée elle ne doit pas non plus être nécessairement atteinte: la dose d'irradiation pour la population ou pour le groupe critique de population doit être aussi faible que possible.

Le concept de capacité radiologique permet une vue globale des rejets de plusieurs installations nucléaires, vue globale essentielle en ce qui concerne les problèmes de radioprotection. Dans le domaine des rejets d'effluents radioactifs, il rend possible les études prospectives relatives à l'établissement de centres nucléaires, donc le planning motivé du nombre et de l'emplacement des installations. Il est souvent difficile de faire l'intégration des conséquences des rejets de plusieurs installations où pour chacune d'elles, tout en restant au-dessous des normes de radioprotection, on applique le principe de décharge «as low as practicable» ou «achievable» (aussi bas qu'il est faisable), qui introduit des considérations économiques et sociales. La certaine relativité de critères comme le coût social par exemple peut entraîner beaucoup de difficultés lorsqu'on se place dans un cadre plus vaste, par exemple celui d'un bassin hydrobiologique récepteur.

Il faut d'ailleurs souligner que pour des problèmes de bassins hydrobiologiques internationaux, la notion de capacité radiobiologique constitue un élément de référence objectif et acceptable par les scientifiques au plan international. Cela ne présume pas malheureusement des discussions quant à la répartition de cette capacité à la fois dans le temps et dans l'espace. Mais le point de départ de la discussion est précis et intangible, ce qui constitue toujours un élément important dans une confrontation.

C'est pourquoi les autorités chargées de la radioprotection, donc responsables des problèmes de santé publique et d'environnement, ne peuvent qu'être résolues à établir et à développer les principes de la capacité radiologique d'un milieu récepteur de façon à posséder une base scientifique de référence quant à l'évaluation objective des risques d'un rejet de substances radioactives. De ce fait, elles seront à même de prendre des décisions motivées quant à la qualité et la quantité des substances radioactives à rejeter. Il est d'ailleurs possible d'envisager parfois une certaine complémentarité de la capacité radiologique avec le principe de rejet «as low as practicable», dans l'optique de l'intégration de cas particuliers dans un problème général.

La capacité radiologique peut être la base concrète réaliste qui apporte à la solution du problème des rejets d'effluents radioactifs un élément positif, sans arbitraire. Car si on a établi des normes d'irradiation et si on les a acceptées, il est scientifiquement possible, sans trop d'artifices de calcul ou de raisonnement, de définir, pour un site donné, sa capacité à recevoir une certaine quantité de polluants sans que les normes d'irradiation soient dépassées.

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OBSERVED VERSUS THEORIZED CAPACITY OF THE ENVIRONMENT FOR RADIOACTIVE CONTAMINANTS

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Abstract

OBSERVED VERSUS THEORIZED CAPACITY OF THE ENVIRONMENT FOR RADIOACTIVE CONTAMINANTS.

The potential radiation exposure of people usually establishes the quantity of radionuclides that are acceptable in the environs of nuclear installations. Estimates of the potential doses that may occur are based upon many predictions and assumptions, including the behaviour of the radionuclides in ecosystems, the production and harvest of edible products and the dietary, occupational and recreational habits of people. To ensure that radiation doses will not exceed prescribed limits, assumptions used are conservative and the degree of conservatism is inversely related to the confidence associated with an assumption.

Because of the conservative nature of the assumptions and other imperfections in the dose calculation process, the radiation exposure actually received by people in the vicinity of an operating nuclear installation may differ substantially from early estimates. Comparisons between predicted and actual doses are possible where radioactive contaminants have been found in readily measurable quantities in the environs of large nuclear complexes (such as the USAEC plutonium plant in the State of Washington) and some nuclear power stations. In most cases the observed concentrations of radionuclides in critical foodstuffs and in people have been substantially less than predicted. The best agreement is with simple, direct exposure pathways such as drinking water. Poor agreement is associated with complex food chains, irregularly consumed foods and short-lived nuclides. Major sources of error are associated with bio-accumulation factors, market dilution, dietary assumptions and seasonal variations.

Conservative assumptions on the capacity of the environment should always be used as a basis for plant design and the setting of effluent standards. However, revised estimates of the dose actually received by people in the environs, and thus the capacity of the environment, should be made as soon as factual data become available.

INTRODUCTION

The capacity of the environment to accommodate radioactive materials should be based upon several types of criteria, including the potential effects of radiation upon ecosystems as a whole. However, at this stage in the development of the nuclear industry the only basic standards that are generally recognized are those that have been established for people[1-5]. For the time being, then, the recommended maximum doses for people must serve as the starting point for derivation of estimates of capacity. In essence, this appears to represent the position taken by the IAEA Panel on Procedures for Establishing Limits for Radionuclides in the Sea in respect to the assessment of "limiting" capacity[6].

Even though the basis for estimation of capacity is confined to the dose standards for humans, a further choice must be made. This involves a determination of whether to invoke the limit for individuals (or a small critical population) based upon exposures close to the point of radioactive waste disposal, or to invoke a lower limit that applies to some average population. A few years ago selection of the most restrictive case was relatively simple because the nuclear installations operating at that time released enough mixed fission products and/or neutron activation products so that the close-in dose (and thus the standard for individuals) was quite obviously the limiting situation. Further, the critical pathways were usually associated with the release of liquid wastes to rivers or to the sea[7-8].

Recently proposed rules now under consideration in the United States[9] and the diligent use of sophisticated radioactive waste cleanup equipment for nuclear power plants are modifying the former situation. Releases of radionuclides that tend to accumulate in nearby sediments and biota are expected to be negligible from the nuclear power plants now under design and construction. Rather, the small doses to people anticipated from such plants will most likely come from the ^{3}H and ^{85}Kr[10] that are discharged to the atmosphere. Even in this situation, however, it appears that the close-in dose to members of the public will continue to be more restrictive than the average dose to populations[6,11].

As the future installations with their very low releases are placed in operation, there will be increasing reliance on theoretical calculations as the basis for estimation of the radiation dose to people. It will be impractical or too expensive to make accurate measurements in the field of the incremental contribution of radioactive materials from each individual plant. Paradoxically, as we depend more and more heavily upon the theoretical estimates, our ability to determine from field measurements the reliability of those estimates for some critical individual or population will diminish.

PREDICTION OF DOSE

The factors that must be taken into account in order to predict the fate of radionuclides and then estimate the radiation dose to critical groups have been described by Slansky[6]. As demonstrated by this symposium, there are now many excellent research programs in progress throughout the world that are filling voids in the knowledge of the fate of radioactive materials released by nuclear installations; the results of this work will make substantial contributions to calculations of environmental capacity. Nevertheless, we still need verification of the dose models as a whole under conditions where known releases of radionuclides can be compared with measured concentrations that ultimately exist in places inhabited by people, and in the foods and beverages consumed by those people.

The immense amount of data on the deposition and fate of radionuclides injected into the atmosphere by the testing of nuclear devices is most useful, but in only a few places are good sets of data available on both chronic releases from point sources and the resulting doses to people. Such sets of data have been developed at the large nuclear installations in North America[7] and the United Kingdom[12] and also for some nuclear power stations[12-13]. The most extensive sets of data appear to be those associated with the U.K. Windscale plant and U.S. Hanford plant. In both of these cases, the quantities and varieties of radionuclides released to the environment have warranted large-scale programs for sampling and measuring the materials at key locations along the exposure pathways from the point sources of release to the critical populations. A paucity of comparable data for most U.S. nuclear power plants is associated with the relatively small quantities of radioactive materials released and the difficulty of measuring them once they have been dispersed in the environment.

Much of the U.S. power plant data is tabulated as gross activity, rather than in quantities of specific radionuclides[14]. Although this is quite sufficient to demonstrate compliance with regulations and the absence of any significant dose to the public, additional special studies, such as those undertaken by Kahn, et al.[13], for reactors and by Martin[15] for fuel reprocessing plants, are needed in order to develop the source-dose relationships.

Several assumptions must be used with the mathematical models that predict the dose to people in the vicinity of nuclear installations. These have been noted by the IAEA Panel on Procedures for Establishing Limits for Radionuclides in the Sea[6]. One of these assumptions involves the kinds and quantities of radionuclides that will be released. Inasmuch as the release rates are dependent upon the integrity of equipment, the efficiency of waste clean-up equipment and the performance of workers, these "source terms" are subject to substantial variations. The rates of release do not modify the capacity of the environment per se, but do, of course, determine what fraction of the capacity is used.

Another assumption involves the amount of dilution that takes place in the atmosphere or hydrosphere between the point of release and the point of human use or exposure. Since we are usually concerned with exposures that are integrated over a period of a year, this parameter can be estimated quite accurately provided that adequate meteorological and hydrological studies have been carried out. Several of the most important exposure pathways, including the dose from submersion in air, inhalation, drinking water and swimming, require little extrapolation beyond this stage.

Additional assumptions are required for the ingestion of foods and it is in this area that estimates of the environmental capacity become especially complex. Even where it is possible to predict the relationships between the concentration of radionuclides in air and crops or water and fish, there remain the problems of-predicting (or determining) a) the quantities of the foods that are actually consumed by the critical individual or population, b) the seasons when they are consumed, c) the age and size of the consumer, and d) the number of different exposure pathways that impinge on the same people.

THE HANFORD OBSERVATIONS

For many years, the plutonium production reactors in the State of Washington discharged substantial quantities of radioactive materials to the Columbia River[16] and some of the gamma-emitters could easily be traced through a number of food chains to man[17]. All but one of these reators have now been shut down[16], but the block of data acquired over more than two decades provides an exceptional opportunity for comparison of dose pathway predictions with subsequent measurements that include whole-body counting.

The first estimates of the dose to people in the environs of the Hanford plant were reported in 1958 at the Second United Nations International Conference on the Peaceful Uses of Atomic Energy[18]. These early estimates were not characteristic of the preoperational predictions as are now made for nuclear power installations. At the time the first reactors started up (1944), the methodology for predicting radiation doses from exposure pathways was only in the embryonic stage. No significant releases to the environment had occurred, and the earliest experiments on transfer of radionuclides from soil or water to plants and animals had only just begun. Consequently, no preoperational dose predictions, as we calculate them today, were made for the Hanford plant.

Nevertheless, the 1958 dose estimates, which were based on environmental monitoring data, did help to establish the exposure pathways concept and methodology. At this same time the United Kingdom was using similar techniques to estimate the exposure and set discharge limits for the Windscale Works[19]. The authors of the 1958 dose estimates for Hanford[18] concluded that:

> "The over-all summation of results from an environmental survey program of this nature is complicated by the large numbers of possible sources of exposure and, recently, by the general prevalence of fallout isotopes. The best estimates of the actual exposures to people are still uncertain because of the wide variations possible in diet, occupancy and other factors. At the present levels the estimates are adequate to indicate low exposures to people but refinements of the technique are constantly being made so that improved values will be available."

The refinements have continued since 1958 as recounted by Corley[20] and Essig[21]. Some of the more significant refinements that have been incorporated into recent evaluations[22] are:

- Expression of all results in terms of dose (mrem/year) rather than as fractions of maximum permissible concentrations
- An upward revision of the quantity of water consumed by the maximum individual from "standard man" values to 730 liters per year (this was based on local surveys), but also the use of radionuclide concentrations in samples of drinking water rather than unprocessed river water
- Determination of intake and exposure data for the average local resident through the use of questionnaires and field surveys.
- Inclusion of external exposure from 500 hours per year of recreational activity along the banks of the Columbia River
- Inclusions of seafood as a dietary assumption for average Richland residents even though the distance to the Pacific Ocean is about 400 kilometers
- An upward revision in the estimated consumption of fish caught from the Columbia River by the maximum individual from about 4 kilograms of whitefish per year to 40 kilograms of panfish species (crappie, bass and perch)
- Inclusion of eggs as a dietary assumption when it was noted that the eggs of hens from some farms along the Columbia River contained radionuclides of Hanford plant origin
- Inclusion of game birds (pheasants and quail) as a dietary assumption when it was determined that the average Richland resident ate about 1.2 kilograms of such birds each year (For comparison the average consumption of Columbia River fish is assumed to be 0.48 kilograms per year)
- Inclusion of both ^{131}I and ^{133}I in drinking water as sources of dose to the thyroid.

A substantial part of the dose refinement program at Hanford has been associated with the eating habits of people and the whole-body counting of large numbers of people in the environs of the plant. It was recognized at the time of the early estimates[18] that fish caught locally from the Columbia River would constitute the critical exposure pathway for people who ate them. There were, however, no factual data on the numbers of fish of various species that were harvested, how many people ate them, or in which seasons of the year most of the fish were caught. Consequently, the early dose estimates had to be based upon the personal conjecture of people who were familiar with the habits of some of the local fishermen.

REVISION OF ASSUMPTIONS

The paucity of data on fish consumption prompted a census of the intensity of local fishing and the questioning of people about their eating habits. The first such census was carried out in 1961 and a more comprehensive census was carried out in 1965-1967[23] through the cooperation of the State of Washington Department of Game. These surveys showed some substantial variations from the early theorized dietary assumptions. For example, panfish species were consumed at a higher rate than were whitefish; the most popular fishing area was not close to the reactors, but many kilometers downstream; most of the fish were caught not in the autumn, but in the summer.

A few of the fishermen questioned thought they ate fish as many as 200 times each year and subsequently this unusually large rate of consumption was used to estimate the dose to the critical individuals. There is, however, good reason to question the validity of the 200 meal-a-year rate of consumption, which is derived from the personal estimates of the fishermen. On the basis of the responses to the questionnaire, the estimate of the number of fishing trips and the number of fish caught are both four times greater than can be accounted for by the actual counts of fishermen on the river[23]. Further, neither followup interviews at the homes of fishermen nor the results of extensive whole-body counting confirm a 200 meal-a-year rate of consumption.

When the plutonium production reactors were operating, Columbia River fish sampled downstream from the project contained ^{65}Zn in easily measurable concentrations. Because of this, two special experiments were undertaken to determine the relationship between known rates of intake of ^{65}Zn by people who consumed fish and their resulting body burdens of ^{65}Zn[24,25]. In one of the experiments, a sustained intake of about 10 kilograms per year (50 meals) led to a maximum body burden of about 130 nCi ^{65}Zn[24]. By ratio, a person consuming 40 kilograms (200 meals) of such fish would be expected to have had a body burden of about 500 nCi ^{65}Zn. However, no body burdens approaching such a figure were found among avid fishermen, members of the general public, or plant workers who were counted between 1959 and 1964. The highest ^{65}Zn burden found was that of the man who ate the fish experimentally. The next highest values (3 people) were slightly more than 100 nCi, but since these were plant workers, the ^{65}Zn could well have been from their occupation rather than from their diet[17]. These data suggest that the theorized dose for the critical group of people, attributed to the consumption of 40 kilograms of fish each year, was quite likely overstated by a factor of at least four.

Combining whole-body counting with questionnaires on dietary habits proved to be a highly effective means of developing good data on exposure pathways and the results have been reported in several papers by the late John Honstead[26,27]. These data showed good agreement between the availability of ^{65}Zn in drinking water and the average ^{65}Zn body burdens of the people who regularly drank the water[26]. The agreement between the concentration of ^{24}Na in drinking water and the ^{24}Na body burdens was less consistent. Because of the short half-life (15 hr) of the ^{24}Na, the concentrations of this nuclide fluctuated with transit time of the water in the river and municipal systems, with day-to-day variations in amount of water drunk, and with the elapsed time between a drink and the whole-body count.

Data supplied by school children[21] indicates that the average child drinks about 0.4 liters of water per day and that the maximum intake is not apt to be more than 0.8 liters. However, adults who drink many cups of coffee and tea consume about 2 liters of water per day[28]. The difference in the quantity of water consumed by the average and maximum individual is relatively small in comparison with the differences in quantities for other dietary items. Consequently, dose estimates associated with drinking water will probably be more accurate than those associated with other pathways.

Where complex food chains are involved, the dose prediction is subject to additional sources of error. Through use of the whole-body counter we have been able to bypass some of these errors and to quantify others. In one special study, Honstead[27] obtained both dietary estimates and whole-body counts on a small population living on farms irrigated with water from the Columbia River. The ^{65}Zn body burden was predicted for each person on the basis of his recollection of what he ate and the content of ^{65}Zn in his water and foods as determined from the environmental monitoring program. This prediction was then compared with the ^{65}Zn body burden as measured by the whole-body counter. In over 50% of the people the measured body burden of ^{65}Zn was less than half of that predicted on the basis of the dietary and monitoring data. In a very few individuals, however, the measured body burden was two to four times greater than the predicted value.

In 1970 and 1971 somewhat similar comparisons were made between estimated ^{65}Zn intake and measured body burdens for people who live on the Pacific Ocean near the mouth of the Columbia River[29]. For the 1970 survey the average measured-to-calculated ratio was between one and two--a remarkably good result. However, a few individuals had about ten times the predicted body burden. In 1971 the average body burdens were two to four times higher than calculated and again a few students had burdens as much as an order of magnitude greater than calculated.

The relative agreement reported here between the calculated and measured body burdens of critical population groups should not be viewed as representative of a precision that will exist in the vicinity of all nuclear installations. Only one nuclide (^{65}Zn) is considered and some of the uncertainties associated with predicting its concentrations in foods have been eliminated by extensive sampling and radiochemical analysis of the foods. Were it not for the environmental monitoring data, it would also have been necessary to make estimates about dilution of the effluent, bioaccumulation factors and seasonal variations. Such estimates can, however, be made with remarkable success provided that adequate studies are made of the bioaccumulation factors and of the characteristics of the environs of the nuclear site.

CONCLUSIONS

The limiting capacity of the environment to accept radioactive materials can be predicted with reasonable accuracy in terms of specified permissible dose rates to man. However, the precision of the prediction depends not only upon how well the movement and fate of the radionuclides in the biosphere can be modeled, but also upon an accurate knowledge of the dietary and recreational habits of the critical populations. Several predictions have been made that turned out to be within a factor of 2 of the values measured in the field. In other cases (individual people rather than populations) the first estimates may have either understated or overstated the dose by an order of magnitude.

Even with improved methodology for predicting the relationships between release rates of radioactive materials and the dose to critical population groups, the first approximations of environmental capacity should be made with the use of assumptions that are sufficiently conservative to assure that the radiation doses will fall within desired limits. As soon as actual operating experience provides factual data, the tentative assumptions used at the outset should be refined. This refinement will aid in the validation of predictive models rather than in the perpetuation of false concepts through a reiteration of unconfirmed dogma.

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DISCUSSION

I. L. OPHEL: I am interested in the small number of individuals who contained more than ten times the predicted amount of zinc-65. Was any attempt made to find the exact reasons for the discrepancies?

R. F. FOSTER: The individuals who contained substantially more ^{65}Zn than predicted did not have unusual absolute body burdens of ^{65}Zn. The large difference between what was predicted and what was actually experienced seems to be associated with an underestimate of the intake of common foods containing ^{65}Zn.

I. HANDYSIDE: I have a question on the psychological impact of your whole-body monitoring program. Did you find that suspicions were aroused among the local population that something was wrong with the Handford Plant. What action did you take to assure such persons that this was not the case?

R. F. FOSTER: At first we were concerned that the public would be apprehensive about the condition of the plant if we asked people to be counted in the whole-body counter. Consequently the program was described in the press and television, school children and their teachers were given a one-hour lecture on radioactivity, both natural and artificial, and literature was sent home with the students. We found that there was absolutely no need for us to worry about the psychological impact. In fact, it turned out to be an excellent public information and relations program.

LA CAPACITE RADIOLOGIQUE DE L'ENVIRONNEMENT, BASE DES ETUDES PROSPECTIVES DE PROTECTION

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Abstract–Résumé

THE RADIOLOGICAL CAPACITY OF THE ENVIRONMENT AS A BASIS FOR FUTURE RADIATION PROTECTION STUDIES.

The health hazards arising from the normal effluents discharged by a nuclear facility are evaluated in terms of the irradiation dose to the most exposed population groups. Evaluation of these hazards requires a knowledge of the existing equilibrium relationships between the activity discharged to the environment and the dose to the members of the critical group. Since these relationships are assumed to be known, by assigning a fixed limiting value to the dose one can obtain the maximum activity of the corresponding discharge per unit time. This value is identified with what is called the "limiting radiological capacity of the receiving environment for the radionuclide considered". The limiting radiological capacity depends on a large number of factors associated with the characteristics of the radionuclide, the receiving environment and the populations concerned. It implies that the environment considered is homogeneous and in equilibrium with the radionuclide. The limiting radiological capacity of an environment can be evaluated at three levels: local level, for estimates concerning only a limited population group; regional level, for estimates concerning more extensive contamination (rivers and lakes); and international level, for long-term estimates on the assumption of generalized contamination of the planet (atmosphere and oceans). The relationship between the estimates of discharge and the limiting radiological capacity of the environment considered provides the means of evaluating the level of protection of populations in comparison with the basic standards.

LA CAPACITE RADIOLOGIQUE DE L'ENVIRONNEMENT, BASE DES ETUDES PROSPECTIVES DE PROTECTION.

Les risques sanitaires résultant des effluents normaux rejetés par une installation nucléaire sont évalués en termes de dose d'irradiation délivrée aux groupes de population les plus exposés. L'évaluation de ces risques implique la connaissance des relations qui existent à l'équilibre entre l'activité rejetée dans le milieu et la dose délivrée aux individus du groupe critique. Ces relations étant supposées connues, en donnant à la dose la valeur limite fixée on obtient l'activité maximale du rejet correspondant par unité de temps. Cette valeur s'identifie à ce qu'on appelle la «capacité radiologique limite du milieu récepteur pour le radionucléide considéré». La capacité radiologique limite (CRL) dépend de nombreux facteurs, liés aux caractéristiques du radionucléide, du milieu récepteur et des populations concernées. Elle implique que le milieu considéré est homogène et en équilibre avec le radionucléide. La CRL d'un milieu peut être évaluée: à l'échelle locale pour des prévisions n'intéressant qu'un groupe limité de population; à l'échelle régionale pour des prévisions concernant une contamination plus étendue (fleuve, lac); à l'échelle mondiale, pour des prévisions à long terme dans l'hypothèse d'une contamination généralisée de la planète (atmosphère terrestre, océans). Le rapport entre les prévisions de rejet et la CRL du milieu considéré permet d'apprécier le niveau de protection des populations par rapport aux normes de base.

INTRODUCTION

La capacité d'acceptation d'un milieu récepteur pour un polluant donné a été définie [1] comme «la quantité de ce polluant que le milieu récepteur peut recevoir en un point donné, d'une façon continue sans qu'il en résulte à terme de nuisances appréciables pour l'homme et son environnement».

Cette notion repose sur l'existence d'un équilibre entre les apports continus à débit constant du polluant et son élimination par tous les processus possibles. Elle représente une caractéristique du milieu au point considéré, fonction de son potentiel de dilution et d'autoépuration d'une part, de son utilisation par l'homme d'autre part.

Pour un polluant radioactif, la capacité d'acceptation du milieu est appelée « capacité radiologique». Elle s'exprime alors en activité par unité de temps. Dans ce cas, la décroissance radioactive vient s'ajouter aux autres processus d'élimination et il peut arriver aussi que ce soit le seul processus à considérer. Par définition, la capacité radiologique exprime une limite au-delà de laquelle la nuisance n'est plus considérée comme acceptable. Le critère de base pour l'appréciation du risque radioactif étant la limite de dose fixée pour les individus du public, il en résulte que les rejets d'effluents radioactifs dans le milieu considéré, pour être acceptables, doivent être limités de telle façon que la dose d'irradiation délivrée à l'organe critique des individus du groupe critique reste inférieure à la limite de dose. Connaissant la relation qui existe à l'équilibre entre l'activité rejetée dans le milieu et la dose délivrée aux individus du groupe critique, si l'on donne à la dose la valeur limite fixée par les autorités responsables, il est possible d'évaluer l'activité maximale du rejet correspondant par unité de temps. Cette valeur s'identifie alors à ce que l'on peut appeler la « capacité radiologique limite» ou CRL du milieu récepteur pour le radionucléide considéré [2].

La CRL d'un milieu récepteur pour un polluant radioactif donné dépend essentiellement de cinq séries d'éléments:

a) la limite de dose fixée pour l'organe critique
b) les paramètres biologiques et, éventuellement, nutritionnels de l'individu moyen du groupe de population critique
c) les paramètres écologiques de transfert du milieu à l'homme
d) les paramètres intrinsèques du milieu récepteur qui déterminent à l'équilibre la concentration du polluant dans ce milieu
e) les caractéristiques du polluant lui-même, en particulier sa forme physico-chimique et sa période de décroissance.

Suivant l'étendue du territoire sur lequel s'exerce la nuisance et l'importance de la population qui en subit les effets, la capacité radiologique peut être évaluée à l'échelle locale, régionale ou mondiale.

1. EVALUATION DE LA CRL A L'ECHELLE LOCALE ET REGIONALE

L'exposition de la population locale peut résulter, soit de l'irradiation directe, soit de l'inhalation d'air contaminé, soit, enfin, de l'ingestion de produits alimentaires contaminés. Mais le plus souvent, l'exposition de la population déborde le cadre local et prend une ampleur régionale. Dans certains cas, le milieu récepteur, fleuve, grand lac, par exemple, est suffisamment étendu pour intéresser la population d'une région, et même parfois prendre un caractère international. Dans d'autres cas, les produits agricoles contaminés localement peuvent ne pas être consommés exclusivement par la population locale, mais être distribués, après mélange avec des produits d'autres régions, et consommés par une population plus importante. Il en résulte alors une certaine dilution dont il pourrait être tenu compte pour l'évaluation de la CRL.

Dans tous les cas, la première étape du calcul conduisant à la CRL consiste à établir les équations exprimant la dose annuelle D délivrée à l'individu moyen des groupes de population intéressés, en fonction du paramètre u symbolisant le niveau de contamination du milieu récepteur. Ces équations sont de la forme:

$$D = f_c\,(u) \qquad (1)$$

f_c est un facteur de conversion que l'on peut appeler facteur de dose. La voie critique et le groupe de population critique sont déterminés par l'équation qui maximalise D pour une même valeur de u, c'est-à-dire par les paramètres partiels qui maximalisent f_c.

Dans les cas d'exposition par irradiation directe et inhalation, l'évaluation de f_c est relativement simple et ne fait intervenir que les paramètres biologiques de l'homme et les caractéristiques du radionucléide. Les paramètres liés au site n'ont pas à être pris en considération. Par contre, lorsque l'atteinte de l'homme résulte de l'ingestion de produits alimentaires contaminés, la valeur de f_c dépend, d'une part, des caractéristiques du site, d'autre part, des habitudes alimentaires de la population. Les équations (1) prennent alors la forme:

$$D = \sum q_i\, f_i\,(u) \qquad (2)$$

où q_i est la quantité d'aliments I consommés annuellement par l'individu moyen du groupe considéré et f_i (u) est la dose élémentaire délivrée à l'organe de référence pendant l'unité de temps par la quantité de radionucléide contenue dans l'unité d'aliment i, exprimée en fonction du paramètre u.

Les paramètres qui entraînent la plus grande variabilité de D pour une même valeur de u sont l'âge des individus et leurs habitudes alimentaires, et les différents facteurs de transfert du milieu aux produits alimentaires parvenus au stade de la consommation. Ces paramètres sont très nombreux et il est le plus souvent illusoire de vouloir en tenir compte de façon exhaustive. En général, on se contente de retenir quelques paramètres essentiels qui eux-mêmes intègrent un grand nombre de paramètres secondaires.

Deux cas principaux se présentent.

1.1. Cas d'une pollution atmosphérique

Pour l'évaluation de la contamination des végétaux, on prend le plus souvent en considération quatre paramètres:

a) La vitesse apparente de dépôt, c'est-à-dire le rapport

$$V_g = \frac{\text{activité déposée par unité de temps et par unité de surface}}{\text{concentration du radionucléide dans l'air}}$$

Cette vitesse de dépôt varie dans d'assez larges limites. Pour le dépôt de l'iode sur l'herbe par exemple, cette variabilité peut atteindre un facteur 100 suivant la forme physico-chimique de l'iode et un facteur 10 suivant la densité de l'herbe et la vitesse du vent.

b) Le paramètre de transfert air-végétal, exprimant la fraction du dépôt retenue dans la partie comestible du végétal. Ce paramètre peut varier de quelques pour-cent à 25 ou 30%.

c) Le paramètre de transfert sol-végétal, exprimant la fraction du dépôt accumulé dans le sol présente dans la partie comestible du végétal au moment de la récolte. Ce paramètre varie dans d'assez larges limites en fonction des caractéristiques du sol, de celles du végétal, des techniques agricoles, etc.

d) Le coefficient d'élimination du radionucléide, à partir de son dépôt au sol, dont dépend l'activité du sol à l'équilibre.

Pour les produits animaux, il faut tenir compte en outre des facteurs de passage nourriture-produits animaux, qui, généralement, varient assez peu d'une région à l'autre.

Revenons à l'expression (1). Supposons que l'on ait déterminé pour le site étudié la valeur maximale de f_c; en donnant à D la valeur de la dose limite, on obtient la concentration limite X_l du radionucléide dans l'air.

Pour passer de la concentration limite X_l à la CRL, on utilise la relation suivante:

$$Q = \frac{X}{\bar{d}} \qquad (3)$$

où Q est le débit du rejet, X est la concentration moyenne dans l'air au niveau du sol, et $\bar{d}$ est le facteur moyen de dilution pondéré, qui tient compte de la distance du point considéré à la cheminée, de la hauteur de dispersion des panaches, de la vitesse du vent et d'autres conditions météorologiques.

Ayant déterminé sur le site les zones où X est maximal, en donnant à X la valeur X_l obtenue précédemment, on obtient:

$$\mathrm{CRL} = \frac{X_l}{\bar{d}} \qquad (4)$$

ou

$$\mathrm{CRL} = \frac{D_l}{\bar{d} \times f_c} \qquad (5)$$

Finalement, la CRL dépend essentiellement de deux facteurs globaux:

a) Le facteur de dilution pondéré $\bar{d}$ qui intègre un certain nombre de paramètres météorologiques liés au site; l'expérience montre que pour l'ensemble de ces paramètres et pour des conditions de climat et de relief assez homogènes, l'influence du site est relativement faible et qu'il existe une plus grande dispersion dans les résultats propres à un même site qu'entre les résultats moyens des différents sites [3].

b) Le facteur f_c, qui dépend de nombreux facteurs liés au site, parmi lesquels les facteurs intrinsèques du sol, son utilisation agricole, la transformation et la distribution des produits, et, enfin, les habitudes alimentaires des populations jouent un rôle prépondérant.

Pour l'iode-131, par exemple, la pratique de l'élevage de vaches laitières sur des pâturages situés dans la zone critique, et la consommation de lait frais par la population locale, constituent des circonstances aggravantes susceptibles d'entraîner un abaissement de la CRL de deux à trois ordres de grandeur par rapport à un site où l'élevage des vaches laitières n'est pas pratiqué.

Dans le cas où l'autorité responsable doit faire un choix entre plusieurs sites, l'évaluation de la CRL pour chacun de ces sites peut être alors un critère intéressant à considérer.

1.2. Cas d'une pollution des eaux résultant de rejets liquides

Le risque dépend, dans ce cas, essentiellement de l'utilisation des eaux contaminées. Il est le plus généralement lié à l'ingestion de l'eau elle-même ou des aliments contaminés par l'eau, c'est-à-dire les produits de la pêche et les produits agricoles irrigués.

Comme dans le cas de la pollution atmosphérique, l'évaluation de la CRL se fait en deux temps.

Dans un premier temps, on établit les équations exprimant la dose d'irradiation en fonction de la concentration C du radionucléide dans l'eau, soit:

$$D = f_c\ (C) \tag{6}$$

La voie critique et le groupe de population critique sont déterminés par l'équation qui maximalise D pour une même valeur de C. En donnant à D la valeur limite D_l, on en déduit la concentration limite C_l.

Pour passer de la concentration limite C_l à la CRL, différents facteurs interviennent, dont les plus importants sont le volume de dilution du milieu récepteur et le coefficient d'épuration propre à ce milieu.

Soient R le débit du rejet exprimé en activité par unité de temps; Q, la quantité de polluant accumulé dans le milieu récepteur lorsque l'équilibre est atteint; λ, le coefficient d'élimination du polluant par unité de temps, tenant compte à la fois de la décroissance radioactive et des autres processus d'épuration (fixation sur les sédiments, élimination biologique, etc.). L'équilibre est atteint lorsque

$$R = \lambda\ Q \tag{7}$$

Dans le cas des eaux courantes, le volume de dilution se confond avec le débit Δ de la rivière dans l'unité de temps considérée, d'où:

$$\text{CRL} = \lambda\, C_l\ \Delta = \frac{\lambda}{f_c}\ \Delta\, D_l \tag{8}$$

Dans le cas d'un lac, le volume de dispersion est le volume de l'eau du lac, au-dessus de la thermoclyne; soit V_s, ce volume,

$$\text{CRL} = \lambda\ C_l\ V_s = \frac{\lambda}{f_c}\ V_s\ D_l \tag{9}$$

La CRL dépend donc de trois facteurs principaux:
- le volume de dispersion du milieu récepteur
- le coefficient d'épuration de ce milieu
- le facteur de dose, intégrant l'utilisation des eaux, les habitudes alimentaires des populations de la région, la dilution éventuelle de la contamination au cours des transformations des produits et de leur distribution.

2. EVALUATION DE LA CRL A L'ECHELLE MONDIALE

Certains radionucléides sont susceptibles d'une grande dispersion soit dans le milieu atmosphérique, soit dans les milieux aquatiques. Ils peuvent alors entraîner une exposition généralisée de l'ensemble de la population mondiale. C'est le cas en particulier du krypton-85, du tritium et de l'iode-129. On peut alors chercher à évaluer la CRL de l'atmosphère terrestre ou des eaux du globe pour ces radionucléides.

L'ensemble de la population mondiale étant concernée, il est justifié de prendre pour critères de base des concentrations maximales admissibles dans l'air et dans l'eau correspondant aux limites de dose recommandées pour les individus du public.

On considère alors que le polluant est uniformément réparti dans le volume de dispersion du milieu récepteur. Soit V ce volume; la quantité limite de polluant à admettre dans ce volume V est

$$Q_l = (\mathrm{CMA})\ V \qquad (10)$$

Soit λ_r le coefficient de décroissance radioactive du radionucléide. A l'équilibre, la CRL est égale à

$$\mathrm{CRL} = \lambda_r Q_l = \lambda_r\ (\mathrm{CMA})\ V \qquad (11)$$

La CRL apparaît donc comme le produit de trois facteurs:
- la CMA, qui intègre la dose limite, les caractéristiques du radionucléide et les caractéristiques biologiques de l'homme
- le volume de dispersion du milieu récepteur
- la période radioactive du radionucléide.

2.1. Le milieu récepteur est l'atmosphère terrestre — Cas du krypton-85

On suppose que tout le krypton-85 émis est mélangé instantanément et uniformément dans l'atmosphère terrestre et que le seul processus d'élimination est la décroissance radioactive. D'après la relation (11), la CRL est

$$\mathrm{CRL} = \lambda_r (\mathrm{CMA})\ V$$

Remplaçons les trois facteurs par leur valeur:

$$\mathrm{CRL} = \frac{0,693}{11} \times 3 \times 10^{-7}\ (4,2 \times 10^{18})$$

$$\mathrm{CRL} = 7 \times 10^{10}\,\mathrm{Ci/an}$$

Si l'on admet comme hypothèse que la majeure partie des installations est située dans l'hémisphère Nord et que le mélange entre les atmosphères des deux hémisphères est très lent, le volume de dispersion est réduit d'un facteur 0, 4 et la CRL est égale à $2,8 \times 10^{10}$ Ci/an.

L'évaluation de la CRL permet alors d'apprécier la situation résultant de différentes hypothèses.

a) Supposons que la production d'énergie nucléaire reste constante à son niveau actuel, soit 20 GW(e) par an. En admettant une émission de $0,23 \times 10^6$ Ci par GW(e), l'apport annuel serait alors de

$$20 \times 0,23 \times 10^6 = 4,6 \times 10^6 \text{ Ci}$$

A l'équilibre, l'exposition de la population mondiale serait

$$D = \frac{4,6 \times 10^6}{7 \times 10^{10}} D_l \sim 6 \times 10^{-5} D_l$$

Compte tenu de la période radioactive du krypton-85, qui est d'environ 11 ans, l'équilibre ne serait atteint que dans une cinquantaine d'années. Il en résulte que pour la période 1972-2022, l'exposition de la population resterait comprise entre l'exposition actuelle, évaluée à environ 4×10^{-6} limite de dose et 6×10^{-5} limite de dose.

b) Supposons que la production d'énergie nucléaire mondiale croisse jusqu'à atteindre 4 000 GW(e) en l'an 2000, puis se stabilise à ce niveau, l'émission annuelle de krypton-85 à partir de l'an 2000 serait

$$4\,000 \times 0,23 \times 10^6 = 10^9 \text{ Ci}$$

L'exposition de la population à l'équilibre serait alors

$$D = \frac{10^9}{7 \times 10^{10}} D_l = 1,4 \times 10^{-2} D_l$$

Mais la dose égale à $1,4 \times 10^{-2} D_l$ ne sera atteinte qu'à l'équilibre, c'est-à-dire en 2050. Entre 2000 et 2050 l'exposition de la population mondiale resterait comprise entre $10^{-3} D_l$ et $1,4 \times 10^{-2} D_l$.

L'évaluation du rapport (émission annuelle)/(CRL) donne donc une valeur approchée du degré de protection de la population mondiale. Ce rapport est égal au rapport:

$$\frac{\text{dose délivrée}}{\text{dose limite}}$$

lorsque l'équilibre est atteint.

2.2. Le milieu récepteur est la masse des océans

On suppose que la totalité du radionucléide est rejetée dans les effluents liquides et se trouve finalement dispersée dans l'ensemble des mers et océans, ou plus exactement dans la couche située au-dessus de la thermoclyne (0 à -100 m) en négligeant les échanges très lents qui affectent les eaux profondes. Le volume de dispersion est alors d'environ $3,5 \times 10^{16}$ m³.

2.2.1. Cas du tritium

La CRL calculée suivant (11) est

$$\mathrm{CRL} = \frac{0,693}{12} \times 0,003 \times 3,5 \times 10^{16} = 6 \times 10^{12}\ \mathrm{Ci/an}$$

Les hypothèses considérées pour le krypton-85 conduisent aux résultats suivants:

a) Prolongement de la situation actuelle au niveau de 20 GW(e). Prenant pour base une production de tritium de 23 Ci par MW(e) et par an, la production annuelle est $4,6 \times 10^5$ Ci. A l'équilibre, c'est-à-dire dans une soixantaine d'années, l'exposition de la population mondiale serait

$$D = \frac{4,6 \times 10^5}{6 \times 10^{12}} D_l = 10^{-7} D_l$$

b) La puissance installée atteinte en l'an 2000 se stabilise au niveau de 4 000 GW(e). L'émission annuelle de tritium serait alors égale à 10^8 Ci et l'exposition de la population à l'équilibre serait

$$D = \frac{10^8}{6 \times 10^{12}} D_l = 1,6 \times 10^{-5} D_l$$

Mais cette dose ne serait atteinte que vers 2060, progressant de $3 \times 10^{-6} D_l$ en 2000 à $1,5 \times 10^{-5} D_l$ en 2060.

2.2.2. Cas de l'iode-129

La CRL calculée suivant (11) serait:

$$\mathrm{CRL} = \frac{0,693}{1,7 \times 10^7} \times 4 \times 10^{\ 7} \times 3,5 \times 10^{16} = 400\ \mathrm{Ci/an}$$

Il apparaît donc, et il fallait s'y attendre, que pour un radionucléide à période très longue, la CRL reste relativement basse. Mais comme l'équilibre exigerait, pour être atteint, un délai de l'ordre de 100 millions d'années, la prise en considération de la CRL n'a plus de signification.

Il est plus réaliste de considérer la quantité maximale admissible dans l'atmosphère, telle que

$$Q_l = (\mathrm{CMA})\ V = 1,4 \times 10^{10}\ \mathrm{Ci}$$

et de comparer la quantité cumulée à cette quantité limite, en négligeant la décroissance radioactive.

2.2.3. Evaluation pour l'an 2000

En se basant sur l'hypothèse de décroissance de Spinrad [4] et en admettant un taux de production de 1 Ci par an et par GW(e), on peut évaluer à 4×10^4 Ci l'activité de l'iode-129 produit d'ici à l'an 2000. L'activité totale rejetée dans le milieu, en l'absence de filtres (2% dans les effluents liquides et 5% dans les effluents gazeux) serait alors égale à $2,8 \times 10^3$ Ci.

En supposant que l'iode rejeté dans l'atmosphère et déposé au sol atteint la nappe phréatique et rejoint dans les océans l'iode-129 rejeté dans les effluents liquides, on peut admettre à long terme une dispersion dans l'ensemble des mers du globe. Le rapport

$$\frac{2,8 \times 10^3}{1,4 \times 10^{10}} = 2 \times 10^{-7}$$

donne l'exposition de la population évaluée en limite de dose.

En l'an 2000, la production annuelle d'iode-129 serait de 4×10^3 Ci et, toujours en l'absence de filtres, la quantité rejetée serait

$$0,07 \times 4 \times 10^3 = 280 \text{ Ci/an}$$

Supposons, suivant l'hypothèse retenue pour les autres radionucléides, que la puissance atteinte en l'an 2000 se stabilise. En l'an 2100, la quantité rejetée serait

$$2800 + 280 \times 100 = 30\,800 \text{ Ci}$$

L'exposition de la population mondiale serait alors

$$\frac{30\,800}{1,4 \times 10^{10}} \# 2 \times 10^{-6} D_1$$

ce qui est encore tout à fait négligeable.

Au surplus, des considérations d'ordre économique conduisent à réduire la durée du cycle du combustible, notamment le temps de refroidissement. Ceci conduit, en raison du risque lié à l'iode-131, à introduire des filtres assurant des facteurs de décontamination d'au moins 10^4 pour retenir l'iode-131 [5]. Le même facteur de décontamination s'applique évidemment à l'iode-129.

Dans tous les cas, pour les radionucléides à période très longue, la CRL, telle qu'elle a été définie, perd toute signification.

CONCLUSION

La CRL d'un milieu récepteur pour un polluant donné apparaît comme une notion extrêmement utile en radioprotection, car elle intègre tous les facteurs qui jouent un rôle important dans la dilution du radionucléide dans le milieu, son épuration éventuelle au sein de ce milieu, ainsi que tous les autres facteurs locaux, régionaux ou mondiaux, qui conduisent à l'évaluation de la dose d'irradiation qui en résulte pour les populations concernées.

Fondée sur les mêmes hypothèses que la notion de concentration maximale admissible, elle présente les mêmes limitations. En particulier, appliquée à des radionucléides à période très longue, tels que l'iode-129, elle risque de perdre toute signification à l'échelle de la vie humaine, à moins que d'autres processus d'élimination plus rapides n'interviennent pour abréger le temps nécessaire à l'établissement de l'équilibre.

Sous cette réserve, la CRL peut rendre les plus grands services. C'est ainsi que la comparaison du niveau de rejet autorisé en un point à la CRL du milieu en ce point donne automatiquement le degré de protection assuré.

Utilisée à l'échelle locale ou régionale, la CRL permet de comparer utilement entre eux plusieurs milieux récepteurs, plusieurs sites et éventuellement plusieurs points de rejet. A l'échelle mondiale, elle constitue une limite plus théorique, mais qui conserve tout son intérêt pour des prévisions à long terme.

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DISCUSSION

E. KUNZ: It seems to me that evaluation of the limiting environmental capacity in the case of external exposure or exposure by inhalation is connected with site factors, such as residence by an individual in a certain place, and consequently either the factor f_c or the data on the contamination of the environment (see Eq. (1) of your paper) will need to correspond to the local situation.

G. LACOURLY: In cases of direct irradiation or of inhalation, site parameters are not involved in the evaluation of the dose factor as I have defined it, unless residence by an individual is regarded as such a parameter. On the other hand, site parameters are to be taken into consideration when, in the second phase of the calculation, one proceeds from the concentration of the radionuclides in the environment to the limiting environmental capacity.

THE ENVIRONMENTAL CAPACITY OF FRESHWATERS FOR WASTE RADIONUCLIDES

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Abstract

THE ENVIRONMENTAL CAPACITY OF FRESHWATERS FOR WASTE RADIONUCLIDES.

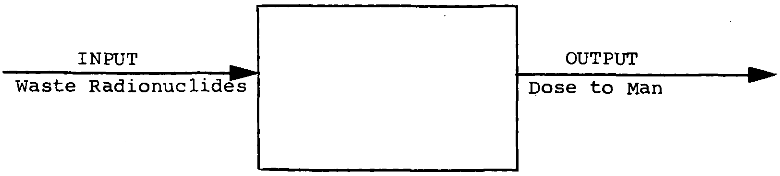
The maximum permissible human body burden of many radionuclides can be expressed as a specific activity, i.e. the amount of a radioisotope per unit weight of its stable counterpart. A similar concept can be applied to a large body of water that is providing water and food for man. It can be thought of as a pool or reservoir of stable elements that could dilute waste radionuclides down to specific activities less than those corresponding to the body burden. It is the size and the dynamics of this stable element pool that determine the environmental capacity of an aquatic ecosystem for a radionuclide.

Results from a continuing study of strontium in an experimental lake (Perch Lake, Ontario) illustrate some of the factors that must be considered. Using elevated levels of ^{90}Sr, which have been present in the lake since 1955, it has been possible to investigate the dimensions and the dynamics of the various compartments that make up the stable strontium pool. Several of these compartments, such as lake water and some biota, have a fast turnover time (i.e. they rapidly attain a uniform specific activity); but they make up only a small fraction of the stable element pool. Other compartments of interest are the marginal vegetation and the semi-aquatic animals, which cause a carry-over of ^{90}Sr into terrestrial food-chains and result in further dilution of the radionuclide. Bottom sediment is important, although a considerable fraction of the stable strontium in the sediment of Perch Lake has such a slow exchange with the lake water that it limits the effective size of this compartment. However, it is the trapping action of the sediment and the consequent removal of some radionuclides from the ecosystem that provides the continuing capacity of a lake to accept such radionuclides.

INTRODUCTION

After being discharged into a lake, a waste radionuclide is subject to physical, chemical and biological processes which distribute the radionuclide throughout the lake ecosystem. This mixing takes time depending on the rate constants of the particular processes involved. Distribution of the radionuclide is not necessarily uniform throughout the ecosystem, but the over-all effect is that of dispersion of the radioactive atoms. The simplest model for our purpose [1] would show an aquatic ecosystem as a single compartment, with the input being radionuclides and the output being a radiation dose to a human population living in the vicinity of the lake (Figure 1).

If the radionuclide is an isotope of a chemical element already present in the ecosystem then the same processes that are acting on the stable element will act on the introduced radioisotope. In effect, there occurs a chemical dilution of the radioactive atoms by the stable atoms. Whether the final distribution of the radioisotope within the ecosystem is exactly



AQUATIC ECOSYSTEM

FIG.1. Systems analysis model of lake contamination.

the same as that of the element depends on a number of factors which are discussed in the following sections of this paper.

SPECIFIC ACTIVITY

The measurement, in an environmental sample, of the amount of a stable element and its radioisotope produces a ratio which is termed the specific activity (SA) of that radionuclide. Formally, using the notation of KAYE and NELSON [2], it can be expressed as:

$$SA_i \text{ (dimensionless)} = \frac{X_i}{X_i + Y_i} \qquad \text{..........(1)}$$

where X_i = concentration of radioisotopes (atoms/g) or number of atoms in compartment i

Y_i = concentration of stable element (atoms/g) or number of atoms in compartment i

In most situations the mass of the radioisotope is negligible so that we can write:

$$SA_i = \frac{X_i}{Y_i} \qquad \text{..........(2)}$$

We may regard the freshwater food chain in its basic form as a two-compartment system where compartment 1 represents the lake ecosystem and compartment 2 represents the critical organ of ICRP standard man. If instantaneous equilibrium occurred then:

$$SA_1 = SA_2$$

$$\text{or} \quad \frac{X_1}{Y_1} = \frac{X_2}{Y_2} \qquad \text{..........(3)}$$

It is in this form (i.e. as the worst possible case in an evaluation of radiation hazards) that the specific activity concept has been utilized for discussions of marine disposal of radioactive wastes by committees of the U.S. National Academy of Sciences [3,4] and for evaluation of the radiological consequences of Plowshare proposals [e.g. 5,6].

In fact, instantaneous equilibrium does not occur, and a number of other factors must be inserted into Eq. (3). On the right-hand side of the equation these include: (1) the loss of X_2 by decay (physical half-life), (2) the loss of X_2 and Y_2 by elimination from the critical organ (biological half-life) and (3) the growth of the critical organ during the time that the contamination persists. The mathematical treatment of these parameters and a sensitivity analysis of them is given by KAYE and NELSON [2]. They show that if Eq. (3) is used, the calculated amount of radionuclide (X_1) which may be safely discharged is always conservative in cases where the biological half-life of the radionuclide is long in comparison with its physical half-life. However, they do not analyze factors affecting X_1 and Y_1 and they are the main concern of this paper.

BIOLOGICAL AVAILABILITY

The determination of the amount of a radioisotope (X_1) and of a stable element (Y_1) participating in a lake ecosystem is not always a simple matter. Ample evidence is now available to show that the presence of a radionuclide in a water body does not necessarily mean that it is involved in the ecosystem processes. Conversely it is possible that the radionuclide can participate in processes to a much greater extent than its stable analogue. For example, the results of Merlini [7] on the accumulation of ^{54}Mn by freshwater clams in Lago Maggiore are only understandable if the radiomanganese (from weapons fallout) was incorporated into the ecosystem food chains much more readily than the stable manganese present in the lake and inlet waters.

Another striking example is the behaviour of ^{55}Fe from fallout in the N.E. Pacific Ocean [8] where the specific activities of radioiron in salmon were found to be 1000 to 10,000 times greater than that in the sea water from which they were taken. Some of these differences can be attributed to movement of fish to an area of lower specific activity after becoming contaminated [9], but it is mostly due to differences in physicochemical form of the stable iron and the radioisotope. Similar anomalies in specific activities have been found for ^{60}Co and ^{65}Zn in Pacific Ocean water and biota [10].

What is required then, in the estimation of X_1, is not the straightforward measurement of the total amount of radionuclide present. We also require information on the relative availability of the radioisotope and stable element to those organisms which act as entry points into the food chains of the ecosystem. Such studies on ^{90}Sr [11] and ^{137}Cs [12] in contaminated freshwaters have shown no differences in biological availability between these radioisotopes and their stable analogues.

Other problems occur when estimates are undertaken of the amount of stable element (Y_1) in an ecosystem. Boundaries of a lake ecosystem are difficult to define because some food chains have terrestrial components that extend beyond the lake shore. Furthermore, some of the important compartments of the ecosystem, such as bottom sediments, contain large amounts of stable elements which may be measured in some methods of chemical analysis but, in fact, can not participate in exchange processes because of physical barriers. This would be true of ions trapped in clay lattices, occluded in sand grains, or incorporated into decay-resistant organic materials (e.g. "humic acids") produced outside the lake and carried into it by inlet streams [11].

A further source of uncertainty is the volumetric extent to which bottom sediments participate. In some lakes the top centimetre of sediment contains a significant fraction of some of the elements in the whole ecosystem. The top metre would contain many times the total in the remainder of the lake. Consequently, the depth to which exchange processes operate, which we can call the "effective depth", is a major consideration.

The problems mentioned above, and the important question of the rates at which these processes occur, can be studied conveniently in lakes which contain measureable quantities of radionuclides. In the next section they are discussed using results

TABLE I. DIMENSIONS OF SOME STABLE STRONTIUM COMPARTMENTS IN PERCH LAKE

Compartment	Area (m^2)	Stable Sr (mg/m^2)	Total Stable Sr (kg)
Water (2 m)	4×10^5	66	26
Vegetation	1.6×10^5	13	2
Animals (herbivores)	-	0.33 (0.28)*	0.13 (0.11)*
Bottom sediment			
Gyttja (60 cm)	3×10^5	2100	630
Sand (60 cm)	1×10^5	10000	1000

* Value after correcting for biological availability (see text).

obtained by the author and his colleagues during studies of the behaviour of ^{90}Sr in Perch Lake, a small, natural lake which lies wholly within the boundaries of the Chalk River Nuclear Laboratories [11].

STUDIES IN A CONTAMINATED LAKE

Perch Lake has contained relatively constant, low concentrations of ^{90}Sr since 1955 [13]. During the last nine years (when regular measurements of lake water concentrations have been available) the mean ^{90}Sr content was 196 picocuries/litre. Use of an average figure of 33 micrograms/litre (measured during 1965-1971) for the stable strontium content of the water results in mean specific activity of almost exactly 6 picocuries/microgram (range: 4.4 - 8.0).

The stable strontium pool of the lake ecosystem is divided between a number of compartments* of different size and complexity. A simple model would recognize lake water, plants (producers), animals (consumers) and lake bottom (sediment) as important compartments. Food chains, physical and chemical exchange are the processes by which these compartments interact with each other. A summary of compartment dimensions is given in Table I.

Lake Water

The mean depth of water in Perch Lake is 2 metres and the area is 4 x 10^5 square metres (m^2). An average Sr content of 33 $\mu g/\ell$ means that the lake water contains approximately 26 kilograms of the stable element. Water samples taken in different parts of the lake show a uniform distribution of Sr and mixing of inlet stream water is rapid (dye studies). Analysis of filtered (1.2 micron) and unfiltered samples has shown that all of the strontium and radiostrontium are in solution.

Vegetation

A dense zone of submersed, floating-leaf and emergent aquatic plants surrounds the lake and occupies about 40 per cent of the lake area. The summer standing crop of plants in the lake has been estimated as 1130 g (fresh weight)/m^2 [13]. Analysis of the six most common species averages 11.7 μg Sr/g (f.w.) [11] which means the total amount of stable strontium present in the aquatic vegetation is approximately 2 kilograms.

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* Strictly speaking these should be called sub-compartments since the lake ecosystem was referred to earlier as a single compartment. For convenience the term compartment will be used in this section of the paper.

TABLE II. STRONTIUM CONTENT AND SPECIFIC ACTIVITIES OF LAKE ANIMALS (Lake water is given for comparison of specific activities)

Animal (No.)	Approx. weight (kg)	Stable Sr content* (μg/g f.w.)	Specific Activity pCi^{90}Sr/μgSr
Beaver (2)	10.0	60.0	0.12
Muskrat (2)	1.25	20.0	4.2
Turtle (2)	0.5	20.0	1.5
Mink (2)	0.85	6.5	0.7
Fox (1)	3.8	6.5	1.2
Lake water	-	0.033	6.0

* Calculated from bone analyses assuming all Sr present in bone and skeleton weight is 10% of animal weight.

Exchange of strontium and radiostrontium between most plants and lake water is rapid and complete as indicated by identical specific activities in both compartments throughout the growing season. There is evidence that some deep-rooted plants obtain some of their strontium from below the level to which ^{90}Sr has penetrated in sediment, resulting in a lower specific activity in these plants [14]. However, such plants make up only a small fraction of the total plant biomass.

Animals

The lake supports a rich population of herbivorous animals including insects, molluscs, fish (Ictalurus), turtles (Chrysemys), muskrat (Ondatra) and beaver (Castor). No population estimates have been made for Perch Lake but reasonable estimates of herbivore biomass for freshwater ecosystems are available from the literature [e.g. 15]. A conservative figure of 2.5 g (dry weight)/m^2 for the total summer standing crop of herbivores was used in the following calculations. This has been divided into 50% for fish, 25% for invertebrates and 25% for the larger animals.

Using a dry weight/fresh weight ratio of 0.33 and dividing the 0.625 g/m^2 equally between turtles, muskrats and beaver we find that the live-weight standing crop for each species is 2.5 x 10^2 kg. This provides us with a population estimate of approximately 500 turtles, 200 muskrats and 25 beaver, all of which are reasonable numbers for a 40 hectare (100 acre) lake.

Specimens of each of these species were trapped and tissues were analyzed for strontium and ^{90}Sr (Table II). From these results it can be calculated that the resident beaver population contains about 15 grams of stable strontium and the muskrat and turtle populations about 5 grams each - very small amounts even when compared with the vegetation content of 2 kilograms. Furthermore, the very low specific activity of the strontium in beaver (2 per cent of that in the water) (Table II) indicates that these aquatic animals derive very little of their ^{90}Sr (and strontium) from the lake. Consequently their actual contribution of stable strontium to the compartment total is negligible. This is not true of the muskrats (70% of water SA) and turtles (25% of water SA), although the Sr metabolism of these species is not completely intermeshed with the lake food-web.

Analyses of Ictalurus from Perch Lake have shown a stable Sr content of 33 μg/g f.w. [11] and a specific activity identical with that of the lake water [14]. The estimated total Sr contained in the herbivorous fish biomass (1.5 x 10^3 kg) is then approximately 50 grams.

We have a few analyses of the herbivorous mollusc Amnicola (135 μg Sr/g f.w.) and insect larvae (15 μg Sr/g f.w.) from the lake, but no information on the specific activity of these organisms. Assuming each comprises 50 per cent of the invertebrate biomass results in a total of approximately 55 grams of stable strontium in this sub-compartment. The grand total for all herbivorous animals is 130 grams, but after corrections based on their specific activities the total becomes 110 grams - less than 0.5 per cent of that in the lake water (Table I).

Two species of carnivores were collected and analysed for stable and radioactive strontium (Table II). Both mink (Mustela) and red fox (Vulpes) show from their specific activities (respectively 12% and 20% of water SA) that they have some involvement with lake food chains. However the relative biomass of predators is very small [15] and the resulting terrestrial carry-over is negligible.

Bottom sediment

Two types of sediment are found in the lake — black semi-liquid organic gyttja which occupies about 75% of the lake bottom, and firm sand with a low organic content found on the east side of the lake. On a dry weight basis the total stable strontium content of the two sediment types is very different, the gyttja containing approximately 50 μg/g and the sand 10 μg/g [11]. However, because of the big difference in water content, the mean wet weight values are near 3.5 μg/g for the mud and 7 μg/g in sand. This means that each centimetre of the upper layers of the two layers of sediment in the lake contains approximately 35 mg Sr/m^2 and 175 mg Sr/m^2 respectively.

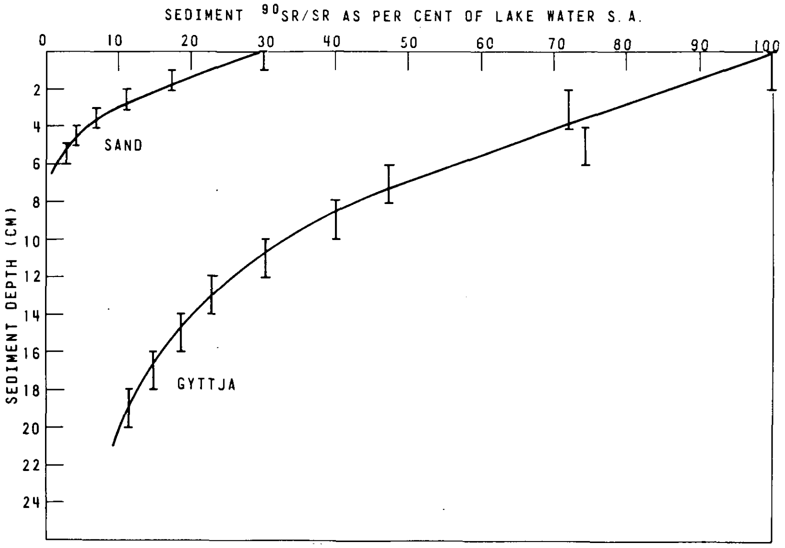

FIG.2. Distribution of ^{90}Sr in two types of Perch Lake sediment (data derived from two cores of each sediment type).

In order to estimate the magnitude of the sediment strontium compartment we need to know; (1) the depth to which exchange processes occur between strontium in lake water and strontium in the sediments and (2) the completeness of this exchange. Both of these questions can be answered in Perch Lake by an examination of ^{90}Sr and stable strontium distribution in sediment cores. The results for both kinds of sediment are given in Figure 2.

The curves in Figure 2 show that in the 16 years (1956-1972) that ^{90}Sr has been present in the lake it has penetrated approximately 6 cm into the sandy sediment and 24 cm into the gyttja. The observed distribution is not due to deposition of contaminated sediment. Rate of sedimentation in the lake (using a ^{14}C dated core)* can be estimated as about 6 x 10^{-2} cm/yr. This rate would result in less than 1 cm of sediment accumulation in the 16-year period.

If diffusion of the ^{90}Sr ion in interstitial water was the sole mechanism of penetration into the sediments then the radioisotope would have reached approximately 100 cm in 16 years at known diffusion rates of the Sr^{2+} ion [16]. The shorter penetration depths shown in Figure 2 are the result of retardation of diffusional transport of ^{90}Sr by chemical exchange with the sediments. In fact, comparison of ^{90}Sr concentration in the upper

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* ^{14}C date determined by Dr. J. Terasmae, Brock University, Ontario, Canada.

layer of sediment with that in the lake water provides an estimate of the distribution factor (K) representing the exchange and uptake of strontium by the wet solids. This was found to be K = 100 ± 20 for both types of sediment [11].

In their investigation of fallout ^{90}Sr in Lake Ontario, LERMAN and TANIGUCHI [16] calculated that a sediment penetration depth of 8 cm in 16 years resulted from a $K \sim 120$ and a diffusion coefficient (D) of 3×10^{-6} cm^2/sec (9×10^1 cm^2/yr). This corresponds closely with the sandy sediment of Perch Lake. Greater penetration into the gyttja is probably due to some mixing of the upper layers of the semi-liquid mud during periods of rough water in the shallow lake.

Substituting $K = 100$ and $D = 3 \times 10^{-6}$ cm^2/sec in the equation (8) given by LERMAN and TANIGUCHI for steady state conditions ($t \to \infty$), we find significant amounts of ^{90}Sr (~ 10% of surface concentration) penetrating 150 cm into the sediment. Considering the high stable strontium content of sediment this depth represents a huge potential compartment, equivalent to at least 60 cm of fully exchanged sediment ("effective depth"). This depth contains 2.1×10^3 mg Sr/m^2 in gyttja and 1×10^4 mg Sr/m^2 in sand, resulting in estimated stable strontium contents of 6.3×10^2 kg and 1×10^3 kg respectively (Table I).

ENVIRONMENTAL CAPACITY

From Table I we find that the bottom sediments of Perch Lake contribute 98% of the stable strontium available in the ecosystem for dilution of ^{90}Sr. However, if the rate of ^{90}Sr exchange (including mixing) between water and sediments is very slow in comparison with the residence time of the water in the lake then the material balance might hardly be affected despite the large pool of exchangeable strontium in the sediment.

Using the data in Figure 2 it can be estimated that the ^{90}Sr held in the gyttja is approximately 630 millicuries and in the sand 60 millicuries, making a total of 690 millicuries (uncorrected for decay) that has been trapped by the sediments over a period of 16 years. During that same period, monitoring data from the Environmental Research Branch indicates that about 4500 millicuries of ^{90}Sr has left the lake in the outflow. So notwithstanding the large ultimate capacity of the lake ecosystem for trapping radiostrontium (approximately 10 curies would be held in the top 60 cm of sediment at equilibrium), the rate processes in the ecosystem limit the ^{90}Sr removal in Perch Lake to about 15 per cent of that passing through the lake - about 40-50 millicuries per year under the present conditions.

The quantitative information on environmental capacity obtained in this study is, of course, specific to ^{90}Sr and Perch

Lake. Other radionuclides will have different half-lives, biological availabilities, diffusion coefficients, and distribution factors. Other lakes will have different volume/bottom area ratios, residence times, and ecosystems [16]. However, the study of radioactively labelled water-bodies like Perch Lake can contribute much to our understanding of the processes that underlie the environmental capacity of freshwaters for waste radionuclides.

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DISCUSSION

P. REINIGER: In your paper you give a number of examples where the availability to living organisms of a radioisotope and the stable isotope of the same element is different. Was the difference in the physico-chemical form of the isotopes given as an explanation assumed or actually measured, and do you know of any experiments carried out under controlled conditions to investigate the difference in uptake?

I. L. OPHEL: In the examples mentioned in the paper it was found that unexpected differences in physico-chemical form were responsible for the observed anomalies in specific activity. No such anomalies have been found for strontium.

I know of one laboratory study of anomalies in ^{65}Zn/Zn specific activities and the title and source are: CROSS, A. F., WILLIS J. N., BAPTIST, J. P., Distribution of radioactive and stable zinc in an experimental marine ecosystem, J. Fisheries Res. Bd Can. 28 11 (1971) 1783-88.

O. L. van der BORGHT: On the basis of your data from Perch Lake is it possible to establish the relative importance of direct uptake and uptake through the food chain of Sr (and Ca) in the shell of pulmonate snails?

I. L. OPHEL: We are at present working on the Sr/Ca ratios in the pulmonate mollusc Amnicola, the only pulmonate species in the lake. I feel optimistic about being able to determine the relative importance of diet and direct uptake for radiostrontium accumulation by Amnicola because many of the plant species on which it feeds have a different Sr/Ca ratio from that in the water.

W.K.G. KÜHN: In the paper you quote the ratio of ^{90}Sr/stable strontium in the lake ecosystem over the past 16 years. Is this ratio always constant, or does a time-function come into play here? I can imagine that the ratio may well be an equilibrium factor!

I. L. OPHEL: Both the ^{90}Sr and the stable strontium content of the water entering Perch Lake have remained remarkably constant during the ten years or so during which detailed analyses have been available. Further information on the measured concentrations can be found in the paper by Ophel et al. presented at the radioecology conference held in Rome in September 1971.

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ОСНОВНЫЕ ПРИНЦИПЫ САНИТАРНОГО НОРМИРОВАНИЯ РАДИОАКТИВНЫХ ВЕЩЕСТВ В ВОДЕ ПОВЕРХНОСТНЫХ ВОДОЕМОВ

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Abstract–Аннотация

BASIC PRINCIPLES FOR ESTABLISHING HEALTH PROTECTION STANDARDS WITH RESPECT TO RADIOISOTOPE CONCENTRATIONS IN SURFACE WATER BODIES.

The public health problem of preventing radioisotopic contamination of surface water bodies can be resolved only if one has criteria for controlling the concentrations of various isotopes in the water. Different approaches have been adopted in different countries to the establishment of such criteria. The authors discuss various ideas in relation to conditions in the Soviet Union. The great variety of situations encountered in practice (arising out of the different uses to which different water bodies are put and social and economic aspects of the way of life of critical population groups) makes it virtually impossible to establish a single, meaningful stadard (maximum permissible concentration) for every isotope. A better approach is therefore one that takes into account local conditions while being based in all cases on the same principles, criteria and computational methods. The maximum permissible radioisotope concentrations established in the Soviet Union for water bodies relate to that part of the water body that is directly below the point of effluent discharge. The establishment of standards is based on the "critical population group", i.e. those rural inhabitants of littoral settlements who come into closest contact with the water body (fishermen, buoy-minders etc. and members of their families) or who are engaged in agriculture in the associated floodplain. It entails testing the effects of ionizing radiation on man, account being taken of various factors. One or more tests may be carried out, depending on the conditions in each particular case. Each of the factors is considered separately in assessing the effects of ionizing radiation on man, but standards are ultimately established on the assumption that all the factors may act simultaneously, the resulting values being corrected as required (one must consider not only the present situation, but also the possible long-term uses of the water body). Apart from solving immediate public health problems, the establishment of these standards has a broader purpose: it creates safe conditions for the development of fishing, irrigation-based agriculture, milk and meat production, etc.

ОСНОВНЫЕ ПРИНЦИПЫ САНИТАРНОГО НОРМИРОВАНИЯ РАДИОАКТИВНЫХ ВЕЩЕСТВ В ВОДЕ ПОВЕРХНОСТНЫХ ВОДОЕМОВ.

Практическое решение проблемы санитарной охраны поверхностных водоемов от загрязнения радиоактивными изотопами невозможно без наличия критериев, позволяющих регламентировать их концентрации в воде. Установление таких критериев в разных странах производится по - разному. В данном сообщении излагаются соображения применительно к условиям Советского Союза. Чрезвычайное разнообразие ситуаций, наблюдаемых на практике и обусловленных природными условиями, характером использования водоемов, социально- экономическими особенностями жизни критических групп населения, ограничивает возможность надежного обоснования единых унифицированных для каждого изотопа допустимых концентраций. Более целесообразным является дифференцированный подход с учетом местных особенностей. Вместе с тем, во всех случаях принципы подхода, критерии и методы расчета должны быть едины. Исходя из положений, действующих в Советском Союзе, допустимые концентрации радиоактивных изотопов устанавливаются ются для участков водоемов непосредственно ниже места спуска сточных вод. Нормирование ведется для критической группы населения. К ней относится часть сельских жителей прибрежных населенных пунктов, наиболее тесно контактирующих с водоемом (рыбаки, бакенщики и члены их семей) или с пойменной частью долины (население, использующее ее для сельскохозяйственных целей). Для нормирования применяются тесты, отражающие воздействие ионизирующей радиации на человека с учетом различного рода факторов. В зависимости от конкретных условий может быть использован только один тест или весь комплекс. Нормирование ведется по каждому из тестов, а также для условий одновременного воздействия всех факторов с последующим корректированием полученных значений. При этом целесообразно учитывать ситуацию не только на данный момент, но также характер вероятного использования водоема на перспективу. Наряду с решением чисто санитарных задач подобное нормирование носит универсальный характер, гарантируя безопасные условия для развития рыбного хозяйства, поливного земледелия, мясо-молочного животноводства и т.д.

Решение проблемы санитарной охраны открытых поверхностных водоемов от загрязнения радиоактивными изотопами невозможно без наличия критериев, позоляющих регламентировать их концентрации в воде. Установление таких критериев в разных странах производится по-разному [1].

В данном сообщении излагаются соображения применительно к условиям Советского Союза.

Использование для этой цели нормативов, установленных для питьевой воды, далеко не всегда обосновано и оправдано, так как из числа радиоактивных изотопов, попадающих в открытые водоемы, большинство может мигрировать отсюда в организм человека по пищевым цепям. Кроме того, при наличии в воде гамма-излучателей не исключена возможность внешнего облучения людей непосредственно от зеркала водоема или от поверхности территории, подвергающейся затоплению (в паводок) или орошению (ирригация). Следовательно, возникает необходимость обоснования таких нормативов, которые бы учитывали различные пути возможного воздействия на человека ионизирующей радиации, обусловленной радиоактивными изотопами, содержащимися в воде водоемов. Указанные параметры необходимы: 1) в качестве исходных данных при проектировании канализационных систем, предназначенных для обезвреживания и удаления сточных вод, содержащих радиоактивные вещества; 2) для целей оперативного санитарного контроля в качестве критерия при оценке радиологической чистоты водоемов; 3) для гигиенической оценки прогнозируемой радиационной обстановки, которая может возникнуть при эксплуатации устройств, являющихся потенциальными источниками загрязнения местности радиоактивными веществами.

Однако чрезвычайное разнообразие ситуаций, наблюдаемых на практике и обусловленных природными факторами (гидрохимические, гидрологические, гидрографические особенности водоемов), характером использования водоемов, социально - экономическими особенностями жизни критических групп населения и т.д. ограничивают возможность надежного обоснования единых, унифицированных для каждого изотопа допустимых концентраций. Более целесообразным является обоснование нормативов для соответствующих изотопов применительно к конкретным условиям с учетом местных особенностей. Вместе с тем во всех случаях принципы подхода, критерии и методы расчета указанных нормативов должны быть едины. Изложение их и является задачей настоящей работы.

Исходя из положений, принятых в Советском Союзе, допустимые концентрации радиоактивных изотопов в воде водоемов устанавливаются для участков водоемов, находящихся непосредственно ниже места сточных вод. Это обусловлено главным образом загрязнением рыбы преимущественно вблизи выпуска сточных вод, а также неизбежной кумуляцией в донных отложениях, являющихся своеобразным "регулятором" концентраций в воде.

Допустимые концентрации радиоизотопов в воде должны исключить возможность облучения критических групп населения в дозах, превышающих пределы доз, рекомендованных МКРЗ и принятых советским законодательством [2]. При этом делается допущение, что человек подвергается воздействию ионизирующей радиации, обусловленной только радиоактивными изотопами, находящимися в водоеме. При наличии других источников облучения (не считая естественных и применяемых в медицине) в расчеты вносятся необходимые коррективы.

Следуя рекомндациям МКРЗ [3] и МАГАТЭ [4], нормирование ведется для критической группы населения. Применительно к данной проблеме указанную группу составляет часть сельских жителей прибрежных населенных пунктов, наиболее тесно контактирующих с водоемом (рыбаки, бакенщики, паромщики и члены их семей) или с пойменной частью долины (население, использующее для сельскохозяйственных целей заливные луга и огороды).

В зависимости от характера использования водоема, его природных свойств и других условий пути воздействия на человека ионизирующей радиации, обусловленной радиоактивными изотопами, находящимися в воде водоема, могут быть различны, равно как и санитарная значимость каждого из них. Поэтому при обосновании нормативов приходится учитывать отдельные критические пути воздействия, либо их совокупность. В этой связи необходимо использование ряда тестов, отражающих воздействие ионизирующей радиации на человека с учетом:

1) Характера использования водоема:
    а) как источника хозяйственно-питьевого водоснабжения;
    б) как место ловли рыбы.
2) Возможного загрязнения пищевых продуктов "наземного" производства (молоко) в результате периодического затопления прибрежной территории.
3) Значимости внешнего гамма-облучения от зеркала воды и поверхности поймы.
4) Возможного комбинированного (внешнего и внутреннего) воздействия.

В зависимости от конкретных условий при обосновании допустимых концентраций может быть использован только один тест, характеризующий поступление радиоактивных изотопов по критическому пути (например, с рыбой). В других случаях при одновременном воздействии ионизирующей радиации по нескольким путям производится корректирование значений допустимых концентраций с учетом вклада каждого из путей воздействия.

В качестве исходных материалов для обоснования указанных нормативов используются:

-Пределы доз на критические органы, рекомендованные МКРЗ и принятые советскими законодательством для отдельных лиц из населения в качестве официальных.

-Пределы годового поступления соответствующих изотопов через желудочно-кишечный тракт, регламентированные тем же законодательством.

-Величины коэффициентов накопления (КН) радионуклидов в рыбе, установленных в натурных условиях. Значения их для одних и тех же изотопов могут варьировать в зависимости от солевого состава воды, вида рыбы и т.д., что обычно связано с природно-климатическими условиями той или иной местности. Это обстоятельство определяет целесообразность разработки нормативов с учетом местных условий. Следует отметить, что для радионуклидов, распределяющихся в организме равномерно (цезий, тритий, сера и др.), величина КН устанавливается по тушке рыбы. Для остеотропных учитывается КН нуклидов в мягких тканях рыбы, а также количество их, переходящее при кулинарной обработке из костей в съедобную часть пищи.

-Количественная оценка миграции радиоизотопов из водоема в организм человека с рыбой требует знаний величин, характеризующих среднедушевое ее потребление за год критической группой населения.

Имеющиеся данные свидетельствуют о том, что в ряде районов страны среднедушевое потребление рыбы критической группой населения колеблется, составляя в среднем – 200-300 г/сут [5,6].

-В условиях незарегулированных рек может иметь место поступление радионуклидов из водоемов на прибрежную территорию в период паводка. Использование заливных лугов под сенокосы и выпасы создает возможность миграции ряда нуклидов по цепи вода-почва-трава-молоко. Показателем накопления их в почве является коэффициент распределения (КР), представляющий собой отношение удельной активности почвы при максимальном ее насыщении данным радионуклидом к удельной активности воды. Величина КР различных изотопов для разных видов почв неоднозначна. Однако для почв, чаще всего встречающихся в поймах, диапазон колебаний невелик. Например, для цинка от 1460 (дерново-луговые почвы) до 1220 (чернозем), для иттрия соответственно 230 и 110 [7]. Показатели миграции радионуклидов на следующих звеньях данной цепи определяются величиной отношений $\frac{\text{трава}}{\text{почва}}$ и $\frac{\text{молоко}}{\text{трава}}$ (табл.I).

Эти показатели с учетом средне-душевого потребления молока позволяют установить количество радионуклидов, поступающих в организм

ТАБЛИЦА I. ПАРАМЕТРЫ МИГРАЦИИ НЕКОТОРЫХ РАДИОНУКЛИДОВ ИЗ ПОЧВЫ В МОЛОКО

Нуклиды	Коэффициент распределения в почве (дерново-луговой)	Коэффициент перехода	
		из почвы в траву	из травы в молоко
Кобальт	1000	0,07	-
Цинк	1450	-	-
Стронций	1400	1,74	0,001
Цезий	1000	0,19	0,01
Церий	250	0,03	0,0002

с данным пищевым продуктом. Следует отметить, что соотношение трава/почва из-за гетерогенности местных условий (тип почвы, виды растений и т.д.) для одних и тех же изотопов могут быть различны.

-Кроме внутреннего облучения за счет радионуклидов, поступающих из водоема в организм, часть населения (критические группы) может подвергаться внешнему облучению от зеркала водоема и поверхности пойменной территории.

Все упомянутые выше коэффициенты (КН,КР и др.) устанавливаются до достижения равновесного состояния радионуклидов между водой и соответствующими компонентами, что происходит спустя более или менее значительного периода времени. Следовательно, величины допустимых концентраций радиоактивных изотопов в воде водоемов, полученные по всем упомянутым выше тестам, относятся преимущественно к случаям постоянного или систематического поступления радиоактивных изотопов в водоем.

Таким образом, расчет допустимых концентраций (ДК) изотопов в воде водоемов производится исходя из следующих тестов:

1. Потребление воды для питьевых целей.
2. Потребление рыбы из водоема.
3. Потребление молока (возможно, и мяса), загрязненного в результате контакта воды с почвой пастбищ (паводки, ирригация).
4. Внешнее облучение от поверхности почвы, загрязненной в результате контакта с водой.
5. Внешнее облучение от зеркала водоема.

ДК, рассчитанная по каждому из этих тестов, обусловливает в каждом случае облучение организма, равное пределу дозы.

Одновременное воздействие всех или нескольких рассмотренных факторов, как уже указывалось ранее, приводит к суммированию поглощенных доз. Однако поскольку суммарная доза не должна превышать предела дозы необходимо,соответствующим образом скорректировать допустимую концентрацию изотопа в воде.

Расчет суммарного значения ДК проводится по формуле:

$$ДК = \frac{1}{\sum\limits_{1}^{n} \frac{1}{ДК}}$$

В случае, если критическим является один из внутренних органов, то, используя вышеприведенную формулу, следует ДК, полученные по гамма-излучению от поверхности воды и почвы, разделить на коэффициент экранирования мягкими тканями 0,6 [8].

В зависимости от физико-химических свойств изотопа значимость каждого из рассмотренных тестов может быть различна и не исключено, что один из них окажется подавляющим. В этом случае вводить поправку нет необходимости. Ниже приводится табл.II, в которой даны ДК для некоторых изотопов, рассчитанные по каждому из тестов в отдельности и по суммарному воздействию.

Как видно из этой таблицы, используемые тесты далеко не равнозначны. Ведущим фактором в основном является поступление радиоизотопа с рыбой. Однако для кобальта-60 и цинка-65 таковым

ТАБЛИЦА II. ДОПУСТИМЫЕ КОНЦЕНТРАЦИИ РАДИОАКТИВНЫХ ИЗОТОПОВ В ВОДЕ ПРЕСНЫХ ВОДОЕМОВ (Ки/л)

Радиоизотоп	T 1/2	Тесты						Отношение
		С питьевой водой ДК-1	С рыбой ДК-2	С молоком ДК-3	Внешнее облучение от почвы ДК-4	от зеркала водоема ДК-5	Суммарн. ДК	$\frac{\text{ДК-1}}{\text{ДК}}$
^{3}H	12,3 год	$3{,}2 \cdot 10^{-6}$	$3{,}5 \cdot 10^{-5}$	-	-	-	$2{,}9 \cdot 10^{-6}$	1,1
^{24}Na	14,9 час	$2{,}8 \cdot 10^{-8}$	-	-	$4 \cdot 10^{-7}$	$4 \cdot 10^{-8}$	$1{,}9 \cdot 10^{-8}$	1,4
^{32}P	14,5 сут	$1{,}9 \cdot 10^{-8}$	$1{,}10^{-10}$	-	-	-	$1 \cdot 10^{-10}$	190
^{35}S	87,1 сут	$6{,}3 \cdot 10^{-8}$	$4{,}8 \cdot 10^{-8}$	-	-	-	$2{,}7 \cdot 10^{-8}$	2,3
^{60}Co	5,25 года	$3{,}5 \cdot 10^{-8}$	$1{,}3 \cdot 10^{-9}$	-	$3{,}4 \cdot 10^{-11}$	$8{,}2 \cdot 10^{-8}$	$3{,}2 \cdot 10^{-11}$	1000
^{65}Zn	245 сут	$1 \cdot 10^{-7}$	$1 \cdot 10^{-9}$	$1{,}5 \cdot 10^{-8}$	$1{,}7 \cdot 10^{-10}$	$4 \cdot 10^{-7}$	$1{,}3 \cdot 10^{-10}$	770
^{90}Sr	28,4 года	$4 \cdot 10^{-10}$	$1{,}5 \cdot 10^{-10}$	$1{,}4 \cdot 10^{-11}$	-	-	$1{,}3 \cdot 10^{-11}$	31
^{131}I	81 сут	$2 \cdot 10^{-9}$	$2{,}1 \cdot 10^{-9}$	-	$8 \cdot 10^{-7}$	$8 \cdot 10^{-7}$	$1 \cdot 10^{-9}$	2
^{137}Cs	30 лет	$1{,}5 \cdot 10^{-8}$	$1{,}5 \cdot 10^{-10}$	$5 \cdot 10^{-10}$	$1{,}4 \cdot 10^{-10}$	$4{,}6 \cdot 10^{-7}$	$6 \cdot 10^{-11}$	250
^{144}Ce	265 сут	$1{,}2 \cdot 10^{-8}$	$4 \cdot 10^{-9}$	$5 \cdot 10^{-7}$	$5{,}6 \cdot 10^{-8}$	$4{,}1 \cdot 10^{-5}$	$2{,}9 \cdot 10^{-9}$	4
^{210}Po	138,3 сут	$7{,}3 \cdot 10^{-10}$	$2{,}5 \cdot 10^{-12}$	-	-	$\sim 10^{-2}$	$2{,}5 \cdot 10^{-12}$	300

является внешнее гамма-облучение от почвы, для цезия-137 – рыба, внешнее гамма -облучение от почвы и отчасти потребление молока, для трития – питьевая вода. Таким образом, данные таблицы наглядно демонстрируют необходимость учета различных путей воздействия изотопов на организм человека при расчете ДК. Расчет только по тесту "питьевая вода", применявшийся ранее, может в отдельных случаях привести к существенному превышению облучения критических групп населения по сравнению с установленными пределами доз.

В заключение следует отметить, что во всех случаях при обосновании допустимых концентраций радиоактивных изотопов в воде водоемов надлежит учитывать не только ситуацию на данный момент, но также характер вероятного использования водоема на перспективу.

Наряду с решением чисто санитарных задач подобное нормирование носит "универсальный" характер, обеспечивая безопасные условия для развития рыбного хозяйства, поливного земледелия, мясо-молочного животноводства и т.д.

Поскольку указанные нормативы намечается устанавливать для воды водоемов непосредственно ниже места выпуска сточных вод, дальнейшее уменьшение концентраций радиоактивных изотопов за счет разбавления их в массе воды создает надежную гарантию безопасности любого вида использования водоема.

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DISCUSSION

R. F. FOSTER: In the USSR are the concentration factors for foods already being utilized in the establishment of standards for the maximum permissible quantities of radionuclides in water, or is this concept something that it is planned to use in the future?

R.M. BARKHUDAROV: Some concentration factor values are already being used, but much work remains to be done, and will be done, in this field.

M. J. A. DELPLA:- Concentration factors vary from place to place, depending in particular on whether the release point is situated on a river, a lake or the sea. Do you take account of local variations and establish concentration factors for each release point?

I should also like to know what authorities, in the USSR, are competent to establish and apply concentration factors.

R.M. BARKHUDAROV: The concentration factors are established in respect of the equilibrium state of the fish-water system. For this reason we do not concern ourselves with any dependence of the concentration factor on specific points in the water body.

In reply to your second question, specialized laboratories, among them our own, are engaged on the study of isotope concentrations in fish and other organisms. Assignment of permissible concentrations on the basis of the research data obtained is then the responsibility of the Radiation Protection Commission.

R. J. ROUX: I understand that you calculate a different concentration factor depending on the area in which the populations concerned live — Siberia or the shores of the Black Sea, for example. Do you therefore use a number of factors on the territory of the USSR, or is my supposition incorrect, and you in fact use a simple average factor or a minimum factor?

R.M. BARKHUDAROV: We indeed do endeavour to use concentration factors for isotopes in fish that will be applicable to the particular area concerned. In the paper we present average concentration factor values relating to the central zone of the European part of the USSR.

CONTROL OF LOW-LEVEL LIQUID RADIOACTIVE WASTE DISPOSAL
UK experience in the derivation and use of 'environmental capacity'

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Abstract

CONTROL OF LOW-LEVEL LIQUID RADIOACTIVE WASTE DISPOSAL: UK EXPERIENCE IN THE DERIVATION AND USE OF 'ENVIRONMENTAL CAPACITY'.

UK policy on the control of radioactive waste depends primarily on limiting public radiation exposure as this has been found to be more restrictive than potential damage to environmental resources. Assessments of environmental capacity are thus of paramount importance and have become a central feature of the critical pathway approach as practised in the UK. For this purpose environmental capacity is usually identified as a rate of input of radioactivity of specified isotopic composition that, under equilibrium conditions, would result in an exposure of the public equal to the ICRP-recommended dose-limits.

The paper considers how assessment of environmental capacity is conducted, involving detailed studies of the behaviour of radioactivity after release to the environment and the pathways which lead to public radiation exposure. Each assessment is valid only in relation to the appropriate food consumption rate or other habits of the exposed population resulting in exposure. Each pathway can be visualized as involving several stages such as dilution, dispersion, reconcentration, and public use of the environment, culminating in radiation exposure. Each of these stages is discussed to show the factors that must be considered. The procedures adopted in the UK for the evaluation of environmental capacity are discussed in the context of disposal of liquid radioactive wastes to freshwater lake and river systems, particular reference being made to the nuclear power station site at Trawsfynydd and the supporting radioecological research conducted there. Recent experience in re-assessment of environmental capacity in relation to changing factors, such as effluent composition, is discussed and the paper concludes with an account in terms of public radiation exposure of how application of a control policy dependent on the assessment of environmental capacity has ensured the continued safety of waste disposal and the maintenance of very high standards of environmental protection.

1. INTRODUCTION

In establishing methods for the control of liquid radioactive waste disposal to the aquatic environment, attention has been directed primarily to the limitation of public radiation exposure. Studies of the potential damage to environmental resources[1] have shown that this is much less important than the potential consequences to the human population. Indeed, resource damage can be regarded as negligible provided that the ICRP recommendations for human radiation exposure are met.

The UK control system for disposal of radioactive waste has already been described in detail[2] and depends on prior authorization. Discharges are so regulated as to ensure compliance with three national policy objectives laid down in 1959 in a Government White Paper[3]. The first two objectives relate to mandatory dose limits for the public and cover both the somatic and genetic consequences of exposure; the third is more in the nature of a general directive requiring actual levels of exposure to be minimized, as far as is practicable.

It is therefore of paramount importance to ensure that a planned disposal will not result in the chosen dose limits for members of the public being exceeded, and in consequence it is essential to know what rate of input of radioactivity will in a given situation cause maximum allowable exposure of the public. This rate of input is thus the maximum discharge rate which could be permitted in the particular circumstances, and is referred to as the environmental capacity or, sometimes, the limiting environmental capacity. In defining it the habits of the exposed population must be specified, for a particular environmental capacity will only remain valid whilst these factors obtain. Because values of environmental capacity vary considerably for different nuclides, depending on their radiotoxicity, the composition of the waste must also be specified or, alternatively, a series of values calculated for separate critical nuclides, that is to say, those causing the limiting rates of exposure.

In practice, the assessment of environmental capacity can be a difficult process, particularly when it is being made for the first time at a site. In this pre-operational phase a number of assumptions will have to be made, and because of uncertainty in the data used, and in view of the mandatory nature of the major policy objectives, it has become common practice within the UK to apply a safety factor - often 10 - to this first assessment before using it to set authorized limits. This reduced value is often referred to as the provisional environmental capacity, and its use is largely confined to pre-operation and the early operational period. When sufficient experience has been gained of routine disposal of waste to measure environmental contamination, environmental capacity can be re-assessed on a more reliable basis, dispensing with the theoretical data used in the pre-operational assessment. This post-operational estimate will be so much more accurate that there will be little or no need to include a safety factor before using it for regulatory purposes.

A further concept which is used is that of the stipulated environmental capacity, by which is meant the discharge rate specified in the authorization. This should not be confused with the permissible maximum discharge rate (that is, limiting environmental capacity), compared with which it will often be very small.

Because many disposals utilize a very small proportion of the limiting environmental capacity, the need for such thorough investigation may be questioned; however, the concept of environmental capacity based on the critical path approach is so central to UK methodology that a fair knowledge of actual values of environmental capacities is highly desirable, if not absolutely essential. Provisional and stipulated environmental capacities, though based on scientific principles, also reflect the administrative application of the basic policy concepts to practical control. In considering how estimates of capacity are made, discussion will therefore be confined to assessment of the limiting environmental capacity itself.

2. THE ASSESSMENT OF ENVIRONMENTAL CAPACITY

Methods used fall into two categories, classified according to whether calculation is being carried out before or after operation has begun. In the latter case there is a further distinction according to whether measurable contamination exists, this being of value in improving the accuracy of the estimate. Different computational models are employed for internal versus external exposure pathways, though a common factor is their dependence on data from surveys of population habits and foodstuff consumption rates. These surveys have several functions, initially qualitative in identifying the groups most likely to be exposed and thus the pathways of potentially critical importance. This phase is succeeded by a quantitative stage, establishing consumption rates of a foodstuff (or drinking water) if the exposure pathway is internal or the rates of occupancy of an external radiation field if it is external. Because control requires protection of the most highly exposed individuals in the population at risk, the emphasis is on identifying those individuals whose consumption rates or other habits place them on the upper end of the observed exposure range. Surveys are conducted on an interview or "market survey" basis and it would be unusual to be able to cover the whole of the population concerned, the normal aim being to secure a sample representative of the whole exposed group. Wherever possible, a "critical group" is selected from the exposed group as the basis of hazard assessment, though in many cases, due to the remoteness of the sites responsible for major discharges of waste, the exposed population group is quite small and no small critical sub-group can be identified. When this occurs, assessment of environmental capacity is based on the most highly exposed individual.

2.1. Pre-operational assessment of environmental capacity

2.1.1. Internal exposure pathways

The basis of the chosen computational model is shown in Fig. 1, which, for simplicity, will be discussed in the context of a single nuclide. The first stage is to predict, relative to an assumed rate of discharge (generally 1 Curie per day), the concentration at equilibrium in the water mass. For this purpose a hydraulic model is required to describe the mixing process, together with such further factors as radioactive decay and removal by sediment which will influence the water concentration. This predicted water concentration is converted into a concentration in the critical material by use of concentration factors based on research findings and operational experience, and thence - by application of consumption survey data - to a rate of ingestion for the radionuclide concerned. This rate is compared with values of maximum permissible intake calculated from ICRP data[4] to derive the proportion of the ICRP-recommended dose limit to which individuals in the population would be exposed. The environmental capacity is then derived by weighting the assumed discharge rate by this proportion.

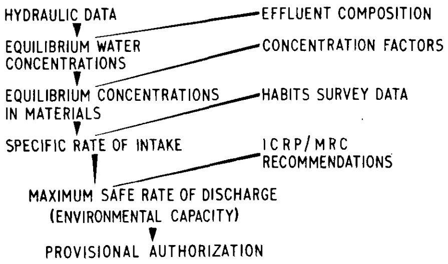

FIG.1. Pre-operational evaluation of environmental capacity due to internal exposure.

In practice, and although the identity of the critical nuclide may be quickly apparent, it is usual to complete this type of calculation for each of the nuclides expected in the effluent. This places great importance on the accuracy of predictions of effluent composition, which have been found to be the principal source of error in the final result. Nevertheless, for control purposes estimates are usually needed in terms of a specified mixture, since a comprehensive authorization covering all radioactivity discharged from a site is required.

Several different values may have to be calculated, referring to exposure of either the whole body or a specific organ, according to which is "critical" - i.e. subject to the highest degree of exposure. In practice, estimates are usually needed only for gastro-intestinal (GI) tract, total body and bone.

The way in which ICRP data are used in this sequence of calculations calls for comment, since direct use of the Commission's recommendations cannot be made. For each nuclide, and in relation to each organ of reference, a "maximum permissible rate of intake" - which may alternatively be called a derived intake limit - is calculated from ICRP data on maximum permissible concentrations in drinking water (168 hour week, occupational worker). One-tenth of this value for each nuclide is taken for application to the general public and multiplied by the same "standard man" consumption rate of drinking water (2.2 litres per day), which has been adopted by ICRP in their metabolic modelling to obtain a total daily intake figure.

2.1.2. External exposure

A computational model similar to that described above is employed, the important differences being in the hydraulic model used and in the stage involving use of ICRP data. The latter is much simpler, in that contamination of the critical material, e.g. sediment, will be defined in terms of the direct radiation dose from the sediment and this can be compared directly with ICRP-recommended dose limits.

2.2. Operational assessment of environmental capacity

2.2.1. Internal exposure

The basis of the computational model used is shown in Fig. 2. Although it follows a similar pattern to that adopted in a pre-operational phase, there is an important difference in the early stage of the procedure, since the need for a hydraulic model is eliminated. The relationship between discharge rate and contamination level in the critical material is established directly from environmental monitoring and detailed effluent analysis, so that knowledge of concentration factors is also unnecessary.

A further advantage when routine operation has been achieved is that many of the uncertainties of effluent composition have been eliminated; in addition, environmental monitoring will have established the identity of the critical nuclide(s) if the discharge is proving to be radiologically important. The greater accuracy with which the level of contamination (and thus public exposure) in relation to discharge rate is known substantially increases the accuracy and dependability of the estimate of environmental capacity.

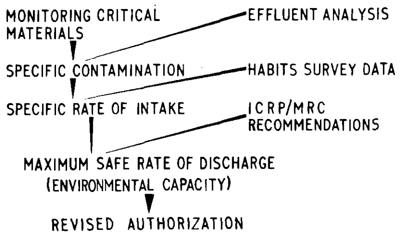

FIG. 2. Operational evaluation of environmental capacity due to internal exposure.

Although this is typical of the general situation in an operational phase, there will be instances where a modified procedure is necessary when contamination has not reached measurable levels in critical materials. Effluent composition will not be a problem, since direct experience will be available, eliminating the main source of uncertainty in the calculation, but a hydraulic model must be retained unless an indicator material shows measurable levels of contamination. Any radionuclide whose rate of discharge is known will be suitable, provided that concentration factors between critical and indicator materials and the water mass are known.

2.2.2. External exposure

No important differences of procedure exist which have not already been mentioned for the computational model used for assessment of environmental capacity due to internal exposure.

3. UK EXPERIENCE IN PRACTICE

The foregoing procedures have evolved over a period of years and have been refined in the light of experience gained as successive sites in the UK nuclear power programme have been developed. Sites occur in a wide range of environments, from lakes and river systems to open coastal waters. Examples appropriate to this symposium have been selected to illustrate how the procedures described have worked out in practice.

3.1. River systems

Most of UK experience in this type of environment has been gained from river estuaries, which pose greater problems of assessment than entirely freshwater regimes such as the middle section of the River Thames, to which the three establishments of Harwell, Aldermaston and Amersham dispose of their low-level liquid wastes after treatment. Discharges from these sites are limited by consideration of genetically-significant exposure, and consumption of drinking water is the critical pathway. There is no foodstuff of any potential importance, fishing being mainly for sport, nor is there an external exposure pathway.

3.1.1. Bradwell

Bradwell, a nuclear power station of the Magnox type and one of the first to come into commercial operation, is situated on the estuarine reaches of the River

Blackwater, to which discharges of liquid radioactive waste are made after dilution with the station cooling water. The River Blackwater is typical of many British estuaries and the hydraulic model used in early assessment is a good example of those used elsewhere. This has been described extensively, for instance by Preston[5], and at its simplest envisages an input of fresh water into a tidal plug from which water is lost at its downstream end to the sea. Sediment interaction plays an important role, though the extent varies according to the radionuclide concerned. The sediment load in the Blackwater is sufficient to strip a large proportion of such readily absorbed nuclides as cobalt-60 and zinc-65 from the water, and because British estuaries tend to retain sediment they can become sinks for such nuclides. Indeed, in some environments where the sole exposure pathways are external, control has to be exercised on the basis of a mathematical model which supposes that none of the activity is lost from the estuary except by radioactive decay. In dealing with internal exposure and uptake by biota, short-term mixing trends are unimportant, because rates of attainment of equilibrium between radionuclides and the critical materials (that is, foodstuffs) are relatively slow. For the same reason isolated peak concentrations in effluent discharges, and the transient higher levels they may generate in the receiving water mass, are unimportant and can be disregarded, assessment of environmental capacity being based on mean rates of disposal. Short-term fluctuations in effluent composition can also be ignored, though long-term trends can have a substantial effect.

The importance of effluent composition and the impact it has on control is well illustrated by the history of Bradwell. From the outset the especial importance of zinc-65 was recognized and control was based primarily on it. The critical exposure pathway is well known[6], involving oysters which are farmed extensively in the estuary. Zinc-65 concentrates to a high degree in oyster flesh, consumption of which leads to total body being the critical organ, with local oyster fishermen and their families as the most highly exposed population. A simple model predicted an environmental capacity of 0.2 Ci/day; when refined by allowing for interaction with sediment (this being calculated to remove 75% of the zinc, which is then no longer biologically available), the estimate rises to 0.8 Ci/day. Soon after operation started it became possible to measure contamination which was related to effluent data; the environmental capacity was then recalculated without recourse to a hydraulic model of any kind and was found to be 0.5 Ci/day.

The importance of zinc-65 was thus quickly confirmed and dominated the scene for a number of years, even though it has never been responsible for more than a very small proportion of the radioactivity discharged. In the first few years of operation the effluent consisted predominantly of activation products, and although the composition changed considerably, none of those other constituents, not even those discharged in many times larger amounts than zinc-65, had any environmental impact. Of the many nuclides found in the effluent, most of the activity was due to sulphur-35 and tritium; their negligible impact is due to a combination of lower radiotoxicity than zinc-65 and their environmental behaviour. Neither reconcentrates in oyster flesh, but zinc-65 concentrates to a very high degree, by a factor of more than 10^5.

In due course, as the station reached equilibrium operating conditions, the composition of the effluent underwent a further substantial change. Traces of activation products appeared in oysters, notably cobalt-60 and iron-55 but at concentrations which were insignificant by comparison with zinc-65. Excluding tritium, the

TABLE I. ENVIRONMENTAL CAPACITY OF THE BLACKWATER ESTUARY TO RECEIVE LIQUID EFFLUENT FROM THE BRADWELL NUCLEAR POWER STATION

Radionuclide	Environmental capacity, curie. $year^{-1}$*	
	Based on GI tract exposure	Based on total body exposure
Zinc-65	3.7×10^{2}	1.9×10^{2}
Silver-110m	3.2×10^{1}	7.4×10^{3}
Caesium-134	3.5×10^{6}	6.3×10^{4}
Caesium-137	5.6×10^{6}	1.4×10^{5}
Sulphur-35	4×10^{8}	2×10^{7}
Tritium	2×10^{8}	4×10^{8}

*Based on a consumption rate of 75 $g.d^{-1}$ of oyster flesh.

waste gradually changed from being activation product-dominated and became largely composed of fission products, especially caesium-137 and -134. These nuclides appeared in oyster flesh but only at low concentrations, insignificant compared with those of zinc-65 which was not displaced from its position of critical nuclide until silver-110m appeared[7]. Like zinc-65 this nuclide is produced by neutron activation and bears other similarities; in particular, it makes up only a very tiny proportion of the effluent and owes its radiological significance to an extemely high concentration factor in oyster flesh, at least as high as that for zinc-65 and probably marginally higher. However, and in contrast to zinc-65, its behaviour in the water mass is largely conservative.

Over a number of years the environmental distribution and behaviour of these nuclides has been studied in considerable detail, and this has made it possible to calculate values of environmental capacity for a range of nuclides along the lines discussed in section 2.2. Individual values are quoted in Table I in respect of exposure of the two most important body organs. Although several nuclides have been detected in oyster flesh, none of this contamination has great radiological significance, the highest level experienced representing only a fraction of 1% of the ICRP-recommended dose limit. A breakdown of exposure calculated for 1971 is given in Table II and shows that the maximum level of exposure to any body organ

TABLE II. RADIOLOGICAL IMPORTANCE OF OYSTERS IN THE BLACKWATER ESTUARY

Radionuclide	Mean concentration in oyster flesh, pCi g^{-1} (wet)	Radiological importance*		
		Bone	GI tract	Total body
Zinc-65	0.5	0.002	0.009	0.017
Silver-110m	1.5	< 0.001	0.17	< 0.001
Caesium-137	0.1	0.007	< 0.001	0.017
	Total	0.01	0.18	0.035

*Expressed as a percentage of the ICRP-recommended dose limit.

of an oyster consumer was 0.2% of the ICRP dose limit. This table also demonstrates the fine balance between zinc-65 and silver-110m in terms of the exposure for which they are responsible. Quite a small change in their relative discharge rates would be sufficient to alter the identities of critical nuclide and critical organ.

3.2. Lacustrine systems

Only one such environment is used for disposal of radioactive waste from a nuclear power station in the UK, the freshwater Lake Trawsfynydd in a mountainous region of North Wales. This has posed new problems, although the same basic principles have been applied in assessing environmental capacity as have been described previously, and it provides a good example of the research effort which may be needed, as well as further illustrating the importance of a knowledge of effluent composition.

3.2.1. Trawsfynydd

Like Bradwell, the power station is of the Magnox type but has a larger output - 500 MW(E) compared with 300 MW(E). Water from Lake Trawsfynydd, an artificially created lake of the soft-water dystrophic type (3 ppm Ca^{++} and 0.4 K^{+}), is used for condenser cooling and receives low-level liquid radioactive effluents as it is returned to the lake.

In the first, pre-operational, assessment of the environmental (lake) capacity, a simple hydraulic model was adopted, a constant volume of the lake being assumed with a constant outflow rate and no partition between lake water, suspended matter, and lake bed. The equation used to derive the estimated steady-state concentrations of each radionuclide in the lake water (C_{ss}) ($pCi.l^{-1}$) was therefore:

$$C_{ss} = \frac{I}{KV}, \qquad (1)$$

where I = input of radionuclide to the lake ($pCi.d^{-1}$)

V = volume of the lake (l)

$K = (\lambda_R + \frac{r}{V})$ the effective removal constant (d^{-1}), (2)

where r = rate of run-off of water from the lake ($l.d^{-1}$)

λ_R = physical decay constant of the radionuclide (d^{-1}).

Under the conditions assumed, the half-time calculated for lake water is approximately 50 days, so that for many radionuclides, such as caesium-137 or strontium-90, with relatively long radioactive half-lives, $K = r/V$ and the estimated $C_{ss} = 2.8$ $pCi.l^{-1}$ for a discharge rate, I, of 1 $mCi.d^{-1}$. For the others, such as phosphorus-32 ($t_{\frac{1}{2}}$ 14.3 days), radioactive decay has a considerable influence on C_{ss} which for this radionuclide is much lower at 0.63 $pCi.l^{-1}$.

The critical exposure pathway for discharges into the lake was identified at an early pre-operational stage as being due to consumption of lake fish, involving internal exposure only. The lake has been a noted fishery for many years, trout (Salmo trutta) being the principal species, though a small minority of the exposed population, essentially anglers and their families, also eat perch (Perca fluviatilis).

Early predictions of the effluent composition resembled those for Bradwell, in that the waste was expected to consist predominantly of neutron activation products. It was anticipated that phosphorus-32, with a suggested concentration factor [trout flesh/lake water] of 10^5 might be important, in which case bone could have been the critical organ. Zinc-65, cobalt-60 and antimony-124 might also make significant contributions to contamination of fish and public radiation exposure, in which case the lower large intestine could have become the critical organ. It was clear that Lake Trawsfynydd would have a lower capacity than the environments of other UK nuclear sites, so that radioecological studies utilizing fallout[8] were initiated prior to station operation. This demonstrated the behaviour of the fission products caesium-137 and strontium-90 and showed how important they could become if significant amounts were to appear in the effluent requiring disposal.

The station began operating at full power in November 1965, but early discharges were minimal. As was found during the early operational stage at other Magnox stations, the effluent consisted primarily of activation products but the anticipated importance of phosphorus-32 did not materialize. Contamination by artificial radioactivity of lake fish and public exposure from it remained low, being due only to fallout. In time, and following the pattern experienced elsewhere, the emphasis changed and excluding tritium the predominant radionuclides in the effluent became those of caesium - mainly caesium-137 with smaller amounts of caesium-134. In contrast to Bradwell these became the critical nuclides and

TABLE III. ENVIRONMENTAL CAPACITY OF LAKE TRAWSFYNYDD TO RECEIVE RADIOACTIVE LIQUID EFFLUENT FROM THE TRAWSFYNYDD NUCLEAR POWER STATION

Radionuclide	Environmental capacity, curie.year^{-1}*	
	Based on bone exposure	Based on total body exposure
Caesium-137	3.4×10^1	1.4×10^1
Caesium-134	4.6×10^1	6
Strontium-90	4.2×10^1	7.4×10^1
Calcium-45	6×10^2	5×10^3
Sulphur-35	2×10^6	1×10^6
Tritium	8×10^6	1×10^7

*Based on a consumption rate of 100 g.d^{-1} of trout flesh.

caesium-137 has been detected widely - in biota, bed sediment and water from the lake. Radioecological research was therefore intensified; measurements of caesium-137 in both effluent and lake water for the five-year period October 1967 to September 1972 demonstrated that for a discharge rate of 1 mCi.d^{-1} the steady state concentration observed in lake water was 2.4 pCi.l^{-1}[9], in good agreement with the value of 2.8 pCi.l^{-1} deduced for use in the original site assessment. A further outcome of this research has been the realization that the means by which uptake of caesium occurs is far from simple, the lake bed playing an important intermediate role, since almost all of the activity reaching fish comes via food and very little directly from water. Sampling of lake bed mud shows that up to the present time contamination is confined to the surface layers and that penetration rates are slow[9]. Whilst caesium-137 concentrations in both trout and perch flesh ultimately reflect changes in the caesium-137 level in lake water, there is a long lag time. This lag time will be a function not only of the biological half-times of caesium in fish flesh[8], but also of the transit time through the food chain, which has been examined by study of the caesium-137:caesium-134 ratio. This ratio in effluent has changed from about 12:1 during early operation to about 4:1 in recent times as the power station has reached equilibrium operation. During early operation the quantities of both nuclides discharged were insufficient to alter the situation in the lake, in which virtually all the caesium was caesium-137. In due

TABLE IV. RADIOLOGICAL IMPORTANCE OF TROUT IN LAKE TRAWSFYNYDD, 1971

Radionuclide	Mean concentration in trout flesh, pCi g^{-1} (wet)	Radiological importance*		
		Bone	GI tract	Total body
Caesium-137	12	1.09	0.07	2.7
Caesium-134	2.0	0.13	0.02	1.1
Strontium-90	0.13	1.48	0.01	0.84
	Total	2.70	0.10	4.64

*Expressed as a percentage of the ICRP-recommended dose limit.

course power station discharges took effect and changes of caesium-137:caesium-134 in effluent have been rapidly reflected in lake water. However, the ratio for fish flesh takes at least two years to reach equilibrium with input to the lake, and the concentration of caesium-137 in trout flesh has not yet reached the expected steady state with respect to lake water.

Further study has been made of strontium-90, the next most important nuclide after those of radiocaesium, and has led to examination of the possible role of calcium-45, because calcium behaves similarly to strontium. In practice, calcium-45 has been only a minor component of the effluent and of very low potential importance, as will be seen from Table III. This lists the most recent estimates of environmental capacity for a series of radionuclides, those included being either major constituents in the effluent and/or those which are found in the critical material.

The three radionuclides caesium-137 and -134, and strontium-90 are effectively responsible for all the radiation exposure of the public following consumption of lake-fish, though not all of this exposure is power-station derived. A detailed assessment of radiation exposure through this pathway is summarized in Table IV, which shows how contamination and public exposure have been satisfactorily controlled. It also illustrates once more how a slight shift in the relative abundance of two radionuclides could change the identity of critical organ, which might be bone if strontium-90 were to rise relative to caesium-134 and -137.

TABLE V. ESTIMATES OF PUBLIC RADIATION EXPOSURE FROM LOW-LEVEL LIQUID RADIOACTIVE WASTE DISPOSAL TO LACUSTRINE AND RIVER SYSTEMS OF THE UNITED KINGDOM, 1971

Site	Exposure pathway	Maximum exposure of an individual; % of ICRP-recommended dose limit	Approximate total population gonad dose, man-rem*
Trawsfynydd	Fish	5	10^{0}
Harwell Aldermaston Amersham	Drinking water	< 1	10^{2}-10^{3}
Berkeley Oldbury	External dose	< 0.3	10^{-1}
Bradwell	Oyster flesh	0.2	10^{-3}
Chatham	External dose	< 0.1	10^{-1}

*Values may be compared with a policy dose limit for the country as a whole which is equivalent to 1.8×10^{6} man-rem per year.

4. CONCLUSIONS

The crucial test of any radioactive waste disposal policy must be the extent to which the public are exposed to radiation as a result of its application. Departmental policy within the UK is to restrict discharges so that they fall within an environmental capacity based on ICRP radiation exposure criteria, often by a very large margin; as a consequence, public radiation exposure should be small. Estimates of actual levels of radiation exposure, a requirement which the critical path approach is especially suited to meeting, are given in Table V and include both somatic and genetic considerations. These estimates show that a very high standard of radiation protection is maintained.

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DISCUSSION

E. KUNZ: I fully appreciate the practical value and the objective nature of the terms "limiting environmental capacity" and "provisional limiting environmental capacity", but the concept of "stipulated environmental capacity" is unclear to me in this connection. I can relate it to the environment only in respect of your Fig. 1, from which I infer that it is something less than "provisional limiting environmental capacity". Related to the operator however, it appears to describe rather his capacity than the capacity of the environment.

N. T. MITCHELL: Quite so, it reflects the quantity of waste whose release the operator can justify, and will be the quantity written into the authorization. As such it will never be more than the provisional environmental capacity for a first, provisional authorization for a site, or more than the limiting environmental capacity for a later, amended version.

R. R. A. PRIDE: Would you please comment further on the large values shown in your tables for environmental capacity, particularly for tritium and sulphur-35?

N. T. MITCHELL: In each case they are a reflection of the low basic radiotoxicity involved and of the lack of environmental reconcentration processes. I see no reason to doubt these values, high though they may be, and neither do I share the doubts expressed by Mr. Mawson, during the discussion on paper IAEA-SM-172/17 on Monday afternoon, regarding the credibility of this kind of value to others.

R. E. HOLMES: It would be interesting if we could examine in greater detail the intermediate role of the lake-bed in the ^{137}Cs ecological chain at Lake Trawsfynydd.

N. T. MITCHELL: Yes. This is an important subject but I fear that lack of time prevents a detailed discussion here. Details will be published in the paper quoted as Ref. [7], in addition to the preliminary work, a paper which is already available (Ref. [8]). The lake-bed does play a vital intermediate role in the critical pathway for both radiocaesium nuclides and their entry via food is much more important than via water.

P. KAYSER: In some cases we know in advance that the limiting environmental capacity of a water body is very large in regard to the real needs of nuclear industry, e.g. when the water is not used for irrigation and fishing does not play an important role. In such cases is it prudent to determine the limiting environmental capacity in advance? That might encourage nuclear industry to discharge liquid wastes in amounts greater than the minimum necessary. Would it not be preferable to start with the minimum needs of industry and to estimate on that basis the irradiation which would thereby accrue to man?

N. T. MITCHELL: To us in the United Kingdom calculation of the limiting environmental capacity is the most logical first step. Judging by our experience, a combination of administrative control and technical inspection procedures, all applied by regulatory departments of the central government, is quite sufficient to prevent unjustifiable discharges being made. If one adopted the procedure you suggest, one might find that the needs of the operator as first assessed were larger than the limiting environmental capacity.

M. J. A. DELPLA: You have spoken of a safety factor of 0.1. Is this really a matter of safety? In other words, is there any danger beyond these amounts?

N. T. MITCHELL: I would say that it is a matter of caution and could be referred to in other ways. The exact figure used requires a value judgement, but in our experience to date a factor of 0.1 has never proved insufficient.

M. J. A. DELPLA: During the Symposium many references have been made to the concentrations of certain elements. Do the dose calculations in the 'critical group' also take account of the dilution factors arising during the transport of the radionuclides from the point of release to the critical group?

N. T. MITCHELL: These factors are taken into account as shown in Figs 1 and 2 and as discussed in section 2 of the paper.

A. MARTIN: I would like to make a general comment on all the papers concerned with environmental capacity. The general terminology seems to me to be inappropriate, since the term 'capacity' usually implies a quantity rather than a rate. The general concept also seems to be unsatisfactory particularly when — as indeed is usually the case — it is related to a dose rate in the environment. My objection is that no account is taken of the much greater significance of reaching the limiting dose rate, or even some fraction of it, if the nuclide is long-lived in the environment, e.g. ^{137}Cs, than if it is short-lived, e.g. ^{131}I.

The situation is analogous to that which arises in setting limits of internal radioactive contamination; here, the concept of maximum permissible body burden, related to dose rate, has given way to maximum permissible annual intake, related to dose commitment. I feel that in the same way release limits should be related to dose commitment and not to dose rate.

N. T. MITCHELL: You have, of course, raised some wide-ranging issues here, but nevertheless I feel that your principal difficulty is one of semantics, a problem which becomes more severe in an international context. As far as the term 'capacity', is concerned, I conceive of this as a property, in the light of which I believe our aims become clearer.

I disagree with you regarding the nuclides of differing half-life, because limiting environmental capacity as we envisage it is based on the equilibrium situation.

S. PRÊTRE: In your Tables IV and V you indicate doses as percentages of the ICRP limits. To what limit are you referring — to the limit for occupationally exposed persons or to that for the general public? Does your figure of 5% mean 250 mrem/yr or 25 mrem/yr?

N. T. MITCHELL: We are concerned with protection of the public and have thus compared all our measurements and calculations with the dose limits which ICRP recommend as acceptable for the public. In consequence, the 4.6% quoted in Table IV is equivalent to 23 mrem/yr. This is a pessimistic estimate based on the individual with the highest known exposure (consumption taken to be 100 g of fish per day). The exposed group was too small for us to be able to identify a critical group that would meet the ICRP 'definition'.

ASSESSING POTENTIAL RADIOLOGICAL IMPACTS TO AQUATIC BIOTA IN RESPONSE TO THE NATIONAL ENVIRONMENTAL POLICY ACT (NEPA) OF 1969*

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Abstract

ASSESSING POTENTIAL RADIOLOGICAL IMPACTS TO AQUATIC BIOTA IN RESPONSE TO THE NATIONAL ENVIRONMENTAL POLICY ACT (NEPA) OF 1969.

Environmental impact statements for nuclear power stations, prepared at Oak Ridge National Laboratory for the US Atomic Energy Commission, include an assessment of potential radiological impacts to organisms in the environment, in addition to assessments of impacts to man. These impact statements reflect the USAEC's implementation of the National Environmental Policy Act of 1969, which requires that federal agencies prepare a detailed statement for any major action that significantly affects the quality of the environment.

Assessments of impacts for nuclear power stations sited near bodies of water receiving reactor effluents indicate that aquatic biota may receive higher doses than do terrestrial biota and that potential doses due to internal deposition of radionuclides may be several hundred times greater than potential external doses from immersion in contaminated water. Estimated doses are listed in each environmental impact statement for aquatic plants, invertebrates (crustaceans and molluscs) and fish. Where appropriate, doses are also estimated for waterfowl, shore birds or muskrats. A steady-state bioaccumulation factor (concentration of radionuclide in biota/concentration of radionuclide in water) is used to estimate the body burden of each radionuclide for calculating internal dose. In most cases, site specific bioaccumulation factors are not available, and literature values, which usually cover a wide range, must be considered for the calculations.

The radionuclides released from water-cooled reactors to the aquatic system differ from plant to plant, depending upon the type of radioactive waste cleanup system, but the most important dose-contributing radionuclides are usually ^{134}Cs and ^{137}Cs. Radionuclides of lesser importance are ^{58}Co, ^{60}Co, ^{90}Sr, ^{91}Sr and ^{54}Mn. Although some uncertainty exists in the calculated potential doses, the upper limit doses to fish and invertebrates calculated under the most pessimistic assumptions usually do not exceed a few rems per year. Evaluation of the doses calculated thus far suggests that no biological effects of irradiation will likely be detected in the aquatic biota and that the radiological impact to the biota is likely to be trivial compared to potential effects from impingement of fish on water intake structures, from entrainment of fish and other aquatic organisms in condenser cooling water, and from temperature increases the biota experience in the receiving waters.

INTRODUCTION

The National Environmental Policy Act (NEPA) of 1969 is probably the most significant United States statute pertaining to environmental protection [1]. It is a strong act that was enacted at a time of acute environmental awareness; its provisions apply to all agencies of the federal government. The Act established a council on Environmental Quality which reports to the President of the United States. Any governmental agency responsible for actions significantly affecting the quality of the

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TABLE I. NUCLEAR POWER STATIONS CONSIDERED IN THIS PAPER

Nuclear Power Stations	Total Rated Capacity [MW(e)]	Location	Body of Water Receiving Liquid Effluents
Pressurized Water Reactors			
Indian Point Units 1 and 2	275 and 873	Peekskill, N.Y.	Hudson River Estuary
William B. McGuire Nuclear Stations Units 1 and 2	1150 each	Mecklenburg County, N.C.	Lake Norman (artificial freshwater reservoir)
Joseph M. Farley Nuclear Plant Units 1 and 2	898 each	Houston County, Ala.	Chattahoochee River (fresh water under influence of dams)
Surry Power Station Units 1 and 2	823 each	Surry County, Va.	James River (a major tributary of the lower Chesapeake Bay Estuary)
Oconee Nuclear Station Units 1, 2, and 3	922 each	Oconee County, S.C.	Lake Keowee (artificial freshwater reservoir)
Palisades Nuclear Generating Plant	821	Van Buren County, Mich.	Lake Michigan (fresh water)
Diablo Canyon Units 1 and 2	1060 each	San Luis Obispo County, Calif.	Pacific Ocean
North Anna Power Station Units 1 and 2	980 each	Louisa County, Va.	Lake Anna (artificial freshwater lake flowing into North Anna River, thence to three other rivers, eventually reaching the upper Chesapeake Bay Estuary)
San Onofre Nuclear Generating Station, Units 2 and 3	1140 each	San Diego County, Calif.	Pacific Ocean
Arkansas Nuclear One, Unit 2	950	Pope County, Ark.	Lake Dardanelle (artificial freshwater reservoir)
Boiling Water Reactors			
Vermont Yankee Nuclear Power Station	540	Vernon, Vt.	Connecticut River (fresh water)
The Edwin I. Hatch Nuclear Plant Units 1 and 2	786 and 795	Appling County, Ga.	Altamaha River (fresh water – reaches Atlantic Ocean 100 miles downstream)
Shoreham Nuclear Power Station	820	Brookhaven, N.Y.	Long Island Sound (marine – Atlantic Coastal Waters)
Limerick Generating Station Units 1 and 2	1100 each	Pottstown, Pa.	Schuylkill River (fresh water flowing into Delaware River)
Newbold Island Nuclear Generating Station Units 1 and 2	1088 each	Burlington County, N.J.	Delaware River (fresh water under tidal influence)
Peach Bottom Atomic Power Station Units 1 and 2	1065 each	York County, Pa.	Susquehanna River (fresh water flowing into upper Chesapeake Bay Estuary)

natural environment is required under NEPA to submit an environmental impact statement outlining the potential impacts of the proposed action and an environmental cost-benefit analysis.

The broad directive of NEPA and its interpretation in the courts have stimulated the U.S. Atomic Energy Commission, as the responsible governmental agency for construction of nuclear power stations, to quickly and dramatically expand its expertise into areas of nonradiological impact. In some areas there is a present need for certain types of environmental impact data which must be factored into a cost-benefit analysis, but which are still to be quantified by scientific investigation. Meeting the requirements of NEPA has had a profound effect on the licensing of nuclear power stations in the United States, and in some cases delays in operation have resulted when licensing has been held up due to litigation.

The potential impacts to man and his environment from large nuclear power stations can be extensive. These impacts include environmental disturbances during site preparation, influx of construction workers on the socioeconomic structure of communities close to construction sites, loss of forested land to construction of transmission line rights-of-way, esthetic qualities of the completed power station, and effects related to removal and discharge of cooling water. Public health and safety have always been a paramount concern of the nuclear industry, but NEPA now requires industry and government to extend that concern to all components of the environment. Thus, we now find that doses to biota are computed as part of the assessment of nuclear power plant operation, in addition to the dose assessments for man.

Effluent releases to the environment are limited by state law in each nuclear plant's operating license and by the Coded Federal Register (10CFR20 and 10CFR50) for the protection of man, but there are no specific laws to protect biota from radioactive effluents. The philosophy expressed by many has been, "If we adequately protect man, we adequately protect all of the organisms in the natural environment." Since some aquatic biota concentrate radionuclides in their tissues up to thousands of times greater than in the ambient water environment, the question can be asked, "Will these concentrations result in excessive doses?"

It is the purpose of this paper to examine and summarize the estimated radiation doses to biota from sixteen nuclear power stations (see Table I) reported in environmental impact statements prepared at Oak Ridge National Laboratory (ORNL) for the U.S. Atomic Energy Commission [2].

METHOD OF DOSE ASSESSMENT

The equations used at ORNL for dose estimation have been adequately covered by Blaylock [3], Booth et al. [4], and in each of the environmental impact statements [2] from which the data in this paper have been compiled. One assumption common to each assessment is that the radioactivity is uniformly distributed in the receiving body of water and in the reference organism. The effective absorbed energy used in the calculation of internal dose from each radionuclide is a function of the size of the organism. A conservative approach has been used in the environmental impact statements, because all calculations of internal dose to aquatic plants, invertebrates, and fish have utilized effective absorbed energies tabulated in ICRP Report 2[5] for 30-cm diameters. In almost all cases the effective diameter of aquatic biota is less than 30 cm; however, effective absorbed energies corresponding to this large diameter were chosen because they maximize the dose from gamma photons and x rays. This conservative application is a safeguard against the possibility that the bioaccumulation factor has been underestimated for a certain type of organism or that the radiosensitivity has been underestimated.

The bioaccumulation factor, C, for aquatic biota enters into the calculation of dose as a representation of the relationship between the concentration of radioactivity in biota relative to the water environment. The bioaccumulation factor for a radionuclide is expressed mathematically as

$$C = \frac{\mu\text{Ci per g of biota}}{\mu\text{Ci per g of water}}$$

where all concentrations are measured under steady-state conditions. Bioaccumulation factors are not physical constants, but are biologically and environmentally influenced factors that have been shown to vary with the physiochemical nature of particular sites, changes in food habits of biota and their prey, radioactive decay and biological elimination, occasional ingestion of sediments, homeostatic control of element concentration in tissues, etc. [6-8].

The bioaccumulation factors used in estimating dose for the sixteen nuclear power stations reported in this paper are listed in Table II. Parts A and B of this table list the bioaccumulation factors for freshwater and saltwater, respectively. These factors were chosen from several reference sources which are noted at the bottom of the table. It would be naive to believe that the bioaccumulation of a radionuclide in a class of organisms from many different sites could be accurately represented in such a simplistic manner as a universal bioaccumulation factor. Rather, it has been assumed that the tabulated bioaccumulation factors are probably conservative and, therefore, would lead to overestimation of dose. If the calculated doses are still acceptable when conservative estimates for effective absorbed energies and bioaccumulation factors are used, then it is less critical to know precisely the site and species specific bioaccumulation factors.

The immersion doses for aquatic organisms living in the receiving bodies of water were calculated for both photons and beta particles by using the infinite source formula presented by Hine and Brownell [9].

RELEASE CHARACTERISTICS

The basic assumptions used in environmental impact statments are that the plant will operate over a projected 30-year period and that waste effluents (at activities not exceeding the permissible release limits) will be released on a continuous basis over this period. Furthermore, an instantaneous steady state is assumed between the concentration of all radionuclides in the receiving water and their concentrations in aquatic biota. This later assumption results in initially conservative estimates of internal dose, especially for longer lived radionuclides that require a period of months, or years in some cases, to approach a steady state. The magnitude of conservativeness is equal to the ratio of the steady-state body burden or concentration divided by the body burden or concentration at some reference time prior to steady state.

Concentrations of the principal dose-contributing radionuclides in the receiving waters of the sixteen nuclear power stations are listed in Table III. Although all nuclear power stations listed in Table III are expected to discharge approximately 5 Ci/year per unit (excluding tritium), the concentrations (μCi/ml) listed for identical radionuclides vary by several orders of magnitude. These variations between power stations are due mostly to assumed differences in radwaste systems and the decontamination factors used for the various filters, ion exchangers, and evaporators.

TABLE II. BIOACCUMULATION FACTORS USED IN ENVIRONMENTAL IMPACT STATEMENTS TO ESTIMATE INTERNAL DOSES TO FRESHWATER AND SALTWATER BIOTA

Part A: Freshwater Bioaccumulation Factors

Radionuclide	Plants[a]	Mollusks and Crustaceans	Fin Fish	Waterfowl and Muskrats[b]
^{89}Sr	3.0E+3[c,g]	4.0E+3[c]	1.5E+2[c]	6.5E+3
^{90}Sr	3.0E+3[c]	4.0E+3[c]	1.5E+2[c]	9.0E+3
^{91}Sr	3.0E+3[c]	4.0E+3[c]	1.5E+2[c]	5.1E+1
^{91}Y	1.0E+4[d]	1.0E+3[d]	1.0E+2[d]	8.3E+0
^{93}Y	1.0E+4[d]	1.0E+3[d]	1.0E+2[d]	6.1E–2
^{129m}Te	1.0E+3[e]	6.1E+3[e]	4.0E+2[e]	3.6E+2
^{132}Te	1.0E+3[e]	6.1E+3[e]	4.0E+2[e]	9.3E+1
^{131}I	2.0E+2[c]	1.0E+3[c]	5.0E+1[c]	2.1E+2
^{133}I	2.0E+2[c]	1.0E+3[c]	5.0E+1[c]	2.5E+1
^{135}I	2.0E+2[c]	1.0E+3[c]	5.0E+1[c]	8.0E+0
^{134}Cs	2.5E+4[c]	1.1E+4[c]	1.0E+3[c]	2.3E+5
^{136}Cs	2.5E+4[c]	1.1E+4[c]	1.0E+3[c]	3.9E+4
^{137}Cs	2.5E+4[c]	1.1E+4[c]	1.0E+3[c]	2.5E+5
^{140}La	1.0E+4[d]	1.0E+3[d]	1.0E+2[d]	2.4E–1
^{24}Na	1.6E+2[d]	2.7E+1[d]	3.2E+1[d]	1.3E+1
^{32}P	1.0E+5[d]	1.0E+5[d]	1.0E+5[d]	1.3E+5
^{54}Mn	3.5E+4[c]	1.4E+5[c]	2.5E+1[d]	2.8E+3
^{58}Co	2.5E+3[c]	1.5E+3[d]	5.0E+2[d]	9.0E+2
^{60}Co	2.5E+3[c]	1.5E+3[d]	5.0E+2[d]	1.0E+3
^{239}Np	1.0E+3[d]	2.8E+2[d]	1.0E+4[d]	2.4E–2

Part B: Saltwater Bioaccumulation Factors[f]

Radionuclide	Plants[a]	Mullusks and Crustaceans	Fin Fish	Waterfowl and Shore Birds[b]
^{89}Sr	2.0E+1	1.0E+0	1.0E+0	4.3E+1
^{90}Sr	2.0E+1	1.0E+0	1.0E+0	6.0E+1
^{91}Sr	2.0E+1	1.0E+0	1.0E+0	3.4E–1
^{91}Y	3.0E+2	1.0E+2	3.0E+1	2.5E–1
^{93}Y	3.0E+2	1.0E+2	3.0E+1	1.8E–3
^{129m}Te	1.0E+3	1.0E+2	1.0E+1	3.6E+2
^{132}Te	1.0E+3	1.0E+2	1.0E+1	9.3E+1
^{131}I	1.0E+4	1.0E+2	2.0E+1	1.1E+4
^{133}I	1.0E+4	1.0E+2	2.0E+1	1.2E+3
^{135}I	1.0E+4	1.0E+2	2.0E+1	4.0E+2
^{134}Cs	1.0E+1	5.0E+1	3.0E+1	9.3E+1
^{136}Cs	1.0E+1	5.0E+1	3.0E+1	1.5E+1
^{137}Cs	1.0E+1	5.0E+1	3.0E+1	1.0E+2
^{140}La	3.0E+2	1.0E+2	3.0E+1	7.2E–3
^{24}Na	1.0E+0	1.0E+0	1.0E+0	1.0E+0
^{32}P	1.0E+5	1.0E+4	1.0E+4	1.3E+5
^{54}Mn	1.0E+4	5.0E+4	3.0E+3	8.0E+2
^{58}Co	1.0E+2	1.0E+4	1.0E+2	3.6E+1
^{60}Co	1.0E+2	1.0E+4	1.0E+2	4.0E+1
^{239}Np	1.0E+3	2.8E+2	1.0E+4	2.4E–2

[a] Includes algae and macrophytes.

[b] Bioaccumulation factors for waterfowl, muskrats, and shore birds as listed in Environmental Impact Statements. The impact statements give assumptions and procedures which were used to calculate these factors for a steady state condition. Essentially, the factors apply to a vertebrate mammal or bird deriving all of its food intake from consumption of aquatic plants or algae. It was further assummed in the statements that daily food consumption equalled one-tenth the animal's body weight, and that assimilation and elimination were equal to the values listed in ICRP Report 2 (1959) for man.

[c] D. E. Reichle, P. B. Dunaway, and D. J. Nelson, *Nuclear Safety* 11(1): 43–55 (1970).

[d] W. H. Chapman, L. H. Fisher, and M. W. Pratt, UCRL-50564 (1968).

[e] Calculated from stable element analysis listed in the *Farley Nuclear Power Station Environmental Report*, Georgia Power and Light Company, 1972.

[f] All bioaccumulation factors listed in Part B are from A. M. Freke, *Health Physics* 13, 743–758 (1967).

[g] 3.0E+3 is equivalent to $3.0 \times 10^{+3}$.

TABLE III. ESTIMATED CONCENTRATIONS (μCi/ml) LISTED IN ENVIRONMENTAL IMPACT STATEMENTS FOR PRINCIPAL DOSE CONTRIBUTING RADIONUCLIDES AT POINT OF LIQUID DISCHARGE

Nuclear Power Station	^{89}Sr	^{90}Sr	^{91}Sr	^{91}Y	^{93}Y	^{129m}Te	^{132}Te	^{131}I	^{133}I	^{135}I	^{134}Cs	^{136}Cs	^{137}Cs	^{140}La	^{24}Na	^{32}P	^{54}Mn	^{58}Co	^{60}Co	^{239}Np
Pressurized Water Reactors																				
Indian Point Units 1 and 2	2.5E-11[c]	5.0E-12	7.0E-14	1.7E-11	*a*	1.6E-12	1.1E-11	8.2E-09	3.5E-09	1.8E-09	5.6E-10	2.4E-10	6.5E-10	1.6E-13	2.5E-09	*a*	8.1E-10	6.0E-10	2.5E-10	2.0E-13
William B. McGuire Nuclear Stations Units 1 and 2	1.0E-12	3.5E-14	*a*	2.2E-10	*a*	8.6E-11	4.1E-11	2.3E-10	*a*	*a*	9.5E-10	3.2E-10	5.2E-11	1.3E-12	*a*	*a*	5.2E-11	1.8E-09	5.2E-11	*a*
Joseph M. Farley Nuclear Plant Units 1 and 2	9.0E-13	3.0E-14	*a*	1.8E-10	*a*	7.5E-11	3.5E-11	2.0E-10	*a*	*a*	8.1E-10	2.7E-10	4.4E-11	8.1E-13	*a*	*a*	4.4E-11	1.6E-09	4.4E-11	*a*
Surry Power Station Units 1 and 2	1.2E-11	3.8E-13	*a*	1.4E-11	*a*	9.3E-11	6.7E-10	7.8E-09	5.6E-09	1.3E-09	5.4E-09	1.8E-09	4.3E-09	*a*	*a*	*a*	1.4E-11	4.5E-10	1.3E-11	*a*
Oconee Nuclear Station Units 1, 2, and 3	8.1E-12	*a*	*a*	1.8E-10	*a*	6.1E-11	*a*	2.0E-10	*a*	*a*	6.4E-10	6.5E-11	4.5E-11	4.1E-12	*a*	*a*	5.9E-11	1.4E-09	5.7E-11	*a*
Palisades Nuclear Generating Plant	2.6E-10	9.6E-12	*a*	5.6E-09	*a*	1.9E-09	1.1E-09	9.0E-08	5.3E-09	8.4E-10	3.7E-09	*a*	3.7E-09	*a*	*a*	*a*	2.5E-10	7.5E-09	2.4E-10	*a*
Diablo Canyon Units 1 and 2	3.0E-12	1.0E-13	2.8E-13	4.1E-13	*a*	2.5E-11	1.5E-10	2.1E-09	7.1E-10	3.2E-11	2.4E-10	9.4E-11	1.7E-10	2.6E-12	*a*	*a*	1.6E-12	4.5E-11	4.5E-12	2.2E-12
North Anna Power Station Units 1 and 2	1.3E-13	4.3E-15	6.3E-14	1.5E-13	1.5E-15	1.0E-11	6.8E-11	2.0E-09	8.5E-10	1.7E-10	1.7E-11	6.6E-12	1.2E-11	1.4E-13	*a*	*a*	1.3E-13	4.1E-12	1.2E-13	1.4E-13
San Onofre Nuclear Generating Station, Units 2 and 3	6.5E-15	2.2E-16	1.5E-15	4.0E-13	4.1E-16	4.9E-14	3.1E-13	3.8E-11	2.5E-11	2.4E-12	7.3E-12	5.1E-13	6.2E-12	4.7E-15	*a*	*a*	4.9E-15	1.6E-14	5.1E-15	*a*
Arkansas Nuclear One, Unit 2	2.9E-13	1.0E-14	2.4E-15	1.8E-13	3.2E-15	2.3E-12	7.2E-12	3.4E-10	7.3E-11	*a*	2.2E-09	7.3E-10	1.6E-09	2.9E-13	*a*	*a*	2.0E-13	5.3E-12	5.6E-13	*a*
Boiling Water Reactors																				
Vermont Yankee Nuclear Power Station	7.9E-10	5.3E-10	8.0E-13	5.1E-10	7.9E-12	1.0E-11	7.2E-11	2.2E-09	2.5E-10	2.3E-13	4.5E-10	1.3E-10	3.4E-10	8.7E-10	3.8E-12	4.3E-13	5.9E-12	8.0E-10	8.0E-11	*a*
The Edwin I. Hatch Nuclear Plant Units 1 and 2	2.2E-11	1.7E-12	3.2E-13	3.4E-11	1.9E-11	2.2E-13	5.0E-12	1.1E-10	*a*	*a*	1.9E-10	7.0E-11	1.4E-10	1.0E-11	9.6E-12	5.2E-14	2.2E-13	3.0E-11	3.0E-12	*a*
Shoreham Nuclear Power Station	3.8E-10	2.5E-11	3.7E-13	1.9E-10	3.7E-12	7.7E-12	3.4E-11	1.0E-09	1.2E-10	1.1E-13	2.1E-10	6.2E-11	1.6E-10	4.2E-10	1.8E-12	1.3E-12	3.0E-12	3.6E-10	3.7E-11	*a*
Limerick Generating Station Units 1 and 2	2.1E-10	1.3E-11	3.4E-10	2.0E-11	2.5E-11	5.9E-12	7.3E-11	4.0E-09	1.2E-08	4.5E-09	2.5E-11	1.1E-11	2.1E-11	1.5E-10	9.1E-11	2.5E-14	4.5E-12	5.6E-10	5.6E-11	4.5E-11
Newbold Island[b] Nuclear Generating Station Units 1 and 2	1.4E-09	7.9E-11	*a*	2.9E-09	4.9E-09	3.0E-11	*a*	1.3E-09	3.0E-09	5.5E-10	1.5E-08	4.8E-09	1.1E-08	1.1E-09	1.6E-10	4.9E-12	1.0E-10	1.3E-09	1.3E-10	*a*
Peach Bottom Atomic Power Station Units 1 and 2	8.7E-11	5.0E-12	5.7E-11	6.0E-11	7.4E-11	1.5E-12	2.0E-11	1.2E-10	2.5E-10	3.3E-11	6.7E-11	2.5E-11	6.4E-11	7.7E-11	4.3E-12	2.5E-14	1.0E-12	5.7E-11	5.7E-12	*a*

[a] Radionuclide not listed in *Environmental Impact Statement for this nuclear power station.*

[b] According to Environmental Impact Statement for Newbold Island Nuclear Generating Station, listed concentrations represent 1:5 dilution with Delaware River water.

[c] 2.5E-11 is equivalent to 2.5×10^{-11}.

ESTIMATED DOSES TO AQUATIC BIOTA

The estimated internal doses (summed over all radionuclides) to the four types of biota range from less than 1 mrad/year to a maximum of 62,000 mrad/year (Table IV). This variation is due almost entirely to variations in source term (see Table III), because the assumptions and methods for calculating doses and the concomitant bioenvironmental data were standardized for each nuclear power station.

The results of immersion dose calculations tabulated in the environmental impact statements showed that this exposure mode was minor compared to exposure from internally deposited radionuclides. In all cases, immersion resulted in estimated doses not exceeding 1 mrad/year for all radionuclides combined.

The dose calculations for all sites except the Newbold Island Generating Station apply to the location of radioactive effluent discharge before mixing with the receiving body of water; therefore, nonconsideration of the true diluting capacity of the aquatic environment greatly overestimates the calculated doses. The environmental impact statement for the Newbold Island Station states that a 1:5 dilution of radioactive effluents was assumed before calculating doses to biota.

TABLE IV. SUMMARY OF INTERNAL DOSES (MILLIRADS PER YEAR) TO AQUATIC BIOTA

	Freshwater and Marine Plants	Mollusks and Crustaceans	Fin Fish	Muskrats, Waterfowl and Shore Birds
Pressurized Water Reactors				
Indian Point Units 1 and 2	900	1,500	42	*a*
William B. McGuire Nuclear Stations Units 1 and 2	700	390	37	3,600
Joseph M. Farley Nuclear Plant Units 1 and 2	600	330	32	2,200
Surry Power Station Units 1 and 2	4,600	2,200	1,800	22,000
Oconee Nuclear Station Units 1, 2, and 3	450	270	150	17,000
Palisades Nuclear Generating Plant	4,000	2,600	1,500	5,800
Diablo Canyon Units 1 and 2	1,300	9	<1	12
North Anna Power Station Units 1 and 2	24	57	4	70
San Onofre Nuclear Generating Station, Units 2 and 3	8	<1	<1	3
Arkansas Nuclear One, Unit 2	1,800	800	70	8,800
Boiling Water Reactors				
Vermont Yankee Nuclear Power Station	860	300	32	2,700
The Edwin I. Hatch Nuclear Plant Units 1 and 2	160	80	8	*a*
Shoreham Nuclear Power Station	120	59	2	68
Limerick Generating Station Units 1 and 2	270	480	59	370
Newbold Island Nuclear Generating Station Units 1 and 2	15,000	6,100	550	62,000
Peach Bottom Atomic Power Station Units 1 and 2	140	60	5	380

[a] No dose estimate listed in Environmental Impact Statement for this nuclear power station.

For ten of the power stations, the estimated internal doses to muskrats, waterfowl, and shore birds are higher than for the other three types of biota. In no case is the estimated dose to the fin fish higher than any of the other types of biota for a particular nuclear power station. Table II shows that for all radionuclides except ^{24}Na in freshwater and ^{239}Np in saltwater the bioaccumulation factors for fish do not exceed the factors for the other types of biota, indicating the importance of the bioaccumulation factor in determining the magnitude of the estimated dose. The two radionuclides which are exceptions are only minor contributors to dose and are not present in significant concentrations in the releases from most nuclear power stations (see Table III).

Although the number of radionuclides considered for calculating doses often numbered 50 to 60, only a few radionuclides were responsible for most of the estimated doses. This trend is clearly demonstrated by the data in Table V which show that approximately 50 to 99% of the estimated dose received by a given type of biota is attributable to only three radionuclides released by a nuclear power station. Radioisotopes of cesium, principally ^{134}Cs and ^{137}Cs, appear to be the greatest dose contributors. Although the data in Table V are expressed in terms of percent, the doses for individual radionuclides can be computed from the total doses in Table IV, which in turn were calculated from the concentrations of radionuclides listed in Table III for the point of discharge.

DISCUSSION

The conclusions published in the sixteen environmental impact statements relative to the potential impacts of the estimated doses compiled in Table IV were uniform in stating that no detectable adverse effect will be produced on the biota. Since, in most cases, the methods used in the environmental impact statements were selected to overestimate the doses, the conclusion of no expected detectable adverse effects is not surprising. Even at the highest calculated annual dose rate of 62 rads/year to a hypothetical terrestrial mammal or bird obtaining all of its diet from aquatic plants growing at the point of discharge at the Newbold Island site, there is no information to indicate that a detectable radiation effect would be found. However, it is stated in the Draft Environmental Impact Statement for Newbold Island that "An increased mutation rate in these organisms cannot be dismissed completely." A discussion of this possibility pointed out that increased mutation rates in mammals have been found only at dose rates considerably higher than those calculated for Newbold Island.

Aside from the conservative assumptions for calculating internal dose already discussed, the lowest yearly average flow on record for the Delaware River at the point of discharge is approximately five times the record low flow which was used in the calculations. Thus, a simple reduction factor of 5 of the published annual dose of 62 rads/year appears to be justified for the Newbold Island site. Another important scaling factor relates to the fraction of each organism's diet obtained from the point of discharge. This diet factor was assumed to be equal to 100% for all groups of biota; if the actual number were less than 100%, say 20%, then the dose estimates would need to be divided by another factor of 5. Combination of these two factors alone results in a factor of 25 reduction in the published dose estimates for the hypothetical mammal or bird consuming 20% of its annual diet at the point of discharge from the Newbold Island Generating Station.

The estimated doses for the Newbold Island Nuclear Generating Station present another interesting point for discussion. Table V shows that almost all of the estimated doses are due to radioisotopes of cesium, and

TABLE V. RADIONUCLIDES CONTRIBUTING THE GREATEST PERCENT OF THE ESTIMATED INTERNAL DOSE TO BIOTA

Nuclear Power Station	Freshwater and Marine Plants		Mollusks and Crustaceans		Fin Fish		Muskrats, Waterfowl and Shore Birds	
Pressurized Water Reactors								
Indian Point Units 1 and 2	^{134}Cs, 39% ^{54}Mn, 30% ^{137}Cs, 20%	89%	^{54}Mn, 73% ^{134}Cs, 9% ^{137}Cs, 5%	87%	^{134}Cs, 26% ^{137}Cs, 17% ^{24}Na, 10%	53%	[a]	
William B. McGuire Nuclear Stations Units 1 and 2	^{134}Cs, 70% ^{136}Cs, 14% ^{58}Co, 8%	92%	^{134}Cs, 54% ^{54}Mn, 18% ^{136}Cs, 11%	83%	^{134}Cs, 54% ^{58}Co, 28% ^{136}Cs, 10%	92%	^{134}Cs, 93% ^{137}Cs, 4% ^{136}Cs, 3%	~100%
Joseph M. Farley Nuclear Plant Units 1 and 2	^{134}Cs, 70% ^{136}Cs, 14% ^{58}Co, 7%	91%	^{134}Cs, 55% ^{54}Mn, 18% ^{136}Cs, 11%	84%	^{134}Cs, 53% ^{58}Co, 28% ^{136}Cs, 10%	91%	^{134}Cs, 91% ^{137}Cs, 4% ^{136}Cs, 3%	98%
Surry Power Station Units 1 and 2	^{134}Cs, 61% ^{137}Cs, 26% ^{136}Cs, 12%	99%	^{134}Cs, 55% ^{137}Cs, 23% ^{136}Cs, 11%	89%	^{134}Cs, 61% ^{137}Cs, 25% ^{136}Cs, 12%	98%	^{134}Cs, 59% ^{137}Cs, 38% ^{136}Cs, 2%	99%
Oconee Nuclear Station Units 1, 2, and 3	^{134}Cs, 73% ^{58}Co, 9% ^{91}Y, 4%	86%	^{134}Cs, 52% ^{54}Mn, 30% ^{58}Co, 9%	91%	^{134}Cs, 87% ^{58}Co, 5% ^{136}Cs, 5%	97%	^{134}Cs, 94% ^{137}Cs, 5% ^{136}Cs, 1%	~100%
Palisades Nuclear Generating Plant	^{134}Cs, 45% ^{137}Cs, 24% ^{91}Y, 15%	84%	^{134}Cs, 26% ^{131}I, 23% ^{137}Cs, 14%	63%	^{134}Cs, 28% ^{58}Co, 16% ^{137}Cs, 15%	59%	^{134}Cs, 54% ^{137}Cs, 42% ^{131}I, <1%	97%
Diablo Canyon Units 1 and 2	^{133}I, 92% ^{135}I, 6% ^{129m}Te, <1%	99%	^{58}Co, 52% ^{60}Co, 13% ^{133}I, 11%	76%	^{133}I, 28% ^{134}Cs, 19% ^{239}Np, 15%	62%	^{133}I, 92% ^{134}Cs, 2% ^{135}I, 2%	96%
North Anna Power Station Units 1 and 2	^{134}Cs, 36% ^{131}I, 14% ^{137}Cs, 14%	64%	^{131}I, 28% ^{132}Te, 26% ^{133}I, 23%	77%	^{132}Te, 28% ^{131}I, 23% ^{133}I, 19%	70%	^{134}Cs, 60% ^{137}Cs, 33% ^{131}I, 4%	97%
San Onofre Nuclear Generating Stations, Units 2 and 3	^{133}I, 50% ^{131}I, 40% ^{135}I, 8%	98%	^{133}I, 40% ^{131}I, 32% ^{135}I, 6%	78%	^{133}I, 34% ^{131}I, 27% ^{134}Cs, 20%	81%	^{131}I, 85% ^{133}I, 14% ^{135}I, <1%	~100%
Arkansas Nuclear One, Unit 2	^{134}Cs, 61% ^{137}Cs, 24% ^{136}Cs, 12%	97%	^{134}Cs, 63% ^{137}Cs, 24% ^{136}Cs, 12%	99%	^{134}Cs, 63% ^{137}Cs, 25% ^{136}Cs, 12%	~100%	^{134}Cs, 63% ^{137}Cs, 35% ^{136}Cs, 2%	~100%
Boiling Water Reactors								
Vermont Yankee Nuclear Power Station	^{140}La, 36% ^{134}Cs, 27% ^{137}Cs, 11%	74%	^{134}Cs, 33% ^{137}Cs, 14% ^{89}Sr, 11%	58%	^{134}Cs, 29% ^{58}Co, 14% ^{137}Cs, 12%	55%	^{134}Cs, 41% ^{90}Sr, 30% ^{137}Cs, 24%	95%
The Edwin I. Hatch Nuclear Plant Units 1 and 2	^{134}Cs, 55% ^{137}Cs, 23% ^{136}Cs, 13%	91%	^{134}Cs, 55% ^{137}Cs, 23% ^{136}Cs, 12%	90%	^{134}Cs, 54% ^{137}Cs, 23% ^{136}Cs, 12%	89%	^{134}Cs, 61% ^{137}Cs, 37% ^{136}Cs, 2%	~100%
Shoreham Nuclear Power Station	^{131}I, 71% ^{133}I, 16% ^{140}La, 4%	91%	^{58}Co, 69% ^{60}Co, 19% ^{59}Fe, 3%	91%	^{140}La, 27% ^{58}Co, 21% ^{131}I, 9%	57%	^{131}I, 88% ^{90}Sr, 4% ^{32}P, 3%	95%
Limerick Generating Station Units 1 and 2	^{140}La, 20% ^{91}Sr, 15% ^{133}I, 14%	49%	^{133}I, 40% ^{135}I, 23% ^{91}Sr, 11%	74%	^{32}P, 51% ^{133}I, 16% ^{135}I, 9%	76%	^{90}Sr, 54% ^{134}Cs, 17% ^{137}Cs, 11%	82%
Newbold Island Nuclear Generating Station Units 1 and 2	^{134}Cs, 52% ^{137}Cs, 21% ^{93}Y, 11%	84%	^{134}Cs, 56% ^{137}Cs, 23% ^{136}Cs, 10%	89%	^{134}Cs, 56% ^{137}Cs, 22% ^{136}Cs, 11%	89%	^{134}Cs, 61% ^{137}Cs, 35% ^{136}Cs, 2%	98%
Peach Bottom Atomic Power Station Units 1 and 2	^{134}Cs, 24% ^{140}La, 19% ^{93}Y, 16%	59%	^{134}Cs, 25% ^{137}Cs, 13% ^{132}Te, 7%	45%	^{134}Cs, 29% ^{137}Cs, 15% ^{91}Sr, 7%	51%	^{134}Cs, 45% ^{137}Cs, 32% ^{90}Sr, 20%	97%

[a] No dose estimate listed in Environmental Impact Statement for this nuclear power station.

Table III shows that the predicted concentrations of ^{134}Cs and ^{137}Cs are 500 to 600 times higher for Newbold Island than they are for any other power station. These higher concentrations of cesium radioisotopes were predicted because the station was assessed on the basis that all floor drain accumulations (10,000 gal/day) are to be processed through a deep bed demineralizer and discharged to the Delaware River in the blowdown. A cesium decontamination factor of unity was assumed in the analysis. Other plants recycle most of the floor drainage and use evaporators to remove the cesium with an assumed decontamination factor of approximately 1000.

Since only the Draft Environmental Impact Statement was available for the Newbold Island Nuclear Generating Station at the time of this writing, recommended changes in the radwaste system might still be incorporated into the final statement.

It appears that the radiological impact to aquatic biota exposed by routine nuclear power plant operations will be trivial compared to the impacts of impingement of fish on water intake structures and entrainment of eggs and larvae of aquatic organisms in condenser cooling water. The extent that impingement will be a factor depends upon the intake velocity of the water, the physical design of the intake structure, and the presence of small fish. These factors are discussed in detail in each environmental impact statement, and estimates of mortality from entrainment are given. A further environmental impact could occur if heated effluents do not receive sufficient mixing in receiving waters.

CONCLUSIONS

1. The radiological impact to aquatic biota from operation of light water-cooled nuclear power stations in the United States is not expected to cause any significant biological effects. Although in most cases the annual dose to aquatic biota is estimated to be less than 1 rad, a maximum dose of 62 rads was calculated for a hypothetical muskrat, waterfowl, or shore bird that derives all of its diet from aquatic plants growing at the point of discharge. For most of the nuclear sites, such a hypothetical animal probably does not exist. The value of such a calculation is that it establishes an upper limit of dose.

2. The most important radioisotope for internal dose is ^{134}Cs, which contributes more than half the dose to the biota at many sites. Next in importance are ^{54}Mn, ^{58}Co, ^{60}Co, ^{131}I, ^{133}I, and ^{137}Cs. Additional radionuclides making apparently significant contributions to internal dose at one or more sites include ^{24}Na, ^{32}P, ^{89}Sr, ^{90}Sr, ^{91}Y, ^{129m}Te, ^{132}Te, ^{135}I, ^{136}Cs, ^{140}La, and ^{239}Np.

3. Internal exposure is considered more important than external exposure, because the predicted immersion doses published in the environmental impact statements are only a small fraction (approximately less than 1%) of the predicted internal doses.

4. The predicted concentrations of radionuclides at the point of discharge vary over several orders of magnitude for the sixteen nuclear power stations reviewed in this paper. This large variability accounts for a concomitant variation in estimated doses.

5. The conservative assumptions used in dose estimations in the environmental impact statements are justified in most cases, because the information required and the methods of calculation are greatly simplified. In the few cases where conservative methods resulted in relatively high dose estimates (for instance, greater than 10 rads/year) the analysis could have been recalculated using more realistic data and assumptions. Only radioisotopes of cesium would be involved in these recalculations; all other radioisotopes contribute a negligible fraction of the doses estimated to be greater than 10 rads/year.

6. The most significant impacts to aquatic biota are expected to result not from radiation, but from impingement of fish on water intake structures, entrainment of aquatic organisms, and possibly in a few isolated cases from thermal loading of receiving waters.

REFERENCES

[1] The National Environmental Policy Act of 1969, Public Law 91-190, 91st Congress of the United States, January 1, 1970.

[2] Environmental impact statements are available for inspection by the public in the Commission's Public Document Room at 1717 H Street, N.W., Washington, D.C. 20545, and certain local, state, and federal government offices in the vicinity of each nuclear power station. Single copies can be obtained upon request to the U.S. Atomic Energy Commission, Washington, D.C. 20545, Attention: Deputy Director for Reactor Projects, Directorate of Licensing.

[3] BLAYLOCK, B. G., "Effects on aquatic biota of radioactive effluents from nuclear power stations," IAEA Panel on Radiation Effects on Population Dynamics in Ecosystems, Reykjavik, Iceland, 2-5 October 1972 (in press).

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DISCUSSION

I. L. BRISBIN, Jr.: I am very happy to see that impact effects are now being studied in connection with environmental components and populations other than man alone! How would you relate the requirements of the National Environmental Policy Act, as you have described them, to the concept of 'environmental capacity' as we have heard it described in other papers at this Symposium?

S. V. KAYE: I would like to point out that I am not an unreserved supporter of the environmental capacity concept. Limiting environmental capacity relates only to the maximum quantity of a radionuclide that may be released to a receiving environment. It is intended to protect man only, as I understand it, with no mention of biota. However, the experience of the nuclear industry indicates that if man is adequately protected, so will be the biota. Under NEPA we are attempting to assess the impact on biota with estimates of dose, not a release index.

C.A. MAWSON: Perhaps it would be a good idea to calculate your results as accurately as you can with the information available to you so that you end up with what you believe to be the real dose. Then, because of uncertainty about your data, you could apply a stated safety factor. This would avoid the danger of multiplication of safety factors in a way that is hidden from people unfamiliar with the details of your assumptions.

S. V. KAYE: I believe that 'real dose' is an 'unrealistic' term to use because any estimate of dose will have connected with it a certain range of uncertainty. Furthermore, I fail to see the difference between adding in the uncertainty in the course of the calculation process, or adding in a total uncertainty estimate at the end of the dose calculation — one ends up with the same answer. All the assumptions used in the calculations are given in detail in the environmental impact statements, so there is no information hidden from the public.

O. L. van der BORGHT: Was a study such as you have just described also done for nuclear fuel reprocessing plants?

S. V. KAYE: There are two nuclear fuel reprocessing plants in the United States of America preparing for initial commercial operation. Neither of these plants will discharge liquid effluents to water bodies; however, potential environmental impacts from gaseous radioactive emissions and chemical pollutants are evaluated in the impact statements.

A. MERKEL: I see that you do not list ^{65}Zn in your tables. This nuclide has of course been discussed in many other papers at the Symposium and so do we take it that ^{65}Zn emission is not a feature of the special reactor types with which you are here concerned?

S. V. KAYE: Very low concentrations of ^{65}Zn were predicted in the case of these light water reactors using zirconium-clad fuel elements — so low in fact that the fraction of the total estimated dose due to ^{65}Zn is trivial.

R. J. GARNER: The implication of your paper is that the only potential adverse effect considered is an increase in mutation rate. Have you attempted to derive anything for animals equivalent to the ICRP dose limits for man and relating to other possible adverse effects? I ask this particularly because the maximum dose rate to your hypothetical animal of 62 rad/yr is, in fact, about 40% of an exposure rate established by G. Casarett ultimately to produce infertility in male dogs. Perhaps infertility in the male would be a more critical end point to consider than an increase in mutation rate.

S. V. KAYE: With reference to interpretation of estimated doses listed in environmental impact statements, these estimates were discussed in terms of both somatic and genetic effects where appropriate. Since radiation standards developed for man appear to give adequate protection to biota, I do not believe that it will ever be necessary to have separate standards for biota if man is present in the particular environment also.

I have tried to make the point in the paper that the power stations with the highest estimated doses have estimated release rates which are probably overstated by a factor of from 10 to 1000 for the principal dose-contributing radionuclides. When these plants become operational we do not expect any detectable biological effects to result from radiation. The irradiation experiments by Casarett (with which I am familiar) deal with external exposure to high energy X-rays, whereas the environmental situation is one of chronic internal exposure. The exposure situations are not comparable.

F.O. HOFFMAN: What would be the 'environmental capacity' if one were to consider the hypothetical case where man was absent from a given ecosystem?

S.V. KAYE: If man were absent from a given ecosystem — and my primary concern was protection of biota — I would base my estimate of environmental capacity on the minimum radioactive release rate that would elicit a biological response in the most sensitive life stage of the most sensitive organism. Of course you have asked an academic question, so my answer is inevitably academic also!

PRELIMINARY EXPERIENCE GAINED IN MONITORING KRYPTON-85 RECEIVED IN THE NEIGHBOURHOOD OF THE KARLSRUHE REPROCESSING PLANT

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Abstract

PRELIMINARY EXPERIENCE GAINED IN MONITORING KRYPTON-85 RECEIVED IN THE NEIGHBOURHOOD OF THE KARLSRUHE REPROCESSING PLANT.

Several 100 Ci of krypton-85 activity are released from a Purex-type plant for spent nuclear fuel operated in the direct neighbourhood of the Karlsruhe Nuclear Research Center at 60 m height after each dissolving process of an average duration of six hours. To evaluate the burden on the environment, air samples are taken with a compressor at two places in the forecasted area of maximum concentration received. The samples are taken shortly after the emission maximum has been attained. Prior to measurement in a gas-filled counter tube, the krypton is separated in two enrichment steps, utilizing adsorption to cooled activated charcoal. When 1 m^3 of sampled air is measured for over 100 min, a detection limit of 4 pCi/m^3 is obtained. In measurements performed in connection with fuel element dissolving concentrations were found that exceeded the present ^{85}Kr background of about 20 pCi/m^3 by up to a factor of 5000. For such measurements 100 litres of sampled air are sufficient, the processing of which takes several hours.

The krypton concentrations measured are compared with concentration values on the basis of the meteorological conditions prevailing during sampling and the relevant Pasquill diffusion parameters, taking into account the sampling time. An estimate of the uncertainties in the definition of the diffusion parameters required for the calculation allows a better understanding of the partly significant differences between experimentally determined and calculated concentration values. Taking the available results of concentration measurements and of the total emission per dissolving process as a basis, the maximum local β-dose to the environment can be assessed in a more realistic way. This is of particular interest since the β-dose values per dissolving step are below dosemeter detection limits.

1. INTRODUCTION

Several 100 Ci of ^{85}Kr activity are released in each dissolving process of a Purex-type reprocessing plant located in the immediate neighbourhood of the Karlsruhe Nuclear Research Center (600 metres from the northern fence of the centre), designed for a maximum throughput of 40 tons per annum of irradiated uranium with an average burn-up of 20 000 MW·d/t. The activity is emitted discontinuously from a 60-m high stack with a duration of about 6 hours per dissolving process. In 1972 350 to 1550 Ci of ^{85}Kr were discharged per dissolving process. The annual emission totalled about 68 000 Ci. According to the activity release management plan, the Nuclear Research Center is permitted to release up to 350 000 Ci per annum [1].

A special measuring program was initiated to monitor the ^{85}Kr received by the environment of the reprocessing plant. This not only served the purpose of supporting the previous calculations of the radiation burden on the environment by measured values but also of facilitating the evaluation of possible interference with radiation protection measurements at the Nuclear Research Center through a temporary increase in the background from the ^{85}Kr received. A further goal consisted in gathering practical experience in routine ^{85}Kr monitoring in the environment of nuclear facilities.

The measuring techniques and preliminary results of this monitoring program are reported here.

2. SAMPLING

The ^{85}Kr activity concentration in air can only be measured with adequate sensitivity after enrichment and separation of krypton from the air sampled. A compressor is used to sample air in the receiving area. About 400 litres of air are compressed into a 5-litre respiration air cylinder. This operation takes about 5 minutes.

The time of sampling is necessarily determined by the development in time of krypton emission. Figure 1 shows a typical example of the development in time of the krypton concentration C_A (t) in the off-gas from a dissolving process. Short-term peaks preceding the emission maximum proper mark the time of addition of the acid. In 1972 the maximum emission rates were between 0.03 and 0.3 Ci/s. The period of nearly constant maximum emission is 1-2 hours, depending on the operation of the process.

Sampling was performed within one hour of maximum emission. Two consecutive air samples were taken for each dissolving process in the area expected to receive the maximum concentration.

Besides the meteorological data (wind direction, stability category), the access to the premises had to be taken into account when fixing the two sampling points since the immediate vicinity of the reprocessing plant

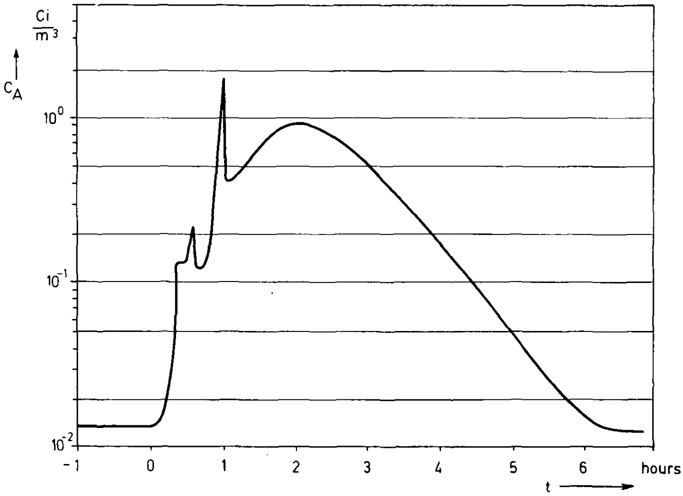

FIG.1. Example of development with time of ^{85}Kr activity concentration C_A(t). Beginning of dissolution t = 0.

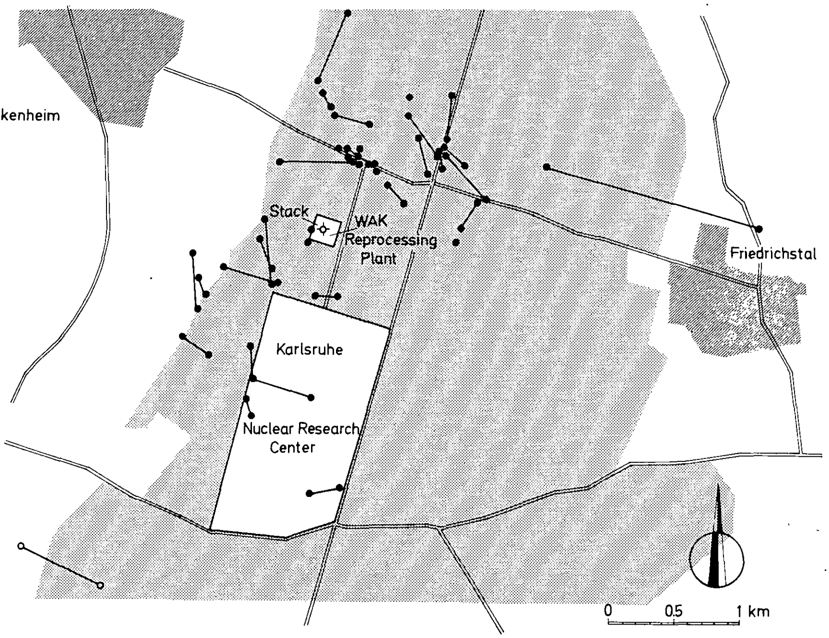

FIG.2. Survey map of sampling points in the vicinity of WAK. Locations belonging to the same monitoring operation have been joined; forest zones appear as shaded areas.

is a wooded area (see Fig. 2). The samples were therefore taken mainly on roads, wooded paths or clearings within a radius of 0.5 to 3 km of the exhaust stack. The second sampling point was set up to take into account the variation in the wind direction, assuming that at least one location would be very close to the actual direction of transport. Practice has shown that this was possible in 50% of the cases with an angular deflection of $< 15°$ from the direction of transport.

3. MEASURING TECHNIQUE

The ^{85}Kr concentration in air is measured in the laboratory. The actual measurement is preceded by the treatment of the samples, which takes several hours. When 1 m^3 of sampled air is used a detection limit of 4 pCi/m^3 can be obtained. However, for measuring much higher ^{85}Kr concentrations in emission monitoring in the environment of the reprocessing plant 100 to 200-litre samples of air are sufficient.

Following drying with a molecular sieve and freezing of CO_2, this air volume is passed through activated charcoal cooled by liquid nitrogen in a preliminary enrichment step (see Fig. 3). Krypton, xenon and radon are almost completely adsorbed by the activated charcoal, while argon, nitrogen and oxygen are only partly adsorbed. In a second enrichment step this gas mixture is expelled by heating the activated charcoal to about 200°C. Repeated flushing with pure nitrogen ensures that no krypton is

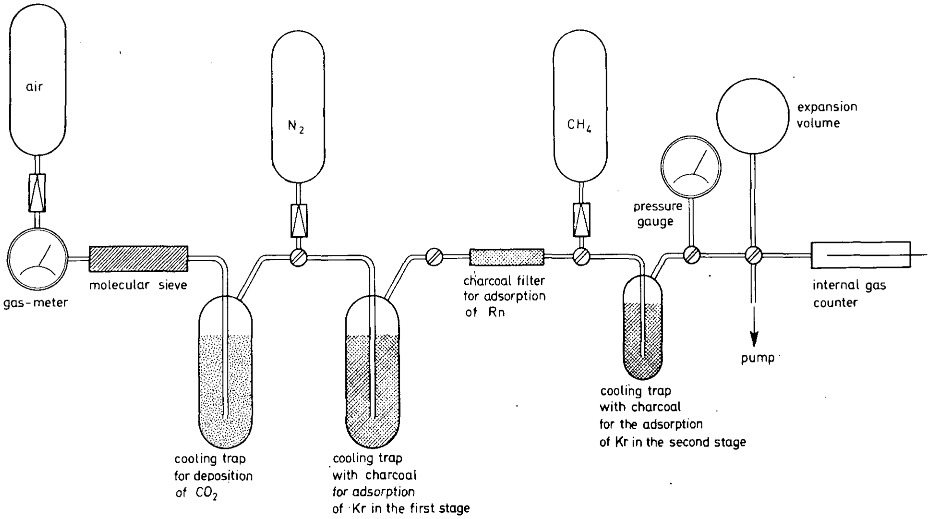

FIG.3. Schematic representation of the apparatus for adsorption and measurement of ^{85}Kr.

left. The expelled gases are first passed through uncooled activated charcoal for radon adsorption. Then they pass through a much smaller amount of activated charcoal cooled by liquid nitrogen. In this process the krypton is again almost completely adsorbed, while the majority of the nitrogen and oxygen still present can be pumped off. To prevent an important fraction of the adsorbed krypton from getting lost, the pumping off of nitrogen and oxygen is discontinued when a pressure of 100 Torr has been reached. By adsorption of the gas now contained in a closed apparatus the cooled activated charcoal causes the pressure to drop below 1 Torr.

In krypton purification advantage is taken of the fact that nitrogen and oxygen are desorbed at relatively low activated charcoal temperatures, while krypton remains adsorbed. The cooling fluid is simply removed and the nitrogen and oxygen so released are transferred to an expansion vessel. If the pressure increases slowly, the connecting line between the expansion vessel and the cooling trap is closed, the expansion vessel evacuated and the gas left in the cooling trap readsorbed by cooling it again. By repeating this process several times the amount of nitrogen and oxygen is reduced to such an extent that these electronegative gases can no longer disturb the measurements, which are made in gas-filled counter tubes using methane as counting gas. Heating the activated charcoal to about 200°C and flushing with methane expels the krypton, which is then passed over to the measuring counter tube with a volume of 500 cm^3.

Tests conducted with activated krypton on the one hand and with true air samples, using two sets of equipment in series, on the other, led to ^{85}Kr yields of 80-90%. The relative error for the ^{85}Kr concentrations measured is ± 25%.

4. RESULTS OF MEASUREMENTS AND COMPARISON WITH CALCULATED VALUES

The Karlsruhe reprocessing plant (WAK) was commissioned in late 1971. Of 85 fuel-element dissolutions performed in 1972 a total of 34 processes were monitored by 67 measurements of the ^{85}Kr received. Roughly 45% of the ^{85}Kr concentrations in air measured lay between 1 and 10 nCi/m^3 and one third of the air samples showed concentrations between 10 and 100 nCi/m^3. The highest value measured was 167 nCi/m^3.

Measurement of air samples taken in the Black Forest (Feldberg and Kaiserstuhl) and in the more distant environment of the Nuclear Research Center concurrently yielded a ^{85}Kr background in air near the ground of about 20 pCi/m^3.

In almost 80% of the cases monitored the ^{85}Kr received exceeded the normal ^{85}Kr background by factors of between 50 and 5000. Although some influence of these emissions on the background count rate of the various radiation protection measurement instruments used within the Nuclear Research Center could be detected, it has no practical importance because of the short duration of these disturbances. The statistics of the wind direction for 1972 show that the frequency of the wind direction relevant to such disturbances is only about 12%.

To complete the measurements the ^{85}Kr concentrations were calculated for the appropriate times and co-ordinates of the locations of sampling. The computing model used is based as usual on a Gaussian distribution of the concentration received. The following expression holds for the concentration distribution C near ground level (compare, e.g., Refs [2, 3])

$$C = \frac{\dot{A}}{\pi \bar{u} \sigma_y \sigma_z} \exp - \left[\frac{H^2}{2 \sigma_z^2} + \frac{y^2}{2 \sigma_y^2} \right] \tag{1}$$

where

$\dot{A}$ = source strength in Ci/s
$\bar{u}$ = average wind velocity in m/s
σ_y, σ_z = Pasquill diffusion parameters in m
H = effective stack height in m
y = position coordinate perpendicular to transport direction

The Pasquill diffusion category applicable to the time of sampling was determined from the measurements performed at the 200-m high meteorological tower of the Nuclear Research Center. For the effective stack height H the purely empirical set-up according to Concawe [4],

$$H = h + 0.047 \frac{Q_H^{0.58}}{u^{0.7}} \tag{2}$$

is used, where Q_H is the heat emission in cal/s, h the height of the stack in m, and u the wind velocity at the height of the stack outlet.

Since for the very short sampling period of 5 minutes reliable mean values cannot be determined from the meteorological measurements made at the 2-km distant measurement tower, the required mean values were taken from longer periods of observation and provided with realistic variations for each individual case. In this way individual variations for the direction of transport and the σ-parameters were defined for each sampling.

According to Concawe, there is an error of ± 50% in the term of Eq. (2), which determines the plume rise. From the uncertainty of the quantities indicated a scattering range was calculated for the ^{85}Kr concentration at the point of sampling. The position of the sampling points and their angular co-ordinates α (see Fig. 2) relative to the direction of transport $\bar{\phi}$ is described by the angular difference

$$\Delta\alpha = |\bar{\phi} - \alpha| \tag{3}$$

The frequency distribution of $\Delta\alpha$ is shown in Fig. 4. Of the 62 sampling points 34 deviate from the average transport direction by not more than 15°. Despite the intention to locate, if possible, the two sampling points for each monitoring operation in the exact direction of transport, the spread of the frequency distribution clearly shows the difficulty of a correct forecast of the transport direction for the time of sampling.

To compare the ^{85}Kr concentrations measured with those calculated with Eq. (1) the relation

$$q = \frac{C_{\text{exp.}}}{C_{\text{theor.}}} \tag{4}$$

was set up with the remarkable error limits being mainly determined by the uncertainty of C_{theor}. For sampling points with angular deviations $\Delta\alpha > 15°$ from the transport direction the calculations mostly yielded much

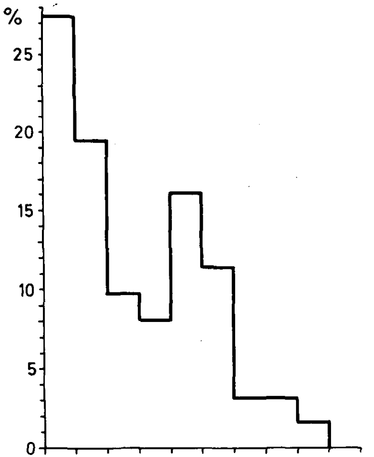

FIG.4. Frequency distribution of angle differences $\Delta\alpha$ between transport directions and α co-ordinates of the sampling points.

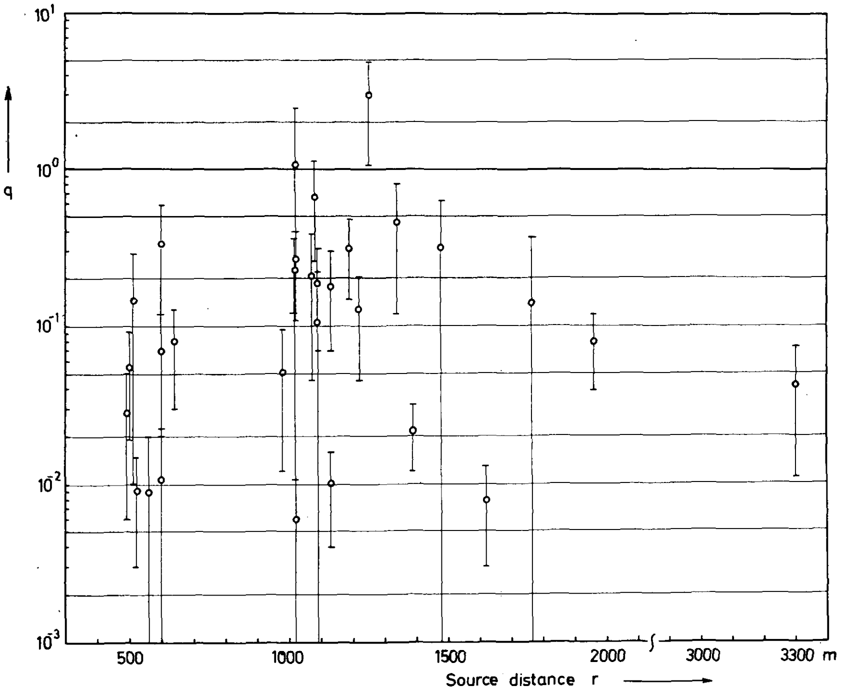

FIG. 5. $q = \frac{C_{exp}}{C_{theor}}$ as a function of the source distance for $\Delta\alpha < 15^{\circ}$.

smaller concentration values than the measurements. These results reveal that the actual distribution of concentration of the ^{85}Kr received is broader than had been assumed theoretically.

For the cases with $\Delta\alpha < 15°$ the reference value q is shown in Fig. 5 as a function of the source distance r of the sampling points. Nearly all the samples were taken under unstable to neutral atmospheric conditions. This is reflected by the distribution of source distances shown in Fig. 5.

The q ratios vary between 0.006 and 3 and all values except for two are well below unity. The unidirectional deviation of the distribution from unity, which fixes the agreement between theory and measurement, is surprising since in spite of only 5 minutes of sampling and as many as 30 measurements a broad but still more symmetrical distribution had been expected.

On the basis of the results from diffusion experiments carried out at the Karlsruhe Nuclear Research Center with tritiated water vapour as tracer [5, 6] and a sampling period of 20 to 30 minutes a greater number of values $q > 1$ was also expected for krypton sampled at points in the diffusion sector $\Delta\alpha < 15°$. As a matter of fact, significant scattering ranges were found in these experiments despite six times the duration of sampling. Although the asymmetry of the distribution represented in Fig. 5 cannot yet be regarded as statistically certain (only two sampling points per monitoring operation) compared to the results of the diffusion experiments (25 sampling points per experiment), they can hardly be considered as being merely accidental values.

Topographical conditions may considerably affect the concentration field. The deviations between theory and measurement might be due to the fact that almost all sampling points were located in a wooded area (paths, clearings, etc.) (see Fig. 2). However, this hypothesis is not supported by experience gathered in the diffusion experiments since on average the concentration values found in wooded areas were no lower than the values obtained in the open field.

A possible cause of the discrepancy between theoretical and experimental values might be a wrong assessment of the plume rise, which has not yet been verified experimentally.

The site-specific experience gained in the field of atmospheric diffusion of radioactive gases cannot yet be reconciled with the available data on ^{85}Kr concentration measurements performed in the vicinity of WAK. This is an illustration of the problems that must be overcome to make a realistic statement on the maximum concentration received based on monitoring results from a few samples. The problem cannot be solved by increasing the monitoring expenditure since in view of time and personnel required for sampling and measurement in routine monitoring only a few samples can be taken.

To explain the still incomprehensible asymmetry of the reference value q shown in Fig. 5 the following steps seem to be appropriate:

(1) Check the plume rise, which is a significant term in the calculation
(2) Extension of the sampling period to make it more independent of short-term variations
(3) Use of different σ-parameter sets for the theoretical calculations in order to be able to consider the topographical conditions more efficiently.

5. ESTIMATE OF THE ^{85}Kr RADIATION BURDEN ON THE ENVIRONMENT

Starting from the available data the maximum radiation burden observed so far in a dissolving process can be estimated and the annual radiation burden on the environment extrapolated accordingly.

5.1. Radiation burden in the maximum concentration received calculated for each individual dissolving process

The calculation of the beta radiation dose D_β for the individual dissolving process is based on the model of semi-finite space filled with a constant activity concentration. If A is the activity released, g_β the submersion dose constant from beta radiation ((rem · m^3)/(Ci · s)) and χ the diffusion factor (s/m^3), the following relation applies

$$D_\beta = g_\beta \chi A \tag{5}$$

For ^{85}Kr, $g_\beta = 0.074$ rem · m^3/Ci · s. Consequently, with A = 1550 Ci and the greatest calculated χ_{max}-value of 1.1×10^{-5} s/m^3, 1.2 mrem is obtained as the local β-dose.

The respective γ-dose D_γ is calculated with the relation [7]

$$D_\gamma = \hat{g}_\gamma \hat{\chi} A \tag{6}$$

where $\hat{g}_\gamma$ is the submersion dose constant from gamma radiation (for ^{85}Kr $\hat{g}_\gamma = 3.4 \times 10^{-7}$ rem $\cdot$ m^2/Ci $\cdot$ s) and $\hat{\chi}$ the modified diffusion factor. In the example considered the wind velocity was 4 m/s. According to Ref. [8], the maximum value of $\hat{\chi}$ is 1.0×10^{-2} s/m^2. Consequently, a γ-dose of about 5 μrem is obtained. Persons present at the place of the maximum concentration received are practically only affected by gamma radiation, while the beta radiation (range in tissue 2.4 mm [9]) has an effect only on uncovered skin so that the β-dose to skin averaged over the skin surface is definitely less than 1/10 of the local β-dose estimated from Eq.(5).

The estimates above start from the assumption that the diffusion conditions and in particular the wind direction are constant in the course of the dissolving process. However, this is not true in practice so that the maximum ^{85}Kr emission is subject to migration and, consequently, a too pessimistic value has been indicated for the maximum diffusion factor. According to the results of measurements, the rarefaction of ^{85}Kr in the atmosphere appears to be better than calculated. However, on account of the low maximum range in air of about 2 m [9] this affects only the beta-radiation burden, while its influence on the gamma burden is small.

5.2. Annual radiation burden on the population in the environment

The exact calculation of the annual radiation burden D_a calls for a calculation at each receiving area and for each dissolving process, taking into account the prevailing weather conditions. However, such a detailed calculation is not necessary here and only an estimate will be made using the relation

$$D_a = R \cdot Z \cdot D_e \tag{7}$$

where D_e is the β and γ-dose, respectively, for the dissolving process with the maximum burden on the environment, Z the number of dissolving processes per annum and R a factor indicating for each dissolving process the probability of a radiation burden occurring at the receiving area.

With respect to the radiation burden outside built-up areas it should be noted that there is little probability of one person being exposed to several burdens at the maximum concentration received in the course of one year. In no case will it be higher than the burden within built-up areas. There are 5 villages within a radius of 5 km of WAK. The most unfavourably located community (Leopoldshafen) is on the leeward side of the WAK exhaust stack for about 20% of the time. On the pessimistic assumption that the inhabitants live in the open air for about 50% of the time, R in Eq.(7) is 0.1. For the other villages in the vicinity of WAK the conditions are much more favourable.

Consequently, with $Z \approx 100$, a value of about 0.05 mrem/yr is obtained for the whole-body gamma dose and 1.2 mrem/yr for the skin beta dose averaged over the skin surface, the latter value being certainly overestimated by a greater factor than the first value because of the better rarefaction of the radioactive effluents.

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DISCUSSION

K.-J. VOGT: May I comment on the fact that your measured concentrations were too low compared with the calculated values. As I understand it, the Kr concentrations were calculated on the basis of diffusion factors specified in my paper quoted as [8] in your references. These values were calculated with the standard deviations recommended by Pasquill.

As I have stated earlier at this Symposium, these σ values are too small for long-term diffusion over rough terrain. Consequently the maxima of the concentrations are predicted as occurring further from the source than is correct. Our diffusion measurements carried out at Jülich, i.e. for a site similar to the Karlsruhe installations in terms of ground roughness, show that the maxima are shifted towards the source by a factor of 3 or 4 compared with the predictions based on Pasquill. This may, at least partly, explain the low figures for the concentration measurements.

M. WINTER: Quite so, but I fear that in your and our diffusion experiments the divergences between the theoretical and experimental values of the Q ratios are much higher in absolute terms.

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EMPIRICAL BENEFITS DERIVED FROM AN ECOSYSTEM APPROACH TO ENVIRONMENTAL MONITORING OF A NUCLEAR FUEL REPROCESSING PLANT

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Abstract

EMPIRICAL BENEFITS DERIVED FROM AN ECOSYSTEM APPROACH TO ENVIRONMENTAL MONITORING OF A NUCLEAR FUEL REPROCESSING PLANT.

The environmental monitoring program for a nuclear fuel reprocessing plant (Allied-Gulf Nuclear Services) located in the South Carolina coastal plain, is based on a man-environment ecosystem concept. Impact measurement and analysis capabilities include radionuclides, noxious chemicals and heat. The principal pathways to man, atmospheric, terrestrial and aquatic, are each subdivided into natural, recreational and domestic components. Basic inputs include meteorology, geology, surface drainage, direction and velocity of groundwater flow, interaction of underlying aquifers and ecological descriptions of the various interrelated ecosystems of the site and region. Process water is discharged through an ecological monitoring, conditioning and cooling pond. Sampling from a 100 square mile grid is based on significant links in the various ecological pathways. Standardized procedures for sample collection, processing and analysis have been developed. All data may be stored, retrieved and analysed through an open computerized program, designed so that additions can be made and data verified and retrieved under any ordered scheme for either printed or graphic display.

Empirical benefits include a means for merging results of basic research on distribution and fate of radionuclides, noxious chemicals and heat in the natural environment with monitoring activities, based on direct and indirect pathways to man. The use of ecologically defined samples provides rates of movement and bio-accumulation that (a) are sensitive enough to verify the adequacy of source control with respect to environmental quality, (b) indicate magnitudes and trends of ecological impacts and therefore serve as an early warning system of potential adverse effects to the environment and man before they become irreversible or expensively reversible, and (c) provide a base for comparison with measurements and analyses of effluents from other sources on a regional and international scale. The program can accommodate additional questions and unexpected impacts. The procedures and ecological models are transferable to other nuclear industry. Thus, through the characteristics of comparability, accommodation, predictability, and transference, the program emphasizes those basic factors and procedures that are necessary for determining the capacity of the environment to receive radionuclides, with emphasis on both natural and human radiation exposures.

INTRODUCTION

There is a rapidly increasing awareness of the close interaction and interdependence of man with his natural environment. His welfare is commensurate with the quality of this planet as a life support system. As a consequence, we are at the threshold of moving environmental monitoring procedures from a superficial to an indepth man-environment ecosystem concept. This is demonstrated by the following considerations [1,2,3,4].

1. Radiological monitoring was founded and in large measure has been perpetuated on the dual premises that unless the effects of radionuclides in the environment affect man directly they are not of major importance and that only those parts of the environment with which man is in reasonably close contact need be examined in order to make adequate evaluations. Although the limitations and inadequacies of this approach have been evident to ecologists for many years, only now is an understanding of these inadequacies beginning to enter into the conceptual framework of state, federal and international regulatory agencies and of industry. The capacity of the environment to accept radioactivity is one problem which has emerged from this recent awareness of the need to maintain environmental quality as a normal aspect of the nuclear age.

2. With the development of nuclear energy the biosphere can be exposed to an increasing burden of radiation. Certain conditions arising from the technical performance of the nuclear industry seriously temper the expectation that nuclear radiation in the natural environment can be restricted to within approximately one percent of background or natural radiation. These are (a) attainment and long-term maintenance of anticipated engineering performance; (b) adequate management of radioactive wastes; (c) prevention of sabotage;and (d) avoidance of catastrophic accidents. The significance of this is two-fold. Man faces an increasing probability of greater indirect as well as direct exposures to radiation, while there exists an increasing potential for injurious effects on the natural environment at the population, community, and ecosystem levels. The life support systems of this planet are the result of millions of years of evolutionary development and it is axiomatic that increased radiation stress, whether on a global, regional or local scale, can lead toward imbalance or disorder.

3. The problem is extremely complicated because ecosystem balance results from the interaction of many factors, including ionizing radiation. Although the current research base is substantial, it is uneven in its depth and coverage, and to a large extent lacks the needed specific orientation to the kinds of radiation stresses from nuclear industry. For example, more needs to be known about the behavior of radioactive Krypton, Plutonium and Iodine in the environment or of their effects upon life within natural systems. Recent disclosures on the ecological effects of non-radioactive pollutants, such as DDT, lead and mercury further illustrate this point.

4. Environmental impacts from nuclear radiation fall into two categories: those which are observable and therefore measureable in terms of physical and biological altera-

tions and those which do not produce such alterations and therefore can be measured only in terms of their accumulation. Whereas the former would be expected from radiation releases by catastrophic events, the latter are the ones with which monitoring is routinely concerned. However, it is important to recognize that even though adverse effects may not be detectable by observation, they may occur. Potential genetic effects must be considered because of the very short reproductive cycles of many plant and animal species. However, existing research indicates that somatic effects on embryonic stages are more likely.

5. Another contemporary aspect of sound ecological monitoring is the need for greater emphasis on measurements of specific nuclides and less reliance on gross alpha, beta, and gamma determinations. Furthermore, laboratory measurements of collected samples must be supported by in situ field measurements by gamma-ray spectroscopy in order to assure valid interpretations.

6. Environmental monitoring procedures now specified by regulatory agencies are based primarily on a determination of the kinds and concentrations of radionuclides in highly generalized and restricted compartments of the environment. Customary procedures include sediment sampling on an indiscriminate base; grab samples of unidentified vegetation and invertebrates; collection of readily available game animals, fish and shellfish; and sampling of the more common agricultural commodities which constitute direct food pathways to man. Thus, the environmental sampling, especially of natural ecosystems, is based on fairly random and ecologically non-identifiable collections, while the common usage of averaging the results of radionuclide analyses conceals the maximum accumulations by sediments and the biota.

Approaches need to be developed which will include (a) the pathways by which these materials are moved through the environment; (b) where they are concentrated in natural systems; (c) how long it may take them to move through these systems to a position in contact with man; (d) and the effects these nuclides may have on populations, communities, and ecosystems. In order to achieve these approaches sampling procedures should be based,for example,on identification of biota by scientific name, physiological state, and trophic position, and by physical analyses of substrates. By these and other such means, the ecological base can be developed which is necessary for the development of early warning systems on a national or global scale, and which would have the capacity to prevent potential problems from developing.

THE MONITORING PROGRAM FOR THE ALLIED-GULF NUCLEAR FUEL REPROCESSING PLANT

Concepts presented in the introduction have been used as the basis for design and development over the past 3 years of the monitoring program for the Allied-Gulf Nuclear Fuel Reprocessing Plant, located in the coastal plain of South Carolina, U.S.A.

The basic criteria and objectives for this program as established by Allied-Gulf Nuclear Services were:

1. That it be planned and managed by a University group who could act independently of the restraints which are sometimes associated with industry consultation.

2. That the program extend for many years, with at least the first three being in the pre-operational stage.

3. That it provide guidelines for clarification of the adequacy of effluent source control with respect to maintenance of environmental quality.

4. That it provide data to estimate human population exposures to effluents from actual operation.

5. That it maintain surveilance and indicate magnitudes and trends of ecological impacts of the industry and from this provide an early warning system of potential adverse effects.

6. That it evaluate the contribution of other regional sources to background levels of various effluents within the geographic area under the influence of this industry's operation.

7. That it incorporate a close working relationship with state and federal agencies and other area activities.

8. That it provide environmental data for public information.

As a means of achieving these objectives, a systems approach was developed which encompasses the region surrounding the site. This region is treated as a man-environment ecosystem with all components closely inter-linked with each other.

This has required the close interaction of an interdisciplinary team which collectively represents competence in plant and animal radiation ecology, hydrology, meteorology, radionuclide measurement and evaluation, and ecosystem modeling and analysis. Thus, the program differs significantly from routine monitoring activities in two respects, one being the use of an interdisciplinary team and the other the development of an ecological model around which the program could develop.

Since the operational design criteria for this plant has utilized and implemented the state of the art such that normal and accidental pollutants will be removed or held at a minimum, observable cause and effect relationships from these are not expected. The environmental impact program therefore has been established primarily in terms of physical and biological concentrations. However, the program also has incorporated within it the capability of dealing with observable alterations and the means for rapidly establishing the necessary procedures for their measurement and evaluation in the event of unforeseen stresses.

The concept of monitoring and evaluation used in this program should not be considered a research effort. Rather it draws upon the extensive body of existing research according to the specific situations to which it may be applied. However, as would be expected, independent research activities are being derived from it[11]. The essential aspect of this program is that the concept of assessment on a sound ecological basis be recognized and utilized in response to the requirements and guidelines of regulatory agencies. The fact that it exceeds the present guidelines in its depth and scope is a recognition of the directions in which monitoring activities are rapidly moving.

The program was initiated in the summer of 1969 prior to establishment of the Environmental Protection Agency and of the guide lines developed by EPA and AEC for compliance with its directives. The ensuing three years have been devoted initially to development of an ecological model which provides a means for satisfying the program objectives and secondly a pre-operational inventory designed to test the validity of the model and to meet state and federal regulations [4-10].

The plant is expected to be operational in 1975. This next year is to be used for inventory evaluation, additional surveillance as needed and completion of the program as projected for the operational stage.

Ecosystem Components and Pathways

Ecosystem structure is based on the spatial arrangement of plant and animal species, the substrate and its quality, and quantitative knowledge of the chemical elements contained in the various organisms. Ecosystem functions concern the pathways and turnover rates of such processes as energy flow and chemical element cycling.

This type of approach ultimately leads to quantification of systems in terms of their functioning. Such data in turn provides a basis for the development of descriptive and/or predictive models of the system. There are two approaches to the sampling of these systems which deal with the problem of "early warning". One is uniform sampling of the entire area affected while the other is the development of qualitative models which will allow selective sampling to accomplish this purpose. For this program, the latter approach is used, since it is more likely to define environmental concentrations of released materials by providing an efficient and fairly rapid entry to an otherwise almost unmanageable mass of data. By this means, it is relatively easy to assess the total impact expected from known releases of given products since the species most likely to receive and accumulate the materials have been identified.

The Barnwell Nuclear Fuel Plant (BNFP) is located on the eastern edge of the Aiken Plateau of the Atlantic Coastal

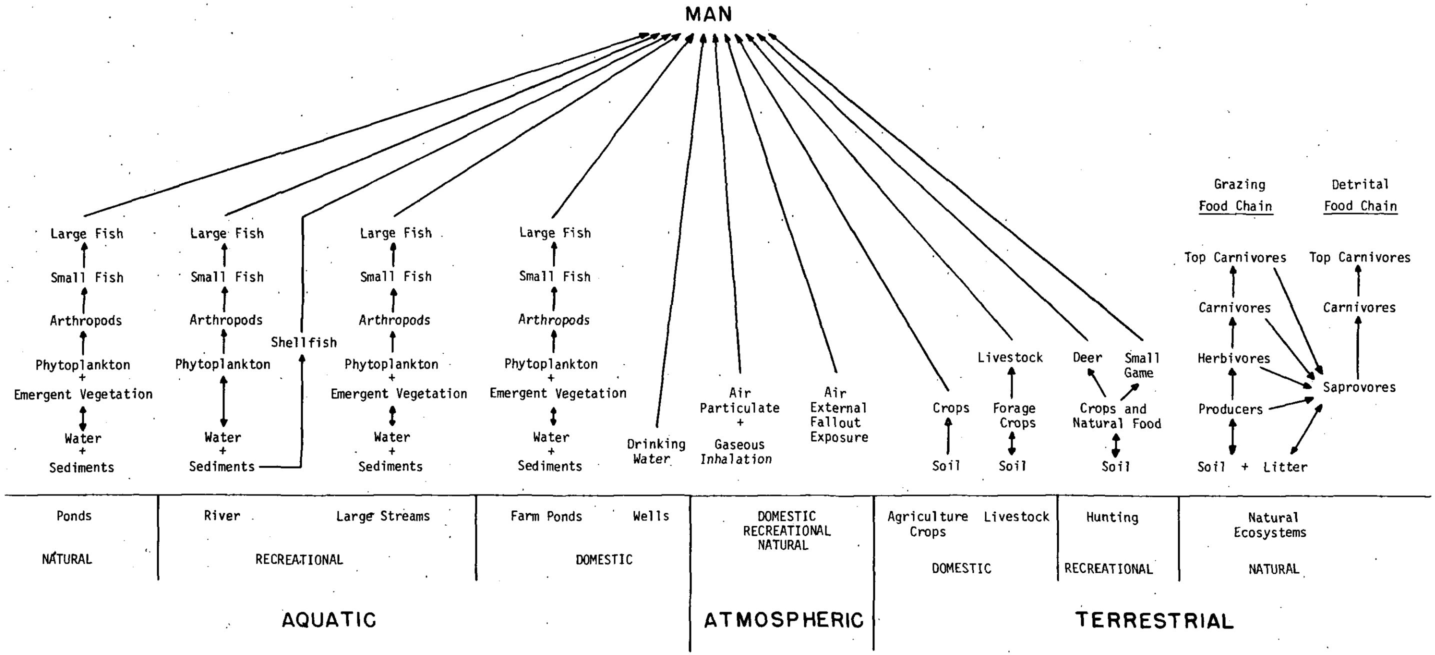

FIG. 1. Expected pathways of BNFP effluents.

Plain physiographic province and is adjacent to the eastern boundary of the Atomic Energy Commission Savannah River Plant of which it was formerly a part. The site comprises some 1700 acres, of which approximately 165 are affected by construction activities. The land is generally level with a gently rolling surface. Differences in elevation on the site occur mostly in a north-south direction with variations in elevation from 270' to about 210' MSL.

The primary ecological subsystems are terrestrial, aquatic and atmospheric. For each of these, there are three kinds of generalized food chains by which materials can be moved, accumulated and potentially affect man and nature These are the domestic, recreational, and natural subsystems. Domestic food chains represent a direct and sustained trophic coupling between man and his environment. Recreational food chains indicate a direct but infrequent coupling, while man and the food chains of natural systems are only indirectly coupled. This basic system is shown in Figure 1.

Terrestrial

Terrestrial systems and pathways on-site and in the region have been studied [4,5,9]. Most of the defined natural systems of the Upper Coastal Plains Province are represented. The region is a mosaic of systems with abrupt changes within relatively small geographical areas. The proximity of the water table to the surface plays a significant role in determing community distribution over the landscape. Relatively small changes in topography can produce significant changes in the species present on a given site. For example, in the flood plain of the drainage system, changes of one to two feet in elevation can result in completely different forest systems.

The geologically young soils in the area are predominately sandy. In low-lying areas sufficient detrital material has accumulated and provides a chemical nutrient base for growth and development. However, in the upper more xeric areas the ecosystems are often nutrient limited and do not show rapid development.

Ten distinct terrestrial ecosystems have been identified and described according to basic structure and function. These are pine-scrub oak-sand hill forests, loblolly and mixed pine forests, pine-oak forests, Carolina bays, upland hardwood forests, mixed mesic slope forests, domestic crop lands, bottom lands and swamp forests, stream bank forests, and old fields.

In addition to their ecological identification, these systems have been mapped by aerial photography and surface inspection, and then related to existing topography, meteorology, and sub-surface hydrology. From this base, the land use of the area has been quantified at varying distances from the plant site. For crop land this includes human use by row crops, field crops, and orchards, as well as domestic livestock use of pasture, rotation-hay, hayland and grazed forests and also by idle farm land. Population density and dispersion also has been determined for urban situations.

Species lists of the natural, domestic and recreational flora and fauna have been developed, along with information on distribution and abundance of principle species.

Aquatic

Extensive hydrological investigations of both the surface and sub-surface activities have been made [5,6,7,8,10,11,12]. The 1700 acre plant site is located in the northeasterly portion of the Lower Three Runs Creek drainage system, a tributary of the Savannah River which drains an area of some 180 square miles. There are no surface streams on the site, and natural surface run-off features are essentially absent, and except for unusually heavy precipitation, rainfall is held in local surface depressions, evaporates, transpires, and in part infiltrates into the groundwater table. Aquatic systems adjacent to the site or in the region include rivers, black water creeks, farm ponds, impoundments,Carolina Bays and marshes.

The groundwater hydrosphere has been the subject of several geological and hydrological studies in past years. The site is underlain by a series of unconsolidated and partly consolidated sedimentary formations, each of which have distinctive characteristics and constitute well-defined aquifers. These geologic formations from the surface downward are the Hawthorn, Barnwell, McBean, Allenton, and Tuscaloosa. The latter extends from about 370 to more than 1000 feet below ground, and is the principal aquifer for the project water supply systems.

Although these aquifers are separated by well-defined aquicludes, the possibility of hydraulic connections has been the subject of extensive studies. The principle means used for this was a 60-day pumpage test from the Tuscaloosa aquifer, with concomitant observations on behavior of this and the overlying aquifers. From these studies, it is concluded that the pressure gradient created by the cone of depression caused by the pumpage test will not result in any drawdown of the upper water tables, and hence use of this water will not result in flow from the upper into the lower water-bearing strata. The important corollary is that in the absence of hydraulic connections between the upper water bearing strata and the Tuscaloosa, this aquifer is safe from inflow of radionuclide contaminated water that might be introduced accidently into the upper water table.

The upper water table has been monitored since mid-1971. Records show that the slope of the water table definitely persists in a southerly direction, and therefore all drainage from the project site enters into the LTRC drainage system. This surface water table is of course the potential recipient of any accidental leakage of radioactive liquid wastes. Transport phenomena involving radionuclides in the groundwater environment are influenced by the speed and direction of the groundwater movement, by the ability of the local soils to attract and hold radionuclides, and by the rate of decay of the radionuclides.

The transport velocity of this groundwater was conservatively calculated to be 0.02 feet per day. Since the shortest seepage path from the site to the creek is about 6,000 feet, the travel time for water is predicted to be some 820 years. Travel time for all pertinent radionuclides and decay products have been estimated. With the exception of long lived isotopes of certain heavy metals, the residence time in the groundwater environment is sufficiently long to reduce radioactivities to safe and acceptable levels. The heavy-metal isotopes which form complex chemical compounds tend to precipitate and are physically restrained from moving with the groundwater. Based on many studies, it is recognized that these precipitates progress little beyond points of injection, and can therefore be physically removed.

These kinds of studies are thus essential to the development of a monitoring program for radioactive waste storage.

All plant management water except sewage is discharged through an ecologically designed cooling pond (BEACON Pond) to Lower Three Runs Creek. Sewage is treated and then dispersed over a three acre plot by a spray irrigation system. Lower Three Runs Creek, which is within the Savannah River Plant boundaries, flows for about 27 miles before entering the Savannah River. It has a gentle slope with narrow to broad channels intermittently bordered by large marshy areas. The upper portion of LTRC is covered by a large impoundment, Par Pond, and the plant effluents are to be discharged just below the dam.

Hydrological studies of LTRC include physical properties, average flows, critical minimum flows, maximum flows, and flood stages. Water quality studies have included temperature and chemical contents. Radiological studies have included extensive inventories along the stream channel and across its flood plain. Biological studies have included a detailed analysis of the structure and function of the stream, stream bank and flood plain ecosystems.

The cooling pond through which management water flows prior to emptying into Lower Three Runs Creek is especially designed as an ecological monitoring and conditioning pond. This pond, known as BEACON Pond, is approximately 17 acres with an average depth of 9 1/2 feet and a volume of about 60,500,000 gallons. It serves the operational needs of the plant by providing a supply of water for emergency recycling to the plant, fire protection and cooling of discharge water. Environmental quality is maintained by providing for ecological conditioning and equilibration and by continuous monitoring across the discharge weir. These measurements include ph, temperature, turbidity, organic carbon, dissolved oxygen and conductivity. The pond has been biologically stocked from adjacent ponds and streams such that a balanced ecological system is being developed. Retention time for a flow-through rate of 2300 gpm is approximately 7 days.

A special study has been made to assess the effects of two modes of power failure on water temperature in Beacon Pond. The first assumes that only commercial power fails and that the Tuscaloosa wells can be operated by emergency power supplies. The second and more severe emergency assumes that both the commercial power supplies and the emergency supplies are unavailable for operation of the cooling towers, which are located between the Pond and the Plant and the supply wells. The studies have shown that even under these conditions when the temperature rise in the water entering the ponds is 33°F, it will be cooled to about 2° above ambient under the most adverse environmental conditions. Thus water may be recycled over an extended time period in case of an emergency until the situation can be corrected.

The water in Lower Three Runs Creek beyond Par Pond dam is of excellent quality. Detailed studies have shown that the expected effluent releases from Beacon Pond will be comparable to those of the creek, and no adverse effects are anticipated with respect to temperature, dissolved oxygen, nutrients, conductivity, and other parameters.

Atmospheric

The site, approximately 85 miles inland from the Atlantic ocean, experiences a mesothermal climate according to the Koppen-Geiger classification. Historical meteorology data were obtained from a weather bureau station 35 miles northwest of the site and from a 1200 foot television tower about 25 miles northwest. These data are applicable to the site, since there are no hills, valleys or large bodies of water in the vicinity which might invalidate the data. A 100 meter meteorological tower has been installed at the site with the appropriate instrumentation.

Although the site is far enough inland to avoid hurricane force winds, such storms on occasion move over the area and are accompanied by heavy rainfall. The site elevation along with the topography precludes any possibility of flooding. With respect to tornados, which have a recurrence interval of about 5 years, non-critical structures are designed to withstand the forces from 100mph winds, and structures for process areas and essential utility services are designed to withstand a differential of three PSI for three seconds, and the forces of missles and 300mph winds associated with a direct hit by a tornado.

Gaseous releases will be from a 100 meter stack. Again there are no hills, valleys or large bodies of water which would tend to channel airflow or create mechanical turbulence. Thus the site permits the use of the Gaussian distribution equation to relate concentrations or dosages at any point in space to the rate of emission of airborne radionuclides and noxious gases. Detailed studies on diffusion of stack effluents on the site, at the site boundary, and to the region have been made by AGNS personnel [4].

The Sampling Program

The sampling program is a key issue, since the hard data obtained is the basis for satisfying the operational objectives.

Thus the method for collection and analysis of samples constitute the action part of the program. The initial planning for sampling was concerned with the kinds and sources of technical support; the kinds of data to be obtained along with sampling procedures and analyses relating each component of the whole system; and special considerations for critical effluents and for the more sensitive sub-systems; and the sequential steps requisite to the inventory, pre-operational and operational stages of the plant [4].

Methods and Procedures

The sampling station network now consists of 83 stations, identified by colloquial names and map coordinates. These are arranged on a 100 square mile grid, and within this grid on concentric circles extending from the plant at 5-mile intervals. Forty-one of these stations consist of wells, most of which are in the upper unconfined groundwater table while others are located in the lower aquifers. Of the eight natural terrestrial stations, six are on the plant site and two off-site. Seven drinking water stations are established, most of them being in nearby communities. Of the five atmospheric stations, one is on-site and four are in nearby communities. Eleven aquatic stations are established along the discharge-water pathways from the plant through Beacon Pond and along Lower Three Runs Creek. Eighteen domestic stations are located in various agricultural systems within the region.

A manual has been developed which reduces to writing all of the procedures for sample collection, processing and analyses.

Data cards, designed for computer programming, storage and analysis, are attached to the abiotic and biotic samples. All pertinent information is entered on this card, including the time, date and specific location of each sample as well as the taxonomic identification, tissue subsample, and type of analysis performed for each biotic sample.

Radiological Survey

The transference of radionuclides through ecosystems, the resulting biological accumulation and effects, and the eventual dose delivered to man depends on the radionuclides released and their chemical form. It is therefore essential that the radiological survey program be based on a complete ecosystem approach incorporating, whenever possible, sensitive measurements of the concentrations of individual nuclides in the most critical ecosystem compartments, beginning at the release points. The major emphasis should be placed on the most critical nuclides, and the most sensitive indicators. The requirement that all releases be as low as practicable has brought about in most cases the expectation that the contributed concentrations be low as compared to natural radioactivity, cosmic radiation and fallout. The measurement of these low concentrations has put greater demands on the sampling and analysis procedures and detection sensitivities. For many

TABLE I. ESTIMATED AVERAGE RADIOACTIVE EFFLUENTS (curies/sec) FROM MAIN STACK AND RESULTING AIR CONCENTRATIONS AT SITE BOUNDARY

Nuclide	Main Stack[1] (curies/sec)	Concentration at Site Boundary (Ci/m^3)[2]	Nuclide	Main Stack[1] (curies/sec)	Concentration at Site Boundary (Ci/m^3)[2]
*H-3	1.8×10^{-2}	3.4×10^{-10}	Ce-144	8.4×10^{-8}	1.6×10^{-15}
*Kr-85	4.3×10^{-1}	8.0×10^{-9}	Pm-147	1.5×10^{-8}	2.8×10^{-16}
Sr-89	7.6×10^{-9}	1.4×10^{-16}	U-234	7.0×10^{-14}	1.3×10^{-21}
*Sr-90	8.8×10^{-9}	1.6×10^{-16}	U-235	7.0×10^{-16}	1.3×10^{-23}
Y-90	8.8×10^{-9}	1.6×10^{-16}	U-238	2.0×10^{-14}	3.7×10^{-22}
Y-91	1.6×10^{-8}	3.0×10^{-16}	*Pu-238	1.3×10^{-10}	2.4×10^{-18}
Zr-95	2.8×10^{-8}	5.2×10^{-16}	*Pu-239	1.2×10^{-11}	2.2×10^{-19}
Nb-95	5.3×10^{-8}	9.9×10^{-16}	*Pu-240	2.1×10^{-11}	3.9×10^{-19}
Ru-103	8.4×10^{-9}	1.5×10^{-16}	*Pu-241	5.5×10^{-9}	1.0×10^{-16}
Ru-106	5.0×10^{-8}	9.3×10^{-16}	Pu-242	1.1×10^{-13}	2.0×10^{-21}
*I-129	1.5×10^{-9}	2.8×10^{-17}	*Am-241	3.2×10^{-11}	5.9×10^{-19}
*I-131	1.2×10^{-8}	2.2×10^{-16}	Am-242	6.3×10^{-13}	1.2×10^{-20}
*Cs-134	1.9×10^{-8}	3.5×10^{-16}	*Cm-242	3.7×10^{-9}	6.9×10^{-17}
*Cs-137	1.2×10^{-8}	2.2×10^{-16}	Cm-243	2.5×10^{-12}	4.6×10^{-20}
Ce-141	5.5×10^{-9}	1.0×10^{-16}	*Cm-244	3.9×10^{-10}	7.2×10^{-18}

*Quantitatively and Biologically Significant.
Short lived decay products are also present in equilibrium quantities.

[1] Activity Basis: Specific Power - 40 kw/kg
Cooling Time - 180 days
Fuel Exposure - 32,000 MWD/MTU

Release Basis: Production Rate - 1,500 MTU/Yr.
100% Kr-85 & H-3 Released
Iodine Decontamination Factor = 1,000
DOG/VOG Decontamination Factor = 10 and Filter Efficiency = 99.8%
HAW/Vaporizer Decontamination Factor = 10^{12}
GPF/Vaporizer Decontamination Factor = 10^{6}
Pu Product Recovery = 99%
Pu Loss to Waste = 1%

[2] Average X/Q is $1.86\times10^{-8} sec/m^3$ @ 2350 meters or beyond if value is greater. This value is based on historical meteorological data. A one year meteorological survey is now being performed.

radionuclides the predicted concentrations are below the current practical limits of detection. Also, for many kinds of samples the concentrations depend on very detailed characteristics of the sample and the sampling program must be carefully guided by those ecologists capable of detailed sample ecological characterization. For example, the concentrations of nuclides in sediments varies over a wide range depending on the finer properties of the sediment. Thus, it is not appropriate to merely quote concentrations in sediments. Another example is the variations of ^{137}Cs concentrations in trees and parts of flood plains. [13]

The radiological survey program associated with the AGNS fuel reprocessing plant is sensitive to the problems just described.

The most critical measurements in any monitoring program are those made at the points of effluent releases and in the air in the immediate vicinity, especially at the site boundary. Detailed and in many cases, isotopic measurements are therefore planned in the main stack and at the site boundary. During the past year environmental measurements have been made to establish an inventory and base line of concentrations in many sub-ecosystems. These data are being used to establish necessary and meaningful measurements during plant operation. Since it would be impossible to describe these inventory measurements in their entirely in this short paper, only a few examples will be described.

Ground Air Concentrations

Gamma Emitters

Ideally a ground air concentration monitoring program would measure the continuous time varying concentrations of each radionuclide in its unique chemical forms. These measurements can then be compared with calculated predicted releases (Table I) [14], and expected doses to man could then be calculated and compared with dose measurements made with TLD's or continuously operated ionization chambers. There is no doubt that the trend in atmospheric monitoring is toward continuous dose measurements, and there are some indications that eventually continuous concentration measurements, made by gamma-ray spectrometry, will be highly desirable or required.

The main constituents that make up the ground air concentrations of radionuclides are those resulting from nuclear weapons testing and from natural processes. The nuclear weapon radionuclides consist of fission produced and neutron induced activity from weapon component materials. The natural radionuclides include the decay of the gaseous (radon) members of the three naturally occurring series, the natural radionuclides in the airborne dust, and the cosmic-ray-produced radionuclides which result from spallation reactions in the atmosphere.

During this year's inventory phase of the monitoring program the following measurements were made of the radionuclides found in the atmosphere: gross alpha and beta activity, gamma-ray spectrometric survey using regular H.V. filters

TABLE II. GAMMA-RAY EMITTERS IN H.V. FILTERS (July 1971 - June 1972)

Isotope	Limit of Detection (pCi)	Composite No.1[b] x10^{-3} pCi/m^{3}	Limit of Detection x10^{-3} pCi/m^{3}	Composite No.2[c] x10^{-3} pCi/m^{3}	Limit of Detection x10^{-3}pCi/m^{3}	Composite No.3[d] x10^{-3} pCi/m^{3}	Limit of Detection x10^{-3}pCi/m^{3}	Composite No.4[e] x10^{-3} pCi/m^{3}	Limit of Detection x10^{-3}pCi/m^{3}
^{7}Be	6.45	124.3±9.39	1.00	152.3±11.2	0.56	201.3±14.6	0.42	260.3±18.9	0.61
^{22}Na	0.86	ND	0.13	ND	0.07	ND	0.06	ND	0.08
^{40}K	15.3	ND	2.37	ND	1.32	ND	1.01	ND	1.45
^{54}Mn	0.73	0.24±0.07	0.11	ND	0.06	0.21±0.05	0.05	0.22±.06	0.07
^{57}Co	0.46	0.29±0.03	0.07	ND	0.04	ND	0.03	ND	0.04
^{60}Co	0.79	0.18±0.05	0.12	ND	0.07	ND	0.05	ND	0.07
^{95}Nb[f]	0.93		0.14	3.64±0.29	0.08	4.59±0.34	0.06	40.0±2.92	0.09
^{95}Zr	1.25	6.22±0.63	0.19	10.7±0.86	0.11	45.9±3.37	0.08	51.9±3.79	0.12
^{103}Ru	0.72	5.92±0.59	0.11	16.4±1.23	0.06	15.8±1.16	0.05	32.9±2.40	0.07
^{106}Rh	5.83	11.4±0.88	0.90	6.24±0.74	0.50	7.32±0.76	0.38	10.9±1.13	0.55
^{125}Sb	1.95	1.96±0.35	0.30	0.84±0.19	0.17	1.40±0.22	0.13	1.20±0.20	0.18
^{134}Cs	0.68	0.30±0.08	0.11	ND	0.06	ND	0.04	ND	0.06
^{137}Cs	0.81	3.20±0.24	0.13	2.3±0.20	0.07	3.85±0.30	0.05	5.42±0.42	0.08
^{140}Ba	1.61	ND	0.25	18.4±2.85	0.14	28.9±2.60	0.11	11.5±1.12	0.15
^{141}Ce	0.93	4.80±0.62	0.14	12.3±0.91	0.08	47.8±3.46	0.06	56.5±4.08	0.09
^{144}Ce	3.84	38.9±2.83	0.58	22.9±1.71	0.33	40.1±2.87	0.25	58.6±4.27	0.36
^{155}Eu	2.07	0.80±0.08	0.21	0.36±0.07	0.18	4.32±1.38	0.14	1.03±0.16	0.20
^{226}Ra	13.3	ND	2.06	ND	1.15	ND	0.88	ND	1.26
^{232}Th	1.03	ND	0.16	ND	0.09	ND	0.07	ND	0.10

[a] Limit calculated for a 1000 minute counting time.

[b] Volume = 11,000 m^{3}. BNFP and Barnwell Air/Rain Stations.

[c] Volume = 7,000 m^{3}. BNFP and Barnwell Air/Rain Stations.

[d] Volume = 7,300 m^{3}. BNFP and Barnwell Air/Rain Stations.

[e] Volume = 4,500 m^{3}. All Air/Rain Stations.

[f] Activities of ^{95}Nb calculated from theory.

TABLE II. (cont'd) GAMMA-RAY EMITTER IN H.V. FILTERS (May 1972 - December 1972) Activities in × 10^{-3} pCi/m^3

	Composite No.5[g]		Composite No.7[h]		Composite No.8[i]		Composite No.9[j]		Composite No.6[k]		Composite No.10[ℓ]	
Isotope	Activity	L/D[n]	Activity	L/D[n]	Activity	L/D[n]	Activity	L/D[n]	Activity	L/D[n]	Activity	L/D[n]
^{7}Be	161.58±11.74	0.48	127.1±10.7	0.38	108.9±8.2	0.37	132.1±10.0	0.52	75.1±5.8	0.80	86.34 7.59	1.90
^{22}Na	ND	0.06	ND	0.04	ND	0.04	ND	0.05	ND	0.08	ND	0.18
^{40}K	ND	1.74	ND	1.25	1.58±1.26	1.22	3.31±1.66	1.69	3.85±2.17	2.60	12.34 6.09	6.20
^{54}Mn	0.13±0.04	0.05	ND	0.03	ND	0.03	ND	0.04	ND	0.06	ND	0.15
^{57}Co	ND	0.03	ND	0.03	ND	0.02	ND	0.03	ND	0.05	ND	0.13
^{60}Co	ND	0.06	ND	0.05	ND	0.05	ND	0.07	ND	0.10	ND	0.21
^{95}Zr	30.9±2.25	0.09	14.3±1.32	0.06	2.39±0.32	0.06	1.11±0.21	0.08	0.20±0.14	0.12	0.36 0.16	0.28
^{95}Nb	28.7±2.09[m]	0.07	18.6±1.72[m]	0.03	4.04±0.54[m]	0.03	2.00±0.38[m]	0.04	0.37±0.26[m]	0.06	0.55 0.17	0.15
^{103}Ru	35.7±2.6	0.05	18.0±1.9	0.02	2.31±0.3	0.02	1.3±0.2	0.03	0.25±0.08	0.05	ND	0.12
^{106}Rh	7.75±4.3	0.44	6.34±0.83	0.38	2.70±0.47	0.37	1.3±0.4	0.51	2.2±0.6	0.79	ND	1.88
^{125}Sb	1.14±0.18	0.15	0.58±0.12	0.09	ND	0.09	0.39±0.09	0.12	ND	0.18	ND	0.44
^{134}Cs	0.39±0.07	0.05	ND	0.04	ND	0.04	ND	0.05	ND	0.08	ND	0.18
^{137}Cs	6.43±0.48	0.06	2.04±0.18	0.04	0.83±0.09	0.04	0.90±0.11	0.50	0.53±0.12	0.09	0.52 0.24	0.19
^{140}Ba	27.6±0.5	0.12	ND	0.07	ND	0.07	ND	0.10	ND	0.14	ND	0.36
^{141}Ce	37.5±2.7	0.07	16.8±2.1	0.04	1.57±0.43	0.04	ND	0.05	ND	0.08	ND	0.19
^{144}Ce	34.4±2.5	0.29	17.6±1.34	0.19	6.1±0.54	0.19	4.9±0.5	0.26	2.2±0.4	0.40	ND	0.95
^{155}Eu	ND	0.16	ND	0.11	ND	0.11	ND	0.15	ND	0.23	ND	0.55
^{226}Ra	ND	0.99	ND	0.70	0.93±0.88	0.68	ND	0.95	ND	1.46	ND	3.48
^{232}Th	ND	0.08	ND	0.05	ND	0.05	ND	0.07	0.30±0.14	0.11	ND	0.26

[g] Collection period 16 May 1972 - 2 June 1972. Volume = 5688 m^3. All Air/Rain Stations.

[h] Collection period 6 June 1972 - 24 July 1972. Volume = 9434 m^3. All Air/Rain Stations.

[i] Collection period 24 August 1972 - 25 September 1972. Volume = 9188 m^3. All Air/Rain Stations.

[j] Collection period 25 September 1972 - 23 October 1972. Volume = 6978 m^3. All Air/Rain Stations.

[k] Collection period 1 November 1972 - 22 November 1972. Volume = 5661 m^3. All but BNFP Air/Rain Station.

[ℓ] Collection period 30 November 1972 - 28 December 1972. Volume = 1934 m^3. BNFP Air/Rain Station.

[m] Activities of ^{95}Nb calculated from theory.

[n] Limit of detection.

and a high resolution Ge(Li)-Na(I) anticoincidence-coincidence spectrometer, ^{131}I and stable ^{127}I measurements using charcoal filters, and tritium in the air in the form of HTO, using silica gel and gamma-ray spectrometric measurement of rain water deposition. The gross alpha and beta measurements serve merely as a screening procedure. Any unusual variations must be accompanied by spectrometric analysis. Composites of air filters were used for spectrometric measurements. A summary of the spectrometric data taken at five air stations situated around the plant site is presented in Table II. These levels of activities agree well with predictions made from the natural background and fallout seasonal variations and with measurements made at other nuclear installations in the U.S.[15,16]. As previously stated, the concentrations in most cases are very nearly equal to predicted concentrations from the reprocessing plants main stack (compare Tables I, and II).

Iodine

The concentrations of ^{131}I were measured by charcoal filters and either Ge(Li) or NaI(Tl) detectors. [17]. The concentrations vary from a maximum of 3.15×10^{-3}pCi/m^3 to a minimum below the limit of detectability of 1.12×10^{-3}pCi/m^3. The Savannah River Plant [15] calculated 1971 concentrations of ^{131}I in air at the plant perimeter, based on releases, are a maximum of 7×10^{-3}pCi/m^3 and an average for 1971 of 5×10^{-3}pCi/m^3. The maximum permissible concentration is 100 pCi/m^3.

Since the man-rem calculations for ^{131}I and ^{129}I can be performed by the specific activity technique, it is important to know the concentrations of stable ^{127}I at the plant site and vicinity. [18]. The average concentrations of ^{127}I measured by neutron activation techniques at Atlanta, Georgia, the reprocessing plant site and Barnwell, S.C., were 3×10^{-9}gram/m^3, 4.2×10^{-9}gram/m^3 and 55.5×10^{-9}gram/m^3, respectively.

These concentrations of stable iodine in air are to be compared with reported concentrations for other areas of the country which vary widely. Data from studies conducted in Hawaii, Alaska, Cambridge, Massachusetts and around Lake Michigan gave fairly consistent iodine concentration values [19-21] of 10^{-8}-10^{-9} grams/m^3. Values of 1×10^{-7}grams/m^3 have been reported in the U.K. [22], but some values there near the sea as high as $\sim 10^{-6}$grams/m^3 have also been reported.

Another critical nuclide is ^{129}I. A neutron activation technique is now being developed for air filters to analyze for this nuclide.

The entire problem of assessing the environmental impact of the iodine isotopes ^{129}I and ^{131}I is continuously being assessed. [17,22,23]. Preliminary studies are being performed to assess the impact and make reliable and meaningful measurements of iodine in all its possible chemical forms.

Tritium in Air

Preliminary results of measurements of tritium in air in the form of HTO are presented in Table III. Measurements made during 1971 by SRP are presented in Table IV.

TABLE III. TRITIUM (HTO) IN AIR AROUND BNFP

Location	Collection Period	Absolute Average Humidity bm/m^3	Tritium (HTO) pCi/m^3	Concentration TU^a
BNFP	1st Quarter 1972	11.9	214	10,700
Barnwell,S.C.	1st Quarter 1972	11.9	71	3,470
BNFP	2nd Quarter 1972	6.25	146	3,830
Barnwell,S.C.	2nd Quarter 1972	6.25	109	2,860
Denmark,S.C.	2nd Quarter 1972	11.0	25	722
Hilda,S.C.	2nd Quarter 1972	11.0	34	960
Norway,S.C.	2nd Quarter 1972	11.0	ND^b	ND

a) Tritium Units, 1 TU $=(T/H)\times10^{18}$ = 3.2pCi/kg water or 6.68×10^4 tritium atoms/gm H_2O.

b) Non-detectable below the level of sensitivity of 5pCi/m^3.

TABLE IV. SAVANNAH RIVER 1971 HTO MEASUREMENTS

Location	Values for 1971 (pCi/m^3)^a		
	Max.	Min.	Avg.
Plant Perimeter	504^b	36	162
Aiken Airport	210	10	40
Barnwell, S. C.	190	10	60
Williston, S.C.	326	10	60

[a] Limit of sensitivity is 10pCi/m^3

[b] This is 0.25% of the Maximum Permissible Concentration

The data compare favorably with those taken by the Savannah River Plant in 1971, [15], but are above the normal background levels found in the southeastern part of the states [24]. The maximum level of 214pCi/m^3 at BNFP is, however, less than 0.1% of the maximum permissible concentrations. The levels reported are those for tritium in the form of tritiated water vapor (HTO). The normal background also contains tritium in the form of T_2 and HT. These levels have not been measured in the BNFP vicinity, but have been extensively measured in other areas of the U.S. including Miami, Florida, by Ostlund et al. [24]. Since the majority of the expected tritium releases from the BNFP plant will be in the form of HTO, and since the present levels of HTO are considerably higher than the concentrations of HT, initial monitoring will be concentrated on HTO levels in the environment.

Krypton

A review of the problems associated with the release of ^{85}Kr is given in report PB-207 079 entitled "Krypton 85, A Review of the Literature and An Analysis of Radiation Hazards", by William P. Kirk [25]. This report published by the Eastern Environmental Radiation Laboratory, Montgomery, states that "Krypton 85 is an environmental contaminant for which progress in development of monitoring and control methodology appears to have outstripped knowledge and understanding of its biologic effects". In addition the basis for release regulations (MPC's) are entirely based on radiation dose calculations. These calculations have not been subjected to the scrutiny of in vivo experimentation [26]. The presently accepted maximum permissible concentrations in air for ^{85}Kr, as established by the AEC, NCRP and ICRP are 3×10^5pCi/m^3 for unrestricted areas, 10^7pCi/m^3 for occupational exposure for 40 hours weekly and 3×10^6pCi/m^3 for occupational exposure for a 168-hour week.

Estimates made by AGNS and measurements made at NFS (Nuclear Fuel Services, reprocessing plant) indicated that releases from the BNFP facility will result in concentrations orders of magnitude below this, and the average annual potential whole-body exposure to an individual living at the site boundary is expected to be approximately 0.28 mrem/year from the ^{85}Kr released from the BNFP to the environment. This is only about 0.2% of the exposure the individual would receive from natural background radioactivity. Although measurements of ^{85}Kr in the environment around BNFP are being planned, no actual measurements will be taken in the immediate future as part of the pre-operational inventory.

Plutonium

Plutonium is one of the more critical nuclides associated with the releases from a nuclear fuel reprocessing plant. The major characteristics of the plutonium isotopes are listed in Table V.

TABLE V. ISOTOPES OF PLUTONIUM

Isotope	Half Life (years)	Particle	Energy (MeV)	Ci/gm
^{238}Pu	86.4	α	5.5	17.4
^{239}Pu	24,296	α	5.2	0.061
^{245}Pu	6,580	α	5.2	0.23
^{241}Pu	13.2	β	0.021	112.0
^{242}Pu	379,000	α	4.9	0.004

There is a paucity of information on the behavior of plutonium in ecosystems and its radiological effects. [27-32]. Its transference from the atmosphere through soil and various kinds of sediments is not well understood. It enters plants from soil only to a very small extent and less than one thousandth of that ingested in diet is absorbed through the gastrointestinal tract, but when inhaled it is one of the most toxic of radioactive materials. The methods of detection of low levels demand tedious chemical separation and alpha spectrometric measurements. The background air concentrations of ^{239}Pu range from (1 to 10)x 10^{-17}Ci/m^{3}, where it is accompanied by smaller quantities of ^{240}Pu, from which it cannot be distinguished by alpha spectrometry, of ^{238}Pu in the approximate ratio of 0.02 to 0.04 and of ^{241}Pu. [33].

The inventory phase of the program will consist of measuring ^{239}Pu and ^{238}Pu in air filters and ecologically characterized soil and sediments. These measurements will be continued during the operational phase along with spectrometric analysis of air samples taken in the main stack.

In Situ Gamma Spectrometric Measurements

It is becoming more and more evident that in situ environmental gamma-ray spectrometric measurements are a tremendously powerful tool in meaningful ecological monitoring. [34,36]. This is particularly true for making dose and concentration measurements of gamma-ray emitters in soil and qualitative measurements of air concentration of releases from the nuclear industry. Plans are currently being made to use this technique in some special studies. One of these is a base line study of the plant vicinity of the doses due to the Th, V and K nuclides in soil and a comparison with similar measurements made with TLD's [36].

Non-radioactive Gaseous Effluents

The only significant non-radioactive gaseous process effluents from the recycling plant are oxides of nitrogen. Only approximately 10% is expected in the form of NO2, and the remainder is in the less toxic form of NO and N20. Small amounts of SO2 and CO are expected from the several standard boilers used for process and building heating. Adjacent to the reprocessing plant will be a uranium hexofluoride facility, which will discharge low levels of fluoride.

Detailed studies of these [9,12] include expected operational release rates and predicted concentrations at the unrestricted site boundary, along with a comparison to regulatory ambient air quality standards. Table VI[12] gives the maximum predicted off-site concentrations as a percentage of State and Federal Standards.

Operational Program

The operational monitoring program is being developed from the pre-operational and inventory phase. The primary emphasis will be on close coordination between stack discharges and their distribution to atmospheric, terrestrial and aquatic systems and on movement through these systems to man. Since the inventory and the identification of the principle pathways, including sites of greater bioaccumulation, will have been established, the sampling program can be far less extensive, and correspondingly more specific. Thus, the operational phase not only will be more efficient and reliable, but the costs also will be substantially reduced.

TABLE VI. COMPARATIVE OFF-SITE CONCENTRATIONS OF NONRADIOACTIVE GASEOUS EFFLUENTS

	Total BNFP Percentage of State and Federal Standards	
	Annual Average	Short Duration Peaks
NO_x*	0.9%	No Standard Given
SO_2	1.5%	15%
CO	4×10^{-4}%	0.03%
*HF**	2.8%	No Standard Given

*Maximum values based on predicted conditions while operating the Separations Facility process boilers on the standby fuel (No. 1 diesel oil). Values are considerably lower when operating these boilers on the normal fuel, natural gas.

**No State or Federal Concentration Standards. Comparison made to Emory University recommended Standard of 0.5μg/M^3 as compared to values given in the literature and by other states which vary from 1.0 to 5.0μg/M^3.

Systems Analysis

A system of digital computer programs has been developed to provide for data storage, retrieval and analysis, and to generate reports on sample preparation and analysis. In order to accommodate all possible sampling locations and types of samples, the computer programs were written to allow for open-ended data categories for each sample. New sampling program requirements or new sample locations can be added at any time. This feature is particularly important in field work when trained observers discover a situation which should be sampled. New data categories are then added as needed in the field and data collection can proceed with assurance that the newly collected data will be properly stored and available through the data base.

There are five distinct areas of activity in the sampling program and the data objectives of these areas are not necessarily compatible. In order to accommodate each of the subgroups, a general data sheet was devised and then modified for each group. Each data form uniquely identifies a sample through generation of a unique sample number when certain basic data columns are filled out. Routine data entries require circling appropriate digit sets which are alphabetically identified on the data sheet. Non-routine and numerical data must be written into columns.

The original data sheets are used for keypunch operations and the resulting cards provide input to the master data file through a file monitor program. Data sheet errors detected by the file monitor cause a print out which instructs the user as to the type of error detected. Acceptable data are added to the file and a print out of new file data is generated. As samples are carried through numerous processing and analysis stages, continual updates on the status of a sample are entered through a remote terminal. An update program allows for changes in the sample status category and the addition of analytical results to a sample file. The master file may be edited for errors and this responsibility has been limited to two people.

Data retrieval allows for both printed and graphic display of data through the creation of secondary files which may be used in data reduction programs available through a computing center library. The master file may be addressed according to any ranking of the sample data categories. This makes the data base as useful to one looking for the next sample to process as it is for the investigator who is looking for correlations.

Data analysis may be conducted according to the needs of each user. In addition to the computer program library, several programmable calculators are also in use with data sets derived from the master file. One interesting application in data analysis is the generation of a digitized map of the study area on which may be written mean concentrations of elements. This allows for variables such as BNFP operations, to be visually correlated with topography, vegetation, or a chemical species of interest. The advantage here is in quickly reviewing ordered data sets in their natural spatial relationship.

The final aspect of this system is that of report generation. Data entered into the master file are dated at time intervals and they may be retrieved on a temporal basis for generation of computer written reports. The basic report format is written and stored and computer generated variables are inserted for each report with the necessary tables and adjustment of text accomplished by the program.

EMPIRICAL BENEFITS

These are presented in two ways. The first consists of examples of direct empirical benefits to Allied-Gulf Nuclear Services derived from its monitoring program. The second is a summary statement of the empirical benefits of an ecosystem approach to monitoring which are applicable not only to radionuclides and other pollutants in the environment, but also to the development and design of industrial facilities.

Examples of Empirical Benefits to Allied-Gulf Nuclear Services

The initial significant contribution from the monitoring program was to place the facility within an ecological perspective unique to its particular on-site and off-site environment. By accomplishing much of this prior to construction, environmental problems were anticipated and provided for during the early flexible stages of facility design. Thus the difficult and expensive procedures for retrofitting were avoided.

A good example of these kinds of feed-back loops to facility design is that of the source, utilization and discharge of process water with regard to volume, quality, and radionuclides. The original facility design called for a once through cooling system, with water drawn from the Tuscaloosa aquifer and then released to Lower Three Runs Creek, which is located on the AEC Savannah River Plant site. The design also included release of low levels of radionuclides and of varying levels of stable elements from process and cooling water treatment, and the addition of some heat. In order to assess the impact of this tentative design, intensive biological and hydrological studies were made of the stream system, including its flood plain forests. These showed (a) that the projected rate of discharge was much higher than the stream could accommodate during normal flows without severe stress on both the stream channel and its flood plains; (b) that the quality of the stream at and below the point of discharge was good, and therefore the addition of treatment chemicals and heat could lead to violation of water quality standards; (c) that the stream and flood plain contained radionuclides at levels high enough to make it almost impossible to differentiate between these and possible additions from the facility, and (d) that certain areas of the stream showed prior damage and successional shifts which also precluded a positive differentiation in future years as to the cause of the damage.

Consequently, the design was altered (a) to provide for a recycling system which reduced the discharge by three-fourths, which is judged to have either no adverse effects or under conditions of low flow to be beneficial; (b) to eliminate the discharge of any radionuclides, and (c) to eliminate the discharge of chemical additives and heat. To assist with the latter, a specially designed ecological monitoring and conditioning pond was located between the plant and LTRC, and sewage was diverted through an irrigation spray system to a three acre plot.

These early changes (a) prevented costly retrofitting which at least in part would have been necessary; (b) gave complete protection to LTRC; (c) reduced the demand on the Tuscaloosa aquifer, and (d) eliminated the need for a more costly and long continuing monitoring program for LTRC.

Design characteristics for the gaseous waste treatment and stack monitoring facilities were also substantially influenced by ecological considerations, especially with regard to the treatment and release of plutonium, iodine and fluorine.

Significant inputs to the design for radioactive waste storage and monitoring came from the hydrological studies, such as those on the direction and rate of flow of ground water, and the tests which showed that leaks did not exist between the several underlying aquifers.

Other kinds of empirical benefits from the ecological characterization and inventory phase apply to (a) the operational monitoring program; (b) the identification of specific modes for an early warning system of bioaccumulation; and (c) a contribution to meaningful comparisons of data from other nuclear industry. For the operational program, the procedures can be more efficient and direct, and therefore much less extensive and expensive. For the early warning system, certain species in the turkey-oak community showed much higher levels of bioaccumulation of ^{137}Cs than those in other communities, and this community therefore is identified as a sensitive and primary site for continual sampling. For meaningful comparisons with other nuclear industry the use of the red maple (_Acer rubrum_) is suggested. This tree has a wide and very common distribution in the eastern U.S. from Florida to New England. Although only a moderate accumulator, it shows consistant patterns with respect to radionuclide concentrations in roots, small limbs, and leaves, as well as to physiological states. Thus, this easily accessible species could be used as an integrator and common denominator throughout the Eastern U.S.A. for the environmental distribution of certain radionuclides.

As a final example, the spin-off of basic research should be recognized as a significant benefit, since this is urgently needed for the development of regional models. The paper [11] presented in this symposium on _Flood Plain Transfer and Accumulation of ^{137}Cs from a Reactor Effluent Stream_ may be cited as an example of basic research derived from this monitoring program.

A Summary of Empirical Benefits from Ecosystem Monitoring

1. A systems approach to monitoring of the entire man-environment ecosystem which provides a comprehensive understanding of the major pathways and functional processes within this system. This approach gives to the facility an ecological perspective.
2. Inputs to decision making processes, as for example facility design and operational criteria.
3. A critical separation between effluents from various sources.
4. A realistic means for an early warning system which is capable of detecting long range trends and magnitudes of change prior to the potential development of serious problems.
5. Provision for a higher level of protection to the public and to a better understanding of risks vs benefits.
6. Provision for accommodation of unexpected problems which in turn increases efficiency and reduces costs. Unexpected problems may be new questions raised by regulatory agencies, new design criteria, and additional facilities on-site or in the region.
7. A methodology for the development of regional models and data banks which are emerging as essential to national and international monitoring programs.
8. A methodology for comparison of data from many sources at a level of confidence much higher than that in common use.
9. A monitoring program which is applicable to nonradioactive as well as radioactive substances, and is therefore transferable to the monitoring of pollutants from other industry.
10. A contribution to an understanding of the capacity of man's total environment to accommodate radionuclides.

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ENVIRONMENTAL PROGRAMS

(Session X)

Chairman:

H. JAMMET (France)

Mémoire commandé
ANALYSE CRITIQUE DU COLLOQUE

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Je voudrais, en commençant cet exposé relatif aux conclusions que l'on peut tirer de cette réunion, remercier les trois organisations internationales d'avoir donné aux participants l'occasion d'échanger leurs opinions et de faire le point de la situation actuelle.

Il est évident que je n'envisage pas de faire une analyse critique des communications qui ont été présentées, sinon je me ferais autant d'ennemis, et je n'envisage pas non plus de faire la louange des sujets qui me paraissent avoir été le mieux traités, parce qu'il est très difficile de manier la louange d'une façon équitable. Je voudrais, si vous me le permettez, tirer des conclusions générales sur trois points particuliers.

Le sujet du colloque était, en fait, le comportement des radioéléments dans l'environnement, et ceci en relation avec la protection radiologique de l'homme. Il est apparu que ce sujet, qui pouvait être traité d'une façon restrictive, a été interprété par les organisateurs et les participants d'une façon extensive, c'est-à-dire que l'on ne s'est pas contenté de considérer le comportement des radioéléments dans l'environnement, mais qu'on a aussi cherché à estimer quel était, au travers de ces transferts, l'irradiation éventuelle ou l'irradiation réelle de la population.

Ainsi, ces conclusions générales concerneront d'une part les transferts de radioéléments dans le milieu tels qu'ils sont apparus au cours de ces journées, d'autre part l'estimation des conséquences de ces transferts pour l'homme, et enfin l'application pratique de ceux-ci dans le contexte général des recommandations de la Commission internationale de protection radiologique.

TRANSFERTS DE RADIOELEMENTS DANS L'ENVIRONNEMENT

Tout d'abord, en ce qui concerne les transferts de radioéléments dans l'environnement, il faut se féliciter d'avoir trouvé ici un ensemble considérable de données. Je dois dire que, pour ce qui est des sources mêmes de la pollution radioactive, j'ai été un peu étonné de constater qu'on accordait, par exemple, une grande importance aux réacteurs nucléaires et que, bien qu'il y ait eu des communications à ce sujet, on avait donné l'impression de moins s'intéresser aux usines de traitement qui, il faut bien le reconnaître, sont tout de même les installations les plus polluantes dans la chaîne industrielle de l'énergie nucléaire. D'autre part, en ce qui concerne les radioéléments proprement dits, je crois que l'accent a été mis d'une façon heureuse sur le fait que l'on s'intéresse un peu moins aux produits de fission et davantage aux produits d'activation, que certains gaz rares, tels l'argon ou le krypton, font l'objet d'études, que certains transuraniens tel le plutonium commencent à être très étudiés, et enfin que le tritium a trouvé maintenant, je crois, sur le plan international la place à laquelle il a droit, compte tenu des quantités qui peuvent être rejetées dans le milieu. La nature physico-chimique sous laquelle se présentent les effluents radioactifs a été étudiée, mais peut-être mériterait-elle de l'être davantage et dans ce domaine des études plus poussées pourraient être profitables.

Ensuite, nous avons constaté que les vecteurs initiaux de la contamination, c'est-à-dire le vecteur atmosphérique et les eaux, qu'il s'agisse des eaux de surface ou des eaux marines, gardent toujours la faveur. Je dois constater qu'un très grand nombre, je dirais même une majorité de communications ont porté sur ces vecteurs initiaux. Certes, ces vecteurs initiaux sont très intéressants, en particulier pour la dilution et pour la dispersion des radioéléments dans le milieu, mais, nous allons le voir, ils ne constituent, en fait, que le début de la chaîne des transferts. Ainsi, il semble que le colloque s'est montré tout à fait traditionaliste en continuant d'accorder une très grande importance à ces vecteurs.

Les vecteurs intermédiaires, c'est-à-dire les transferts au sein de la biosphère, ont fait l'objet de nombreuses communications et il est normal que l'on attache une importance notable à une chaîne de transferts particulièrement complexe et au sein de laquelle on trouve des processus de reconcentration, de redilution, de redispersion et de remaniement dans les voies des radionucléides. Par contre, j'avoue être un peu surpris, bien que le sujet ait été traité dans un certain nombre de communications, de l'intérêt limité que l'on a porté à une chaîne de transferts qui, en ce qui concerne l'industrie nucléaire, apparaît pour le public comme la chaîne essentielle, je veux parler de la chaîne alimentaire.

Certes, on a parlé de la chaîne alimentaire, mais essentiellement en ce qui concerne la production agricole, qu'il s'agisse de l'agriculture ou de l'élevage. Mais je trouve qu'on a, par rapport au reste — bien entendu tout est relatif —, par rapport à l'importance accordée aux vecteurs initiaux, assez peu parlé de la transformation industrielle des produits alimentaires qui joue souvent un rôle de filtration vis-à-vis de la production agricole. On n'a pour ainsi dire pas parlé de la distribution commerciale des produits alimentaires. Or, la distribution commerciale des produits alimentaires est un facteur de dispersion de la radioactivité qui est sans doute de très loin le plus important. Vous savez comme moi que l'autoconsommation, en particulier dans les pays industriellement développés, est un mode d'alimentation qui tend à disparaître, et que l'alimentation se fait au travers des chaînes commerciales. Or, les chaînes commerciales peuvent disperser les radionucléides à des distances bien plus considérables que les courants marins ou que les courants atmosphériques. Je dois dire que j'ai été surpris de voir que ces voies nouvelles paraissent assez peu exploitées. Enfin, il y a les problèmes liés à la consommation alimentaire proprement dite, c'est-à-dire à la fois aux habitudes culinaires et aux régimes alimentaires des populations, qui sont importants parce que les habitudes culinaires peuvent jouer de nouveau, comme l'industrie de transformation alimentaire, un rôle de filtration dans le transfert de la radioactivité.

Et, finalement, on aboutit aux vecteurs terminaux de la pollution radioactive que sont l'inhalation, dans un certain nombre de cas assez limité d'ailleurs, l'ingestion, qui reste de très loin la voie préférentielle, et, secondairement, l'irradiation externe à partir des retombées radioactives du fait des rejets d'effluents. Au fond, quand on considère l'ensemble des données qui ont été discutées au colloque on s'aperçoit que les recherches de laboratoire sont assez poussées, c'est-à-dire que de par le monde, il y a maintenant un certain nombre d'équipes qui font un excellent travail en matière de transferts, soit sur le plan des vecteurs atmosphériques et hydrologiques, soit sur le plan des transferts dans l'atmosphère, soit sur le plan de la production agricole. Je crois qu'en effet on consacre moins de travaux à la transformation alimentaire, aux études de circuit de distribution et à celles de la consommation proprement dite, mais ceci est en partie compensé par les études faites in situ.

Je crois qu'au cours de ce colloque il est apparu que les études et les recherches faites en laboratoire d'une part et les vérifications particulières faites in situ d'autre part étaient tout à fait complémentaires les unes des autres. On ne saurait les dissocier. Les recherches de laboratoire ont l'avantage de nous fournir des informations sur la variabilité des valeurs des paramètres de transfert. Les études in situ, qui entrent dans le cadre opérationnel, ont pour intérêt de nous permettre la vérification dans des conditions particulières et concrètes des valeurs réelles que l'on doit prendre en considération dans tel ou tel cas.

Enfin, un grand nombre de communications nous ont montré que, à l'inverse de ce qui se passait autrefois, on attache beaucoup d'importance désormais aux synthèses, c'est-à-dire à l'ensemble des travaux qui permettent de tirer des conclusions soit de l'expérimentation en laboratoire, soit des observations faites in situ. Ceci m'amène tout naturellement à aborder la deuxième partie de mes commentaires, qui concerne donc l'exploitation des données expérimentales et des données d'observation dans le but d'estimer les conséquences des rejets pour la population.

ESTIMATION DES CONSEQUENCES POUR L'HOMME

Je pense que pour bien analyser cette exploitation, il est intéressant de considérer les deux stades que sont d'une part les études prévisionnelles et d'autre part les études opérationnelles. Je dois dire que, là aussi, j'ai été un peu surpris de voir combien les études prévisionnelles avaient la faveur, et combien les études opérationnelles paraissaient parfois moins intéressantes à certains. En fait, je crois qu'elles sont parfaitement complémentaires. Je pense que nul ne niera l'importance de prévoir les conséquences des rejets, mais il est tout aussi important de vérifier à partir du moment où les opérations sont en cours que ce que l'on a prévu se réalise bien et que les hypothèses — parce que souvent les prévisions sont basées sur ces hypothèses de travail —, ou les valeurs choisies pour les paramètres concordent bien avec la réalité.

On a eu l'occasion, au cours du colloque, de voir que toute une terminologie nouvelle commençait à apparaître et que des concepts nouveaux entraient peu à peu dans la pratique courante. D'abord, la notion même de l'environnement. L'environnement a été souvent en tant que tel l'objet de définitions variées. En fait, je crois maintenant qu'il est bien admis par tout le monde que l'environnement, ce n'est pas le milieu atmosphérique ou le milieu aquatique qui sert de réceptacle initial aux rejets de substances radioactives. L'environnement, c'est tout ce qui est compris entre la source de pollution, c'est-à-dire les émissaires de radioactivité, et les voies aérodigestives supérieures de l'homme si l'on excepte le cas de l'irradiation externe. Par conséquent, l'environnement, pour nous, cela comprend tous les vecteurs: l'air, les eaux, la biosphère, la chaîne alimentaire; ainsi, lorsque l'on étudie les transferts au sein de l'environnement, on est amené à envisager ce que certains appellent la capacité radiologique; d'autres estiment encore un peu abusif de l'appeler ainsi, mais la capacité radiologique peut s'interpréter d'une façon assez simple en la considérant comme la capacité, pour une chaîne donnée de l'environnement, de transférer d'une façon dynamique la radioactivité depuis la source jusqu'à l'homme. Il est évident que de telles chaînes peuvent être extrêmement complexes mais que le terme de capacité de transfert garde toute sa valeur.

Il ne faut pas confondre la capacité de transfert avec la capacité de réception. Ce qui nous intéresse, puisque au fond nous nous intéressons à ce qui arrive à l'homme, c'est la capacité de transfert. Que la capacité de réception présente de l'intérêt dans certains cas, c'est vrai, et nous avons à l'étudier, mais il est indéniable que la capacité de transfert apparaît jusqu'à présent comme la principale. Certes, on peut préciser: il n'y a pas une capacité de transfert, il y a des capacités de transferts; il y a la capacité limite, c'est celle qui correspond hypothétiquement à l'irradiation limite de la population compte tenu des limites de doses de la CIPR; il y a des capacités stipulées, il y a des capacités acceptées, ce sont celles qui correspondent aux transferts qui sont considérés comme acceptables compte tenu des doses que l'on considère comme acceptables pour le public. Et par conséquent on est amené, nous l'avons vu au cours du colloque, à mettre au point des méthodes qui permettent d'estimer la capacité de transfert de l'environnement pour les radioéléments et de relier cette capacité de transfert à l'irradiation voulue ou effective des populations.

Ces méthodes d'estimation sont apparues dans les communications et au cours des débats comme étant variées. On peut les concevoir très simples avec des approximations relativement grandes. On peut les concevoir extraordinairement complexes en essayant d'injecter tous les paramètres que l'on peut imaginer pour avoir quelque chose d'aussi complet que possible. A ce moment-là, on est bien obligé de passer par des moyens de calcul qui sont les moyens modernes de l'informatique. Mais de toute façon ce qu'il faut bien savoir, c'est que, quelle que soit la méthode utilisée, on ne retrouve à la sortie des modèles que l'on emploie que ce que l'on a mis à l'entrée. Et si on injecte à l'entrée des données qui sont approximatives on ne peut retrouver à la sortie que des conclusions approximatives. Au fond la difficulté du problème avec lequel nous sommes confrontés, ce n'est pas tellement la mise au point de modèles plus ou moins compliqués de transfert et l'estimation de la capacité de transfert, mais c'est en fait dans la pratique de voir dans quelle mesure l'utilisation de ces modèles, compte tenu des données dont on dispose, correspondent à la réalité ou non. Par conséquent, les études sur la variabilité des valeurs des paramètres, les études sur les particularités de chacun des sites ou des divers environnements qui entourent les installations sont hautement souhaitables parce que ce sont elles seules qui permettront, soit dans des cas généraux, soit dans des cas particuliers, de mettre dans les modèles qui ont été choisis des données valables qui permettront d'obtenir des résultats valables.

Ensuite, si nous voulons franchir un pas de plus, nous aboutissons, à partir des transferts de radioéléments, à l'estimation des incorporations par la population. Et l'incorporation par l'homme pose des problèmes qui sont ceux des répartitions métaboliques, dont je ne parlerai pas puisque ce n'était pas l'objet du colloque. Finalement, après cette répartition métabolique, nous passons à l'estimation des doses effectivement reçues par le public, et, après l'estimation des doses, celle du détriment pour la population.

C'est dire que les travaux qui ont été présentés au cours de ce colloque constituent la base normale à partir de laquelle on peut continuer pour faire une estimation des incorporations, une estimation des doses et une estimation des détriments.

Au début de ce colloque, M. Lindell a attiré notre attention sur le fait qu'actuellement on s'engageait peu à peu dans une extension de la notion de dose, et ceci est intéressant à considérer car cela implique que les travaux qui sont faits sur les transferts de radioéléments dans le milieu soient complétés par des études concernant le public. Je m'explique: Vous savez que jusqu'à présent les recommandations de la Commission internationale de protection radiologique portaient sur ce que l'on appelait les doses individuelles pour l'homme; vous savez qu'en particulier la publication 7 de la CIPR donnait des méthodes qui consistaient à choisir les voies critiques de transfert et finalement le groupe critique de la population pour lequel on souhaite que les doses individuelles restent au-dessous d'une certaine valeur. Et vous savez que cette méthode a été utilisée avec des résultats fructueux dans un grand nombre d'études concernant la pollution radioactive à partir d'installations nucléaires. Or il se trouve que l'évolution normale nous oblige de plus en plus à considérer, non pas les doses seulement, mais le détriment pour la population. Lorsque l'on veut estimer le détriment, on est obligé de considérer le nombre de personnes qui sont exposées. C'est la raison pour laquelle on vous a indiqué, et vous le saviez déjà, que la notion de dose collective est une notion qui va peu à peu entrer dans les mœurs et que par conséquent les études de transfert de la radioactivité doivent tenir compte non seulement du transfert aux individus mais du nombre d'individus auxquels la radioactivité est transférée. Il est évident que de ce fait la complexité du problème apparaît dans toute son ampleur et, comme on vous l'a dit au début, on peut considérer le cas d'une installation, et, en appliquant les méthodes de la publication 7 de la CIPR, considérer le groupe critique de la population correspondant à cette installation. On peut pour ce groupe critique évaluer les doses individuelles. Et à partir du moment où l'on connaît la taille du groupe de population considéré et d'autre part la distribution au moins approximative des doses individuelles on peut faire une estimation de la dose collective correspondant à ce groupe. Par ailleurs, il est indéniable que l'on peut s'intéresser, dans un premier temps, exclusivement au groupe critique de la population; mais il n'est pas sans intérêt de voir qu'elle peut être la dose collective, non seulement pour le groupe critique correspondant à une installation, mais pour l'ensemble de la population, que ce soit sur un plan régional ou sur un plan mondial. Et ceci parce que, lorsque des autorités auront peu à peu à s'occuper, non pas d'une installation donnée, mais de l'ensemble des installations, on s'apercevra très vite que l'ensemble des installations, par exemple, l'ensemble des réacteurs d'une région donnée, aboutit par les transferts de radioactivité à un groupe critique qui n'est pas forcément l'ensemble des groupes critiques de chaque installation, mais qui peut être un groupe critique nouveau. En plus on peut s'intéresser également à l'ensemble des installations et à la dose collective qui en résulte pour l'ensemble de la population, soit sur un plan régional, soit sur un plan mondial. Il est évident qu'on ne pouvait pas demander dans ce colloque qu'on nous apporte des travaux d'une telle ampleur et représentant un tel effort, mais il faut bien dire que la tendance qui se développe laisse penser que de tels travaux devront s'effectuer dans le futur, cela toutefois avec quelques considérations qui relèvent des possibilités humaines en la matière. Il est certain qu'il est déjà difficile de définir d'une façon correcte le groupe critique correspondant à une installation donnée. Il est certainement beaucoup plus difficile d'estimer le retentissement d'une installation donnée, je prends par exemple une installation électro-nucléaire, sur l'ensemble de la population mondiale, parce qu'on aboutit à des dispersions de radioactivité telles que les valeurs obtenues sont si petites que, après multiplication par un nombre aussi grand de personnes, on tend vers des situations que les mathématiciens connaissent bien et qui sont celles de la multiplication d'un infiniment petit par un infiniment grand dont le résultat est, vous le savez, très difficile à estimer. Par conséquent, il ne faut se faire aucune illusion sur la difficulté de telles entreprises, mais ce n'est pas une raison pour ne pas s'y préparer et, d'autre part, pour ne pas considérer qu'on a quand même un moyen complémentaire d'estimation.

Au fond l'estimation de l'irradiation ou de la contamination de la population dans son ensemble est peut-être plus facile à prendre par l'autre bout, c'est-à-dire en estimant quelle est réellement – car on peut la mesurer dans un certain nombre de cas –, la contamination effective des populations, et en remontant la chaîne jusqu'aux sources. Je vous rappellerai le succès que de telles méthodes ont rencontré dans les travaux du Comité des radiations des Nations Unies. Il ne s'agissait pas de l'industrie nucléaire, il s'agissait essentiellement, du moins au début, des applications militaires de l'énergie nucléaire. On a fait dans ce Comité des estimations prévisionnelles de ce que pourraient être les retombées radioactives pour la population régionale ou pour les populations mondiales. On a fait des calculs sur les doses engagées jusqu'à l'an 2000, par exemple. Mais un énorme travail a été fait au moyen des réseaux de surveillance mondiaux, sur les retombées effectives et sur les contaminations effectives des populations. Et c'est l'ensemble des deux méthodes, la méthode prévisionnelle et la méthode opérationnelle, qui a permis au Comité de conclure sur les doses effectives reçues par les populations et sur les détriments actuels ou prévisibles. Par conséquent, je pense qu'il est important pour nous de voir que ces deux méthodes complémentaires devraient, à la suite des travaux qui ont été présentés ici, apparaître comme des méthodes d'estimation valables.

APPLICATIONS PRATIQUES DES RECOMMANDATIONS DE LA CIPR

Je voudrais en terminant évoquer ce que j'appellerai la conformité des travaux qui ont été présentés ici avec l'application des recommandations de la CIPR. Vous savez que je suis le président du Comité 4 de la CIPR qui s'occupe justement de l'application des recommandations. Certains penseront que c'est un «dada» de ma part, mais effectivement je crois que cela a beaucoup d'importance, parce que l'utilisation de vos travaux et de vos résultats va aboutir finalement à l'estimation des risques et des détriments pour l'espèce humaine du fait des rejets radioactifs des industries nucléaires. Vous savez qu'autrefois on estimait que l'essentiel des recommandations de la CIPR reposait sur les limites d'irradiation de l'homme. Peu à peu, depuis quelques années, on a pris l'habitude d'attacher beaucoup plus d'importance à une petite phrase d'un paragraphe, le paragraphe 52 de la publication 9 de la CIPR, qui dit qu'il faut éviter toute irradiation inutile et qu'il faut réduire l'irradiation réelle autant que cela est réalisable, compte tenu des impératifs économiques et sociaux. Cette petite phrase contient deux choses essentielles: on ne doit pas se contenter de respecter les limites d'irradiation, mais on doit se livrer à une étude d'optimisation de la protection, c'est-à-dire faire en sorte que la protection réalisée soit la meilleure, compte tenu d'une part des détriments et d'autre part des avantages que l'on tire de l'utilisation de l'énergie nucléaire.

Je voudrais mettre en garde contre un certain nombre de déviations possibles et de difficultés très importantes. On peut interpréter les termes «éviter toute irradiation inutile» dans le sens de la justification de l'utilisation de l'énergie nucléaire. Dans ce cas, on fait une analyse coût-bénéfice, qui consiste à mettre en balance d'une part le détriment pour la population et d'autre part les bénéfices pour la population. C'est la justification. La deuxième partie de la phrase est relative à l'optimisation, qui consiste à mettre en balance l'augmentation du coût de la protection par rapport à la diminution du détriment. C'est donc une différentielle; en fait on apprécie les variations de la protection par rapport aux variations des dommages. Or, quand on analyse les possibilités actuelles que nous avons de traiter le problème, on s'aperçoit que le détriment n'est déjà pas tellement facile à préciser. Comme je l'ai dit tout à l'heure, le détriment demande que l'on connaisse la dose collective; et qu'en plus on connaisse, à partir de la dose collective, les corrélations dose-effet qui entraînent par exemple l'induction de cancer, l'induction de mutations, etc. Par conséquent, on est là dans le domaines d'hypothèses biologiques souvent plus ou moins fondées et le plus souvent choisies en fonction de leur caractère pessimiste qui nous met en confiance. En supposant que l'on prenne les hypothèses les plus pessimistes on aboutit aux conclusions les plus prudentes. Donc le détriment sera difficile à exprimer, car il faudra connaître les doses collectives. Je vous l'ai dit, les doses collectives ne pourront être connues que grâce aux travaux que vous faites, c'est-à-dire à l'estimation des transferts. Le coût de la protection n'est pas si facile à estimer non plus, parce qu'il y a des coûts directs de la protection: épaisseur de béton, confinement, etc., mais il y a aussi des coûts indirects de la protection. Il se peut que l'excès de protection entraîne des détriments dans d'autres domaines. Il se peut que la protection entraîne des difficultés de travail, des baisses de rendement. Le coût de la protection comprend tout cet ensemble, non seulement le paiement des protections matérielles ou des contraintes humaines, mais également le retentissement de la protection sur la production ou sur la santé des travailleurs ou du public par d'autres intermédiaires. Mais où les difficultés sont les plus grandes, c'est lorsque l'on essaie d'examiner l'estimation des bénéfices, parce que les considérations économiques et sociales sont, si vous voulez, faciles à exprimer tant qu'on ne fait que les estimer, mais quand on veut les chiffrer on se rend compte que déjà dans le domaine économique on a des difficultés, et que dans le domaine social on en rencontre de bien plus grandes. L'estimation économique, par exemple, du nombre de kilowatts produits est une chose, mais l'estimation du bien-être que ces kilowatts apportent à l'humanité, c'est autre chose. Finalement, les considérations économiques, sociales, psychologiques, politiques que l'on doit prendre en compte font que l'estimation est très difficile. Or, ne vous faites aucune illusion, quand on veut comparer le détriment, le coût de la protection et le bénéfice tiré des opérations, on est amené à choisir une unité commune. Jusqu'à présent, la seule unité que l'on ait trouvé, c'est l'unité monétaire. Or l'unité monétaire, c'est l'estimation en argent, quelle que soit la monnaie choisie, des bénéfices économiques, sociaux et autres. C'est l'estimation en argent du coût de la protection, c'est aussi l'estimation en argent de la vie humaine lorsqu'il s'agit des détriments. Vous comprendrez que dès lors, il y a non seulement des difficultés pratiques qui apparaissent, mais des difficultés fondamentales. Il y a des gens qui ne conçoivent pas que l'on fasse des estimations de la vie humaine, qui ne se rendent pas compte qu'on les fait tous les jours, parce qu'on est habitué à les faire d'une façon inconsciente; et là où la difficulté devient encore plus grande, c'est que souvent l'opinion publique a des facteurs de pondération tout à fait étonnants pour la vie humaine. Si vous prenez le cas des transports, je pense que dans la plupart des pays les morts du trafic automobile ne frappent pas beaucoup l'opinion publique, du moins si on regarde la presse; il suffit qu'un avion tombe pour qu'immédiatement la première page des journaux le signale, et si par hasard un bateau coule, cela prend les caractères d'un drame. Parce que l'opinion publique considère que les bateaux sont faits pour ne pas couler, que les avions sont faits pour ne pas tomber, mais on admet fort bien que les voitures soient faites pour se rencontrer. La vie humaine, de ce fait, n'est pas jugée du tout de la même façon. La vie des passagers d'un bateau est implicitement considérée comme supérieure à la vie des passagers d'automobiles particulières et, d'une façon générale, les transports en commun sont considérés par l'opinion publique comme ne devant pas avoir d'accidents alors que la même opinion admet qu'il y en ait beaucoup pour les transports privés. Je cite cela à titre d'exemple, mais nous pouvons le retrouver dans les domaines qui nous intéressent, et il se peut fort bien que l'opinion publique trouve normal des détriments considérables par des nuisances chimiques et trouve anormal les détriments relativement petits par des nuisances radioactives. Il est évident que dans ce domaine nous ne pourrons progresser qu'avec beaucoup de circonspection. Finalement, il y a peut-être une méthode, qui vaut ce qu'elle vaut, mais qui a au moins le mérite de comparer entre elles des choses comparables: c'est la méthode comparative.

A partir du moment où l'on est capable d'estimer le détriment lié à une activité nucléaire, on peut le comparer à celui lié à une autre activité qui donne les mêmes bénéfices, par exemple, la production d'un certain nombre de kilowatts. La comparaison des deux détriments, l'un par des moyens classiques l'autre par des moyens nucléaires, peut être plus facile, finalement, que la balance entre le détriment et le bénéfice. De même, si l'on a de grandes difficultés à estimer le détriment de façon raisonnable, on peut se contenter de l'estimation des doses. A partir du moment où l'on a une idée correcte des doses collectives, on peut essayer de comparer d'abord les doses collectives entre elles: par exemple, les doses collectives dues à l'énergie nucléaire aux doses collectives dues à l'irradiation médicale; ou bien les doses collectives dues à l'irradiation artificielle aux doses collectives dues à l'irradiation naturelle et aux fluctuations de l'irradiation naturelle. Cela est peut-être plus facile que de faire une analyse coût-bénéfice comme on le fait pour la justification, ou une analyse coût-efficacité comme on le fait pour l'optimisation.

Si en terminant j'ai voulu attirer votre attention sur ces voies de l'avenir, c'est parce que ce sont vos travaux qui y conduisent. C'est dans la mesure où vous ferez de bonnes estimations des transferts et ensuite de bonnes estimations des incorporations et des doses collectives que l'on sera apte ou non à faire des comparaisons valables ou à faire des balances valables. Et même si vous arrivez à faire quelque chose de correct sur le plan scientifique et technique, ne croyez pas que vous en aurez terminé avec l'opinion publique. Une séance mouvementée de contestataires à Aix-en-Provence a permis de constater que certains d'entre eux considèrent que ce que vous, vous considérez comme bénéfique est pour eux une calamité, c'est-à-dire que la production d'énergie est le mal en soi. Vous allez vous retrouver, en face de l'opinion publique, avec des problèmes qui ne sont plus des problèmes d'estimation — qui sont déjà assez compliqués nous venons de le voir — mais des problèmes purement qualitatifs ou moraux qui consistent simplement à considérer que la croissance est mauvaise en soi, que l'énergie est mauvaise en soi et pourquoi pas, bientôt, dans un contexte de surpopulation, que le cobalt-60 produit par les réacteurs nucléaires permet de guérir des cancéreux et qu'au fond la bonne nature permettrait de les éliminer si on ne les traitait pas. A partir du moment où l'on commence à considérer que toute activité humaine est nocive et que le progrès, comme on nous l'a dit, n'est pas le bonheur, ce qui est vrai d'ailleurs, on peut aboutir aux errements les plus grands.

Je voudrais justement attirer votre attention sur le fait que dans la mesure où vos travaux vous auront conduits à faire des estimations correctes qui soient présentables objectivement, qui soient inattaquables sur le plan scientifique et technique, vous aurez en main, je l'espère, des armes suffisantes pour lutter contre ceux qui posent le problème sur le plan philosophique et moral, ce qui n'est pas l'objet de nos débats.

PANEL DISCUSSION
AIMS AND NEEDS FOR ENVIRONMENTAL PROGRAMS RELATED TO THE NUCLEAR INDUSTRY

Chairman:

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Panel Members:

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F. MORLEY (United Kingdom)
P. PELLERIN (World Health Organization)
W.O. SCHIKARSKI (Federal Republic of Germany)

PANEL DISCUSSION

AIMS AND NEEDS FOR ENVIRONMENTAL PROGRAMS RELATED TO THE NUCLEAR INDUSTRY

H. JAMMET (Chairman): In response to our request, quite a large number of questions to the Panel have been submitted in writing, so that we have had an opportunity of examining them in advance and of deciding which of us can best answer in each case. What I propose to do, then, is to state the questions for the benefit of the Symposium participants as a whole, whereupon we shall offer our replies.

Question No.1, by P. Krejsa

> Mr. Pellerin showed us that there is a common philosophy underlying monitoring of nuclear plants and also that most of this monitoring is done for reasons of public concern about radioactivity. However, there is another trend in this field that can much less be justified on a technical basis. What I have in mind is the elaborate pre-operational survey of a projected nuclear power station. For the purpose of showing changes in environmental radioactivity during the subsequent operation of the reactor, many samples of air, milk, fish, grass, vegetables and so on are taken to obtain baseline data. But it is well known that natural radioactivity, for example in air, varies with time over a range much wider than a nuclear reactor will contribute under normal conditions. The contribution from weapon test fall-out likewise shows a variation in time, so that here also it is not possible to use the pre-operational survey data as background values.
> Would it therefore not be advisable to develop international directives for pre-operational surveys? Measurements should be restricted to those that are meaningful, to the exclusion of all others.

P. PELLERIN: Mr. Krejsa's remarks tie up both with what Mr. Jammet has said in his critical review paper earlier this afternoon and with my own paper on Tuesday. Mr. Krejsa clearly believes not only that too much monitoring and survey work during the operation of a facility can – by its very extent – give rise to unjustified public apprehensions but also that the same considerations apply to pre-operational surveys conducted before the facility is ever erected.

My personal view is that Mr. Krejsa is quite correct, and his feelings reflect the anxiety that most of us in atomic energy experience, namely that our activities will be misinterpreted. We should take great care not to arouse groundless public fears by the mobilization of an over-meticulous sampling or monitoring effort. Nuclear energy in general and radiation protection in particular (the latter being the facet which comes into most direct contact with the public) should strive to achieve an image of normality in the eyes of the – admittedly quite small – section of the population that is liable to worry too much. After all, nuclear energy is at least as safe as any other technique, and we really don't have to be sensationalistic about its dangers.

Question No. 2, by the USSR participants

What is the Panel's opinion on:
(1) The possibility of siting nuclear power plants near large urban centres;
(2) The establishment of limits for the release of radionuclides from nuclear power plants taking into account the likely future development of the latter; and
(3) The establishment of unified compulsory procedures for sampling and analysis of air, water and other media in order to monitor the operation of nuclear power plants?

F. MORLEY: In the United Kingdom at least, the siting of power stations near large population centres is more than just a possibility – it is about to become a reality. Two large advanced gas-cooled reactors are being constructed in the north of England, one near Hartlepools and the other near Heysham. While I have no precise figures with me, I can tell you that a population numbering some hundreds of thousands lives within about 10 miles of the Heysham station perimeter. Since this question has been asked at a Symposium dealing with the release of radionuclides to the environment, perhaps it is in the mind of the questioners that such releases in normal operation are an important factor in power station siting. Now, certainly in the United Kingdom, this is not true. The amount of radioactivity released by our reactors is very small, and I believe this is the case elsewhere as well. The 1972 UNSCEAR report showed, on the basis of the data received, that the doses to local population groups from the operation of power stations was almost everywhere less than one per cent of the ICRP recommended limit.

Far more important is the safety of the overall system and in the United Kingdom I believe one reason for the relaxation of siting policy relating to gas-cooled reactors has been the adoption of pre-stressed concrete pressure vessels.

Turning now to the second part of the question, I would recall that this morning we heard Mr. Mitchell explain how authorized limits are fixed in the United Kingdom. It is a question of dialogue between the operator and the authorizing Government departments; the limiting environmental capacity – to use our terminology – is calculated, but the actual authorization to discharge waste will cover only the amount that is needed, which is always less than the limiting capacity. We believe that this procedure has been very effective in the past, and we foresee no change in the immediate future.

With regard, finally, to the third part of the question, if it refers to the standardization of analytical techniques, then I think the answer is most certainly in the affirmative and I believe that the IAEA in particular has already gone some way towards setting up procedures for intercomparison of analytical techniques. If on the other hand the question is asking whether there should be a single regime of measurement around each station, then I say equally emphatically that the answer is in the negative. In performing these measurements we are principally concerned with estimates of dose; we therefore require measurements on relevant exposure pathways, and the identity of these exposure pathways is highly specific to each site.

Question No. 3, by R. Bittel

> The fate of radionuclides in the environment, and hence the hazards imputable to them, are governed by a number of factors, some of them of a physico-chemical nature.
> The general level of pollution, particularly organic pollution, is an important characteristic of the environment. This being so, should we not take into account the incidence of microbiological parameters, since very many reactions affecting mineral and organic pollutants are due to microorganisms, which represent the primary agents of biochemical transformations?

H. JAMMET (Chairman): I feel that this question should be considered against the background of the complexity of the processes occurring in the biosphere. All students of soil science, for example, are aware of the importance of microbiological processes, and here there is scope for some general investigations aimed at determining how far factors associated with the action of microorganisms are of importance in the estimation of transfers. The answere here, then, might lie with the general guidance to be imparted to laboratory studies or, in certain cases, to investigations which might profitably be undertaken on these factors.

Question No. 4, by D. Maire

(1) Calculations forecasting the migration of fission products in water and in soils towards man call for numerous data; these are determined experimentally by various laboratories, which frequently arrive at contradictory results. Is there any national or international body engaged in co-ordinating these activities and building up a library or thesaurus of the most reliable data?
(2) Could not models for the diffusion of fission products in the atmosphere be treated analogously to 'benchmark problems', as is done in the case of calculations for radiation protection shielding?
(3) The diffusion models we have heard described are applicable to diffusion over a long distance and on smooth terrain. For calculations of accident consequences, however, the doses received in the neighbourhood of the site ($R < 20$ km) are also of interest. Has any work been done in this sphere taking account of the effect of buildings, and also dealing with atmospheric diffusion above large urban centres?

K.E. COWSER: Perhaps I may take this opportunity of offering some general, and then some more specific, remarks on environmental program needs. In a general way, and outside our very natural concern with radioactive materials, we might begin reflecting on needs that relate to such items as thermal effects, the impact of biocides or corrosion inhibitors used in cooling systems, site selection criteria, future land use as it might affect a facility, the behaviour of plutonium, studies of ecological processes which are in fact mission-oriented, and so on.

More specifically, I have been reflecting on three or four of these needs, and I feel that several of the questions put to the Panel also illustrate a concern with them. One of them is the need for an adequate definition of problems, and by this I mean in particular the need to establish priorities to be dealt with within the context of the available time and money. Here much can be done by regulating one's methods of work. We must anticipate the problems that are likely to arise in the future, and must attempt to devise what I would call strategies in order to detect possible impacts far enough in advance to prevent them from occurring in many cases. Aids to the definition of problems – and these are just a few that occur to me – may include such things as staff assessments, conferences at local, regional, national and international level and state-of-art reviews, coping with the flood of information of which we were reminded earlier in the week, and condensing perhaps 500 papers all dealing with the same restricted field into one document which we shall have the possibility of reading instead of merely filing.

A second specific point is information development and availability, and here I think there is a wealth of information from routine environmental monitoring of which we should begin to make use. This will provide us with a tool for developing indices and regulations governing pollution, and with a capability to verify some of the models that have been developed during research activities. I believe that an effective monitoring network can serve as an early warning system for pollutants, and that the development of biological indicators has a very real part to play in assisting us to determine the health of ecosystems. We must, in this connection, also give attention to the communication of information, in order to avoid duplication of effort in some of our research work, and we might consider whether a data bank – well, perhaps not a data bank but a specimen bank – would be of value in terms of collecting and disseminating baseline information in the future.

The question of interdisciplinary research teams has arisen on a number of occasions and Mr. Liverman reflected on the subject during his review. I feel that, as environmental problems become more and more complicated, it will be impossible to avoid large teams of scientists and engineers working collectively on the solution of those problems. This certainly represents a potential obstacle to communication – how do you talk to someone who is trained in a narrow field? – but I think the difficulty can be largely overcome by dialogue between the specialists as together they attack a problem of common interest. As to who should furnish the interdisciplinary team capability, it seems to me that the nuclear energy laboratories, in view of their past and present efforts and involvement, are the natural agents for this purpose.

One part of Mr. Maire's question deals, I believe, with the availability of information on atmospheric models. Well, in 1968 the United States Atomic Energy Commission issued a complete list of models in its publication: "Meteorology and Atomic Energy", and I understand that work is being done on the effect of buildings at the Idaho plant and by the Tennessee Valley Authority.

H. JAMMET (Chairman): Perhaps I might re-emphasize my belief – strengthened as a result of a discussion I had with Mr. Liverman a few days ago – that the correct application of the new concepts, relating not only to individual but also to collective doses, will make quite heavy demands on our resources. In particular, we shall need to have available interdisciplinary teams accustomed to working together on a continuing basis. It will not be enough to bring about occasional dialogues between meteorologists, hydrologists, biologists, agronomists, experts on the food chain, nutritionists, doctors and, finally, physicists. Success will only come from the existence of teams large enough and sufficiently well-endowed to tackle the overall problem, and in any given country such teams should normally be operating at national level — in the service of the country as a whole. Mr. Liverman gave it as his opinion that at some of the research centres working under contract in the United States of America there were teams possessing the critical mass, if you like, which enabled them to deal with problems on behalf of all the people concerned. For example, the operator of a nuclear installation needs to estimate the consequences of his operations; he can, with some difficulty, do this in respect of the critical group, but if he wants to do so in respect of a substantial regional population or even of the world at large, he needs detailed information on the commercial distribution of food commodities etc., and it is here that he requires the aid of a really powerful team. The Government authorities, on their side, are responsible for the welfare of the whole population, for all the critical groups taken together, and they also need to check on the operator's activities. They are concerned with the collective dose received by the whole country. Thus it seems a good idea for competent teams to work on behalf of the entire community, placing their human and material resources at the disposal of the operators or of the national authorities as appropriate. Fragmentation or dispersal of effort will, I feel, not lead to the desired result.

W.O. SCHIKARSKI: I should like to make a few points on the problems of modelling, which was a major issue at this Symposium.

Modelling in connection with air and water pollution concerns of course not only the nuclear industry but many other branches of production as well. I personally have learnt from this Symposium that we have now available a number of well-developed computer models, which are of great importance for understanding the behaviour of pollutants released to the environment. I believe that these models are necessary, well worthwhile, and should be further developed. However, I have missed at the Symposium a critical review of their potentialities and of their limitations. True, questions about the accuracy of these models were asked a few times during the discussions, but this matter of accuracy is, to my mind, more serious than model designers seem to realize.

Let me list a few problems. We lack input data, only in a few cases are they available to the extent required, and their lack is particularly keenly felt for regional modelling. Then we have the problem of transport processes. These are complex processes, they involve physical, chemical, meteorological, hydrological, biological and other aspects, and many of them are not well understood even in the scientific circles that have discovered and described them, and first formulated them in mathematical terms. In describing the behaviour of pollutants by models, therefore, concentration on the essential points is the only way to proceed, since a complete description in all details is impossible. It would in any case lead to computer models of extremely large size, in some cases even exceeding today's computer capacities. For example, only about one cubic mile of atmosphere can be simulated reasonably on the largest computer now available. Modelling has thus to constitute a simplification, a compression, a reduction of the whole problem to its essentials.

This is in fact more or less what is done in the models presented at this Symposium, but one result of this is that we have to find out where we are in the over-simplification process. What are the errors consequent upon neglecting certain parameters or upon extrapolating simple correlations – e.g. extrapolating up to 100 km simple diffusion formulas developed for distances in the atmosphere of only 10-15 km?

We should also ask ourselves what is the real purpose of modelling. Is it to diagnose the present situation concerning emission? Is it to prognosticate future trends in the burden of certain populations, of certain ecosystems, and so on? Is it to identify the critical pathways of critical pollutants and their burden? Is it to study the implications of possible changes in industrial, economic or even sociological circumstances and to furnish data thereon to policy-makers? Or are our modelling efforts just the reaction to the desire of the public to find out more about the impact of technology on the environment? I believe that we have to distinguish between all these targets for modelling, and that models will change as their purposes change. I would therefore suggest that in the near future the Agency or another appropriate institution should organize a meeting on these basic problems of modelling.

P. PELLERIN: I am in full agreement with what has been said in reply to Mr. Maire so far. I should just like to stress the need for realism in our approach to these matters. Once again let us not forget, at the outset, that we are trying to avoid tiny fractions of rems being added to the (much greater) background and medical radiation dose. The resources required by this process of refinement could possibly be well spent elsewhere.

There are now two or three specific points I can add in reply to Mr. Maire. First, as regards the establishment of national or international data libraries, I think we should remember that this kind of facility already exists – on a large scale – in the field of radiation protection and atomic energy in general. I really doubt whether it is worth while adding yet another system of classification of the existing literature.

Secondly, I should like to say something about the proposals that are often made regarding the co-ordination and dissemination of data. Here I can quote our experience at WHO in our role of international reference centre for radioactivity measurements. For several years we have been in touch with ten or a dozen countries that are highly advanced in matters of nuclear research and radiation protection, and the first problem to emerge is that of calibration. There are amazing differences between the values arrived at by establishments whose competence is beyond question. So one has to be very careful not to collect measurement data for which no intercomparisons have been made – otherwise the effort will have been useless, or even worse than useless, since any interpretation one likes can be made of the results.

These problems of calibration are particularly difficult at the levels of refinement at which we all too frequently now have to operate. Here, I fully agree with Mr. Jammet that such work can only be entrusted to teams working in permanent liaison at national or international level.

I would sound a note of warning against the suggestion to issue hard-and-fast directives to local – or relatively local! – authorities responsible for radiation protection matters. The work that these authorities are called upon to accomplish demands great initiative combined with sound common sense and an intimate knowledge of local conditions, and if we go too far in terms of doing their work for them, we shall run the risk of turning them into unthinking robots completely devoid of a sense of responsibility. We already do enough in this direction in the form of advice – incorporated in a vast number of publications issued by all kinds of national or international organizations. This advice in fact itself constitutes a mass of directives. In this connection I find Mr. Schikarski's concluding proposal most welcome – namely a proposal to hold a scientific meeting based on a well-defined research program without laying undue stress on the fact that it is a protection program. That is the way we should try to go.

Question No. 5, by H.F. MacDonald

> For purposes of dealing with a single nuclide originating from a clearly identifiable source, the 'critical nuclide' approach has proved valuable in the past.
> Does the Panel consider that this situation will persist in the future, or must we begin to develop more sophisticated models capable of describing the behaviour of mixtures of isotopes discharged from a range of nuclear installations located within a given region, and irradiating man via several pathways?

B. LINDELL: I myself do not believe that this situation will persist, i.e. we shall not be able to go on basing all assessments on the critical group concept only. I think that proper control – and I am henceforth using the word 'control' in its full sense of exercising authority – will necessitate more than a critical group approach. The only practical way will be to control each of the many sources that will represent the total in the future. As an example of a workable method I have discussed the concept of population dose commitment per megawatt · year of electric energy. This method should make it possible to allow for the expansion of power plant capacities in the future, which was one of the points raised in the question to the Panel by the USSR participants.

I also believe that in the future local monitoring programs to check maximum individual doses will still be necessary, but that they will be of less significance than radioecological research on transfer mechanisms, which will be essential for purposes of assessing the total consequences of a release. Here perhaps I may give an anticipatory reply to a question we shall be hearing later regarding the appropriateness or otherwise of some of the programs that are carried out. To be blunt, I think that many of these programs are not justified because they are trying to measure extremely small individual dose contributions, and frequently they have no clearly defined objectives, which is an even more serious matter. So I would say that one can get more for one's money by relying on environmental radioecological research rather than on monitoring programs with rather diffuse objectives.

W.O. SCHIKARSKI: If you are going to institute a monitoring program at all, models can be most useful in deciding what should be monitored and where and when it should be done. I have a feeling that a good deal of monitoring is done in a perfunctory way, merely for the purpose of collecting data – which is then filed and forgotten. The combination of modelling and monitoring, on the other hand, ensuring that the effort is concentrated on the proper areas, can yield valuable results.

Question No. 6, by I. L. Brisbin, Jr. and F. O. Hoffman

> As scientists, we realise the utilitarian and pragmatic advantages of using the term 'environmental capacity' with reference to radionuclide contamination processes. We feel, however, that the manner in which this term has been used at the Symposium may convey an erroneous impression to the public. We refer specifically to the word 'capacity', which implies that the environment exists as a large empty vessel into which man may introduce his wastes, up to the limits of that vessel to contain them. Could the Panel, therefore, consider the use of some alternative term, such as 'environmental commitment'?
>
> We are particularly disturbed by the use of the term 'environmental capacity' in many cases to refer strictly to factors that can be shown to be either directly or indirectly impingent upon human populations. We feel that the possibilities of impacts upon other components of the biosphere – no matter how insignificant they may be or how apparently far-removed from man and his food chains – should not be ignored, as the use of the term 'environmental capacity' seems to imply.

F. MORLEY: While sharing the reservations of Messrs. Brisbin and Hoffman regarding the term 'environmental capacity', I cannot suggest an alternative. They themselves propose 'environmental commitment', but I think that their own commentary, as contained in their first and third paragraphs, highlights part of the difficulty in using this term.

I told the Symposium yesterday that I have lived with an alternative called 'derived working limit of release rate' for many years; this caused no problems, but neither has it been particularly informative.

I have been using 'environmental capacity' for three or four years past in a qualitative way, to explain the principles of radioactive waste disposal. I think I began to be unhappy about it when I needed to write down definitions and when I heard people talking not about local capacities but about regional capacities and about world capacities.

With specific regard to Messrs. Brisbin and Hoffman's point that 'environmental capacity' relates only to factors that are either directly or indirectly impingent upon human populations, I must say that this situation does not worry me. I am convinced that man is the limiting factor in the assessment of radioactive waste disposal operations. We have had ten or twenty years of operational experience now, quite apart from all the laboratory studies that have been made, and while many people still express fears about damage to ecosystems even when man is protected, I have yet to see the evidence for this damage. I am satisfied, in short, that we are doing the right thing in treating man as the end point for the discharge dose calculation.

B. LINDELL: As a technical concept I believe 'environmental capacity' to be very useful, and it is well understood among those of us working in this field. As Mr. Morley indicated in his reference to an alternative that does exist, it is, in the ICRP sense, the derived working limit for input rate of activity to a given recipient. We can't call it anything as cumbersome as that, of course. On the other hand, 'environmental capacity' is overworked and leads to a lot of misunderstanding, particularly with the public. Finding a new term, therefore, is one of the tasks of a forthcoming IAEA Panel, of which I am the Chairman. I accordingly should like to invite anyone who may have a proposal on the subject to mail it to me at the address given in the list of Symposium participants.

K.E. COWSER: I believe that the authors of this question have expressed concern about the impact of factors other than radioactive contaminants on ecosystems. This is something on which, in a general way, I commented when I made reference just now to needs related to thermal plumes, to chemicals in the form of biocides, etc., and perhaps even to synergistic effects produced by combinations of thermal plumes, chemicals and radioactive materials.

Question No. 7, by P. Kirchmann

> What does the Panel consider to be the main requirements at present in respect of radioecological research, and what direction should be imparted to the latter?

Question No. 8, by E. de Rademaeker

> On several occasions during the Symposium it has been stressed that the environmental impacts of nuclear and non-nuclear industry, each with its particular volumes and types of waste, are interrelated. However, most scientific studies have related to the influence of radionuclides on the environment and in this way the nuclear industry has found itself in an isolated position vis-à-vis the conventional energy industry and the chemical industry.
>
> Could the Panel evolve a set of recommendations on:
>
> (1) The co-ordination of environmental studies for nuclear and for non-nuclear industry;
> (2) The short and long-term planning of such studies;
> (3) The methods to be applied, as a matter of priority, to free nuclear environmental problems from their 'isolated' position?

B. LINDELL: On all these points I don't think I can add anything to what I have just said: namely, that radioecological research programs will, in my opinion, gradually become more important than environmental studies of the old type.

W.O. SCHIKARSKI: At Karlsruhe we also deal to some extent with pollution from sources other than nuclear energy and we now believe that much of the work being done on environmental research and analysis in connection with nuclear energy can be applied to other branches of industry. Indeed, the synergism of nuclear and non-nuclear pollutants, to which Mr. Cowser referred a few moments ago, constitutes an important item that has barely been touched upon at the Symposium but which should be exercising our attention more and more in the future.

H. JAMMET (Chairman): I can quote a good example from French experience of the extension to general pollution problems of the experience gained in the nuclear field. The Commissariat à l'Energie Atomique has been requested to study problems arising out of non-nuclear activities such as chemical manufacturing in the area of the Lower Seine between Paris and the Channel and in the Marseilles area in connection with the new industrial and harbour complex of Fos.

Question No. 9, by R. Amavis

> Can the Panel offer any observations on the methods to be used for estimating the population dose commitment when the critical transfer pathway is constituted by foodstuffs enjoying very wide distribution, often transcending national borders and liable to vary in volume with time?

H. JAMMET (Chairman): I imagine that Mr. Amavis is particularly interested in this matter since he is a member of the European Communities Secretariat. Well, I have pleasure in informing him that the European Communities have been considering the point he has raised for more than ten years now. There is at work a team comprising researchers from the six original members of the Community – who will doubtless now be joined by colleagues from the new member countries – and this team has carried out what may rightly be termed epoch-making studies on food consumption habits and food pollution. For the first time ever, precise investigations were conducted by having the researchers live for days with the families whose food and cooking patterns they were studying, and the result was the emergence of the most copious and accurate data regarding the extreme ranges of consumption within the Community countries, and hence regarding the commodities critical from a contamination point of view. For the original Community there is now available information on the distribution circuits of cereals, meat, milk, vegetables and other foodstuffs. This means that we are in a position to study the dose commitment for a grouping of nearly 200 million people – a figure which has risen to 250 million with the expansion of the Community.

The scale of all this reflects what Mr. Lindell has said about the need for very broadly based studies embracing the problem on at least a regional scale, to the exclusion of programs concerned with just one problem, just one facility, or just one critical group.

B. LINDELL: Replying more specifically to Mr. Amavis' question, I think there are two ways of going about it. One way, of course, is to follow all the food chains, to try to establish what the total consumption of food is, and then add up and integrate. The other possibility, which was broached by Mr. Barry yesterday, is to proceed in a much simpler way and, in assessing a population dose per unit practice, to aim at finding out only the amount of contaminated food and the average contamination of that food; it then does not matter where it goes and who eats it, because per kilogram of food there will be a certain dose commitment, and hence the dose commitment in the total amount of food can be quite easily and cheaply calculated in a first approximation.

W.O. SCHIKARSKI: I believe the European Communities have in fact set up a number of committees to deal with such problems as atmospheric pollution and so on. However, it seems to me that the questions which can best be handled by this means are basic issues such as optimum monitoring and modelling methods, rather than specific questions at regional level. The investigation of regional problems is clearly dependent on the specific location; for example, and analysis of the situation in the Upper Rhine valley will yield vastly different results from one conducted in the valley of the Seine.

P. PELLERIN: The international reference centre operated by the World Health Organization has undertaken the study of food contamination at international level. Work is well under way as regards the investigation of milk in various parts of the world, and attention is now being turned to the components of the diet as a whole. As can be imagined, the degree of variation between one country and another is enormous, and adds to the complexity of the problem.

H. JAMMET (Chairman): With reference to what Mr. Schikarski has just said, I should like to make it clear that the European team to which I referred is not engaged on the study of problems peculiar to a given facility, but devotes its efforts to the collection of parameter values and to general methodological research.

Question No.10, by M.J.A. Delpla

At Session VI on Thursday morning, four of our United States colleagues gave a detailed account of their methods of evaluating irradiation dose to the population due to nuclear industry. One of them justified the considerable effort involved on grounds of the need to reassure the public.

Does the Panel consider that the substantial sums of money devoted to this purpose are well spent in terms of public health benefit?

Question No.11, by G. Cantillon

The public health authorities everywhere have to face an organized nuclear opposition movement whose motives are not always fully clear. Could not the World Health Organization, as a universal authority in matters of the health of mankind, undertake the preparation of a publication in which the arguments of the 'opposition' would be analysed and replied to in an appropriate manner?

Question No.12, by C.A. Mawson

Many of our public relations difficulties are due to our own approach to safety calculations. We make 'conservative' assumptions which, we proudly say, are extremely pessimistic. We know they are not realistic – each has a large factor of safety added to it. When we multiply them together the result yields an astronomical factor of safety to which the public, and often the regulatory bodies, then apply a further factor of safety. If you put stupid numbers into a computer it will deliver a stupid result.

Should we not put realistic numbers into our calculations, make realistic assumptions regarding what we really believe will happen, and then at the end of the process apply a known and clearly stated safety factor? I consider this to be the only way to avoid fantastically pessimistic assumptions which lead to public panic and waste of public money on unjustified caution.

H. JAMMET (Chairman): We have decided to consider these questions last because we believe they make the widest general appeal.

P. PELLERIN: I will try to reply to each of them in turn.

First, there is scarcely any need for me to say once again how entirely I agree with Mr. Delpla that we sould not spend on radiation protection, in particular radiation protection around nuclear power stations, more than we spend, as a maximum, on measures to protect public health against roughly equivalent non-radioactive hazards (and I fear, incidentally, that that maximum is in fact not always spent).

As regards Mr. Cantillon's suggestion, I am extremely pleased to hear proposals along these lines because I have on a number of occasions stressed the need for an international organization right outside the field of nuclear energy to publish its views on this subject. Clearly WHO is eminently suited to carry out this function. I have no doubt that Mr. Cantillon's suggestion will evoke lively interest at WHO Headquarters. WHO is all the more sympathetic to the rapid development of nuclear power in that energy from the atom is essentially kind to the environment. This is now beyond dispute – except of course in the eyes of those who dispute everything.

Finally, I entirely share Mr. Mawson's belief that, if you artificially stack up all the unfavourable factors and perform your calculations on that basis, then you end up in a situation which is virtually ridiculous. I am glad to see that at this Symposium – unlike at some radiation protection meetings – there has been a commendable absence of mathematical over-refinement and of questionable pursuit of minute fractions of millirems. Rather we have been shown here, particularly by Mr. Barry as I recall, that simple data applied with common sense and in the light of a wealth of experience can give results that are often much more reliable than those obtained with highly sophisticated calculations. As Mr. Mawson points out, a computer is a stupid animal, and if the input is stupid, then so will be the output. However, I think it reasonable to assume that most of the radiation protection experts present here are much more intelligent than the most perfect of computers!

K.E. COWSER: May I say a word in defence of models. In my opinion they have a real value, especially when one is concerned with complicated regional systems or the handling of large amounts of data. As Mr. Mawson points out, we are not always sure that we have the correct input information. However, in such cases a sensitivity analysis can be made that is informative as to which parameters are the most important.

From a financial viewpoint I feel that the judicious use of modelling to determine the need for control systems prior to operation would be less expensive than adding the control systems at a later date.

H. JAMMET (Chairman): I am extremely grateful to Mr. Mawson for having raised again the question of safety factors. When the ICRP established irradiation dose limits, it defined these limits as those that represented an acceptable risk. The acceptable risk concept is based on pessimistic assumptions, some of which have never been proved, as for example the absence of a threshold and the linearity of dose and effect. These are working hypotheses and not proven facts. ICRP proceeded quite correctly in defining the limits in this way, because maximum safety is thereby guaranteed. But, if in every case of transport, dispersal or reconcentration you base your calculations on the most exaggeratedly pessimistic figures, you achieve a degree of caution regarding establishment of a nuclear facility in a given area that is positively grotesque. Applying the same radiation hazard criteria to everyday life, one would always – just out of fear of the minute additional cosmic radiation – inhabit the ground floor of a building and in no circumstances live high up in a skyscraper! So I associate myself unreservedly with Mr. Pellerin in saying that by all means let us be cautious – we always have been cautious in fact – but let us not be ridiculous.

CLOSING OF THE SYMPOSIUM

H. JAMMET (Chairman of concluding session): It now remains for me to thank the International Atomic Energy Agency, the Nuclear Energy Agency of the OECD and the World Health Organization for having convened the Symposium and given us the opportunity for such a valuable exchange of views this week.

D.G. JACOBS (Scientific Secretary): On behalf of the three sponsoring Organizations, I should like to thank the French Government, and especially the Commissariat à l'Energie Atomique, for the invitation to hold the Symposium here at Aix-en-Provence, and for the kindness and hospitality which have been accorded us. Our particular appreciation is due to the staffs of the Fontenay-aux-Roses and Cadarache Nuclear Research Centres for their valuable efforts to make the meeting a success.

H. JAMMET: I now declare the Symposium closed.

CHAIRMEN OF SESSIONS

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