

Radioactivity in the Mediterranean flora of the Kaštela Bay, Croatia



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ABSTRACT

This study refers to background activity concentrations of ^{238}U , ^{226}Ra , ^{232}Th , ^{208}Tl , ^{40}K , and ^{137}Cs in soil and plants of the Kaštela Bay, Croatia and related plant-soil concentration ratios (CR's). Fourteen different Mediterranean plant species growing in natural conditions have been included and were divided into three major plant groups (grasses and herbs, shrub, tree). Radionuclide activity concentrations were determined by means of high resolution gamma-ray spectrometry. Soil parameters (pH, electrical conductivity, and organic matter content) were also analysed. CR ranges were within one order of magnitude for ^{40}K (10^{-2} – 10^{-1}), ^{238}U , and ^{226}Ra (10^{-3} – 10^{-2}), and two orders of magnitude for ^{232}Th , ^{208}Tl , and ^{137}Cs (10^{-4} – 10^{-2}). There was no statistical difference between the plant groups in radionuclide uptake. Overall statistical analyses indicated a moderate negative relationship between soil concentrations and CR values, and no relationship with soil parameters, except a negative one for ^{137}Cs . Comparison with literature showed more agreement with studies that were done in the Mediterranean than with ICRP and IAEA databases. Our data not only describe the natural radioactivity of the Bay, but also create a dataset that could be relevant for further radioecological assessments of the Kaštela Bay.

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1. Introduction

Due to the complexity of natural processes, it is difficult to predict the behaviour of a particular radionuclide within a given ecosystem and/or organism. For many radioactive elements, concentration ratios (CR's) are available for only about 10% of different plant/soil combinations and generally exhibit large variability in radionuclide transfer even within plants of same taxonomic rank (Hinton et al., 2013; Howard et al., 2013a; Vandenhove et al., 2009). Furthermore, naturally occurring radionuclides have received less research attention compared to artificially produced radionuclides that have been the focus since the first atmospheric nuclear weapon tests (Mitchell et al., 2013; Shtangeeva, 2010; Vandenhove et al., 2009). The majority of current knowledge on behaviour and transfer of natural radionuclides have been acquired through studies on polluted sites (Chen et al., 2005; Vera Tomé et al., 2002) and areas with elevated natural background radiation (Termizi Ramli et al., 2005) or in studies with "spiked" soils (Vandenhove et al., 2007; Vandenhove and Van Hees, 2007). Fewer data are available from unpolluted sites (Beresford et al., 2008; Shtangeeva, 2010).

In this study, we focus on the background activity concentrations of radionuclides in the soil of Kaštela Bay, Croatia, and their transfer into the local Mediterranean flora. The bay is located on the eastern coast of the middle Adriatic Sea. In the second half of the 20th century, it was exposed to different types of pollution, including radioactivity originating from coal used to power a nearby factory (no longer in operation). This coal comprised naturally occurring radionuclides (mainly from uranium and thorium decay chains), whereas its combustion products, i.e. ash and slag of elevated radioactivity, were disposed locally. Over the years, the disposal site for the ash and slag has been colonised by different Mediterranean plants. To put the transfer of radionuclides from the coal ash and slag to vegetation into context, we needed data on the radioactivity and transfer of radionuclides from soil to plant in natural background conditions of the Bay.

For Mediterranean ecosystems, studies on the distribution of radionuclides and their transfer from soil to plant have been mostly performed around disused uranium mines or nuclear power plants (Baeza et al., 2001; Rodriguez et al., 2010; Vera Tome et al., 2002, 2003). Therefore, we were motivated to carry out our study to establish a dataset on background soil and plant activity concentrations and related CR values for Kaštela Bay against which data from contaminated site can be compared. Howard et al. (2013b) identified which ICRP reference plant–radionuclide combinations can be designated as high priority for future research needs. These

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are Pine Tree/ ^{238}U , and Wild Grass/ ^{226}Ra , ^{232}Th , ^{238}U , which overlap with the radionuclides and plants in our study. These authors also suggested that variation in soil-to-plant radionuclide transfer can be caused by different soil types, and therefore site-specific assessment might be justified in some cases. Since, the Mediterranean region is more diverse in soil types than any other climatic region (Verheye and de la Rosa, 2009), site-specific assessment based on field-experiment results, not extrapolated CR values, might also be justified for this Bay.

Our study focused on natural radionuclides ^{238}U , ^{226}Ra , ^{232}Th , ^{208}Tl , ^{40}K , and an anthropogenic radionuclide with high variability in radionuclide transfer from soil to plants, ^{137}Cs (IAEA, 2009). It includes a total of 14 different Mediterranean plant species growing in natural background conditions. The aim was to collect data on activity concentrations in soil and plants and calculate the resultant transfer of radionuclides between them. The purpose of these data was not only to describe natural radioactivity of the Bay, but also to create a dataset that could be important for further radioecological assessments of Kaštela Bay.

2. Description of the area

Kaštela Bay (Fig. 1) is a synclinal fold transgraded in the Post-pleistocene stage, made of Eocene flysch and Eocene marly limestone (Hidroelektra-projekt, 2004). It is a semi-enclosed bay in the middle Adriatic Sea (Ujević et al., 2000).

The climate in the Bay is typically Mediterranean. The average annual temperature is $15.9\text{ }^{\circ}\text{C}$, with a minimum monthly average of $8.0\text{ }^{\circ}\text{C}$ in January, and a maximum of $25.9\text{ }^{\circ}\text{C}$ in July. The average annual rainfall is 820.6 mm , and the average relative humidity is 66% . The area is rather windy, with 75% of winds being stronger than $12\text{--}19\text{ kmh}^{-1}$ (Hidroelektra-projekt, 2004).

The Kaštela coast and the northern slopes of the nearby island of Čiovo (see Fig. 1) are covered with maquis of *Orno-Quercetum ilicis* and *Cymbopogoni-Brachypodion* grasslands in karst, and *Clematido-Spartietum* heath and *Vulpio-Lotion* meadows in flysch. The southern slopes of Čiovo island, which are the warmest and driest, are dominated by maquis of *Myrto-Pistacietum lentisci*. The coastal halophytic vegetation in the Pantan lagoon includes *Juncetum maritimi-acuti* and *Bolboschoeno-Scirpetum litoralis*. Elsewhere in the lagoon rocky shores, *Plantagini-Limonietum cancellati* occurs, whereas *Euphorbio-Glaucietum flavi* is found in the beach coves. Since autumn and winter are the rainy seasons in the area, vegetation growth in the Bay is favoured within these periods (Lovrić and Oleg, 1999).

The island of Čiovo is a part of the Bay and has the same geology and vegetation as described above. But, unlike other parts of the

Bay, it was not affected heavily by previous industry activities and urbanisation. Also, it has many areas that have not been affected by agriculture at all or have not been used for that purpose in decades. Therefore, we considered it as suitable area for collecting data on background radioactivity. An additional sampling point Pantan lagoon, which is a natural reservation, was added as representative of halophytic vegetation.

The impact of previous industrial activities and waste disposal site of coal and ash on sampling locations can be neglected since they are at a sufficient distance (Dai et al., 2007; Flues et al., 2002). Čiovo is approximately at a 8.5 km aerial distance from the waste disposal site, and Pantan lagoon at 11 km .

3. Materials and methods

3.1. Sampling

Sampling of soil and plants was carried out on six locations (see Fig. 1) in March 2011, prior to Fukushima accident releases. Five sampling locations were located on the island of Čiovo, and one in the Pantan lagoon. Sampling was performed in zones which have neither been urbanised nor affected by industry. Specific sampling locations were determined based on the presence of different plant species and site accessibility (e.g., southern slopes of the Čiovo island are very steep and unapproachable).

We collected 6 soil samples and 16 samples of 14 different plant species. While most of the plant samples represent typical terrestrial plants, two (Spiny Rush and Common Reed), collected at the Pantan lagoon, belong to halophytic vegetation. All these plant species can be considered common for the Mediterranean.

The type of terrain determined our sampling method (rocky area with dry soil) and tools used (shovel instead of corer). Soil samples were taken in squares $15\text{ cm} \times 15\text{ cm}$, taking approximately the first 10 cm soil layer. Each soil sample for single sampling location represented a composite material taken from a few points over approximately 1 m^2 , with a total sample mass of $3\text{--}4\text{ kg}$ (fresh mass). Roots and bigger stones were removed immediately.

A minimum of 1 kg (fresh mass) of each plant species was collected within a 10 m radius of the area where the soil was

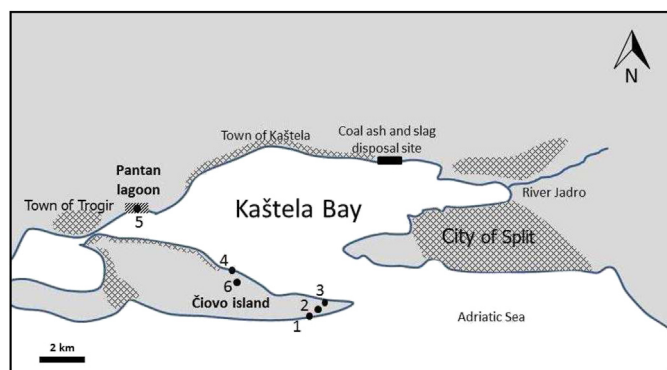


Fig. 1. Kaštela Bay with sampling locations [Čiovo island (1,2,3,4,6) and Pantan lagoon (5)] and marked major urbanised areas.

Table 1

List of plant species collected on island Čiovo and Pantan lagoon. Plant samples are divided into three major groups according to their association with reference plant groups used by Brown et al. (2008).

Latin name	Plant group	Common name	Sampling location
<i>Helichrysum italicum</i> (Roth)	Grasses & herbs	Curry plant	1
<i>G. Don</i>			
<i>Piptatherum miliaceum</i> (L.)	Grasses & herbs	Smilo grass	2
<i>Coss.</i>			
<i>Dittrichia viscosa</i> (L.) Greuter	Grasses & herbs	Sticky fleabane	3
<i>Phragmites australis</i> (Cav.)	Grasses & herbs	Common reed	5
<i>Trin. eX Steud.</i>			
<i>Juncus acutus</i> L.	Grasses & herbs	Spiny rush	5
<i>Pistacia lentiscus</i> L.	Shrub	Mastic	1, 6
<i>Spartium junceum</i> L.	Shrub	Spanish broom	2, 6
<i>Rubus heteromorphus</i>	Shrub	Blackberry	3
<i>Ripart eX Genev.</i>			
<i>Pittosporum tobira</i>	Shrub	Japanese Pittosporum	4
<i>(Thunb.)Aiton</i>			
<i>Nerium oleander</i> L.	Shrub	Oleander	4
<i>Ficus carica</i> L.	Tree	Fig	3
<i>Pinus halepensis</i> Mill.	Tree	Pine tree	1
<i>Cupressus sempervirens</i> L.	Tree	Mediterranean cypress	2
<i>Tamarix dalmanica</i> Baum	Tree	Tamarisk	3

Table 2
Results of analysis of some soil properties: pH, weight loss by ignition (LOI) and electrical conductivity (EC).

Parameter	Sampling location					
	1	2	3	4	5	6
pH	9.00	8.47	7.98	8.73	7.74	9.11
LOI%	7.66	16.47	12.45	21.33	11.49	8.96
EC (μScm^{-1})	223	690	685	654	2100	258

sampled. Plants were cut a few centimetres above ground by hand or using metal scissors. Plant sample included whole aboveground plant parts (leaves, branches, trunk). The exception was tamarisk, cypress, and ficus, whose trunk was too thick to cut with scissors or by hand. Therefore, for those species only branches and leaves were sampled. We carefully separated plant species from one another.

3.2. Preparation of samples

Soils were dried at 105 °C to a constant mass, sieved through a 2 mm pore sieve, ashed in a muffle furnace at 450 °C for 24 h, and finally put into Marinelli beakers (1 L volume) to use the maximum amount of sample in order to enhance measurement precision. From each soil sample, 200 g were taken for pH, organic matter content loss by ignition (LOI) and electrical conductivity (EC) analysis.

Plant samples were dried at 105 °C to a constant mass and ashed at 450 °C in a muffle furnace for 24 h. The resulting ash was mechanically grinded and thoroughly mixed in mortar and then put into cylindrical plastic containers of appropriate volumes (50 mL, 100 mL, 200 mL). Both soil and plant samples were left to rest for a minimum 30 days in order to reach secular equilibrium within the thorium and uranium decay chains.

Table 3
Activity concentrations (Bqkg^{-1}) of selected radionuclides in the soil of the Kaštela Bay. For comparison, some literature data are given in the lower part of the table.

Location	^{238}U	^{226}Ra	^{232}Th	^{208}Tl	^{40}K	^{137}Cs
1	24	33.2	26.2	8.9	332	15.4
2	40	37.5	30.3	11.2	269	99.8
3	92	120	73	29.2	526	16.4
4	46	56.6	45.1	14.7	469	77.1
5	51	36.1	39	14.2	561	12.6
6	69	64.2	69	22.8	503	42.3
GM \pm GSD	49 \pm 2	52 \pm 2	44 \pm 1	15 \pm 1	429 \pm 1	32 \pm 2
AM \pm SD	53.5 \pm 23.8	57.9 \pm 32.8	47.1 \pm 19.7	16.8 \pm 7.7	443.5 \pm 116.2	43.9 \pm 36.8
RANGE	24–92	33.2–120	26.2–73	8.9–29.2	269–561	12.6 \pm 99.8
Literature data						
Mediterranean soils ^a						
GM \pm GSD	30.1 \pm 16.4	n/a	55.5 \pm 6.6	n/a	147.3 \pm 76.0	n/a
AM \pm SD	31.6 \pm 9.1	n/a	60.9 \pm 23.3	n/a	207.2 \pm 149.1	23.00 \pm 7.07 ^b
RANGE	10.5–53.2	n/a	10.9–118.9	n/a	37.1–402.0	0–85
Croatia ^c						
GM \pm GSD	105 \pm 1	72 \pm 1	61 \pm 1	n/a	643 \pm 1	35 \pm 2
AM \pm SD	107 \pm 25.7	73.3 \pm 15.8	61.8 \pm 13.4	n/a	645.0 \pm 61.4	38.8 \pm 18.8
RANGE	78–140	51–86	45–77	n/a	570–720	23–62
South Europe ^d						
AM \pm SD	51.75 \pm 40.59	35.50 \pm 13.49	34.83 \pm 11.63	n/a	432.86 \pm 212.42	n/a
RANGE	1–240	0–250	1–190	n/a	0–1650	n/a
Greece ^e						
AM \pm SD	n/a	25 \pm 19	21 \pm 16	n/a	335 \pm 220	n/a
RANGE	n/a	1–238	1–193	n/a	12–1570	n/a

n/a = no available data.

Relative errors (ratio between absolute error and measured value) for soil activity concentrations in our study (not shown) are below 10%.

^a Laubenstein and Magaldi (2008).

^b Charro et al. (2013). Data derived from measurements of ^{137}Cs activity concentration in 0–5 cm and 5–10 cm soil layers.

^c Cesar et al. (1994).

^d UNSCEAR (2000). AM \pm SD calculated from mean values reported for Albania, Croatia, Cyprus, Greece, Portugal, Slovenia and Spain (Mediterranean countries). Range is based on reported individual measurement data.

^e Anagnostakis et al. (1996).

3.3. Gamma-ray spectrometry

Samples were analysed by means of gamma-ray spectrometry. We used three ORTEC photon detector systems: Ge(Li) (relative efficiency of 15.4%, peak full width at half maximum (FWHM) of 1.87 keV), HPGe GEM (relative efficiency of 21%, FWHM of 1.75 keV), and HPGe GMX (relative efficiency of 74.3%, FWHM of 2.24 keV). The efficiency and FWHM data refer to ^{60}Co at 1.33 MeV.

Data on ^{40}K and ^{137}Cs activities were obtained from photopeaks at 1460 keV and 661 keV, respectively. Activities of ^{238}U , ^{226}Ra , and ^{232}Th were determined from those of their decay products, assuming that secular equilibrium had been established. Activity of ^{226}Ra was determined from that of ^{214}Bi (photopeaks at 609 keV, 1120 keV, and 1764 keV), activity of ^{232}Th from that of ^{228}Ac (photopeaks at 338 keV, 911 keV, and 968 keV), and activity of ^{238}U from those of ^{234}Th (photopeak at 63 keV) and $^{234\text{m}}\text{Pa}$ (photopeak at 1001 keV). ^{208}Tl activity was obtained from photopeak at 583 keV.

Calibration of the measurement setup was carried out using standards prepared by the Czech Metrology Institute covering an energy range between 80 and 2500 keV. Quality assurance was performed via regular participations in interlaboratory comparisons organised by International Atomic Energy Agency (IAEA), World Health Organisation (WHO), and Joint Research Centre (JRC) (Petřinec et al., 2011).

The measurement time depended on sample activity and type of detector used, ranging from 70,000 s to 250,000 s. Soil samples were tested for attenuation of gamma rays and results were corrected accordingly.

3.4. Soil pH, LOI and EC analysis

Soil leachate was prepared by an accredited method (HRN EN 12457-4:2005), in which pH (HRN EN ISO 10523:2012) and

electrical conductivity were measured (HRN EN 27888:2008) by a Mettler Toledo MCP 227 pH/Conductivity meter. Organic matter content was determined by loss of ignition of dry mass on 550 °C (HRN EN 12879:2000, muffle furnace CEM Phoenix).

3.5. Data analysis

Plant species collected in this study can be associated with reference plant groups used in wildlife dose assessment (Brown et al., 2008) (Table 1). Although the new approach in categorising plant species has been developed recently with even more plant subcategories (Howard et al., 2013a, b), due to a limited number of data (n = 1 for 75% plant species covered by this study), additional partitioning of our results probably would not be beneficial.

Activity concentrations in literature are sometimes expressed per dry mass (DM) (Baeza et al., 2001; IAEA, 2010; Rodriguez et al., 2010; Vandehove et al., 2009; Vera Tome et al., 2003) and sometimes per fresh mass (FM) (Beresford et al., 2008; IAEA, 2011; ICRP, 2011). We consistently expressed activity concentrations in soil per DM, and in plants per FM, which is consistent with the international approach for calculating soil-to-plant radionuclide CR values (Howard et al., 2013a). In comparing our data with those from the literature, when necessary, we expressed the latter ones, in the above mentioned units by using DM-to-FM conversion factors for plants as recommended by IAEA (2010).

Concentration ratio (CR_{wo-media}) of a radionuclide from soil (media) to plant (whole organism –wo) was calculated as (Howard et al., 2013a, b).

$$CR_{\text{plant-soil}} = \frac{\text{Activity concentration in plant (Bqkg}^{-1} \text{ fresh mass)}}{\text{Activity concentration in soil (Bqkg}^{-1} \text{ dry mass)}} \quad (1)$$

In this paper, we included values that represent the limit of detection (LOD) for some plant activity concentrations. Although there are certain controversies over using LOD values in datasets, there are techniques for manipulating this sort of data widely used in other fields of environmental sciences (e.g. Kaplan–Meier method). An example of using this approach in CR analysis can be found in Wood et al. (2010). Therefore, we found it beneficial to present our LOD values. Due to pragmatism and portion of LOD values (less than 20%, except for ²⁰⁸Tl which is 25%), we chose conservative approach and used LOD values as absolute values in CR calculations (Wood et al., 2008).

Statistical analysis of data was performed using statistical software STATISTICA Version 12. Normality of data was evaluated with Shapiro–Wilk’s test. Soil activity concentrations showed tendency towards normal distribution, although small sample numbers (n = 6) give uncertainty in statistical analysis. Plant activity concentrations and CR values were normally distributed after log–transformation of data as was expected (Vandehove et al., 2009; Vera Tome et al., 2003; Wood et al., 2013). For each dataset (soil activity concentrations, plant activity concentrations, CR values) following summary were calculated: geometric mean (GM), arithmetic mean (AM), related standard deviations (GSD, SD), and range, as described by the IAEA (2009).

Welch’s test was used to test the null hypothesis that there was no statistically significant difference between our results and literature data, considering a 95% confidence interval. If literature sources provided only means and number of samples without standard deviations, Student’s unpaired t-test was applied, assuming that literature data has the same variance and standard deviation as ours. To test significant differences between means of three or more groups, we used one-way Analysis of Variance

Table 4 Activity concentrations (Bqkg⁻¹ FM) in plant samples. Summary statistic is presented for overall results.

Plant species	Plant group	²³⁸ U	²²⁶ Ra	²³² Th	²⁰⁸ Tl	⁴⁰ K	¹³⁷ Cs
Sticky fleabane	Grasses & herbs	4.89E-01	4.10E-01	1.64E-01	<2.80E-02	4.75E+01	2.50E-02
Spiny rush	Grasses & herbs	7.10E-01	8.00E-01	1.39E+00	1.33E-01	1.38E+02	5.80E-02
Curry plant	Grasses & herbs	5.09E-01	5.40E-01	8.40E-01	9.40E-02	6.86E+01	2.60E-01
Smilo grass	Grasses & herbs	2.15E+00	1.20E-01	4.70E-01	4.66E-01	1.05E+02	1.10E-01
Common reed	Grasses & herbs	2.25E+00	4.90E-01	2.06E+00	7.30E-02	6.20E+01	<4.80E-02
Spanish broom	Shrub	3.40E-01	1.90E-01	1.57E-01	1.20E-02	9.29E+01	2.70E-02
Spanish broom*	Shrub	<1.88E-01	7.80E-02	<4.90E-02	<1.00E-02	4.78E+01	3.90E-02
Blackberry	Shrub	<4.12E-01	3.30E-01	1.47E-01	<2.50E-02	8.39E+01	1.10E-02
Oleander	Shrub	9.20E-01	1.51E+00	8.50E-01	1.30E-01	8.42E+01	1.20E-01
Pitosporum	Shrub	8.30E-01	3.80E+00	2.60E+00	2.91E-01	2.57E+02	4.90E-02
Mastic	Shrub	5.60E-01	4.00E-01	4.40E-01	3.70E-02	1.57E+02	4.90E-02
Mastic*	Shrub	<1.86E-01	1.70E-01	2.28E-01	<1.20E-02	9.51E+01	6.30E-02
Pine tree	Tree	1.26E-01	2.40E-01	1.96E-01	3.10E-02	6.21E+01	5.60E-02
Cypress	Tree	1.32E-01	2.00E-01	1.40E-01	3.40E-02	8.72E+01	5.90E-02
Fig	Tree	6.40E-01	1.90E-01	4.59E-01	5.50E-02	2.00E+02	1.82E-01
Tamarisk	Tree	4.48E-01	1.60E-01	1.47E-01	1.05E-01	8.14E+01	4.00E-02
Summary statistics							
GM ± GSD		4.82E-01 ± 2.29E+00	3.38E-01 ± 2.60E+00	3.62E-01 ± 2.95E+00	5.30E-02 ± 2.99E+00	9.30E+01 ± 1.59E+00	5.60E-02 ± 2.10E+00
AM ± SD		6.81E-01 ± 6.39E-01	6.02E-01 ± 9.23E-01	6.46E-01 ± 7.54E-01	9.60E-02 ± 1.22E-01	1.04E+02 ± 5.74E+01	7.50E-02 ± 6.50E-02
RANGE		1.30E-01 – 2.25E+00	8.00E-02 – 3.80E+00	5.00E-02 – 2.60E+00	1.00E-02 – 4.70E-01	4.70E+01 – 2.57E+02	1.00E-02 – 1.80E-01

Relative errors (not shown) for ⁴⁰K and ²²⁶Ra are below 10%. For ¹³⁷Cs, ²⁰⁸Tl, ²³²Th, ²³⁸U they are somewhat larger, but still below 20%.

<Below limit of detection.

*Duplicate samples from sampling location no. 6.

(ANOVA). The difference was considered to be statistically significant when $p < 0.05$. Since these statistical tests assume normal distribution of data, logarithmic transformation of the data was performed when necessary (Wood et al., 2013). Since all our data do not have same distribution and sample size is small, as a measure of statistical dependence between two variables, nonparametric Spearman's rank correlation coefficient was calculated (ρ). Nevertheless, for the sake of comparing our results with other authors, we also performed single parameter regression analysis to test the correlation between variables. Correlations were significant only for log-transformed data since CR values were not normally distributed. Statistical dependence and correlations were marked as significant at $p < 0.05$.

4. Results and discussion

4.1. Soil parameters and activity concentrations

In Table 2, we present the results of ^{238}U , ^{226}Ra , ^{232}Th , ^{208}Tl , ^{40}K , ^{137}Cs in soil samples collected in Kaštela Bay and literature data for comparison. In Table 3, results of soil pH, LOI and EC are presented.

Comparison of our results with data from other countries that belong to the Mediterranean region of Europe, shows some statistically significant differences for ^{40}K ($p < 0.009$), ^{238}U ($p < 0.01$) and ^{232}Th ($p < 0.02$). These differences reflect soil type variability within the Mediterranean and an influence of different geology on concentrations of natural radionuclides in soil (Verheye and de la Rosa, 2009). However, when we performed a comparison with more generic data for Southern Europe (UNSCEAR, 2000), we found no significant difference between data ($p > 0.05$). This suggests that our sampling sites exhibit no larger deviation than other parts of the Mediterranean region (no very low or very high values).

Soil pH measurements showed that all soil samples have a pH above 7, which reflects alkaline conditions. Higher pH favours the adsorption of cationic metals on soil particles (McLean and Bledsoe, 1992). Spearman's correlation test between pH and EC showed a strong negative correlation ($\rho = -0.8857$, $p < 0.05$) which confirmed reduced mobility of cations in soil. Highest EC was measured in a sample from Pantan lagoon. Since that sampling location is characterized by brackish water, the soil's high EC was probably due to the higher sodium chloride concentration. All soil samples showed LOI $< 20\%$, except one sampling location which exceeds very little over.

4.2. Activity concentrations in plant samples

In Table 4, we list results for activity concentrations ^{238}U , ^{226}Ra , ^{232}Th , ^{208}Tl , ^{40}K and ^{137}Cs in our plant samples and related

summarised statistics. Activity concentrations for ^{238}U , ^{208}Tl , ^{40}K and ^{137}Cs are within one order of magnitude and for ^{226}Ra and ^{232}Th within two orders of magnitude. Due to the low number of samples (1 sample for each plant species), we could not draw any conclusions on interspecies variability.

Table 5 shows a summary of activity concentrations measured in our group plants and a comparison with those found in the literature for same or similar plant groups. Statistical analysis showed no difference ($p > 0.05$) between our activity concentrations measured in grasses and herbs and those for pastures in Spain (Baeza et al., 2001) or those for Wild Grass in England and Wales (Beresford et al., 2008). Because of limited number of data in our study (6 soil samples, 14 plant species, 1 sample per plant species), the statistic results are somewhat uncertain.

4.3. Soil-to-plant CR

CR values that resulted from our study are presented in Table 6. CR ranges are within one order of magnitude for ^{40}K (10^{-2} – 10^{-1}), ^{238}U and ^{226}Ra (10^{-3} – 10^{-2}), and two orders of magnitude for ^{232}Th , ^{208}Tl , and ^{137}Cs (10^{-4} – 10^{-2}).

ANOVA analysis showed no significant difference between plant groups in their uptake of radionuclides ($p > 0.05$), although they comprised a range of 14 different plant species with different root lengths. Also, all soil samples were taken within first 10 cm of soil. Lack of significant differences between plant groups with different root length is in accordance with the fact that root density decreases with soil depth (Ehlken and Kirchner, 2002). Therefore, soil activities in first soil layers, with the highest root density probably have the highest impact on the uptake of radionuclides. Also, studies on vertical distribution of radionuclides show that concentrations of U, Th, Ra and K are constant with depth, while Cs is usually retained up to 86% in first 10 cm of soil (Arapis and Karadinis, 2004; Charro et al., 2013; Porto et al., 2001).

We found no statistical difference in the uptake of ^{238}U , ^{226}Ra , and ^{232}Th within same plant group. Similarities between ^{238}U and ^{232}Th plant uptake were expected considering that they are both actinides and therefore have similar chemistry. Unlike in our study, alkaline-earth ^{226}Ra was distinct in similar study in Mediterranean showing two orders of magnitude higher plant uptake (Vera Tome et al., 2003). The higher uptake of ^{40}K (one to three orders of magnitude with regard to other radionuclides, $p < 0.0001$) is no surprise considering its role in the physiology of plants.

Statistical dependence between CR values and soil activity concentrations (overall plant species), measured by Spearman's rank, was moderate and negative for almost all of the radionuclides: ^{238}U ($\rho = -0.51$, $p < 0.05$), ^{226}Ra ($\rho = -0.63$, $p < 0.01$), ^{232}Th ($\rho = -0.53$, $p < 0.05$), ^{40}K ($\rho = -0.53$, $p < 0.05$). ^{137}Cs showed also

Table 5
Comparison of activity concentrations (Bqkg⁻¹ FM) for plant groups from our study (GM \pm GSD, AM \pm GSD) with literature data for same or similar plant groups.

Literature source	Plant group	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Kaštela Bay	Grasses & herbs	9.69E-01 \pm 1.98E+00 1.22E+00 \pm 8.98E-01	4.01E-01 \pm 1.90E+00 4.72E-01 \pm 2.45E-01	7.14E-01 \pm 2.43E+00 9.85E-01 \pm 7.55E-01	7.82E+01 \pm 1.46E+00 8.42E+01 \pm 3.68E+01	7.24E-02 \pm 2.21E+00 1.00E-01 \pm 9.46E-02
	Shrubs	4.14E-01 \pm 1.82E+00 4.91E-01 \pm 2.94E-01	4.09E-01 \pm 3.47E+00 9.25E-01 \pm 1.36E+00	3.06E-01 \pm 3.34E+00 6.39E-01 \pm 9.06E-01	1.03E+02 \pm 1.64E+00 1.17E+02 \pm 6.98E+01	4.15E-02 \pm 1.99E+00 5.11E-02 \pm 3.47E-02
	Trees	2.63E-01 \pm 2.06E+00 3.37E-01 \pm 2.52E-01	1.95E-01 \pm 1.16E+00 1.98E-01 \pm 3.30E-02	2.07E-01 \pm 1.61E+00 2.36E-01 \pm 1.51E-01	9.69E+01 \pm 1.55E+00 1.08E+02 \pm 6.25E+01	7.00E-02 \pm 1.77E+00 8.43E-02 \pm 6.57E-02
Baeza et al. (2001) ^a	Pasture ^a	n/a	1.00E+00 \pm 6.00E-01	1.60E+00 \pm 8.00E-01	4.00E+01 \pm 5.00E+01	3.20E-01 \pm 2.80E-01
Beresford et al. (2008) ^b	Wild Grass ^b	3.60E-01 \pm 4.00E-01 <2.00E-02–2.30E+00	3.30E-01 \pm 2.90E-01 3.70E-02–1.70E+00	1.20E-01 \pm 1.70E-01 <5.50E-02–8.10E-01	2.70E+02 \pm 1.22E+02 4.06E+01–7.08E+02	n/a
	Pine Tree ^b	7.30E-03 \pm 7.40E-03 7.40E-04–2.60E-02	7.80E-02 \pm 8.50E-02 <4.70E-03–1.80E-01	5.20E-03 \pm 4.60E-03 7.79E+00–1.35E+02	3.52E+01 \pm 4.37E+01 7.79E+00–1.35E+02	n/a

n/a = no available data.

^a Annual mean values for aerial fraction of the plant from vicinity of nuclear power plant.

^b Values are estimated from a combination of measured and literature review values. Results are reported as mean \pm SD and range.

Table 6
CR values for different plant species from the Kaštela Bay. Summary statistic is presented for overall results.

Plant species	Plant group	²³⁸ U	²²⁶ Ra	²³² Th	²⁰⁸ Tl	⁴⁰ K	¹³⁷ Cs
Sticky fleabane	Grasses & herbs	5.33E-03	3.42E-03	2.19E-03	<1.03E-03	9.03E-02	1.83E-03
Spiny rush	Grasses & herbs	1.39E-02	2.22E-02	3.56E-02	9.15E-03	2.46E-01	4.76E-03
Curry plant	Grasses & herbs	2.13E-02	1.63E-02	3.21E-02	1.01E-02	2.07E-01	1.69E-02
Smilo grass	Grasses & herbs	5.38E-02	3.20E-03	1.55E-02	4.20E-02	3.90E-01	1.10E-03
Common reed	Grasses & herbs	4.41E-02	1.36E-02	5.28E-02	4.93E-03	1.11E-01	<3.97E-03
Mastic	Shrub	2.33E-02	1.20E-02	1.68E-02	4.49E-03	4.73E-01	3.25E-03
Spanish broom	Shrub	8.50E-03	5.07E-03	<5.28E-03	8.93E-04	3.45E-01	3.01E-04
Blackberry	Shrub	<4.46E-03	2.75E-03	2.05E-03	<6.85E-04	1.60E-01	6.10E-04
Oleander	Shrub	2.00E-02	2.67E-02	1.88E-02	8.84E-03	1.80E-01	1.56E-03
Pittosporum	Shrub	1.80E-02	6.71E-02	5.76E-02	1.97E-02	5.48E-01	6.49E-04
Spanish broom*	Shrub	<2.75E-03	1.25E-03	7.25E-04	<4.39E-04	9.50E-02	9.46E-04
Mastic*	Shrub	<2.75E-03	2.65E-03	3.33E-03	<4.39E-04	1.89E-01	1.42E-03
Pine tree	Tree	5.42E-03	7.23E-03	7.63E-03	3.37E-03	1.87E-01	3.90E-03
Cypress	Tree	3.25E-03	5.33E-03	4.62E-03	2.68E-03	3.24E-01	6.01E-04
Fig	Tree	6.96E-03	1.58E-03	6.30E-03	2.05E-03	3.80E-01	1.10E-02
Tamarisk	Tree	4.89E-03	1.33E-03	2.05E-03	3.77E-03	1.55E-01	2.44E-03
Summary statistics							
GM ± GSD		9.62E-03 ± 2.54E+00	6.16E-03 ± 3.10E+00	8.24E-03 ± 3.56E+00	3.21E-03 ± 3.68E+00	2.21E-01 ± 1.72E+00	1.91E-03 ± 2.90E+00
AM ± SD		1.49E-02 ± 1.51E-02	1.20E-02 ± 1.66E-02	1.65E-02 ± 1.85E-02	7.16E-03 ± 1.06E-02	2.55E-01 ± 1.39E-01	3.45E-03 ± 4.45E-03
RANGE		2.75E-03 ± 5.38E-02	1.25E-03 ± 6.71E-02	7.25E-04 ± 5.76E-02	4.39E-04 ± 4.20E-02	9.03E-02 ± 5.48E-01	3.01E-04 ± 1.69E-02

<CR values calculated by plant activity concentration values that are below limit of detection.

*Duplicate samples from sampling location no. 6.

negative, but strong statistical dependency with activity concentration in soil ($\rho = -0.81, p < 0.001$). Linear regression indicated that overall there was significant relationship ($p < 0.05$) between soil activity concentrations and log-CR values, although small R^2 values (proportion of variability explained by the independent variable) indicated weak correlation: ^{238}U ($R^2 = 0.30, p < 0.03$), ^{226}Ra ($R^2 = 0.32, p < 0.02$), ^{232}Th ($R^2 = 0.418, p < 0.007$), ^{40}K ($R^2 = 0.28, p < 0.03$), ^{137}Cs ($R^2 = 0.489, p < 0.002$). Spearman's rank did not show any statistically significant dependence of CR values for ^{208}Tl , but linear regression showed a weak one ($R^2 = 0.30, p < 0.03$). The results of these statistical tests are in agreement with authors who also found a negative relationship between CR values and soil concentrations (Chen et al., 2005; Sheppard et al., 2004), although there are studies who found no linear relationship (Vandenhove et al., 2009), contradictory evidence of linear relationship (Rodriguez et al., 2002) or linear relationship, but in the hydroponic conditions (Rodriguez et al., 2006).

Higher soil pH and moderate organic matter did suggest that the mobility and root uptake of radionuclides could be reduced (McLean and Bledsoe, 1992; Mitchell et al., 2013). However, we found no significant dependency (overall plant species) between single soil parameters (pH, LOI, EC) and radionuclide transfer to plants. The exception was the moderate negative effect of soil LOI on the uptake of ^{137}Cs ($\rho = -0.89, p < 0.05; R^2 = 0.30, p < 0.03$). This indicated that increased organic matter content favoured the retention of Cs, which was inversely related to the hypothesis that bioavailability of Cs increases with the increase of organic matter (Staunton et al., 2002). Although other authors did report a correlation between CR values and pH e.g. Vandenhove et al. (2009) for Th and Gerzabek et al. (1998) for Cs and Ra, the lack of such correlation in our case could be due to the narrow alkaline pH range (7–9) of our soil sample.

A comparison of our CR values with the literature showed more agreement with studies done in the Mediterranean region (Baeza et al., 2001; Vera Tome et al., 2003) than those reported by the ICRP (2011) and IAEA (2011). All statistically significant differences are marked in Table 7. Generally, the IAEA and ICRP datasets have higher CR values for U, Th, Ra in grass group, while for tree group they are lower than CRs calculated by our study. No significant difference was found between our and literature CR values for ^{40}K . Emphasis should be placed on our CR values of ^{137}Cs that are statistically very different, lower ($p < 0.0004$) than those of the ICRP and IAEA. These differences might be the result of various factors like soil pH, organic matter content, mineralogical soil composition, difference in plant species, etc. Influence of these factors on plant uptake of ^{137}Cs could not be more thoroughly analysed due to small number of data in our study.

Expanding the scarce database for plant uptake of radionuclides in terrestrial Mediterranean ecosystem has not been the only goal of this research. It has been suggested that, in radiological assessment of non-human biota, estimated dose rates due to human activities should be put in context by comparison with dose rates from natural background radiation, as the latter ones are normally experienced by animals and plants (Beresford et al., 2008). Therefore, our data on background radioactivity of soil and plants, and related radionuclide transfer, can be used for further radiological assessment within the Bay.

5. Conclusions

In this study, we use high resolution gamma-ray spectrometry to determine the background activity concentrations of ^{238}U , ^{226}Ra , ^{232}Th , ^{208}Tl , ^{40}K and ^{137}Cs in soils and plants of a Mediterranean ecosystem. Samples were collected in unpolluted areas of the Kaštela Bay, Croatia, on the east coast of the middle Adriatic Sea.

Table 7
Comparison of CR values (GM ± GSD, range, number of data) for plant groups within our study with literature data for same or similar plant groups.

Literature source	Plant group	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K	¹³⁷ Cs
This study	Grasses & herbs	2.06E-02 ± 2.30E+00	8.82E-03 ± 2.26E+00	1.83E-02 ± 3.10E+00	1.82E-01 ± 1.71E+00	3.64E-03 ± 2.54E+00
		5.33E-03–5.38E-02	3.20E-03–2.22E-02	2.19E-03–5.28E-02	9.03E-02–3.90E-01	1.10E-03–1.69E-02
		5	5	5	5	5
	Shrub	8.16E-03 ± 2.38E+00	7.19E-03 ± 3.72E+00	6.48E-03 ± 3.99E+00	2.40E-01 ± 1.80E+00	9.70E-04 ± 2.04E+00
		2.75E-03–2.33E-02	1.25E-03–6.71E-02	7.25E-04–1.88E-02	9.50E-02–4.73E-01	3.01E-04–3.25E-03
		7	7	7	7	7
	Tree	4.95E-03 ± 1.32E+00	3.00E-03 ± 2.09E+00	4.62E-03 ± 1.61E+00	2.44E-01 ± 1.45E+00	2.81E-03 ± 2.84E+00
		3.25E-03–6.96E-03	1.33E-03–7.23E-03	5.15E-03 – 2.41E-03	1.55E-01–3.80E-01	6.01E-04–1.10E-02
		4	4	4	4	4
IAEA ^a	Grasses & herbs	4.5E-02 ± 4.6E+00	5.4E-02 ± 4.9E+00*	9.9E-02 ± 3.7E+00*	3.2E-01 ± 2.0E+00	5.1E-01 ± 3.7E+00**
		7.7E-05 ± 5.5E+00	5.1E-05 ± 1.2E+01	2.2E-04 ± 2.7E+00	2.0E-02 ± 3.0E+00	1.9E-03 ± 3.7E+01
		439	464	341	43	2028
	Shrub	8.1E-02 ± 4.3E+00**	5.4E-01 ± 3.1E+00**	9.9E-02 ± 3.9E+00*	5.7E-01 ± 2.1E+00*	1.1E+00 ± 3.3E+00**
		1.4E-05 ± 5.9E+00	2.4E-02 ± 1.2E+01	1.2E-03 ± 3.9E+00	4.5E-02 ± 1.7E+00	9.8E-03 ± 1.6E+01
		970	504	403	22	354
	Tree	2.9E-03 ± 3.7E+00521	4.5E-04 ± 2.5E+00*	7.6E-04 ± 2.3E+00*	2.6E-01 ± 2.3E+00	7.5E-02 ± 3.1E+00*
		1.4E-05 ± 3.2E-02	1.1E-04 ± 2.4E-03	1.0E-05 ± 3.1E-03	1.4E-03 ± 6.6E-01	1.2E-03 ± 1.8E+00
		521	20	85	4	487
ICRP ^b	Wild grass	4.3E-02 ± 5.0E+00	9.2E-02 ± 4.8E+00*	9.5E-02 ± 2.6E+00*	2.8E-01 ± 1.9E+00	8.6E-01 ± 3.3E+00*
		7.7E-05 ± 5.5E+00	3.6E-03 ± 1.2E+01	1.6E-03 ± 6.5E-01	1.7E-01 ± 4.4E-01	3.6E-03 ± 3.7E+01
		151	168	30	26	1068
Pine tree	9.9E-04 ± 2.0E+00**	6.3E-04 ± 2.4E+00*	3.2E-04 ± 3.6E+00*	3.3E-01 ± 2.2E+00	7.5E-02 ± 3.2E+00**	
	2.0E-04 ± 1.8E-03	5.6E-04 ± 2.4E-03	1.0E-05 ± 1.8E-03	1.4E-03 ± 6.6E-01	1.2E-03 ± 1.8E+00	
	13	10	5	3	235	
Vera Tome et al. (2003) ^c	Grass-pasture	1.9E-02 (1.3E-02)	3.2E-02 (5.9E-02)*	1.2E-02 (1.3E-02)	n/a	n/a
		–	–	–	–	–
Baeza et al. (2001) ^d	Pasture	n/a	2.2E-02 ± 1.2E-02	2.0E-02 ± 6.0E-03	6.0E-02 ± 7.4E-02	5.0E-02 ± 3.4E-02
		–	–	–	–	–
		4	4	4	4	4

n/a = no available data.

*Data statistically significant different from CR values calculated in our study at $p < 0.05$.

**Data statistically significant different from CR values calculated in our study at $p < 0.001$.

^a Extracted and summarised data for wildlife category as used by IAEA EMRAS Working Group 5. These were output from the database in Feb 2011.

^b Extracted and summarised data for the ICRP Reference Animals and Plants. These were output from the database in Feb 2011.

^c Study was done in the area around a disused uranium mine. We considered only geometric means for two sampling sites not affected by the mine (in brackets are values from this second location). There are four samples for each sampling site.

^d CR values from this study represent annual mean values for aerial fraction of the pasture from vicinity of nuclear power plant.

Plant-soil concentration ratios were calculated and they range within one order of magnitude for ⁴⁰K (10^{-2} – 10^{-1}), ²³⁸U and ²²⁶Ra (10^{-3} – 10^{-2}), and two orders of magnitude for ²³²Th, ²⁰⁸Tl and ¹³⁷Cs (10^{-4} – 10^{-2}).

We found no differences between plant groups in radionuclide uptake. Statistical analysis showed that the effect of soil concentrations on plant radionuclide uptake was moderate but negative. No statistical dependency was found between soil parameters (pH, LOI, EC) and CR values. The exception was ¹³⁷Cs, whose CR value decreased with increase of soil organic matter content.

Our CR values were generally more in agreement with those reported by studies done in similar Mediterranean ecosystem than those reported by the ICRP and IAEA. This did not hold for the CR of ⁴⁰K, for which there were no statistically significant differences among different literature sources. The CR for ¹³⁷Cs, was significantly lower than values reported by international datasets.

Our results not only supplement the sparse data on radionuclide soil-to-plant transfer in Mediterranean ecosystem, but can also be used as “background” data against which data from contaminated areas inside the Bay can be compared.

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References

- Anagnostakis, M.J., Hiniš, E.P., Simopoulos, S.E., Angelopoulos, M.G., 1996. Natural radioactivity mapping of Greek surface soils. *Environ. Int.* 22, 3–8.
- Arapis, G.D., Karadinos, 2004. Migration of ¹³⁷Cs in the soil of sloping semi-natural ecosystems in Northern Greece. *J. Environ. Radioact.* 77, 133–142.
- Baeza, A., Paniagua, J., Rufo, M., Guillén, J., Sterling, A., 2001. Seasonal variations in radionuclide transfer a Mediterranean grazing-land ecosystem. *J. Environ. Radioact.* 55, 283–302.
- Beresford, N.A., Barnett, C.L., Jones, D.G., Wood, M.D., Appleton, J.D., Breward, N., Copplestone, D., 2008. Background exposure rates of terrestrial wildlife in England and Wales. *J. Environ. Radioact.* 99, 1430–1439.
- Brown, J.E., Alfonso, B., Avila, R., Beresford, N.A., Copplestone, D., Prohl, G., Ulanovsky, A., 2008. The ERICA tool. *J. Environ. Radioact.* 99, 1371–1383.
- Cesar, D., Sokolović, E., Kovač, J., 1994. Radioactivity of soil in Croatia. In: Franić, Z., Kubelka, D. (Eds.), *Proceedings of 2nd Symposium of Croatian Radiation Protection Society*, pp. 107–114.
- Charro, E., Pardo, R., Peña, V., 2013. Chemometric interpretation of vertical profiles of radionuclides in soils near a Spanish coal-fired power plant. *Chemosphere* 90, 488–496.
- Chen, S.B., Zhu, Y.G., Hu, Q.H., 2005. ²³⁸U, ²²⁶Ra and ²³²Th on a uranium mining-impacted soil from southeastern China. *J. Environ. Radioact.* 82, 222–236.
- Dai, L., Wei, H., Wang, L., 2007. Spatial distribution and risk assessment of radionuclides in soils around a coal-fired power plant: a case study from the city of Baoji, China. *Environ. Res.* 104, 201–208.
- Ehlikens, S., Kirchner, 2002. Environmental processes affecting plant root uptake of radioactive trace elements and variability of transfer factor data: a review. *J. Environ. Radioact.* 58, 97–112.

- Flues, M., Moraes, V., Mazzilli, B.P., 2002. The influence of a coal-fired power plant operation on radionuclide concentrations in soil. *J. Environ. Radioact.* 63, 285–294.
- Gerzabek, M.H., Strebl, F., Temmel, B., 1998. Plant uptake of radionuclides in lysimeter experiments. *Environ. Pollut.* 99, 93–103.
- Hidroelektra-projekt, Zagreb, 2004. Studija o utjecaju na okoliš za zahvat – izgradnja pomorskih i kopnenih objekata na prostoru «Giričić»-Kaštel Gomilica. Available from: <http://www.marina-kastela.hr/pdf/UtjecajOkolis.pdf> (in Croatian).
- Hinton, T.G., Garnier-Laplace, J., Vandenhove, H., Dowdall, M., Adam-Guillermin, C., Alonzo, F., Barnett, C., Beaugelin-Seiller, K., Beresford, N.A., Bradshaw, C., Brown, J., Eyrolle, F., Fevrier, L., Fariel, J.C., Gilbin, R., Hertel-Aas, T., Horemans, N., Howard, B.J., Ikäheimonen, T., Mora, J.C., Oughton, D., Real, A., Salbu, B., Simon-Cornu, M., Steiner, M., Sweeck, L., Vives i Batlle, J., 2013. An invitation to contribute to a strategic research agenda in radioecology. *J. Environ. Radioact.* 115, 73–82.
- Howard, B.J., Beresford, N.A., Copplestone, D., Telleria, D., Proehl, G., Fesenko, S., Jeffrey, R.A., Yankovich, T.L., Brown, J.E., Higley, K., Johansen, M.P., Mulye, H., Vandenhove, H., Gashchak, S., Wood, M.D., Takata, H., Andersson, P., Dale, P., Ryan, J., Bollhöfer, A., Doering, C., Barnett, C.L., Wells, C., 2013a. The IAEA handbook on radionuclide transfer to wildlife. *J. Environ. Radioact.* 121, 55–74.
- Howard, B.J., Wells, C., Beresford, N.A., Copplestone, D., 2013b. Exploring methods to prioritise concentration ratios when estimating weighted absorbed dose rates to terrestrial reference animals and plants. *J. Environ. Radioact.* 126, 326–337.
- HRN EN-12457:2005 Characterization of waste – leaching iance test for leaching of granular waste materials and sludges – Part 4: one stage batch test at a liquid to solid ratio of 10 l/kg for materials with particle size below 10 mm (without or with size reduction) (EN 12457–4:2002)
- HRN EN ISO 10523:2012 Water quality – Determination of pH (ISO 10523:2008; EN ISO 10523:2012).
- HRN EN 27888:2008 Determination of electrical conductivity (ISO 7888:1985; EN 27888:1993).
- HRN EN 12879:2000 Characterization of sludges – Determination of the loss on ignition of dry mass (EN 12879:2000).
- IAEA, 2009. Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments. IAEA, Vienna. IAEA-TECDOC-1616.
- IAEA, 2010. Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments. IAEA, Vienna. IAEA-TRS-472.
- IAEA summary tables, February 2011. Available from: <http://www.wildlifetransferdatabase.org/downloadsummary.asp>.
- ICRP RAP summary tables, February 2011. Available from: <http://www.wildlifetransferdatabase.org/downloadsummary.asp>.
- Lovrić, A.Ž., Oleg, A., 1999. Analysis of vegetation landscape in Kaštela across the transect Ciovo-Kozjak. In: Hodžić, M. (Ed.), *Kaštela kolijevka Hrvatske: zbornik: radovi sa simpozija u Kaštel Starom*, pp. 548–551 in Croatian.
- Laubenstein, M., Magaldi, D., 2008. Natural radioactivity of some red Mediterranean soils. *Catena* 76, 22–26.
- McLean, J.E., Bledsoe, B.E., 1992. Behaviour of Metals in Soil. U.S. EPA, EPA/540/S-92/018. Robert S. Kerr Laboratory, Ada, OK.
- Mitchell, N., Pérez-Sánchez, D., Thorne, M.C., 2013. A review of the behaviour of U-238 series radionuclides in soils and plants. *J. Radiol. Prot.* 33, R17–R48.
- Petrinec, B., Franić, Z., Bituh, T., Babić, D., 2011. Quality assurance in gamma-ray spectrometry of seabed sediments. *Arh. Hig. Rada Toksikol.* 62, 17–23.
- Porto, P., Walling, D.E., Ferro, V., 2001. Validating the use of caesium-137 measurements to estimate soil erosion rates in a small drainage basin in Calabria, Southern Italy. *J. Hydrol.* 248, 93–108.
- Rodríguez, B.P., Vera-Tomé, F., Lozano, J.C., 2002. About the assumption of linearity in soil-to-plant transfer factors for uranium and thorium isotopes and ²²⁶Ra. *Sci. Total Environ.* 284, 167–175.
- Rodríguez, B.P., Vera Tomé, F., Pérez Fernández, M., Lozano, J.C., 2006. Linearity assumption in soil-to-plant transfer factors of natural uranium and radium in *Helianthus annuus* L. *Sci. Total Environ.* 361, 1–7.
- Rodríguez, B.P., Vera Tomé, F., Lozano, J.C., Pérez Fernández, M.A., 2010. Transfer of ²³⁸U, ²³⁰Th, ²²⁶Ra, and ²¹⁰Pb from soils to tree and shrub species in a Mediterranean area. *Appl. Radiat. Isot.* 68, 1154–1159.
- Sheppard, S.C., Sheppard, M.I., Sanipelli, B.L., Tait, J.C., 2004. Background radionuclide concentrations in major environmental compartments of natural ecosystems. Report by EcoMatters for the Canadian Nuclear Safety Commission Contract No. 87055020215.
- Shtangeeva, I., 2010. Uptake of uranium and thorium by native and cultivated plants. *J. Environ. Radioact.* 101, 458–563.
- Staunton, S., Dumat, C., Zsolnay, A., 2002. Possible role of organic matter in radionuclide adsorption in soils. *J. Environ. Radioact.* 58, 163–173.
- Termizi Ramli, A., Wahab, M.A., Hussein, A., Wood, A.K., 2005. Environmental ²³⁸U and ²³²Th concentration measurements in an area of high level natural background radiation at Palong, Johor, Malaysia. *J. Environ. Radioact.* 80, 287–304.
- Ujević, I., Odžak, N., Barić, A., 2000. Trace metal accumulation in different grain size fractions of the sediments from a semi-enclosed Bay heavily contaminated by urban and industrial wastewaters. *Wat. Res.* 34 (11), 3055–3061.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000. Sources and effects of ionising radiation. In: Sources, Annex B: Exposure from Natural Radiation Sources vol. i. Report to the General Assembly, United Nations, New York, USA.
- Vandenhove, H., Van Hees, M., Wouters, K., Wannijn, J., 2007. Can we predict uranium bioavailability based on soil parameters? Part 1: effect of soil parameters on soil solution uranium concentration. *Environ. Pollut.* 145, 587–595.
- Vandenhove, H., Van Hees, M., 2007. Predicting radium availability and uptake from soil properties. *Chemosphere* 69, 664–674.
- Vandenhove, H., Olyslaegers, G., Sanzharova, N., Shubina, O., Reed, E., Shang, Z., Velasco, H., 2009. Proposal for new best estimates of the soil-to-plant transfer of U, Th, Ra, Pb and Po. *J. Environ. Radioact.* 100, 721–732.
- Vera Tomé, F., Blanco Rodríguez, M.P., Lozano, J.C., 2002. Distribution and mobilization of U, Th and ²²⁶Ra in the plant-soil compartments of a mineralized uranium area in south-west Spain. *J. Environ. Radioact.* 59, 41–60.
- Vera Tomé, F., Blanco Rodríguez, M.P., Lozano, J.C., 2003. Soil-to-plant transfer factors for natural and stable elements in a Mediterranean area. *J. Environ. Radioact.* 65, 161–175.
- Verheye, W., de la Rosa, D., 2009. Mediterranean soils, in Land use, land cover and soil sciences. In: Verheye, W., de la Rosa, D. (Eds.), *Encyclopedia of Life Support Systems (EOLSS)*, Developed under the Auspices of the UNESCO. Eolss Publishers, Oxford, UK. <http://www.eolss.net>.
- Wood, M.D., Marshall, W.A., Beresford, N.A., Jones, S.R., Howard, B.J., Copplestone, D., Leah, R.T., 2008. Application of the ERICA integrated approach to the Drigg coastal sand dunes. *J. Environ. Radioact.* 99, 1484–1495.
- Wood, M.D., Beresford, N.A., Semenov, D.V., Yankovich, T.L., Copplestone, D., 2010. Radionuclide transfer to reptiles. *Radiat. Environ. Biophys.* 49, 509–530.
- Wood, M.D., Beresford, N.A., Howard, B.J., Copplestone, D., 2013. Evaluating summarised radionuclide concentration ratio datasets for wildlife. *J. Environ. Radioact.* 126, 314–325.