J. KAU: Mar. Sci., **vol. 14,** pp. 53-64 (2003 A.D. / 1423 A.H.)

Polychlorinated Biphenyls (PCBs) and Chlorinated Pesticides Contamination in Coral Reef Skeleton from the Egyptian Red Sea Coast

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ABSTRACT. The residues of 17 organochlorine pollutants in coral reef skeleton (*Acropora* sp.) collected from 20 different locations in April 1999 from Egyptian Red Sea Coast were analyzed by GC. The pollutants studied were α-, β-, γ-hexachlorocyclohexane (HCHs), Cyclodienes (heptachlor, aldrin, dieldrin, heptachlorepoxide) as well as sum DDT (*p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT) and seven individual polychlorinated biphenyl (PCBs) congeners. The concentrations of sum DDT were in a range from 1 to 12 ng g^{-1} with an average 6 ng g^{-1}. The highest DDT concentrations were recorded at Sharm El-Maya (St. No. 11), whereas the PCB levels ranged from 6.2 to 48.3 ng g^{-1} with an average 18.9 ng g^{-1} and HCHs level ranged from 0.8 to 14.5 ng g^{-1} with an average 4 ng g^{-1}. The PCBs were dominant in most studied locations. The present results indicate low to moderate PCBs and pesticides contamination in most of the studied coral reef samples.

KEYWORDS: Coral reef, Organochlorines, Pesticides, Polychlorinated biphenyls (PCBs), Organic pollution, Red Sea, Egypt .

Introduction

Chlorinated organic compounds have a wide range of industrial and agricultural applications. They include pesticides such as DDT (dichlorodiphenyltrichloroethane) and Lindane (γ-HCH or gamma-hexachloro-cyclohexane) and polychlorinated biphenyls (PCBs) which are used in a range of industrial applications in-

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cluding dielectrics in electrical transformers. Organochlorines have been implicated in reproductive and immunological abnormalities observed in birds and marine mammals (Livingston, 1976). The highest concentrations of organochlorines have been associated with centers of urbanization in most of studies elsewhere, such as estuarine and marine sediments near major metropolitan areas along the eastern coast of the United States (NRC, 1989) and at a wide range of locations in Europe, Asia and Africa associated with human settlement (Alvarez Pineiro *et al*., 1995; Agnihotri *et al.*, 1996; Thompson *et al*., 1996; Abed-Allah *et al*., 1998).

Man-made organochlorines have been considered a serious threat to the long-term health of the marine environment for many years. The main reasons are their strong accumulation in lipid tissues of marine biota as well as the high toxicity to marine organisms and the slow degradation of several members of this group.

Various effects of pollution on coral reef organisms and communities have been documented (*e.g.* Wood and Johannes, 1975; Loya and Rinkevich, 1980; Hatcher *et al.*, 1989; Rogers, 1989; Hughes, 1994).

No systematic studies have been conducted on coral reefs in the Egyptian part of the Red Sea. The little available information indicates that reefs are better developed on the west coast of the Gulf of Suez than on the east one. They are patchy and very shallow at far north, but start to develop into a more or less continuous fringing reef at Sukhna. Branching corals dominate the communities there due to high sedimentation rates, but *Porites* occur also. In the Gulf of Aqaba, fringing reefs are developed on foot of steep cliffs skirting almost the whole length of the Gulf coast. These reefs are usually narrow, but may reach a width as high as 1 km in embayments and areas of old wadi systems. Reefs are much more developed as well as diversified in the Red Sea proper. Actually, very good reefs occur up to the entrances of the two Gulfs at north. Generally, reefs become increasingly wider and deeper as well as much more diverse as we move from north to south along the coast (DANIDA, 1996; EIMP, 1996).

This investigation was undertaken to determine the residues of organochlorine pollutants for the first time in coral reef skeleton collected from Egyptian Red Sea Coast (El Nemr *et al.*, 2004).

Materials and Methods

Sampling

The coral reef sampling stations were located along the Red Sea coast starting from Marsa Alam to Taba (about 900 km, Figure 1). Coral reef was collected at 20 sites; only one type of coral reef (*Acropora* sp.) was collected for this

study within a period of two weeks during April 1999. At each site, large parts of coral reef were collected within a reef area of 10 km^2 at each of the 20 sites with collection depths ranged from 2.5-5 m. Coral samples were collected from the top of the colonies, avoiding areas of discontinuities where additional sediment had been included into the skeleton during a partial mortality, injury, or invasion by bioeroders (Scott, 1990). Coral samples were sun-dried, placed in pre-cleaned aluminum bags, and transported to the laboratory. Coral samples were washed with distilled water, and dried in an oven at 50ºC for 48 hr. Subsamples (100-150 g) of coral skeleton containing at least the last 4 years of skeletal growth (based on growth rates of 5-6 mm/year, (Guzman *et al.,* 1991) were crushed in ball mill and homogenized before extracting with hexane.

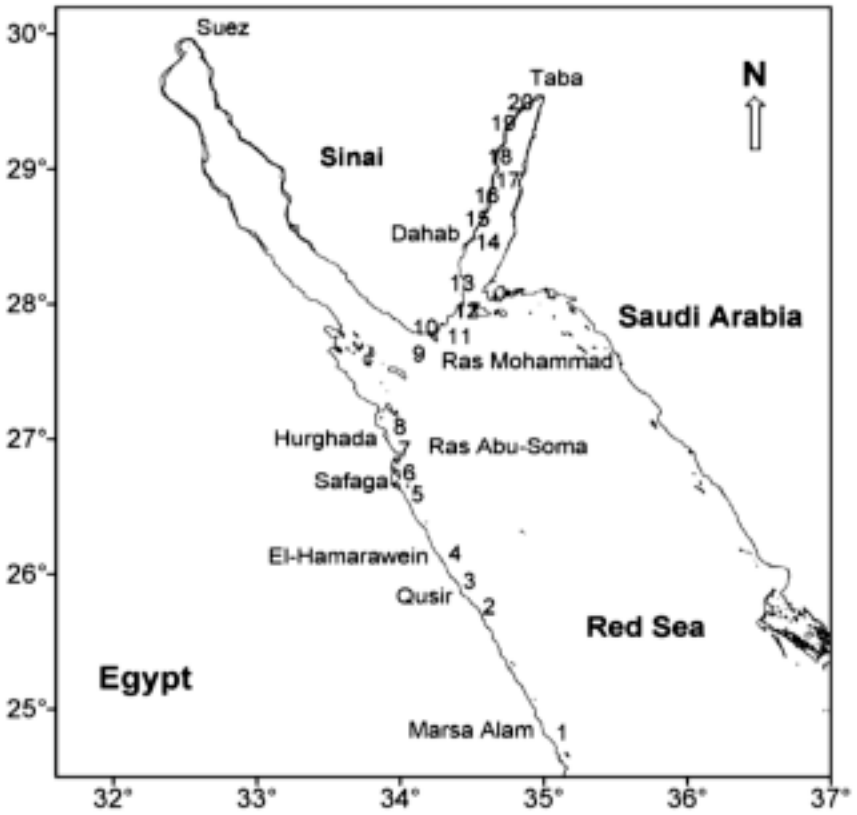

FIG. 1. Sampling locations in the Egyptian Red Sea.

Chemicals

All solvents were pesticides grade purchased from Merck (Germany). Sodium sulfate was extracted with hexane in a soxhlet apparatus for 8 hours and then with methanol or dichloromethane for another 8 hours, then pre-

combustion in a muffle furnace at approximately 400ºC overnight and cooled in a greaseless desiccators. Florisil used for column chromatography was solvent extracted with n-hexane in a glass cartridge inserted into an extraction apparatus, as described by Ehrhardt (1987). After extraction, they were first dried in the same cartridge by passing nitrogen stream through it and were then activated by heating the cartridge in an electric tube oven to 200ºC for 6 hours followed by partially deactivated with 0.5% water by weight and stored in a tightly sealed glass jar with ground glass stopper and the mixture were allowed to equilibrate for one day before use. The activation/deactivation procedure was made one day before use.

GC Conditions

Hewlett Packard 5980 series II high-resolution gas chromatograph (Hewlett Packard, USA) equipped with a ^{63}Ni electron capture detector (ECD) was used for analysis. A fused silica capillary column (50 m × 0.32 mm × 0.52 μm) coated with DB-1 (5% diphenyl and 95% dimethyl polysiloxane) was used for the quantification. The oven temperature was programmed from an initial temperature of 70ºC (2 min hold) to 280ºC at a rate of 5ºC min^{-1} and was then maintained at 280ºC for 20 min. Injector and detector temperatures were maintained at 270 and 300ºC, respectively. Helium was used as the carrier (1.5 ml min^{-1}) and nitrogen as the make-up (60 ml min^{-1}) gas.

Method

30 g of coral reef was soxhlet extracted with 200 ml n-hexane for 8 hours with a siphon cycle of around 10 minutes. After the extraction was completed the extract solvent was concentrated to 15 ml using a rotary evaporator under 40ºC and transferred into a Kuderna-Danish concentrator and the volume of the extract further reduced to 1 ml under a gentle stream of pure nitrogen gas. A chromatography column is prepared using 50 ml burette in which a piece of glass wool is added near the stopper to maintain the packing material. Then 20 g of florisil were transferred into the column followed by 1 g of sodium sulfate.

The extract (1 ml) was sequentially eluted from the column with 70 ml of hexane for PCBs congeners, fraction (F1). Then the column was eluted with 50 ml of mixture containing 70% of hexane and 30% of dichloromethane for pesticide fraction (F2). F1 and F2 were evaporated by gentle stream of nitrogen for instrumental analysis.

An equivalent mixture provided by Dr. Ehrenstorfer Laboratories (Augsburg, Germany) with known PCBs composition and content was used as the standard. Organochlorine pesticides were quantified from individually resolved peak are-

as with the corresponding peak areas of the external standards (POC mixture provided by IAEA). To control the analytical reliability and assure recovery efficiency and accuracy of the results, 5 analyses were conducted on PCB reference material 2974 provided by EIMP-IAEA. The laboratory results showed recovery efficiency ranging from 96-106% with coefficients of variation between 10-14%.

Results and Discussion

GC analysis for hexane extracts of coral reef skeleton exhibited the presence of organochlorines including α-, β- and γ-HCH, dieldrin, aldrin, heptachlor, *p-p′*-DDT, *p-p′*-DDE and *p-p′*-DDD as well as PCBs (Tables 1 and 2). Concentration of all these organochlorine pesticides in samples collected from the Egyptian Red Sea coast (Figure 1) were somewhat low in most studied area. However, The concentrations of organochlorines in coral reef (*Acropora* sp.) skeleton decreased in the order of PCBs > DDTs > HCHs > cyclodienes for the most of studied locations. Concentrations of sum DDT in the coral skeleton ranged from 1 ng g^{-1} at station 20 (Taba) to 12 ng g^{-1} at station 11 (Sharm El-Maya) with an average 6 ng g^{-1}. Among DDT metabolites, *p-p′*-DDE accounted for a range from 50 to 95% of the sum DDT. The highest percentage of *p-p′*-DDE was found at stations 6 (81%), 11 (96%), 13 (94%), 14 (84%), 16 (84%), 17 (93%) and 19 (93%). Lowest concentrations of sum DDT were recorded for stations 2, 3, 4, 10, 12, 16 and 20 (ranged from 1 to 2.5 ng g^{-1}). These results reflects few fresh-inputs of DDT to the environment along the Red sea coast. Metabolic transformation of *p-p′*-DDT under oxidative conditions lead to *p-p′*-DDE, whereas under anaerobic conditions *p-p′*-DDD is formed (Villeneuve *et al*., 1999).

Average concentrations of total HCHs in the coral reef samples were 4 ng g^{-1} with a range of 0.8 to 15 ng g^{-1} (Table 1). Although the use of γ-HCH in agriculture has been much greater than *p-p′*-DDT, the average ratio of total HCHs to sum DDT was 70%. The relatively low concentrations of HCHs reflect their lower potential for bioaccumulation in the coral reef skeleton. Furthermore, higher vapor pressures of HCHs than of *p-p′*-DDT and its metabolites facilitate relatively rapid atmospheric dissipation (Kannan, *et al*., 1995).

The concentration of the cyclodienes (heptachlor, aldrin, heptachlor-epoxide and dieldrin) in the skeleton were from 0.5 ng g^{-1} at stations 2, 4, 13, 14 and 20 to 5.5 ng g^{-1} at station 19, with the average 1.6 ng g^{-1} showing an average of 4-fold less than total DDT and almost half of the HCH concentration levels (Table 1 and Figure 2). The concentrations of aldrin, dieldrin and heptachlorepoxide were higher at stations 1, 5 and 9 whereas their concentration at the other stations were much lower.

TABLE 1. Concentration (ng/g) of organochlorine pesticides in coral reef samples.

Site	α-HCH	β-HCH	γ-HCH	**T.HCHS**	HC	aldrin	HCE	dieldrin	**T. CYD**	*p,p'*-DDT	*p,p'*-DDD	*p,p'*-DDE	**T. DDT**	**GT**
1	5.7	1.1	4.3	**11.1**	0.1	0.2	1.2	1.6	**3.1**	3.4	1.1	3.7	8.2	**22.4**
2	0.1	0.2	0.5	**0.8**	0.1	0.1	0.1	0.2	**0.5**	0.1	0.6	1.7	2.4	**3.7**
3	0.3	0.6	0.3	**1.2**	0.1	0.2	0.1	0.2	**0.6**	0.2	1.0	1.2	2.4	**4.2**
4	0.1	0.2	0.6	**0.9**	0.1	0.1	0.1	0.2	**0.5**	0.1	0.4	1.5	2.0	**3.4**
5	3.1	7.3	2.9	**13.3**	0.1	1.0	0.9	1.4	**3.4**	1.2	6.1	3.4	10.7	**27.4**
6	0.8	1.1	3.6	**5.5**	0.3	0.3	0.2	0.3	**1.1**	0.8	1.0	7.9	9.7	**16.3**
7	3.1	8.5	3.0	**14.6**	0.1	0.5	0.4	0.3	**1.3**	0.4	1.9	2.1	4.4	**20.3**
8	0.2	1.1	1.2	**2.5**	0.2	0.2	0.1	0.2	**0.7**	0.1	0.9	3.0	4.0	**7.2**
9	1.3	1.6	1.8	**4.7**	1.6	0.9	0.9	1.0	**4.4**	1.0	2.4	2.8	6.2	**15.3**
10	0.2	0.4	2.3	**2.9**	0.9	0.5	0.1	0.4	**1.9**	0.1	0.9	1.8	2.8	**7.6**
11	0.3	0.7	1.5	**2.5**	0.2	0.2	0.1	0.1	**0.6**	0.1	0.3	11.3	11.7	**14.8**
12	0.3	0.6	1.4	**2.3**	0.2	0.1	0.3	0.1	**0.7**	0.1	1.0	1.4	2.5	**5.5**
13	0.3	0.4	1.0	**1.7**	0.2	0.1	0.1	0.1	**0.5**	0.1	0.3	6.7	7.1	**9.3**
14	0.2	0.4	1.1	**1.7**	0.1	0.1	0.1	0.2	**0.5**	0.1	0.9	5.2	6.2	**8.4**
15	0.3	0.6	1.2	**2.1**	0.2	0.2	0.1	0.5	**1.0**	0.2	1.1	4.2	5.5	**8.6**
16	0.2	0.5	1.7	**2.4**	0.1	0.2	0.2	0.1	**0.6**	0.1	0.3	1.6	2.0	**5.0**
17	0.4	0.8	1.5	**2.7**	0.4	0.3	0.1	0.2	**1.0**	0.1	0.6	9.2	9.9	**13.6**
18	0.3	0.5	1.6	**2.4**	0.5	0.4	0.1	0.7	**1.7**	0.2	1.7	3.5	5.4	**9.5**
19	0.8	1.6	1.7	**4.1**	0.1	5.1	0.1	0.2	**5.5**	0.3	0.4	9.0	9.7	**19.3**
20	0.2	0.3	0.3	**0.8**	0.1	0.1	0.1	0.2	**0.5**	0.1	0.3	0.6	1.0	**2.3**
Average	**0.9**	**1.4**	**1.7**	**4.0**	**0.3**	**0.6**	**0.3**	**0.4**	**1.6**	**0.4**	**1.2**	**4.1**	**5.7**	**11.3**

1: Marsa Alam; 2: Qusir; 3: Qusir Ref; 4: El-Hamarawien Ref.; 5: Safaga; 6: Pub beach Safaga; 7: Ras Abo-Soma; 8: Horgada NIOF; 9: Ras Mohammad; 10: Sharm El Mina; 11: Sharm El-Maya; 12: Na'ama Bay; 13: Nakhlat El-Tal; Dahab; 15: Ras Mamlah; 16: Hibeiq Ras Nabar; 17: Nuweiba, El-Siadin; 18: Nuweiba; 19: Marsa Muqibila; 20: Taba; HC: heptachlor; HCE: heptachlorepoxide; CYD: cyclodiene; GT: Grand total (total pesticides); T = total.

TABLE 2. Concentration (ng/g) of PCBs congeners in coral reef samples.

Site	28	52	101	118	138	153	180	Total
1	3.1	4.0	1.7	0.4	0.7	0.8	0.1	**10.8**
2	5.8	7.1	4.6	1.2	2.4	3.1	0.1	**24.3**
3	9.2	9.0	8.0	1.2	2.3	3.0	4.1	**36.8**
4	8.3	1.4	1.9	2.1	0.5	4.9	2.0	**21.1**
5	2.1	2.1	1.3	0.4	0.6	1.3	0.7	**8.5**
6	3.0	3.0	1.4	0.6	0.2	0.1	1.0	**9.3**
7	6.3	8.1	1.1	1.1	1.2	0.9	0.1	**18.8**
8	4.3	11.6	0.7	0.9	1.4	1.1	1.2	**21.2**
9	2.2	2.9	15.2	1.0	2.4	2.3	4.8	**30.8**
10	1.2	3.4	4.5	4.2	2.1	2.3	1.9	**19.6**
11	2.0	2.1	0.7	0.1	0.1	0.1	7.9	**13.0**
12	1.1	1.9	0.7	0.3	0.2	0.1	1.9	**6.2**
13	1.8	7.4	1.1	0.6	0.4	0.2	2.6	**14.1**
14	1.3	1.5	1.7	0.2	0.4	0.2	5.2	**10.5**
15	6.9	14.3	4.9	2.7	5.5	3.9	10.1	**48.3**
16	1.2	2.9	2.5	1.9	1.2	3.3	2.4	**15.4**
17	2.2	1.9	1.2	0.2	0.1	0.1	0.5	**6.2**
18	5.3	6.9	1.8	4.5	1.9	1.9	3.3	**25.6**
19	1.4	1.5	1.7	0.3	0.2	0.1	1.2	**6.4**
20	3.6	7.7	13.7	1.9	1.8	1.5	0.9	**31.1**
Average	**3.6**	**5.0**	**3.5**	**1.3**	**1.3**	**1.6**	**2.6**	**18.9**

28 = 2, 4, 4'-Trichlorobiphenyl; 52 = 2, 2',5,5'-tetrachlorobiphenyl; 101 = 2, 2' 4, 5, 5'-pentachlorobiphenyl; 118 = 2, 3' 4, 5, 5'-pentachlorobiphenyl; 138 = 2, 2', 3, 4, 4', 5'-hexachlorobiphenyl; 153 = 2, 2', 4, 4',,5,5'-hexachlorobiphenyl; 180 = 2, 2', 3, 4, 4', 5, 5'-heptachlorobiphenyl.

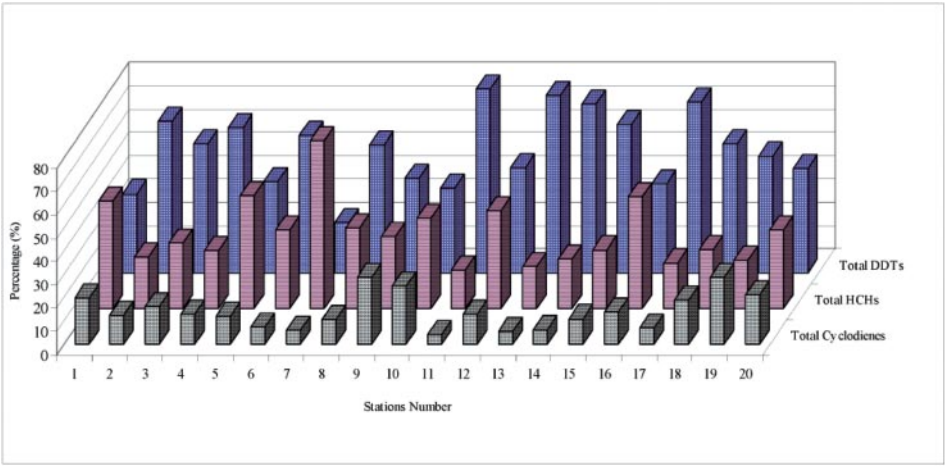

FIG. 2. Percentage of total HCHs, Cyclodienes and DDTs relative to total pesticides in coral reef.

The PCBs were present in higher concentrations relative to other organochlorines in this study (Table 2). The PCB concentrations were in the range of 6 ng g^{-1} at stations 12, 17 and 19 to 48 ng g^{-1} at station 15, with an average concentration of 19 ng g^{-1}. PCBs concentrations were on average 3 fold the sum DDT concentration averaged over all station, while it was up to 15-fold at station 3 alone (Tables 1 and 2).

The highest concentration of PCB 28 (9 ng g^{-1}, 49%) was recorded at station 3, PCBs 52 and 138 (14, 5.5 ng g^{-1}, respectively) at station 15, PCBs 101 (15 ng g^{-1}) at station 9, PCBs 118 (4.5 ng g^{-1}) at station 18, PCBs 153 (4.9 ng g^{-1}) at station 4 and PCBs 180 (10.1 ng g^{-1}) at station 15 (Table 2, Figure 3).

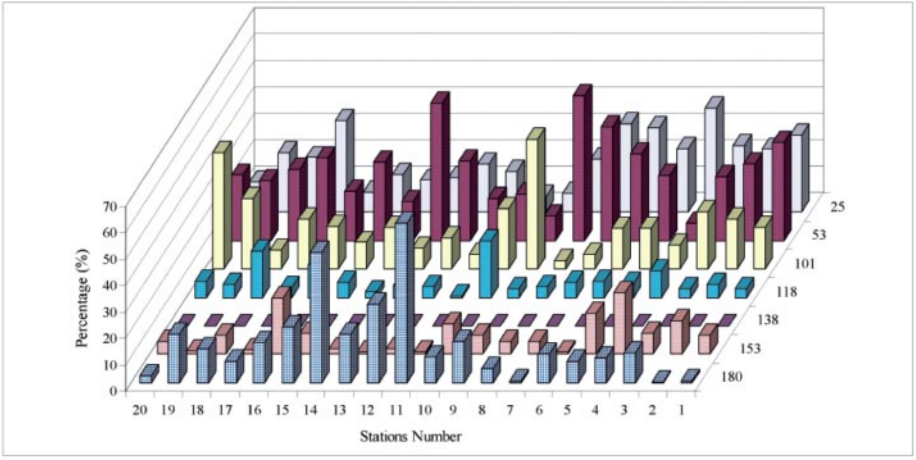

FIG. 3. Percentage of individual PCBs concentrations relative to total PCBs concentrations in coral reef.

Figure 4, exhibit the percentage of the cyclodienes, HCHs, DDTs and PCBs relative to the total organochlorine pollutants (grand total). It showed that, the PCBs were dominant in most studied locations except stations 5, 17 and 19 where DDT was the dominant. The total cyclodienes ratios ranged from 1.3 to 13.7% of the grand total, whereas the HCHs and sum DDT exhibited ratios ranging from 2 to 37% and 2.8 to 50%, respectively.

The persistence of *p-p′*-DDT in marine systems exhibited a half life $T_{1/2}$ is 5 years (Carvalho *et al.*, 1994; Quensen *et al.*, 1998). Assuming that after 1974 there has been no further releases of DDT, these half-life values would allow for an estimated reduction of DDT in coastal environment. Nevertheless, despite the ban of DDT there are still continuous inputs into the coastal environment, mainly by atmospheric deposition of *p-p′*-DDT (Villeneuve and Cattini, 1986) and DDT leaching from agricultural soils followed by discharges into estuarine areas (Claisse, 1989). These inputs would contribute to maintain DDT in

the coastal environment. Other chlorinated pesticides indicate a more rapid disappearance from the coastal environment than DDT (ILMR, 1975; Villeneuve *et al*., 1999) or a lower use in the coastal area in comparison with DDT.

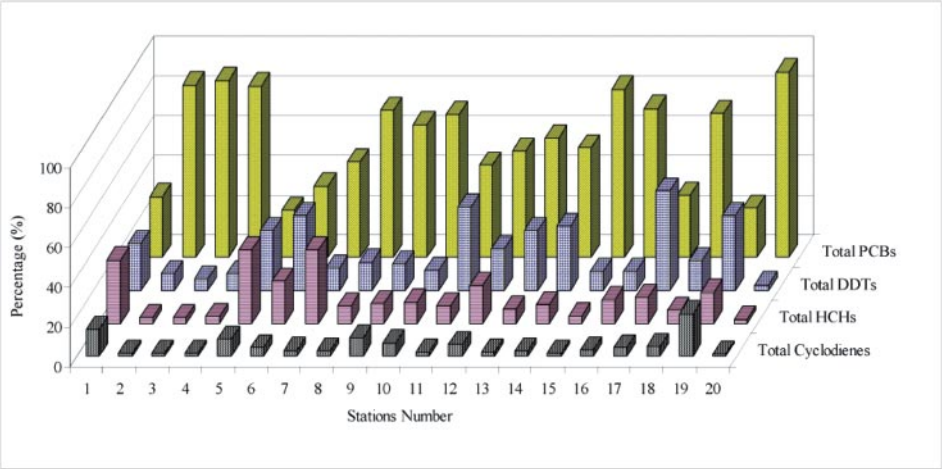

FIG. 4. Percentage of total HCHs, DDTs, Cyclodienes and PCBs relative to grand total of organic pollutant detected in coral reef.

Conclusion

The 20 locations investigated showed a low concentration of chlorinated pesticides and PCBs in most studied locations. The highest concentrations measured were 29 ng g^{-1} for the pesticides and 48 ng g^{-1} for PCBs congener. However, the sum DDT concentrations were in low concentration at most stations investigated. The concentrations of PCBs and pesticides observed in coral reef skeleton exhibited moderate differences with low accumulation for most locations.

The authors would like to express their sincere gratitude to NIOF for supporting the project. Also our deep appreciation for Mrs. Fadia Abu El-Maged, Mrs. Nadia El-Shaer, Mr. Mohamed Emam, and Mr. Ahmed El-Gamel for their kind assistance during the experimental work.

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مركبات ثنائية الفينيل متعددة الكلور والمبيدات المكلورة في هيكل الشعاب المرجانية الموجودة فى شواطئ البحر الأحمر بمصر

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المستخلص. اهتمت هذه الدراسة بدراسة ١٧ نوعا من المركبات العضوية فى هيكل الشعاب المرجانية (نوع أكروبورا). وقد جمعت العينات من ٢٠ موقع مختلف فى إبريل من سنة ١٩٩٩م من الشواطئ المصرية بالبحر الأحمر وقد تم قياس الملوثات (ألفا ، بيتا و جاما سداسى كلور الهكسين الحلقى ومركبات السيكلوداين بالأضافة إلى مبيدات الـ ددتى وسبعة مركبات من ثنائى الفينيل متعددة الكلور) فى العينات بواسطة جهاز التحليل الكروماتوجرافى وقد أوضحت هذه الدراسة أن مجموع تركيزات الـ ددتى تتراوح بين ١ إلى ١٢ نانوجرام/ جرام بمتوسط ٦ نانوجرام/ جرام.

وقد وجد أن أعلى تركيز لمجموع الـ ددتى كان فى منطقة شرم الميه ومن ناحية أخرى فإن تركيزات مركبات ثنائى الفينيل متعددة الكلور كانت تتراوح بين ٦,٢ إلى ٤٨,٣ نانوجرام/ جرام بمتوسط ١٨,٩ نانوجرام/ جرام وتركيزات مركبات سداسى كلور الهكسين الحلقى تراوحت بين ٠,٨ إلى ١٤,٥ نانوجرام/ جرام بمتوسط ٤ نانوجرام/ جرام.

وقد لوحظ أن مجموع تركيزات مركبات ثنائى الفينيل متعددة الكلور هى السائدة فى معظم المواقع . وقد أوضحت هذه الدراسة أن مستوى الملوثات العضوية تحت الدراسة كانت منخفضة إلى متوسطة فى معظم المواقع موضوع البحث.