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ORGANOCHLORINE PESTICIDES IN WATER, SEDIMENT AND FISH FROM THE NILE RIVER AND MANZALA LAKE IN EGYPT

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To assess the risks of organochlorine pesticides discharged into the hydrospheric environment of Egypt, river and lake water, drinking water, suspended solids, sediments and fish were collected during 1993–1994 from the Nile River and Manzala Lake in Egypt and were transported to Japan for chemical analysis. Among different organochlorine pesticides analyzed, p,p'-DDE was the most predominant in fish (7.6 to 67 ng/g wet wt.), sediments (3.2 to 432 ng/g dry wt.) and suspended solids (5.3 to 138 pg/L). However, in the dissolved phase of water samples HCH compounds predominated (α -HCH, 71 to 2,815 pg/L). Concentrations of organochlorine pesticides, except chlordane, were higher in Manzala Lake than in the River Nile. Concentrations of organochlorine pesticides in fish corresponded with those in sediments from each location. Comparison of organochlorine concentrations in Nile River water with those reported in earlier studies suggested a decrease in concentrations during the last decade. However, concentrations of p,p'-DDE has increased in fish. It appears that the release of this metabolite from contaminated sediment is the major source of p,p'-DDE in fish during recent years.

Keywords: Organochlorine pesticides; DDT; HCH; HCB; Egypt; Nile River

INTRODUCTION

Among several man-made agro-chemicals produced and used during the last four decades, organochlorine pesticides have received utmost attention due to their potential for bioaccumulation, persistence and toxicity in humans and wildlife. In

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Egypt, large amounts of organochlorine pesticides were used for agricultural purposes during the last three decades. Egypt is the largest pesticide market in Arabian countries and the fourth largest importer of pesticides among developing countries ^[1]. Because of the limited cultivable land available for agriculture, more than 30,000 metric tonnees of formulated pesticides are imported and used intensively to enhance production in the Nile River Delta ^[2]. The Ministry of Agriculture of Egypt has reported that the total active ingredients of major organochlorine pesticides used during a 30 year period were: Toxaphene, 54,000 metric tones (1955–1961); Endrin, 10,500 mt (1961–1981); DDT, 13,500 mt (1952– 1971); and Lindane (y-HCH), 21,000 mt (1952-1978). Based on the reports of the harmful effects of these pesticides to wildlife and humans, they were banned or restricted from use during the last decade. However, due to their low biodegradability, organochlorine pesticides used during the last three decades still persist in the environment. Nevertheless, very little information is available for organochlorine pesticides discharged into the Egyptian environment during these periods, and no temporal trend studies have been conducted to evaluate the effectiveness of regulations on the use of these pesticides [3, 4].

The present study examined pollution by organochlorine pesticides in the Nile River and Manzala Lake in Egypt. The Nile River supplies about 60 billion cubic meters of water annually, and one-third of its flow is used for agricultural irrigation. The Nile River water is also used for industrial activities, transportation, disposal of agricultural and industrial wastewater, which results in severe contamination by toxic pollutants. In addition, Manzala Lake, which receives drainages from Damietta branch of the Nile River, is a major source of fishery resources in Egypt, accounting for nearly 50% of the country's total fishery production during the early 1970s ^[5] and 35% during the 1980s ^[6]. For the assessment of risks of organochlorine pesticides discharged into the Nile River and Manzala Lake, baseline concentration information is needed. Therefore, as a part of international collaborative research between the National Institute for Resources and Environment in Japan and the University of Mansoura in Egypt, this study examined concentrations of organochlorine pesticides in water, sediment and fish from those aquatic environments.

EXPERIMENTAL

Sample collection

The general location of Manzala Lake and the Nile River is shown in Figure 1. Almost all environmental samples were collected during January 1993. Water,



FIGURE 1 General location of Nile River and Manzala Lake in Egypt

suspended solid, sediment and fish samples were collected from several locations in Manzala Lagoon (M1 to M5) and the Nile River near Cairo (C1 to C3), Egypt (Figures 2). Drinking water was collected from a tap at the University of Mansoura in January 1994.

About 20 L of water was collected using polyethylene containers. Water samples were transported to the laboratory and filtered using a glass fiber filter (GFF; diameter; 14 cm, pore size; 0.42 mm), which was cleaned by heating at 240°C for 48 h prior to use. Filtered water samples were passed through a glass column (2 cm i.d., 20 cm length) packed with 20 g of XAD-2 resin (Amberlite). The flow rate was maintained at 20 mL/min. A preliminary study showed that the recovery rates of all organochlorine pesticides analyzed in this study, using solid phase extraction with XAD-2 resin, were >91%. XAD-2 resin was cleaned with ethyl alcohol in a Soxhlet apparatus prior to use. Drinking water samples were collected twice using XAD-2. Surface sediment samples were collected using an



FIGURE 2 Sampling locations in Nile River near by Cairo and Manzala Lake

Eckman-Birdge sampler from all locations. Fish samples were purchased from fishermen at each location, except in Cairo where fishes were purchased at a local market. All samples of sediments, fishes, glass fiber filters and XAD-2 columns were transported to Japan in dry ice, and analyzed.

Analytical Procedure

Dichlorodiphenyltrichloroethane (p,p'-DDT) and its metabolites p,p'-DDD and p,p'-DDE, hexachlorocyclohexanes (HCHs; α -, β -, γ - and δ - isomers), chlordane compounds (CHLs; *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, heptachlor and oxychlordane) and hexachlorobenzene (HCB) were analyzed following the method described below.

Organochlorine pesticides adsorbed on XAD-2 resin were eluted with 300 mL of ethanol. The eluate was transferred to 100 mL of hexane in 500 mL of hexane-washed water in a separatory funnel. After partitioning, the hexane was concentrated and passed through 12 g of activated Florisil (130 °C for 10 h) packed glass column for separation. The first fraction eluted with 120 mL of hexane con-

tained HCB and p,p'-DDE. The second fraction eluted with 150 mL of 20% dichloromethane in hexane contained HCHs, p,p'-DDD, p,p'-DDT and CHLs. Each fraction was concentrated using a K-D (Kuderna-Danish) concentrator and washed with concentrated sulfuric acid and hexane-washed water. Suspended solids collected on glass fiber filter were ground with anhydrous sodium sulfate and pulverized mixtures were Soxhlet extracted with 400 mL dichloromethane and hexane (3:1). The extracts containing organochlorine residues were concentrated and treated with concentrated sulfuric acid. After washing with hexane-washed water, extracts were cleaned using Florisil column chromatography as described above. Because of the limited amount of samples, dry weight of suspended solids were not measured. Therefore, results for suspended solid concentrations are reported as pg/L.

Sediment samples were dried at room temperature, and then ground with anhydrous sodium sulfate and extracted in a Soxhlet extractor with 400 mL of dichloromethane : hexane (3:1). The extracts containing organochlorine residues were concentrated and washed with concentrated sulfuric acid. The extracts were treated with activated copper to remove sulfur and cleaned by passing through a Florisil column as described above.

For fish samples, whole fish were minced and homogenized twice using a blender. Twenty g of homogenized fish was ground with anhydrous sodium sulfate and extracted in a Soxhlet extractor with 400 mL of diethylether and hexane (3:1). The hexane extract containing organochlorine residues was concentrated and added to 20 g of Florisil packed in a glass column, and then hexane was removed by passing purified nitrogen. Organochlorine compounds adsorbed on Florisil were eluted using 150 mL of acetonitrile containing 20% water. The eluate was transferred to 100 mL of hexane and 500 mL of hexane-washed water in a separatory funnel. After partitioning, Florisil column separation was performed as described above.

Aliquots of final extracts were injected into a gas chromatograph (Hewlett Packard 5890 II) equipped with 63 Ni electron capture detector and capillary column (FS-WCOT, chemically bonded DB-17, 0.25 mm i.d., 30 m length). Column oven temperature was programmed from 160°C to 230°C at a rate of 1.5°C/min. Injector and detector temperatures were maintained at 250°C and 280°C, respectively. A few samples were also injected into a gas chromatograph equipped with OV-1701 (FS-WCOT, 0.25 mm i.d. × 25 m length) for confirmation of compounds detected using DB-17. Concentrations of organochlorine pesticides were quantified from individually resolved peak areas with corresponding standard peak areas. The recoveries of target pesticides through the entire analytical procedure were greater than 93%.

RESULTS AND DISCUSSION

Water

Concentrations of organochlorine pesticides measured in filtrated water samples (dissolved phase) are presented in Table I. HCHs were most predominant, followed by DDTs and CHLs. Among HCH isomers analyzed, β -HCH predominated in samples from the Nile River in Cairo. However, concentrations of HCH isomers in Manzala Lake were in the following order: α -HCH > γ -HCH > β -HCH > δ -HCH, which is similar to that in technical HCH preparations containing 55–70% α -, 10–18% γ -, 5–14% β - and 6–10% δ - isomer ^[7]. Among these, the β -isomer is the most persistent in the environment, while the α -isomer is more soluble in water. Prevalence of β -HCH isomer in water samples from the Nile River in Cairo suggests its leaching from soils contaminated by the past inputs. Use of HCH has been restricted in Egypt since the 1970s. However, the pattern of HCH isomers in Manzala Lake resembling technical HCH preparations suggests recent usage. Al-Mansoura, a catchment area of Manzala Lake, represents one of the most productive agricultural areas in the Nile Delta. HCH may still be used in this region for agricultural purposes.

	Nile River				Manzala Lake					
	Cl	C2	СЗ	M1	M2	М3	M4	M5		
hexachlorobenzene	4.3	4.0	3.8	86	158	37	81	30		
α-НСН	71	65	74	583	2,815	727	553	482		
β-нсн	75	70	114	321	99 1	349	304	226		
ү-НСН	65	45	47	487	1,408	467	462	324		
δ-НСН	5.8	4.3	5.0	74	167	87	72	61		
<i>p,p</i> ′-DDT	10	5.8	8.1	19	5.8	12	18	15		
p,p'-DDE	22	21	21	218	122	83	207	27		
p,p'-DDD	32	16	37	230	232	131	218	46		
c-chlordane	4.9	3.9	4.7	5.3	33	13	5.1	1.8		
t-chlordane	4.9	4.2	6.3	5.3	35	14	5.1	2.2		
c-nonachlor	n.d.	n.d.	n.d.	4.1	2.9	4.4	3.9	n.d.		
t-nonachlor	5.0	2.1	3.3	2.5	1. 9	2.7	1.8	n.d.		
heptachlor	4.5	3.8	1.9	n.d.	7.8	2.6	n.d.	n.d.		
oxychlordane	2.4	1.5	1.2	14	3.6	4.5	11	3.1		

TABLE I Organochlorine concentrations (pg / L) in filtrated water samples collected from Nile River and Manzala Lake in 1993

n.d. < 1 pg/L

1	773							
		Nile Rive	r		Ма	inzala L	ake	
	C1	C2	СЗ	M1	М2	M3	M4	М5
	2.6	1.7	1.9	11	21	4.3	7.8	2.5
	3.1	1.6	2.6	77	325	83	64	23
	4.58	2.6	5.4	54	257	51	21	38
	1.82	1.4	1.5	65	163	56	63	45
	0.31	0.28	0.68	6.3	25	5.6	2.1	3.1
	16	4.2	3.0	8.7	29	3.5	1.2	1.3

44

12

4.1

5.3

n.d.

1.9

1.7

2.5

138

37

17

22

2.6

6.2

3.0

2.5

12

4.7

6.7

14

n.d

1.3

2.6

1.3

5.3

2.4

3.5

5.9

2.5

2.3

1.9

1.5

8.9

2.2

1.1

3.3

2.1

0.23

0.15

0.69

TABLE II Organochlorine concentrations (pg / L) in suspended solid samples collected from Nile River and Manzala Lake in 1993

31

11

7.8

8.0

1.0

3.4

2.6

1.2

25

4.6

5.0

6.0

n.d.

2.8

1.6

0.89

17

3.9

6.2

6.4

n.d.

2.6

3.6

2.1

oxychlordane n.d. < 0.1 pg / L

hexachlorobenzene

α-HCH

β-HCH

γ-HCH

δ-HCH

p, p'-DDT

p, p'-DDE

p,p'-DDD

c-chlordane

t-chlordane

c-nonachlor

t-nonachlor

heptachlor

Among all sampling locations in Manzala Lake, the highest concentrations of pesticides were detected at location M2 (see Figure 2) where two main drainages, Bahr El-bakar and Bahr Hados accounting for 75% of the drainages in Manzala Lake, are located. Water concentrations of almost all pesticides decreased linearly from M2 to M5. The location M5, located near Gamil, is the main outlet of lake water to the Mediterranean Sea. Egyptian National Environmental Action Plan reported that Bahr El-bakar and Bahr Hados drainages were the major sources of heavy metals and nutrients in Manzala Lake ^[8]. High incidences of gill diseases and internal parasites in fish collected from this area have been attributed to pollution by contaminants ^[9].

Concentrations of organochlorine pesticides in suspended solids and its contribution to total concentrations in water are shown in Tables II and III, respectively. Proportions of CHLs in suspended solids were similar to that in dissolved phase. However, there were some differences in the proportions of organochlorine pesticides in dissolved and particulate phases, depending on sampling locations. Proportions of HCHs in particulate phases were 3 times higher in Manzala Lake than those from the Nile River in Cairo. On the other hand, DDTs and HCB were predominant in dissolved fractions in Manzala Lake than those from the Nile River. This may suggest a lack of equilibrium in organochlorine pesticide concentrations in water. In addition, plankton productivity, organic carbon, resuspension of sediments and octanol-water partitioning coefficients can influence the partitioning of organochlorines between suspended and dissolved phases.

	Nile River			Manzala Lake					
	CI	C2	СЗ	M]	М2	М3	M4	М5	
hexachlorobenzene	37	30	33	11	12	10	8.8	7.7	
α-ΗCΗ	4.2	2.4	3.4	12	10	10	10	4.6	
β-нсн	5.8	3.5	4.5	14	21	13	6.3	14	
ү-НСН	2.7	3.0	3.1	12	10	11	12	12	
δ-НСН	5.1	6.1	12	7. 9	13	6.1	2.8	4.9	
<i>p</i> , <i>p</i> ′-DDT	54	42	27	31	83	23	6.4	8.1	
p,p'-DDE	59	54	44	17	53	13	2.5	25	
<i>p,p</i> ′-DDD	26	22	9.6	5.1	14	3.5	1.1	4.6	
c-chlordane	61	56	57	44	34	35	41	38	
t-chlordane	62	59	50	50	38	51	54	60	
c-nonachlor		-	· -	-	47	-	39	~	
t-nonachlor	40	57	44	43	77	32	56	-	
heptachlor	36	30	65	-	28	50	-	-	
oxychlordane	34	37	64	15	41	22	12	18	

TABLE III Percentage of each organochlorine pesticide in the suspended solid fractions of Nile I	River
and Manzala Lake waters	

concentration in suspended solid / (conc. in suspended solid + conc. in water) × 100

Sediments

Residue levels of pesticides in sediments from Manzala Lake are shown in Table IV. Spatial distribution of concentrations of HCB, HCHs, DDTs and CHLs in sediments was similar to those found in filtrated water, which decreased with increase in distance from M2. In contrast to that in filtrated water, p,p'-DDE was the predominant contaminant in sediment. Despite the predominance of α -HCH in water samples, β -HCH was predominant in sediments from Manzala Lake, suggesting a greater potential of β -isomer than α -isomer to persistence.

	Nile River		
	CI	СЗ	
hexachlorobenzene	4.5	3.8	
α-НСН	0.03	0.02	
β-НСН	1.4	0.95	
ү-НСН	0.02	n.d.	
δ-НСН	n.d.	n.d	
p,p'-DDT	8.5	5.6	

TABLE IV Organochlorine concentrations (ng / g dry wt.) in sediment samples collected from Nile River and Manzala Lake in 1993

Ml

0.35

0.21

2.4

0.07

n.d.

26

132

37

1.2

0.37

n.d.

0.31

0.05

0.04

М2

0.60

4.8

8.5

2.7

0.08

79

432

129

18

0.5

0.11

9.4

0.60

0.42

43

32

3.2

0.83

0.42

1.65

0.52

0.67

48

40

6.5

1.2

0.53

2.56

heptachlor 0.93 0.75 oxychlordane

n.d. < 0.01 ng / g

p, p'-DDE

p,p'-DDD

c-chlordane

t-chlordane

c-nonachlor

t-nonachlor

An earlier study has reported the concentrations of organochlorine pesticides in Nile River water collected in 1982 ^[10]. Concentrations of pesticides reported in water were: Lindane (γ -HCH), 0.1 ng/L; p,p'-DDT, 0.9 ng/L; p,p'-DDE, 4.4 ng/L; p,p'-DDD, 2.7 ng/L. These values are remarkably higher than the samples collected in 1993 in this study, except Lindane. Concentrations of DDT compounds decreased by 100-200 fold while Lindane decreased by only 1.4-fold. Although, the differences in the sampling periods, in the particular rainfall [11], and analytical methods may influence concentrations of organochlorines, these results suggest a decline in DDT inputs into the Nile River during the last decade, despite the continued, but limited use of DDT as a rodenticide and termiticide in recent years ^[12].

Fish

Organochlorine pesticides were found in almost all fish from the Nile River and Manzala Lake (Table V). Concentrations of p, p'-DDE was the highest in fish.

М5

0.17

0.28

3.0

0.04

0.02

0.08

3.2

2.1

0.19

0.21

0.07

0.09

0.01

0.02

Manzala Lake

M3

0.10

0.15

4.0

0.18

0.01

2.2

46

21

4.2

1.1

0.08

2.0

0.25

0.2

M4

0.37

0.56

3.5

0.04

0.07

0.08

12

6.1

0.63

0.46

0.13

0.43

0.07

0.06



FIGURE 3 Isomer composition of HCHs in environmental samples from Egypt

Aly and Badawy ^[13] reported residue levels of Lindane (8.9 ng/g), p,p'-DDT (15 ng/g), p,p'-DDE (7 ng/g) and p,p'-DDD (49 ng/g) in fish collected from the Nile River in Cairo during 1981–1982. Compared with those of our results, concentrations of Lindane and p,p'-DDT declined during the last decade. However,

the reduction rate in fish was lesser than that estimated for water. Furthermore, residue levels of p,p'-DDE increased by 9-fold during the last decade. These results seem to suggest continuous exposure of fish to p,p'-DDE from contaminated sediment.

	Nile River			Manzala Lake				
	A	B	M]	М2	M4	M5		
Number	5	5	10	5	10	10		
mean body weight (g)	66	74	33	86	31	27		
fat (%)	5.8	5.0	9.2	3.9	4.1	5.6		
hexachlorobenzene	0.87	0.95	0.79	0.33	0.16	0.28		
α-НСН	0.58	0.43	3.1	1.3	1.7	5.1		
β-НСН	1.8	1.2	5.7	4.0	3.0	7.2		
ү-НСН	0.20	0.15	1.3	0.71	0.56	2.1		
δ-НСН	0.13	0.21	0.78	0.45	0.34	1.3		
<i>p</i> , <i>p</i> ′-DDT	2.2	1.3	2.6	0.33	0.11	0.18		
p,p'-DDE	67	63	61	44	10	7.6		
<i>p</i> , <i>p</i> ′-DDD	26	14	33	17	3.0	6.1		
c-chlordane	2.7	2.1	1.0	0.61	0.06	0.29		
t-chlordane	1.2	0.95	0. 66	0.13	n.d.	0.13		
c-nonachlor	1.4	1.3	0.35	0.65	0.22	0.38		
t-nonachlor	2.3	2.0	0.73	1.0	0.09	0.26		
heptachlor	0.12	0.07	0.16	0.18	n.d.	0.09		
oxychlordane	0.20	0.25	0.17	0.44	0.05	n.d.		

TABLE V Orga	nochlorine	concentrations	(ng / g w	et wt.) in	fish samples	collected f	rom N	lile F	liver
and Manzala La	ike in 1993								

n.d. < 0.01 ng / g i.f.: not quantified because of interference of unknown peak.

Isomer profiles of HCHs and DDTs in fish, dissolved phase, suspended solids and sediment are presented in Figures 3 and 4. The HCH isomer distribution patterns in the suspended particulate and dissolved fractions were similar, whereas the distribution of DDT compounds in the particulate fraction was similar to that in sediment. This reflects the flux of DDT compounds from sediments to suspended particles. However, a clear trend in the composition of CHLs was not discerned (Figure 5)



FIGURE 4 Isomer composition of DDTs in environmental samples from Egypt

Drinking water

Organochlorine concentrations were also measured in drinking water collected from the University of Mansoura (Table VI). HCB, α -, γ -, δ -HCH, p,p'-DDE and *t*-chlordane were detected in drinking water at concentrations similar to that in



FIGURE 5 Composition of CHLs in environmental samples from Egypt

water from Manzala Lake. Tap water collected in Alexandria, Egypt, contained Lindane (0.29 ng/L), p,p'-DDT (0.47 ng/L) and heptachlor (0.12 ng/L) ^[14]. p,p'-DDT and heptachlor were not detected in this study because of interferences from unknown peaks. However, detectable levels of organochlorine pesticides in

drinking water is of concern from human exposure point. Considering an average daily intake of 2 L of water, exposure from water along would contribute to 1.5 to 1.9 ng HCHs/person/day. Intake from other food items including fish which was discussed above will be of further concern. In addition, several ECD-responsive compounds were also noticed in drinking water, for which identities were not examined, since our analytical procedure was optimized mainly for organo-chlorine pesticides.

	1st time	2nd time
hexachlorobenzene	54	42
α-ΗCΗ	580	630
β-НСН	i.f.	i.f.
ү-НС Н	200	250
δ-НСН	67	50
<i>p</i> , <i>p</i> ′-DDT	i.f.	i.f.
<i>p</i> , <i>p</i> '-DDE	140	110
<i>p</i> , <i>p</i> '-DDD	i.f.	i.f.
c-chlordane	i.f.	i.f.
t-chlordane	38	41
c-nonachlor	i.f.	i.f.
t-nonachlor	n.d.	n.d.
heptachlor	i.f.	i.f.
oxychlordane	n.d.	n.d.

TABLE VI Organochlorine concentrations (pg / L) in drinking water samples collected from University of Mansoura in 1994

n.d. < 1 pg / L

i.f. : not quantified because of interference of unknown peak.

CONCLUSIONS

Discharge of HCHs into the Nile River appears to have decreased after the ban on usage in 1978. However, agricultural regions near Al-Mansoura continues to be a source of HCHs in the Nile River Delta. In Manzala Lake, drainages from Bahr El-bakar and Bahr Hados are the main sources of organochlorine pesticides. Organochlorine pesticides in water has decreased during last decade. However, residue levels of p,p'-DDE in fish has increased, and contaminated sediments are the probable source of DDTs in fish. Although the dam constructed across the Nile River in Aswan in 1972 reduced the movement of sediments downstream, and therefore the transport of pollutants, higher accumulation in the Delta sediments and fish was still observed.

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