

ORGANOCHLORINE PESTICIDE RESIDUES IN THE LOWER CHAO PHRAYA RIVER AND KLONGS ALONG THE RIVER AT BANGKOK METROPOLITAN AREA, 1982-1984

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Abstract

Organochlorine pesticides were ubiquitously found in the lower Chao Phraya River and klongs along the river at Bangkok metropolitan area. These pesticides such as BHCs, Heptachlor, Aldrin, Dieldrin and DDTs had the highest frequencies of occurrence in water and sediment samples collected from the river and klongs. The arithmetic averages of these pesticide residues in the river water on April and October 1984 were 0.233 and 0.100 µg/l, respectively. In particular, the concentrations of α-BHC, Aldrin, and Dieldrin in the river water in dry season were found to be 3-10 times higher than those values measured in rainy season. The distancial distribution of these pesticides in the river water was strongly influenced by tidal effect and flow rate of the river in the low part of the Chao Phraya River. For example, the tested compounds were highly concentrated in the river water from the river mouth to 30 km upstreams, while, the compounds were highly accumulated in the river sediment from 30 to 60 km up the river, when the survey was conducted on September 1984. In addition, these pesticide residues in the river sediment were 200-350 times higher than those found in the river water.

Introduction

Organochlorine pesticides, in addition to polychlorinated biphenyls, are world-wide organic pollutants¹⁻⁵. Such compounds are characterized by their stability for long periods and may lead to marked changes in the aquatic ecosystems. Uptake and accumulation of such chlorinated hydrocarbons by sediments, microorganisms, and fish led to the build up of such compounds in the food chain⁶⁻⁷.

Many organochlorine pesticides, including organophosphate insecticides,

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phenoxy acidic herbicides, and carbamate fungicides, have been imported into Thailand for a long time. The ubiquity of organochlorines has created now much interest in Thailand. However, there have been few papers on pesticide pollution levels in the aquatic environment in Thailand⁸⁻¹⁰. Although their occurrence in raw water and finished waters has been confirmed in the four main rivers of Thailand, the levels detected vary widely and are dependent upon locations, time of analysis, and method of analysis.

The Chao Phraya River is one of the most important rivers in Thailand, which covers a basin area of about 162,000 km² and which has been used for recreational purposes and as a source of potable water. The river flows from the Northern valleys through the central plain and into the Gulf of Thailand. The Bangkok metropolitan area is located on the river estuary and is subjected to tidal effects. The present situation of water quality of the river is heavily polluted by industrial wastewater and domestic wastewater. This study was, therefore, designed to evaluate the organochlorine pesticide residues in the lower Chao Phraya River and klongs along the river at Bangkok metropolitan area.

Materials and Methods

*Organochlorine Pesticides**

Organochlorine pesticide standards; α -, β -, and γ -BHC (BHCs), Heptachlor, Heptachlor epoxide, Chlordane, Aldrin, Dieldrin, Endrin, Endrin aldehyde, p,p'-DDT, o,p'-DDT, p,p'-DDE, o,p'-DDE, p,p'-DDD and o,p'-DDD (DDTS) and these mixtures which were assigned in the EPA protocol (Federal Register, vol. 44, No. 233 Dec. 3, 1979), were purchased from Supelco Inc. in USA. These standards were stored at 4°C in refrigerator until preparation of working standard solutions. The working standard solutions were prepared by dilution of the stock with purified methanol.

* α -, β -, γ -BHC (1, 2, 3, 4, 5, 6-Hexachloro - cyclohexane), Heptachlor (1, 4, 5, 6, 7, 8, 8-Heptachloro - 2, 3-epoxy - 2, 3, 3a, 7a - Tetrahydro - 4, 7 - methanoindene), Heptachlor epoxide (1, 4, 5, 6, 7, 8, 8-Heptachloro - 2, 3-epoxy - 2, 3, 3a, 7a - Tetrahydro - 4, 7 - methanoindene), Chlordane (1, 2, 4, 5, 6, 7, 8, 8 - Octachloro - 3a, 4, 7, 7a-tetrahydro - 4, 7 methanoindene), Aldrin (1, 2, 3, 4, 10, 10 - Hexachloro - 1, 4, 4a, 5, 8, 8a - hexahydro - 1, 4, 5, 8 - endo, exo - dimethanonaphthalene), Dieldrin (1, 2, 3, 4, 10, 10 - Hexachloro - 6, 7-epoxy - 1, 4, 4a, 5, 6, 7, 8, 8a, octahydro - endo, exo - 5, 8, - dimethanonaphthalene), Endrin (1, 2, 3, 4, 10, 10 - hexachloro - 6, 7 - epoxy - 1, 4, 4a, 5, 6, 7, 8, 8a - octahydro - 1, 4, 5, 8 - endo - dimethanonaphthalene), Endrin aldehyde (1, 2, 3, 4, 10, 10 - hexachloro - 6, 7 - epoxy - 1, 4, 4a, 5, 6, 7, 8, 8a - octahydro - 1, 4, 5, 8 - endo - dimethanonaphthalene, 9 - carboxaldehyde), p, p' - DDT (p, p' Dichlorodiphenyl trichloroethane o, p' - DDT), o, p' Dichlorodiphenyl trichloroethane, p, p' - DDE (p, p' - Dichlorodiphenyldichloro - ethylene), o, p' - DDE (o, p' - Dichlorodiphenyldichloro - ethylene), p, p' - DDD (Dichlorodiphenyldichloro - ethane) o, p' - DDD (o, p' - Dichlorodiphenyldichloro - ethane).

Other Chemicals

All of the organic solvents and inorganic chemicals used in this work were pesticide residue grades.

Sample Collection

On April and October 1984, water samples were collected from 32 sampling sites of the lower Chao Phraya River as shown in Fig. 1, according to the procedure outlined in Standard Methods¹¹⁻¹⁵, with minor modifications. In each sampling station, the water samples were collected at the middle layer below the water surface in the middle of the river, using Ditto type water sampler. The collected samples of 2.5 l glass bottles with screw caps were transported to 5°C storage room of the ONEB laboratory until analysis.

Additional samples of water and sediment were collected from 20 sampling sites of the lower Chao Phraya River (river mouth to 142 km up the river) and selected klongs at the metropolitan area, as shown in Fig. 2, on September and October 1984. Only the top 10 cm depth of the bottom sediment layers were collected in this work using the Ekman-Berge Dredge. All collected sediment samples were put in ice box for transportation to the ONEB laboratory and then stored in a 5°C room until analysis.

Pesticide Extraction and Clean Up Procedures:

Procedures for pesticide extraction from the water and sediment samples and their clean-up methods were the same as described in literatures^{12, 13}.

Organochlorines in water samples (500 ml) were extracted with 15% diethyl ether in n-hexane (60 ml, 3 times) using a 1000 ml separatory funnel. The combined organic layers were dehydrated on 10 g of anhydrous sodium sulfate for 1 h. After concentration of the organic layers to 2 ml using a Kuderna-Danish (K.D.) concentrator, 3 μ l of the samples were injected into gas chromatograph for identification and determination of organochlorines in the water samples. The extracts were further cleaned up using Florisil column chromatography, if necessary.

Organochlorine pesticides in the sediment samples (10 g) were extracted with 50 ml of mixed solvent of n-hexane and acetone (9:1 in v/v) by shaking overnight, after grinding the dried sediments into tiny pieces and passing them through a No. 20 sieve with a pore size of 0.84 mm. The n-hexane layers were pipetted from the sediment samples and the sediments were rinsed again with 10 ml of n-hexane. The combined organic layers were dehydrated on 10 g of anhydrous sodium sulfate for 1 h. After concentration of the n-hexane layers to 2 ml using the K.D. concentrator, 3 μ l of the extracts were injected into a gas chromatograph for identification and determination of organochlorines in the sediment samples. The extracts were further cleaned up using Florisil or Silica Gel column chromatography, if necessary.

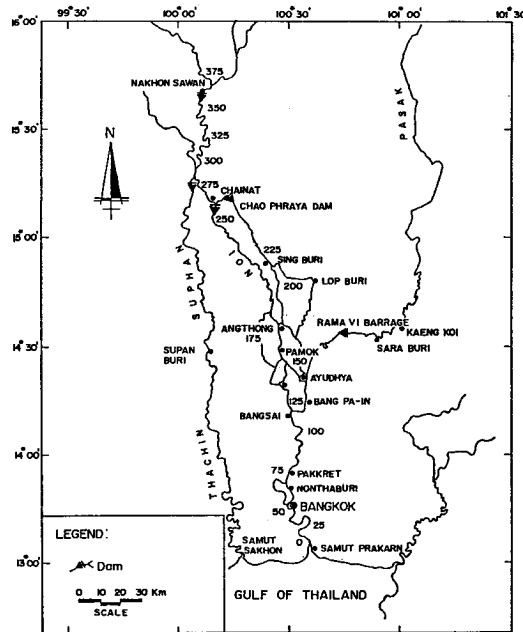
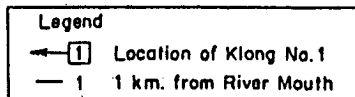


Fig. 1 Map of the Lower Chao Phraya River System.



List of Selected Klongs

- | | | |
|----|------------------|------------|
| 1 | Bang - Nangkrong | (21.5 km.) |
| 2 | Samrong | (23.5 km.) |
| 3 | Bang - Na | (25.5 km.) |
| 4 | Phra - Kanong | (28.5 km.) |
| 5 | Lad - Laung | (36.7 km.) |
| 6 | Jang - Ron | (38.6 km.) |
| 7 | Raj - Burana | (39.6 km.) |
| 8 | Bang - Pakok | (40.5 km.) |
| 9 | Bang - Pakasw | (41.2 km.) |
| 10 | Dao - Kanong | (41.6 km.) |
| 11 | Bangkok - Nol | (51.0 km.) |

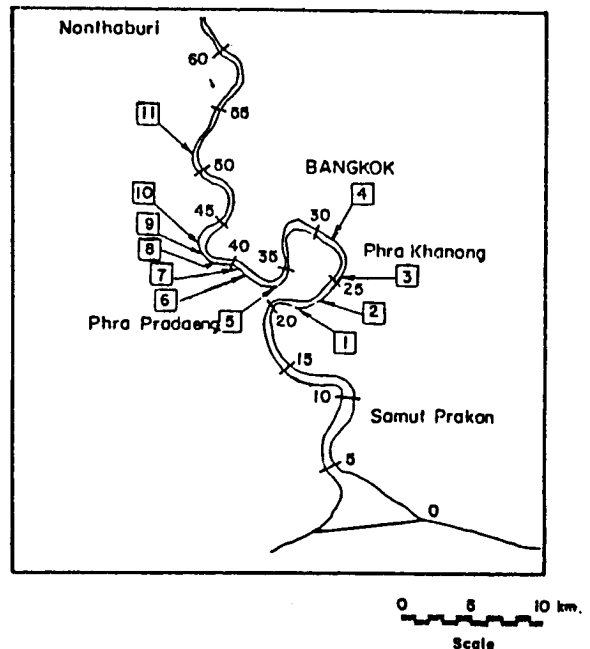


Fig. 2 Locations of Selected Klongs along the Chao Phraya River.

Electron Capture Detector-Gas Chromatography (ECD-GC)

Analyses were performed using a Varian Model 3700 gas chromatograph equipped with a linear⁶³ Ni electron capture detector (ECD-GC). A 2-mm i.d. by 200-cm stainless steel or glass column packed with 1.5% SP2250 + 1.95% SP 2401 on Supelcoport (100-200 mesh), 4% OV 101 + 6% OV 201 on Chromosorb-W-HP (80-100 mesh), and 5% DEGS + 1% H₃PO₄ on Chromosorb-W (AW-DMCS, 60-80 mesh) were used for qualitative and quantitative analyses of organochlorine residues in water and sediment samples. The gas chromatographic conditions and the relative retention times data for organochlorine standards on these columns are shown in Table 1.

Table 1 Relative Retention Times Data of Organochlorine Pesticides on Three Kinds of Column Packing Materials

Organochlorine pesticides	Relative retention times (Aldrin = 1.00)		
	Column I ^a	Column II ^b	Column III ^c
1 α -BHC	0.47	0.48	1.36
2 β -BHC	0.45	0.73	— ^d
3 γ -BHC	0.58	0.64	2.16
4 Aldrin	1.00 (3.10)	1.00 (2.66)	1.00 (2.20)
5 Heptachlor	0.82	0.81	1.05
6 Heptachlor epoxide	1.33	1.54	2.86
7 Chlordan	1.54	1.53	—
8 p,p'-DDE	1.62	2.23	3.32
9 o,p'-DDD	1.80	2.63	6.14
10 o,p'-DDT	2.23	3.19	4.19
11 p,p'-DDD	2.20	3.44	10.05
12 Endrin	2.19	4.76	4.95
13 Dieldrin	1.95	2.43	4.41
14 P,P'-DDT	2.69	4.16	8.95
15 Endrin aldehyde	3.20	—	18.95

^a Column; 4% OV101 + 6% OV210/Chromosorb-W-HP (80-100 mesh) in 2 mm I.D. \times 2 m glass column, oven temperature; 220°C, detector and injector temperatures; 250°C, N₂ flow rate; 60 ml/min.

^b Column; 1.5% SP2250 + 1.95% SP 2401/Supelcoport (100-200 mesh) in 3 mm I.D. \times 2 m stainless column, oven temperature; 200°C, detector and injector temperatures; 250°C, N₂ flow rate; 60 ml/min.

^c Column; 5% DEGS + 1% H₃PO₄/Chromosorb-W (AW-DMCS, 60-80 mesh) in 2 mm I.D. \times 2 m glass column, oven temperature; 200°C, detector and injector temperatures; 200°C, N₂ flow rate; 80 ml/min.

^d Not determined.

Quantitative measurements for water and sediment samples were made using the calibration curves of organochlorine standards. Since the sensitivity of ECD-GC is dependent upon the operating time to time and day to day, calibration curves for organochlorine standards were made at the beginning and end of each operating day. All results were calculated as $\mu\text{g/l}$. Percent recovery for this analysis was 90 percent.

Results and Discussion

Pesticide Residues in the Lower Chao Phraya River on Dry and Rainy Seasons

The first survey was carried out on April and October 1984 during low tide, in order to know the distancial distribution of organochlorine pesticides in this river on typical dry and rainy seasons in Thailand¹⁶. The distancial distribution patterns of organochlorines from the river mouth to 330 km upstream in this river on April and October 1984 are shown in Fig. 3. Comparison of the pesticide residues in the lower Chao Phraya River during dry and rainy seasons of 1984 are also summarized in Table 2.

Table 2 Comparison of Concentrations (in $\mu\text{g/L}$ = ppb) of Organochlorine Pesticides in the Lower Chao Phraya River on April and October 1984.^a

Organochlorine pesticides	April 1984			October 1984		
	Freq ^b	Range	Med.	Freq ^b	Range	Med.
α -BHC	95	0.001 – 0.056	0.022	100	0.001 – 0.035	0.007
β -BHC	5	<0.001 – 0.024	—	0	—	—
γ -BHC (Lindane)	27	<0.001 – 0.021	—	72	<0.001 – 0.032	0.003
Heptachlor	41	<0.002 – 0.015	—	27	<0.002 – 0.166	—
Heptachlor epoxide	32	<0.002 – 0.005	—	18	<0.002 – 0.005	—
Chlordane	0	—	—	0	—	—
Aldrin	100	0.002 – 0.284	0.126	100	0.002 – 0.028	0.008
Dieldrin	100	0.020 – 0.289	0.080	88	<0.005 – 0.442	0.029
Endrin	0	—	—	0	—	—
p,p'-DDE	36	<0.005 – 0.031	—	18	<0.005 – 0.030	—
p,p'-DDD	9	<0.005 – 0.035	—	18	<0.005 – 0.030	—
p,p'-DDT	13	<0.005 – 0.015	—	9	<0.005 – 0.271	—
Σ Pesticides	100	0.058 – 0.605	0.233	100	0.005 – 0.892	0.100

^a The survey was performed at 32 sampling sites from the river mouth to 333 km up the river.

^b Frequency of occurrence in %; — Not available.

α -BHC, γ -BHC (Lindane), Heptachlor and its epoxide, Aldrin, Dieldrin and p,p'-DDT had the highest frequencies of occurrence of over 30% in water samples collected from the lower Chao Phraya River on April and October 1984 (Table 2). Other pesticides detected in this work were β -BHC, p,p'-DDD, and p,p'-DDE. These compounds identified to be present in the river water reflect some species of the imported organochlorine pesticides into Thailand¹⁷. Comparatively high concentrations of organochlorine pesticides in the river water were found at 62 km (Bangkok metropolitan area), 170 km, and 300 km upstream (lower reaches of Nakhon Sawan Dam) on April 1984 (Fig. 3). Higher polluted regions with these pesticides were also observed at 48 and 330 km sampling sites on October 1984.

The arithmetic averages of organochlorine residues in the river water on April and October 1984 were 0.233 and 0.10 $\mu\text{g/l}$, respectively. Especially, the concentrations of α -BHC, Aldrin, and Dieldrin in the river water collected on typical dry season in Thailand were found to be 3-10 times higher than those observed on rainy season (Table 2). The difference in the concentrations of organochlorines in the river water when the surveys were done on dry and rainy seasons can be explained by the dilution of these pollutants by a heavy rain water during the rainy season in Thailand¹⁸.

The possible presence of taste- and odour-products or toxic organic contaminants, even in low concentration, in river water and drinking water is of concern to the consumer. Concentrations of organochlorine pesticides detected in the lower Chao Phraya River (Table 2) are expressed in terms of 0.233 $\mu\text{g/l}$ or less, and this polluted water has been used as a source of potable water in Bangkok metropolitan area. However, these values correspond to those found elsewhere^{1,19} and are well below published maximum permissible levels for drinking water^{20,21}.

Tidal Effect on Distancial Distribution of Organochlorines in the Lower Chao Phraya River.

Figure 4 shows the distancial distribution of organochlorine pesticides in the lower Chao Phraya River from the river mouth to 142.6 km up the river during low and high tides on September 1982. Relatively high concentrations of organochlorines in the river water were detected at 10 to 30 km upstream during low tide, while these were observed in the 40 to 65 km regions during high tide. Nakamuro *et al.*¹⁸ have demonstrated that water quality of the lower Chao Phraya River is strongly influenced by the tidal effects, especially in Bangkok metropolitan area. This shift in the distribution of organochlorines in the river water from near the river mouth to the upper stations (60 km) may be explained by the tidal effect in the lower Chao Phraya River.

Pesticide Residues in Water and Sediment of the Lower Chao Phraya River

In order to compare the concentrations of organochlorine pesticides in river

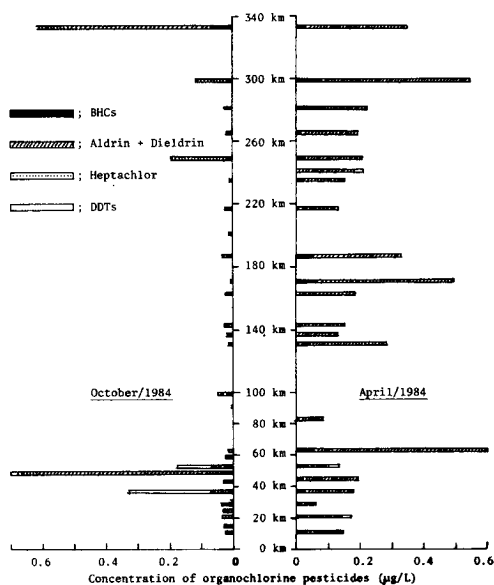


Fig. 3 Distancial Distribution of Organochlorine Pesticides in the Lower Chao Phraya River at Low Tide on April and October 1984

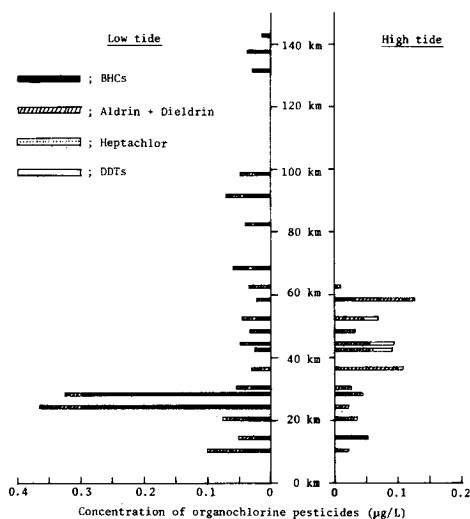


Fig. 4 Effects of High and Low Tides on the Distancial Distribution of Organochlorine Pesticides in the Lower Chao Phraya River on September 1982

water and sediment of the lower Chao Phraya River, these samples were collected at 20 sampling sites from the river mouth to 142.6 km upstream during low tide on September 6, 1984. The distancial distributions of organochlorines in water and bottom sediment at 20 sampling sites are shown in Fig. 5. During this period, Heptachlor, Dieldrin, and DDTs had the highest frequencies of occurrence in the water samples, while, BHCs, Dieldrin, and DDTs were highest in the sediment samples.

It can be seen from Fig. 5 that organochlorine compounds were concentrated in river water from the region of the river mouth to 30 km up the river, with the highest value at 14 km site. However, high concentrations of these pesticides were detected in river sediment from the region of 30 to 60 km upstream, with the highest level at 60 km sampling site. This shift can be explained by the tidal effects in this river and by the precipitation of the suspended solids, which are usually transported from the upper river, in the Bangkok metropolitan area.

The concentrations of organochlorine pesticides in the water samples collected from the lower Chao Phraya River and klongs along the river at Bangkok metropolitan area during September 1982 to October 1984 are summarized in Table 3. As a general trend, the concentrations of organochlorine pesticides in the river water (Table 3) followed the order: Aldrin + Dieldrin > BHCs > DDTs > Heptachlor.

Polprasert *et al.*¹⁰ have reported that DDTs were not detectable in the water samples collected from the lower Chao Phraya River during November 1978 and April 1979. Our results, however, showed that BHCs, Heptachlor, Aldrin, Dieldrin and DDTs were ubiquitously detected in the water samples collected from the lower Chao Phraya River during April and October 1984. It seems that organochlorine pesticide residues in the water of the lower Chao Phraya River (Table 3) are reasonable values, in comparison with those found in river water in USA¹, England¹, and Japan¹⁹, in water of rice paddies in Malaysia²³, and in sea water of the Mediterranean sea²⁴ and Northern Greece²⁵.

Comparison of the concentrations of organochlorine pesticides in sediment samples of the lower Chao Phraya River and klongs along the river at Bangkok metropolitan area are summarized in Table 4, together with those reported in literature¹⁰. As general trend, the concentrations of organochlorine pesticides in river sediments collected from the lower Chao Phraya River (Table 4) followed the order: DDTs > Aldrin + Dieldrin > BHCs > Heptachlor. This order is somewhat different from that found in the river water.

Tayaputch *et al.*⁷ have reported that p, p'-DDE, p,p'-DDD, and p,p'-DDT were detectable in sediment samples of the lower Chao Phraya River, ranging in concentrations of 0.4-0.5, 0.7-37.1, and 0.7-47.9 µg/Kg, respectively. Our results of DDTs measurements in the river sediments (Table 4) are in good agreement with those previously found by Tayaputch *et al.*⁷ and Polprasert *et al.*¹⁰. The

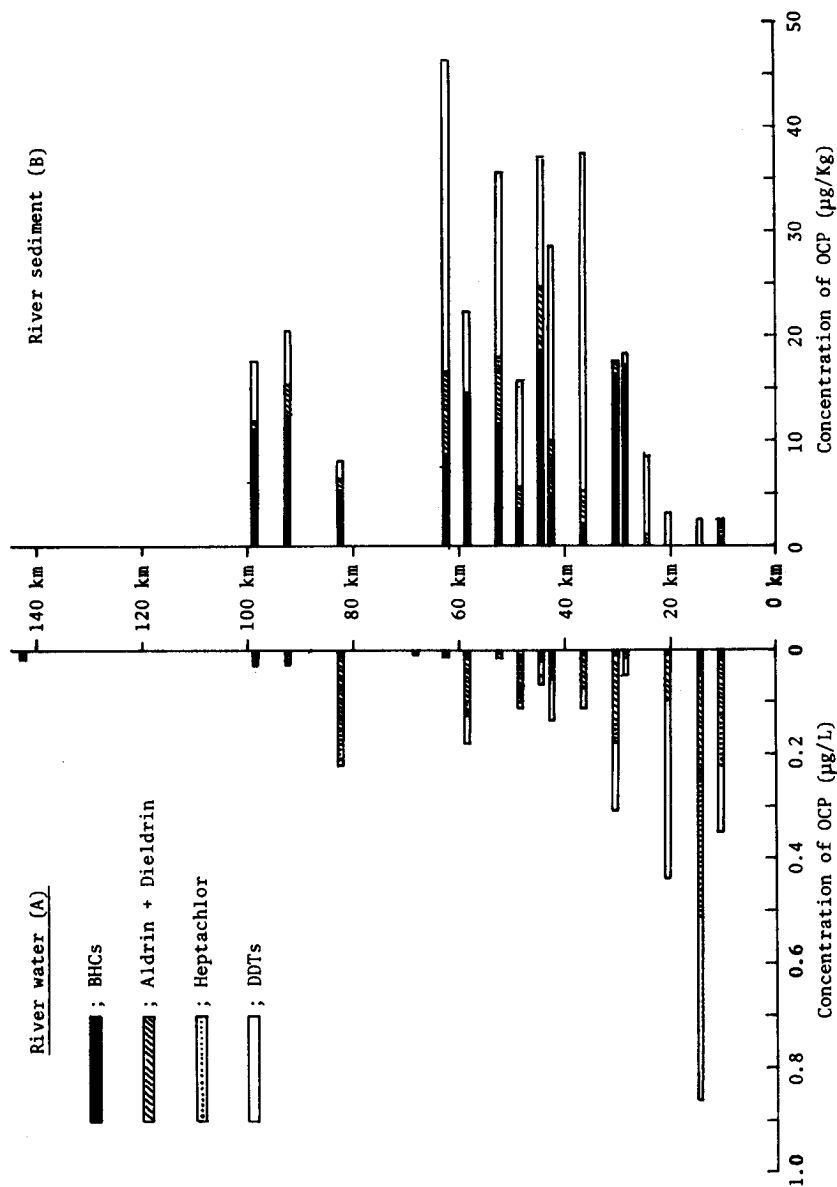


Fig. 5 Distancial Distribution of Organochlorine Pesticides in Water (A) and Sediment (B) Samples of the Lower Chao Phraya River at Low Tide on September 6, 1984

Table 3 Comparison of the Concentrations (in $\mu\text{g/l} = \text{ppb}$) of Organochlorine Pesticides in the Water Samples of the Lower Chao Phraya River and Klongs along the River

Pesticides	Polprasert <i>et al.</i> ⁷			Present Work					
	(1979)			River (1984)			Klong (1984)		
	Freq	Max	Med	Freq*	Max	Med	Freq*	Max	Med
α -BHC	0	—	—	100	0.035	0.007	100	0.072	0.055
β -BHC	0	—	—	0	—	—	0	—	—
γ -BHC	0	—	—	72	0.032	0.003	91	trace	0.001
Heptachlor	0	—	—	27	0.166	—	82	trace	0.001
Hept. epox.	0	—	—	18	trace	—	0	—	—
Chlordane	—	—	—	0	—	—	0	—	—
Aldrin	0	—	—	100	0.028	0.008	100	0.028	0.018
Dieldrin	100	0.117	0.090	88	0.442	0.029	100	0.082	0.046
Endrin	8	0.313	—	0	—	—	0	—	—
o,p'-DDE	0	—	—	0	—	—	0	—	—
p,p'-DDE	8	0.034	—	18	0.030	—	45	trace	—
o,p'-DDD	0	—	—	0	—	—	0	—	—
p,p'-DDD	8	trace	—	18	0.030	—	18	0.040	—
o,p'-DDT	0	—	—	0	—	—	0	—	—
p,p'-DDT	25	trace	—	9	0.271	—	0	—	—

* Frequency of occurrence in %; — non detectable or not available

concentrations of organochlorine pesticides detected in the sediment samples from the lower Chao Phraya River (Table 4) are reasonable values, in comparison to those found in Southern California Coastal sediments²², rice paddies sediments in Malaysia²³, and Mediterranean sea sediments²⁴.

Table 4 Comparison of the Concentrations (in $\mu\text{g/l} = \text{ppb}$) of Organochlorine Pesticides in the Sediment Samples of the Lower Chao Phraya River and Klongs along the river

Pesticides	Tayaputch ¹⁰ (1976) range	Polprasert et al ⁷ (1978) (1979)						Present Work River (1984) Klong (1984)					
		Freq*	Max	Med	Freq*	Max	Med	Freq*	Max	Med	Freq*	Max	Med
α -BHC	—	—	—	—	—	—	—	92	3.30	1.32	100	0.70	0.29
β -BHC	—	—	—	—	—	—	—	0	—	—	0	—	—
γ -BHC	—	—	—	—	—	—	—	92	15.90	8.27	0	—	—
Heptachlor	—	—	—	—	—	—	—	0	—	—	89	100.00	3.50
Hept. epox.	—	—	—	—	—	—	—	17	1.00	—	0	—	—
Chlordane	—	—	—	—	—	—	—	0	—	—	0	—	—
Aldrin	—	0	—	—	9	—	—	53	2.80	1.20	100	16.60	2.78
Dieldrin	—	16	8.00	—	45	430.00	—	94	6.10	2.03	100	15.40	4.20
Endrin	—	0	—	—	36	6.00	—	6	3.00	—	0	—	—
o,p'DDE	—	0	—	—	0	—	—	0	—	—	0	—	—
p,p'DDE	0.4-0.5	63	4.00	1.45	45	5.00	—	62	5.60	3.18	100	1.54	0.71
o,p'DDD	—	0	—	—	0	—	—	35	1.50	—	70	2.00	0.72
p,p'DDD	0.7-37.1	91	13.00	5.00	55	9.00	—	87	18.00	3.43	100	15.00	4.60
o,p'DDT	—	0	—	—	36	2.00	—	0	—	—	0	—	—
p,p'DDT	0.7-47.9	91	109.0	15.00	55	20.00	—	37	19.80	—	50	58.00	—

* Frequency of occurrence in %; — non detectable or not available

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บทคัดย่อ

ศึกษาการป้องกันกำจัดศัตรูพืช และสัตว์ ชนิดออร์กาโนคลอรีน ในแม่น้ำเจ้าพระยาตอนล่าง และคลองที่เชื่อมต่อในเขตกรุงเทพมหานคร พบว่าจำนวนการพบของสารตกค้างประเภทบีเอชซี (BHCs) เฮปตาคลอร์ (Heptachlor), อัลดริน (Aldrin), ดีลดริน (Dieldrin) และดีดีที (DDTs) ในตัวอย่างน้ำและตะกอนดินมีบ่อยครั้ง ปริมาณที่พบในแม่น้ำเจ้าพระยาในเดือนเมษายน และเดือนตุลาคม 2527 มีค่าเฉลี่ย 0.233 และ 0.100 ไมโครกรัมต่อลิตร ตามลำดับ โดยเฉพาะปริมาณความเข้มข้นของแอลฟาบีเอชซี (α BHC) อัลดรินและดีลดริน ในแม่น้ำในฤดูแล้งสูงกว่าฤดูฝน 3-10 เท่า นอกจากนี้จากการศึกษาพบว่าการแพร่กระจายของสารป้องกันกำจัดศัตรูพืชและสัตว์ ชนิดออร์กาโนคลอรีน ขึ้นกับการขึ้นและลงของกระแสน้ำ อัตราการไหลของน้ำและปริมาณการตกค้างของสารดังกล่าวในตะกอนดินมีค่าสูงกว่าในน้ำ 200-300 เท่า