

Residue analysis of organochlorine pesticides in the Mae Klong river of Central Thailand

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Received 27 July 2007; received in revised form 7 December 2007; accepted 7 December 2007

Available online 28 December 2007

Abstract

The most natural waterways in Thailand are utilized for conventional agricultures, e.g. irrigation and wastewater discharge. Concentrations of 20 organochlorine pesticides (OCPs) were measured in surface sediments collected from the tributaries of the Mae Klong river during 2003–2005. The sampling sites chosen were nearby the rice paddy which organochlorine pesticides were intensively used for crop cultivation. Solid–liquid extraction technique was used for sediment extraction, and gas chromatography with electron capture detector was applied for pesticides determination. Total concentrations of OCPs found were ranged from 4.12 to 214.91 $\mu\text{g g}^{-1}$ dw in the first year. In the second year, total residue concentrations in the surface sediments were in the range of 3.26–215.09 $\mu\text{g g}^{-1}$ dw. Concentrations of pesticides were greater in dry-season rice cultivation period in both sampling years, and heptachlor epoxide was the contaminant found at highest concentration in this study. In addition, the occurrence of some selected contaminants including DDT, DDD and DDE reflects the recent contamination in the study area. This demonstrates not only that the natural waterway was polluted and contained much pesticide residues, but it also illustrates the necessity of a continuous long-term monitoring of the affected environment.

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Keywords: Organochlorine pesticides; Mae Klong river; Distribution; Sediment; DDT; DDE; Heptachlor; HCHs; Thailand

1. Introduction

Organochlorine pesticides (OCPs) are notorious among the major type of pesticides, notorious for their high toxicity, persistence in the environment and their ability to bioaccumulate in food chain [1,2]. Although bans and restriction on the usage of some OCPs in developed countries were enacted during the 1970s and 1980s, some developing countries are still using them for agricultural and public health purposes because of their low cost and versatility in controlling various pests [2,3]. These compounds can enter aquatic environment through effluent release, atmospheric deposition, runoff and other means [3,4]. Because of the low water solubility, OCPs have a strong affinity for particulate matter. As a result, sediments can serve as an ultimate sink for them [5–7].

Thailand is a tropical country, which is located between 5°40'N and 20°30'N latitude and between 97°70'E and 105°45'E

longitude. The climate is dominated by tropical monsoons, with daily average temperatures ranging from 25 to 40 °C and the humidity is around 56–79%. Generally, there are three seasons throughout the year: a dry season from March to June, a rainy season from July to October, and a cool season from November to February [8]. In last three decades, the agricultural areas were expanded which led to greatly increased pesticides consumption in Thailand [9]. OCPs including DDT, hexachlorocyclohexane (technical HCH), lindane, dieldrin and aldrin were all imported and applied to farmland. These OCPs have produced its share of pesticide poisoning and wider environmental contamination, especially in the agricultural sector [10].

The Chao Phraya Delta, an important center of rice production for some centuries, comprises three major river basins, the Chao Phraya, Tha Chin and Mae Klong river basins. The Mae Klong river basin covers a total area of 30800 km² in the west central of Thailand. The delta is very flat and periodically flooded in the rainy season [11]. The Mae Klong river, which is the main waterway in this basin, ends at the Gulf of Thailand in Samut Songkhram Province, and has a length of 132 km. It receives water from both the irrigation canal systems and natural

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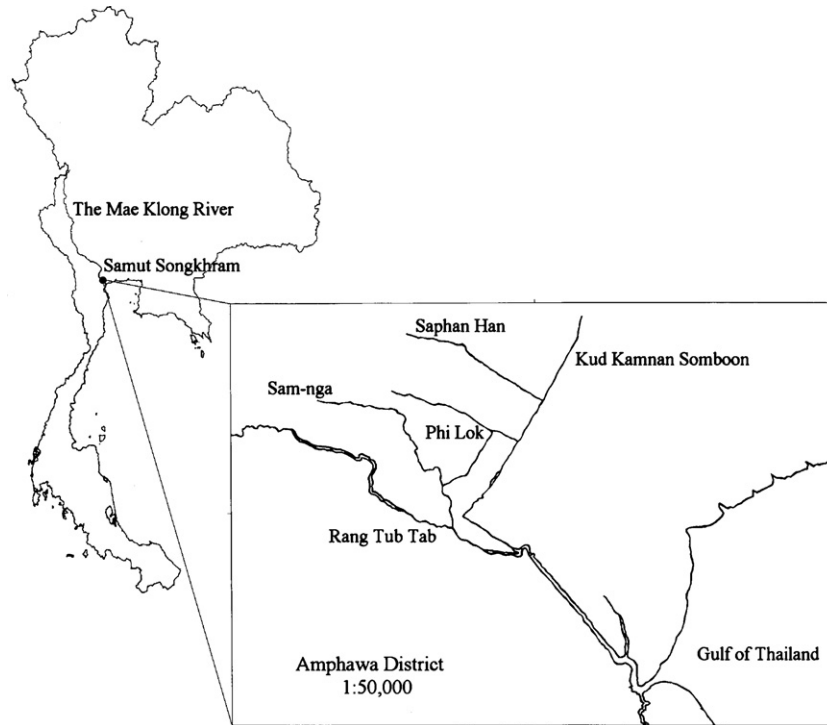


Fig. 1. Map of Thailand showing the five tributaries of the Mae Klong river in the study area.

tributaries, which flow through agricultural areas (rice paddies and orchards) where OCPs are intensively used to improve crop yields. The discharge of untreated wastes from agriculture is a major source of OCP contamination in this area, and has been a major concern to citizens and officials for more than two decades [8,12].

The objective of this study was to determine the contamination status and spatial distributions of 20 OCPs in surface sediments from tributaries of the Mae Klong river.

2. Material and methods

2.1. Sampling sites

The sampling sites were on the Mae Klong river tributaries (Figs. 1 and 2). These tributaries are also major sources for household and industrial water supply in Samut Songkhram Province. The areas surrounding the sampling sites are a traditional rice-cultivation area in which farmers cultivate rice twice per year. The first crop (rainy-season rice) cultivation typically starts in August and harvested in November or December. An irrigate dry-seasons crop is planted in April or May and ends in July or August. Five sites were chosen in such a way as to include hotspots of pollution discharges along the Mae Klong river such as industrial regions, paddy fields and orchards for the first year. For the second year of monitoring, sediment samples were collected from Rang Tub Tab canal (site R) and Phi Lok canal (site P), which is an important crossing point of several tributaries in study area. Site R is also the tributary connected to the Gulf of Thailand. Sediment samples were collected monthly for a period of 2 years, during August 2003–February 2005, as shown in

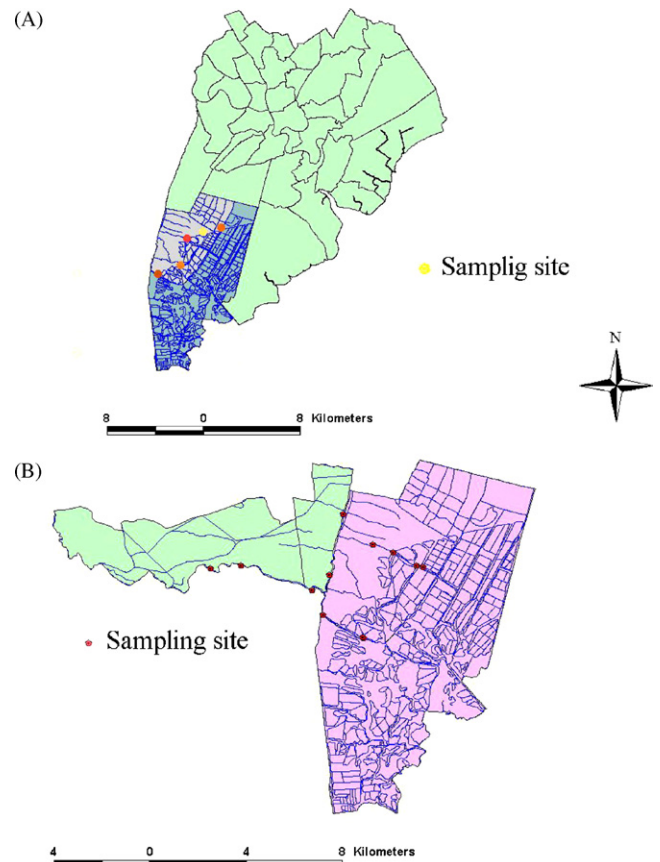


Fig. 2. The study areas and sampling locations along the selected tributaries during August 2003 to July 2004 (A) and March 2004 to February 2005 (B).

Table 1
Sampling areas and codes of the tributaries of Mae Klong river, their coordinates and the basic physicochemical properties of natural surface sediment samples within the area

Sampling year	Sampling sites	Code	Geographical coordinate	Water content (%)	pH (%)	Sand (%)	Silt (%)	Clay (%)
2003	Kud Kamnan Somboon	K	(598863N, 1475923E)	50.79	5.9	20	24	56
	Saphan Han	SP	(598450N, 1475000E)	64.15	5.8	30	21	49
	Phi Lok	P	(595992N, 1475052E)	56.88	5.7	25	23	52
	Sam-nga	SN	(595235N, 1472857E)	64.41	5.6	32	20	48
	Rang Tub Tab	R	(593496N, 1472125E)	61.79	5.8	29	30	41
2004	Phi Lok	P	(595986N, 1475040E; 594718N, 1471496E; 595133N, 1475374E)	59.34	5.7	24	23	53
	Rang Tub Tab	R	(588357N, 1474380E; 589628N, 1474503E; 592581N, 1473464E; 593310N, 1474090E; 593059N, 1472441E; 593899N, 1476626E)	42.44	5.8	29	30	41

Table 1. A total of 180 and 324 surface sediment samples were collected during 2003–2004 and 2004–2005, respectively.

2.2. Sample collection

The surface sediments (upper 10–20 cm) were collected with a stainless steel grab sampler and then placed into chemically clean glass bottles. All the samples were immediately sealed and kept at 4 °C during transportation to the laboratory and stored at –20 °C until analysis. The samples (50 g wet weight) were freeze-dried, homogenized and pass through 2 mm sieve. All the equipment used for sample collection, transportation, and preparation was free from organochlorine contamination.

2.3. Sample extraction

Sediment samples were extracted by acetone and hexane as described below. In the first cycle the sample was extracted with 100 mL acetone for 6 h on a mechanical shaker. In the second cycle, 100 mL of hexane:acetone (1:1 v/v) was used for 1 h before allowing the layers to separate. The supernatant was transferred into a 500 mL separatory funnel, dried by passing through a layer of anhydrous sodium sulfate, and then concentrated to 2 mL by a rotary evaporator (Strike 102/202, Steroglass, Perugia, Italy). The extract was redissolved in 5 mL *n*-hexane and cleaned by filtering through Minisart filters (0.45 µm sterile-EO, non-pyrogenic, hydrophilic, Sartorius AG, Goettingen, Germany). After filtration, the extract was evaporated nearly to dryness under a gentle stream of nitrogen. Consequently, the sample was dissolved in 1 mL of *n*-hexane for GC determination.

All glassware in this study was washed with different solvents in the order of water, acetone and *n*-hexane prior to use.

2.4. Sample analysis

The quantification of pesticide was performed by injecting 1 µL of each sample into a gas chromatograph equipped with a ⁶³Ni electron capture detector (GC-µECD Agilent Technology 6890 N, USA) and a moving needle type injection system

(Agilent Technology 7683 series injector, China). The column consisted of a DB-5MS fused silica capillary column (30 m length × 0.25 mm i.d. × 0.25 µm film thickness, J&W Scientific Co., Folsom, CA, USA). The column temperature was programmed from 50 °C at a rate of 25 °C/min to 100 °C, held for 1 min, and then at a rate of 5 °C/min to 300 °C, held for 7 min. The temperature of the injector and detector were 250 and 300 °C, respectively. Helium was used as a carrier gas, while nitrogen was the make-up gas. OCPs concentrations were quantified by comparing the individual peak area of the samples with the corresponding peak area of the external standards (organochlorine pesticides mixture # 1-503, Lot 315-98 A, Chemservice, PA, USA).

2.5. Quality assurance

Before analysis was performed, standards were run to check for the column performance, peak height, resolution, and the detection limit. The analyte recovery experiment was performed with uncontaminated sediment taken from Salaya campus, Mahidol University, Nakhonpathom Province, Thailand. The experiment was spiked with known quantities of standards then processed, and analyzed as described previously. The method of detection limits of OCPs were determined as the concentrations of analytes in a sample that give rise to 3:1 signal versus noise value (S/N). With each set of 10 samples analyzed, a procedural blank and a spike sample with standard mixture of OCPs were run in a sequence to check for contamination, peak identification and quantification. The analytical procedure was calibrated against a standard reference material (EPA Method 8081A). The correlation coefficients (*r*) of all the calibration curves of OCPs were all higher than 0.998. The detection limit was 0.001 µg g⁻¹ dw. Mean recovery of OCPs through the analytical procedures were between 82 and 103%.

3. Results and discussion

The variations of the OCPs levels for different rice-cropping seasons in surface sediments from the five tributaries dur-

Table 2

Mean and standard deviation (S.D.) of OCPs ($\mu\text{g g}^{-1}$ dw) detected in the surface sediments in five tributaries of the Mae Klong river during August 2003–July 2004 ($n = 60$ /cultivation period)

Pesticides	Rainy-season rice cultivation ^a					Non-rice cultivation ^b					Dry-season rice cultivation ^c				
	K	SP	P	SN	R	K	SP	P	SN	R	K	SP	P	SN	R
α -HCH	0.05 (0.06)	0.25 (0.18)	0.12 (0.10)	0.25 (0.18)	0.33 (0.08)	0.28 (0.05)	0.28 (0.20)	0.26 (0.11)	0.25 (0.14)	0.30 (0.13)	0.26 (0.09)	0.39 (0.07)	0.35 (0.00)	0.41 (0.04)	0.33 (0.08)
β -HCH	0.11 (0.02)	0.18 (0.02)	0.13 (0.04)	0.17 (0.02)	0.21 (0.08)	0.17 (0.06)	0.20 (0.02)	0.17 (0.03)	0.18 (0.03)	0.23 (0.07)	0.21 (0.10)	0.18 (0.02)	0.15 (0.02)	0.19 (0.03)	0.33 (0.19)
γ -HCH	0.21 (0.05)	0.35 (0.00)	0.26 (0.05)	0.32 (0.06)	0.30 (0.05)	0.23 (0.04)	0.35 (0.00)	0.29 (0.00)	0.28 (0.09)	0.33 (0.00)	0.23 (0.04)	0.35 (0.00)	0.29 (0.00)	0.32 (0.06)	0.31 (0.06)
δ -HCH	0.54 (0.24)	0.91 (0.41)	0.76 (0.72)	0.68 (0.22)	3.31 (1.32)	0.86 (0.81)	1.28 (1.51)	1.15 (1.13)	1.67 (2.49)	1.99 (1.01)	2.64 (1.08)	2.89 (2.13)	1.96 (1.35)	1.85 (1.22)	2.95 (0.95)
Heptachlor	0.06 (0.07)	0.15 (0.10)	0.21 (0.19)	0.13 (0.10)	0.19 (0.14)	0.04 (0.04)	n/a	0.09 (0.07)	n/a	0.25 (0.19)	0.10 (0.04)	0.13 (0.05)	0.15 (0.11)	0.21 (0.12)	0.61 (0.91)
Aldrin	0.22 (0.03)	0.30 (0.06)	0.27 (0.08)	0.29 (0.05)	0.33 (0.17)	0.19 (0.08)	0.27 (0.11)	0.26 (0.03)	0.23 (0.16)	0.34 (0.04)	0.17 (0.10)	0.22 (0.07)	0.20 (0.09)	0.25 (0.11)	0.94 (1.04)
Heptachlor epoxide	11.61 (4.39)	13.99 (5.96)	16.52 (20.78)	9.23 (7.87)	63.22 (24.72)	20.41 (25.67)	20.82 (27.51)	29.79 (38.96)	33.01 (56.12)	58.77 (49.73)	64.46 (29.63)	76.11 (44.67)	55.74 (19.57)	41.76 (26.84)	152.17 (55.50)
γ -Chlordane	0.12 (0.07)	0.19 (0.13)	0.12 (0.08)	0.21 (0.09)	0.15 (0.08)	0.20 (0.02)	0.27 (0.03)	0.18 (0.07)	0.16 (0.12)	0.15 (0.13)	0.19 (0.03)	0.27 (0.03)	0.19 (0.08)	0.28 (0.00)	0.22 (0.09)
Endosulfan I	0.12 (0.00)	0.16 (0.00)	0.13 (0.00)	0.16 (0.00)	0.15 (0.00)	0.11 (0.02)	0.15 (0.03)	0.13 (0.00)	0.16 (0.00)	0.16 (0.02)	0.10 (0.02)	0.17 (0.02)	0.11 (0.03)	0.16 (0.00)	0.12 (0.05)
α -Chlordane	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
Dieldrin	0.04 (0.00)	0.01 (0.03)	0.05 (0.04)	0.02 (0.03)	0.01 (0.03)	0.07 (0.05)	0.03 (0.04)	0.08 (0.07)	0.08 (0.10)	0.10 (0.09)	0.07 (0.06)	0.08 (0.06)	0.07 (0.03)	0.10 (0.07)	0.15 (0.06)
<i>p,p'</i> -DDE	0.08 (0.03)	0.04 (0.08)	0.09 (0.06)	0.06 (0.08)	0.11 (0.04)	0.09 (0.08)	0.17 (0.10)	0.13 (0.09)	0.12 (0.14)	0.16 (0.07)	0.10 (0.03)	0.21 (0.04)	0.12 (0.06)	0.18 (0.08)	0.16 (0.07)
Endrin	0.05 (0.02)	0.06 (0.07)	0.06 (0.06)	0.06 (0.07)	0.06 (0.00)	0.09 (0.07)	0.09 (0.06)	0.06 (0.03)	0.03 (0.04)	0.03 (0.06)	0.06 (0.06)	0.02 (0.03)	0.02 (0.03)	0.06 (0.05)	0.09 (0.18)
Endosulfan II	0.03 (0.02)	n/a	0.05 (0.04)	0.01 (0.03)	0.05 (0.04)	0.03 (0.03)	n/a	0.01 (0.03)	0.04 (0.02)	0.01 (0.03)	0.14 (0.25)	0.03 (0.07)	n/a	0.05 (0.04)	0.07 (0.05)
<i>p,p'</i> -DDD	0.01 (0.02)	0.06 (0.12)	0.03 (0.03)	0.02 (0.03)	0.02 (0.03)	0.13 (0.05)	0.03 (0.04)	0.03 (0.03)	0.03 (0.06)	n/a	0.14 (0.10)	0.13 (0.27)	n/a	0.11 (0.18)	n/a
Endrin aldehyde	0.13 (0.06)	0.16 (0.11)	0.15 (0.06)	0.20 (0.08)	0.09 (0.11)	0.14 (0.09)	0.06 (0.08)	0.16 (0.07)	0.08 (0.10)	n/a	0.18 (0.14)	0.43 (0.34)	0.25 (0.12)	0.27 (0.12)	0.91 (1.29)
Endosulfan sulfate	0.07 (0.06)	0.13 (0.03)	0.08 (0.05)	0.15 (0.04)	0.15 (0.03)	0.11 (0.05)	0.08 (0.08)	0.17 (0.08)	0.07 (0.10)	0.38 (0.46)	0.15 (0.01)	0.21 (0.07)	0.19 (0.02)	0.19 (0.02)	0.58 (0.51)
<i>p,p'</i> -DDT	0.13 (0.08)	0.17 (0.12)	0.14 (0.13)	0.26 (0.11)	0.08 (0.05)	0.26 (0.07)	0.31 (0.13)	0.25 (0.05)	0.38 (0.05)	0.20 (0.14)	0.37 (0.11)	0.35 (0.08)	0.22 (0.10)	0.37 (0.03)	0.63 (0.74)
Endrin ketone	0.02 (0.02)	n/a	0.02 (0.03)	0.01 (0.03)	n/a	0.07 (0.13)	0.04 (0.06)	0.06 (0.12)	0.09 (0.15)	0.50 (0.99)	0.19 (0.07)	0.28 (0.30)	0.11 (0.03)	0.14 (0.08)	0.82 (0.93)
Methoxychlor	0.10 (0.05)	0.11 (0.10)	0.12 (0.09)	0.13 (0.07)	0.16 (0.07)	0.18 (0.07)	0.15 (0.11)	0.20 (0.07)	0.14 (0.17)	0.37 (0.46)	0.23 (0.05)	0.41 (0.31)	0.21 (0.02)	0.27 (0.07)	0.84 (0.94)

n/a means no available.

^a August 2003–November 2003.^b December 2003–March 2004.^c April 2004–July 2004.

ing August 2003 and July 2004 are summarized in Table 2. Tables 3 and 4 show the mean concentration of OCPs residues in the sediment samples taken along site P and site R during March 2004–February 2005, respectively. Despite the official ban and restriction on the usage of some OCPs, pesticide residues were detected in all sampling sites. These OCPs included α -HCH, γ -HCH, β -HCH, δ -HCH, heptachlor and its epoxide, γ - and α -chlordane, aldrin, dieldrin, endrin, endrin aldehyde, endrin ketone, α -, β -endosulfan and endosulfan sulfate, DDT, DDE, DDD, and methoxychlor. The status of OCPs contamination in sediments in this study was compared with those in other rivers and estuaries (Table 5).

The range of total of OCPs concentration in the sediments from five tributaries were 4.78–110.24 $\mu\text{g g}^{-1}$ dw at site K, 5.03–151.27 $\mu\text{g g}^{-1}$ dw at site SP, 4.12–93.04 $\mu\text{g g}^{-1}$ dw at site P, 2.38–125.74 $\mu\text{g g}^{-1}$ dw at site SN and 18.25–214.91 $\mu\text{g g}^{-1}$ dw at site R (Table 2). In the second year of the observation, sites P and R were then chosen for further monitoring. Total residue concentrations of OCPs ranged from 4.56 to 131.61 $\mu\text{g g}^{-1}$ dw at site P and 3.26 to 215.09 $\mu\text{g g}^{-1}$ dw at site R (Tables 3 and 4, respectively). The higher concentration of OCPs in these tributaries indicated the high irrigation of these compounds in the basin. These OCPs are believed to have originated from various pesticide-rich sources, mainly agricultural areas and household. The natural processes of leaching and runoff and irrigation are likely to enhance their transfer to the main course of these tributaries, especially during the rice cultivation season.

Site R was found to be the most contaminated site, with sediments containing a wide variety of pesticides. Its sediments had the highest concentration of heptachlor epoxide, 43.76–204.41 and 2.67–152.76 $\mu\text{g g}^{-1}$ dw in 2003 and 2004, respectively. Other OCPs residues were present very high concentrations. For example, \sum DDT (DDT + DDD + DDE) were found in the range of 0.08–1.83 and <0.001–6.78 $\mu\text{g g}^{-1}$ dw in 2003 and 2004, respectively. The relatively high pesticide levels in this tributary may be explained from the input of pollutant in discharge from agricultural fields. This tributary was connected to and received water from other tributaries so it may contain pesticides from extraneous areas as well as from the immediate surrounding.

OCPs residues in dry-season rice cultivation periods were higher than during the wet season in both years. This meant that some OCPs could be released from the surrounding areas to the tributaries in term of irrigation from paddy fields. Moreover, the still water in dry season is one of the major basis. Decreased OCPs concentrations in the rainy-season cultivation can be explained in term of lower upstream discharge and dilution effects. Where non-agricultural activities in cool season infer to the low levels of the chemicals found in this sampling season.

Usage of some imported pesticides such as DDT was officially banned in 1983 and aldrin and dieldrin were banned in 1988. However, there is the official report presented that these OCPs were parts of organochlorine insecticides active ingredients not included in the ban list. That is why they were still be detected in sediment samples along with their metabolites [8,16,17]. Additionally, the appearance of some OCPs such

Table 3
Mean and standard deviation (S.D.) of OCPs ($\mu\text{g g}^{-1}$ dw) detected in the surface sediments at site P during March 2004–February 2005 ($n = 36$ /cultivation period)

Pesticides	Dry-season rice cultivation ^a	Rainy-season rice cultivation ^b	Non-rice cultivation ^c
α -HCH	0.21 (0.15)	0.32 (0.06)	n/a
β -HCH	0.93 (0.83)	0.12 (0.02)	0.11 (0.02)
γ -HCH	0.18 (0.08)	0.08 (0.13)	0.02 (0.02)
δ -HCH	5.50 (3.32)	1.08 (1.28)	0.64 (0.23)
Heptachlor	0.53 (0.59)	0.04 (0.09)	0.04 (0.03)
Aldrin	0.55 (0.56)	0.01 (0.02)	0.02 (0.01)
Heptachlor epoxide	72.43 (30.90)	19.51 (28.43)	7.66 (3.41)
γ -Chlordane	0.08 (0.05)	0.01 (0.02)	n/a
Endosulfan I	0.07 (0.05)	0.10 (0.05)	0.02 (0.01)
α -Chlordane	n/a	n/a	0.03 (0.01)
Dieldrin	0.07 (0.06)	0.02 (0.01)	0.01 (0.01)
<i>p,p'</i> -DDE	0.66 (0.12)	0.03 (0.01)	0.03 (0.01)
Endrin	0.05 (0.02)	0.01 (0.02)	0.01 (0.01)
Endosulfan II	0.07 (0.04)	0.06 (0.04)	0.02 (0.01)
<i>p,p'</i> -DDD	1.03 (0.63)	0.02 (0.02)	0.01 (0.01)
Endrin aldehyde	0.77 (0.63)	0.03 (0.02)	0.02 (0.01)
Endosulfan sulfate	0.17 (0.07)	0.04 (0.02)	0.01 (0.01)
<i>p,p'</i> -DDT	1.05 (0.85)	0.05 (0.04)	0.02 (0.02)
Endrin ketone	0.72 (0.64)	0.03 (0.02)	0.01 (0.01)
Methoxychlor	0.24 (0.09)	0.10 (0.07)	0.02 (0.01)

n/a means no available.

^a March 2004–June 2004

^b July 2004–October 2004

^c November 2004–February 2005.

as endosulfan and heptachlor in the sediment may indicate continued usage of these pesticides despite the official ban. Illegal smuggling of these pesticides from the neighborhood countries has been reported several times in local newspapers. Furthermore, some pesticides are still use in some developing countries around the tropical belt [5,18], which demonstrates

the degradation persistence of these substances, even in tropical environment [19].

Chlordane is widely used against termite infestation and could be released into the environment primarily from its application as an insecticide [10]. The main components of technical grade chlordane are α - and γ -chlordane, γ -nonachlor and hep-

Table 4
Mean and standard deviation (S.D.) of OCPs ($\mu\text{g g}^{-1}$ dw) detected in the surface sediments at site R during March 2004–February 2005 ($n = 72$ /cultivation period)

Pesticides	Dry-season rice cultivation ^a	Rainy-season rice cultivation ^b	Non-rice cultivation ^c
α -HCH	0.28 (0.15)	0.02 (0.03)	n/a
β -HCH	3.66 (3.29)	0.09 (0.02)	0.08 (0.00)
γ -HCH	0.21 (0.12)	0.05 (0.08)	0.01 (0.01)
δ -HCH	10.09 (6.31)	0.82 (0.94)	0.43 (0.05)
Heptachlor	2.16 (2.61)	0.04 (0.04)	0.02 (0.02)
Aldrin	1.12 (0.74)	0.03 (0.02)	0.00 (0.01)
Heptachlor epoxide	110.39 (56.79)	14.84 (21.82)	4.59 (1.35)
γ -Chlordane	0.14 (0.10)	0.01 (0.01)	n/a
Endosulfan I	0.18 (0.12)	0.06 (0.02)	0.01 (0.00)
α -Chlordane	n/a	n/a	n/a
Dieldrin	0.05 (0.02)	0.02 (0.03)	n/a
<i>p,p'</i> -DDE	0.11 (0.05)	0.03 (0.03)	n/a
Endrin	0.09 (0.07)	0.01 (0.01)	0.01 (0.01)
Endosulfan II	0.10 (0.03)	0.07 (0.03)	n/a
<i>p,p'</i> -DDD	0.17 (0.09)	0.02 (0.01)	n/a
Endrin aldehyde	6.36 (12.38)	0.02 (0.01)	n/a
Endosulfan sulfate	4.62 (7.47)	0.05 (0.03)	n/a
<i>p,p'</i> -DDT	2.99 (2.59)	0.05 (0.02)	n/a
Endrin ketone	1.29 (1.32)	0.03 (0.03)	n/a
Methoxychlor	3.31 (2.95)	0.07 (0.08)	n/a

n/a means no available.

^a March 2004–June 2004.

^b July 2004–October 2004.

^c November 2004–February 2005.

Table 5
Comparison of OCPs concentration (ng g⁻¹ dw) in the sediments in paddy fields, rivers, reservoir or estuaries from other Asian locations

Location	Survey year	∑ HCHs	Heptachlor	Heptachlor epoxide	Endosulfan I	Endosulfan II	Endosulfan sulfate	∑ DDTs	Methoxychlor	References
Caspian Sea	2000–2001	0.004–3.460	<0.0005–0.088	<0.0004–0.186	<0.0003–0.189	<0.0004–0.055	<0.0004–0.170	0.006–13.401	<0.0009–1.476	[13]
Azerbaijan	2000–2001	0.196–3.460	0.002–0.010	<0.001–0.045	<0.001–0.016	<0.001–0.022	0.002–0.027	0.560–13.401	0.003–0.330	[13]
Iran	2000–2001	0.029–0.589	0.0006–0.006	<0.0005–0.037	<0.0004–0.013	<0.0005–0.013	0.0006–0.170	0.057–3.897	0.0015–0.150	[13]
Kazakhstan	2000–2001	0.004–0.260	<0.0005–0.001	<0.0004–0.016	<0.0003–0.014	<0.0004–0.007	0.0004–0.006	0.0115–1.896	<0.0009–0.080	[13]
Russian Federation	2000–2001	0.099–0.807	<0.01–0.088	<0.001–0.186	0.001–0.189	0.001–0.055		0.006–1.865	0.001–1.476	[13]
Jiulong river estuary, South China	1999	0.48–9.00	<0.01–0.07	<0.01–0.08	0.01–0.26	<0.01–3.92	<0.01–10.40	0.01–0.43	1.25–3.13	[14]
Minjiang river estuary, Southeast China	1999	2.99–16.21	0.88–2.77	0.59–2.77	0.57–1.61	1.53–3.12	1.77–8.84	1.57–13.06	1.09–5.76	[15]
Agricultural areas, Northern part of Thailand	1996	ND ^a	ND	ND	1.22–634.90	ND	ND	1.57–599.40	ND	[8]
Agricultural areas, Eastern part of Thailand	1997	ND	0.005–0.297	0.009–11.910	0.011–8.818	ND	ND	0.005–3.349	ND	[8]
Tributaries of Mae Klong river, Thailand	2003	640–6920	<1–1920	2340–204410	50–200	<1–510	<1–1340	80–1830	<1–230	This study
Rang Tub Tab Canal, Mae Klong river basin, Thailand	2004	340–24170	<1–5950	2670–152760	10–320	<1–140	<1–15720	<1–6780	<1–6470	This study

^a No data.

tachlor [20]. In this study, only γ -chlordane was detected (range of >0.001 – $2.91 \mu\text{g g}^{-1}$ dw in sediment samples in 2003; and >0.001 – $0.09 \mu\text{g g}^{-1}$ dw for site P and >0.001 – $0.23 \mu\text{g g}^{-1}$ dw for site R in 2004). Generally, chlordane is metabolized in the environment and in the human body to oxychlordane and it is an unlikely source of exposure of heptachlor epoxide is found in the absence of either oxychlordane or γ -nonachlor.

Heptachlor epoxide was found at the highest concentration in sediments of station R (Tables 2 and 4). The enzymatic epoxidation of heptachlor to heptachlor epoxide in soil and animals is most likely responsible for the significant greater incidence of heptachlor epoxide than heptachlor [21]. This chemical was also extensively used for termite control in Thailand in the past, and addition, was used in seed treatments and control of soil insect such as ants. Although usage of heptachlor was officially banned in 1988, the observed high residue concentrations of heptachlor epoxide in all sampling sites could come either its from the persistence in the environment, or from continued use reflecting either illegal smuggling, or use of old stocks. Heptachlor and its epoxide were also detected in both water samples [17] and agricultural soil samples [16] from the Chao Phraya basin.

Other OCPs including aldrin, endrin, dieldrin, and their metabolites, endrin ketone and endrin aldehyde were found in small amounts in all sampling sites in the Mae Klong river basin. The major targets of these OCPs are sugarcane, corn, cotton and fruit trees which are not the foremost crops in the study area. These OCPs were also found in water and soil in agricultural areas in other parts of Thailand [8,16,17], and studies of groundwater in areas surrounding the Mae Klong basin reported that maximum concentration of aldrin + dieldrin was more than 100 times of standards [16].

For some OCPs, chemical transformation and microbial decomposition may occur, e.g. DDT is transforming to DDE and δ -HCH to α -HCH. However, complete mineralization of organochlorine compounds does usually either not take place, or is an extremely slow process [22].

Composition of DDT metabolites and HCH isomers were examined in detail because their differing composition in the environment could indicate different sources of contamination [4]. In the first year of monitoring, levels of DDTs were found elsewhere, for example, between 0.08 and $1.83 \mu\text{g g}^{-1}$ dw in sediment from site R. As to the individual DDTs, the concentrations in surface sediment were generally found in the order of $\text{DDT} > \text{DDE} > \text{DDD}$ in most sampling sites and sampling seasons in Fig. 3A. The same trend was also reported for the second year (Fig. 3B) except for both site P and R (non-rice cultivation areas), which in DDE showed the highest percentage. DDE was usually present in a higher concentration than DDD, suggesting that the most DDT was transformed into DDE [14]. The higher occurrence of DDD and DDE are the result of DDT degradation and the higher stabilities of the metabolites [7,23,24]. The metabolite DDE is produced from DDT under aerobic conditions, such as occurs in upland soils. On the other hand, DDT is converted to DDD with a half-life of a few days under anaerobic conditions, such as in the aquatic sediment [22]. Microbial degradation of DDTs is generally slow

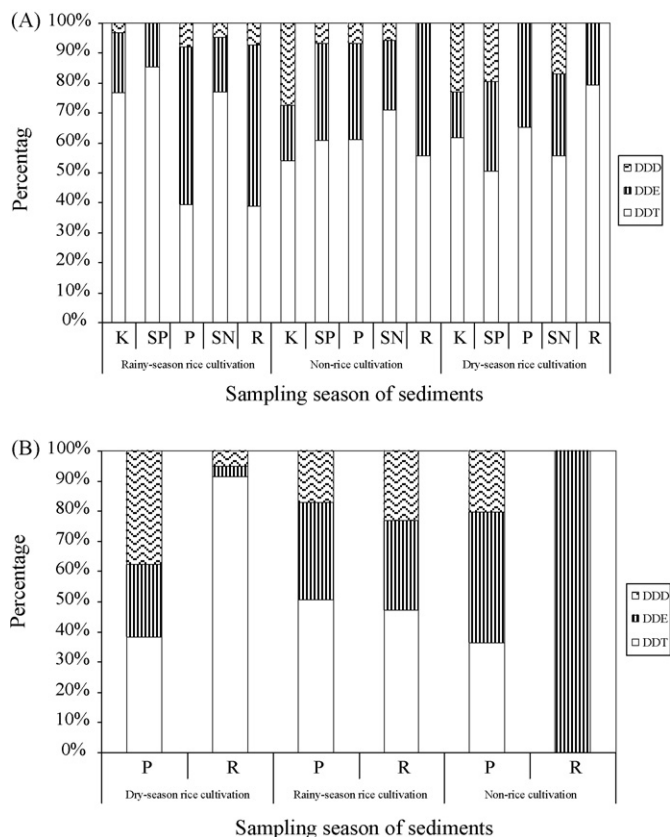


Fig. 3. Consumption of DDTs in surface sediments, during August 2003–July 2004 (A) and March 2004–February 2005 (B).

resulting in environmental persistence of these compounds [25]. The relative concentrations of the parent DDT and its metabolites can indicate whether DDTs input are from the former or present uses [7,18,26]. Ratios of $(\text{DDE} + \text{DDD}) / \sum \text{DDT}$ in surface sediments ranged from 0.21 to $0.61 \mu\text{g g}^{-1}$ dw in 2003 and from 0.10 to $1.00 \mu\text{g g}^{-1}$ dw during 2004 (Fig. 4A and B). Most values were less than 0.5 for the first year, whereas they were greater than 0.5 for the second year. Since ratios of $(\text{DDE} + \text{DDD}) / \sum \text{DDT} > 0.5$ are reported to indicate long-term weathering [18,27–30], this suggest that the DDT compounds in sediments collected in 2003 were from the current use whereas, the 2004 values mostly >0.5 , indicate that degraded metabolites then formed a significant proportion of total DDT compounds. Since the DDD/DDE ratios were less than the unity at both sampling sites, this indicates that the sediment samples were dominated by the products of aerobic degradation. Also, these values assume the ongoing use of DDT and its metabolites were derived under aerobic conditions before transported by surface runoff to the waterways sediment or may be explainable by the relatively higher transportability of DDE than the other form in the atmosphere [3].

Methoxychlor, another analogue of DDT that is widely used both in the field and in a commerce and industry success [31,32] was also found in all sampling sites with concentration between >0.001 – 0.23 and >0.001 – $6.47 \mu\text{g g}^{-1}$ dw in 2003 and 2004, respectively.

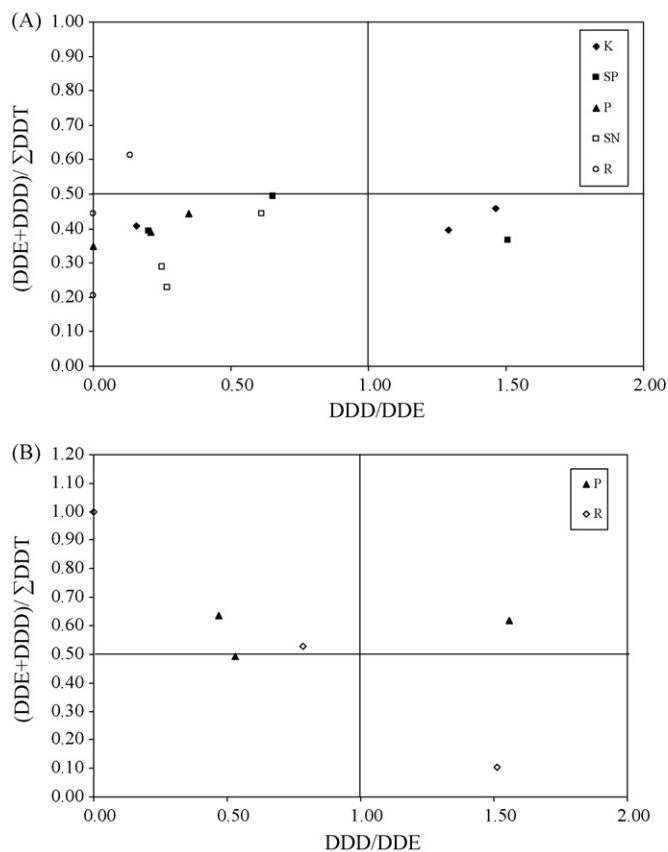


Fig. 4. Relationship between $(DDE + DDD) / \sum DDT$ and DDD/DDE in surface sediments during August 2003–July 2004 (A) and March 2004–February 2005 (B).

HCH residues are among the most widely distributed and frequently detected organochlorine contaminants in the environment [25]. Technical HCH was formerly used as a broad-spectrum pesticide for agricultural purpose in Thailand until banned in 1980 [10] whereas the γ -isomer, commonly known as lindane, (the only isomer with insecticidal activity) used for controlling agricultural and medical pests [33] was banned in 2001. The concentrations of HCH isomers detected in this study in 2003 were in the sequence $\beta\text{-HCH} < \alpha\text{-HCH} < \gamma\text{-HCH} < \delta\text{-HCH}$ in most sampling sites. In 2004, the order was $\alpha\text{-HCH} \leq \gamma\text{-HCH} < \beta\text{-HCH} < \delta\text{-HCH}$. Generally, the most common isomers of HCHs in the environment are $\alpha\text{-HCH}$, $\beta\text{-HCH}$ and $\gamma\text{-HCH}$ [2,15]. Whereas there were high concentrations of $\delta\text{-HCH}$ in sediments in most sampling sites during the present study, it results similarly as detected at Beijing Guanting reservoir, which the possible reasons are still unclear [2]. The typical technical HCH generally contains all four isomers and their physicochemical properties are different. The most stable $\beta\text{-HCH}$, relatively resistant to microbial degradation because it has the lowest water solubility and vapor pressure, and so represents a good indicator of contamination [4,23,33,34]. Many studies have reported that β -isomer was dominant in sediments from river or estuary environment after long-term migration and transformation [27,35,36]. In addition, α -isomer is typically predominant in ambient air as well as ocean waters and may be converted to the β -isomer in the environment [3,4]. However, $\beta\text{-HCH}$

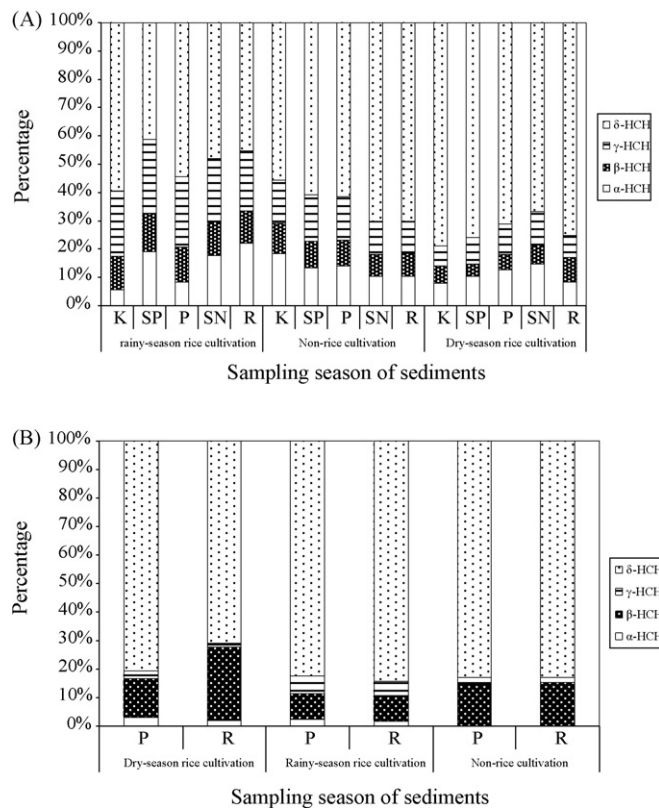


Fig. 5. Consumption of HCHs in surface sediments, during August 2003–July 2004 (A) and March 2004–February 2005 (B).

represented the lowest proportion (7.61% of the total HCHs compared to 11.2 and 12.0%, for α - and β -isomers, respectively) during 2003 in the present study (Fig. 5A). This can be explained by the resistance to chemical degradation of these isomers in this environmental condition and the recent use of technical HCH in this area [34]. Various technical HCH formulations have differing amounts of the α -, β -, and γ -isomers and long range atmospheric transport favors $\alpha\text{-HCH}$, the most volatile isomer [13]. On the other hand, the decreased proportion of $\alpha\text{-HCH}$ and the increased proportion of $\beta\text{-HCH}$ in 2004 (Fig. 5B) implied that the HCHs contamination in the sediment samples might have originated from a relatively distant source or long-term accumulation. Among HCH isomers analyzed, $\gamma\text{-HCH}$ (lindane) contributed 12% and then 2.22% of the total HCH concentrations in sediment samples in 2003 and 2004, respectively. This data indicated the use of technical HCHs rather than lindane and may be explained by the degradation of $\gamma\text{-HCH}$ in this environmental condition because $\gamma\text{-HCH}$ is degraded by microorganisms [37,38] and photochemically isomerized to the α -isomer [34,39]. Although, the predominance of α -isomer in some environmental samples reflects the recent use of technical HCH [40], the higher concentration of α -isomer than γ -isomer in this study may establish the use of lindane in the past. There are no historical data concerning HCHs in this area, thus the data presented here establish a baseline for the future monitoring and management of pesticides in this area.

In the case of endosulfan, the concentration of its α -isomer (the main component of the commercial product), was higher than β -endosulfan (a secondary component) in all sampling sites during both 2 years. Total endosulfan showed values in the range >0.001 – $1.34 \mu\text{g g}^{-1}$ dw in 2003, with 0.05 – $0.20 \mu\text{g g}^{-1}$ dw for the α -isomer and only >0.001 – $0.51 \mu\text{g g}^{-1}$ dw for the β -isomer. During warehouse storage, β -endosulfan is slowly converted to α -endosulfan, which has more toxic insecticidal properties than the β -isomer [31]. Endosulfan sulfate, an endosulfan degradation by-product, was found in large concentration in the sediment samples (range >0.001 to $1.34 \mu\text{g g}^{-1}$ dw). Total endosulfan in 2004 was $22.60 \mu\text{g g}^{-1}$ dw, the highest proportion was endosulfan sulfate (>0.001 – $15.72 \mu\text{g g}^{-1}$ dw). This chemical was extensively used to control golden apple snails in rice fields in this area and was officially banned in 2004. Endosulfan and its by-products were found in relatively high concentrations, both during the dry and rainy-rice cultivation seasons. These data reflects the wide use of endosulfan in the surrounding agricultural areas over a long period.

Aldrin and its analogues (dieldrin and endrin) were also detected in this study. Both aldrin and dieldrin were used to control termites in houses by direct soil injection in the past [41]. Aldrin is rapidly converted to dieldrin in the environment. Their concentration in 2003 ranged between >0.001 – $2.38 \mu\text{g g}^{-1}$ dw for aldrin and >0.001 – $0.17 \mu\text{g g}^{-1}$ dw for dieldrin. The same trend as the first year of monitoring, aldrin concentrations were between >0.001 and $1.81 \mu\text{g g}^{-1}$ dw and endrin concentrations were between >0.001 and $0.17 \mu\text{g g}^{-1}$ dw in 2004. In contrast to the former, endrin, a stereoisomer of dieldrin, was not extensively used for termite control of residences. It is transformed in the environment to metabolites including endrin aldehyde and endrin ketone. In this study, higher concentrations of these metabolites (Tables 2–4) may explained as the result of endrin degradation and the higher stabilities of endrin aldehyde and endrin ketone molecule [42].

Although most organochlorine pesticides found in this study were officially banned in Thailand, the primary substances and their metabolites were still detected in the surface sediments in this area. Regular monitoring and strict law enforcement are needed to develop a strategy to manage the environmental hazards due to these OCPs and to improve environmental protection in this area.

4. Conclusion

This study was the first known analysis of OCPs distributions in sediments from major agricultural areas in Mae Klong basin. The surface sediment in the tributaries of Mae Klong river is polluted by the OCPs. A relatively high level of these compounds were observe in most study areas. Summer showed the highest OCPs residues in both sampling years. Heptachlor epoxide presented the highest concentration among detected OCPs in this study. In addition, \sum DDT and HCHs were found in slightly elevated levels, representing a recent input of these two OCPs into the study area. The concentrations of other detected OCPs that were not as high as found for heptachlor epoxide, were nevertheless of environmental concern.

Acknowledgements

This work was supported by the Post-Graduate Education, Training and Research Program in Environmental Science, Technology and Management (ESTM) under Higher Education Development Project of the Ministry of Education and Mahidol University. It was also supported by and carried out in the framework of the Integrative Research Network of the Tha-chin and Mae Klong river basins. We gratefully acknowledge the staff of ESTM and Tha-chin project for their supports, cooperation and laboratory facilities. We also thank Asst. Prof. Phillip D. Round for proof reading this manuscript.

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