Environmental Occurrence of Organochlorines in the East Coast of Thailand

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The measurements of organochlorine pesticides in mussels and oysters collected along the east coast of Thailand indicate that DDTs, chlordanes (CHLs), hexachlorocyclohexanes (HCHs) and hexachlorobenzene (HCB) are the main organochlorine residues. The results of the study revealed that DDT residues were present in the highest concentration in green mussels and oysters, followed by CHLs, HCHs, and HCB. However, the residue levels in the present study were much lower than those found elsewhere in the tropical coastal water of Asia. Polychlorinated biphenyls (PCBs) were present also in high concentrations in urban and industrial areas. It is interesting that tetra-, penta-, hexa- and hepta-chlorinated isomers were the predominant homologues in both the mussel and oyster samples. The PCB profile in the sediment column from the study area, reflects the active use of PCBs along the east coast of Thailand during the period of 1972–1992. Considering the low levels of organochlorine pesticides (OCs) and PCBs in green mussels and oysters found in this study, the contamination from these organic pollutants in the study area is not particularly serious in the view of human health and ecosystem perspectives.

Key words —— organochlorine, polychlorinated biphenyl, mussel, oyster

INTRODUCTION

Many coastal zones are subjects to considerable pressure from rapidly growing human and industrial development with the risk of contamination from sewage, oils and pesticides to the marine environment.[1)] Organochlorine pesticides (OCs) were widely used in Thailand along with other countries in the region for pest control in agriculture.[2)] Organochlorine pesticides such as hexachlorocyclohexanes (HCHs), chlordanes (CHLs), hexachlorobenzene (HCB) and DDTs are known to be bioaccumulative because of their high lipophilicity and persistency.[3)] In the aquatic environment, organochlorine compounds are removed from the water column and adsorbed on the particulate matters due to their high affinity for organic matter, and ultimately accumulate in sediments, which may play a role as a secondary contamination source. These contaminants also accumulate in the sediment-dwelling organisms and may be transferred to higher tropic levels through the food chain.[4)]

Polychlorinated biphenyls (PCBs) are amongst the most dangerous environmental contaminants due to their persistence, bioaccumulative properties and toxicity. PCBs have been used worldwide as plasticizers, hydraulic and dielectric fluids, fire retardants and paint additives.[5)]

This study was conducted to determine organochlorine pesticides and PCBs residues in green mussel (*Perna viridis*) and oyster (*Crassostrea commercialis*) as a biological indication of the extent of marine pollution along the east coast of Thailand. Moreover, in order to determine the degree of marine pollution in this coastal waters, concentrations detected were compared with reported values from other regions. Organochlorine pesticides and PCBs in sediment core samples were also measured to determine the history of contamination in the area.

MATERIALS AND METHODS

Sample Collection —— Green mussels (*Perna viridis*) and oysters (*Crassostrea commercialis*) were collected from the east coast of Thailand during 2002–2003. More than 30 green mussel with a maximum shell length of 9.5 ± 0.9 cm (age 6 months) and 15 oyster with a maximum shell length of 8.4 ± 1.1 cm (age 7 months) were collected in each location. Adhering matrix was removed in the field. Samples were placed in polyethylene bags in a cooler with dry ice and transported to the laboratory where they were kept in a deep freezer. Frozen samples were thawed, scraped clean and shucked. The soft tissues from all animals were pooled, homogenized, transferred to hexane-rinsed glass bottles and re-fro-

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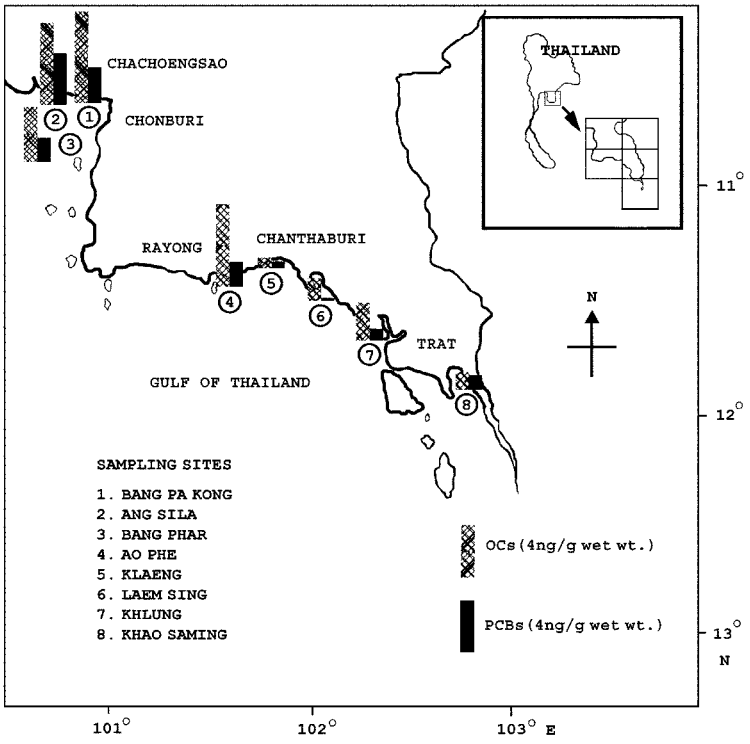

Fig. 1. Distribution of OC and PCB Concentrations (ng/g wet wt.) in Green Mussel from the East Coast of Thailand

zen at –20°C until chemical analysis. The details of sampling site are given in Fig. 1.

Analysis ——— The analytical procedure of OCs in the tissue sample was a modification of the method described by Oh (2000).[6] About 15 g of homogenized sample was dehydrated with activated sodium sulfate. About 10 ng of each internal standard (dibromooctafluorobiphenyl, PCB 103 and PCB 198) was added to the sample. The sample was soxhlet extracted with 200 ml of dichloromethane for 16 hr. The extract was evaporated to about 2–3 ml on a water bath. The condensate was cleaned up by liquid chromatography using silica gel/alumina column. The column was filled with 10 g of alumina and 20 g of silica gel (deactivated with about 1 and 5% water, respectively), and eluted with 100 ml of dichloromethane. The extract was separated by high performance liquid chromatography (HPLC; 250 × 22.5 mm i.d. of size-exclusion column packed with Phenogel 100 A, Phenomenex Co., CA, U.S.A.) using dichloromethane for further clean-up and fractionation. The collection time of OCs fraction was determined based on retention times of dibromooctafluorobihenyl and perlyene in HPLC calibration solution using an UV detector. After stabilization of the retention time of both compounds, the sample was loaded into a HPLC, and OCs fraction was collected. The fraction was concentrated and re-constituted with hexane. The sample was concentrated to 0.5 ml with a gentle stream of N_2 and 10 ng of internal standard (tetrachloro-*m*-xylene) was added.

A Hewlett-Packard 5890 gas chromatograph (GC) with an electron capture detector was used for identification and quantification of OCs in samples. The column used for analysis was a fused silica capillary DB-5 (30 m × 0.25 mm i.d. × 0.25 μm thickness). Helium gas was used as the carrier gas and argon:methane (95 : 5) was used as the make-up gas. The GC temperature was programmed as follows; 100°C (1 min holding time), heated to 140°C at a rate of 5°C/min (1 min holding time), heated to 250°C at a rate of 1.5°C/min (1 min holding time), and heated to 300°C at a rate of 10°C/min (5 min holding time). Total PCB concentration in sample was quantified by summing the concentrations of individually resolved peaks relative to an equivalent mixture of standard with known PCB composition and content (Ultra Scientific Co., U.S.A.).

Spike sample recoveries of PCBs, DDTs, HCHs, CHLs and hexachlorobenzene were in the range of 73–99, 81–112, 85–107, 79–121 and 86%, respec-

Table 1. Concentrations (ng/g wet wt.) of Organochlorine Pesticides in Green Mussel & Oyster from the East Coast of Thailand

Station	PCBs		DDTs		CHLs	
	Mussel	Oyster	Mussel	Oyster	Mussel	Oyster
1	2.70	10.29	3.25	9.95	1.40	1.64
2	3.70	5.31	3.52	5.27	1.00	2.21
3	1.70	1.75	3.64	7.29	< 0.01	0.53
4	1.82	5.37	3.86	16.71	1.21	< 0.01
5	0.99	1.32	1.17	9.35	0.14	< 0.01
6	0.32	3.94	1.64	1.21	< 0.01	0.59
7	0.96	3.63	1.97	5.76	0.47	0.88
8	1.29	1.79	1.37	4.87	< 0.01	0.09
Range	0.32–3.70	1.32–10.29	1.17–3.86	1.21–16.71	< 0.01–1.40	< 0.01–2.21
Mean	1.69	4.18	2.55	7.55	0.53	0.75

Station	HCHs		HCB	
	Mussel	Oyster	Mussel	Oyster
1	0.24	< 0.01	0.05	< 0.01
2	0.49	0.37	0.09	0.09
3	0.27	< 0.01	< 0.01	0.10
4	0.30	0.71	0.06	0.10
5	< 0.01	0.10	0.10	0.04
6	0.44	0.32	0.07	0.04
7	0.40	0.75	0.08	0.23
8	0.22	< 0.01	< 0.01	0.07
Range	< 0.01–0.49	< 0.01–0.75	< 0.01–0.10	< 0.01–0.23
Mean	0.30	0.29	0.06	0.09

PCBs = sum of polychlorinated biphenyl. DDTs = sum of *o*,*p*′-DDE, *p*,*p*′-DDE, *o*,*p*′-DDD, *p*,*p*′-DDD, *o*,*p*′-DDT and *p*,*p*′-DDT. CHLs = sum of α-chlordane, γ-chlordane, *cis*-nonachlor and *trans*-nonachlor. HCHs = sum of α-HCH, β-HCH, γ-HCH. HCB = hexachlorobenzene.

tively. The detection limit was 0.1 ng/g for PCBs and 0.01 ng/g for OC pesticides. The reagent blank analyses were carried out with all bivalve samples. The whole analytical procedure was validated by analyzing reference materials, SRM 1974 from NIST, showing the results to fall within the range of the certified value.

Sediment Dating ——— The chemical procedures described by Nittrouer *et al*. (1979),[7] and Carpenter *et al*. (1982),[8] were used in the Pb-210 radiometric dating of the sediments. Pb-210 activity was determined by alpha spectrometry of its granddaughter Po-210 by assuming that Pb-210 and Po-210 are at secular equilibrium. The analysis was done in collaboration with the Office of the Atomic Energy in Bangkok.

RESULTS AND DISCUSSION

Organochlorine Pesticides Contamination

The concentrations of OCs and PCBs detected in mussel and oyster are given in Table 1. The concentration was the highest for DDT residues in both green mussels and oysters (mean = 7.55 ng/g wet wt. in oyster and 2.55 ng/g wet wt. in mussel), followed by CHLs, HCHs and HCB. Although the usage of organochlorine pesticides such as DDT for agricultural purposes has been banned in Thailand since 1983, it is still being used for malaria vector control along the coast and in neighbouring countries.[9] Considerable residues of *p*,*p*′-DDT in DDTs found in green mussel and oyster from some locations may indicate the presence of significant current sources of DDT in Thailand. However, based on the results shown in earlier studies by Menasveta & Cheevaparanapiwat (1981),[10] Siriwong *et al*. (1991),[2] Ruangwises *et al*. (1994),[9] and present study (Table 2), it appears that DDT concentrations have declined in Thailand coastal waters since the restriction of DDT in 1983. Among the OCs examined, CHLs were detected at a high level (mean = 0.75 ng/g wet wt. in oyster and 0.53 ng/g wet wt. in mussel) next to DDTs (Table 1). While the concentration of HCHs (mean = 0.29 ng/g wet wt. in oyster and 0.30 ng/g wet wt. in mussel) were relatively low,

Table 2. Concentrations (ng/g wet wt.) of Organochlorine Residues in Bivalves Mollusks Collected Worldwide

Species	Location	PCBs	DDTs	CHLs
Blue mussel				
(*Mytilus edulis*)	North, Central and South America, 1986–1993	10–3800	6.7–960	2.8–9.6
	Kattegat, Denmark,1985	3–328	2.4–67	—
	North-West Spain, 1990–1991	nd–620	nd–36	—
Green Mussel				
(*Perna viridis*)	Hong Kong, 1986	49–330	50–520	—
	South India, 1988–1989	0.66–7.1	2.8–40	—
	Coastal, India, 1994–1995	0.31–15	0.93–40	< 0.01–1.9
	Philippines, 1994–1997	0.69–36	0.19–4.2	0.15–9.5
	Coastal water of Thailand, 1979	2–43	32–42	—
	Coastal water of Thailand, 1989	—	0.39–7.41	—
	Coastal water of Thailand, 1991	—	0.74–5.38	—
	Coastal water of Thailand, 1994–1995	< 0.01–20	1.3–38	0.25–5.9
	Coastal water of Thailand, 2002–2003	0.32–3.70	1.17–3.86	< 0.01–1.40
Oyster				
(*Crassotrea gigas*)	Taiwan, 1997	—	nd–131	—
(*Crassotrea commercialis*)	Coastal water of Thailand, 2002–2003	1.32–10.29	1.21–16.71	< 0.01–2.21

Species	Location	HCHs	HCB	References
Blue mussel				
(*Mytilus edulis*)	North, Central and South America, 1986–1993	—	—	(13)
	Kattegat, Denmark,1985	0.6–7.4	—	(14)
	North-West Spain, 1990–1991	nd–49	—	(15)
Green Mussel				
(*Perna viridis*)	Hong Kong, 1986	53–110	—	(16)
	South India, 1988–1989	4.3–16	—	(17)
	Coastal, India, 1994–1995	1.5–12	< 0.01–0.38	(12)
	Philippines, 1994–1997	< 0.01–0.19	< 0.01–0.04	(12)
	Coastal water of Thailand, 1979	—	—	(10)
	Coastal water of Thailand, 1989	< 0.02–0.19	< 0.02–0.31	(2)
	Coastal water of Thailand, 1991	< 0.02–0.09	< 0.02–0.21	(9)
	Coastal water of Thailand, 1994–1995	< 0.01–0.43	< 0.01–0.12	(12)
	Coastal water of Thailand, 2002–2003	< 0.01–0.49	< 0.01–0.10	This study
Oyster				
(*Crassotrea gigas*)	Taiwan, 1997	nd–7.0		(18)
(*Crassotrea commercialis*)	Coastal water of Thailand, 2002–2003	< 0.01–0.75	< 0.01–0.23	This study

with α-HCH detected as the prominant isomer in most locations. These results may be reflective of the past usage of technical HCH in Thailand. Although the usage of technical HCH was banned in the 1980s, the application of γ-HCH (lindane) still appear to be continuing in Thailand.[2)] The detection of γ-HCH as a prevalent isomer in some locations (site 1–3) suggests the continuing usage of lindane in Thailand.

HCB contamination seems to have originated from the usage of fungicides or as an impurity in pesticide formations, a by product of various chlorinating processes and the combustion of industrial and municipal wastes.[11)] The concentration of HCB found in the present study (mean = 0.09 ng/g wet wt. in oyster and 0.06 ng/g wet wt. in mussel) was the lowest of the organochlorine compounds (Table 1) and consistent with values from other studies,[2,9,12)] suggestive of limited usage of HCB in the east coast of Thailand.

The residue of organochlorine pesticides was higher in oysters than mussels (Table 1). Probably this was due to the higher lipid content in oyster tissue ($2.75 \pm 0.45\%$) at about double that in mussel ($1.16 \pm 0.43\%$). Relatively high organochlorine pesticide concentration was detected in mussel and oys-

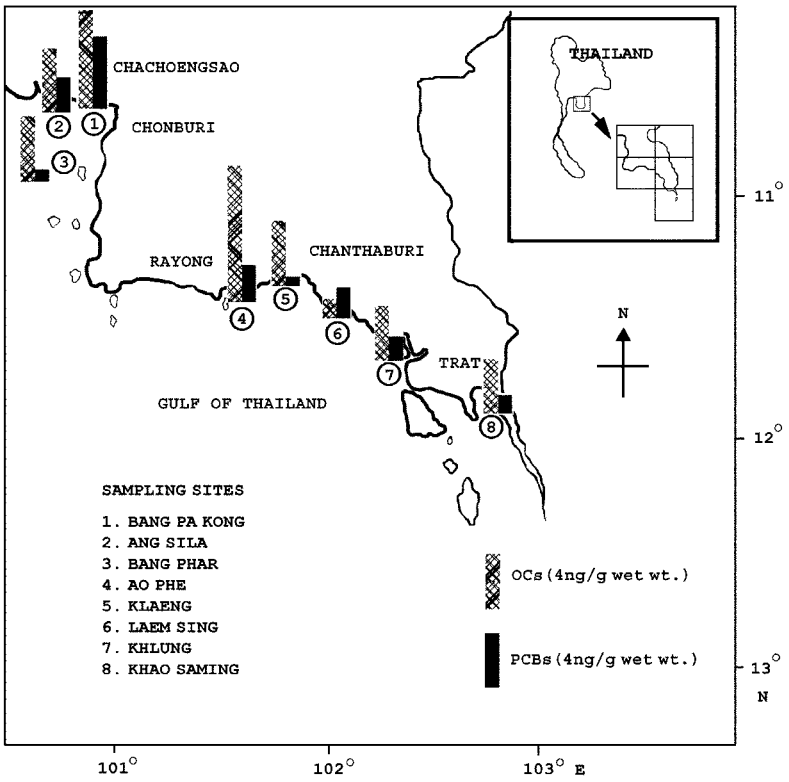

Fig. 2. Distribution of OC and PCB Concentrations (ng/g wet wt.) in Oyster from the East Coast of Thailand

ter samples in the northern parts of this study area (sampling sites 1–4) and generally decreased towards the south (sampling sites 5–8) (Figs. 1 and 2). This reflects greater urban populations and active industrial activities in the northern regions. Whereas in the south, regions such as Klaeng (site 5), Laem Sing (site 6), Khlung (site 7), and Khao Saming (site 8), are more pristine.

The spatial differences in OCs residue levels in green mussels and oysters from several locations along the east Gulf of Thailand suggest that these compounds were widely distributed in the eastern coast of Thailand. Although the agricultural usage of organochlorine pesticides such as DDT and HCH, have been banned in Thailand, they still remain in the aquatic organisms and environments. Figure 3 revealed that in case of the green mussels collected from the east coast of Thailand, it can be observed that OCs contamination pattern was similar to that previously reported from the tropical coastal waters in Asia by Tanabe *et al*. (2000).[12] However, their residue level in the present study were much lower than those from other tropical coastal waters in Asia. Considering the low levels of OCs in green mussels and oysters found in this study, contamination of OCs in the study area is not particularly serious in the

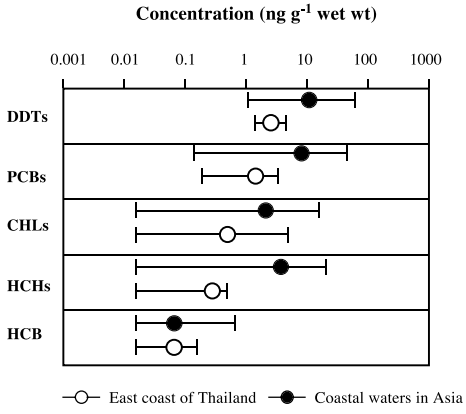

Fig. 3. Range and Mean Concentrations of Persistent Toxic Contaminants in Green Mussel from the East Coast of Thailand during 2002–2003 (this study) Compared with those from the Tropical Coastal Waters in Asia during 1994–1995 (Tanabe *et al*., 2000)

view point of human health and ecosystem perspective.

PCB Contamination

Generally sporadic incidences of higher PCBs levels were oberved at urban and industrial areas (sampling sites 1, 2; Figs. 1 and 2). This could be

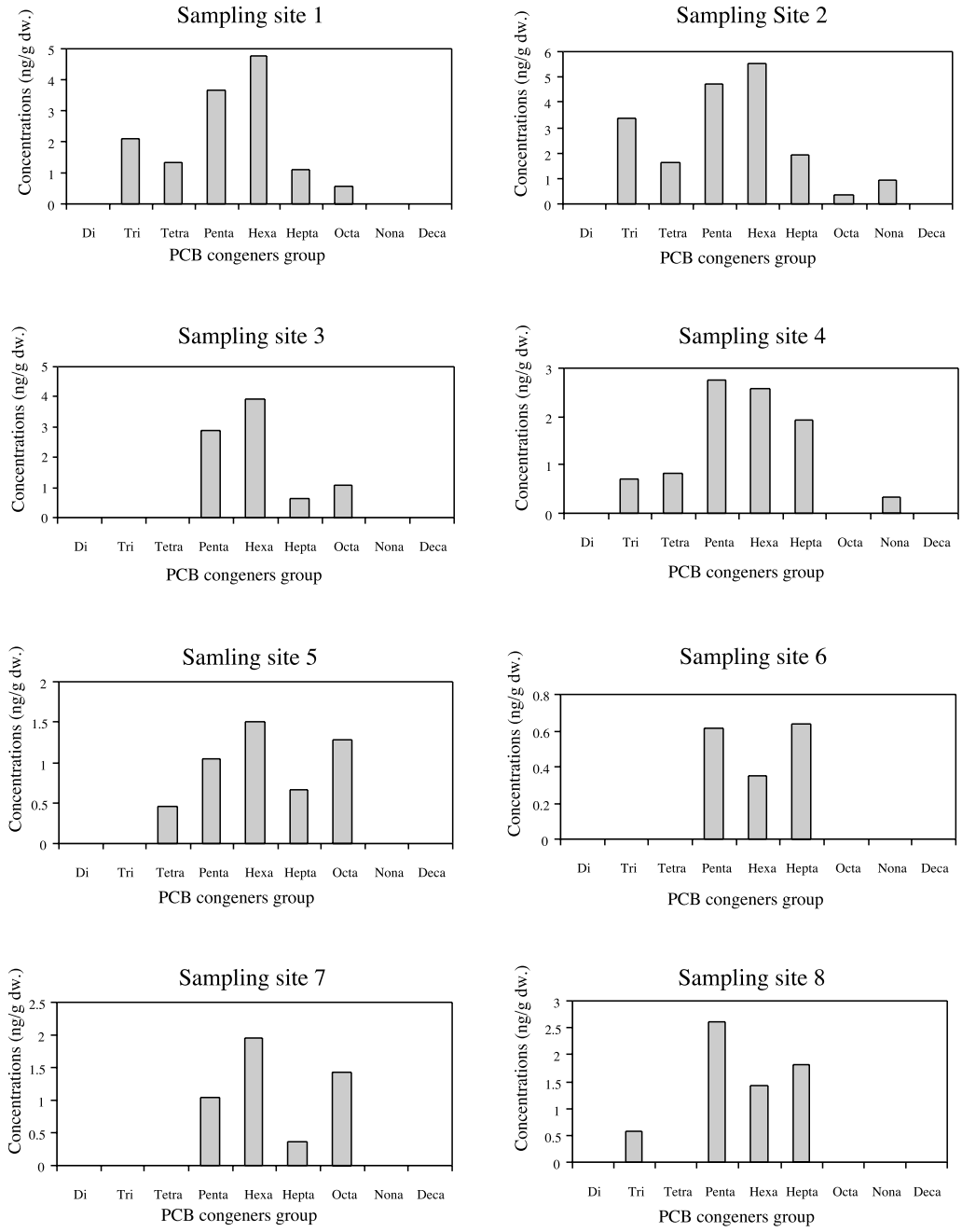

Fig. 4. The PCB Congener Groups in Green Mussel from the East Coast of Thailand

due to waste draining from industrial activities and the power plant (site 1) located in this study area. One of the major source of PCBs in Thailand is considered to be transformers and capacitors imported by the Electricity Authority of Thailand.[19] Watanabe *et al*. (1996),[19] recorded PCB pollution at the transformer dumping site and from capacitors located at a Bangkok suburb near the Chao Praya River estuary. However, based on a comparison of PCB levels in mussels in this study and in other tropical coastal waters in Asia[12] (Fig. 3), it appears that the former was relatively low, suggesting the contamination by PCBs in the coastal waters of Thailand is not a serious concern problem at present. In spite of such a situation, it is necessary to act continuously as a monitor of the PCBs due to increasing industrial and related polluting activities in these regions.

The distribution pattern of PCB homologues in mussels and oysters from this study area are given in Figs. 4 and 5 respectively. It is interesting that

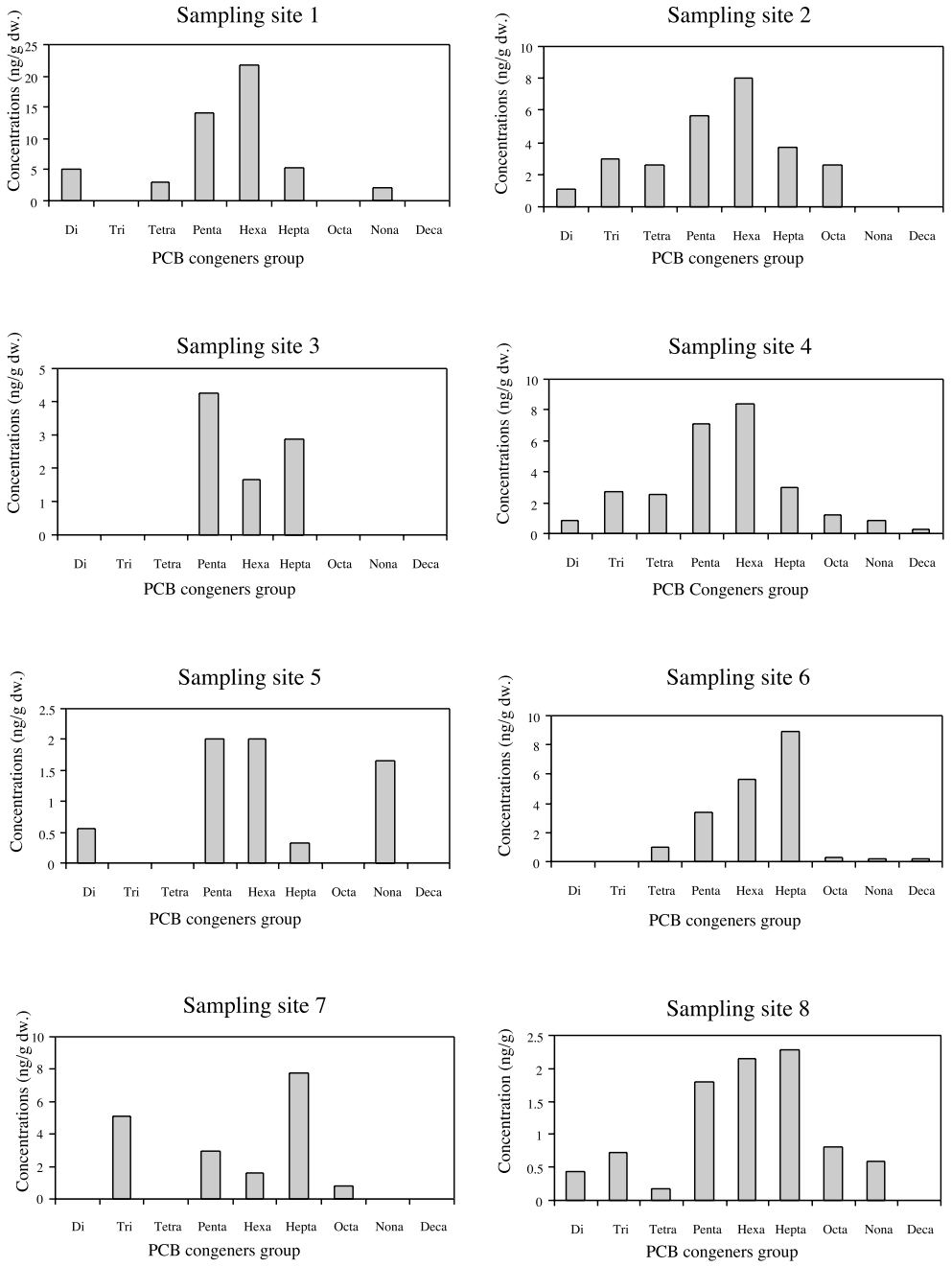

Fig. 5. The PCB Congener Groups in Oyster from the East Coast of Thailand

penta-, hexa- and hepta-chlorinated congeners are the predominant homologues in both the mussel and oyster samples. The degree of chlorination of PCBs has an effect on their fate in the environment. Highly chlorinated congeners are persistent in the environment because they are less volatile and nondegradable in comparison with lower chlorinated congeners.[20] PCBs are mixtures of chlorinated hydrocarbons in which the penta-, hexa and hepta-chlorinated biphenyls are the most common isomers in industrial applications, including dielectrics in transformers and large capacitors. Hence it is likely these biphenyls are the most abundant homologues in biota and their environments with lower and higher chlorinated biphenyls being present only in low concentrations. In accord, Oh *et al.* (2001).[21] reported a similar distribution pattern of these biphenyls in the Yellow sea.

The vertical distributions of PCBs and DDTs in the sediment core sample from site 1 are given in

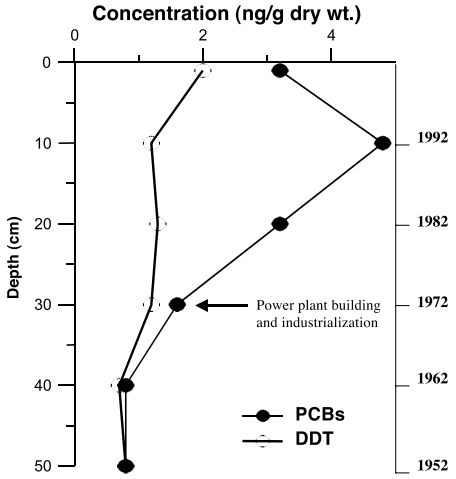

Fig. 6. PCB and DDT Concentration Profiles in Sediment Core Sample of Station 1

Fig. 6. The concentration of PCBs shows a subsurface maximum (4.82 ng/g) at depth of around 10 cm. This corresponds to the 1992 on the base of the age dating results. The profile of PCB concentrations reflects the active use of PCBs in the east coast of Thailand during the period of 1972–1992. This is because of the building of Bang Pa Kong power plant in 1972 and the subsequent blooming of industry in the area to around 1992. The national economy was then slowed after reaching a crisis in 1997. Among the organochlorine pesticides, DDT compounds were detected in the core samples. The pollution peak was found at the surface of the sediment column. This may reflect the current usage of DDT compounds in the area. However, the concentration was constant and low (0.84–2.05 ng/g) in relation to those found in the north coast of Vietnam (6.2–10.4 ng/g),[22] Osaka Bay in Japan (2.5–12 ng/g),[23] and Masan Bay in Korea (0.27–89.2 ng/g).[24]

International Comparison

Comparison of organochloride pesticides and PCBs in mussels from Thailand with those from other countries is summarized in Table 2. The levels of organochlorine compounds in mussels in the present study were comparable with those found in other developing countries (*e.g.*, India and Philippines), but lower than those of developed countries (*e.g.*, North, Central and South America, Denmark, Spain, and Hong Kong). This could be explained by the flux model of Iwata *et al.* (1994)[23] which suggests that the residence time of organochlorine compounds in tropical aquatic ecosystem is quite short with comparatively rapid transfer to the atmosphere due to the high temperature. However, OCs are still being widely used for agricultural and public health purposes. In this regard, the continuous monitoring of OCs residues in the country is deemed necessary.

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