CONTAMINATION BY PERSISTENT ORGANIC POLLUTANTS AND ENDOCRINE DISRUPTING CHEMICALS IN VIETNAM - PATTERNS, BEHAVIOR, TRENDS AND TOXIC POTENTIAL

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ABSTRACT

Widespread contamination by Endocrine Disrupting Chemicals (EDCs) and their toxic effects on wildlife and humans have been major concerns and have received considerable attention during the past four decades. Vietnam is a developing country located in the central part of the Southeast Asian tropical region and therefore, understanding of occurrence of EDCs may help increase knowledge regarding the role of this region as emission sources of EDCs for pristine areas. This paper provides a comprehensive overview of the occurrence of EDCs, their distribution, behavior and fate in various environmental compartments such as air, water, sediments, soils and biota from Vietnam. Data of some case studies conducted in our laboratory in recent years have also been reviewed to provide in-depth insights into the environmental behavior and trends of contamination by these toxic chemicals in the Vietnamese environment.

Existing monitoring data reported during the 1990s clearly indicate elevated contamination of DDTs in most of these compartments in Vietnam. Studies in frame of the Asia-Pacific Mussel Watch Program have revealed that the aquatic environment of Vietnam contained higher concentrations of DDTs as compared to other countries in region, suggesting the role of Vietnamese environment as a significant emission source of DDT in the Southeast Asian region. Widespread contamination of some endocrine active compounds such as alkylphenols and phthalates was observed along the coasts of northern and middle Vietnam. Elevated concentrations of bis-phenol A were found in some locations in Red River delta, North Vietnam, comparable or higher than those reported for developed nations in Western Europe and North America. This suggests the presence of significant source of bisphenol-A along Red River estuary. A case study on seasonal variation of alkylphenols and phthalates in surface water of river delta and estuary of north and middle Vietnam indicated the differences in distribution of these compounds between dry and rainy seasons. Higher concentrations of alkylphenols and phthalates were found in dry season in estuary; while the contrasting pattern was observed in the river delta, showing elevated residues in rainy season.

INTRODUCTION

In recent years, an increasing number of studies have highlighted the role of the tropical Asian developing countries as a potential source of pollution by endocrine disrupting chemicals (EDCs), particularly chlorinated pesticides for pristine areas such as the Arctic region. Vietnam is a developing country located in the center of the Southeast Asian tropical region and therefore, understanding of occurrence of these compounds may help increase knowledge regarding the role of this region as emission source. Our studies conducted during the last decade indicated ubiquitous and high pollution by organochlorine pesticides, particularly DDTs, alkylphenolic compounds, phthalates in different environmental media from both North and South Vietnam; and the results suggested that Vietnam may become a significant emission source of DDT pollution in south East Asian region (Iwata et al., 1994; Kannan el at., 1995; Nhan et al., 1999; 2001; Viet et al., 2000, 2002; Minh et al., 2002; Monirith et al., 2003).

In recent years, our laboratory has been involving in the project on monitoring of Endorine Disrupting Chemicals (EDCs) in rivers and fresh water bodies close to the coastal areas in East Asian countries, supported by United Nations University (UNU). In the context of this program, we have extensively investigated the levels, fate and examined the trends of contamination by chlorinated pesticides and endocrine active compounds in water, sediments and mussels from different locations along the Red river delta, North Vietnam (Viet et al., 2002), coastal areas in Hue, Middle Vietnam and Hochiminh City canals, South Vietnam. In this study, we focused on the analysis of chlorinated pesticides such as DDTs and its metabolites, hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB), endrin, aldrin, dieldrin and chlordane related compounds like heptachlor and chlordane (listed in POPs compounds according to Stockholm convention) in water and sediment from Babe Lake, the remote lake located at relatively high sea level in the northern of Vietnam and West Lake, the biggest lake in Hanoi city, which receives discharges from various industrial and extensive human activities. On the other hand, water samples in lagoons, river in Middle Vietnam also were collected to determine alkylphenols, bis-phenol A, phthalates. In addition, concentrations of several pesticides were also examined in some locations in Saigon River, the biggest rivers in Hochiminh city, the largest industrialized city in the south of Vietnam to compare the magnitude of contamination among north and south region and to discern the temporal trend of contamination in the southern of the countries.

MATERIALS AND METHODS

Samples and sampling locations

Water and surface sediment samples were collected in 2 main areas: Babe Lake, West Lake (in Hanoi) and in both rainy (August, 2002) and dry season (December, 2002) and lagoon system in Hue city in two years 2000, 2001. In addition, water and sediment samples were also collected from some locations in the canals in Hochiminh city. Babe Lake is a natural mountain lake situated at the densely forest area of the Babe National Park, Bac Kan province of northern Vietnam. In 1997, the United Nations (UNESCO) proposed the Babe Park as one of 6 areas of world important heritage in Vietnam. West Lake is situated at the north of the downtown areas of Hanoi city, the capital of Vietnam. This is the biggest lake in Hanoi and it is surrounded by some industrial factories and extensive human activities. Located about 650 km in the south of Hanoi city and with population of 1,045,130 (1999), Thua Thien-Hue is an important province well known for its tourism activities and traditional, fine agricultural products. Belonging to this province, Tamgiang-Cauhai-Langco lagoons system spreads over 23,650 hectares, with 70 km of length along the northwest coast. These lagoons are among the biggest lagoons in Southest Asia and play an important role to preserve biodiversity of ecosystem in large coastal area. The lagoon is also a reservoir of water from five regional rivers including Huong river, Olau river, Bo river, Truoi river, Loinong river and Cauhai river. Saigon River is the largest river in Hochinhminh city. Hochiminh city is also the biggest industrial city in Vietnam, with high density population. All samples were immediately stored in ice-box and transported to the laboratory and stored at -20°C until analysis. Water samples were analyzed immediately after collection.

Chemical analysis

Analytical procedures of water and sediment samples were followed the standard protocol provided by United Nations University (UNU) while analytical procedure of biological samples was developed in our laboratory. Water samples were extracted using hexane, followed by dehydration by anhydrous sodium sulfate, cleaning up by silica gel column. The chlorinated pesticides were then quantified using high resolution gas chromatograph coupled with mass spectrometric detector (GC/MS) (Shimadzu GC/MS QP-5000, Shimadzu Corp. Kyoto, Japan). The analytical procedure of sediment comprises Soxhlet extraction by mixture

of acetone and hexane, dehydration with anhydrous sodium sulfate, cleaning up and fractionation by Florisil column and quantification by both GC/ECD and GC/MS (Shimadzu GC-14B and QP-5000). Recoveries for chlorinated pesticides ranged from 79 – 106 % for water and 70 – 110 % for sediment samples. Standard Reference Materials (SRMs), supplied by Shimadzu Co., Ltd. were also analyzed. The results from our laboratory were in good agreement with the certified values.

RESULTS AND DISCUSSION

Phthalates

Concentration of four compounds diethylphthalates (DEP), di- n- butylphthalates (DBP), di- 2- ethylhexyladiphates (DEHA) and di- 2- ethylhexylphthalates (DEHP) in water samples has been collected from three chosen areas are shown in Table 1.1.

As shown in table 1.1, in rainy season, levels of DEP were lower than detection limit (2 $ng.L^{-1}$) in both sites at Red river and Balat estuary. However, in Tamgiang- Cauhai- Langco lagoon, average DEP concentration was 3.7 $ng.L^{-1}$ in rainy season and 4.9 $ng.L^{-1}$ in dry season.

On the other hand, DBP was determined in al three sites with only low concentrations: 4.6, 3.9, and 2.1 $ng.L^{-1}$ in Red river, Balat estuary and Tamgiang- Cauhai- Langco lagoon respectively, in the rainy season. In particular, DBP was found with 2.3 $ng.L^{-1}$ in Tamgiang- Cauhai- Langco lagoon, in the dry season.

The third compound, DEHA was lower than detection limit (4 $ng.L^{-1}$) in Red river, and around 5.5 and 6.3 $ng.L^{-1}$ in Balat estuary and Tamgiang- Cauhai- Langco lagoon in the rainy season. In dry season, DEHA was found at 6.5 $ng.L^{-1}$ in Tamgiang- Cauhai- Langco lagoon.

Table 3.1. Concentrations of DEP, DBP, DEHA, DEHP ($ng.L^{-1}$) in collected water samples

No	Compounds	Red river	Balat estuary	Tamgiang- Cauhai- Langco lagoon	
		RS (n=5)	RS (n=15)	DS (n=24)	RS (n=24)
1	DEP	< 2 (< 2)	< 2 (< 2)	4.9 (< 2- 17.6)	3.7 (< 2 - 6)
2	DBP	4.6 (< 1.2- 8.3)	3.9 (< 1.2 - 16.9)	2.3 (< 1.2- 9.2)	2.1 (< 1.2- 8)
3	DEHA	< 4 (< 4)	5.5 (< 4 - 7.2)	6.5 (< 4- 13.8)	6.3 (< 4 - 28.8)
4	DEHP	18.3 (8.8- 25.6)	17.2 (2.3- 36.2)	18.8 (< 1.1 - 78)	14.3 (2- 32)
5	Σ Phthalates	22.9 (0.01- 25.6)	21.1 (< - 36.2)	26 (< - 78)	20.1 (< - 32)
	Figures in parentheses ( ) indicate the range RS: rainy season; DS: dry season ; N: amounts of analyzed samples				
	Σ Phthalates = DEP+DBP+DEHP.				

DEHP is probably most abundant compounds in phthalate group accounting for 70-80% of total phthalate concentration and ranging from about 14 to 19 $ng.L^{-1}$. DEHP concentration seemed to be higher in Red river than that in Balat estuary (18.3 comparison to 17.2 $ng.L^{-1}$). In Tam Giang lagoon, on the other hand, DEHP level was higher in dry season in comparison to that in the rainy season. This difference may due to the dilution of water in that lagoon during rainy season.

In conclusion, there were maybe no significant differences of concentrations among DEP, DBP, DEHA and DEHP at all three investigated sites and those levels may suggest the background level of such compounds in aquatic environment.

However, at Tamgiang- Cauhai- Langco lagoons, concentrations of four phthalates increased more slightly in the dry season than that in the rainy season. That may be resulted from the dilution of these compounds by water in the rainy season.

However, the above mentioned is only a few preliminary appreciations, based on the results of this research. In order to achieve an overall conclusion about the change in time or season of phthalates in general and its target compounds in particular, thorough studies should be pronounced

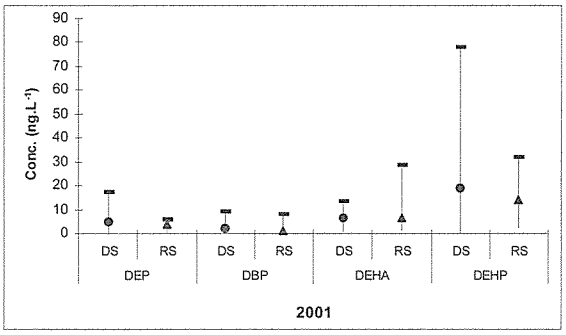

Figure 1.1a. Concentrations of DEP, DBP, DEHA, DEHP in rainy season (2000) at Red river (■), Balat estuary (●)

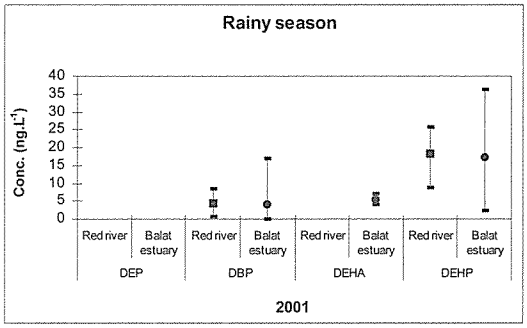

Figure 1.1b. Concentrations of DEP, DBP, DEHA, DEHP in rainy season (σ) and dry season (●) at Tamgiang-Cauhai-Langco lagoons

Bisphenol- A and alkylphenols

Concentration of bisphenol- A and alkylphenols ($ng.L^{-1}$) in water samples at Red River, Balat estuary and Tamgiang- Cauthai- Langco lagoon are showed in table 2.

Table 2. Concentrations of bisphenol- A and alkylphenols (ng.L-1) in collected water samples

No	Compounds	Red river RS (n=5)	Balat estuary RS (n=15)	TG, CH, LC, TT, SH lagoon DS (n=24)	TG, CH, LC, TT, SH lagoon RS (n=24)
1	4-t-Butylphenol	6.3 (4.9- 7.9)	8.9 (<0.13- 22.8)	5.1 (0.8- 10.1)	4.8 (<0.13- 4.8)
2	2,4-Dichlorophenol	1.9 (<0.15- 2)	<0.15 (<0.15)	1.1 (<0.15- 2)	<0.15 (<0.15)
3	4-n-Butylphenol	7.4 (4.9- 11.1)	4.9 (0.3- 8.1)	4.8 (<0.1- 9.6)	1.5 (<0.1- 7.4)
4	4-n-Pentylphenol	1.6 (1.1- 2.0)	1.1 (<0.1- 6.3)	0.8 (<0.1- 3.1)	0.4 (<0.1- 0.5)
5	4-n-Hexylphenol	0.4 (0.3- 0.6)	15.7 (<0.1- 25.7)	18 (5.8- 47.5)	1.4 (<0.1- 3.2)
6	4-t-Octylphenol	<0.15 (<0.15)	1.1 (<0.15- 2.4)	2.9 (<0.15- 17.9)	1.9 (<0.15- 13.4)
7	4-n-Heptylphenol	1.6 (0.4- 4.4)	<0.23 (<0.23)	8.2 (<0.23- 15.8)	3.3 (<0.23- 4.5)
8	4-Nonylphenol	25.9 (2.8- 70.3)	9.7 (<1- 22.5)	63.1 (<1- 131.2)	<1 (<1)
9	4-n-Octylphenol	3.1 (<0.1- 8.8)	0.3 (<0.1- 0.5)	3.1 (<0.1- 14.1)	2.7 (<0.1- 12.9)
10	Pentachlorophenol	0.9 (<0.2- 1.1)	<0.2 (<0.2)	6 (<0.2- 7.4)	3.6 (<0.2- 8.8)
11	Bisphenol-A	9.8 (6.9- 15.6)	3.2 (0.8- 6.2)	13.9 (6.7- 44.9)	16.8 (<0.5- 45.8)
12	Σ alkylphenols	46.3	41.7	106	16

Figures in parentheses ( ) indicate the ranges
RS: rainy season; DS: dry season;
n: amounts of analyzed samples

Σ alkylphenols= 4-t-Butylphenol+ 4-n-Butylphenol+4-n-Pentylphenol+4-n-Hexylphenol+ 4-t-Octylphenol+ 4-n-Heptylphenol+ 4-Nonylphenol+ 4-n-Octylphenol.

In general, the levels of total alkylphenols and bisphenol-A varied from 16 to about 106 ngL^{-1} in rainy and dry season in Tam Giang lagoon. In Red river and its estuary, the concentration was higher than that in Tam Giang lagoon in the same rainy season.

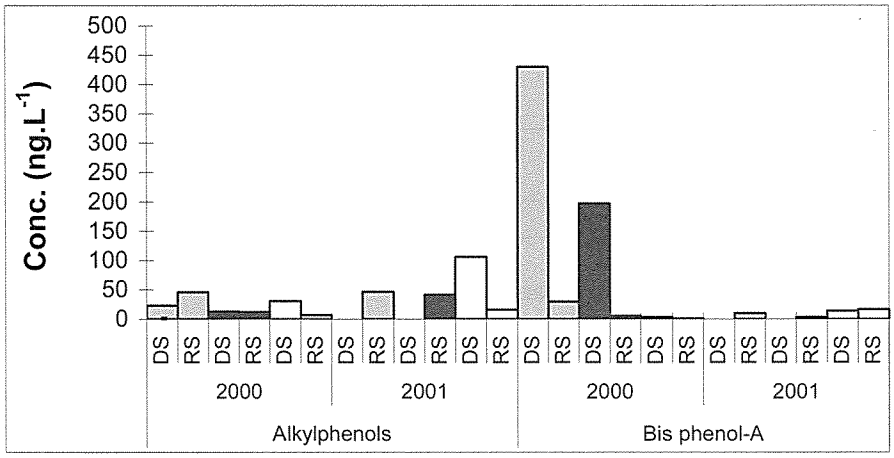

Figure 2. Histogram illustrate total of alkylphenols, bis phenol-A at Red River (▨) Balat estuary (■) ,Tamgiang-Cauhai-Langco lagoons (□)

By comparing the results of 2001 with that of 2000, we see that:

At the Red River, in the rainy season, the level of total alkylphenols was sustainable, but bisphenol-A was three times lower than that in 2000 (30 $ng.L^{-1}$ in 2000 and 9.8 $ng.L^{-1}$ in 2001).

At Balat estuary, in the rainy season, the level of total alkylphenols was three times higher than that in 2000 (41.7 compared to 12 $ng.L^{-1}$ in 2000). Meanwhile, the level of bisphenol-A in this study (3.2 $ng.L^{-1}$) has increased lightly than in 2000 (5.4 $ng.L^{-1}$).

In similar comparison, at lagoons of Hue province, concentration of total alkylphenols and bisphenol-A has increased. In the rainy season, total alkylphenols were 6.8 $ng.L^{-1}$, 16 ngL^{-1} and bisphenol-A were 0.9 $ng.L^{-1}$, 4.7 $ng.L^{-1}$ in 2000, 2001 respectively. In the dry season, the level of total alkylphenols and bisphenol-A was higher than that in the rainy season (total alkylphenols were 31 $ng.L^{-1}$, 106 $ng.L^{-1}$ and bisphenol-A were 3.4 $ng.L^{-1}$, 13.9 $ng.L^{-1}$ in 2000, 2001 respectively. Nevertheless, total alkylphenols from our study was still below the safely level.

This result suggests the different behavior of alkylphenols and phthalates in river delta and coastal environment. The temperature dependence in tropical environment as well as the influence of the specific local sources may be reasons for the observed results in seasonal variations. To our knowledge, this is the first extensive study on the widespread contamination of EDCs in Vietnam environment. Regarding the trends of contamination by POPs, preliminary survey conducted in Red River delta water and sediments indicated a rapid decline trend in water and a slow decrease in sediments during 1995-2001. From ecotoxicological perspectives, concentrations of bis-phenol A and di(2-ethylhexyl)phthalates [DEHP] in surface water from some locations in Vietnam exceeded the guideline values for Ecotoxicological Effects and the Environmental Risk Limit, respectively, suggesting potential for toxic implications on aquatic wildlife. Future studies should be focused on the time trends of POPs and EDCs in biota in Vietnam in order to predict future trend of contamination and to reveal new clues for understanding possible toxic impacts on aquatic organisms

Organochlorines

Table 1.3. Concentrations of organochlorines (ng.L-1) in collected water samples.

No	Compounds	Red river RS(n=5)	Balat estuary RS(n=15)	TG, CH, LC, TT, SH lagoon	
				DS(n=24)	RS(n=24)
1	p,p'-DDE	<2 (<2)	<2 (<2)	<2 (<2)	<2 (<2)
2	p,p'-DDD	<2 (<2)	<2 (<2)	<2 (<2)	<2 (<2)
3	p,p'-DDT	<2 (<2)	3 (<2 - 9.94)	5.8 (<2 - 10.53)	<2 (<2)
4	Σ p,p'-DDTs	<2	3	5.8	<2

Figures in parentheses( ) indicate the range
RS: rainy season ; DS: dry season ;
Σ p,p'-DDTs= p,p'-DDE+ p,p'-DDD+ p,p'-DDT
The whole of three cmpounds of p,p'-DDE, p, p'-DDD, were identified in all water collected samples in Red River, Balat estuary and in Tamgiang- Cauhai- Langco lagoon. However, p, p'-DDT presented with a quite low levels in Balat estuary (rainy season) and Tamgiang-Cauhai- Langco lagoon (dry season) with concentrations 3 $ng.L^{-1}$ and 5.8 $ng.L^{-1}$ respectively. In comparison with the results of the year of 2000 (2.97 $ng.L^{-1}$), Σ p, p'-DDTs in Balat estuary did not fluctuate much in the rainy season.

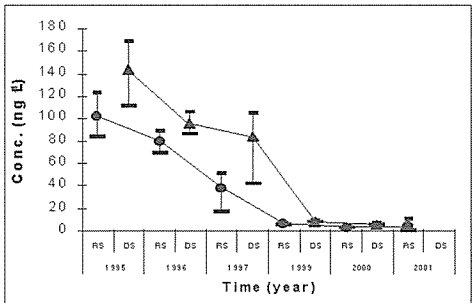

Figure 3. Temporal changes of Σ p,p'-DDTs at Balat estuary

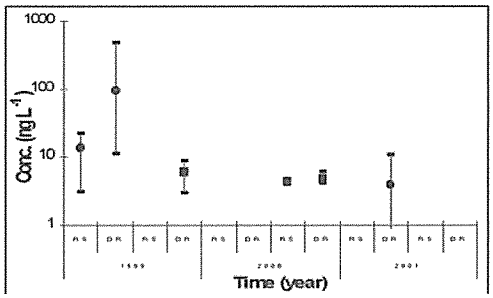

Figure 4. Temporal changes of Σ p,p'-DDTs at Red river (■) and Tamgiang-Cauhai-Langco lagoon (●)

At Tamgiang- Cauhai- Langco lagoon, in the dry season, concentration of Σ p, p'-DDTs were 5.8 $ng.L^{-1}$, which was higher than that in 2000 (not detectable), but much lower than that in 1999. At low concentration the changes of Σ p, p'- DDTs in 2001 was not really considerable and when comparing it with the previous results, However, further researches should be worked out to explain clearly the reasons for the fluctuation of the level of Σ p, p'-DDTs at Tamgiang- Cauhai- Langco lagoon.

To further understand the differences in the magnitude of contamination between northern and southern region and in the behavior of chlorinated pesticides between lakes and rivers, the spatial distribution of DDTs in water and sediment were also examined (Figure 5 and 6). It is clear that the occurrence of HCHs and DDTs was different in water and sediment. In water samples, HCHs were more frequently detected in most of the locations with concentrations generally higher than those of DDTs. On other hand, sediment contained higher concentrations of DDTs. This is due to the stronger affinity and sorption capacity of DDTs to solid particles. An interesting result was observed in Babe Lake, showing elevated concentrations of HCHs in water samples. These levels were apparently greater than those in West Lake and comparable to those in the Saigon River and some industrial sites in

Hochiminh canals (Figure 5). As mentioned earlier, higher concentrations of highly volatile compounds such as HCHs observed at higher latitudes and higher elevation could be as a result of the long range atmospheric transport. In this context, the role of the remote and high mountain lake as a reservoir of volatile organic contaminants should be considered as primary research focus in future.

In the respect of environmental quality point of view, the present situation of pollution by chlorinated pesticides in Vietnam is still of concern. According to the Canadian Environmental Quality Guideline (2002), the fresh water sediment quality guideline values and the probable effect levels for *p,p'*-DDE are 1.42 and 6.75 ng/g (ppb) dry wt basis, respectively. Most of the sediments samples analyzed in this study in West Lake, and some sewage sites in Hochiminh city canals and Sagon River contained p,p'-DDE concentrations exceeding these guideline values. Similarly, concentrations of HCHs in some locations in Saigon River and canals were also beyond the Canadian guidelines values of HCHs in freshwater sediment (0.94 and 1.38 ppb dry wt basis). This result highlights that magnitude of contamination of DDTs and HCHs in Vietnam is of concern and suggests the needs for further studies.

The distribution of HCHs and DDTs in sediments revealed different pattern as compared to that of water. DDT concentrations in sediment from West Lake is significantly higher than those in Babe Lake, indicating clearly that the source of DDTs comes from extensive industrial and human activities around the West Lake region. We found higher concentrations of DDTs in most of the samples analyzed, suggesting the recent input of DDTs in this region. A previous study by Nhan et al. (2001) also found high contamination of DDTs in sediment and biota from various locations in Hanoi, and the authors suggested that DDTs may be used as malaria control rather than agriculture purpose in recent years in Hanoi region (Nhan et al., 2001). Concentrations of DDTs in Saigon River were lower than those in West Lake (Figure 6). High levels of DDTs were also recorded in canals of Hochiminh city. An earlier survey conducted during 1990s also indicated elevated contamination of organochlorines including PCBs, DDTs, and HCHs in some locations of sewage canals in Hochiminh city (Iwata et al., 1994).

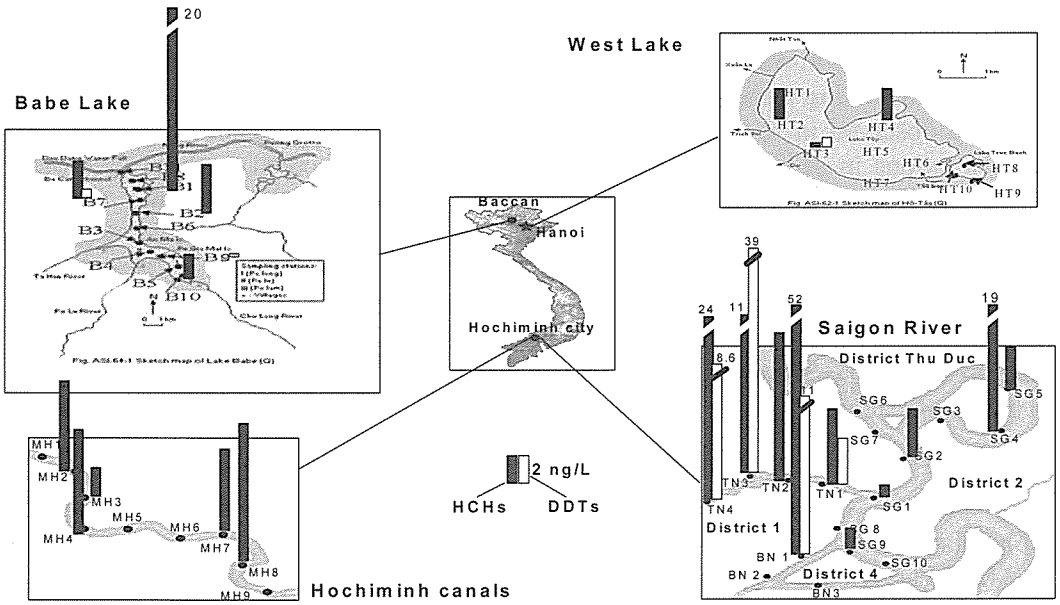

Figure 5. DDTs in some investigated samples.

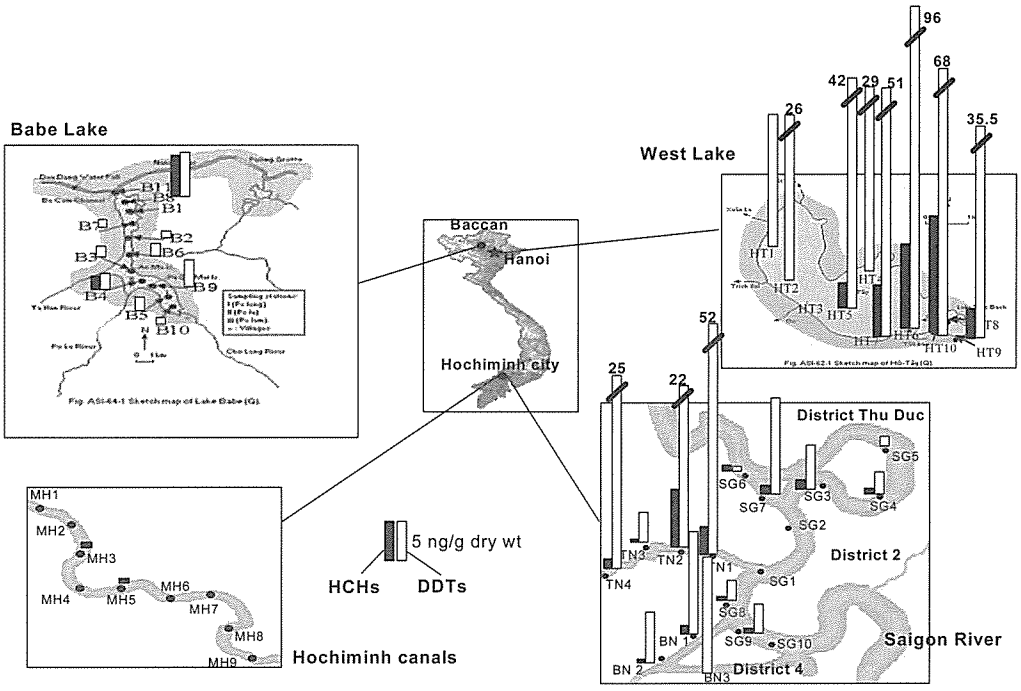

Figure 6. DDTs concentrations in other locations.

In general, our result indicate that in highly industrial locations such as West Lake, Saigon River and Hochiminh canals, higher contamination by both DDTs and HCHs were observed. In such a remote location as Babe Lake, HCHs contamination was more pronounced. The differences in occurrences of organochlorines among water and sediment could be influenced by their physico-chemical properties and recent input into the environment. Higher contamination of HCHs in Babe Lake could be mainly due to the atmospheric transport and preferential deposited to lower temperature region; while greater residues of DDTs in water and sediments from Saigon River, West Lake and Hochiminh canals may be attributable to the extensive human and industrial activities.

Our recent investigations in water and sediment from Red River delta, North Vietnam indicate a relatively rapid declining trend of DDTs and HCHs during 1995 – 2001. Nevertheless, the declining trend observed in sediment was slower than in water (Viet, 2002). In the present study, concentrations in sediment in Hochiminh canals were compared to those previously reported during the survey in 1990 (Iwata et al., 1994) to understand the trend of contamination in southern part of Vietnam. As a result, a significant decrease in concentration of DDTs and HCHs was noticed (Figure 3). In particular, DDT residues in Thinghe and Benghe were about 10 – 20 times decline. This result indicates that environmental quality in Vietnam has been improving during the last decade. However, the magnitude of contamination and temporal trends of chlorinated pesticides, particularly DDTs in biota from Vietnam have not extensively investigated and need continued studies.

CONCLUSIONS

This study presents our preliminary results of occurrence of chlorinated pesticides, alkylpenols, bis-phenol A and phthlates in some representative lakes, rivers, canals, lagoons in North, Middle and South Vietnam. A sign of elevated contamination residues of DDTs in water and sediments from Saigon River, West Lake and Hochiminh canals may be attributable to the extensive human and industrial activities. Temporal studies indicate relatively rapid declining trends of DDTs and HCHs in water and sediment in recent years. Nevertheless, the extent of contamination by DDTs in Vietnam is still of concern because

DDT concentrations in many locations in both north and southern part of the country exceeded the environmental quality guideline values. In estuary, higher residues in water in dry season, suggesting dependence of availability of alkylphenol and phthalates on temperature. In river delta, higher concentrations in water in rainy season, suggesting the presence of specific sources. Bisphenol A concentrations in water from some locations in Red River exceeded the guideline value for Ecotoxicological Effects Di(2-ethylhexyl)phthalate [DEHP] concentrations in water from some locations in Huong River exceeded the Environmental Risk Limit value. Further research should be focused on the systematic temporal trends of contamination of persistent chlorinated contaminants and their potential implications on environmental and human health.

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