CURRENT CONTAMINATION OF PERSISTENT ORGANOCHLORINES IN THIVAI RIVER SEDIMENTS, SOUTHERN VIETNAM

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ABSTRACT. This report presents the development an highly sensitive, stable and reliable OCs analyzing method with GC/MS (gas chromatography/mass spectrometry) and results of OCs (organochlorines) levels in sediments of Thivai River basin. The method recovery ranges from 72% to 129% and meets the requirement for OCs analyzing. 18 composite sediment samples were taken from 18 sites along Thivai river basin from upstream (Formosa Plant sewer mouth – Lo Ren canal) to downstream (confluence of Thi Vai and Go Gia rivers). The analytical results show that OCs concentration in Thi Vai river sediment (max 1.73 ng/g dry weight) is not higher than those of the reference sites and other river basins in Vietnam and in the world.
Keywords: OCs, sediment, Thivai, PCBs, organochlorines

1. INTRODUCTION

Thivai river which has area of 120 square km rises from Longthanh district, Dongnai province and flows a distance of 70 km through Baria – Vungtau province. It joins Gogia river near the end of Caimep industrial zone and goes to East Sea at Ganhrai gulf [19].

There are many industrial zones located along Thivai river basin such as: Nhontrach industrial zones (Nhontrach I, II, III, IV, V, VI), Vedal – Godau industrial zone, Myxuan industrial zone, Holcim cement factory, Phumy industrial zone, Caimep industrial zone. In addition, there are some river port as Vedan port, Godau port, PhuMy port and Caimep port. All these industrial zones and river ports have been the main polluted sources to Thivai river basin since the 1990s.

Organochlorine compounds (OCs) are a set of chemical substances that persist in the environment, undergo long range transport, bioaccumulation through the food chain, and pose a risk of causing adverse effects to human, wildlife and the environment. OCs are easily adsorbed on suspended particles in water and air, then deposit to the soil and sediment due to their hydrophobic property. Soil and sediment are usually OCs' primary receiver when these compounds penetrate the environment. Therefore, soil and sediment can be sensitive indicators for both spatial and temporal trends when we attempt to assess OCs contamination in the ecosystem [34].

2. MATERIALS AND METHODS

2.1. Samples collection:

Eighteen sampling points were selected in Thivai river basin and near the places affected by industrial zones and they are described in Table 1 and Fig. 1 below. At each sampling site, a grab of 5 – 10 cm surface sediment was collected at both river banks by using Eckman dredge and then a composite sediment sample was prepared by mixing the samples taken in both riversides. Eighteen composite sediment samples were taken along the river from the upstream to downstream by 2009. In the lab, these sediments were dried in room temperature on the aluminum foil, ground and sieved, kept in brown glass bottle at low temperature freezer (about -5^{0}C) until analysis.

Table 1: Description on sampling sites in Thivai river basin

Sample code	GPS positions	Note	Sample code	GPS positions	Note
TV-1	N: 10^{0}42'55.2" E: 106^{0}57'36.6"	Loren bridge, small canal, upstream of Thivai river	TV-10	N: 10^{0}38'34.9" E: 107^{0}01'41.1"	Myxuan industrial zone, Thivai river
TV-2	N: 10^{0}41'51.1" E: 106^{0}58'11.2"	Upstream, 1000m from LongTho guard post	TV-11	N: 10^{0}38'10.4" E: 107^{0}00'10.2"	TacCaiChung, light buoy 25, Thivai river
TV-3	N: 10^{0}41'38.9" E: 106^{0}58'22.3"	Upstream, 400m from LongTho guard post	TV-12	N: 10^{0}36'48.9" E: 107^{0}00'29.2"	Upstream, 800m from Holcim factory
TV-4	N: 10^{0}41'30.5" E: 106^{0}58'29.9"	LongTho guard post, Thivai river	TV-13	N: 10^{0}36'16.5" E: 107^{0}01'06.8"	Holcim factory, Thivai river
TV-5	N: 10^{0}41'00.2" E: 106^{0}59'06.3"	MatTroi canal – Thivai river confluence. Downstream, 2000m from LongTho guard post	TV-14	N: 10^{0}35'41.8" E: 107^{0}01'30.5"	Sewer of Phumy Thermal Power Plant
TV-6	N: 10^{0}40'45.1" E: 106^{0}59'42.4"	BaBip canal, 500m from BaBip-Thivai confluence	TV-15	N: 10^{0}35'02.9" E: 107^{0}01'36.8"	PhuMy port, Thivai river
TV-7	N: 10^{0}40'02.5" E: 107^{0}00'57.6"	Vedan sewer, location in NuocLon canal, 300m from NuocLon-Thivai confluence	TV-16	N: 10^{0}34'32.7" E: 107^{0}01'32.4"	Downstream, 200m from PhuMy port
TV-8	N: 10^{0}39'39.8" E: 107^{0}01'01.3"	Vedan port, Thivai river	TV-17	N: 10^{0}32'03.5" E: 107^{0}01'30.0"	CaiMep port, Thivai river
TV-9	N: 10^{0}39'23.8" E: 107^{0}01'09.6"	Godau port, Thivai river	TV-18	N: 10^{0}31'15.5" E: 107^{0}00'36.9"	Thivai – GoGia confluence

2.2. Analytical method:

OCs analytical procedure is set up based on the certified methods [26,28,29] as well as slightly modified from tested methods proposed by other authors [1,4,5,12,14,15] and from analytical protocol of CEAL - EPFL. Briefly, 10g air-dried soil/sediment were mixed with 10 g anhydrous Na_2SO_4 and 5 g activated copper powder, and then spiked with surrogate CB 209. Extraction was conducted by soxhlet extractor using a mixture of n-hexane : acetone (ratio 1:1) during 18 hours. Sulfur compounds will be removed by activated copper powder in the soxhlet extraction process. After rotary evaporation, the extract was clean-up to remove pigment, humic acid and other organic interferences. In our research, we could not use H_2SO_4 96% because it will destroy some analytes such as aldrin, endrin, endosulfans [27] and alumina column is also not recommended because of possible dehydrochlorination of some PTSs, e.g. pp'-DDT [23,25]. Due to these reasons, gel permeation chromatography (GPC) was used for removing co-extracting compounds. Finally, PCBs congeners will be separated from organochlorine pesticides through florisil chromatography column as previously described (Minh et al., 2003). The cleaned extract was concentrated under N_2 flow to 0.5 ml and precise weight was recorded. The final solution was analyzed by gas chromatography/mass spectrometry (GC/MS) after spiking internal standard ^{13}C isotope CB 77 to check response factor of MS detector to specific matrix of sample.

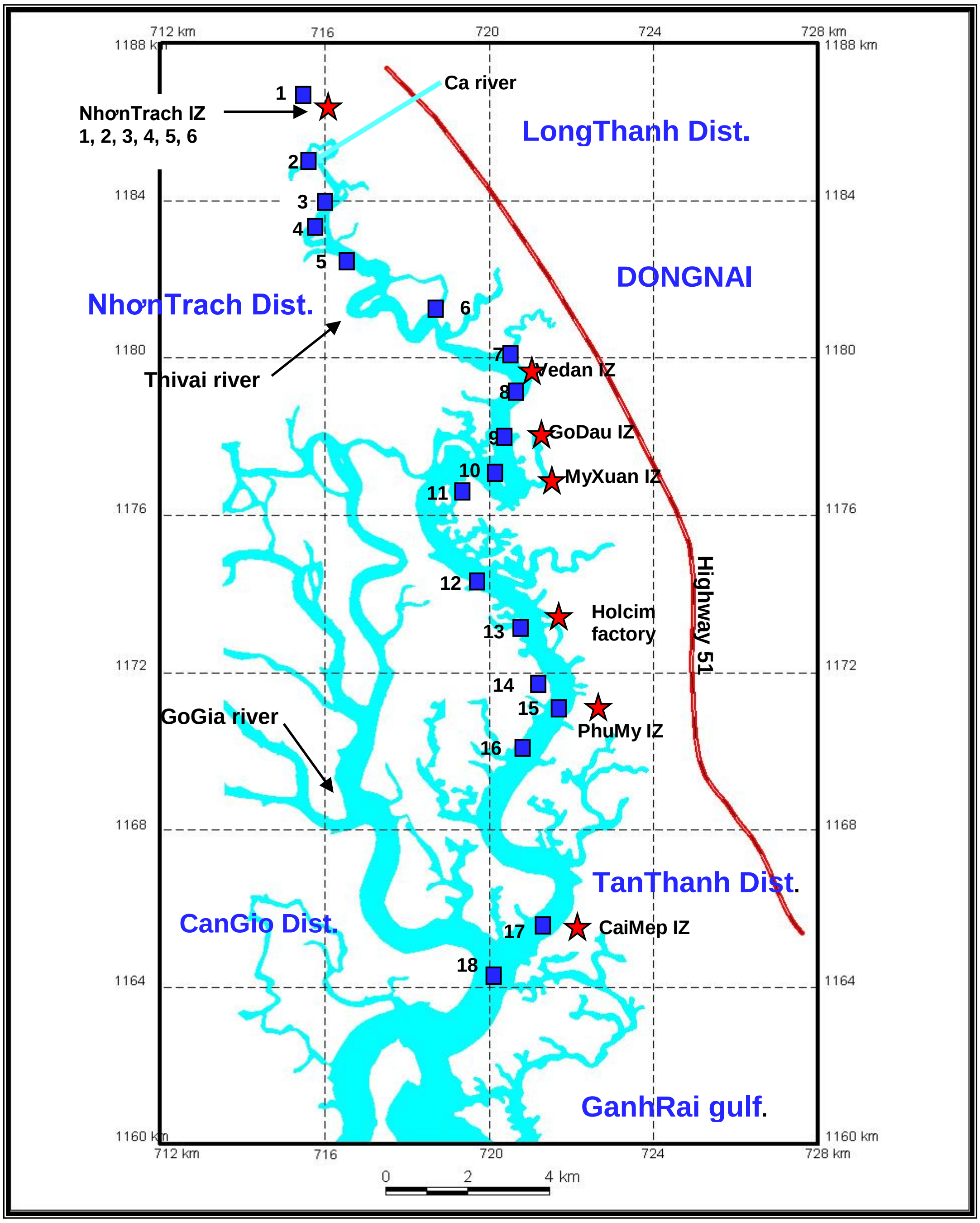

Figure 1: Sampling sites of sediment samples in Thivai river basin

OCs were quantified by GC/MS system (Shimadzu QP2010) using capillary column ZB-5 MS (60 m × 0.25 mm i.d. × 0.25 μm film thickness). The column oven temperature was programmed from 150 ^{0}C (3 minute) to 200 ^{0}C at a rate of 4 ^{0}C/min, held for 3 minutes, then increased to 285 ^{0}C at a rate of 2^{0}C/min, and held for 9 minutes. The PCB standard used for quantification was a mixture of 22 PCB congeners (Promochem GmbH, Germany). Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain total PCB concentrations. The injector and interface temperature was 250 ^{0}C. The column flow was constant at 1.0 ml/min. All OCs compounds were identified by MS detector in the NCI-SIM mode with CH_4 as reagent gas. NCI method is the chemical ionization procedure utilizing negative ions. The advantage of NCI is compound selectivity. Compouns with high electron affinities are selectively ionized. For these types of compouns, very high sensitivity can be obtained in comparison with the EI and NCI methods. However, the ion source is easy contaminated and lifetime of filament is reduced when using this method. Recovery rates obtained by this procedure were as follows: HCHs 92% to 104%; Endosulfans 72 to 100% CHLs 74 to 81%; DDTs 80% to 91%, and PCBs 75% to 129%. Quality control has been achieved by the analyzing the SRM 1941a and a procedural blank was run for crossverification.

3. RESULTS AND DISCUSSIONS

Residue levels and contamination pattern: Concentrations (ng/g dry wt.) of persistent OCs analyzed in this study are given in Table 2. In general, the residue pattern of OCs in the sediment from Thivai River followed the order: DDTs > PCBs > HCB > HCHs > CHLs and similar to those in previous study [18] in Saigon-Thinghe Rivers. Concentrations of OCs, however, varied between sampling sites and higher concentrations were mostly observed towards upstream and middle stream of Thivai River. PCBs and DDTs are the most abundant pollutants in our sediment samples. This suggestes that the widespread and dominant contamination of them in the environment. The abundance of DDTs and PCBs in Vietnam may be due to their larger usage as well as higher persistency over the other OCs. Until 1985, nearly 30,000 tons of PCB – containing industrial oils had been imported to Vietnam [21]. In addition, electrical equipments like transformer and capacitors containing PCB-contaminated oil had been also imported to Vietnam until the mid 1980s [9]. Those materials are parts of PCBs sources to the environment, besides releases from heavy weapons used during the Indochina War [24].

Composition of DDTs: Similar to soil samples, ratios of DDT to its degradation products, as for example ratio of DDT/DDE could be used for evaluating degradation features of the parent compound in sediments. Strandberg et al. (1998) suggested that DDT/ DDE ratio lower than 0.3 could be the result of the aged mixtures in the environment, and those higher than 0.5 might indicate recent use of DDT.

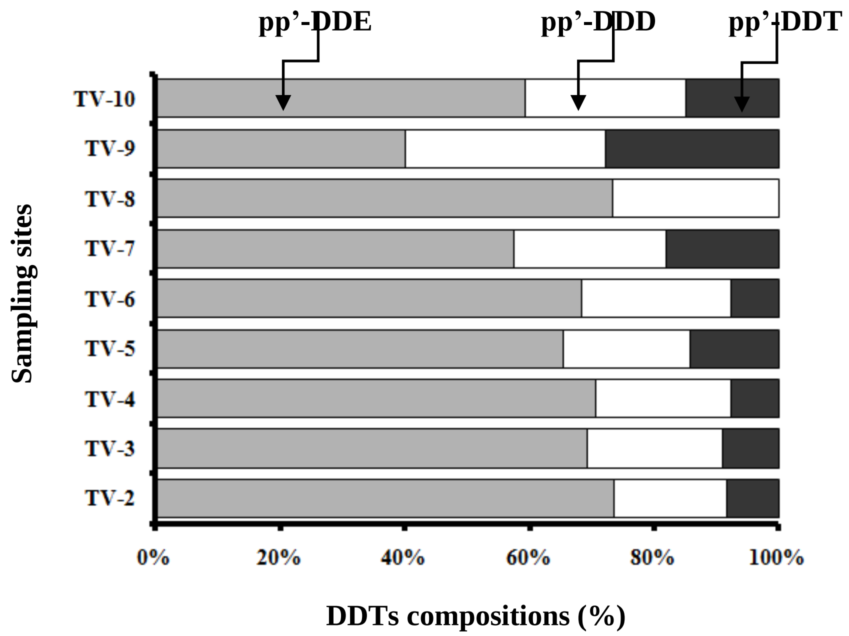

Figure 2: DDTs composition in Thivai river sediment samples

Fig. 2 shows DDTs composition of Thivai River sediment samples. From the Table 2, most of the DDTs composition in Thivai sediments have the ratios of pp'-DDT/pp'-DDE near or lower than 0.3. This suggests possible result of the aged mixtures in the environment.

Table 2: OCs concentration in sediment samples from Thivai river basin (ng/g dry weight)

PTSs	TV-1	TV-2	TV-3	TV-4	TV-5	TV-6	TV-7	TV-8	TV-9	TV-10	TV-11	TV-12	TV-13	TV-14	TV-15	TV-16	TV-17	TV-18	Min.	Max.	Mean	TEL	PEL
PCBs	0.02	0.85	0.82	1.16	0.13	0.15	0.14	0.22	1.23	0.11	1.05	0.29	0.07	0.09	1.27	0.09	0.06	0.05	0.02	1.27	0.43	34.1	277
HCB	0.02	0.78	0.40	0.07	0.10	0.09	0.35	0.54	0.11	0.08	0.21	0.07	0.04	0.06	0.05	0.05	0.05	0.06	0.02	0.78	0.17		
HCHs	nd	0.07	0.07	0.02	0.25	0.16	0.08	0.21	0.04	0.03	0.02	0.02	0.02	0.03	nd	0.02	0.02	0.02	nd	0.25	0.06	0.94	1.38
Heptachlor	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-		
Hept Epox	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-	0.6	2.74
Aldrin	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-		
Dieldrin	0.35	0.47	0.88	0.06	0.08	0.08	0.06	0.06	0.21	0.05	0.04	0.02	0.07	0.02	nd	0.03	0.04	nd	nd	0.88	0.14	2.85	6.67
Endrin	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-	2.67	62.4
Endrin Ald	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-		
Chlordanes	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	-	-	-	4.5	8.9
Endosulfans	nd	0.03	0.04	nd	0.09	0.08	0.13	0.13	0.11	0.18	nd	0.13	0.07	0.14	0.13	0.07	0.29	0.13	nd	0.29	0.10		
pp’-DDE	nd	0.82	0.87	0.80	0.45	0.68	0.96	0.70	0.69	0.62	0.34	0.21	0.31	0.19	0.10	0.17	0.18	0.14	nd	0.96	0.46	1.42	6.75
pp’-DDD	nd	0.20	0.27	0.25	0.14	0.24	0.41	0.25	0.55	0.27	nd	0.13	0.40	0.14	nd	0.13	0.18	0.20	nd	0.55	0.21	3.54	8.51
pp’-DDT	nd	0.09	0.11	0.09	0.10	0.08	0.30	nd	0.48	0.16	nd	0.08	0.16	0.08	nd	0.05	0.14	0.10	nd	0.48	0.11	1.19	4.77
∑DDTs	nd	1.11	1.25	1.13	0.69	1.00	1.67	0.95	1.73	1.04	0.34	0.41	0.82	0.41	0.10	0.35	0.50	0.44	nd	1.73	0.77		

nd: not detectable ; HCHs: sum of α-, β-, γ-, and δ-HCH; Chlordanes: sum of o-, a- and g-chlordane; Endosulfans: sum of Endosulfan –I, II and Endosulfansulfate; DDTs: sum of pp’-DDE, pp’-DDD and pp’-DDT; TEL, PEL [20,3]

Vietnam has not issued Guidelines for OCs levels assessment in freshwater sediment. Therefore, we used two sediment quality assessment values such as Threshold Effect Level (TEL) and Probable Effect Level (PEL) [1,3, 20] in order to assess our analytical results. TEL and PEL are two values used to evaluate the potential impacts of sediment – associated chemicals on various resources uses (e.g., aquatic life or wildlife consumers of aquatic life).

The TEL was calculated as the square root of the product (i.e., the geometric mean) of the lower 15th percentile concentration of the effect data set and the 50th percentile concentration of the no-effect data set. The PEL was calculated as the square root of the product (i.e., the geometric mean) of the 50th percentile concentration of the effect data set and the 85th percentile concentration of the no-effect data set. The TEL represents the upper limit of the range of sediment chemical concentrations that is dominated by no-effect data entries. Within this range concentrations of sediment-associated chemicals are not considered to represent significant hazards to aquatic organisms. The PEL represents the lower limit of the range of chemical concentrations that is usually or always associated with adverse biological effects. The geometric mean is used to account for the uncertainty in the distribution of the data sets [2].

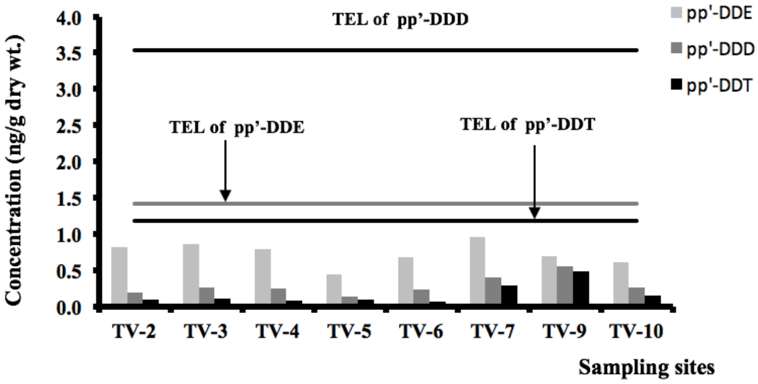

Figure 3: Comparison between degradation products of DDT in Thivai River sediments and TEL values respectively

Fig. 3 indicates that DDT and its metabolites levels in all sites in Thivai River are much lower than TEL value. It is similar to PCBs' case shown in Fig. 4.

Endosulfans were detected in almost all sediment samples in common levels due to their large usage in Vietnam agricultural activities. This substance import has just been prohibited since 2005 and endosulfans using in agricultural activities has also been banned since 2006 [13].

HCB, HCHs, Dieldrin were detected in all Thivai sediment samples with levels ranged 0.02 – 0.78, <0.004 – 0.25, and <0.007 – 0.88 ng/g dry wt., respectively (Table 2). Others OCs in sediment samples such as heptachlor, heptachlor epoxide, aldrin, endrin, endrin aldehyde and chlordanes were lower than LOD of the analytical method or very small value.

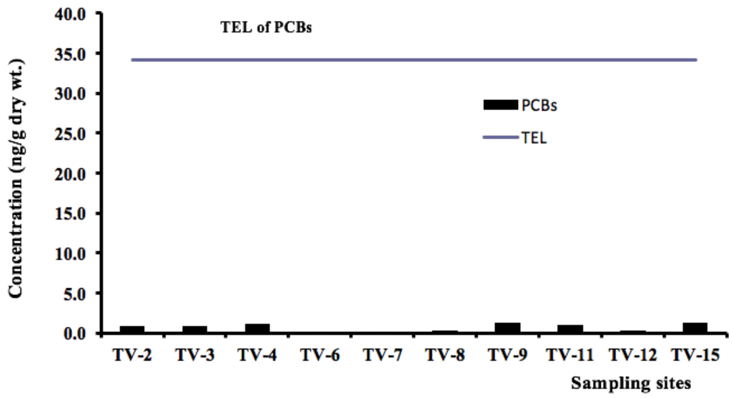

Figure 4: Comparison between PCBs concentrations in Thivai River sediments and TEL values

Contamination by CHLs in Vietnam was scarcely investigated compared to PCBs and DDTs. CHLs might have been used for termite control, wood preservation [32], and protective treatment for underground cables [33].

Table 3: Comparison of OCs levels between our sediment samples and various locations in the world (mean value in ng/g dry weight)

Location	Year	n	PCBs	DDTs	CHLs	HCHs	HCB	Reference
Vietnam								
Thivai river	**2009**	**18**	**0.43**	**0.77**	**nd**	**0.06**	**0.17**	Present study
Cantho (Mekong river)	2004	22	0.89	6.5	0.93	0.10	0.02	Minh et al., 2007b [16]
HCMC canals	2004	6	81	37	2.0	nd	6.60	Minh et al., 2007a [15]
Saigon-DongNai River	2004	9	6.80	5.6	0.28	0.01	0.24	Minh et al., 2007a [15]
SG-ĐN River-Estuary	2004	7	0.90	1.2	0.03	0.01	0.03	Minh et al., 2007a [15]
HCMC canal	2002	2	77	84	5.05	0.13	12.45	Kishida et al., 2007 [11]
HCMC canal	1996	10	223.2[a]	79.9	-	-	-	Phuong et al., 1998 [18]
Hochiminh (Urban areas)	1990	4	460	425 [b]	16.95[c]	7.68 [d]	-	Iwata et al., 1994 [8]
Hanoi (Urban areas)	1997	12	11	30	0.55[e]	0.80	0.03	Nhan et al., 2001 [17]
World								
Coastal Region (Singapore)	2003	13	73.9	6.7	-	18.1	-	Wurl and Obbard, 2005 [31]
Gao-ping River (Taiwan)	2000	12	1.43	0.78	-	2.49	-	Doong et al., 2008 [4]
Hong Kong	2004	6	4.2	5.6[f]	-	0.17 [g]	-	Wei et al., 2008 [30]
Hugli Estuary (India)	2003	10	0.8	0.96[e]	-	0.24	-	Guzzella et al., 2005 [6]
Korea (Masan Bay)	1997	20	15	13.6	-	-	-	Hong et al., 2003 [7]
China (Macao Harbour)	1997	1	340	1630	-	-	-	Kang et al., 2000 [10]
French (Mediterranean basin)	2010	10	293	41	-	7	-	A D Syakti et al., 2012 [35]
NorthEnd Lake (South Africa)	2013	13	2.23	-	-	-	-	E. Kampire et al., 2017 [36]
Alexandria (Egypt)	2010	49	24.6	19.3	-	1.1	-	MA Shreadah et al., 2012 [37]

nd: not detectable; -: data not available; [a]: As aroclor 1254 mixture; [b]: sum of pp'-DDE, pp'-DDD, pp'-DDT and op'-DDT; [c]: sum of t-chlordane, c-chlordane, t-nonachlor and c-nonachlor; [d]: sum of a-HCH, b-HCH and g-HCH; [e]: sum of t-chlordane, c-chlordane and t-nonachlor; [f]: Sum of op'-DDE, pp'-DDE, op'-DDD, pp'-DDD, op'-DDT and pp'-DDT; [g]: sum of a-HCH and g-HCH.

Comparison of OCs levels in Thivai sediment samples with those in previous studies in Vietnam is shown in Table 3. In general, DDTs, PCBs and other OCs levels in Thivai River sediments were several times lower than those in sediment samples collected from Mangroves, southern Vietnam [8], Hau River [15], and Saigon-Dongnai River [11,14, 18]. This indicates that OCs contamination level of Thivai River sediments is low and industrial zones and ports along riversides could be insignificant OCs contamination sources of Thivai River basin. In global comparison, the Table 3 shows PCBs, DDTs and HCHs levels in our study were much lower than those in Singapore, Korea, China and other locations in the world.

CONCLUSIONS

In this study, we reported for the first time the current state of OCs contamination in Thivai River basin sediments. Our analytical results could be suggested that OCs releases from industrial zones and ports activities along Thivai River are insignificant.

Although the OCs level in Thi Vai river sediment is relatively low, the existence of those substances in environment as well as their toxicity to human and other animals should be paid much attention. Moreover, because of the fact that Dong Nai and Ba Ria – Vung Tau provinces have been developing many industrial projects and ports along Thi Vai river, the monitoring, analyzing OCs in other subjects like waste water, exhaust fumes, wildlife should be paid more attention and invested in order to timely find out and limit the OCs sources in the future.

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HIỆN TRẠNG Ô NHIỄM CÁC CHẤT CƠ CHLOR KHÓ PHÂN HUỶ TRONG BÙN LẮNG SÔNG THỊ VẢI, MIỀN NAM VIỆT NAM

Tóm tắt. Báo cáo trình bày áp dụng phương pháp sắc ký khí ghép khối phổ (GC/MS) trong việc phân tích nhóm các chất cơ chlor (OCs) khó phân huỷ bao gồm cả PCB trong bùn lắng của lưu vực sông Thị Vải. Độ thu hồi của phương pháp phân tích dao động từ 72 – 129% đủ đáp ứng được tiêu chuẩn cho phân tích vết các chất cơ chlor. Nghiên cứu đã thu thập 18 mẫu bùn lắng dọc theo lưu vực sông Thị Vải bắt đầu từ họng xả của nhà máy Formosa (kênh Lò Rèn) xuống đến hạ lưu là hợp lưu của sông Thị Vải và sông Gò Gia trước khi đổ ra biển. Các kết quả nghiên cứu cho thấy hàm lượng các chất OCs trong bùn lắng (cao nhất 1,73 ng/g) của sông Thị Vải tương đối thấp khi so sánh với các khu vực khác ở Việt Nam và trên thế giới.

Keywords: OCs, bùn lắng, DDTs, Thị Vải, PCBs, GC/MS

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