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Seasonal, Spatial Variation, and Potential Sources of Organochlorine Pesticides in Water and Sediment in the Lower Reaches of the Dong Nai River System in Vietnam

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Abstract

The goals of the current study were (1) to examine seasonal and spatial variation of selected OCPs concentrations and (2) to identify potential sources of the pollutants in the lower reaches of the Dong Nai River system. Forty-eight water and sediment samples were taken from 12 stations in the dry and rainy seasons to determine the concentrations of dichlorodiphenyltrichloroethane and its metabolites (total DDTs), hexachlorocyclohexane isomers (total HCHs), heptachlor, aldrin, dieldrin, and endrin. The concentrations of total DDTs (0.30), total HCHs (0.29), Aldrin (0.068), heptachlor (0.04, μg L^{-1}) in water, and total DDTs (8.04), total HCHs (4.51), and Aldrin (1.52, μg kg^{-1}) in sediment were significantly higher in the rainy season than in the dry season (0.14, 0.12, 0.008, 0.009 in water and 3.49, 2.29, and 0.4 in sediment, respectively). Cluster analysis grouped 12 sampling stations into 2 groups, of which group 1 (3 stations) had higher concentrations of total DDTs, total HCHs, Aldrin, heptachlor, and dieldrin in both water and sediment than in group 2. Compositional analysis of total DDTs revealed that DDT residue could be decomposed significantly for the past years and that anaerobic decomposition could be predominant. Principal component analysis/factor analysis (PCA/FA) indicated that the potential sources of OCPs in the study stations could come from residential and agricultural areas located in the upper catchment or areas surrounding the studied stations. In short, OCPs concentration in the studies area could depend on seasonal, spatial variation, and transport of OCPs from upper parts or surrounding areas.

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Organochlorine pesticides (OCPs) are chlorinated hydrocarbons widely used over the past century all over the world to control pests for better crop yield and to protect public health by controlling malaria mosquitoes (Rahman 2013; Carvalho 2017). Globally, OCPs were banned or restricted for use a few decades ago (UNEP 2003) due to their toxicity to living organisms. Nevertheless, the presence of OCPs was detected in different environments, such as the oceans (Ma et al. 2015), the seas (Fuoco et al. 2009), water and sediment (Wang et al. 2003), the air (Nost et al. 2018), and the polar regions (Mangano et al. 2017). Due to their long persistence and toxicities to living organisms (Covaci et al. 2005; Singh et al. 2016), most countries in the world have developed national plans and strategies to implement their Stockholm Conventions in protecting the environment and humane health against the threat of Persistent Organic Pollutants (POPs), such as OCPs compounds (UNEP 2018).

When applied to any sites, OCPs are capable of being transported to the vicinity or far from the originally-applied sites through two main pathways, including atmosphere (Shen et al. 2005) and water-related (Leadprathom et al.

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2009). Water transport may cause the estuary and lowland areas polluted with OCPs (Rizzi et al. 2017; Iwata et al. 1994). This may indicate that potential sources of OCPs in the downstream reaches of a river could be partially derived from the upstream. Consequently, the OCPs concentrations in water and sediment in the lower part of a river may be affected by seasons due to varying rainfall, which may wash out or transport the OPCs-containing materials to the downstream area. The water stream, which is normally stronger in the rainy season than in the dry season, may carry more OCPs to the lower part, possibly increasing their concentration in the rainy season, compared with the dry season (Md Pauzi et al. 2015; Barasa et al. 2007). Nevertheless, these findings were not in line with other studies (Williams 2011). These inconsistent findings need additional studies to clarify.

In general, the main sources of OCPs in the lower parts of a river could come from agricultural, residential, and industrial activities (Md Pauzi et al. 2015; Eqani et al. 2011). As a result, Barasa et al. (2007) found that the concentrations of some OCPs varied greatly with sampling sites. Mutiyar and Mittal (2013) reported that OCPs residue varied with different stretches of the Ganga River in India, reflecting agricultural, industrial, and residential activities. A study by Imo et al. (2007) highlighted that the sources of some investigated OCPs in water and sediment could come from the residential area, commercial and naval ports, and agricultural activities. Similarly, local release of OCPs from agricultural, residential, and industrial areas was attributed to being the main sources of OCPs determining their spatial distribution in rivers (Md Pauzi et al. 2015; Eqani et al. 2011). Consequently, it is important to identify the main sources of OCPs in a river estuary, when conducting a study on OCPs assessment.

In Vietnam, the OCPs have been widely used in the period of 1960–1980 for various purposes. As much as 30% of total pesticides used in Vietnam in or before 2005 was persistent organic pollutants (MONRE 2006). Although some OCPs, such as DDTs, were banned from using for public health protection since 1994 (MONRE 2006), the aged OCPs could be present in Vietnam environments at varying concentrations. Because OCPs were normally applied to control pest for better agricultural production, agricultural fields could be a major source of OCPs. Nevertheless, a previous study by Minh et al. (2007) showed that DDTs concentrations in sediment collected in Ho Chi Minh City canals were much higher than those from other sites in the same study, indicating that other main sources of OCPs could be from residential or industrial areas. The findings from two studies by (Minh et al. 2007; Iwata et al. 1994) in the downstream parts of the Dong Nai River system showed that the concentrations of total DDTs and HCHs in sediment decreased with time. In contrast, Hoai et al. (2010) found that the concentrations of total DDTs in sediment taken from some rivers in Hanoi were higher than those reported in previous studies. Consequently, the authors assumed that DDTs were illegally used and then discharged into the study area.

Because residential and industrial areas, developed rapidly recently, could be potential main sources of the OCPs and because hypothetical illegal application of OCPs in the upper catchment could happen, the OCPs concentrations in sediment and surface water of the downstream part of the Dong Nai River system, Vietnam could be increased. This necessitates more study to clarify. Therefore, the current study was conducted in the lower reaches of the Dong Nai River system in 2006 and 2017. The study was designed to (1) examine the seasonal and spatial variation of OCPs concentrations and (2) identify potential sources of the pollutants in the study area. We hypothesized that (1) the OCPs concentrations in water and sediment in the lower reaches of the Dong Nai River system could be higher in the rainy season than in the dry season, (2) the OCPs concentrations could vary greatly with studied stations, and (3) potential sources of OCPs could be from upper-catchment transport and surrounding-area release.

Materials and Methods

Study Area

The current study was conducted in the lower reaches of the Dong Nai River system that are located in Can Gio district—Ho Chi Minh City, Vietnam (106°38′–107°11′E and 10°20′–19°50′N) (Fig. 1). The river system receives discharged water from a total catchment area of around 38,600 km^2, which belongs to three provinces, including Dong Nai, Binh Duong, and Ho Chi Minh City. In the upper reaches, there are a few tributaries collectively adding water to the river system, including Da Nhim River, Song Be River, La Nga River, Sai Gon River, Da Hoai River, and Vam Co River. In the lower reaches, there are two main streams, the Soai Rap River and the Long Tau River, together sharing total flowing water from the River System. In addition, two other tributaries, Vam Co River (tributary 1) and Thi Vai River (tributary 2), are connected to the main streams and add their water to the River system. There are two distinct seasons, the dry and rainy seasons, with total annual rainfall of 1700–2800 mm.

Study Setup

The lower reaches of the Dong Nai River system were geographically separated into three subregions: the mainstream, Tributary 1, and Tributary 2. The main stream is connected

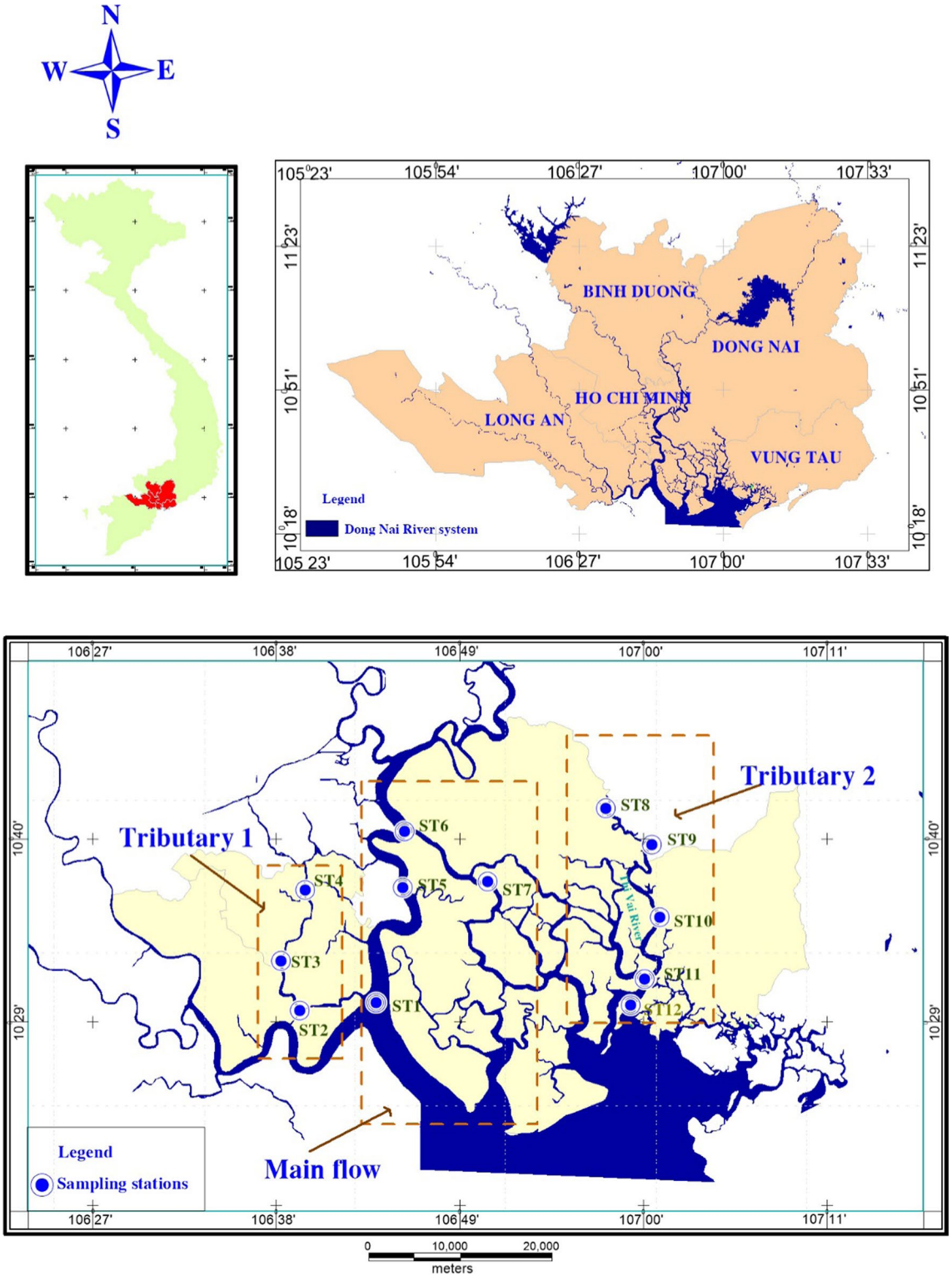

Fig. 1 Sampling stations and study map

directly to the river system and thus directly receives flowing water from the upper catchment of the system. The tributaries 1 and 2 are connected to the main flow through some small canals and directly add their water to the system. Twelve stations located in the three subregions were selected for taking water and sediment samples in the dry and rainy season in 2016 and 2017 for the current study. Stations 01, 05, 06, and 07 were located on the main flow; stations 02, 03, and 04 were on the tributary 1; and stations 08, 09, 10, 11, and 12 were on the tributary 2 (Fig. 1; Supplementary Table 1).

Sampling and Chemical Analysis

Totally, 48 water samples and 48 sediment samples were taken from the 12 sampling stations over the dry and rainy seasons in 2016 and 2017. Water samples were taken from the 0–50 cm surface layer using a Van Dorn water sampler and around 5 L for one sample were collected from 10 points within one station into a bottle with a firm cap. Sediment samples were taken from 0 to 10 cm layers using a Petersen grab into a polybag. All samples were immediately stored in an ice-chest at 4 °C and transported to the laboratory for OCPs analyses. In order to locate the 12 preselected stations for 4 sampling times in the 2 years, 2016 and 2017, a global positioning system (GPS) device was used.

Both water and sediment samples were analyzed for concentrations of six OCPs, including dichlorodiphenyltrichloroethane and its metabolites (DDTs, o,p′-DDT, p,p′-DDT, o,p′-DDE p,p′-DDE, o,p′-DDD, and p,p′-DDD), hexachlorocyclohexane isomers (HCHs, α-HCH, β-HCH, γ-HCH, and δ-HCH), aldrin, heptachlor, dieldrin, and emdrin, following procedure by (Nhan et al. 1998; Zheng et al. 2016; Nguyen et al. 2019). For water samples, a total amount of 1000 ml of the collected water sample was filtered with Whatman filter paper (i.d. 70 mm) to remove debris and suspended materials and then poured into a 2-liter separatory funnel. For the first liquid–liquid extraction (LLE), 60 ml of dichloromethane (DCM) was added and shaken vigorously for 10 min before two-phase separation. The water phase was drained from the separatory funnel into a 1000-ml beaker. The organic-phase was carefully poured into a flask containing 20 g of anhydrous sodium sulfate. The extract was concentrated to the volume of 2 ml using a vacuum rotary evaporator. The clean-up was performed using Florisil cartridges. The cartridges were eluted with 7 ml of hexane for PCBs congeners. Then the column was eluted with 12 ml of the mixture containing 80% of hexane and 20% of dichloromethane for the pesticide fraction. Finally, the eluted solution was concentrated under a gentle stream of nitrogen flow and then analyzed for the examined POPs using a Gas-Chromatography equipped with an Electron Capture Detector (GC-ECD) (Varian Co., U.S.A.). For sediment samples, the air-dried samples were extracted with 300 ml of acetone: n-hexane (1:1 v/v) for 16 h. The extracts were then combined and desulfurized through activated copper powder and then concentrated to a few milliliters in a rotary evaporator at a temperature of 35 °C. The remaining extract was transferred to the top of a glass column packed with 8 g of Florisil followed by elution with 40 ml of hexane for PCBs congeners and 120 ml of mixture n-hexane: DCM (4:1,v/v) for the pesticide fraction. Each fraction was concentrated under a flow of nitrogen and stored at 4 °C before analyzing using the GC-ECD system.

Statistical Analysis

Multivariate analyses (hieratical cluster analysis, principal component analysis, and factor analysis) were reviewed or used as robust methods for environmental studies, including rivers and streams (Kaplunovsky 2005; Phung et al. 2015). Hieratical cluster analysis (CA) was conducted on the entire data to classify 12 study stations into 2 groups (clusters) having similar OCPs concentrations. Principal component analyses and factor analysis (PCA/FA) were applied on each season and each group to investigate the sources of OCPs. In addition, PCA/FA was applied to the whole data to classify the 12 stations without differentiating seasonal and spatial variation. Details of these techniques used in the current study were described by Eqani et al. (2011).

Analysis of variance (ANOVA) following a one-way completely randomized design was conducted to compare the mean values of two seasons or of two groups. The overall ANOVA model was $\gamma_{ij} = \mu + \alpha_j + \in_{ij}$, where γ_{ik} is the response variable; μ is overall mean; α_j is the fixed effect of the jth seasons or groups; and $\in_{ij}$ is the random error with mean zero and having a normal distribution (Ott and Longnecker 2011). If ANOVA indicated significance at $p \leq 0.05$ for any OCP, Student's t test was applied to classify the seasons or groups. Relationships between water and sediment samples in terms of concentrations of OCPs were investigated using a simple linear regression model: $\gamma_i = \beta_1 + \beta_2 x_i + \in$, where γ_i is the concentrations of OCPs in water; x_i is the concentrations of OCPs in sediment; β_1 is an intercept, β_2 is a slope, and $\in$ is a random error. All statistical analyses were conducted using JMP 13 (SAS Institute Inc., Cary, NC). All figures were made using Sigmaplot 12 (Systat Software Inc.).

Results

OCPs Concentrations

Grouping of sampling stations based on spatial similarities/dissimilarities of six OCPs in both surface water and sediment was conducted using hierarchical cluster analysis (CA). Twelve sampling stations were grouped into two groups (Fig. 2). Group 1, including sampling stations ST3, ST4, and ST8, was located on the top of two tributaries. Group 2, consisting of other nine sampling stations, was located on the mainstream (4 stations) and on the lower part of the two tributaries (Fig. 1). Stations ST3 and ST4 were located close to the agricultural and residential areas, and station 08 was close to the agricultural and industrial areas. Station ST6 was close to a residential area; ST5 on residential and industrial areas, ST7 and ST12 were on the residential area; ST9, ST10, and ST11 were on residential and industrial; and ST1 and ST2 were on a residential area. In addition, the stations ST1, ST5, ST6, and ST7 could be affected by the mainstream from the upper parts of the Dong Nai River system. The stations ST2, ST9, ST10, ST11, and ST2 could be influenced by the flow from the upper parts of the two tributaries.

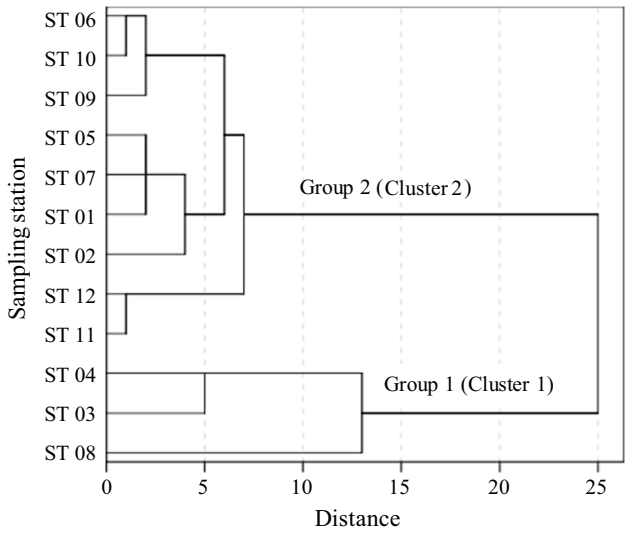

Fig. 2 Hierarchical Dendrogram showing 2 groups (clusters) of studied stations

Seasonal Variation

The method used to quantify the OCP concentrations in water and sediment was highly accurate with coefficients of determination of all standard curves higher than 0.996 (more information about Limit of Detection (LOD) and Limit of Quantification (LOQ) could be found in Supplementary Table 2). In water, the averaged concentration of total DDTs in the dry season was 0.137, varying from 0.02 to 0.37 (μg L^{-1}) and these of total HCHs, aldrin, heptachlor, dieldrin, and endrin were 0.11, 0.008, 0.009, 0.007, and 0.019 (μg L^{-1}) (Supplementary Table 3), respectively. The mean concentrations of total DDTs, total HCHs, aldrin, and heptachlor were significantly higher in the rainy season than those in the dry season (Table 1). The concentrations

Table 1 Mean concentration of water and sediment OCPs in two seasons and two groups

OCPs	Dry season		Rainy season		Group 1		Group 2	
	Mean	SE	Mean	SE	Mean	SE	Mean	SE
In water (μg L^{-1})								
Total DDTs	0.137^{b}	0.017	0.301^{a}	0.068	0.46^{a}	0.11	0.139^{b}	0.019
Total HCHs	0.107^{b}	0.017	0.292^{a}	0.037	0.34^{a}	0.06	0.151^{b}	0.020
Aldrin	0.008^{b}	0.003	0.068^{a}	0.007	0.06^{a}	0.01	0.029^{b}	0.005
Heptachlor	0.009^{b}	0.001	0.040^{a}	0.010	0.04^{a}	0.02	0.018^{b}	0.003
Dieldrin	0.007^{a}	0.004	0.024^{a}	0.008	0.04^{a}	0.02	0.008^{b}	0.002
Endrin	0.019^{a}	0.002	0.027^{a}	0.007	0.03^{a}	0.01	0.021^{a}	0.004
In sediment (μg kg^{-1})								
Total DDTs	3.49^{b}	0.45	8.04^{a}	1.29	11.8^{a}	2.03	3.75^{b}	0.35
Total HCHs	2.29^{b}	0.23	4.51^{a}	0.63	6.20^{a}	0.96	2.47^{b}	0.21
Aldrin	0.40^{b}	0.11	1.52^{a}	0.40	2.37^{a}	0.66	0.49^{b}	0.13
Heptachlor	1.01^{a}	0.19	3.58^{a}	1.30	5.94^{a}	2.43	1.08^{b}	0.15
Dieldrin	0.54^{a}	0.13	0.32^{a}	0.10	0.93^{a}	0.22	0.26^{b}	0.07
Endrin	0.97^{a}	0.14	1.40^{a}	0.23	1.64^{a}	0.42	1.03^{a}	0.11

SE standard error

Within a row, the mean concentrations of dry and rainy seasons (or group 1 and group 2) attached with the same letter are not significantly different from the other

of these OCPs in rainy season were 0.301, 0.292, 0.067, 0.04, 0.024, and 0.027 (μg L^{-1}), respectively. Similarly, in sediment, the concentrations of total DDTs, total HCHs, and aldrin in the rainy season were significantly higher than in the dry season. The concentrations of total DDTs, total HCHs, aldrin, heptachlor, dieldrin, and endrin in the rainy season were 8.04, 4.51, 1.52, 3.58, 0.32, and 1.40 (μg kg^{-1}), respectively (Supplementary Table 4). In the meanwhile, the concentrations of those pesticides collected in dry season were 3.49 for total DDTs, 2.29 for total HCHs, 0.40 for aldrin, 1.01 for heptachlor, and 0.54 for dieldrin (μg kg^{-1}). The concentrations of six OCPs in sediment were much higher than those in water.

Spatial Variation

The water concentrations of total DDTs (0.46), total HCHs (0.34), aldrin (0.06), heptachlor (0.04), and dieldrin (0.04 μg L^{-1}) of the group 1 were significantly higher than those in group 2 (0.14, 0.15, 0.029, 0.018, and 0.008 μg L^{-1}, respectively). Similarly, the sediment concentrations in group 1, including total DDTs (11.8), total HCHs (6.20), aldrin (2.37), heptachlor (5.94), and dieldrin (0.93) were significantly higher than those in the group 2 (3.75, 2.47, 0.49, 1.08, and 0.26, μg kg^{-1}, respectively). The water and sediment concentrations of endrin were not significantly different between the two groups.

Composition of Total DDTs

Six main metabolites and isomers of total DDTs, including *p,p′*-DDT, *o,p′*-DDT, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, and *p,p′*-DDD, were quantified, and the composition in percentage of *p,p′*-DDT, *o,p′*-DDT, DDD, and DDE are shown in Table 2. In general, the largest composition of total DDTs in water was composed of DDD, which was significantly higher in the rainy season (42.1%) than in the dry season (29.8%). In contrast, the DDE composition in water was significantly higher in the dry season (30.9%) than in the rainy season (13.5%). Two groups had similar DDTs and DDE compositions in water. In sediment, the DDD composition was significantly higher in the dry season (39.9%) than in the rainy season (26.5%). Group 1 had significantly higher DDE percentage (23.3%) than group 2 (12.8%). Other compositions of total DDTs in sediment were similar for the two groups and two seasons.

Relationships Between Water and Sediment Concentrations of OCPs

The water concentrations of total DDTs and HCHs were significantly and positively correlated with the sediment concentration in both dry and rainy seasons (Fig. 3a, b). For every 1 μg kg^{-1} of total DDTs in sediment, total DDTs in water increased 0.04 and 0.024 μg L^{-1} in the rainy and dry seasons, respectively. Similarly, total HCHs concentrations in water increased 0.05 and 0.024 μg L^{-1} in the rainy and dry seasons, respectively.

An increase in sediment concentration of aldrin also resulted in a significant increase in water concentration of aldrin in the rainy season but not in the dry season (Fig. 4a). In contrast, the water concentrations of heptachlor and endrin increased significantly with an increase in the sediment concentrations of these OCPs in the dry season, but not in the rainy season (Fig. 4b, d). There was no significant relationship between water and sediment concentrations of dieldrin in both dry and rainy seasons (Fig. 4c).

Table 2 The composition of total DDTs (%) in two seasons and two groups

Metabolites and isomers of DDTs	Dry season		Rainy season		Group 1		Group 2	
	Mean	SE	Mean	SE	Mean	SE	Mean	SE
In water								
p,p′-DDT	27.6[a]	2.0	27.6[a]	4.0	34.5[a]	3.7	25.3[a]	2.6
o,p′-DDT	11.7[a]	1.1	16.8[a]	3.0	8.9[a]	1.8	16.0[a]	2.0
DDD	29.8[b]	2.1	42.1[a]	4.5	37.8[a]	4.7	35.3[a]	3.1
DDE	30.9[a]	2.3	13.5[b]	3.3	18.8[a]	3.8	23.3[a]	2.9
In sediment								
p,p′-DDT	22.5[a]	2.2	26.1[a]	2.7	22.0[a]	3.3	25.1[a]	2.1
o,p′-DDT	22.9[a]	3.7	31.2[a]	2.7	26.1[a]	4.9	27.4[a]	2.7
DDD	39.9[a]	2.0	26.5[b]	1.9	28.6[a]	3.2	34.7[a]	1.9
DDE	14.7[a]	2.6	16.1[a]	1.9	23.3[a]	2.8	12.8[b]	1.7

SE standard error

Within a row, the mean concentrations of the dry and rainy seasons (or group 1 and group 2) attached with the same letter are not significantly different from the other

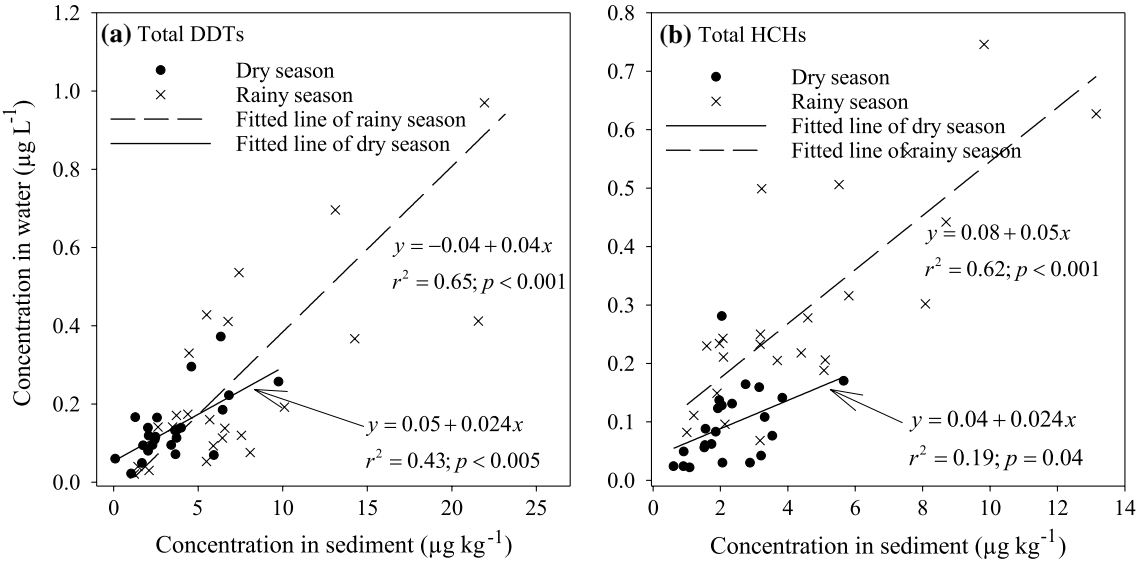

Fig. 3 Relationships between water and sediment concentration of total DDTs and total HCHs. Only significant relationships were shown with the fitted equation, the coefficient of determination (r^2), and probability (p)

Principal Component Analysis and Factor Analysis

Principal component analysis and factor analysis (PCA/FA) were used to examine latent factors representing six examined OCPs in water and sediment and to identify possible sources of the pollutants. PCA/FA extracted three principal components (PC) having eigenvalue greater than 1 for each season and each group (Table 3). The first three PCs, equivalent three varimax factors (VF) (latent factor) having eigenvalue greater than 1, cumulatively explained 75% of total variance for the dry season, 84.5% for the rainy season, 87.6% for group 1, and 69.9% for group 2 (Table 3).

For the dry season, three latent factors were identified, of which factor 1 explained 49.2% of total variance, factor 2 explained 16.5%, and factor 3 explained 9.2% (Table 3). The most important latent factor, factor 1, had high positive correlation coefficients (loadings) with total DDTs, total HCHs, aldrin, heptachlor, dieldrin, and endrin in sediment. The second most important factor, factor 2, had high positive loadings with five OCPs in water, and factor 3 had high loadings with 1 OCP in water and another 1 OCP in sediment. Quite different from the dry season, the first factor for the rainy season had high loadings with total DDTs, heptachlor in water, and total DDTs, aldrin, heptachlor, dieldrin, and endrin in sediment. The second factors had high loadings with total HCHs, aldrin in water, and total DDTs, total HCHs, and aldrin in sediment. The third had high loadings with 3 OCPs in water and 1 OCP in sediment.

For group 1, three factors were extracted. The first had high loadings with total DDTs, total HCHs, heptachlor in water and total DDTs, aldrin, heptachlor, dieldrin, and endrin in sediment. Factor 2 had high loadings with 2 OCPs in water and another 2 in sediment. Factor 3 had high loadings with 3 OCPs in water. For group 2, factor 1 had high loadings with total HCHs, aldrin, heptachlor in water and total DDTs, total HCHs, aldrin, and endrin in sediment. Factor 2 had high loadings with 3 OCPs in water and 1 OCP in sediment. Factor 3 had high loading with only 1 OCP in sediment.

Figure 5 showed that two PCs were extracted when PCA/FA was applied over the entire data. PC 1 accounted for 66.6% and PC2 accounted for 15.2% of total variance. Variance in OCPs in water and sediment collected from 12 study stations in the dry season was much smaller than that in the rainy season. The dry season was located on the negative direction of PC2, whereas the rainy season was on the positive direction of PC2. Group 1 in the rainy season had the greatest variance in OCPs concentration.

Discussion

Status of OCPs

In Water

An overall pattern (averaged over 2 seasons) of the accumulative concentration of the examined OCPs in the current study was total DDTs > total HCHs > Aldrin > heptachlor > endrin > dieldrin (Table 1). These concentrations of total DDTs, total HCHs, dieldrin, and endrin were lower than the guideline values (1, 2, 0.03, and 0.6 μg L^{-1}, respectively) for drinking water (WHO 2018). The concentrations of aldrin in the dry season was lower, but in the rainy season

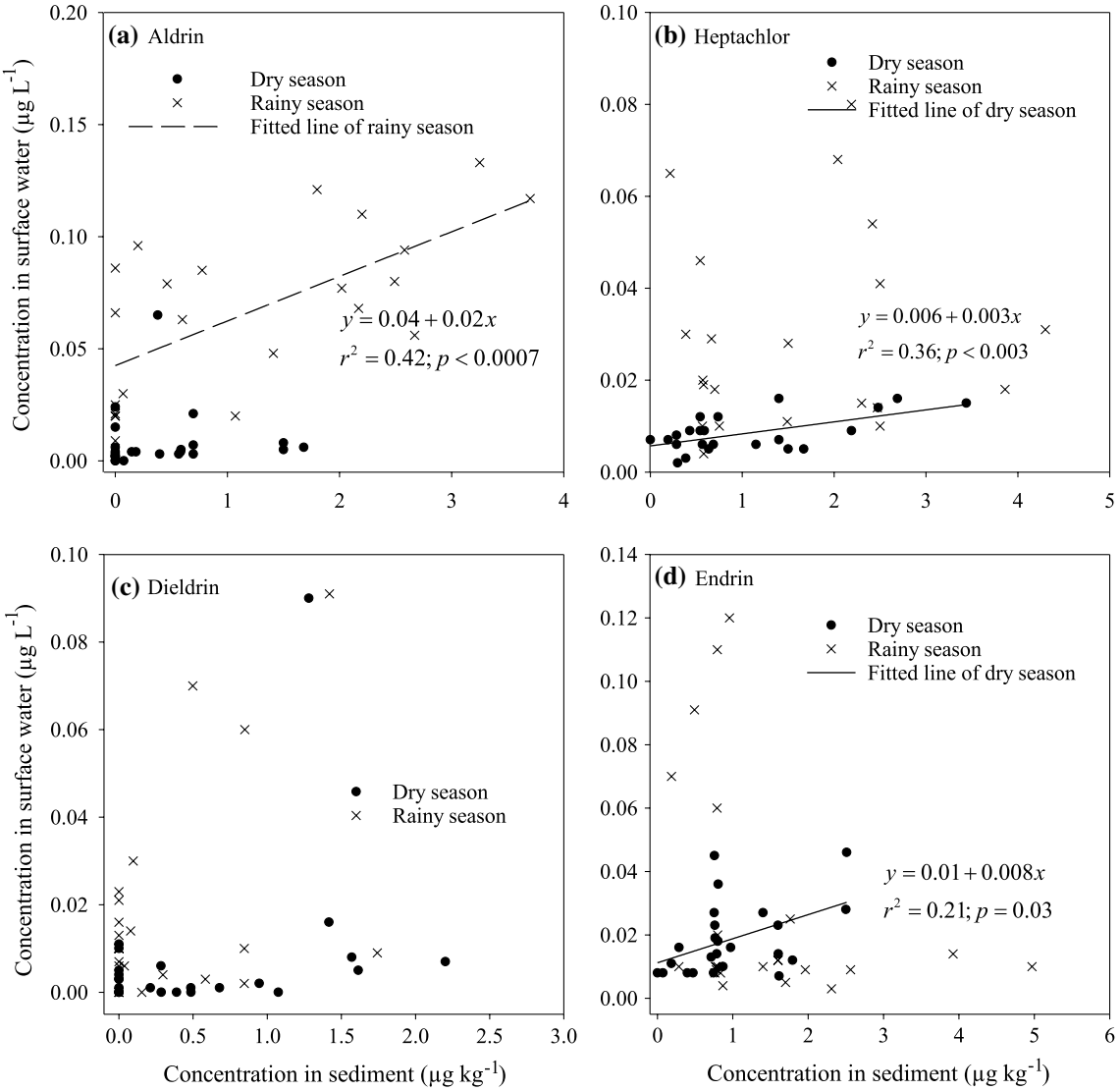

Fig. 4 Relationships between water and sediment concentration of aldrin, heptachlor, dieldrin, and endrin. Only significant relationships were shown with the fitted equation, the coefficient of determination (r^2), and probability (p)

was higher than the guideline values (0.03 µg L^{-1}). The concentrations of total HCHs, aldrin, heptachlor, and dieldrin in the current study were lower than values reported by Behfar et al. (2013) in Karun River, Iran. The concentrations of Aldrin, heptachlor, and dieldrin in the dry season of the current study were lower but in the rainy season were higher than averaged values reported from a few rivers in Japan (Imo et al. 2007).

An important finding from the current study is that the mean concentrations of total DDTs and total HCHs in water from the current study were higher than the findings by Iwata et al. (1994) in the lower reaches of the same river system. This may indicate that recent addition of the pollutants in the upper catchment or areas surrounding the studied stations could happen. Because DDTs and HCHs could be derived from residential, industrial, and agricultural areas, recent rapid development of agriculture and industry as well as the residential population in the areas surrounding the studied stations or in the upper catchment of the Dong Nai River system could release a considerable amount of these pollutants (illegal application), polluting the water in the studied area. The positive and significant relationship between water and sediment concentrations of these two pollutants (Fig. 3a, b) also may indicate that total DDTs and total HCHs in water could come from sediment through diffusion, based on concentration gradient or resuspension of sediment.

Table 3 Loadings of OCPs on different latent factors formed from PCA/FA analysis of two seasons and two groups

Parameters	Dry season			Rainy season			Group 1			Group 2		
	VF1	VF2	VF3	VF1	VF2	VF3	VF1	VF2	VF3	VF1	VF2	VF3
WT-total DDTs	0.53	0.67	0.10	0.53	0.36	0.67	0.70	0.26	0.57	0.18	0.77	−0.25
WT-total HCHs	0.18	**0.85**	0.19	0.46	0.74	0.26	0.67	0.69	0.12	0.71	0.43	−0.20
WT-aldrin	−0.15	**0.80**	0.36	0.16	**0.91**	−0.14	0.30	**0.90**	−0.07	**0.87**	0.16	−0.18
WT-heptachlor	0.28	0.62	−0.25	**0.86**	0.15	0.10	**0.87**	0.19	0.05	0.65	0.03	−0.08
WT-dieldrin	0.25	0.20	0.76	0.20	0.15	**0.89**	0.18	0.11	**0.92**	0.07	0.74	0.05
WT-endrin	0.37	0.73	−0.12	−0.23	−0.08	**0.88**	−0.20	−0.04	**0.90**	−0.03	**0.92**	0.04
SE-total DDTs	**0.90**	0.34	0.08	0.57	0.58	0.48	0.73	0.43	0.43	0.68	0.47	0.35
SE-total HCHs	**0.83**	0.23	−0.06	0.32	**0.87**	0.21	0.45	0.72	0.17	**0.88**	0.03	0.29
SE-aldrin	**0.93**	0.24	0.01	0.74	0.53	0.19	**0.93**	0.28	0.07	0.70	0.17	0.40
SE-heptachlor	**0.81**	0.19	−0.08	**0.88**	0.31	0.15	**0.93**	0.15	−0.05	0.46	0.61	0.41
SE-dieldrin	**0.86**	0.00	0.25	**0.80**	0.24	0.06	0.54	−0.72	−0.10	0.00	−0.07	**0.85**
SE-endrin	0.60	0.15	−0.58	**0.88**	0.25	−0.15	**0.97**	0.02	−0.07	0.57	−0.37	0.22
Eigenvalue	5.91	1.98	1.11	6.92	2.04	1.18	6.62	2.19	1.69	4.86	2.27	1.26
% total variance	49.2	16.5	9.2	57.6	17.0	9.9	55.2	18.3	14.1	40.5	18.9	10.5
Cumulative percentage variance	49.2	65.8	75.0	57.6	74.6	84.5	55.2	73.5	87.6	40.5	59.4	69.9

SE standard error; *WT* water sample; *VF* varimax factor

Bold numbers are those greater than 0.75, and underlined numbers are those greater than 0.5 and smaller than 0.75

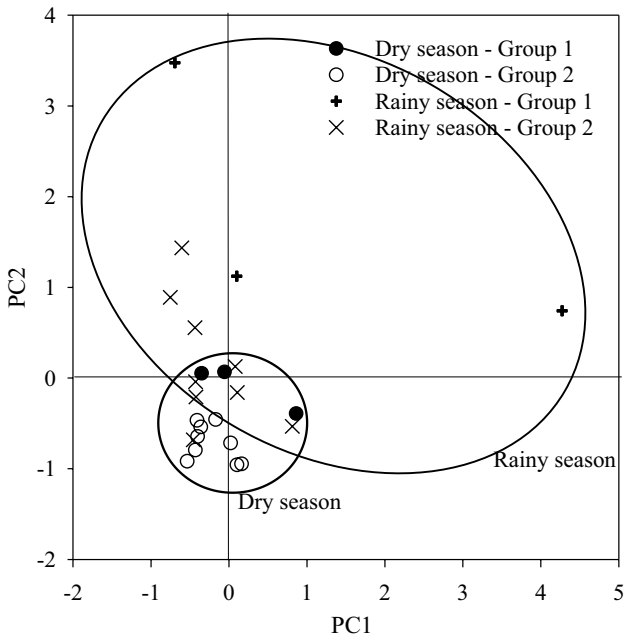

Fig. 5 Grouping of 12 studied stations based on principal component analysis/factor analysis applied to the entire data

In Sediment

The overall accumulative pattern of the examined OCPs concentration in the current study followed an order total DDTs > total HCHs > heptachlor > endrin > aldrin > dieldrin (Table 1). These indicate that total DDTs and HCHs were the most OCP pollutants in the studied area. In the current study, the concentrations of total DDTs in sediment in the dry season (3.49) and in the rainy season (8.04, μg kg^{-1}) were within the range (0.31–274 μg kg^{-1}) of estuarine and marine sediments taken from some parts from northern Vietnam (Hong et al. 2008). These numbers were similar to the findings from studies in Taiwan (0.53–11.4 μg kg^{-1}) (Doong et al. 2002), in China (1.36–8.99 μg kg^{-1}) (Hong et al. 1999) but higher than those in west coast of Sri Lanka (0.09–1.6 μg kg^{-1}) (Guruge and Tanabe 2001) and lower than those reported in Indian (3–119 μg kg^{-1}) (Bhattacharya et al. 2003).

Iwata et al. (1994) reported that the DDTs concentrations varied from 1 to 47 (μg kg^{-1}) and HCHs concentration from 0 to 7 (μg kg^{-1}) from the lower reaches and Minh et al. (2007) found that total DDTs concentration varied from 0.4 to 5.4 (mean = 1.2 μg kg^{-1}) and HCHs below 0.005 (μg kg^{-1}) from the estuary part of the same Dong Nai River system. A general pattern from these two studies was a decrease of DDTs and HCHs concentrations in sediment with time. Nevertheless, the current study found slightly higher DDTs and HCHs concentrations compared with the findings by Minh et al. (2007). This could suggest that either random error or recent release of OCPs from areas surrounding the study areas or from the upper catchment of the Dong Nai River system could happen. This finding was in line with the results about water concentrations of the two pollutants,

which were higher in the current study than in the previous study by Minh et al. (2007).

Technical-grade DDT generally contains 75% *p,p′*-DDT, 15% *o,p′*-DDT, 5% *p,p′*-DDE, and < 5% others (Yang et al. 2005; Eqani et al. 2011). The composition of *p,p′*-DDT in sediment (Table 2) in both seasons and both groups were below 27%, indicating that decomposition of original DDTs could happen significantly for the past years. Meanwhile, Minh et al. (2007) found that the composition of *p,p′*-DDT was approximately 10% in the estuary sites and was 20% in the Ho Chi Minh city canals (the upper part). The higher composition of *p,p′*-DDT in sediment collected from the lower reaches of the Dong Nai River for the current study compared with that by Minh et al. (2007) may suggest that either transport of aged DDTs from Ho Chi Minh city canals or of newly added DDTs from upper parts or from surrounding areas could happen for the recent years. DDTs could be biodegraded into DDD or DDE, depending on the environments such as anaerobic and aerobic conditions, respectively (Doong et al. 2002). As a result, the ratio of (DDD + DDE)/total DDTs > 0.5 was used to assess the long-term degradation of/or indicate aged DDTs (Hitch and Day 1992; Hong et al. 1999; Minh et al. 2007). The current study found that the ratios in sediment in both reasons and both groups were around 0.5, indicating that total DDTs from some studied stations were well degraded, whereas those from the others could be recently added. With the ratios below 0.45 (detail data not shown), the study stations, ST6, ST12, and ST8, may be replenished with newly added DDTs. Moreover, the recent input of DDTs to sediment in some studied stations could be inferred from the ratio of DDT/DDE > 0.5 (Strandberg et al. 1998), which was greater than 2 in two seasons and two groups (Table 2). This finding is in good agreement with a study by Minh et al. (2007) on the same river system. The ratios in water were higher than 0.5, indicating that some sources of these DDTs could be driven from newly-added DDTs areas. In addition, the composition of DDD in sediment in two seasons and two groups was higher than that of DDE (Table 2), indicating that anaerobic decomposition of total DDTs could happen predominantly. The ratio of DDD/DDE in water was almost equal 1 (in the dry season) or greater than 1 (in the rainy season and two groups), indicating that, in addition to the upper catchment transport, total DDTs in water in the rainy season could partially come from anaerobic environments, such as the river sediment (Doong et al. 2002; Bossi et al. 1992).

Seasonal Variation and Sources of OCPs

Seasonal variation of OCP concentration in water could greatly depend on rainfall, which transports pollutants from either upper catchment or surrounding areas and deposits them in the lower reaches of a river, resulting in higher OCPs concentrations in the rainy season than in the dry season. This is in line with the findings of the current study that the concentrations of total DDTs, total HCHs, aldrin, heptachlor in water, and total DDTs, total HCHs, aldrin in sediment were significantly higher in the rainy season than in the dry season (Table 1). These findings were in agreement with the findings from another study (Md Pauzi et al. 2015). Barasa et al. (2007) found that the concentrations of Aldrin, dieldrin, and DDT in sediment collected in the rainy season were higher than those in the dry season. They explained that runoff from agricultural and industrial areas stronger in the rainy season than in the dry season to transport OCPs to the study area may account for the difference. Similarly, Ramesh et al. (1990) reported that the concentrations of HCHs and DDTs in water in wet season was higher than in the dry season and that the difference between the two seasons was attributed to the flowering season of the rice crop. Nevertheless, the higher OCP concentration in the rainy season than in the dry season was not always in agreement with other studies. For example, Williams (2011) reported that OPCs concentration was higher in the dry season than in the rainy season, and the author attributed to more agricultural and domestic activities in the dry season than in the rainy season.

Seasonal variation could be was reflected with correlation coefficients between sediment and water concentrations of total DDTs and total HCHs (Fig. 3a, b), higher in the rainy season than in the dry season. This relationship was even significant in the rainy season but insignificant in the dry season on Aldrin concentration (Fig. 4a). These relationships could be explained that stronger water stream during the rainy season may carry the pollutants from upper parts or surrounding areas to deposit them in the studied stations or resuspend the pollutants already settled in sediment back to surface water. These indicated that the OCPs concentrations in the water and sediment in the current study could be derived from two main sources, including (1) transport from the upper catchment of the Dong Nai River system and (2) release from surrounding areas. Similar sources of OCPs in the water and sediment, taken from the lower part of the King Rivers, North-East Victoria, Australia, were identified by McKenzie-Smith et al. (1994). The original sources could come from agricultural fields, industrial, and residential areas (Mutiyar and Mittal 2013). The upstream transport was reported by a number of studies (Mutiyar and Mittal 2013). Takeoka et al. (1991) found that a small portion of HCHs applied to the catchment area was transported for a long distance to the sea and that the concentration of HCHs could be largely affected by the application of the pesticides during flowering season of paddy rice. OCPs transport from the upper reaches and their deposition in the studied area through a stronger water flow during rainy season could result in greater variance in OCP concentrations

among the twelve study stations, compared with that during the dry season (Fig. 5).

Principal component analysis and factor analysis (PCA/FA) indicated that there were three main sources of OPCs contributed to the water and sediment of the studied area (Table 3). In the dry season, the most important source, VF1 (occupying 49.2% of total variance) could be represented by all 6 OPCs in sediment. Historical transport of the OCPs from agricultural, residential, and industrial areas and deposition in the studied area could be the primary sources. Having strong loadings with total DDTs, total HCHs, Aldrin, heptachlor, and endrin in water, the second most important source, VF2 (equal 16.5% of total variance) could be the OCPs source of temporary transport of OCPs from upper parts or surrounding areas. In the rainy season, the varimax factor 1, explaining 57.6% of total variance, could be represented by DDTs and heptachlor in water and DDTs, Aldrin, heptachlor, dieldrin, and endrin in sediment. These may suggest that the main sources of OCPs in the rainy season could be from both historical deposition and recent transport from upper parts or surrounding areas. Similarly, having high loadings with total HCHs, Aldrin in water, and total DDTs, total HCHs, Aldrin, in sediment (Table 3), the second source of OCPs in the rainy season could be derived from a mixed history and recent transport from the upper parts or surrounding areas. These indicate that rainy season may cause runoff to the upper catchments and surrounding areas and transport the leached materials to deposit them into the study areas.

Spatial Variation and Sources of OCPs

Two groups reflecting the difference in OCPs concentrations in water and sediment were established using cluster analysis (Fig. 2). While DDTs could be used for crop pest and mosquito control, HCHs could be used for pest control in some main crops in Vietnam as rice, maize, and sorghum. Aldrin, heptachlor, and dieldrin could be used to control termites in the residential areas. The concentrations of total DDTs, total HCHs, Aldrin, heptachlor, and dieldrin in water and sediment were significantly higher in group 1 than group 2 (Table 1). In addition, PCA/FA analysis (Table 3) showed that total DDTs, total HCHs, heptachlor in water, and total DDTs, aldrin, heptachlor, dieldrin, and endrin in sediment could describe the most important latent factor (pollutant source 1) of group 1. These may reveal that agricultural and residential areas surrounding the sampling stations ST3, ST4, and ST8 may release a considerable amount of these OCPs into these stations, which were located on the top of two investigated tributaries (Fig. 1). Compared with group 1, group 2 had lower concentrations of five OPCs in both water and sediment, indicating that it received a smaller amount of OCPs deposited. The concentration of total DDTs in Ho Chi Minh City canals, the one upper part of the Dong Nai River system, varied from 12 to 72 μg kg^{-1} (Minh et al. 2007). These numbers were higher than the numbers in group 2, which included nine studied stations located on the mainstream of the Dong Nai River system and in the lower parts of two investigated tributaries. Because of decomposition, DDTs from the stations ST1, ST5, ST6, and ST7 of current study could be derived from the upper parts of the Dong Nai River system through historical or recent riverine transport. Chau (2006) reported that some OCPs in the sediment could be transported from a long distance to their studied area, the Pearl River delta area. Nevertheless, OCPs sources for water could reflect the current riverine transport, whereas those for sediment could be a consequence of historical and recent transport and deposition of agricultural runoff and municipal wastewater in the current study.

In water, OCPs may tend to be absorbed onto suspended sediments (O'Sullivan and Megson 2014), which eventually deposits to the bottom of a river. This may create a connection between water concentrations and sediment concentrations. Consequently, the current study found a significant correlation in OCPs between water and sediment in both seasons (Figs. 3 and 4). As discussed above, the main sources of some OCPs for group 2 could be from upper parts located in/or close to residential and agricultural areas and for group 1 from surrounding areas. Runoff and transport of these OCPs to the studied stations may account for the correlation coefficients of OCPs in water and in sediment significantly higher in the rainy season than in the dry season (Fig. 3).

Conclusions

The presence of six OCPs investigated in the current study was detected at different concentrations in both water and sediment in the lower part of the Dong Nai River system. Total DDTs and HCHs were present in water and sediment at a higher level than the other OCPs. The concentrations of total DDTs, total HCHs, aldrin, heptachlor, dieldrin in water and total DDTs, total HCHs, and Aldrin in sediment were significantly higher in the rainy season than in the dry season. Group 1, including three stations located on the top of two investigated tributaries, had significantly higher concentrations of total DDTs, total HCHs, Aldrin, heptachlor, and dieldrin in both water and sediment than group 2. The relationship in DDTs and HCHs concentrations between water and sediment was significant. Compositional analysis of total DDTs revealed that decomposition of original DDTs in the study stations could happen significantly for the past years, and that anaerobic decomposition of total DDTs could happen predominantly. PCA/FA analyses indicated that the main sources of OCPs in the study areas could come from the upper catchment and surrounding areas close

to the studied area through riverine transport and that OCPs in the study area varied greater in the rainy season than in the dry season. In short, OCPs concentration in the lower reaches of a river in both water and sediment could depend on seasonal, spatial variation, and transport of OCPs from upper parts or areas surrounding the lower reaches of the Dong Nai River system.

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