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Recent levels of organochlorine pesticides and polychlorinated biphenyls in sediments of the sewer system in Hanoi, Vietnam

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Abstract. The occurrence, temporal trend, sources and toxicity of PCBs and organochlorine pesticides were investigated in sediment samples from the sewer system of Hanoi City, including the rivers Nhue, To Lich, Lu, Set, Kim Nguu and the Yen So Lake. In general, the concentrations of the pollutants followed the order DDTs > PCBs > HCHs (β-HCH) > HCB. However, the pollution pattern was different for the DDTs and PCBs when the sampling locations were individually evaluated. The concentrations of the DDTs, PCBs, HCHs, and HCB ranged from 4.4 to 1100, 1.3 to 384, <0.2 to 36 and <0.2 to 22 ng/g d.w., respectively. These levels are higher than at any other location in Vietnam. Compared to measurements from 1997, the DDTs, PCBs, β-HCH and HCB levels show an increasing trend with DDT/DDE ratios, indicating very recent inputs into the environment although these persistent compounds are banned in Vietnam since 1995.

Key words: Vietnam, PCBs, DDTs, HCHs, Sediment, Temporal trend.

Capsule:

Organochlorine pesticides and PCBs were studied in sediments of the sewer system in Hanoi.

1. Introduction

Covering an area of some 1000 km^2 and having a population of 3.5 million people, Hanoi (the capital and second largest city of Vietnam) and its vicinity is the major industrial and economic region in North Vietnam. During the recent decades, the fast development of industry in conjunction with the high population growth have lead to toxic chemicals to enter the rivers of the city as the industrial, medical and domestic wastes are released untreated (Duong et al., 2008; Hoai et al., 2009). In addition, the deterioration of the rivers and the reduction of streambed have turned these rivers to be the open sewers and the principal pollution sources in the city (Hanoi DOSTE, 2003; GHK, 2005; Hanoi Water Discharge Company, 2006), thereby also posing a long-term threat to groundwater that is used for drinking water production (Giger et al., 2003; Duong et al., 2003; Berg et al., 2007, 2008; Norrman et al., 2008).

Flowing inside the Hanoi City, mainly to the south and southeast, with a total length of about 70 km , the rivers To Lich, Lu, Set, Kim Nguu, and a part of the Nhue River, serve as important open sewer system for the drainage of rainwater and municipal wastewater, but are also used for agricultural irrigation in urban and suburban areas. It was reported that 95% of the capital's wastewater effluents are discharged without treatment and an estimated 450 000 m^3/day are discharged untreated into the rivers Lu, Set, To Lich and Kim Nguu in Hanoi City (Hanoi Water Discharge Company, 2006).

Persistent organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were widely used in Vietnam since the 1960s for different purposes in agriculture, industry and public health (Sinh et al., 1999; NIP-Vietnam, 2006). Since 1995, the use of PCBs and some OCPs such as dichlorodiphenyltrichloroethane (DDTs), hexachlorbenzene (HCB) and hexachlorocyclohexans (HCHs) are officially banned in Vietnam (Sinh et al., 1999). However, these toxic chemicals are still observed in Vietnam at higher concentrations than in other southeast Asian countries (Minh et al., 2006; Monirith et al., 2003; Müller et al., 2008) and are currently still detected at elevated concentrations in various environmental compartments in Hanoi City (Kishida et al., 2007; Toan et al., 2007a,b) and also in human breast milk (Minh et al., 2004) of Hanoi citizens. Concentrations of PCBs and OCPs in different environmental compartments in Vietnam in general and Hanoi City in particular were evaluated reviewed by Minh et al. (2008). However, knowledge on the contamination levels in sewer systems of medium to large Asian cities like the Hanoi City are still limited.

Understanding the contamination status of OCPs and PCBs as well as their potential toxic effects on the sewer system in Hanoi City is very significant in order to provide information for the public and environmental authorities to protect the environment and the ecological systems of the city. Concentrations of these pollutants in sediment may give considerable information on their occurrence, long-term temporal trend, sources, and toxic assessment. Therefore, this study was conducted to investigate the concentrations of a wide spectrum of persistent organic pollutants, including PCBs and 17 OCPs in sediments of the sewer system of Hanoi City. In addition, the temporal trend of the pollution, by comparing with previous measurements, the possible sources, and the potential toxicity to the aquatic environments are discussed in this study.

2. Material and methods

2.1. Sample collection

Twenty-two sediment samples were collected on 18 May 2006 from the sewer system in Hanoi City, including the five rivers Nhue, To Lich, Lu, Set, Kim Nguu and the Yen So Lake (Fig. 1). A fraction of the Set River and the Kim Nguu River is discharged into the Yen So Lake. The samples were collected in urban (TL1, TL2, TL3, TL4, L1, L2, L3, S1, S2, KN1, KN2) and suburban districts (TL5, TL6, TL7, N1, N2, YS1-6). After collection with a stainless steel grab sampler (Van Veen Grab), river and lake sediments were wrapped in aluminum foil and shipped within 1 hour to the laboratory (CETASD). Upon arrived at the laboratory, the samples were air-dried, ground, sieved to 1 mm, and stored at -20 °C until analysis.

2.2. Chemical analysis

Seven PCB congeners (IUPAC numbers: PCB 28, 52, 101, 118, 138, 153 and 180) and a set of 17 organochlorine pesticides were analyzed. Those 17 pesticides are dichlorodiphenyltrichloroethane compounds (DDTs: o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'-DDT, p,p'-DDT); hexachlorocyclohexanes (HCHs: α-HCH; β-HCH; γ-HCH); chlordane compounds (CHLs: trans-chlordane, cis-chlordane, oxychlordane); aldrin; hexachlorbenzene (HCB); heptachlor; cis-heptachloroepoxide and mirex. The total concentration of PCB (PCBs) was calculated based on the sum of seven PCB congeners by multiplication with the value of four, which corresponds to the theoretical contribution of those congeners to Aroclor 1254 (Sauvain et al., 1994; Froescheis et al., 2000).

Sediment samples were analyzed for PCBs and OCPs at CETASD applying the EPA 3620B, EPA 8082, and EPA 8081A methods (EPA, 1996) with slight modifications for sample

100 extraction and extract cleanup. Briefly, 20 g of dry sediment was spiked with 10 ng/g
101 surrogate standards (PCB congener 209 and p,p'-DDT-^{13}C), ultrasonically extracted for 5
102 minutes, and then shaken for 2 hours with 60 ml of acetone/n-hexane (1:1, v/v). After
103 completely evaporating the solvents by vacuum and taken up in 2 ml n-hexane, the extract
104 was divided into two 1-ml fractions, which were used to determine PCBs and OCPs,
105 respectively. The clean up step was conducted in the same manner for both fractions.
106 Pigments, humic acids, etc. were removed from the extracts by concentrated H_2SO_4 (98%).
107 This step was repeated several times until the n-hexane layer became colorless. The extracts
108 then were concentrated under a gentle N_2 stream to 1 ml and were further purified on a florisil
109 cartridge (1 g, 6 ml). Non-polar compounds such as PCB congeners, HCB, and p,p'-DDE
110 were isolated from the first fraction after elution with 4 ml of n-hexane. Separately, PCBs and
111 OCPs were isolated from the second fraction after elution with 7 ml of acetone/n-hexane (1:9,
112 v/v). The sulfur-containing substances were removed by subsequently adding several
113 activated copper slices to the obtained solutions and were kept 1 hour until the black sulfur
114 soot no longer appeared on the copper slices. The purified fractions were then spiked with
115 internal standard (1,1-dibromundecane for PCBs calculation, and phenanthrene-d_{10} and
116 chrysene-d_{12} for OCPs calculation), concentrated under gentle N_2 stream to 0.5 ml, and
117 submitted to GC analysis. A 2-µl volume of the first purified fraction, which mainly contains
118 PCBs, was injected on the GC/ECD system (Shimadzu GC 17A, ECD Ni^{63}) for quantification
119 of PCBs. A 2-µl volume of the second fraction was injected on the GC-EI-MS system
120 (Shimadzu GC/MS QP2010) for quantification of OCPs. A DB-5 capillary column (30 m x
121 0.25 mm x 0.25 µm) with helium as a carrier gas at a flow rate of 1.8 ml/min was applied for
122 the separation of OCPs and PCBs on the GC system. The determination was carried out at
123 injector and detector temperatures of 270 and 300 °C, respectively. The GC temperature
124 program for PCBs separation was set to 120 °C before increasing to 200 °C at 10 °C /min,

125 then to 230 °C at 2 °C /min, and finally to 300 °C at 7 °C /min where temperature was
126 maintained for 10 minutes. The oven temperature program for OCPs analysis was 70 °C (1
127 min); 20 °C /min, 160 °C; 2 °C /min, 250 °C; 5 °C /min, 300 °C (5 min).
128
129 Relative sediment concentrations of PCBs and OCPs in this study are expressed on a dry
130 weight (d.w.) basis (not adjusted to recovery rates). Cluster ions were monitored in the mass
131 spectrometer at m/z 246, 219, 235, 235, 373, 373, 387, 263, 284, 272, 353 and 272 for OCPs:
132 DDE, DDD, DDT, HCH, trans-chlordane, cis-chlordane, oxychlordane, aldrin, HCB,
133 heptachlor, cis-heptachloroepoxide and mirex, respectively. In addition, mass traces m/z of
134 247, 188 and 240 were monitored for p,p'-DDT-^{13}C, phenanthrene-d_{10} and chrysene-d_{12},
135 respectively. The method limits of quantification (LOQs) were 0.6 ng/g for PCBs and 0.1 to
136 0.2 ng/g for OCPs. Half of the quantitative limit levels were used to calculate the means,
137 when the measured values were below the limit values.
138
139 *2.3. Quality assurance*
140 The quality assurance consisted of the analysis of method blanks, reproducibility and
141 repeatability tests, as well as the analysis of certified reference material. Recovery rates (72 to
142 121%) were obtained for all compounds for the spiked samples. The certified reference
143 materials (CRM No. 846-050 for OCPs and CRM-No. 911-050 for PCBs, Resource
144 Technology Corp.-RTC, USA) were used to validate the analytical method. The results of
145 total PCBs from the five tests (1.18±0.06 mg/kg) were in good agreement with the certified
146 value (1.28 mg/kg). Similarly, the results of OCPs from the six tests were in good agreement
147 with the certified values with average relative deviations ranging between 0.6 to 19%.
148
149 *2.4. Total organic carbon*

Total organic carbon (TOC) was measured at CETASD using a Total Organic Carbon Analyzer (Shimadzu TOC-V_{CSH}), which was equipped with a Solid Sample Module (Shimadzu SSM-5000A) following the manufacturer's method (Shimadzu, 2001). The TOC value was calculated by the difference of the results of the combustion-oxidation reaction (total carbon analysis) and carbonate acidification reaction (inorganic carbon analysis).

3. Results and discussion

3.1. Pollution status and pattern

The concentrations of PCBs and OCPs in sediments of the Hanoi sewer system are listed in Table 1. Eight the 17 OCPs (β-HCH; HCB; o,p'-DDE; p,p'-DDE; o,p'-DDD; p,p'-DDD; o,p'-DDT; p,p'-DDT) were detected. Although other pesticides such as aldrin, heptachlor, oxychlordane, trans-chlordane, cis-chlordane were detected in different environmental samples in Hanoi City (Kishida et al., 2007; Minh et al., 2004; Nhan et al., 2001) and in Ha Long Bay, Hai Phong Harbor and Ba Lat Estuary (Hong et al., 2008), the somewhat elevated method limits of quantification might have hampered their detection in this study. Because α- and γ-HCH isomers were all below the method limit of quantification (0.2 ng/g), the HCHs represent the concentrations of β-HCH in this report. Finally, the PCB congener 28 was not detectable in any of the sediment samples.

In general, the mean concentrations followed the order DDTs (mean 135 ng/g) > PCBs (mean 104 ng/g) > HCHs (mean 3.8 ng/g) > HCB (mean 3.5 ng/g). This pollution pattern agrees well with observations in sediments of the Hanoi rivers in 1997 (Nhan et al., 2001). Besides, Minh et al. (2004) revealed a similar pollution pattern (DDTs > PCBs >> HCHs, HCB) in human breast milk of Hanoi citizen in 2000. These facts demonstrate that DDTs and PCBs are the two dominant pollutants in the environment of Vietnam. The pollution pattern of DDTs and

PCBs in this study varied depending on the sampling location (comprising the rivers Nhue, To Lich, Lu, Set, Kim Nguu and the lake Yen So). The mean concentration of PCBs was higher than DDTs in sediments of the Nhue River, the Lu River, and the Yen So Lake (PCBs level was around 1.5 time to one order of magnitude higher than DDTs). In contrast, the mean concentration of DDTs were higher than PCBs at the rivers To Lich, Set, and Kim Nguu (Fig. 2). The possible sources for such a pollution pattern are difficult to evaluate since the environment of Hanoi City is impacted by a variety of local activities such as suburban agriculture and industry services in the urban districts.

The spatial distribution shows significantly higher concentrations of DDTs, PCBs, and HCB in the rivers Kim Nguu and Set (Fig. 2). As a result, the Yen So Lake, a reservoir receiving water from these two rivers, is polluted with relatively high levels of PCBs (20 to 384 ng/g) and OCPs (17 to 109 ng/g DDTs, and up to 12 ng/g HCB). The sediment concentrations of OCPs in the Yen So Lake were even higher than in the rivers Nhue, To Lich, and Lu. In conjunction with the rivers Lu and Set, the higher levels of PCBs and OPCs at the upper streams (sites TL1, TL2, TL3, and KN1) compared to those at the lower streams (TL4, TL5, TL6, TL7, and KN2) of the rivers To Lich and Kim Nguu (Table 1) point to input sources within the highly populated center of Hanoi. In addition, pollution sources from industrial and agricultural activities along the course of the rivers must also be taken into account since some pollutants showed increased concentrations down stream, e.g. PCBs (in the rivers Set and Lu) and DDTs (in the rivers Lu and Nhue).

3.2. Temporal trend and composition of pollutants in sediments

3.2.1. HCB

199 HCB was detected in 55% of the sediment samples with concentrations ranging from <0.2 to
200 22 ng/g. In general, HCB was scarcely investigated in Vietnam compared to PCBs, DDTs,
201 and HCHs. The HCB levels were reported to be higher in human breast milk of women living
202 in Hanoi City than in Hochiminh City (Minh et al., 2004) and in sediment along the coast of
203 Northern Vietnam (Nhan et al., 1999). In the survey conducted in 1997, Nhan et al. (2001)
204 reported HCB concentrations in the sediments of the Hanoi rivers ranging from n.d. to 0.13
205 ng/g. The HCB levels of the present study are about one to two orders of magnitude higher
206 than those in 1997, but comparable to the levels in Hochiminh City canals in 2004 (Minh et
207 al., 2007a). HCB was used as a fungicide in Vietnam. Recent sources of HCB are attributed to
208 waste incineration (World Health Organization, 1997) but they may also originate from the
209 production and use of agrochemicals and industrial chemicals containing HCB as a by-
210 product (Monirith et al., 2003).
211
212 *3.2.2. HCHs*
213 β-HCH was the only isomer (among three investigated HCH isomers) detected in quantifiable
214 amounts in five of the 22 samples. The sediment concentrations of β-HCH determined in this
215 study (<0.2 to 36 ng/g, mean 3.8 ng/g) were higher than those measured in Hanoi rivers in
216 1997 (Nhan et al., 2001), in Hochiminh canals and in the Mekong River in 2004 (Minh et al.,
217 2007a,b). The typical technical HCH mixture generally applied in Northern Vietnam
218 contained 55 to 80% α-HCH, 5 to 14% β-HCH, 8 to 15% γ-HCH, and 2 to 16% δ-HCH, and a
219 similar pattern of HCH isomers was actually detected by Nhan et al. (2001). However, the
220 predominant presence of the β-HCH isomer found in this study can be explained by the
221 relatively distant time between the use of HCH and the sampling time, and by the lowest
222 water solubility, lowest vapour pressure, highest chemical stability, and the resistance to
223 microbial transformation of β-HCH compared to other isomers (Ramesh et al., 1991). In

addition, α-HCH can be transformed to β-HCH in the environment (Wu et al. 1997). Relatively high levels of β-HCH as the only isomer were also reported in human breast milk in Hanoi City (Minh et al., 2004).

3.2.3. DDTs

In conformity with previous studies conducted by Nhan et al. (2001) and Kishida et al. (2007), we report the total DDTs concentration as the sum of o,p'-, p,p'-DDD, o,p'-, p,p'-DDE, o,p'-, p,p'-DDT. The sediment concentrations of DDTs varied from 12 to 14 ng/g in the Nhue River, 6.4 to 61 ng/g in the To Lich River, 11 to 103 ng/g in the Lu River, 215 to 680 ng/g in the Set River, 82 to 1100 ng/g in the Kim Nguu River, and 17 to 109 ng/g in the Yen So Lake. All the collected sediment samples contained DDTs and the highest concentration was observed at site KN1 (1100 ng/g), which is located in the highly populated center of Hanoi City. Nhan et al. (2001) and Kishida et al. (2007) reported sediment concentrations of DDTs in the Hanoi rivers ranging from 7.4 to 81 ng/g (mean 31 ng/g) in 1997 and 42 to 44 ng/g (mean 43 ng/g) in 2002, respectively. The results of this study (4.4 to 1100 ng/g, mean 135 ng/g) clearly reveal higher concentrations, indicating recent input of DDTs to the aquatic environment, although the use of DDT is banned in Vietnam since 1995.

The hypothesis of recent input of DDT to the environment in South Vietnam was elucidated by evaluating the pattern of individual DDTs (Phuong et al., 1998; Minh et al., 2007a,b). Similarly, three major compounds, including DDT, DDD, and DDE, can be used for assessing the chronology of DDT input in the Hanoi sewers in North Vietnam. Hong et al. (1999) suggested that a ratio of (DDE + DDD)/DDTs of more than 0.5 is indicative for a long-term biotransformation of DDT to DDD and DDE, while a ratio of less than 0.5 may imply recent input. In addition, a ratio of DDT/DDE >0.5 may indicate recent input of DDT, and, in

249 contrast, of <0.3 may imply past input of DDT (Strandberg et al. 1998). Fig. 3 illustrates the
250 patterns of DDT compounds detected in sediments from this study and those calculated for
251 sediments in 1997 (Nhan et al., 2001). The DDT/DDE ratios being <0.3 and the (DDE +
252 DDD)/DDTs ratios >0.5 in sediments of 1997 (Nhan et al., 2001) revealed that
253 biotransformation was significant in the period of 1997 to 2006. However, a decreasing trend
254 of the (DDE + DDD)/DDTs ratios and an increasing trend of DDT/DDE ratios as well as
255 DDT/DDTs ratios among sediments collected in 1997 and 2006 were recognized, revealing
256 additional recent input of DDT to the environment. Particularly, a ratio of DDT/DDE up to 14
257 (DDT occupied 88% among DDTs) in a sediment sample collected at site KN1 signified a
258 very recent input of DDT to the Hanoi sewer system. DDT might originate from illegal usage
259 of DDT in suburban agriculture, leakage from remaining stockpiles and unsecured landfills,
260 or vector control and hygienic purposes (Minh et al., 2004, 2006; Nhan et al., 2001). Minh et
261 al. (2006) suggested a further source of DDTs might be the insecticide Dicofol, which
262 contains DDT as a byproduct. For example, Dicofol was used in China from 1988 to 2002,
263 thereby, spreading an estimated 8800 tons of DDT to the environment (Qiu et al., 2005).
264
265 *3.2.4. PCBs*
266 Similar to DDTs, the PCBs spanned a relatively wide range. The sediment concentrations of
267 PCBs varied from 22 to 153 ng/g in the Nhue River, 1.3 to 70 ng/g in the To Lich River, 42 to
268 122 ng/g in the Lu River, 36 to 139 ng/g in the Set River, 237 to 328 ng/g in the Kim Nguu
269 River, and 20 to 384 ng/g in the Yen So Lake (Fig. 2). In the Sai Gon-Dong Nai River Basin,
270 Hochiminh City, Minh et al. (2007a) reported PCB levels to decline 3 to 6 times lower than
271 those in the early 1990s. However, PCB levels in sediment of Hanoi showed an opposite trend.
272 Ranging from 1.3 to 384 ng/g (mean 104 ng/g), the sediment levels of PCBs measured in this
273 study revealed a clear increase compared to 0.79 to 40 ng/g (mean 13 ng/g) in 1997 (Nhan et

al., 2001, quantification using Aroclor 1254) and 15 to 120 ng/g (mean 45 ng/g) in 1999 (Viet et al., 2000). Since sediment samples were collected in the dry season in this study and at the onset of the rainy season in 1997, the different amount of water drained through the sewer system might affect the washout of suspended particle containing PCBs. However, by evaluating the temporal trend of PCBs in soils collected in Hanoi City in 2006, Toan et al. (2007b) reported clearly increasing concentrations of PCBs compared to levels in 1990s. In addition, in a sediment survey conducted in 1995, Nhan et al. (1998) reported a relatively low PCB concentration of 5.64 ng/g in the dry season in an irrigation canal close to the Nhue River. Hence, the observation made in this study raises serious concern on the increase of PCB levels in Hanoi City.

Concerning the PCB congeners, Toan et al. (2007b) reported that until 1998, about 48% of the total imported quantity of likely PCBs-containing oil originated from the Soviet Union. This oil contained 11.7, 7.0, 6.5, 3.6, 0.4, and 0.8% of PCB congeners 138, 153, 101, 52, 180, and 28, respectively, which were coincident with those in soil samples collected in Hanoi City. However, a similar composition of PCB congeners in different categories of sediments was observed in this study, except the less persistent congener 28 was not detected (Fig. 4). This result indicates significant recent sources of PCBs to the sediments of the Hanoi sewer system, such as from the industry, atmospheric deposition, and domestic wastewater. The leakages of PCBs from the PCBs-containing oils in old transformers and capacitors, which are widely installed in Hanoi City, and especially from recent PCBs-containing wastes, are thinkable sources of the PCBs pollution. Initial inventory results show that there are more than 11 800 likely PCB-containing electrical equipments in Vietnam, containing some 7000 tones of likely PCB-containing oils (MONRE, 2006). In addition, the rapid development of Hanoi City without adequate measures for safe handling of PCBs-containing oils and other materials are

likely to have led to the increase of PCBs pollution in the city. As cited in Toan et al. (2007b), PCBs could currently be used as a component of lubricating oils for motor vehicles. This fact suggests that the assessment of the sources of PCBs should be given outmost attention.

3.3. Comparison of PCB and OCP levels in Vietnam with other regions of the world

A comprehensive comparison of PCB and DDT levels in recently collected sediments from various locations in Vietnam and in the world is presented in Fig. 5. It can be recognized that among the reported locations in Vietnam, the levels of PCBs (mean 104 ng/g) and DDTs (mean 135 ng/g) in sediments of the Hanoi sewer system are highest followed by the Hochiminh City canals. The result indicates Hanoi City itself is likely comprising sources of PCB and DDT pollution. Furthermore, the sediment levels of PCBs and DDTs in the Hanoi sewer system are comparable to those found in the highly polluted Alexandria Harbor (Egypt) and Macau Harbor (China).

The total organic carbon content (TOC) has a significant influence on the sediment concentration of PCBs and OCPs as these compounds are highly sorptive with high K_{OC} values (Ding and Wu, 1995; Lu et al., 2006). By evaluating the correlation between TOC and the concentration of PCBs, DDTs, and CHLs in sediments of the Hochiminh City canals, the Saigon-Dong Nai River and the estuary in South Vietnam, Minh et al. (2007a) demonstrated that higher amounts of these pollutants mainly occurred in sediments with high TOC. In general, the TOC values in sewers and lake in Hanoi City (1.0 to 11%, mean 6.2%; see Table 1) are higher than those in Hochiminh City canals (3.2 to 4.9%), the Saigon-Dong Nai River and the estuary (0.44 to 4.8%). This could explain the higher concentrations of DDTs and PCBs in Hanoi sewer system in comparison with those in the South of Vietnam in the recent study of Minh et al., 2007a,b. The TOC values in sediments of other reported locations,

including the Tam Giang-Cau Hai Lagoons (Frignani et al., 2007), the Balat Estuary, the Halong Bay, and the Hai Phong Harbor (Hong et al., 2008) were also reported lower than those in Hanoi sewer system.

3.4 Hazard assessment

In order to obtain an overall view on the possible toxic effects to the benthic and epibenthic organisms in the Hanoi sewer system (Fig. 6), the sediment concentration of DDTs and PCBs were compared with the interim sediment quality guideline (ISQG) and the probable effective level (PEL), issued by the Canadian Council of Ministers of Environment (CCME, 2002). The concentrations of DDE, DDD, and DDT (sum of o,p' and p,p' isomers) in all the Hanoi sediment samples were higher than the ISQG values (1.42, 3.54, and 1.19 ng/g, respectively). The DDE, DDD, and DDT generally exceed the PEL values (6.75 ng/g for DDE, 8.51 ng/g for DDD, and 4.77 ng/g for DDT) but vary among the sediment samples. Most of the sediments from the rivers To Lich, Lu, Set and the Yen So Lake contained DDTs above the PEL values, while they were below at the Nhue River. In contrast, the PCBs concentrations (except for sample YS3, 384 ng/g) were lower than the PEL value (340 ng/g as for Aroclor 1254), and only half of them exceed the ISQG value (60 ng/g as for Aroclor 1254).

4. Conclusions

Occurrence, patterns, temporal trend, sources, and the potential toxicity of PCBs and OCPs were assessed in sediments of the Hanoi sewer system. Although pollution patterns are different among the various rivers and lake investigated, the relatively high concentrations of PCBs, DDTs, HCB and β-HCH indicate that pollution by persistent organochlorines is still an important environmental issue in Hanoi City. The increasing trend of these pollutants documented over the last decade in conjunction with increasing DDT/DDE ratios indicates

very recent inputs to the environment. The levels of PCBs and DDTs in sediments of the Hanoi sewer system are the highest recorded in Vietnam so far, indicating that the pollution originates from sources within the city itself. DDE, DDD, DDT and about half of the PCBs exceed the interim sediment quality guidelines established in Canada. Furthermore, DDE, DDD and DDT exceeded the probable effect levels in most of the sediment samples originating from the Hanoi rivers To Lich, Lu, Set and the lake Yen So.

The findings of this study provide valuable information for the public and environmental authorities of Vietnam to mitigate the discharge of toxic chemicals into the aquatic environment via the sewer system (Hoai et al., 2009). In addition, the assessment of the sources of DDTs and PCBs should be given adequate attention, particularly in light of the fact that water of the sewer rivers is used for agricultural irrigation and therefore pose a threat to accumulate in food.

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545 ## Figures legends

546

547 **Fig. 1.** Map of Hanoi City indicating the sewer system and the locations where sediments
548 samples were collected (N: Nhue River, TL: To Lich River, L: Lu River, S: Set River, KN:
549 Kim Nguu River, and YS: Yen So Lake).

550

551 **Fig. 2.** Concentration ranges of PCBs (○), DDTs (●), β-HCH (▫), and HCB (■) in sediments of
552 the sewer system in Hanoi.

553

554 **Fig. 3**. Ratios of DDT compounds in sediments of the Hanoi sewer system in 1997 (Nhan et
555 al., 2001) and 2006 (present study).

556

557 **Fig. 4.** Mean composition of PCB congeners in Hanoi river sediments.

558

559 **Fig. 5.** Concentration ranges and mean values of DDTs (▬) and PCBs (▭) in recently
560 collected sediments from various locations in the world.
561 [a] Sum of PCB congeners 28, 52, 101, 118, 138, 153, 180 multiplied by a value of four; [b] Sum
562 of PCB congeners 28, 52, 101, 138, 153, 180 multiplied by a value of 4.81; [c] Sum of
563 Kanechlor KC300-KC600; [d] As Aroclor 1254; [e] Sum of PCB congeners 8, 18, 28, 29, 44, 52,
564 66, 87, 101, 105, 110, 118, 128, 138, 153/132, 170/190, 180, 187, 195, 200, 206, 209; [f] Sum
565 of 53 PCB congeners; [g] sum of Aroclor 1242, 1248, 1254, 1260; [h] Sum of PCB congeners 28,
566 31, 33, 44, 49, 53, 70, 74, 87, 118, 128, 138, 153, 206, 208; [i] Sum of PCB congeners 1, 5, 28,
567 29, 47, 49, 52, 77, 97, 101, 105, 118, 138, 153, 154, 169, 171, 180, 187, 200, 204; [j] Sum of 99
568 PCB congeners; [k] Sum of PCB congeners 28, 52, 101, 138, 153, 180; [l] Sum of PCB congeners
569 77, 101, 105, 118, 126, 138, 153, 156, 167, 169, 170, 180, 194; [m] Sum of 104 PCB congeners.

570 † Sum of o,p'- and p,p'-DDT, DDD, DDE; †† Sum of p,p'-DDT, DDD, DDE; #: Sum of

571 dichlorobenzophenone and p,p'-DDT, DDD, DDE.

572

573 **Fig. 6.** Comparison of measured DDT and PCB concentrations with the Canadian

574 Environmental Quality Guideline for Sediment. (○) Nhue River, (●) To Lich River, (□) Lu

575 River, (△) Set River, (■) Kim Nguu River, (▲) Yen So Lake. (◄- - -) interim sediment quality

576 guideline, (◄——) probable effect level.

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580 # Tables

581

582 **Table 1.** Concentrations of detectable OCPs and PCBs in sediment

Sample		Location		TOC	PCBs[a]	DDTs[b]	β-HCH	HCB
		Latitude	Longitude	(%)	(ng/g)			
Nhue River	N1	N 20°57'263"	E 105°48'444"	3.1	153	12	<0.2	1.1
	N2	N 20°57'144"	E 105°48'472"	2.3	22	14	<0.2	1.3
To Lich River	TL1	N 21°02'403"	E 105°48'364"	7.5	50	50	12	<0.2
	TL2	N 21°00'923"	E 105°48'270"	2.7	44	47	<0.2	<0.2
	TL3	N 21°00'106"	E 105°49'083"	6.0	26	26	<0.2	0.87
	TL4	N 20°58'237"	E 105°49'501"	2.0	9.2	61	0.8	3.3
	TL5	N 20°57'577"	E 105°48'795"	5.5	70	24	<0.2	2.4
	TL6	N 20°57'615"	E 105°48'752"	4.7	14	6.4	<0.2	<0.2
	TL7	N 20°57'166"	E 105°48'466"	1.0	1.3	11	<0.2	<0.2
Lu River	L1	N 21°00'072"	E 105°50'090"	5.3	42	11	<0.2	3.2
	L2	N 20°59'486"	E 105°50'462"	9.9	122	103	17	<0.2
	L3	N 20°58'409"	E 105°49'707"	7.6	78	73	<0.2	<0.2
Set River	S1	N 20°59'587"	E 105°50'594"	4.1	36	680	<0.2	5.3
	S2	N 20°58'313"	E 105°51'228"	5.6	139	215	<0.2	11
Kim Nguu River	KN1	N 21°00'028"	E 105°51'717"	11	328	1100	<0.2	1.4
	KN2	N 20°58'560"	E 105°51'923"	10	237	82	<0.2	22
Yen So Lake	YS1	N 20°58'471"	E 105°51'881"	6.4	210	79	<0.2	<0.2
	YS2	N 20°58'201"	E 105°51'266"	9.1	24	17	13	8.0
	YS3	N 20°58'395"	E 105°51'793"	9.0	384	67	<0.2	<0.2
	YS4	N 20°58'130"	E 105°51'406"	8.3	98	109	<0.2	12
	YS5	N 20°58'299"	E 105°51'418"	7.5	20	27	<0.2	<0.2
	YS6	N 20°58'308"	E 105°51'659"	6.7	80	33	36	<0.2

583 [a] PCBs: Sum of seven PCB congeners (PCB 28, 52, 101, 118, 138, 153, and 180) by
584 multiplication with the value of four according to Froescheis et al. (2000).

585 [b] DDTs: Sum of o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'DDT, and p,p'-DDT.

586

587

Figures

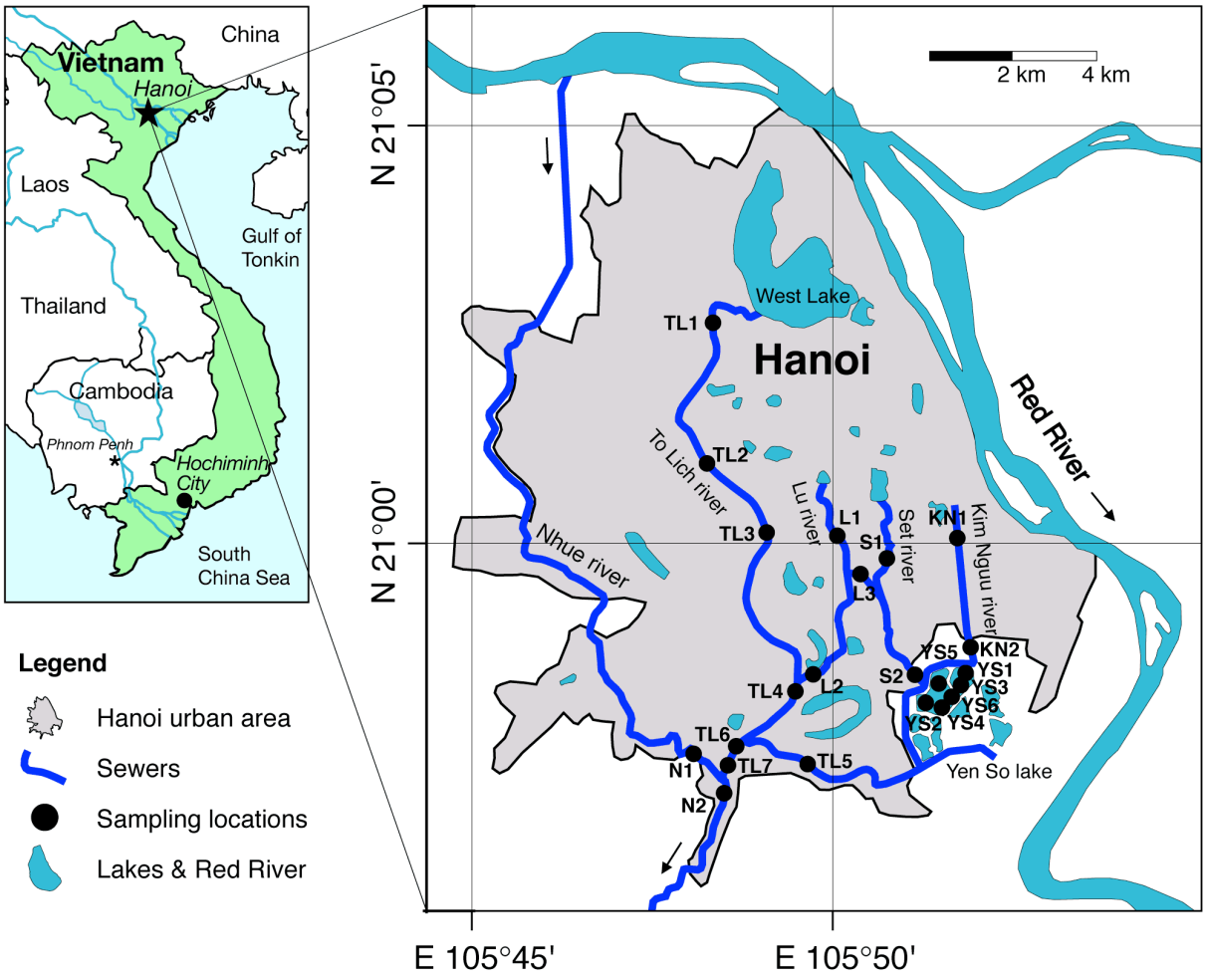

Figure 1

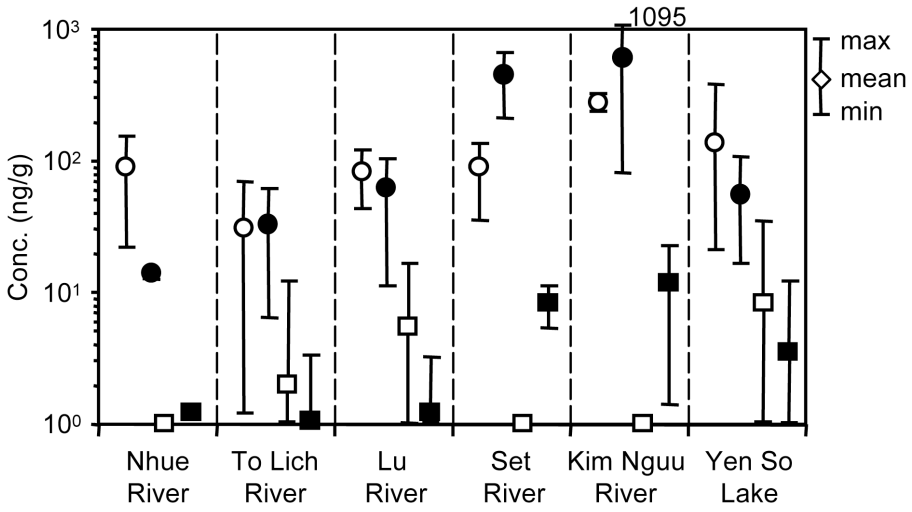
1095
max
mean
min
Conc. (ng/g)
10^3
10^2
10^1
10^0
Nhue River
To Lich River
Lu River
Set River
Kim Nguu River
Yen So Lake

Figure 2

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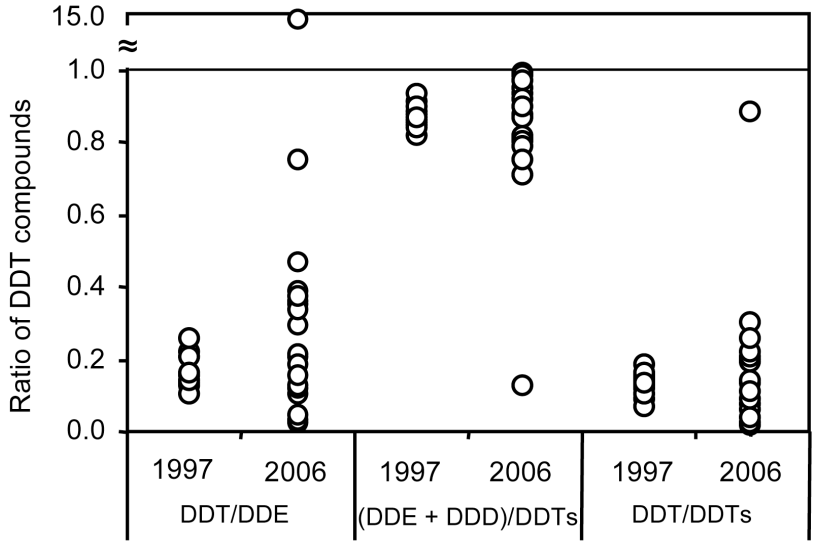

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606 **Figure 3**

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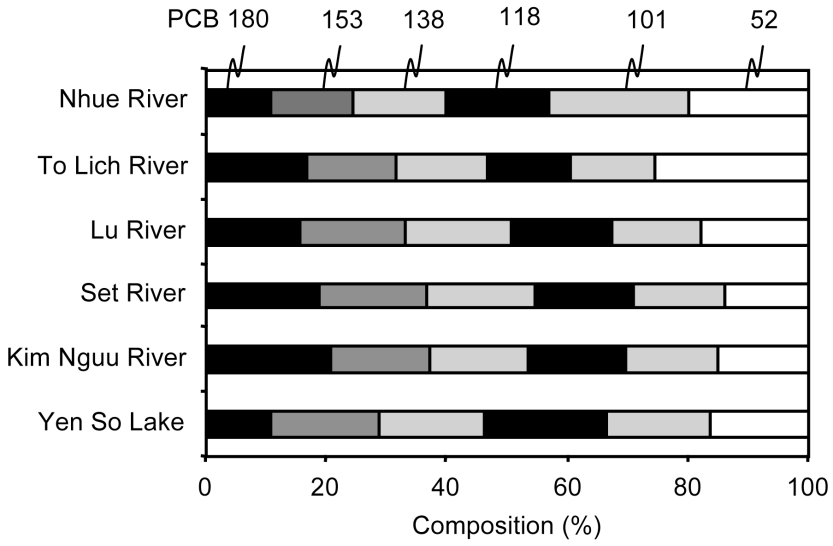

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612 **Figure 4**

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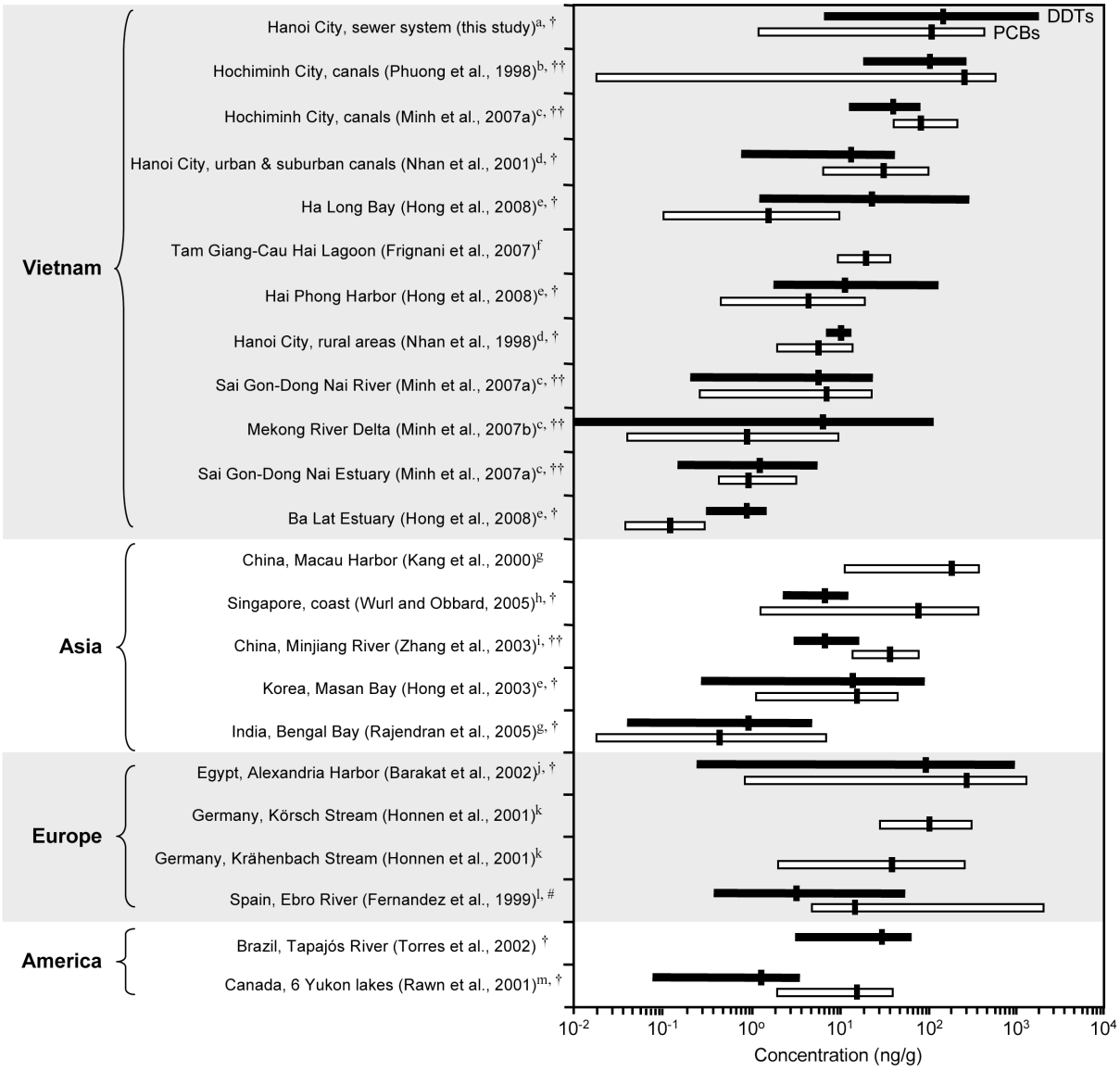
Vietnam
Hanoi City, sewer system (this study)[a, †]
Hochiminh City, canals (Phuong et al., 1998)[b, ††]
Hochiminh City, canals (Minh et al., 2007a)[c, ††]
Hanoi City, urban & suburban canals (Nhan et al., 2001)[d, †]
Ha Long Bay (Hong et al., 2008)[e, †]
Tam Giang-Cau Hai Lagoon (Frignani et al., 2007)[f]
Hai Phong Harbor (Hong et al., 2008)[e, †]
Hanoi City, rural areas (Nhan et al., 1998)[d, †]
Sai Gon-Dong Nai River (Minh et al., 2007a)[c, ††]
Mekong River Delta (Minh et al., 2007b)[c, ††]
Sai Gon-Dong Nai Estuary (Minh et al., 2007a)[c, ††]
Ba Lat Estuary (Hong et al., 2008)[e, †]
Asia
China, Macau Harbor (Kang et al., 2000)[g]
Singapore, coast (Wurl and Obbard, 2005)[h, †]
China, Minjiang River (Zhang et al., 2003)[i, ††]
Korea, Masan Bay (Hong et al., 2003)[e, †]
India, Bengal Bay (Rajendran et al., 2005)[g, †]
Europe
Egypt, Alexandria Harbor (Barakat et al., 2002)[j, †]
Germany, Körsch Stream (Honnen et al., 2001)[k]
Germany, Krähenbach Stream (Honnen et al., 2001)[k]
Spain, Ebro River (Fernandez et al., 1999)[l, #]
America
Brazil, Tapajós River (Torres et al., 2002) [†]
Canada, 6 Yukon lakes (Rawn et al., 2001)[m, †]
DDTs
PCBs
10-2
10-1
10o
101
102
103
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Concentration (ng/g)

Figure 5

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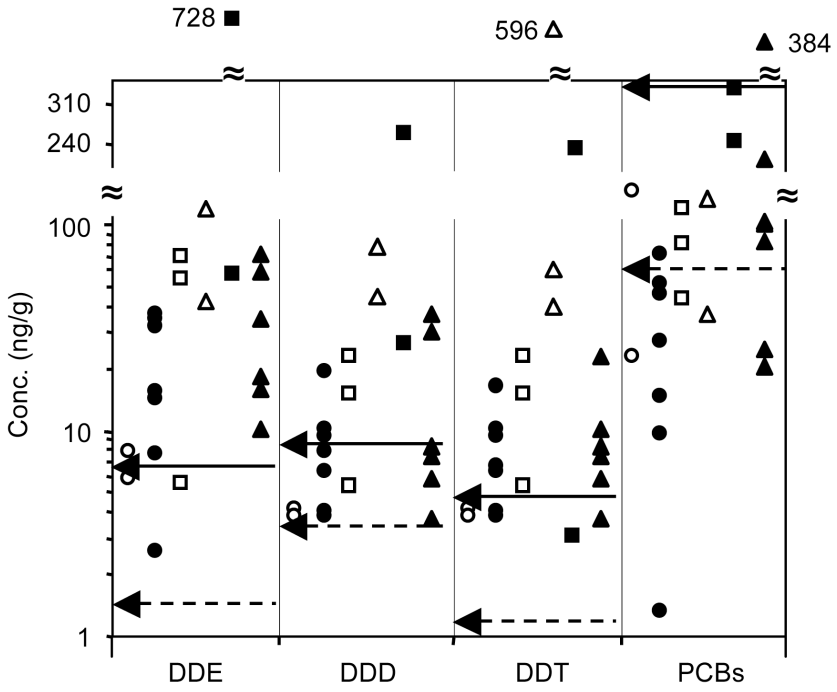

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623 **Figure 6**

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