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Where has the pollution gone? A survey of organic contaminants in Ho Chi Minh City / Saigon River (Vietnam) bed sediments

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Abstract

A wide range of persistent organic chemicals, including polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), some insecticides, as well as polybrominated diphenyl ethers (PBDEs) and some perfluoroalkyl substances (PFASs) were analyzed in 17 bed sediments collected along the Saigon River and at adjacent canal mouths from upstream to downstream in Ho Chi Minh City (Vietnam). Concentrations were rather low for PAHs, as well as for legacy PCBs and dichloro-diphenyl-trichlorethane and metabolites (DDTs), or below detection limits for several PFASs and all PBDEs measured. Several insecticides (chlorpyrifos-ethyl, and the pyrethroids cypermethrin and λ-cyhalothrin) displayed rather high concentrations at a few sites within the city. There was no distinct upstream – downstream trend for PAHs, (DDTs) or PCBs. Although adjacent canal sediments tended to be more contaminated than Saigon River sediments, the differences were not significant. Emissions are almost certainly substantial for PAHs, and probably also for other contaminants such as PBDEs and some PFASs. During the dry season, contaminants are presumably stored in the city, either in canals or on urban surfaces. Heavy rainfall during the monsoon period carries away contaminated particle flows into the canals and then the Saigon River. The strong tidal influence in the river channel hinders the accumulation of contaminated particles. Contaminated deposits should accordingly be investigated further downstream in depositional environments, such as the mangrove.

Keywords

sediment, insecticide, PAH, perfluoroalkyl substance, Vietnam, Saigon River

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1 Introduction

Vietnam has experienced remarkable economic growth over the last few decades (Du and Fukushima, 2009); this growth has occurred mainly in urban environments, such as Ho Chi Minh City (HCMC). With a population increase from 5 million inhabitants in 1999 (Gubry et al., 2002) to about 8.44 million people or more currently (GSO, 2016), and industrial activities supported by 30,000 small industrial production units and more than 800 large-scale factories located in 15 industrial zones (Strady et al., 2016)], HCMC has emerged as one of the most dynamic big cities in southeast Asia.

As a consequence, anthropogenic pressures on HCMC's environment have also rapidly increased, leading among others to potential substantial releases of a wide array of organic contaminants into the atmosphere or aquatic media throughout the area. Insecticides have been used for a long time for malaria vector control: Diphenyl-diethyl-trichlorethane (DDT) was authorized until 1993 (Kishida et al., 2007); alternative compounds such as organophosphates or pyrethroids have been introduced since then. Industrial production throughout HCMC district include pharmaceuticals, plastics, packaging, textile and leather garments, paints and varnish, and electronics (HEPZA, 2015). In addition, the road traffic in the city and its surroundings is quite intense. Potential emissions accordingly involve a wide range of compounds: DDT and other insecticides, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), flame retardants, per- and poly-fluorinated substances (PFASs) and so on. Some of them were already monitored in the past, such as PCBs (Phuong et al., 1998; Kishida et al., 2007; Minh et al., 2007; Nguyen, 2009), DDT and its metabolites (Phuong et al., 1998; Kishida et al., 2007), and PAHs (Kishida et al., 2007). Monitoring data on other contaminants such as polybrominated diphenyl ethers (PBDEs) are still rare or absent.

The study objectives were to provide an up to date diagnosis of the contamination of Saigon River sediments by a range of historical and emerging chemicals. Based on the available knowledge (Phuong et al., 1998; Kishida et al., 2007; Minh et al., 2007; Nguyen, 2009), a moderate/decreasing contamination by legacy chemicals such as DDT and PCBs was expected, as well as an increasing contamination throughout the urbanized river stretch for chemicals related to industries or consumer products (flame retardants and PFASs) and road traffic (PAHs).

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2 Materials and methods

2.1 Study area

Ho Chi Minh City (HCMC) is located along the Saigon River, which is a tributary of the Dongnai River (Figure 1). The respective catchments cover 4 700 km² and 40 000 km² respectively, and are subject to a monsoon regime, with about 150 rainy days between May and late November and an average annual rainfall of 1800 mm (GSO, 2016). Both river flows are regulated by upstream reservoirs, namely Dau Tieng on the Saigon River and Tri An on the Dongnai River (Figure 1-B). Respective mean discharges are estimated around 50-60 and 600-650 $m^3 s^{-1}$ (Ha, 2009; Camenen et al., 2017). Given the low altitude and the flat topography, this system is subject to a strong tidal influence, far upstream from the urbanized areas.

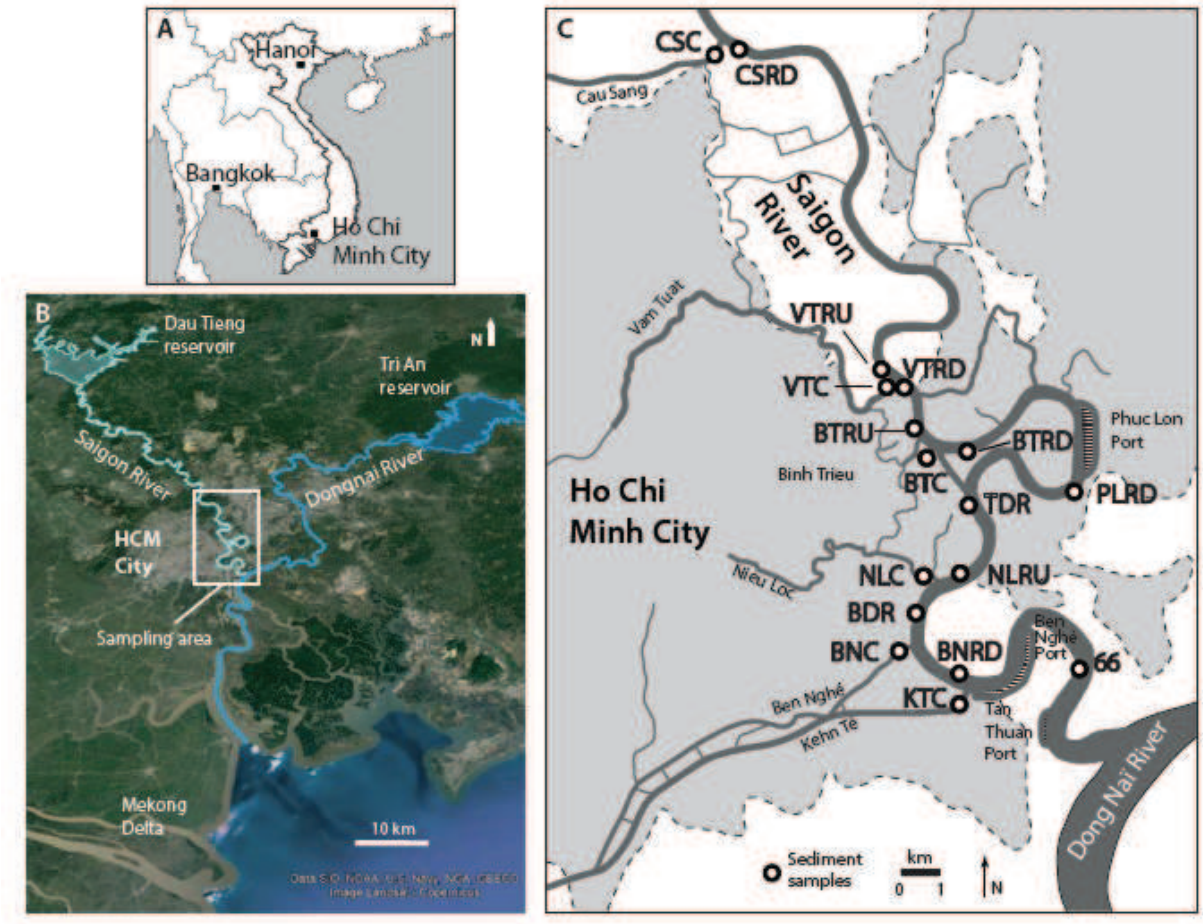

Figure 1 - Study area (A- regional map; B- HCMC area; C- sampling sites in HCMC)

2.2 Sediment sampling

The top 5 cm of bed sediment deposits were sampled with an Eckman grab operated from a boat for the river sites, and from the shore for canal sites (see locations in Figure 1-C). Eleven samples were collected in the Saigon River along an upstream-downstream gradient, and six samples were collected in canals close to their confluence

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with the Saigon River between the 5th and the 8th of January, 2016. Details on the sampling locations are provided in the Supplementary Information (SI), Table S1.

2.3 Sediment characteristics

The organic matter (OM) content was estimated by loss on ignition (LOI) at 550°C on bulk sediment following Heiri et al. (2001); the results are expressed as a percentage of the sediment sample mass (% dw). Grain size distributions were determined by sonicating and then analyzing each sample with a Mastersizer 3000® laser mounted with a hydro-SM small-volume dispersion unit (Malvern Instruments, Worcestershire, UK). Grain-size fractions, mode, sorting, and skewness were computed using the Gradistat program (Blott and Pye, 2001).

2.4 Chemical analysis

2.4.1 Chlorinated pesticides, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs)

These compounds were analyzed following the approach described in previous papers (Thevenon et al., 2013; Mwanamoki et al., 2014; Kilunga et al., 2017). Briefly, sediment samples were first freeze-dried and sieved (2mm). Then, after addition of internal standards (^{13}C-labeled for halogenated compounds and ^{2}H-labeled compounds for all PAHs), about 5 g of dry sediment were extracted with a mixture of 20 % acetone in hexane (v/v) into a Soxhlet system. Interfering sulfur compounds were removed by addition of activated copper to the extract. Then, the organic extract was concentrated to 1 mL in a vacuum rotary evaporator (Buchi Rotavapor, Flawil, Switzerland). The extract was further submitted to fractionation and clean-up over a chromatographic column containing 3 g of Silicagel, according to de Boer et al. (2001). Three separated fractions were collected: first with 16 mL of hexane, then 35 mL of hexane, and finally 50 mL of hexane: dichloromethane (v/v, 1:1). The first two fractions contained PCBs and PBDEs respectively. PAHs and chlorinated pesticides were distributed into the three fractions, which were analyzed separately. After reducing the volume again, the chemicals were measured by gas chromatography with triple mass spectrometry detection (GC-MS/MS, Thermo Scientific, TSQ Quantum XLS Ultra, Waltham, MA, USA). Two columns with different polarities, a ZB-5ms column (60 m x 0.25 mm x 0.25 µm) and one ZB-XLB column (20 m x 0.18 mm x 0.18 µm), were used for separation and identification of the different compounds. The list of compounds analyzed with this approach is provided in the SI, section 2.

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2.4.2 Per- and poly-fluorinated alkyl substances (PFASs)

PFASs were analyzed following the method described in (Munoz et al., 2015) and (Bertin et al., 2014). Briefly freeze-dried / sieved sediment samples (1 g dry weight) were spiked with a suite of ^{13}C-labeled internal standards; PFASs were extracted by sonication using methanol (MeOH), concentrated under a nitrogen stream, purified on ENVI-Carb cartridge.es and eluted with MeOH. Eluates were concentrated to 400 µL under a nitrogen stream and transferred into injection vials. PFASs (list of compounds and acronyms in the SI, Table S3) were analyzed by LC-MS/MS using an Agilent 1200 LC system (Agilent Technology, Massy, France) interfaced with an Agilent 6490 triple quadrupole mass spectrometer. Standards and reagents were purchased from Wellington Laboratories (via BCP Instruments, Irigny, France) and Sigma-Aldrich (St Quentin Fallavier, France).

2.4.3 QA/QC

Almost all chlorinated pesticides, PCBs and PBDEs were included in the second round of UNEP-coordinated Global Interlaboratory Assessment 2012/2013 (UNEP, 2014). In this exercise dozens of laboratories analyzed the same sediment sample. Results from our team are presented in the SI, Table S2. Additionally, other pesticides such as chlorpyrifos-methyl and -ethyl, endosulfan I, II and -sulfate, endrin aldehyde and -ketone, methoxychlor, acetochlor, λ-cyhalothrin, α and β-cypermethrin and deltamethrin were also analyzed. Recovery rates for PCBs varied between 80.5% and 85.3%, and between 61.3% and 103.7% for PAHs. PBDE recovery rates varied between 80.3% and 94.7%, while for pesticides they ranged from 51.5% (endosulfan) to 90.4% (δ-HCH). The limits of detection (LODs) are indicated in Table S2 in the SI.

The PFAS recovery rates varied between 65 and 90 % in spiked reference sand samples (0.5 ng g^{-1} for each analyte, *n*=3). In addition, accuracy was also estimated using spiked sand; both internal standards and analytes were added at the start of the experiment. Accuracy rates ranged from 75 to 115 % (*n* = 3).

Procedural blanks consisting of 10 mL of MeOH were analyzed for each sample series and, when applicable, PFAS concentrations were blank-corrected. The LODs are indicated in Table S5 (section D) in the SI. All results are expressed in ng g^{-1} dry weight (dw).

2.5 Statistics

We used Pro-UCL 5.0 software (https://www.epa.gov/land-research/proucl-software) to determine compound distributions accounting for left-censored results. Wilcoxon-Mann-Whitney (pair comparison) tests were applied

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to compare contamination levels between groups, using Pro-UCL 5.0. Sum parameters, i.e. the sum of concentrations for chemicals of a given family (PAHs, PCBs and so on) were determined as the sums of concentrations above the LD or LoR. Spatial trends were assessed using non-parametric Mann-Kendall and Theil-Sen tests (Pro-UCL 5.0), thus making it possible to handle left-censored data. These tests were not applied when detection frequency fell below 70%.

The significance threshold was set at 0.05 in all analyses.

3 Results

3.1 Sediment characteristics

Grain size distributions in the 17 bed sediments presented three modes at 6.3 µm, 15.4 µm and above 800 µm. Most samples were mainly composed of rather fine particles (poorly sorted silts), with four outliers having a median grain size (D50) of 1039.2 ± 108.3 µm (coarse sand). When these outliers are removed, the median grain size (16.5 ± 12.0 µm) and grain size distributions in canal samples did not differ from those in river samples (*p*-value = 0.421). The OM content in bed sediments presented an average value of 7.8 ± 2.5 %, with one outlier having an OM content below 2%. Furthermore, respective OM contents in canal and river samples did not differ (*p*-value = 0.119). Detailed results are provided in the SI (Table S4).

3.2 Sediment contamination

The detection rates of the 95 analytes ranged from 0 to 100% of the 17 bed sediment samples. PBDE concentrations, as well as those of hexachlorobenzene (HCB), hexachlorocyclohexane (HCH), drins, heptachlor, endosulfan and deltamethrin were systematically below their respective LODs. Detection rates of other insecticides ranged from 12% (cypermethrin) to 94% (chlorpyrifos-ethyl, ΣDDTs). Furthermore, PCB detection rates ranged from 12% for the congener PCB156 (mono-ortho substituted) to 88% for more stable congeners such as PCB153 (Table S5-A in the SI). DDT isomers and their metabolites occurred in 76 to 94% of the samples, except *o,p'*-DDT (41%); overall, some parent compounds and metabolites were present in 100% of the samples (Table S5-B in the SI). Similarly, most PAHs were present in all samples, with individual detection rates ranging from 82% (acenaphtene, dibenzo(a,h)anthracene) to 100% (Table S5-C in the SI). The most frequently detected PFASs included two carboxylic acids (PFUnDA, PFTrDA), PFOS isomers, FOSA, EtFOSAA, some polyfluoroalkyl phosphoric acid diesters (6:2 and 8:2 diPAP) and fluorotelomer sulfonates (8:2 FTSA; Table S5-D and E in the SI).

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3.2.1 Insecticides

Chlordane α and γ isomers were detected in four samples, at concentrations close to the LOD (0.2 ng g^{-1} dw). ΣDDT, i.e. the sum of concentrations of DDT isomers and their metabolites, were determined in all samples, and varied between 2.40 ng g^{-1} dw and 16.05 ng g^{-1} dw; *p,p'* DDD and *p,p'* DDE were predominant throughout the samples. ΣDDT concentrations in canal samples were slightly lower than in river samples, but the difference was not significant (*p*-value 0.17, Table 1).

The pyrethroid insecticide λ-cyhalothrin, was measured at two canal sites (0.91 and 23.2 ng g^{-1}), plus an adjacent river site (0.97 ng g^{-1}) for λ-cyhalothrin, at concentrations several orders of magnitude above the detection limit (0.3 ng g^{-1} dw; Table S5-B in the SI).

3.2.2 PCBs

With a maximum ΣPCB7 of 18.7 ng g^{-1} dw (sum of indicator congener concentrations, i.e. CB 28, 52, 101, 118, 138, 153 and 180), or 25.3 ng g^{-1} dw for Σ12PCB (12 measured congeners), the contamination by this class of compounds remained moderate, as compared e.g. to existing sediment quality guidelines (MacDonald et al., 2000). For two samples (CSC, BNRD), all analyzed congeners were < LOD. Otherwise, the molecular profiles were dominated by congeners 138, 153 and 180, with median values equaling 0.27 ng g^{-1} dw, 0.51 ng g^{-1} dw, and 0.22 ng g^{-1} dw respectively.

3.2.3 PAHs

ΣPAHs (i.e. the sum of the concentrations of the 16 compounds) varied between 49 ng g^{-1} dw and 933 ng g^{-1} dw (median 345 ng g^{-1} dw; SI Table S5-C), well below existing sediment quality guidelines (e.g. MacDonald et al., 2000). Phenanthrene, fluoranthene, pyrene and benzo(b)fluoranthene were the main contributors to ΣPAHs, representing respectively 12%, 17%, 14% and 11% of the sum on average (Figure S1 in SI). Whereas phenanthrene is deemed to result mainly from OM maturation, i.e., petrogenic origin, fluoranthene and pyrene instead point to pyrogenic sources, such as vehicle exhaust or wood coal combustion (Zhang et al., 2013). Applying concentration ratios between specific PAHs (Yunker et al., 2002) tended to confirm this diagnosis of multiple sources, with a predominance of pyrogenic sources (Figure S2 in the SI). Nevertheless ΣPAHs in all samples remained below the probable effect concentration of 2280 ng g^{-1} dw (MacDonald et al., 2000).

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147 **3.2.4 PFASs**

148 Overall, the PFASs detected displayed rather low concentrations, e.g. ΣPFCAs, 8:2 FTSA, 6:2 di-PAP and 8:2 diPAP
149 maximum concentrations at 1.18 ng g^{-1} dw, 0.08 ng g^{-1} dw, 0.18 ng g^{-1} dw and 0.24 ng g^{-1} dw respectively (Table
150 S5-E in the SI) compared to studies in various environments (Ahrens et al., 2010; Chen et al., 2017; Munoz et al.,
151 2017).

152 **3.3 Contamination patterns**

153 There is no upstream-downstream gradient in Saigon river sediments for PAHs, DDTs, PCBs or PFOS (Figure 2).
154 Unlike PAHs, chlorpyrifos-ethyl presented a significant upstream-downstream trend (*p*-value 0.03; Figure 2-D),
155 with two peaks at CSRD, immediately downstream from the Cau Sang canal junction, and BTRD, immediately
156 downstream from the Binh Trieu canal junction. The two corresponding canal sites, i.e. CSC and BTC (not shown
157 in Figure 2, which displays only sampling sites in the river), also displayed appreciable chlorpyrifos-ethyl
158 concentrations compared to other sites in this study, though below those observed at CSRD and BTRD. The BTC
159 sample, but not CSC, also revealed noticeable concentrations of λ-cyhalothrin (23.2 ng g^{-1} dw) and cypermethrin
160 (155 ng g^{-1} dw). While canal sediments showed higher concentrations than river sediments for the most
161 frequently quantified parameters, namely ΣDDTs, ΣPAHs, ΣPCB7 or chlorpyrifos-ethyl, the differences were not
162 significant (Table 1).

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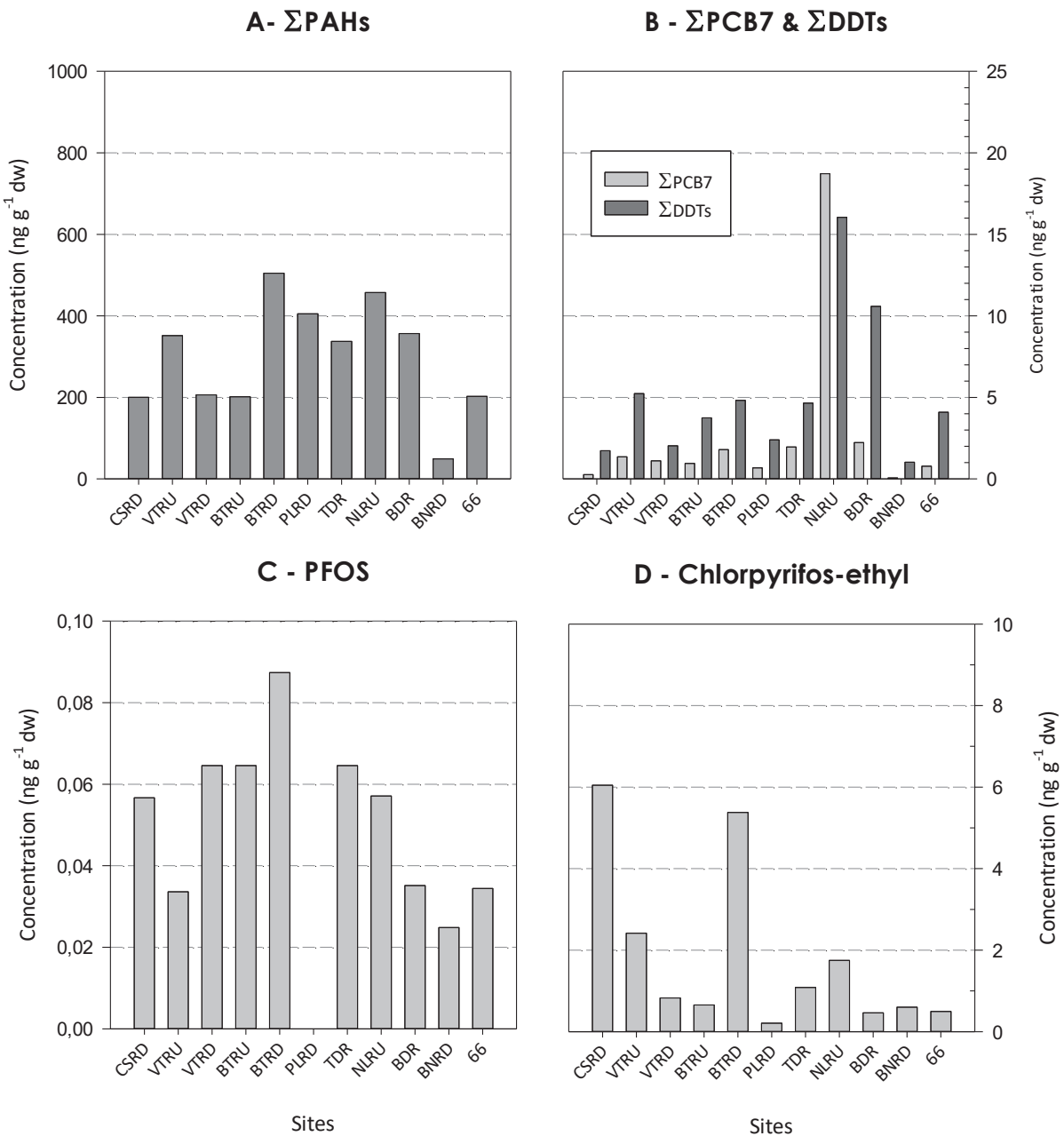

Figure 2 - PAHs, PCBs, DDTs, PFOS, and Chlorpyrifos-ethyl in Saigon River sediments from upstream (CSRD) to downstream (66) of HCMC

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Contaminant	Canals		River		*p*-value
	Mean (SD)	Median (min-max)	Mean (SD)	Median (min-max)	
Chlorpyrifos-ethyl	2.00 (0.72)	1.93 (1.26-2.82)	1.97 (2.07)	0.95 (0.46-6.05)	0.09
ΣDDT	8.02 (3.10)	7.48 (<LOD-13.05)	5.13 (4.45)	4.10 (<LOD-16.05)	0.17
ΣPAHs	498 (308)	503 (108-933)	298 (136)	337 (49-505)	0.09
ΣPCB7	3.95 (2.15)	3.69 (<LD-7.32)	2.99 (5.56)	1.24 (<LOD-18.73)	0.09
ΣPFASs	0.86 (0.539)	0.544 (<LQ-1.60)	0.39 (0.281)	0.37 (<LQ-0.92)	0.08

Table 1 – Comparison of the concentrations (ng g^{-1} dw) in canal and river sediments (Mann-Whitney test)

4 Discussion

4.1 Sediment quality - Comparison to other studies

DDT compounds have been measured in HCMC sediment samples since the 1990s: Phuong et al. mentioned a ΣDDTs up to 253 ng g^{-1} dw, with a median value of 49 ng g^{-1} dw in canal sediments (Phuong et al., 1998), while about 10 years later the concentration ranges were 12 – 72 ng g^{-1} dw in canal sediments, and 0.21 – 23 ng g^{-1} dw in Saigon and Dongnai river sediments (Minh et al., 2007). At the same period, another study in canal and river sediments showed ΣDDTs ranging from 0.1 to 38.3 ng g^{-1} dw (Kishida et al., 2007), similar to the recent findings (1.8 – 74 ng g^{-1} dw) by Bui et al. (2016). Although it was not possible to compute these data in a single trend assessment, in part because most locations are not accurately reported in previous studies, there is no evidence of a sharp ΣDDT decline in the last two decades. Overall, even the highest concentrations recorded in 1998 remained far below the probable effect concentration of 572 ng g^{-1} dw derived from co-occurrence studies in North America (MacDonald et al., 2000).

Measured concentrations of λ-cyhalothrin were close to, or largely exceeded the proposed sediment quality guideline (Ineris) of 1.05 ng g^{-1}, and similar to levels causing adverse effects to *Chironomus dilutus* larvae in sediments from Pearl River tributaries in China (Cheng et al., 2017). Cypermethrin, another pyrethroid for which no guideline could be identified, was measured at the same canal sites at (14.8 and 155 ng g^{-1}). Many pyrethroid insecticides, including permethrin, bifenthrin, and phenothrin were recently identified in HCMC sediments, with particularly high concentrations for permethrin (up to 578 ng g^{-1} dw for the cis-isomer and 3693 ng g^{-1} dw for the trans-isomer) and bifenthrin (477 ng g^{-1} dw; Bui et al., 2016). Furthermore, chlorpyrifos-ethyl displayed concentrations between <LOD (0.3 ng g^{-1} dw) and 6.05 ng g^{-1} dw (median 1.27 ng g^{-1} dw; Table S5-B in the SI), overlapping the range reported in Bui et al., (2016), namely 1.8 – 38.8 ng g^{-1} dw. For both pyrethroid insecticides

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and chlorpyrifos, the higher concentrations reported by this study (Bui et al., 2016) were for canal sites quite far from the river, contrary to the present study.

PCB concentrations are similar to or lower than previous observations, with concentrations in canal sediments ranging from 5 to 123 ng g^{-1} dw for the sum of six indicator congeners (Phuong et al., 1998), or 0.19 to 110 ng g^{-1} dw for the sum of homolog series (Kishida et al., 2007). A previous study reported ΣPCBs between 46 and 150 ng g^{-1} dw in canal sediments, and between 0.33 and 22 ng g^{-1} dw in the sediments from the Saigon and Dongnai rivers (Minh et al., 2007). Again, it is not possible to assess temporal trends on the basis of these results, because of methodological differences such as the list of measured congeners, the presentation of the results or the lack of details on sampling locations.

PAH concentrations are similar to recent findings in HCMC canals, except one site (2339 ng g^{-1} dw) close to an industrial area (Bui et al., 2016). An array of about 60 PAHs having two to six rings were analyzed in canal and river sediments 15 years ago (Kishida et al., 2007). The results are reported by category (two rings, three rings and so on), making it difficult to compare them accurately with our data. Nevertheless, by comparing the sum of concentrations of the compounds common to both studies, the range of values is larger in Kishida et al. (2007) study. Moreover, PAHs in canal sediments from Hanoi displayed higher concentrations (440 – 6210 ng g^{-1} dw) than in the present study (Boll et al., 2008); ΣPAHs in sediments from five sites along the river Kim Nguu in Hanoi city center, were much higher (21.800 – 75100 ng g^{-1} dw; Hung et al., 2014) than in HCMC river sediments.

To our knowledge, PFAS measurements in sediments are currently missing in HCMC waterways. As an indication, the ΣPFASs median concentration in French river sediments was 0.48 ng g^{-1} dw in 2012 (Munoz et al., 2015), while we observed a median of 0.16 ng g^{-1} dw in HCMV. Note that the French study cited included remote, rural, urban and industrial sites, while the 17 sites investigated in this study should all be considered as urban or mixed (urban-industrial), except the upstream sites. To our knowledge, only two other studies attempted to capture PFAS contamination in Vietnamese aquatic environments. A nationwide study conducted in 2013 looked for 11 fluoroalkyl carboxylates (PFCAs) and five sulfonates (PFSAs) in water samples, including three sites in the Saigon River, and six in HCMC canals, rather far from the river (Duong et al., 2015). Concentrations of short-chain PFCAs (PFPeA, PFHxA and PFHpA), as well as those of PFOA or PFOS, reached several ng L^{-1} at some canal sites. Longer-chain PFCAs were not detected. Another study throughout Vietnam conducted in 2013 and 2015 looked for nine PFCAs (from PFHxA to PFTeDA) and four PFSAs (from PFBS to PFDS) in unfiltered water and sediment samples.

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This study included three sampling sites in HCMC along the Saigon River, two sites in the Dongnai River upstream from its confluence with the Saigon River, and four downstream, and eight sites influenced by waste water treatment plant (WWTP) effluents (Lam et al., 2017). The sum of PFAS concentrations in Saigon River water samples varied between 1.7 and 14.6 ng L^{-1} and did not exceed 4.2 ng L^{-1} in Dongnai River water samples, while it reached 107 ng L^{-1} in WWTP/canal samples. Except for PFHxS (detected in ≈ 50 % sediment samples), and several long-chain PFCAs (PFDA, PFUnDA and PFTrDA, detected once out of 11 sites) and PFOS (four sites out of 11), most analyzed PFASs remained below the respective LOQs in sediment samples; however the LOQs reported were generally higher (0.04 – 0.30 ng g^{-1} dw depending of the compound) than those obtained in the present study.

4.2 Organic contaminant fate in Saigon River and canals in HCMC

According to the results of the present study, bed sediments of the Saigon River are weakly contaminated. This is not necessarily surprising for legacy chemicals, such as DDT, which was prohibited in 1993 in Vietnam (Kishida et al., 2007), or PCBs, of which import ceased in 1985 (World-Bank, 2009). This was less expected for more recent substances, such as PBDEs or PFASs, or continuously emitted by multiple sources, such as PAHs. Indeed, PAH concentrations in HCMC are low compared to other urban rivers and canals in the region, such as Hanoi (Kishida et al., 2007; Boll et al., 2008; Hung et al., 2014) and Bangkok (Boonyatumanond et al., 2006) to a lesser extent. The PAH concentrations in Saigon River sediments were similar to those of rural areas or remote (i.e. reference) sites elsewhere in Vietnam (Kishida et al., 2007; Boll et al., 2008). However, some sampling sites in Hanoi studies were presumably located closer to industrial releases than the HCMC sampling sites. After removal of these assumed outliers, the range of ΣPAHs observed in Hanoi canal and river sediments remained higher than in HCMC canal sediments, but similar to Bangkok canal sediments (Figure 3). Interestingly, there is a strong contrast between PAH concentrations in Bangkok canals and in the Chao-Praya River, which flows through this city, and shortly after into the sea.

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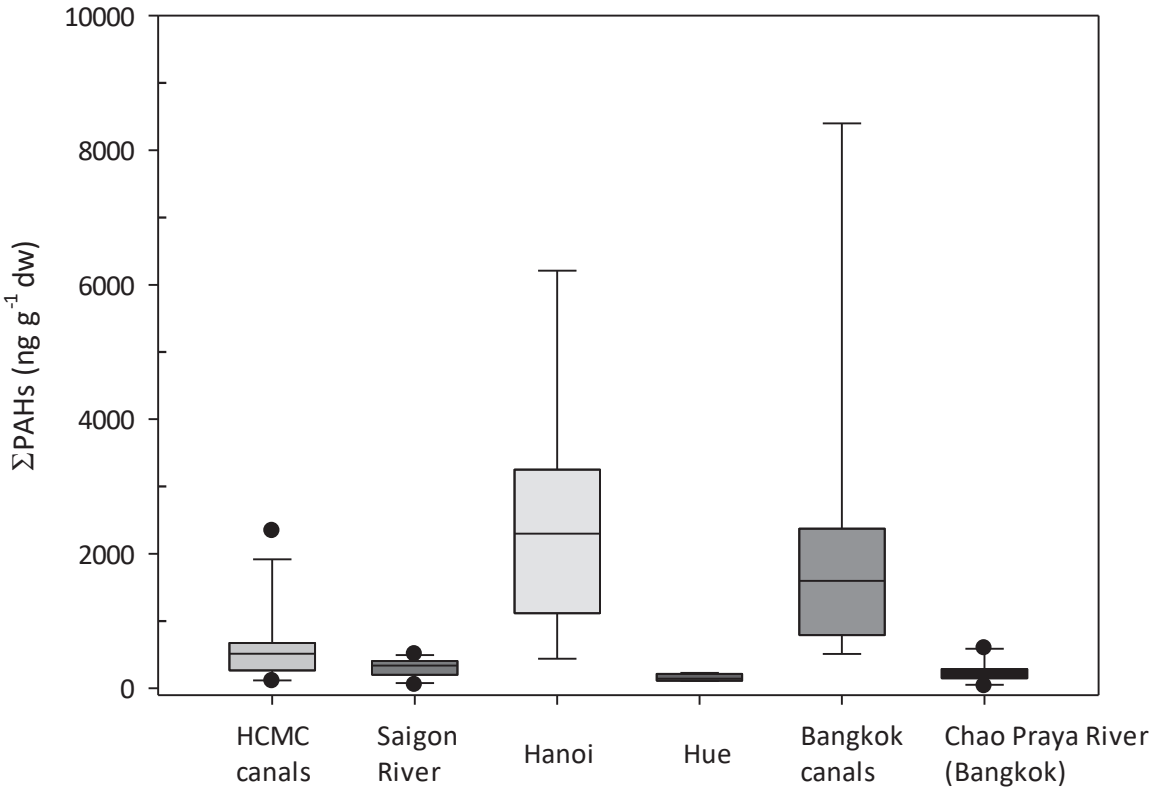

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241 Figure 3 – Box-plot of ΣPAHs concentrations in various locations in southeast Asia (HCMC canals and Saigon River,
242 this study and (Bui et al., 2016); Hanoi and Hue data from (Kishida et al., 2007); Bangkok canals and Chao Praya
243 River data from (Boonyatumanond et al., 2006)

244 Emission patterns might differ among these cities. Boonyatumanond et al. analyzed triterpanes (markers of
245 petroleum origin) in Bangkok canal and Chao-Praya River sediments, and compared the molecular profiles in
246 sediments and urban dust (Boonyatumanond et al., 2006). They concluded that road traffic was one of the major
247 sources of PAHs in canals, while a mixture of petrogenic and pyrogenic sources was demonstrated in the lower
248 Chao-Praya River. Soil erosion due to frequent heavy rains was also suggested as a PAH pathway to the aquatic
249 system. In Hanoi sediments it was also claimed that PAH concentrations were the consequence of a mixture of
250 domestic (cooking), industrial and road traffic related sources, the latter being a major contributor (Boll et al.,
251 2008). These South-Asian cities therefore have in common with HCMC that road traffic is a major PAH source, as
252 illustrated by similar individual PAH ratios (Figure S2 in the SI). PAHs from road traffic further deposit on urban
253 surfaces, either directly or after a stage in the atmosphere.

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While pointing out low PAH concentrations in Malaysian rivers, estuaries and coastal sediments compared to Tokyo and other industrialized areas, Zakaria et al. showed by several means that petrogenic inputs were a major control factor of ΣPAHs concentrations in this set of locations (Zakaria et al., 2002). They claimed that street dust might represent 30% or more of ΣPAHs in some sediment samples. They suggested that urban runoff generated by heavy tropical rains wash out land-based pollutants to aquatic media more effectively than at higher latitudes characterized by less intense rainfall (Zakaria et al., 2002). Boonyatumanond et al. also noticed that ΣPAHs in Bangkok were low compared to other cities around the world, and attributed the lower levels they observed to climatic factors, which would have enhanced PAH degradation (Boonyatumanond et al., 2006). We argue that this cannot actually explain the differences among the three large cities recorded in Figure 3, which experience similar climatic conditions, nor the contrast between canal and river sediments in Bangkok and HCMC. On the other hand, heavy rains (monsoon) occur in Bangkok (mean 1650 mm per year; TMD, 2018) as in Hanoi (mean 1680 mm per year; GSO, 2016). These rainfall records are comparable to HCMC (1800 mm), and would provoke similar surface runoff on impervious urban surfaces and transfer of PAH contaminated particles to urban waterways in the three cities.

In addition to degradation processes, several factors control sedimentary PAH concentrations in surface sediments, in particular (i) the rate of PAH input, (ii) the rate of sediment input, (iii) the resuspension and redeposition of PAHs and sediment particles, and (iv) some vertical mixing due to biological or physical processes (Boonyatumanond et al., 2006). While potentially significant at small spatial scales, bioturbation (i.e., vertical mixing due to biological processes) should be less influential at larger spatial scales than processes involving sediment particle movements in a dynamic system. An attempt to summarize the interplay of these processes is presented in Figure 4. As discussed above, we assume that PAH input rates are similar in Bangkok, Hanoi and HCMC. PAHs are emitted to the atmosphere by industries or road traffic, and later deposited on urban surfaces. PAHs may also be released directly in canals or into the river by industries. Only these two sources are presented for the sake of clarity; complementary sources, e.g., biomass burning, also emit PAHs to the atmosphere, so adding this type of source would not significantly change our comprehension of PAH fate in this system. Sediment rates of inputs in the Saigon River are influenced by the Dau Tieng reservoir, which decreases the overall particle flux from upstream, and releases finer particles. Inputs from the city, i.e., canal sediments, are probably limited during the dry season, because most canal downstream ends are closed by floodgates, so as to prevent flooding

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in more exposed urban areas (Ngoc et al., 2016). As a consequence of the succession of flow and ebb tides, exceeding by far the net flow of the Saigon River (Camenen et al., 2017), sediment particles are in the meantime brought from downstream, i.e. from the Dongnai River and the estuary, deposited and resuspended, Nevertheless, according to the flow velocity of 1.0 – 1.5 m sec^{-1} (Camenen et al., 2017), the net deposition rate should be low. Thus the PAH load in Saigon River sediments might be low in the dry season because of the dilution caused by this redistribution of sediment particles in the river and the low PAH inputs (Figure 4-A). The same presumably arises in Chao Praya River in Bangkok, which is subjected to tidal influence (Chinnarasri and Kemden, 2016), but not in Hanoi, which is far from the sea (Luu et al., 2010).

This pattern changes drastically during the monsoon (Figure 4-B). Under heavy rain, high volumes of water are discharged first on urban surfaces, where they wash out deposited PAHs, then into the canals. At ebbing tide, floodgates open, so as to drain off the rainwater. As a consequence, urban dust as well as canal sediments will be flushed to the Saigon River. The net Saigon downstream flow increases in this season by a factor of about 10 (Camenen et al., 2017), accelerating the sediment transfer towards the estuary. Sediment deposits in the estuary, in particular in the mangrove, might thus be more contaminated than Saigon River sediments.

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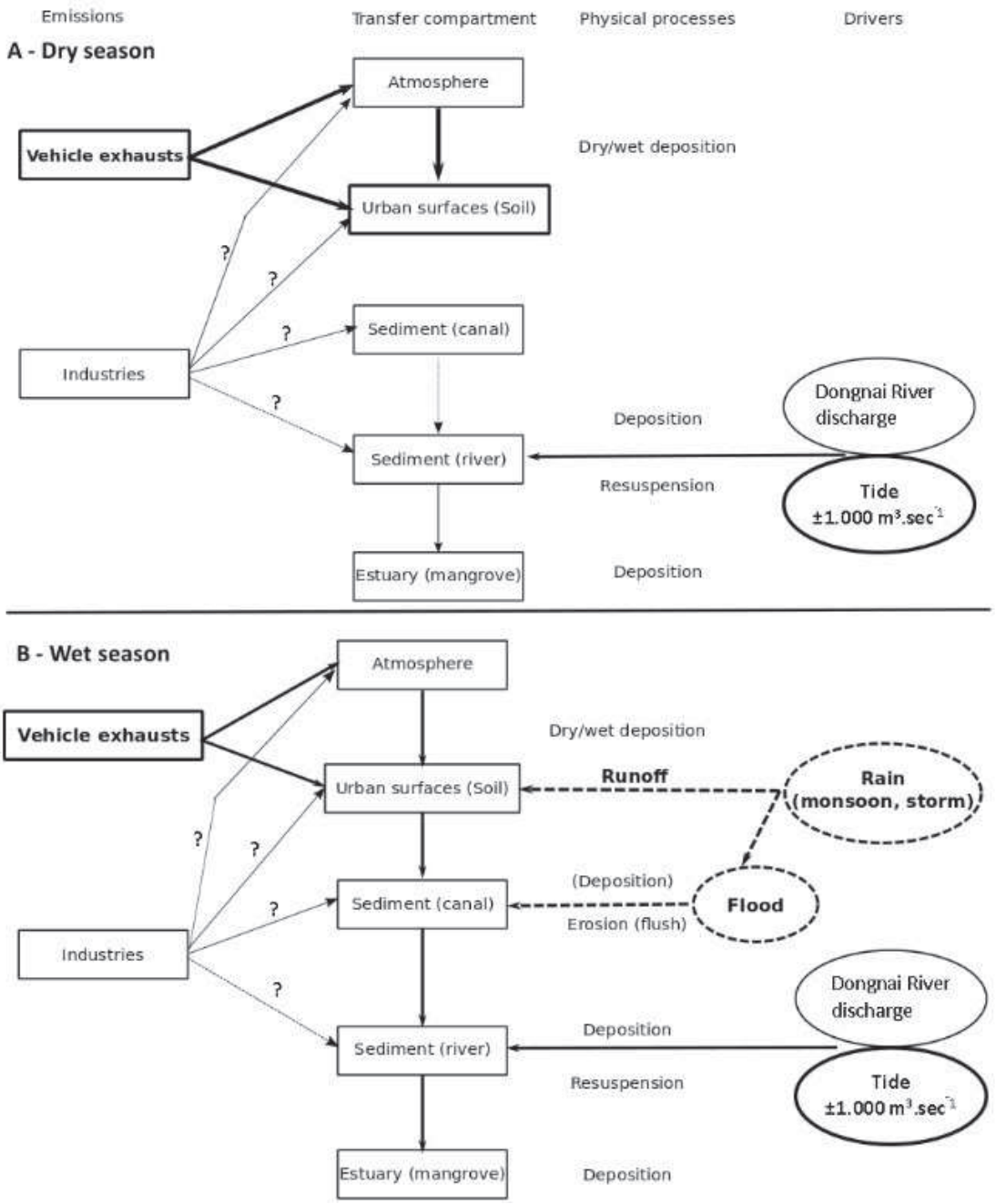

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298 Figure 4 – Tentative scheme of PAH fate in the HCMC-Saigon-Dongnai river system

299 Can this scheme apply to other persistent organic chemicals, such as PBDEs or PFASs? No information could be
300 found in the literature on the occurrence of PBDEs in HCMC sediments, nor in other media. Low PBDE
301 concentrations were found some years ago in catfish from the Mekong River delta (Minh et al., 2006). These
302 compounds were recently found in sediments from rivers and lakes in the Hanoi region, with concentrations
303 ranging between 0.04 and 17.5 ng g^{-1} (dw) for the sum of seven congeners representing the most common PBDE
304 technical mixtures (Mai et al., 2018). They were also found in Hanoi road dust, at concentrations comparable to

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305 those from Chinese cities (Anh et al., 2018). Furthermore, a large PBDE monitoring study focusing on sediment
306 samples collected from urban canals or rivers in Lao PDR, Cambodia, Vietnam, India, Indonesia, Thailand, the
307 Philippines, Malaysia and Japan, including four samples from Hanoi, claimed that there was a relationship
308 between the gross domestic product (GDP) per capita in emergent countries and ΣPBDE concentrations in urban
309 river sediments (Kwan et al., 2013). The predominant congener in this study was BDE209 (43-97% of ΣPBDE),
310 which was not analyzed in HCMC samples. PBDE emissions in HCMC are therefore likely comparable to Hanoi
311 emissions, although there is currently no direct evidence for this. The fact that PBDEs remained undetected in
312 Saigon River sediments is accordingly consistent with the framework described above for PAHs.

313 As mentioned above, there are a few data attesting of the contamination of HCMC waters by PFCAs and PFSAs
314 (Duong et al., 2015; Lam et al., 2017). Concentrations up to 9.2 ng L^{-1} for PFHpA, 5.6 – 5.8 ng L^{-1} for PFHxA and
315 PFPeA, and 18 ng L^{-1} for PFOA were observed at some canal sites (Duong et al., 2015). Lam et al. (2017) searched
316 for a wider range of PFASs and investigated several environmental compartments, and identified long-chain
317 PFCAs in some sediment samples. Both studies support consistent PFAS emissions from numerous kinds of
318 sources, including consumers products (Duong et al., 2015), industries (metal, textile, paper) and domestic waste
319 water discharges (Lam et al., 2017). Beside PFCAs and PFSAs, FTSAs were also identified in 47-65 % of sediment
320 samples, as well as di-PAPs (65-88%), The former compounds arise from the degradation of fluorotelomer-based
321 substances (precursors) used, for example, in firefighting foams or food-packaging, applications and were found
322 in waste waters in several instances (Buck et al., 2011). Polyfluoroalkyl phosphoric acid di-esters (di-PAPs) are
323 used primarily for their surfactant properties in various applications such as food-packaging or a defoaming agent
324 in pesticide formulations (Buck et al., 2011) and were found in waste waters (D'Eon et al., 2009). PFASs (PFCAs,
325 PFSAs or precursors of these acids) have also been identified in rainwater and runoff in urban areas (Zhao et al.,
326 2013). Cumulative rainwater was associated with PFBA and PFBS in the dissolved phase of an urban tropical
327 catchment, suggesting these compounds were mainly brought into the water body by runoff, while several other
328 PFASs originated from point sources (Chen et al., 2017). There are accordingly consistent indications pointing to
329 PFAS emissions in HCMC's environment, although the balance between the routes might differ from that of PAHs,
330 especially considering the fact that atmospheric deposition is unlikely a major source of PFASs in densely
331 populated areas (Lindim et al., 2015).

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5 Conclusion

We searched for a wide range of organic contaminants in bed sediments from the Saigon River, and a few adjacent canals. Measured concentrations were below the LOD for PBDEs and some PFASs, and low for DDT, PCBs, PAHs and several PFASs, except for the insecticides chlorpyrifos-ethyl and λ-cyhalothrin at a few places. We explain the low concentrations observed by the strong tidal influence, which hinders the accumulation of sediments in the Saigon River channel, and the runoff on urban surfaces during the monsoon. This contamination transfer pattern raises concerns about potential impacts further downstream in depositional areas such as the Can Gio mangrove (Schwarzer et al., 2016), an ecosystem providing important services, in particular fisheries (Vo and Kuenzer, 2013).

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