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### ► To cite this version:

Marc Babut, Brice Mourier, Marc Desmet, Caroline Simonnet-Laprade, Pierre Labadie, et al.. Where has the pollution gone? A survey of organic contaminants in Ho Chi Minh city / Saigon River (Vietnam) bed sediments. *Chemosphere*, 2019, 217, pp.261-269. 10.1016/j.chemosphere.2018.11.008 . hal-02018741

**HAL Id: hal-02018741**

**<https://univ-lyon1.hal.science/hal-02018741v1>**

Submitted on 16 May 2020

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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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Role of the Funding source

The field work as well as analyses were funded by the Auvergne-Rh one-Alpes Region, through the support afforded by "COOPERA" program (grant CMIRA # 15 005475 01) to the collaborative project "Saigon River – La ville & le fleuve (2015-2017) : Impacts environnementaux, inondations et contaminations dans une trajectoire historique". The Auvergne-Rh one-Alpes Region was not involved in the study design, nor in data processing, report writing and article submission.

### *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  $\lambda$ -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

## 1    **1    Introduction**

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

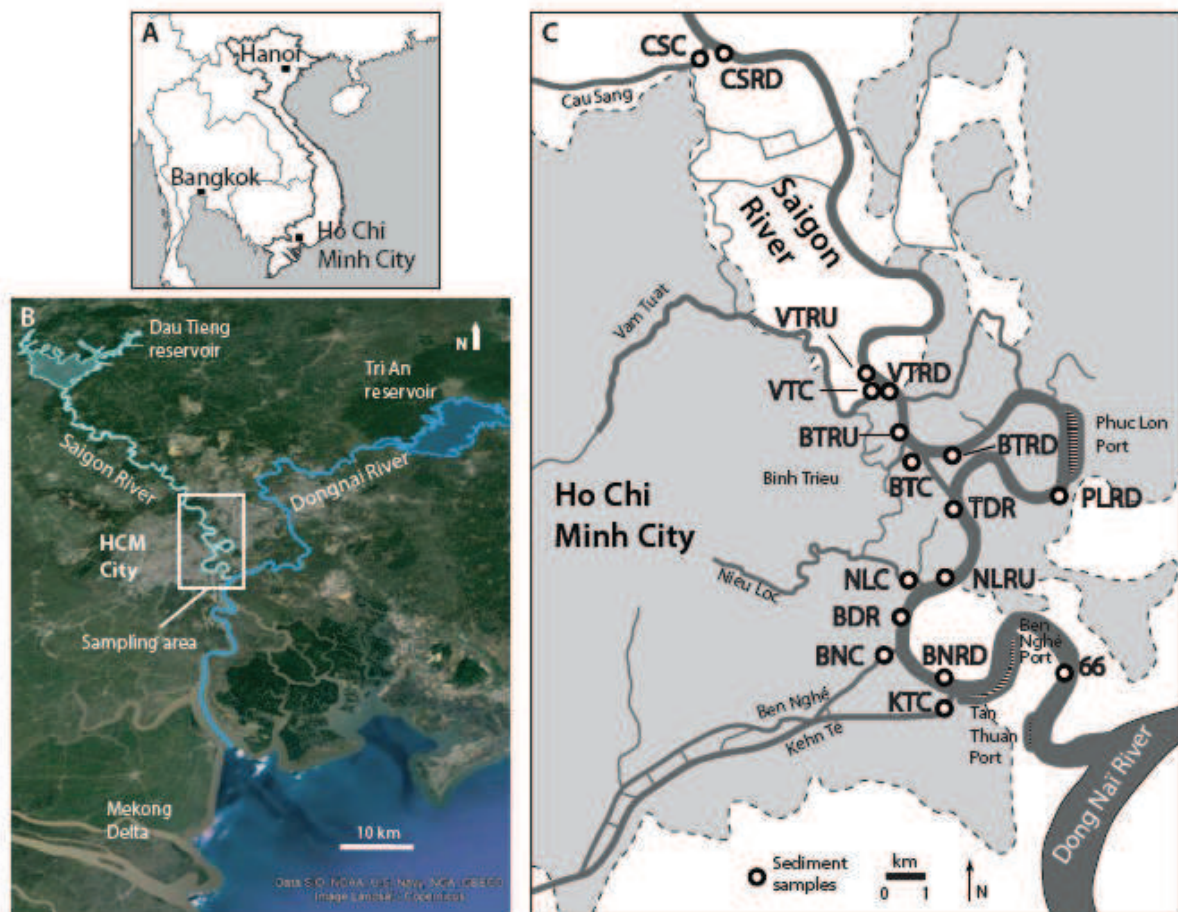
8    As a consequence, anthropogenic pressures on HCMC's environment have also rapidly increased, leading among  
9    others to potential substantial releases of a wide array of organic contaminants into the atmosphere or aquatic  
10    media throughout the area. Insecticides have been used for a long time for malaria vector control: Diphenyl-  
11    diethyl-trichlorethane (DDT) was authorized until 1993 (Kishida et al., 2007); alternative compounds such as  
12    organophosphates or pyrethroids have been introduced since then. Industrial production throughout HCMC  
13    district include pharmaceuticals, plastics, packaging, textile and leather garments, paints and varnish, and  
14    electronics (HEPZA, 2015). In addition, the road traffic in the city and its surroundings is quite intense. Potential  
15    emissions accordingly involve a wide range of compounds: DDT and other insecticides, polychlorinated biphenyls  
16    (PCBs), polycyclic aromatic hydrocarbons (PAHs), flame retardants, per- and poly-fluorinated substances (PFASs)  
17    and so on. Some of them were already monitored in the past, such as PCBs (Phuong et al., 1998; Kishida et al.,  
18    2007; Minh et al., 2007; Nguyen, 2009), DDT and its metabolites (Phuong et al., 1998; Kishida et al., 2007), and  
19    PAHs (Kishida et al., 2007). Monitoring data on other contaminants such as polybrominated diphenyl ethers  
20    (PBDEs) are still rare or absent.

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

## 26 2 Materials and methods

### 27 2.1 Study area

28 Ho Chi Minh City (HCMC) is located along the Saigon River, which is a tributary of the Dongnai River (Figure 1).  
29 The respective catchments cover 4 700 km<sup>2</sup> and 40 000 km<sup>2</sup> respectively, and are subject to a monsoon regime,  
30 with about 150 rainy days between May and late November and an average annual rainfall of 1800 mm (GSO,  
31 2016). Both river flows are regulated by upstream reservoirs, namely Dau Tieng on the Saigon River and Tri An  
32 on the Dongnai River (Figure 1-B). Respective mean discharges are estimated around 50-60 and 600-650 m<sup>3</sup> s<sup>-1</sup>  
33 (Ha, 2009; Camenen et al., 2017). Given the low altitude and the flat topography, this system is subject to a strong  
34 tidal influence, far upstream from the urbanized areas.



35  
36 Figure 1 - Study area (A- regional map; B- HCMC area; C- sampling sites in HCMC)

### 37 2.2 Sediment sampling

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

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

### 43 **2.3 Sediment characteristics**

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

### 49 **2.4 Chemical analysis**

#### 50 **2.4.1 Chlorinated pesticides, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs)** 51 **and polybrominated diphenyl ethers (PBDEs)**

52 These compounds were analyzed following the approach described in previous papers (Thevenon et al., 2013;  
53 Mwanamoki et al., 2014; Kilunga et al., 2017). Briefly, sediment samples were first freeze-dried and sieved  
54 (2mm). Then, after addition of internal standards (<sup>13</sup>C-labeled for halogenated compounds and <sup>2</sup>H-labeled  
55 compounds for all PAHs), about 5 g of dry sediment were extracted with a mixture of 20 % acetone in hexane  
56 (v/v) into a Soxhlet system. Interfering sulfur compounds were removed by addition of activated copper to the  
57 extract. Then, the organic extract was concentrated to 1 mL in a vacuum rotary evaporator (Buchi Rotavapor,  
58 Flawil, Switzerland). The extract was further submitted to fractionation and clean-up over a chromatographic  
59 column containing 3 g of Silicagel, according to de Boer et al. (2001). Three separated fractions were collected:  
60 first with 16 mL of hexane, then 35 mL of hexane, and finally 50 mL of hexane: dichloromethane (v/v, 1:1). The  
61 first two fractions contained PCBs and PBDEs respectively. PAHs and chlorinated pesticides were distributed into  
62 the three fractions, which were analyzed separately. After reducing the volume again, the chemicals were  
63 measured by gas chromatography with triple mass spectrometry detection (GC-MS/MS, Thermo Scientific, TSQ  
64 Quantum XLS Ultra, Waltham, MA, USA). Two columns with different polarities, a ZB-5ms column (60 m x 0.25  
65 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  
66 of the different compounds. The list of compounds analyzed with this approach is provided in the SI, section 2.

#### 67 **2.4.2 Per- and poly-fluorinated alkyl substances (PFASs)**

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

#### 76 **2.4.3 QA/QC**

77 Almost all chlorinated pesticides, PCBs and PBDEs were included in the second round of UNEP-coordinated Global  
78 Interlaboratory Assessment 2012/2013 (UNEP, 2014). In this exercise dozens of laboratories analyzed the same  
79 sediment sample. Results from our team are presented in the SI, Table S2. Additionally, other pesticides such as  
80 chlorpyrifos-methyl and -ethyl, endosulfan I, II and -sulfate, endrin aldehyde and -ketone, methoxychlor,  
81 acetochlor, λ-cyhalothrin, α and β-cypermethrin and deltamethrin were also analyzed. Recovery rates for PCBs  
82 varied between 80.5% and 85.3%, and between 61.3% and 103.7% for PAHs. PBDE recovery rates varied between  
83 80.3% and 94.7%, while for pesticides they ranged from 51.5% (endosulfan) to 90.4% (δ-HCH). The limits of  
84 detection (LODs) are indicated in Table S2 in the SI.

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

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

#### 91 **2.5 Statistics**

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



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

99 The significance threshold was set at 0.05 in all analyses.

## 100 **3 Results**

### 101 **3.1 Sediment characteristics**

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

### 109 **3.2 Sediment contamination**

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



121 **3.2.1 Insecticides**

122 Chlordane  $\alpha$  and  $\gamma$  isomers were detected in four samples, at concentrations close to the LOD (0.2 ng g<sup>-1</sup> dw).  
123  $\Sigma$ DDT, i.e. the sum of concentrations of DDT isomers and their metabolites, were determined in all samples, and  
124 varied between 2.40 ng g<sup>-1</sup> dw and 16.05 ng g<sup>-1</sup> dw; *p,p'* DDD and *p,p'* DDE were predominant throughout the  
125 samples.  $\Sigma$ DDT concentrations in canal samples were slightly lower than in river samples, but the difference was  
126 not significant (*p*-value 0.17, Table 1).

127 The pyrethroid insecticide  $\lambda$ -cyhalothrin, was measured at two canal sites (0.91 and 23.2 ng g<sup>-1</sup>), plus an adjacent  
128 river site (0.97 ng g<sup>-1</sup>) for  $\lambda$ -cyhalothrin, at concentrations several orders of magnitude above the detection limit  
129 (0.3 ng g<sup>-1</sup> dw; Table S5-B in the SI).

130 **3.2.2 PCBs**

131 With a maximum  $\Sigma$ PCB7 of 18.7 ng g<sup>-1</sup> dw (sum of indicator congener concentrations, i.e. CB 28, 52, 101, 118,  
132 138, 153 and 180), or 25.3 ng g<sup>-1</sup> dw for  $\Sigma$ 12PCB (12 measured congeners), the contamination by this class of  
133 compounds remained moderate, as compared e.g. to existing sediment quality guidelines (MacDonald et al.,  
134 2000). For two samples (CSC, BNRD), all analyzed congeners were < LOD. Otherwise, the molecular profiles were  
135 dominated by congeners 138, 153 and 180, with median values equaling 0.27 ng g<sup>-1</sup> dw, 0.51 ng g<sup>-1</sup> dw, and 0.22  
136 ng g<sup>-1</sup> dw respectively.

137 **3.2.3 PAHs**

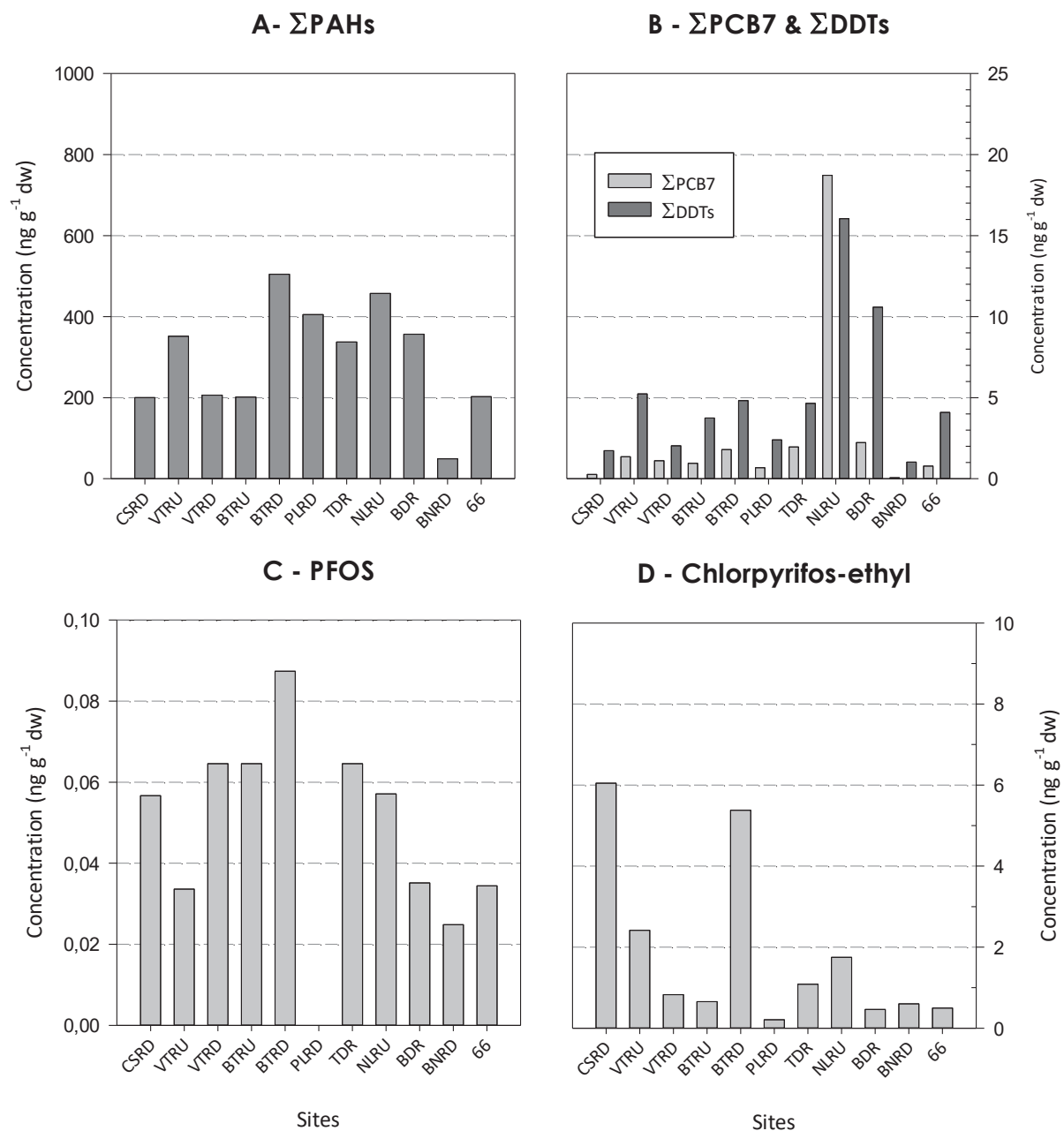
138  $\Sigma$ PAHs (i.e. the sum of the concentrations of the 16 compounds) varied between 49 ng g<sup>-1</sup> dw and 933 ng g<sup>-1</sup> dw  
139 (median 345 ng g<sup>-1</sup> dw; SI Table S5-C), well below existing sediment quality guidelines (e.g. MacDonald et al.,  
140 2000). Phenanthrene, fluoranthene, pyrene and benzo(b)fluoranthene were the main contributors to  $\Sigma$ PAHs,  
141 representing respectively 12%, 17%, 14% and 11% of the sum on average (Figure S1 in SI). Whereas phenanthrene  
142 is deemed to result mainly from OM maturation, i.e., petrogenic origin, fluoranthene and pyrene instead point  
143 to pyrogenic sources, such as vehicle exhaust or wood coal combustion (Zhang et al., 2013). Applying  
144 concentration ratios between specific PAHs (Yunker et al., 2002) tended to confirm this diagnosis of multiple  
145 sources, with a predominance of pyrogenic sources (Figure S2 in the SI). Nevertheless  $\Sigma$ PAHs in all samples  
146 remained below the probable effect concentration of 2280 ng g<sup>-1</sup> dw (MacDonald et al., 2000).

147 **3.2.4 PFASs**

148 Overall, the PFASs detected displayed rather low concentrations, e.g.  $\Sigma$ PFCAs, 8:2 FTSA, 6:2 di-PAP and 8:2 diPAP  
149 maximum concentrations at 1.18 ng g<sup>-1</sup> dw, 0.08 ng g<sup>-1</sup> dw, 0.18 ng g<sup>-1</sup> dw and 0.24 ng g<sup>-1</sup> 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  $\lambda$ -cyhalothrin (23.2 ng g<sup>-1</sup> dw) and cypermethrin  
160 (155 ng g<sup>-1</sup> dw). While canal sediments showed higher concentrations than river sediments for the most  
161 frequently quantified parameters, namely  $\Sigma$ DDTs,  $\Sigma$ PAHs,  $\Sigma$ PCB7 or chlorpyrifos-ethyl, the differences were not  
162 significant (Table 1).



163  
 164 Figure 2 - PAHs, PCBs, DDTs, PFOS, and Chlorpyrifos-ethyl in Saigon River sediments from upstream (CSRD) to  
 165 downstream (66) of HCMC  
 166

Contaminant	Canals		River		<i>p</i> -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

167 Table 1 – Comparison of the concentrations (ng g<sup>-1</sup> dw) in canal and river sediments (Mann-Whitney test)

168 **4 Discussion**

169 4.1 Sediment quality - Comparison to other studies

170 DDT compounds have been measured in HCMC sediment samples since the 1990s: Phuong et al. mentioned a  
 171 ΣDDTs up to 253 ng g<sup>-1</sup> dw, with a median value of 49 ng g<sup>-1</sup> dw in canal sediments (Phuong et al., 1998), while  
 172 about 10 years later the concentration ranges were 12 – 72 ng g<sup>-1</sup> dw in canal sediments, and 0.21 – 23 ng g<sup>-1</sup> dw  
 173 in Saigon and Dongnai river sediments (Minh et al., 2007). At the same period, another study in canal and river  
 174 sediments showed ΣDDTs ranging from 0.1 to 38.3 ng g<sup>-1</sup> dw (Kishida et al., 2007), similar to the recent findings  
 175 (1.8 – 74 ng g<sup>-1</sup> dw) by Bui et al. (2016). Although it was not possible to compute these data in a single trend  
 176 assessment, in part because most locations are not accurately reported in previous studies, there is no evidence  
 177 of a sharp ΣDDT decline in the last two decades. Overall, even the highest concentrations recorded in 1998  
 178 remained far below the probable effect concentration of 572 ng g<sup>-1</sup> dw derived from co-occurrence studies in  
 179 North America (MacDonald et al., 2000).

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

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

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

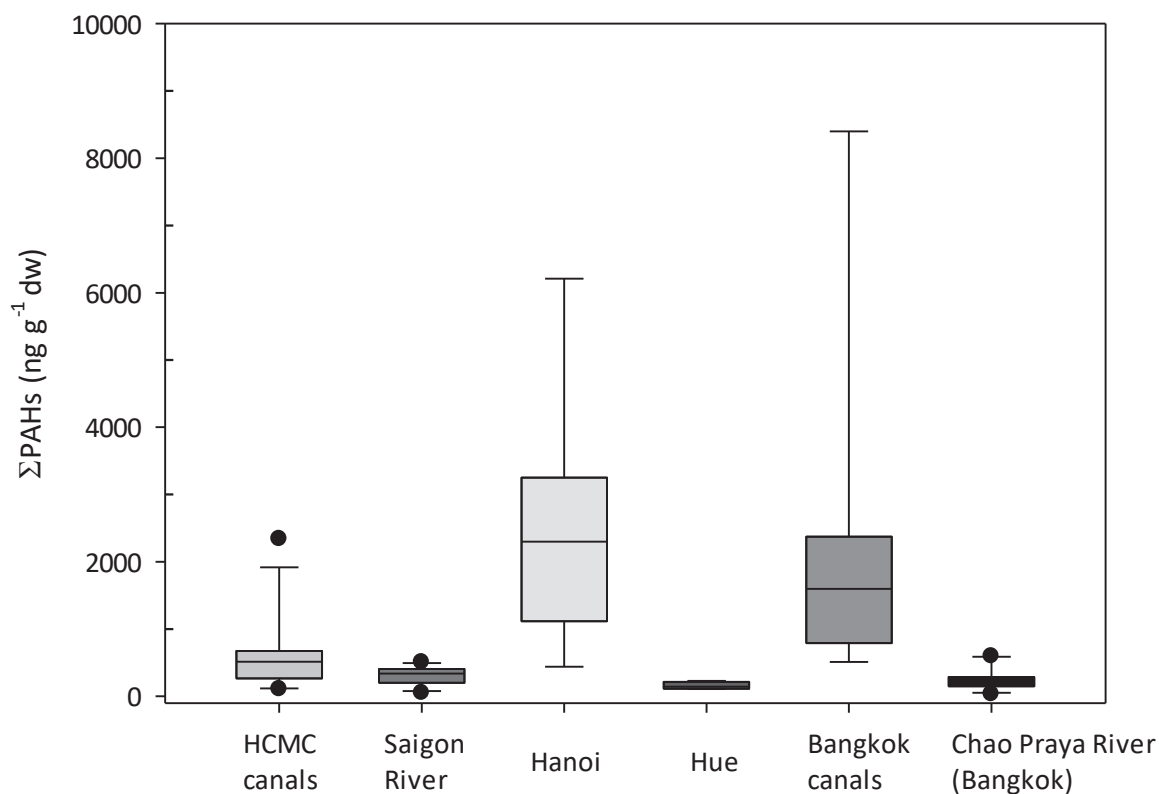
198 PAH concentrations are similar to recent findings in HCMC canals, except one site (2339 ng g<sup>-1</sup> dw) close to an  
199 industrial area (Bui et al., 2016). An array of about 60 PAHs having two to six rings were analyzed in canal and  
200 river sediments 15 years ago (Kishida et al., 2007).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

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

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

#### 226 4.2 Organic contaminant fate in Saigon River and canals in HCMC

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



240  
241 Figure 3 – Box-plot of  $\Sigma$ PAHs 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.



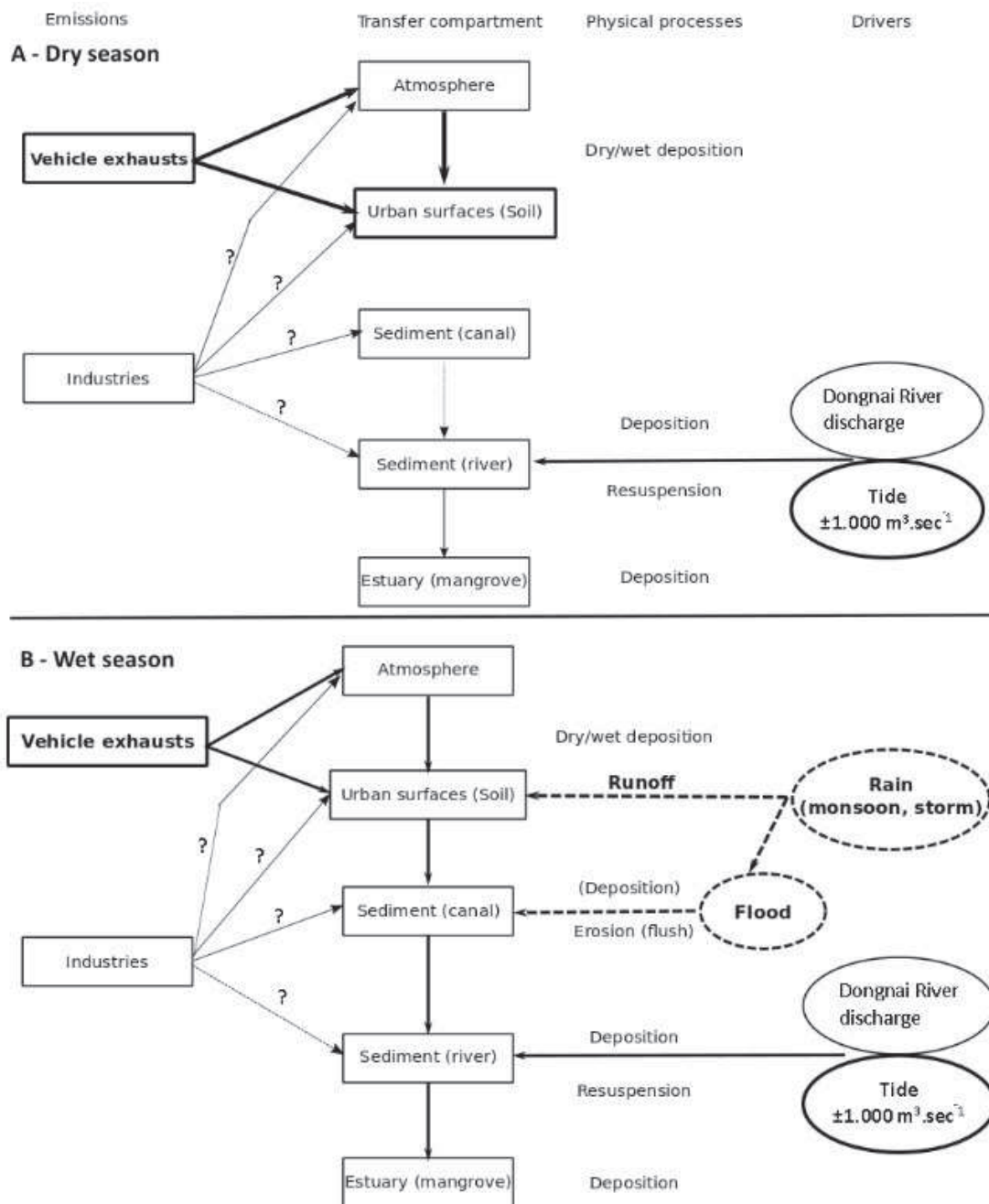
254 While pointing out low PAH concentrations in Malaysian rivers, estuaries and coastal sediments compared to  
255 Tokyo and other industrialized areas, Zakaria et al. showed by several means that petrogenic inputs were a major  
256 control factor of  $\Sigma$ PAHs concentrations in this set of locations (Zakaria et al., 2002). They claimed that street dust  
257 might represent 30% or more of  $\Sigma$ PAHs in some sediment samples. They suggested that urban runoff generated  
258 by heavy tropical rains wash out land-based pollutants to aquatic media more effectively than at higher latitudes  
259 characterized by less intense rainfall (Zakaria et al., 2002). Boonyatumanond et al. also noticed that  $\Sigma$ PAHs in  
260 Bangkok were low compared to other cities around the world, and attributed the lower levels they observed to  
261 climatic factors, which would have enhanced PAH degradation (Boonyatumanond et al., 2006). We argue that  
262 this cannot actually explain the differences among the three large cities recorded in Figure 3, which experience  
263 similar climatic conditions, nor the contrast between canal and river sediments in Bangkok and HCMC. On the  
264 other hand, heavy rains (monsoon) occur in Bangkok (mean 1650 mm per year; TMD, 2018) as in Hanoi (mean  
265 1680 mm per year; GSO, 2016). These rainfall records are comparable to HCMC (1800 mm), and would provoke  
266 similar surface runoff on impervious urban surfaces and transfer of PAH contaminated particles to urban  
267 waterways in the three cities.

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

282 in more exposed urban areas (Ngoc et al., 2016). As a consequence of the succession of flow and ebb tides,  
283 exceeding by far the net flow of the Saigon River (Camenen et al., 2017), sediment particles are in the meantime  
284 brought from downstream, i.e. from the Dongnai River and the estuary, deposited and resuspended,  
285 Nevertheless, according to the flow velocity of  $1.0 - 1.5 \text{ m sec}^{-1}$  (Camenen et al., 2017), the net deposition rate  
286 should be low. Thus the PAH load in Saigon River sediments might be low in the dry season because of the dilution  
287 caused by this redistribution of sediment particles in the river and the low PAH inputs (Figure 4-A). The same  
288 presumably arises in Chao Praya River in Bangkok, which is subjected to tidal influence (Chinnarasri and Kemden,  
289 2016), but not in Hanoi, which is far from the sea (Luu et al., 2010).

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

296



297

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<sup>-1</sup> (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

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  $\Sigma$ PBDE concentrations in urban  
309 river sediments (Kwan et al., 2013). The predominant congener in this study was BDE209 (43-97% of  $\Sigma$ PBDE),  
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 PFSA  
314 (Duong et al., 2015; Lam et al., 2017). Concentrations up to 9.2 ng L<sup>-1</sup> for PFHpA, 5.6 – 5.8 ng L<sup>-1</sup> for PFHxA and  
315 PFPeA, and 18 ng L<sup>-1</sup> 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).

## 332 5 Conclusion

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

341

## 342 Acknowledgements

343 This study was supported by the Auvergne-Rhône-Alpes Region, through the CMIRA-Coopera program, “Saigon  
344 River: la ville et le fleuve, 2015-2017” project. We thank the Centre Asiatique de Recherche sur l’Eau (CARE) staff  
345 in Ho Chi Minh City for their kind and efficient help during field work, as well as EPFL in Lausanne which co-funded  
346 a part of the analytical work. We also thank Thanh-Kiet Bui and Carmen Casado-Martinez who kindly provided  
347 data from their own field work in Ho Chi Minh City. We thank Linda Northrup (English Solutions, Voiron, France),  
348 for copy-editing the manuscript.

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