

# A nationwide survey of perfluorinated alkyl substances in waters, sediment and biota collected from aquatic environment in Vietnam: Distributions and bioconcentration profiles



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## HIGHLIGHTS

- Baseline information for 13 PFASs contamination in Vietnamese riverine ecosystems.
- The most abundance of PFOA concentration in waters.
- Site-specific loads of PFASs contamination in surface and tap waters.
- PFOS and PFHxS were found predominant in sediment.
- PFNA, PFUnDA, PFTrDA and PFHxS were more abundant than other PFASs in biota tissues.

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## ABSTRACT

Water, sediment, various tissues of fish, crustacean, gastropod and bivalve were collected from major river basins in Vietnam and analyzed for the presence of perfluorinated alkyl substances (PFASs). Furthermore, the occurrence of PFASs in coastal, tap and well waters collected from eight different regions in Vietnam was investigated. PFOA and PFOS were consistently detected as the dominant PFASs in surface waters. The greatest concentrations of PFOA (53.5 ng L<sup>-1</sup>) and PFOS (40.2 ng L<sup>-1</sup>) were found in a surface water sample collected from a channel that receives wastewater treatment plant discharges. PFOS and PFHxS were found as the predominant PFASs in sediments. The greatest PFAS concentration in biota was 16.9 ng PFUnDA g<sup>-1</sup> wet weight found in a fish liver. Some long-chain PFASs including PFNA, PFUnDA and PFTrDA as well as PFHxS were more abundant than short-chain PFASs in biota tissues. The measured concentrations of PFOS and PFOA in surface and tap waters were below the provisional health advisory. The rank order of mean bioconcentration factor of PFOS in biota was; crustacean (115 L/kg), gastropod (1117 L/kg), fish (1120 L/kg) and bivalve (2110 L/kg). This study provides baseline information for a better understanding of PFASs contamination in Vietnam.

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## 1. Introduction

Perfluorinated alkyl substances (PFASs) include a wide range of organofluorine compounds that have some unique properties including extreme thermal and chemical stability. These chemical attributes also contributed to their ubiquitous distribution in the environment [1–3], wildlife [4] and humans [5,6]. Perfluorooctane sulfonate (PFOS) and several other long-chain PFASs can biomag-

nify in the food chain and elevated concentrations were found in animals at higher trophic position [7,8]. To humans, the major routes of PFAS exposures include diet, drinking water, and indoor dust [9–12] and the half-lives of some PFASs in human were calculated to be in the range of 4–7 years [13].

In an effort to reduce the widespread global contamination by PFOS, the 3M Company, a major producer of this compound, phased out its production in the USA in 2001 [4]. PFOS and, its salts and perfluorooctane sulfonyl fluoride were listed on Annex B of The Stockholm Convention on persistent organic pollutants by the Fourth Conference of Parties in May 2009. The European Union has submitted a proposal to list perfluorooctanoic acid (PFOA), its

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salts and PFOA-related compounds in Annexes A,B and/or C to the Stockholm Convention in the Eleventh Meeting in October 2015. In addition, over 300 alternative chemicals for PFOA and related chemicals were reported to have been received and reviewed by the United States Environmental Protection Agency through September 2015 [14].

From the past decade, Asia Pacific is one of the fastest growing regions of fluorochemical production. Particularly, there was a significant growth of fluorochemical demand in China and this country has become a fluorochemical producer as well [15]. In addition, six smaller scale manufactures of perfluorooctane sulfonyl fluoride (PFOF) were reported to be existed in Asia (4 in Japan) in comparison to those in U.S. and Europe [16]. Recently, relatively high perfluorooctanoic acid (PFOA) concentrations in ground water ( $524 \text{ ng L}^{-1}$ ), water ( $668 \text{ ng L}^{-1}$ ) and sediment ( $48 \text{ ng g}^{-1}$  dry weight) from river, and human serum ( $93 \text{ ng mL}^{-1}$ ) were reported for environment and residents nearby fluorochemical plants in Fuxin, China [17]. Despite this, yet little is known about the risks of PFAS in environment and humans. It has been to note a recent review research considered that up-to-date data was insufficient to draw firm conclusions regarding the role of PFOA for any of the disease of concern [18]. Last but not least, it is suggested that the transformation of several PFASs in atmosphere could result in derivatives that can potential contribute to the global warming [19].

The global scale diffusion of PFASs has been firstly reported with the evidence of PFASs contaminants in wildlife from remote and polar areas [4]. Hundreds of studies on environmental measurement of PFASs contaminations have been then reported, mostly for North America and Europe [20], but the ultimate environmental fate and pathways of PFASs still remain an uncertainty [21]. Up to date, the most appropriate hypotheses reported for the long range transport of PFASs from anthropogenic activities areas to remote regions are the atmospheric transport of volatile precursors of PFASs and oceanic transport via the currents and movements of water containing PFASs from sewage releasing and surface runoff. The direct oceanic transport has been suggested to be significant in a comparison to atmospheric transport of precursor PFASs [22]. Interestingly, the PFASs in sea water can be further re-distributed from the water to the atmospheric air under certain weather condition via volatilization as a result of sea aerosol and co-evaporation [23] and sea aerosol depended on size fractions can contribute different PFASs burden to air [24] and further to the land surface through rainfall deposition [19]. The trace of airborne PFASs found in waters collected in 2010 from pristine region in Tibetan Plateau has been suggested to be originated from both anthropogenic activities in south-eastern and southern Asia, and from the sea aerosol from the Indian Ocean [19]. This result supports these hypotheses. Much of recent studies have focused on the production, use, and environment occurrence of PFASs across Asia and the Asian contributions to the carry of these compounds getting to other regions [3]. In addition, the continuous manufacture and use of PFASs in Asia countries [25], in combination with the atmospheric and oceanic long range transport can be a significant source to other remote areas. It is, therefore, necessary to carry out more studies on PFAS pollution in Asian environment.

Vietnam is a developing country with an increase in chemical contamination due to rapid industrial development and lack of effective control and waste management strategies [26]. In the best of our knowledge, the production and importation volumes of PFASs in Vietnam have not been reported. Additionally, despite the long usage history of PFASs, there are very few studies that have reported PFASs contamination in Vietnam. A study in Vietnamese aquatic environment reported high concentration of PFASs of up to  $360 \text{ ng L}^{-1}$  in a municipal landfill leachate [26]. Another studies showed high PFOS concentration of up to  $14.6 \text{ ng/ml}$  in

human serum from Ha Noi City [27] and high detection frequency (98–100%) of PFOA in surface water from four cities [28] and PFOS, PFOA, PFHxS in human blood from south central [29]. However, no earlier studies have reported PFASs in other environmental matrices including sediment and biota from Vietnam. Here, we carried out a systematic study during 2013–2015 to determine the current status and extent of PFAS concentrations in both abiotic (e.g. water and sediment) and biotic matrices of freshwater ecosystems including major river basins in Vietnam. Occurrence of PFASs in coastal water, tap water and well water samples collected from eight different regions in Vietnam was investigated. Surface water and sediment samples collected from sewage wastewater treatment plant (WWTPs) effluent discharge canals and landfills were analyzed to identify sources of pollution. PFAS accumulation in tissues (liver, soft tissues, whole body, and muscle) of various freshwater aquatic organisms was investigated.

## 2. Materials and methods

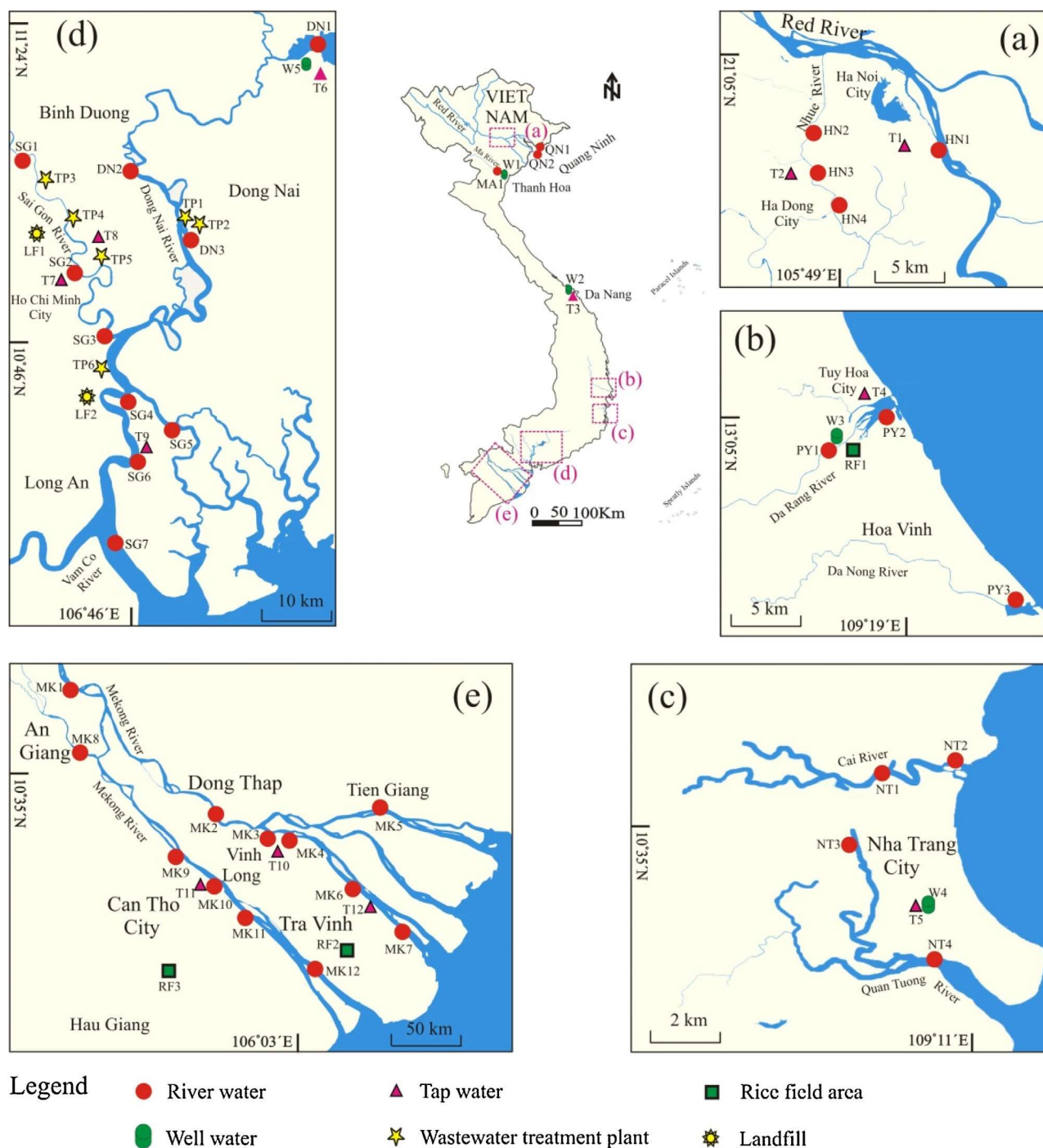
### 2.1. Chemicals and reagents

MPFAC-MXA, a mixture of 9 surrogate standards containing  $^{13}\text{C}_4$ -PFOS (sodium perfluoro-1-[1,2,3,4- $^{13}\text{C}_4$ ]octane sulfonate), and  $^{13}\text{C}_4$ -PFOA (perfluoro-n-[1,2,3,4- $^{13}\text{C}_4$ ]octanoic acid) were purchased with PFAC-MXB, a mixture of 17 native perfluorocarboxylate acids (PFCAs) and perfluoroalkyl sulfonic acids (PFASs) from Wellington Laboratories (Guelph, ON, Canada).  $^{13}\text{C}_4$ -PFOS was used as a surrogate for the analysis of PFCAs and  $^{13}\text{C}_4$ -PFOA was used as a surrogate for the analysis of PFASs. PFAC-MXB mixture was used for standard calibration at concentrations ranging from 0.1 to 50 ng/ml. High performance liquid chromatography (HPLC) grade reagents used include acetonitrile (J.T Baker, Center Valley, PA, USA), Milli Q water (J.T Baker) and ammonium formate (Fisher Scientific, Fair Lawn, NJ, USA).

### 2.2. Sample collection

Water ( $n = 64$ ), sediment ( $n = 16$ ), and biota ( $n = 149$  individuals) were collected during August 2013–February 2015. Details with regard to the sampling location and the types of environmental samples collected are shown in Table S1 of Supplementary material. Thirty-four river water sampling sites are in the Red, Dong Nai, Sai Gon, Mekong, Ma, Da Rang, Da Nong, Cai and Quan Truong Rivers (Fig. 1). The river basins involved in this study accounted for over 81% of the total surface water resources of the country. The Red, Dong Nai and Mekong Rivers are the three largest river basins in Vietnam and play an important role as a water resource for agriculture, industrial, recreational and drinking water resource for millions of people living in the metropolitan cities of Ha Noi, Ho Chi Minh and Can Tho. The sampling locations in rivers were divided generally into three groups as, highly industrialized areas (the Dong Nai, Sai Gon and Red Rivers), moderately industrialized areas (the Mekong River) and less industrialized areas (the Ma, Da Rang, Da Nong, Cai and Quan Truong Rivers). Because the sampling sites were selected to represent the entire country of Vietnam, and involved various levels of industrialization, the results of this study is expected to represent PFAS concentrations in freshwater ecosystems in Vietnam.

Five water samples were collected from two coastal sites in Quang Ninh Province and three irrigation canals of rice fields in provinces of Phu Yen, Tra Vinh and Hau Giang. Eight water samples from canals that receive discharges from five industrial waste water treatment plants (I-WWTPs), one domestic waste water treatment plant (D-WWTP), and two landfill sites located in industrialized and urbanized regions of Ho Chi Minh City, Dong Nai and Binh



**Fig. 1.** Sampling sites of waters in Vietnam from August 2013 to February 2015. (a) Red River Basin, (b) Da Rang and Da Nong Rivers Basins, (c) Cai and Quan Truong Rivers Basins, (d) Mekong River Basin, (e) Dong Nai and Sai Gon River Basins.

Duong Provinces were collected. The treatment capacity of WWTPs ranges from 2000 m<sup>3</sup>/day to 17,500 m<sup>3</sup>/day for individual WWTP and is totally estimated as 52,900 m<sup>3</sup>/day. Overview of the WWTP discharge sites investigated, with potential PFASs sources in their catchment is shown in Table S2. Twelve tap water samples from Ha Noi, Da Nang, Phu Yen, Khanh Hoa, Dong Nai, Ho Chi Minh, Vinh Long, Can Tho, and Tra Vinh and five well water samples from Da Nang, Thanh Hoa, Phu Yen, Khanh Hoa and Dong Nai were collected. All tap water samples collected in this study were treated. The origins of these samples are groundwater (sites T1–2 in Ha Noi City and T4 in Phu Yen Province) and surface water of rivers, natural or artificial lakes (sites T3, T6–T12 in other sampling areas). Sedi-

ment and aquatic organisms were collected from select locations from the rivers listed above, as described in Table S1. In total, forty-seven surface water samples, twelve tap water samples, five well water samples, sixteen sediments and nine aquatic organisms were collected and analyzed.

Duplicate water samples of 500 ml were collected in two clean, methanol rinsed polyethylene terephthalate (PET) bottles that were rinsed three times with waters at the respective sampling sites. Surface layers (top 1–5 cm) of sediment samples were collected using a clean, methanol rinsed polypropylene (PP) spatula and stored in clean polypropylene bags. Five fish species including striped snakehead (*Chana striata*), dusky sleeper (*Eleotris fusca*),

shark catfish (*Pangasius elongatus*), flying barb (*Esomus danricus*), tilapia (*Oreochromis niloticus*) were collected by drift gill net, cast net, or electric shocking. Giant prawn (*Macrobrachium rosenbergii*) samples were collected by trap net or electric shocking. Freshwater paddle crab (*Varuna litterata*) samples were collected by hoop net. Gastropod (golden applesnail: *Pomacea canaliculata*) and bivalve (golden freshwater clam: *Corbicula fluminea*) were collected by hand. Liver tissues were obtained from tilapia and stripped snakehead. Water and sediment samples were transported on ice, to the laboratory, and kept at 4 °C until extraction. Biota samples were stored in dry ice immediately after collection in the field and kept at –20 °C in the laboratory until extraction.

### 2.3. Sample extraction and analysis

Thirteen PFASs consisting of nine perfluoroalkyl carboxylic acids (PFCAs) including perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTTrDA), perfluorotetradecanoic acid (PFTeDA), and four perfluoroalkyl sulfonic acids (PFSAs) including perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS), PFOS, perfluorodecane sulfonic acid (PFDS) were the target analytes in the present study.

Water samples were not filtered to avoid the loss of some PFASs and potential for contamination of target PFASs from filter papers. Water samples were analyzed following the solid phase extraction method described by Yamashita et al. [30]. Sediment and biota samples were analyzed by ion-pairing extraction method described elsewhere [5,31]. The instrumental analysis and ion monitored for the target chemicals are given in Tables S3 and S4.

### 2.4. Quality assurance and quality control

Procedural blanks were prepared to check for possible contamination arising from the analytical procedure. Concentrations of target analytes were subtracted from concentrations found in blanks, when applicable. The target PFASs were not detected in Milli-Q water, PET bottle and vial septum used in this study. One analytical blank (methanol) and one control sample (spiked with known concentrations of standards) were analyzed for every 10 samples to check for potential contamination, accuracy and precision of the analysis. The regression coefficients ( $r$ ) of the calibration curves for all target analytes, prepared at concentration from 0.1 to 50 ng/ml were  $\geq 0.995$ . The limit of detection (LOD) and limit of quantitation (LOQ) values of the target PFASs were evaluated based on the sample volume and signal-to-noise ratio of 3 and 10, respectively. The LOQ values ranged from 0.03 – 0.52 ng L<sup>-1</sup> for water, 0.03–0.48 ng g<sup>-1</sup> dry weight (dw) for sediment, and 0.03–0.48 ng g<sup>-1</sup> wet weight (ww) for biota. The average recovery rates (%) of surrogate standards, <sup>13</sup>C<sub>4</sub>-PFOA and <sup>13</sup>C<sub>4</sub>-PFOS, spiked into each sample prior to extraction were 95.3% and 95.2% for water, 104% and 84.6% for sediment, and 83.4% and 70.7% for biota, respectively.

Fortified water samples (n=3) were made by spiking the native compounds in Milli-Q water to evaluate the recoveries. Mean recoveries of target compounds in fortified water samples were 114 ± 5% for PFHxA, 134 ± 4% for PFHpA, 148 ± 5% for PFOA, 176 ± 8% for PFNA, 143 ± 5% for PFDA, 131 ± 4% for PFUnDA, 95 ± 6% for PFDoDA, 103 ± 2% for PFTTrDA, 58 ± 3% for PFTeDA, 123 ± 3% for PFBS, 131 ± 5% for PFHxS, 112 ± 6% for PFOS, and 102 ± 0% for PFDS. In addition, three standard reference materials (SRM) of Lake Michigan fish tissue (SRM 1947, National Institute of Standards and Technology, Bethesda, MD, USA) were extracted and analyzed. The

recoveries of target PFASs in these SRM samples were 96.8–113% for PFOS and 107–123% for PFDA.

### 2.5. Statistical analysis

Statistical analysis was performed by SPSS® (IBM, version 16). The Pearson correlation test was used to find correlations between the target PFAS concentrations. One-way analysis of variance (ANOVA) using Fisher's Least Square Difference (LSD) post hoc was used to examine the difference of PFASs in biological groups. The level of significance was set to  $p \leq 0.05$ . Non-detectable data (ND) and data below LOQs were assigned a value of zero for statistical analysis.

## 3. Results and discussion

The detecting frequency of all target analytes in water, sediment and fish matrices is presented in Table S5. The relative composition (%) of PFASs in water, sediment and biota is shown in Fig. 2.

### 3.1. PFASs in water

Except for the Sai Gon River, the total PFASs concentrations in water samples collected from the Red, Ma, Da Rang, Da Nong, Cai, Quan Truong, Dong Nai and Mekong Rivers were below 5 ng L<sup>-1</sup> (Table 1). These results were quite consistent with those previously reported for Vietnamese rivers in four cities (<4 ng L<sup>-1</sup> in general and up to 17 ng L<sup>-1</sup>) [28]. Excluding the water samples collected from discharge canals of WWTPs, the surface water in general contained relatively low concentration of PFASs. The mean concentrations of PFASs in river and coastal waters were less than 0.75 ng L<sup>-1</sup> for individual PFAS and 2 ng L<sup>-1</sup> for total PFASs. PFCAs were found at greater concentrations than PFSAs by a factor of 1.73–61.2 (mean PFOA/PFSA ratio = 9.37). PFSAs have been reported as the predominant PFASs in surface water sample collected near several industrial and coastal areas in Korea [32]. The concentrations of PFCAs in water collected from WWTP discharge canals and landfill sites were 1.7 ~ 2.8 fold greater than that of PFSAs. In addition, the significant positive correlations between PFHxA, PFHpA, PFOA, PFNA, PFDA and PFUnDA (Table 2) suggest the wide application range of PFCAs or/and sources arising from their potential precursor compounds such as fluorotelomer alcohols (FTOHs) in commercial products in Vietnam.

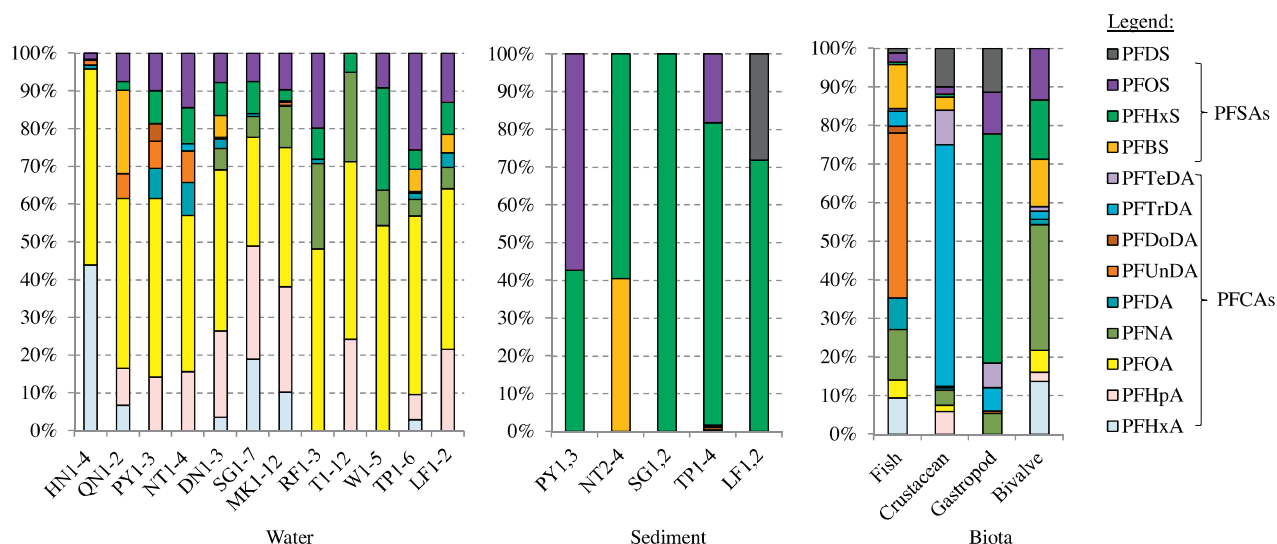
PFOA (detection frequency = 81%, mean = 2.83 ng L<sup>-1</sup>) and PFOS (detection frequency = 51%, mean = 1.32 ng L<sup>-1</sup>) were consistently the dominant PFCAs and PFSAs in surface water samples analyzed. The mean percentages of PFOA and PFOS concentrations in total PFASs concentration were 43% and 11%, respectively (Fig. 2). The greatest concentrations of PFOA (53.5 ng L<sup>-1</sup>) and PFOS (40.2 ng L<sup>-1</sup>) were found in water from the sampling site TP4, which is a discharge canal of an I-WWTP. The ratio of concentration of PFOS to PFOA was in the range of 0.04–1.56 (mean = 0.36). Apart from the sampling site MK4 and TP6, where these ratios are greater than 1, the PFOS/PFOA concentration ratio in surface water from other sampling sites was less than 1. This result suggests that PFOA was the dominant compound in surface water samples in Vietnam.

The greatest concentration of total PFAS (14.6 ng L<sup>-1</sup>) was found at a highly urbanized and industrialized site (SG2) in the Sai Gon River, Ho Chi Minh City. This result might be attributed to the site-specific loads of PFASs related to potential PFASs in its catchment. The catchment of site SG2 is mostly made up of two nearby upstream tributaries named Vam Thuat River and Rach Dua Canal. Several potential sources of PFASs originated from these two upstream tributaries are addressed as sewage water from Tan Son Nhat airport, a population of over one million residents from six districts and towns, and WWTP discharges from five industrial

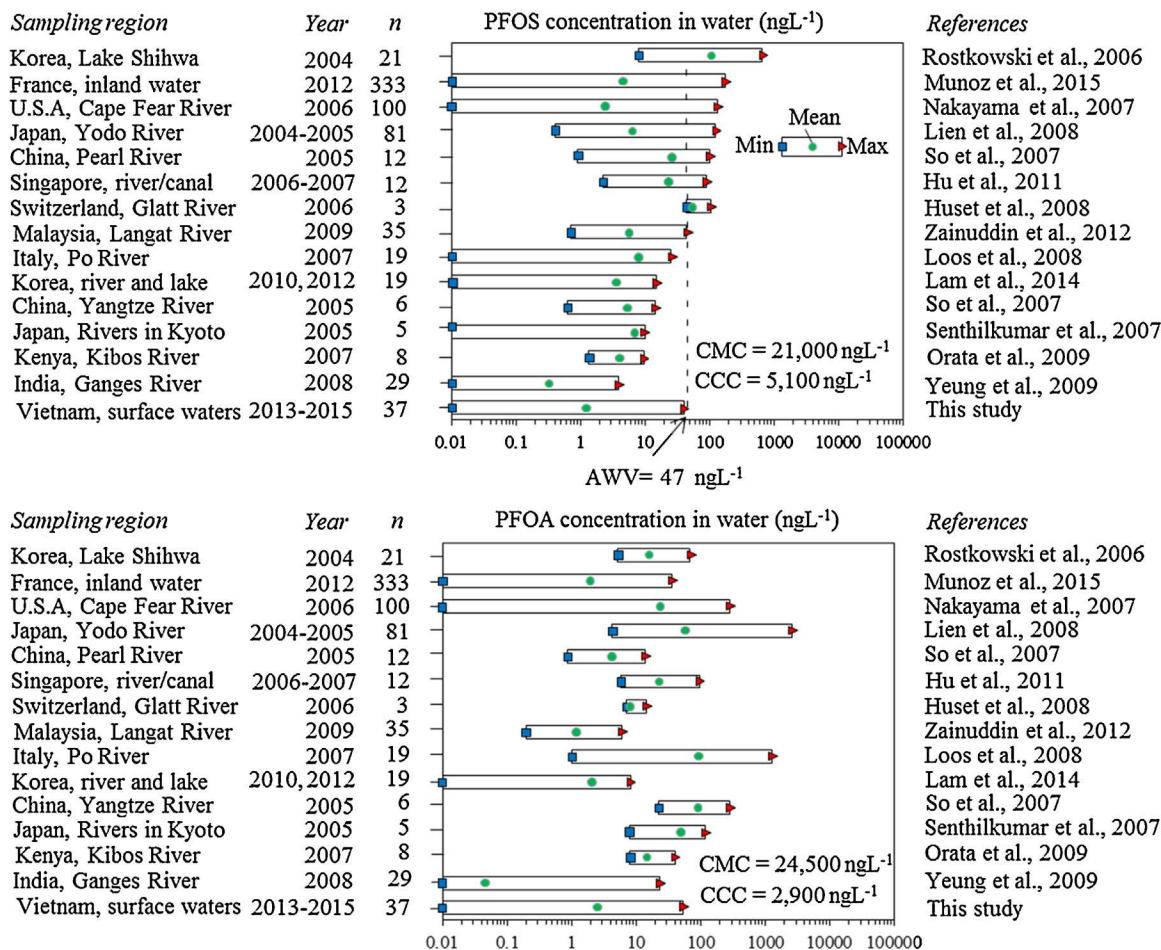
**Table 1**  
Concentrations (ng L<sup>-1</sup>) of perfluorinated alkyl substances (PFASs) in surface, tap and ground waters from Vietnam showing min–max (mean)\*

Sample type	Site (n)	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTtDA	PFTeDA	PFBS	PFHKS	PFOS	PFDS	∑PFASs
Surface water	HN1-4 (4)	<0.52–1.09 (0.27)	<0.45	0.13–0.40 (0.32)	<0.06	<0.03–0.03 (0.01)	<0.03–0.03 (0.01)	<0.04	<0.03–0.03 (0.00)	<0.06	<0.30	<0.07	<0.03–0.04 (0.01)	<0.11	0.23–1.48 (0.62)
	QN1-2 (2)	<0.07–0.07 (0.04)	0.05–0.06 (0.05)	0.24–0.26 (0.25)	<0.06	<0.03 (0.04)	0.03–0.04 (0.04)	<0.03	<0.03	<0.06	0.10–0.15 (0.12)	<0.03–0.03 (0.01)	0.04–0.05 (0.04)	<0.11	0.53–0.58 (0.55)
	MA1 (1)	<0.52	<0.45	<0.37	<0.06	<0.03	<0.06	<0.04	<0.05	<0.06	<0.30	<0.07	<0.03	<0.11	0.00
	PY1-3 (3)	<0.52	<0.05–0.07 (0.02)	<0.13–0.22 (0.07)	<0.06	<0.03–0.04 (0.01)	<0.03–0.03 (0.01)	<0.03–0.03 (0.01)	<0.03–0.03 (0.01)	<0.06	<0.30	<0.03–0.04 (0.01)	<0.03–0.05 (0.02)	<0.11	0.00–0.46 (0.15)
	NT1-4 (4)	<0.52	<0.05–0.06 (0.01)	<0.13–0.16 (0.04)	<0.06	<0.03–0.03 (0.01)	<0.03–0.03 (0.01)	<0.04	<0.03–0.03 (0.00)	<0.06	<0.30	<0.03–0.04 (0.01)	<0.03–0.05 (0.01)	<0.11	0.00–0.38 (0.09)
	DN1-3 (3)	<0.52–0.96 (0.32)	<0.45–1.53 (0.51)	<0.37–1.06 (0.49)	<0.06–0.22 (0.09)	<0.03–0.04 (0.01)	<0.03–0.04 (0.01)	<0.04	<0.04	<0.06	<0.30	<0.07–0.25 (0.14)	<0.03–0.20 (0.13)	<0.11	0.00–4.17 (1.70)
	SG1-7 (7)	<0.52–1.34 (0.19)	<0.45–3.10 (1.22)	0.92–6.39 (2.28)	<0.06–1.04 (0.30)	<0.03–0.67 (0.13)	<0.03–0.03 (0.03)	<0.04	<0.03–0.03 (0.00)	<0.06	<0.30–1.35 (0.31)	<0.07–1.69 (0.47)	<0.03–1.62 (0.41)	<0.11	1.71–14.6 (5.35)
	MK1-12 (12)	<0.52–0.92 (0.14)	<0.45–1.49 (0.38)	<0.37–0.85 (0.50)	<0.06–0.23 (0.15)	<0.03–0.03 (0.00)	<0.06–0.14 (0.01)	<0.04	<0.05–0.06 (0.00)	<0.06	<0.30	<0.07–0.17 (0.04)	<0.03–1.06 (0.13)	<0.11	0.00–3.17 (1.36)
	RF1-3 (3)	<0.52	<0.45	<0.37–1.06 (0.63)	<0.06–0.51 (0.30)	<0.03–0.05 (0.02)	<0.06	<0.04	<0.05	<0.06	<0.30	<0.07–0.19 (0.11)	<0.03–0.41 (0.26)	<0.11	0.00–1.99 (1.31)
	TP1-6 (6)	<0.52–4.26 (1.07)	<0.45–7.81 (2.38)	0.84–53.5 (17.1)	0.22–4.81 (1.58)	0.04–1.37 (0.61)	<0.06–0.33 (0.13)	<0.04	<0.05	<0.06	<0.30–8.28 (2.14)	<0.07–5.98 (1.84)	<0.03–40.2 (9.26)	<0.11	1.30–107 (36.2)
	LF1-2 (2)	<0.52	0.59–0.82 (0.70)	1.38–1.39 (1.39)	0.15–0.22 (0.19)	0.09–0.16 (0.12)	<0.06	<0.04	<0.05	<0.06	<0.30–0.32 (0.16)	0.10–0.45 (0.28)	0.26–0.59 (0.42)	<0.11	3.19–3.33 (3.26)
	Tap water	T1-12 (12)	<0.52	<0.45–0.90 (0.07)	<0.37–0.48 (0.14)	<0.06–0.22 (0.07)	<0.03	<0.06	<0.04	<0.05	<0.06	<0.30	<0.07–0.11 (0.02)	<0.03	<0.11
Well water	W1-5 (5)	<0.52	<0.45	<0.37–5.48 (1.93)	<0.06–1.10 (0.33)	<0.03	<0.06	<0.04	<0.05	<0.06	<0.30	<0.07–4.35 (0.96)	<0.03–1.42 (0.32)	<0.11	0.00–8.88 (3.55)

\* There were differences between LOQ values for target PFASs in water analyzed in 2013 and 2015; n: number of sample analyzed. ∑PFASs refer to sum of thirteen PFASs in each sampling site; HN 1-4: Red River basin, QN1-2: Quang Ninh coastal, MA1: Ma River, PY1-3: Da Rang and Da Nong Rivers, NT1-4: Cai and Quan Truong Rivers, DN1-3: Dong Nai River, SG1-7: Sai Gon River basin, MK1-12: Mekong River, RF1-3: rice field areas, TP1-6: wastewater treatment plant discharge canals, LF1-2: landfill discharge canals, T1-12: tap water, W1-5: well water.



**Fig. 2.** Relative compositions (%) of PFAS in water, sediment and biota. HN 1-4: Red River basin, QN1-2: Quang Ninh coastal, MA1: Ma River, PY1-3: Da Rang and Da Nong Rivers, NT1-4: Cai and Quan Truong Rivers, DN1-3: Dong Nai River, SG1-7: Sai Gon River basin, MK1-12: Mekong River, RF1-3: rice field areas, TP1-6: wastewater treatment plant discharge canals, LF1-2: landfill discharge canals, T1-12: tap water, W1-5: well water.



**Fig. 3.** Concentration of PFOS and PFOA in surface waters from Vietnam and several sampling regions. AWW: Avian Wildlife Value; CCC: Criteria Continuous Concentration; CMC: Criteria Maximum Concentration.

parks in Ho Chi Minh City and Binh Duong Province. The main relevant industrial activities in the catchment of these tributaries are considered as metal industry, textile industry, paper industry and

chemical factory. Moreover, the dilution factor of wastewater from the WWTPs discharge of site TP4 (PFASs concentration = 107 ng/L) and TP5 (PFASs concentration = 26.9 ng/L) may be expected to be

**Table 2**

Pearson correlation coefficients of individual PFASs in water and biota collected from aquatic environment in Vietnam.

Biota/Water	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTTrDA	PFBS	PFHxS	PFOS
PFHxA		0.83**	0.46**	0.73**	0.73**	0.78**	−0.04	0.12	0.19	0.73**	0.18
PFHpA	−0.01		0.59**	0.84**	0.85**	0.79**	−0.05	0.08	0.25*	0.74**	0.33**
PFOA	0.24**	−0.02		0.85**	0.76**	0.66**	−0.03	−0.04	0.78**	0.50**	0.94**
PFNA	−0.02	−0.02	0.48**		0.86**	0.76**	−0.05	−0.03	0.43**	0.73**	0.64**
PFDA	0.02	−0.03	0.51**	0.76**		0.91**	−0.2	−0.04	0.55**	0.78**	0.55**
PFUnDA	0.08	−0.02	0.47**	0.69**	0.83**		0.02	0.27*	0.51**	0.75**	0.44**
PFDoDA	−0.02	−0.01	−0.02	0.19*	0.47**	0.55**		−0.02	−0.03	−0.04	−0.02
PFTTrDA	0.02	0.02	−0.08	0.04	0.28**	0.38**	0.73**		−0.04	−0.05	−0.03
PFBS	0.26**	−0.02	0.51**	0.71**	0.49**	0.42**	−0.03	−0.02		0.22	0.84**
PFHxS	−0.02	−0.02	−0.09	−0.01	−0.09	−0.07	−0.03	−0.15	−0.08		0.21
PFOS	−0.03	−0.02	−0.05	0.12	−0.42	−0.02	−0.03	−0.09	0	0.03	

\*:  $p < 0.05$  and \*\*:  $p < 0.01$ .**Table 3**Concentration ( $\text{ng g}^{-1}$  dry weight) of PFASs in sediment from selected sampling sites in Vietnam.

Site	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	$\sum$ PFASs
PY1	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
PY3	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	0.13	0.17	<0.30	0.3
NT2	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
NT3	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
NT4	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	0.57	0.84	<0.08	<0.30	1.41
DN2	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
DN3	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
SG1	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
SG2	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	1.86	<0.08	<0.30	1.86
MK3	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0
TP1	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	<0.04	0.18	<0.30	0.18
TP2	<0.16	<0.23	<0.20	<0.10	0.17	<0.06	<0.05	<0.03	<0.13	<0.34	16.4	1.18	<0.30	17.8
TP3	<0.16	<0.23	<0.20	<0.10	<0.06	0.24	<0.05	<0.03	<0.13	<0.34	<0.04	<0.08	<0.30	0.24
TP4	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	0.16	0.16	<0.13	<0.34	18.3	6.72	<0.30	23.4
LF1	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	0.46	<0.08	0.85	1.31
LF2	<0.16	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	1.75	<0.08	<0.30	1.75

$\sum$  PFASs refer to sum of thirteen  $\sum$  PFASs in each sampling site; PY1, 3: Da Rang and Da Nong Rivers, NT2–4: Cai and Quan Truong Rivers, DN2–3: Dong Nai River, SG1–2: Sai Gon River basin, MK3: Mekong River, TP1–4: wastewater treatment plant discharge canals, LF1–2: landfill discharge canals.

not significant since these wastewater discharge mostly flow down to their joining point of Rach Dua Canal and Sai Gon River without receiving other runoff surface waters confirming again the site-specific contaminant of PFASs in the catchment of site SG2. PFAS concentrations in estuarine or downstream riverine sites including PY2, PY3, NT4, SG7, MK7 and MK12 were relatively less than those in the upstream sampling regions of corresponding rivers. These results suggest that major sources of PFAs in surface water in Vietnam are region-specific industrial and/or domestic waste discharges from widely dispersive use of the substances in end-user products. The dominance of PFOA concentrations in water from Red River (mean percentage = 52%) and Mekong River (mean percentage = 37%) is consistent with those in snow or water from the upstream catchment of these rivers in Tibetan Plateau [33] and Yunnan province, China [34]. Interestingly, PFASs pollution profiles of environmental waters in Vietnam have been reported to close to those in China suggesting the potential source of PFASs in the environment were quite similar to those in China. Several imported products containing PFASs from China are considered as one of the major sources of PFASs in the Vietnamese aquatic environment [28].

The greatest concentration of total PFASs ( $107 \text{ ng L}^{-1}$ ) in surface water was found in a samples from WWTP discharge canal. PFOA (mean =  $17.1 \text{ ng L}^{-1}$ , 47% of total PFASs) and PFOS (mean =  $9.26 \text{ ng L}^{-1}$ , 26% of total PFASs) were found to be the dominant compounds in WWTPs discharges and this pattern was in good agreement with PFOA and PFOS contamination profiles found in water collected from rivers. The ratios of PFAS concentrations in WWTPs discharges to those in river water samples from the Dong Nai and Sai Gon Rivers were 10 for PFOA, 28 for PFOS and

9 for PFASs. Earlier studies [35,36] reported that WWTPs are “point sources” for PFASs in surface waters and concentrations in WWTP effluents were approximately 5–10 times higher than those in river water samples. Although potential industrial activities in catchments with elevated PFAS levels were identified (Table S2), we are unable to get information of the connected population in the catchment of the investigated WWTPs and the mean discharge rate of the WWTPs in the sampling period to estimate the PFASs mass load assessment and per-capita mass load of PFASs.

Relatively low concentrations of PFOA (mean =  $1.39 \text{ ng L}^{-1}$ ), PFOS (mean =  $0.42 \text{ ng L}^{-1}$ ) and total PFASs (mean =  $3.26 \text{ ng L}^{-1}$ ) were found in water samples collected from discharge canals of landfill sites. PFASs concentrations in water samples ( $n = 10$ ) collected from effluent and surrounding canals of a municipal landfill site contained  $1.2 \text{ ng L}^{-1}$  PFOA,  $0.28 \text{ ng L}^{-1}$  PFOS, and  $8.4 \text{ ng L}^{-1}$  total PFASs [26]. Despite the relatively low concentration of PFASs found in effluent canal from landfill, the total PFASs concentration ( $360 \text{ ng L}^{-1}$ ) in the municipal landfill leachate was approximately 43 fold greater than that found in effluent canal [26]. Landfill leachate from 22 landfill sites in Germany contained total PFASs concentration of up to  $12,819 \text{ ng L}^{-1}$  [37].

The mean PFOA concentration in surface water was relatively higher than that previously reported in four cities in Vietnam [28], Ganges River in India [38], Langat River in Malaysia [39] and comparable with that in main river and lake in Korea [40], while being 4–6 fold lower than that in Kibos River in Kenya [41], river and canal in Singapore [42], Shihwa Lake in Korea [43], Cape Fear River in USA [44] and inland water in France [45], and over 20 fold lower than that in Kyoto River, Yodo River Basin in Japan [46,47], Yangtze River in China [48], Po River in Italy [49] and Glatt River in Switzerland

[50] (Fig. 3). Similarly, relative low mean PFOS concentration in surface water in this study was found which is approximately 5 fold lower than that in Malaysia [39], Japan [47], Italy [49] and 15–67 fold lower than that in Switzerland [50], Singapore [42], China [48], USA [44] and Korea [43]. The results may be attributed to the smaller using amounts of PFOA and/or PFOS in Vietnam than these countries, and suggest that the investigating areas in this study are exposed to lower PFOA and/or PFOS contamination sources. However, it is noted that the sources of PFASs in Vietnamese water environment still remain to be examined since there is very few information on the used, production, and volumes of PFAS in Vietnam [28].

PFOA (mean = 0.14 ng L<sup>-1</sup>), PFHpA (mean = 0.07 ng L<sup>-1</sup>), PFNA (mean = 0.07 ng L<sup>-1</sup>) and PFHxS (mean = 0.02 ng L<sup>-1</sup>) were found in tap water samples. PFOA was the dominant PFAS in tap water samples. The PFOA concentration (0.42 ~ 0.48 ng L<sup>-1</sup>) in three tap water samples collected in Ho Chi Minh City was slightly higher than that determined in river water (0.40 ng L<sup>-1</sup>) collected in site DN2 which is a pump station for drinking water source to Ho Chi Minh City. This result suggests that the water treatment system is not effective in removing PFOA [51]. While PFOS was not detected in any of tap water samples in this study, the substance was previously found in tap water from Osaka, Japan (0.16–22 ng L<sup>-1</sup>) [10], Spain (0.39–0.87 ng L<sup>-1</sup>) [52], France (<4–16 ng L<sup>-1</sup>) [53] and accounting for over than 50% of the total PFASs in tap water from Shenzhen (11 ng L<sup>-1</sup>), Macau (6.2 ng L<sup>-1</sup>), Taipei (5.4 ng L<sup>-1</sup>), and Hongkong (7.0 ng L<sup>-1</sup>), China [51]. Similar to those in surface water, the most abundance of PFOA concentrations in tap water was determined. However, PFOA was only detected in four tap water samples (T6 – T9) which reflect their origins of the highly urbanized and industrialized in Dong Nai and Sai Gon River Basins. This result suggests the site-specific of PFASs concentrations in tap water depend on differences on the PFASs contamination in the raw water and distributing system. It's worth to note that although differences PFASs composition profiles were determined in tap water from China, PFOA (mean = 10 ng L<sup>-1</sup>) was reported as the most prevalent PFASs and accounted for over 40% of the total PFASs in tap water from Nanjing (5.9 ng L<sup>-1</sup>), Shanghai (78 ng L<sup>-1</sup>), Beijing (0.44 ng L<sup>-1</sup>) and Shenyang (2.6 ng L<sup>-1</sup>) [51]. In tap water in this study, mean PFOA concentration was 70-fold lower than that reported for China [51] and highest PFOA concentration (0.48 ng L<sup>-1</sup>) was 14–1,081 fold lower than that in developed countries reported earlier including Japan (84 ng L<sup>-1</sup>) [10], Spain (6.8 ng L<sup>-1</sup>) [52], France (9 ng L<sup>-1</sup>) [53] and Germany (519 ng L<sup>-1</sup>) [54]. In the other hand, relatively lower PFOA concentration in tap water than that in this study was previously reported in the remote areas of Urumuqui and Atushi, China (<0.1 ng L<sup>-1</sup>) [55]. Unfortunately, we could not get information on daily consumption rate of tap water for drinking and cooking in Vietnamese adults and daily PFAS intake of Vietnamese adults through tap water was not estimated.

PFOA, PFNA, PFOS and PFHxS were detected in well water samples. The greatest PFAS concentration was found at 5.48 ng L<sup>-1</sup> for PFOA. This value is slightly greater than that reported for 22 ground water samples in Hanoi and Ho Chi Minh Cities [28]. Since only one well water sample was collected from each of the sampling region in the present study, a more extensive sampling is necessary for further discussion. Well water and tap water are drinking water resources in rural and urban regions in Vietnam. To evaluate the human health risk of exposure to PFOA and PFOS via drinking well water and tap water in the present study, the measured concentrations of these chemicals were compared with provisional health advisory level for PFOA (400 ng L<sup>-1</sup>) and PFOS (200 ng L<sup>-1</sup>) issued by the [56]. The concentration of these PFASs in tap water and well water in the present study were 141–978 fold less than the provisional health advisory level. This suggests that PFOS and PFOA in

the tap water and well water may not pose an immediate health risk for drinking water consumers in Vietnam.

The measured concentrations of PFOS and PFOA in surface water were less than the criteria for aquatic protection including Avian Wildlife Value (AWV), Criteria Continuous Concentration (CCC) and Criteria Maximum Concentration (CMC) previously reported [57]. These criteria values were established based on the U.S. EPA's Great Lakes Initiative [58]. This result suggests that the chronic and acute effects on aquatic organisms exposed to PFOS and PFOA in surface waters in Vietnam were not likely. However, a criterion that was calculated as 5 fold the mean plus one standard deviation of the log-normal distribution was suggested [59]. Accordingly, the measured concentrations for 3 PFOA values and 2 PFOS values from sites TP2, TP4 and TP5 among 47 samples exceeded the interim Vietnam water quality criteria (IVWQC) for PFOA (9.32 ng L<sup>-1</sup>) and PFOS (2.96 ng L<sup>-1</sup>). The suggested IVWQC values for PFOA and PFOS in the present study were 16–7094 fold less than those of reported for AWV, CCC and CMC [57]. The suggested IVWQC values will provide a baseline for evaluating the status and trends of PFASs in surface water in Vietnam.

### 3.2. PFASs in sediment

Approximately 1/5 of the sediment samples analyzed in this study contained PFASs above the detection limits (Table 3). PFDA, PFUnDA, PFTrDA, PFBS and PFDS were found in only one sediment sample at concentrations that ranged from 0.16 to 0.85 ng g<sup>-1</sup> dw. The concentrations of PFHxA, PFHpA, PFOA, PFNA, PFUnDA, and PFTeDA were not found in any sediment samples. PFHxS (mean = 2.48 ng g<sup>-1</sup> dw) and PFOS (mean = 0.52 ng g<sup>-1</sup> dw) were the two predominant PFASs in sediment samples, which accounted for 71% and 15% of the total PFAS concentration in sediments (Fig. 2). The predominant PFAS concentrations in sediment were found at 18.3 and 16.4 ng PFHxS g<sup>-1</sup> dw in sites TP2 and TP4 located in discharge canals of I-WWTPs in Dong Nai and Binh Duong Provinces, respectively. Similar to that found for sediment at site TP2, the greatest concentration of PFHxS (5.98 ng L<sup>-1</sup>) was found in water at the same site, which suggests a potential source for this compound. The partition coefficient of PFHxS (K<sub>d</sub>) estimated by the ratio of the concentration of PFHxS in sediment (ng g<sup>-1</sup> dw) to that in the overlying water (ng L<sup>-1</sup>) at the same sampling site ranged from 2–42. This K<sub>d</sub> value is 200–4200 fold greater than that reported for sampling sites in Korea [40] possibly an influence of difference in organic matter.

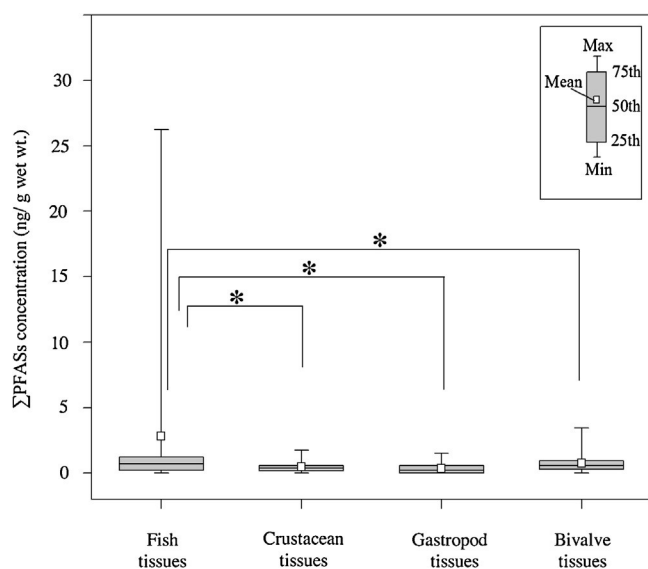
PFOS concentrations (<0.08–0.17 ng g<sup>-1</sup> dw) in river were comparable, or slightly lower than those previously reported in tidal flat areas in Ariake Sea of Japan (0.09–0.14 ng g<sup>-1</sup> dw) [2], Taihu Lake (0.04–0.48 ng g<sup>-1</sup> dw) and Liao River (0.06–0.31 ng g<sup>-1</sup> dw) in China [60]. Over 5 and 10 fold greater of PFOS concentrations than those in this study were also found in Tokyo Bay, Japan (0.13–1.40 ng g<sup>-1</sup> dw) [61], four rivers in San Francisco Bay, USA (<LOD–1.30 ng g<sup>-1</sup> dw) [62] and Korean western coast (>2 ng g<sup>-1</sup> dw) [43]. In the best of our knowledge, the highest PFOS concentration (mean = 536.7 ng g<sup>-1</sup> dw) in a river sediment sample which was found to be over 3000 fold greater than that in our study was reported for Baozhen Port, Yangtze River, China [63] due to significant amount of domestic sewage from the nearby town. Particularly, PFOS concentration in sediment in Mekong River in this study was significant lower than that of polychlorinated biphenyls (PCBs; 0.039–9.2 ng/g dry wt), dichlorodiphenyl-trichloroethane (DDT; <0.01–110 ng g<sup>-1</sup> dw), chlordanes compounds (CHL; <0.004–19 ng g<sup>-1</sup> dw), and hexachlorocyclohexane isomers (HCH; 0.02–1.3 ng g<sup>-1</sup> dw) reported earlier [64]. This was might be attributed to the high water solubility of PFOS compared with other nonpolar organic contaminants [2]. In short, the little amount of PFOS in river sediment in



**Table 4**  
Mean concentration ( $\text{ng g}^{-1}$  ww) of PFASs determined in biological samples collected from Vietnam.

Site	Species	Samples (n)	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	$\sum$ PFASs
PY1	Tilapia	Liver (10)	0.92	<0.23	0.4	1.51	0.84	4.79	0.16	0.37	<0.13	1.34	<0.04	0.16	0.14	10.6
		Muscle (10)	<0.48	<0.23	<0.20	0.04	0.05	0.31	0.03	0.02	<0.13	<0.34	0.05	0.02	<0.30	0.5
	Paddle crab	Soft tissue (15)	<0.48	0.1	<0.20	0.03	<0.06	<0.06	<0.05	0.39	0.09	<0.34	<0.04	<0.08	<0.30	0.61
	Giant prawn	Soft tissue (15)	<0.48	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	0.45	0.02	<0.34	0.01	0.02	0.08	0.58
	Golden apple snail	Soft tissue (6)	<0.48	<0.23	<0.20	0.04	<0.06	<0.06	<0.05	0.08	0.08	<0.34	0.08	0.14	0.18	0.6
DN3	Golden freshwater clam	Soft tissue (30)	0.1	0.02	0.04	0.24	0.01	<0.06	<0.05	0.02	0.01	0.09	0.11	0.1	<0.30	0.73
		Stripped snakehead	Liver (3)	0.88	<0.23	<0.20	<0.10	<0.06	0.03	<0.05	0.06	<0.13	<0.34	<0.04	0.03	<0.30
		Muscle (3)	<0.48	<0.23	<0.20	<0.10	<0.06	0.02	<0.05	<0.03	<0.13	<0.34	<0.04	0.16	<0.30	0.18
MK3	Dusky sleeper	Whole body (3)	<0.48	<0.23	<0.20	0.03	0.07	0.33	0.07	0.13	0.21	<0.34	0.05	0.05	<0.30	0.92
	Giant prawn	Soft tissue (12)	<0.48	<0.23	0.02	<0.10	0.01	<0.06	<0.05	0.14	0.03	<0.34	<0.04	0.02	0.03	0.24
	Golden apple snail	Soft tissue (11)	<0.48	<0.23	<0.20	0.03	<0.06	<0.06	0.01	0.01	0.02	<0.34	0.26	<0.08	<0.30	0.32
	Shark catfish	Whole body (10)	<0.48	<0.23	0.2	<0.10	<0.06	<0.06	<0.05	0.02	<0.13	0.07	0.02	<0.08	<0.30	0.3
	Flying barb	Whole body (5)	<0.48	<0.23	<0.20	0.16	0.25	0.3	<0.05	0.05	<0.13	0.07	<0.04	0.09	<0.30	0.91
Min <sup>(b)</sup>	Giant prawn	Soft tissue (16)	<0.48	<0.23	0.01	0.03	<0.06	<0.06	0.01	0.11	0.02	0.05	<0.04	<0.08	0.06	0.29
	Golden apple snail	Soft tissue (13)	<0.48	<0.23	<0.20	<0.10	<0.06	<0.06	<0.05	<0.03	<0.13	<0.34	0.19	0.02	<0.30	0.22
Max <sup>(b)</sup>			9.2	1.5	1.53	3.67	1.64	16.9	1.43	2.75	0.35	4.17	0.95	1.64	1.43	26.2
Mean <sup>(b)</sup>			0.09	0.01	0.05	0.15	0.07	0.33	0.01	0.14	0.02	0.11	0.06	0.05	0.03	1.13
SD <sup>(b)</sup>			0.76	0.12	0.2	0.49	0.26	1.73	0.11	0.28	0.07	0.54	0.17	0.2	0.17	3.13

n: number of sample analyzed, (a)  $\sum$  PFASs refer to sum of thirteen PFASs in each biota tissues sample; (b) value in whole biota sample.



**Fig. 4.** Total PFASs concentration in tissues of fish, crustacean, gastropod and bivalve collected in Vietnam.

\*:  $p < 0.05$

Vietnam is not likely to take significant part to the possible exposure of benthic and aquatic organisms.

### 3.3. Distribution patterns of PFASs in biota and bioconcentration of PFASs

Concentration and detection frequency profiles of PFASs in biota samples are shown in Table S5 and Table 4. All of thirteen target PFASs were detected in biological species including fish ( $n = 5$  species), crustaceans ( $n = 2$  species), gastropods ( $n = 1$  species) and bivalves ( $n = 1$  species). Fishes tissues samples ( $n = 45$ ) contained the greatest concentration of PFASs, ranging from <LOQ to  $26.2 \text{ ng g}^{-1}$  ww (mean =  $2.81 \text{ ng g}^{-1}$  ww), followed by that of bivalves ( $n = 30$ , <LOQ– $3.45 \text{ ng g}^{-1}$  ww, mean =  $0.73 \text{ ng g}^{-1}$  ww), crustacean ( $n = 58$ , <LOQ– $1.75 \text{ ng g}^{-1}$  ww, mean =  $0.44 \text{ ng g}^{-1}$  ww) and gastropod ( $n = 30$ , <LOQ– $1.50 \text{ ng g}^{-1}$  ww, mean =  $0.33 \text{ ng g}^{-1}$  ww), respectively. Significant differences ( $p < 0.05$ ) between total PFASs concentration in fish tissues with tissues of crustacean, gastropod, bivalve were found (Fig. 4). No significant

positive correlation ( $p > 0.05$ ) between PFASs concentration and total length/body weight was found.

Similar to that found for water, concentrations of PFOA and PFOS in biota were less than those reported in earlier studies for soft tissues of gastropod, bivalve, crustacean from Korea [59], fish liver from Korea [40,43], lakes in China [65,66], rivers and lakes in New York, USA [67], canals in Belgium [68], coastal Indonesia [69] and Japanese coast [2,46,70]. In contrast, PFNA and PFBS concentrations in bivalve soft tissues were greater than those reported from Korea coastal waters [59]. The details on comparison of PFOA and PFOS concentration in biota tissues with other studies are shown in Table S6.

PFAS concentration patterns were different among various biological species analyzed. The dominant PFAS was PFUnDA (accounted for 42% of total PFASs) for fish, PFTTrDA (accounted for 63% of total PFASs) for crustaceans, PFHxS (accounted for 59% of total PFASs) for gastropods, and PFNA (accounted for 33% of total PFASs) for bivalves (Fig. 2). Fish and crustaceans tended to accumulate relatively greater proportions of long-chain PFCAs including PFNA, PFDA, PFUnDA, PFDoDA and PFTTrDA, PFTeDA than other PFASs. However, PFNA and perfluoroalkyl sulfonic acids including PFBS, PFHxS and PFOS were found at relatively greater concentration than other PFASs in gastropods and bivalves. Significant positive correlations between PFNA and PFDA, PFNA and PFBS, PFDoDA and PFTTrDA, PFOS and PFBS were found for all biota ( $p < 0.05$ ) (Table 2).

In order to investigate the accumulation profiles of PFASs in individual tissues and organs in fishes, liver and muscle tissues were collected from tilapia and stripped snakehead, and whole body were analyzed for dusky sleeper, shark catfish and flying barb. The profiles of PFAS concentration in fish varied widely among tissues and species. The ascending order of total PFASs concentration in fish tissues was found for muscle (mean =  $0.43 \text{ ng g}^{-1}$  ww), whole body (mean =  $0.59 \text{ ng g}^{-1}$  ww) and liver (mean =  $8.42 \text{ ng g}^{-1}$  ww). PFUnDA and PFHxS were the predominant PFCAs and PFASs, respectively, in whole body and muscle tissues whereas PFUnDA and PFOS were the PFCAs and PFASs, respectively, in liver. The predominance of PFUnDA and PFOS in fish liver is consistent with those reported previously for fishes in Korea [40]. The concentration ratios of PFASs in tilapia to stripped snakehead were 10 for liver tissues and 3 for muscle tissues. The compound-, species-, and tissues- specific accumulations in fish may be attributed for their natural histories, metabolism, and feeding habit.

**Table 5**  
Mean bioconcentration factors (BCFs) of detected PFASs in biota samples collected from Vietnam.

Species (n)	Fish			Whole body		Crustacean		Gastropod		Bivalve	
	Liver	Muscle	Stripped snakehead (3)	Tilapia (10)	Stripped snakehead (3)	Dusky sleeper (4)	Shark catfish (3)	Flying barb (5)	Soft tissue Paddle crab (15)	Giant prawn (43)	Soft tissue Golden freshwater clam (30)
PFHxA	-	-	914	-	-	-	-	-	-	-	-
PFHpA	-	-	-	-	-	-	-	-	1523	-	268
PFOA	1826	-	-	-	-	-	293	-	-	-	187
PFNA	-	-	-	138	-	1093	-	-	-	64	-
PEDA	22,952	1307	-	1627	-	-	-	-	-	-	256
PFUnDA	142,764	9169	-	-	-	-	-	-	-	-	-
PFDoDA	7729	1393	-	-	-	-	-	-	-	-	-
PFHxS	-	1123	-	282	-	131	-	-	-	-	2781
PFOS	3551	327	894	253	-	-	-	1477	-	1606	2110

n: number of samples analyzed; BCFs of PFTTrDA, PFTeDA, PFBS and PFDS in biota were not determined in this study.

Mean bioconcentration factor (BCF) of PFASs was calculated based on measured PFASs concentration in water and biota (wet mass basis) collected from corresponding locations and results are shown in Table 5. BCFs of PFTTrDA, PFTeDA, PFBS and PFDS in biota were not determined in this study. The greatest mean BCF value was 142,764 (L/kg) for PFUnDA, followed by PFDA (22,953 L/kg), PFDoDA (7729 L/kg), and PFOS (3551 L/kg) in tilapia liver. These results indicate that PFUnDA is bioaccumulative in fish tissues. In contrast, BCF values of PFASs in crustacean (except for PFHpA) and PFCAs in gastropod and bivalve were less than 1000 L/kg.

Although PFOA concentrations in water were relatively higher than those of PFOS, the BCFs of PFOA in biota were 2~13 fold less than those of PFOS. This bioconcentration pattern of PFOA and PFOS is consistent with that reported in earlier study [40]. The BCF of PFOS was as follows: giant prawn soft tissues (156 L/kg) < stripped snakehead liver (181 L/kg) < dusky sleeper whole body (253 L/kg) < tilapia muscle (327 L/kg) < stripped snakehead muscle (894 L/kg) < golden apple snail (1117 L/kg) < flying barb whole body (1147 L/kg) < bivalve soft tissue (2110 L/kg) < tilapia liver (3551 L/kg). The mean BCF of PFOS in biological groups was determined in the ascending order as: crustacean (115 L/kg), gastropod (1117 L/kg), fish (1120 L/kg) and bivalve (2110 L/kg).

#### 4. Conclusions

Overall, the results of this study indicate a general ascending order of PFASs concentration in riverine ecosystems comprising water, sediment, gastropods, crustaceans, bivalves and fish tissues. PFOA was consistently higher than PFOS in waters. The measured concentrations were below the potential chronic and/or acute effects toxic thresholds in aquatic organisms. PFOS and PFHxS were found predominant in sediment. Some long-chain PFCAs including PFNA, PFUnDA and PFTTrDA as well as PFHxS were more abundant than other compounds in biota tissues. Different PFAS composition patterns in biota tissues suggested compound-, species-, and tissues-specific bioaccumulation. The rank order of mean BCF for PFOS in biological species was; crustacean (115 L/kg) < gastropod (1117 L/kg) < fish (1120 L/kg) < bivalve (2110 L/kg). The profiles of occurrence and spatial distribution of PFASs in various environmental media provide baseline information for a better understanding of the PFAS contamination in Vietnam. WWTP discharge is a source of PFASs in the aquatic environment. Further studies with greater numbers of water and aquatic organisms are needed for accurate assessment of sources and pathways of exposures, and bioaccumulation of PFASs in aquatic environment in Vietnam.

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#### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.jhazmat.2016.04.010>.

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