Environment International 135 (2020) 105377

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Legacy and emerging persistent organic pollutants in the freshwater system: Relative distribution, contamination trends, and bioaccumulation

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ARTICLE INFO

Handling Editor: Da Chen

Keywords:
Persistent organic pollutants
Overall monitoring
Relative distribution
Growth-dependent accumulation
Maternal transfer

ABSTRACT

In this study, a comprehensive investigation was performed to understand the overall occurrence, relative distribution, and bioaccumulation of seven different groups of POPs, comprising 27 polybrominated diphenyl ethers (PBDEs), 76 polychlorinated biphenyls (PCBs), 23 organochlorine pesticides (OCPs), three hexabromocyclododecanes (HBCDs), and 13 perfluoroalkyl substances (PFASs) as legacy POPs, and 41 polychlorinated naphthalenes (PCNs) and 24 short-chain chlorinated paraffins (SCCPs) as emerging POPs, by monitoring crucian carp, sediment, and river water in the freshwater system. Among the targeted POPs, SCCPs were predominant in sediment and crucian carp (more than 95%), while a dominance of PFASs was observed in river water (92%). Principal component analysis revealed four different groups/patterns of POPs in all media: one for PBDEs, PCBs, and OCPs, another for HBCDs and PFASs, and the two others for PCNs and SCCPs. Also, sexually dimorphic growth-dependent accumulation of legacy POPs was observed in crucian carp such that POPs concentration increased with increasing fish size and males recorded significantly higher levels of POPs compared to females.

1. Introduction

Since the designation of 12 persistent organic pollutants (POPs) (e.g., polychlorodibenzodioxins/furans [PCDD/Fs], polychlorinated biphenyls [PCBs], and some organochlorine pesticides [OCPs]) for the first time in 2001, several other pollutants have been regulated continuously in their production and use. These include mixtures of polybrominated diphenyl ethers (PBDEs), perfluorooctane sulfonic acid (PFOS), and salts of PFOS as representative perfluoroalkyl substances (PFASs) in 2009 and hexabromocyclododecanes (HBCDs) in 2013 as legacy POPs under the Stockholm Convention. In addition, polychlorinated naphthalenes (PCNs) and short-chain chlorinated paraffins (SCCPs) were recently designated as emerging POPs in 2015 and 2017, respectively. Despite these regulations, the distribution and fate of POPs in the freshwater system continue to have high priority because of their toxicity and persistence in biota, sediment, and water (Malarvannan et al., 2013; Jiang et al., 2018).

The fate of POPs in the freshwater system is known to depend generally on their production, usage, period of regulation, persistence, and physicochemical properties (deBruyn et al., 2009; Wang et al., 2019). Due to the strong hydrophobicity of legacy POPs, many previously conducted studies have investigated the historical residues, which are primarily initiated during precipitation events that lead to loadings in sediment and bioaccumulation in biota (Svihlikova et al., 2015; Jiang et al., 2018). Furthermore, several bioaccumulation factors have been investigated in fish species, as important representative aquatic organisms, to predict their major accumulation routes (Nyholm et al., 2008; Wang et al., 2018). The levels of PBDEs, PCBs, OCPs, and HBCDs were shown to increase as fish grow (Gui et al., 2014; Wang et al., 2018) but to decrease at a certain growth stage in females, suggesting the possibility of growth- or age-dependent accumulation and gender difference (Nyholm et al., 2008; Sühring et al., 2015). However, some previous research obtained opposite results, which resulted from differences in the chemicals targeted, biota species, and surrounding environments (Deribe et al., 2011; Iqbal et al., 2017).

Aside from the historical residues of legacy POPs, the continuous occurrence and contamination of non-regulated or emerging POPs in the freshwater system are also an urgent concern (van Mourik et al., 2016; Park et al., 2018), suggesting a different aspect of management necessity for new pollutants. Interestingly, unlike legacy POPs, for

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https://doi.org/10.1016/j.envint.2019.105377
Received 8 August 2019; Received in revised form 27 November 2019; Accepted 29 November 2019

which substantial information is available, emerging POPs, such as PCNs and SCCPs, have received little attention, especially regarding their fate and bioaccumulation potentials, despite their continuous emissions into the environment (van Mourik et al., 2016; Kim et al., 2019). Due to the differences of diverse POPs in physicochemical properties, usage, and regulation, a field study on the simultaneous monitoring of legacy and emerging POPs in the freshwater system is essential for a better understanding of their relative distribution, contamination patterns, and bioaccumulation factors under same environmental conditions.

Therefore, in this study, the relative distributions and contamination patterns of seven groups of legacy (PBDEs, PCBs, OCPs, HBCDs, and PFASs) and emerging (PCNs and SCCPs) POPs were investigated by monitoring 46 crucian carp and the surrounding environments (sediment and water) in the freshwater system of South Korea. Additionally, we evaluated growth-dependent accumulation and sexually dimorphic growth differences as significant factors influencing the bioaccumulation potential of legacy and emerging POPs in crucian carp. To the best our knowledge, this study is the first study to investigate the relative distribution and contamination patterns of seven legacy and emerging POPs simultaneously following an overall POPs monitoring in a freshwater system.

2. Material and methods

2.1. Chemicals

The targeted POPs were 27 mono- to deca-PBDEs, 76 mono- to deca-PCBs, 23 OCPs, three HBCDs, 13 PFASs, 41 tri- to octa-PCNs, and 24 SCCPs. The details of individual target compounds are given in Supporting Information Table S1. All individual internal standards of POPs, including a mixture of ^{13}C-labeled PBDEs (MBDE-MXG), PCBs (PCB-LCS-H), OCPs (ES-5349), and PCNs (ECN-5102), and individual ^{13}C-labeled α-, β-, and γ-HBCD, PFOS, perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), and perfluorododecanoic acid (PFDoDA) were obtained from Wellington Laboratories (Guelph, Canada). For SCCPs, three technical SCCP mixtures with 51%, 55.5%, and 63% chlorine contents were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany), and $^{13}C_6$-α-hexachlorocyclohexane ($^{13}C_6$-α-HCH) was used as the internal standard.

2.2. Sampling campaign

From June–November 2018, forty-six crucian carp (*Carassius auratus*) were collected from three major rivers in South Korea, the Nakdong (n = 13), Namhan (n = 17), and Yeongsan (n = 16). Muscle tissues were directly separated from the crucian carp and stored in glass amber jars at − 4 °C until analysis. The total length, body weight, and lipid contents of the crucian carp ranged from 22.7 to 44.0 cm, 173–1,050 g, and 0.150–6.88%, respectively. Additionally, river sediment (n = 10) and water (n = 10) samples were collected from three locations in each river (four locations in the Nakdong River) to assess the contamination status of POPs in the surrounding environmental media.

2.3. Experimental procedures and analysis

2.3.1. PBDEs, PCBs, OCPs, and PCNs

Wet-based muscle tissues (5 g) were mixed with 10–15 g of anhydrous sodium sulfate (Na_2SO_4) to absorb water in the samples. The samples were extracted twice by ultrasonication with 30 mL of a dichloromethane (DCM):hexane (1:1 v/v) mixture for 30 min, and the contents were separated from tissues by centrifugation at 4,500 rpm for 10 min. A 1-L volume of filtered water was extracted by liquid–liquid extraction (LLE) with 50 mL of DCM for two times, and filter paper extraction was conducted using the same method employed for muscle tissues followed by the Korean standard method (ES 10906.1) and the United States Environmental Protection Agency (U.S. EPA) Method 527. Both extracts were combined. Accelerated solvent extractor (ASE350; Dionex, Sunnyvale, CA, USA) was used for the extraction of 5 g of freeze-dried sediments with a mixture of DCM:hexane (1:1, v/v). After the extraction steps, all samples were cleaned using multilayer columns packed with Na_2SO_4 (2 g), neutral silica (1 g), 2% KOH silica (4 g), neutral silica (1 g), 44% acidic silica (8 g), neutral silica (2 g), and Na_2SO_4 (2 g) and eluted with 200 mL of DCM:hexane (1:9, v/v).

2.3.2. SCCPs

Extraction procedures for SCCPs in muscle tissues and river water were the same as those available for old legacy POPs. For sediment samples, 5 g of freeze-dried sample was extracted twice with DCM:hexane (1:1, v/v) for 30 min, and the extracts were separated by centrifugation at 4,500 rpm for 10 min. For the clean-up steps, each sample (muscle tissue, river water, or sediment) was passed through a multilayer column packed with Na_2SO_4 (2 g), florisil (3 g), neutral silica (2 g), 44% acidic silica (6 g), and Na_2SO_4 (2 g). In addition, to separate other chlorinated or structurally similar compounds, 40 mL of hexane (1st fraction) was discarded, and the columns were eluted with DCM:hexane (1:1, v/v) (2nd fraction).

2.3.3. Hbcds

Ultrasonication was employed for the extraction of 5 g wet-based muscle tissue and freeze-dried sediment samples using DCM:hexane (1:1, v/v) as the extraction solvent. Samples were centrifuged at 4500 rpm for 15 min. All extracts of muscle tissue and sediment were cleaned using 8 g of 1.5% deactivated silica and 10 g of 44% acidic silica and eluted with DCM:hexane (1:1, v/v). For the extraction of river water samples, 6 L of water was extracted by solid-phase extraction (SPE) with a Supelco C18 cartridge (6 cm^3, 500 mg) after DCM, methanol, and Milli-Q water preconditioning. At the end of sample loading, the cartridge was eluted with DCM:hexane (1:1, v/v).

2.3.4. PFASs

Samples (1 g) of wet-based muscle tissue and freeze-dried sediment were extracted with 5 mL of methyl *tert*-butyl ether (MTBE) by ultrasonication after adding 2 mL of purified water, 1 mL of 0.5 M *tert*-butyl alcohol (TBA), and 2 mL of 0.25 M sodium carbonate buffer as an ion-pairing reagent. The supernatants were separated from the samples by centrifugation at 4,500 rpm for 10 min. For river water, 1 L water samples were extracted by SPE after preconditioning with methanol and Milli-Q water, and the SPE cartridge was eluted using 6 mL of methanol.

2.3.5. Instrumental analysis

Instrumental methods for five POPs (PBDEs, PCBs, HBCDs, PFASs, and PCNs) were extensively described in our previously published papers (Choo et al., 2017; Gu et al., 2017; Park et al., 2018; Moon et al., 2019; Kim et al., 2019). The instrumental analyses were conducted by gas chromatography high-resolution mass spectrometry (GC-HRMS) for PBDEs, PCBs, PCNs, and OCPs, gas chromatography mass spectrometry (GC–MS) for SCCPs, and liquid chromatography with tandem mass spectrometry (LC-MS/MS) for HBCDs and PFASs. Due to the lack of previously published studies on OCPs and SCCPs by our research team, their instrumental analysis information is available in the Supporting Information (S1) of this study.

2.4. Quality assurance/quality control (QA/QC)

Detailed QA/QC information of legacy and emerging POPs, including method detection limits (MDLs), accuracy, and recovery, is shown in the Supporting Information (Table S2). Procedural blanks in every 10th to 15th sample batch were mostly not detected (ND) or lower than MDLs. Accuracy was determined by the analysis of three replicates with the injection of native and internal standards in the same manner as for the sample pretreatments, and the range was from 73 − 140% in all matrix within 15% of the relative standard deviation (RSD). The MDLs, given in Table S2, were obtained by a signal-to-noise ratio of three (3), and the recoveries were 30.3–120% in all matrices.

2.5. Calculation of BCF and BSAF

The bioconcentration factor (BCF, L/kg) and biota-sediment accumulation factor (BSAF) were calculated by using Eqs. (1) and (2), where C_{biota}/f_{lipid} is the lipid-based concentration in crucian carp, C_{water} is the water concentration, and $C_{sediment}/f_{oc}$ is the total organic carbon (TOC)-based concentration in sediment. The lipid contents of crucian carp were analyzed by gravimetrical measurement, and TOC in sediment was measured by the weight difference of dried sediment before and after baking at 450 °C.

$$BCF = \frac{C_{biota}}{C_{water}} \tag{1}$$

$$BSAF = \frac{C_{biota}}{C_{sediment}/f_{oc}} \tag{2}$$

3. Results and discussion

3.1. Concentrations of POPs in the freshwater system

The mean concentrations of POPs in river water and sediment were 0.20 ng/L and 3.05 ng/g dry weight (dw) for PBDEs, 0.09 ng/L and 0.02 ng/g dw for PCBs, 0.89 ng/L and 0.25 ng/g dw for OCPs, 0.07 ng/L and 0.54 ng/g dw for HBCDs, 15.0 ng/L and 0.18 ng/g dw for PFASs, ND and 0.004 ng/g dw for PCNs, and ND and 90.7 ng/g dw for SCCPs, respectively. For crucian carp, the mean concentrations of PBDEs, PCBs, OCPs, HBCDs, PFASs, SCCPs, and PCNs were 0.25, 0.24, 0.23, 0.79, 3.86, 123 ng/g wet weight (ww), and 1.04 pg/g ww, respectively.

In general, the concentrations of all targeted POPs in this study were similar to or lower than those of other studies of the freshwater system of South Korea, and of other countries (Table 1). In particular, the levels of PBDEs, PCBs, OCPs, and HBCDs in both biotic and abiotic media were lower than those reported in previous studies conducted in South Korea (Moon et al., 2006, 2012; Wang et al., 2018), China (Zhou et al., 2007; Wu et al., 2008; Xu et al., 2009; Hu et al., 2010; He et al., 2013), and England (Harrad et al., 2009). The concentration of PFASs in the freshwater system of this study was similar to that of China (Shi et al., 2012), but higher levels were observed in environmental media of Spain (Campo et al., 2016) and Vietnam (Lam et al., 2017). A similar or lower concentration of emerging POPs (PCNs and SCCPs) was obtained in this study compared to those of previous studies in South Korea (Kim et al., 2019), the USA (Kannan et al., 2000; Houde et al., 2008; Helm et al., 2008), China (Zeng et al., 2011), and the Czech Republic (Přibylová et al., 2006).

3.2. Contamination patterns of seven groups of POPs in the freshwater system

The occurrence and distribution of POPs in the freshwater system can vary according to diverse factors, such as their characteristics, regulation, usage, and bioaccumulation potential (deBruyn et al., 2009; Wang et al., 2019). In the present study, due to the absence of overall monitoring of representative POPs, we investigated the relative distribution of seven groups of POPs for the first time to understand their overall fate and contamination trends in the freshwater system, including both biotic and abiotic media, more thoroughly. In addition, an assessment of the relationships of, and similarities in, the fate, source, usage, and contamination trends of each POPs in the freshwater system was conducted by principal component analysis (PCA).

3.2.1. Relative distribution of POPs in the freshwater system

The relative distributions of legacy and emerging POPs in river water, sediment, and crucian carp (wet-based) were obtained by the median distribution percentage of each sample (Fig. 1), and there were no differences between wet- and lipid-based distributions in biota (Fig. S1). Specific distribution patterns of PBDEs, PCBs, OCPs, HBCDs, PFASs, and PCNs are also shown in the Supporting Information (Fig. S2). Generally, the fate of pollutants in environments can be anticipated from their physicochemical properties (deBruyn et al., 2009; Wang et al., 2019); however, SCCPs were overwhelmingly dominant in river sediment (95.4%, Fig. 1B) and crucian carp (95.4%, Fig. 1C), despite their similar or slightly lower log K_{ow} values (5–8.5) compared to those of other POPs (log K_{ow}: 4.46–9.97). The major concern with SCCPs is their high production volumes and industrial use worldwide (1000 kton/year), which are devoid of strict regulation, especially in Asian countries, indicating that the usage amounts of SCCPs might play a significant role in their recorded highest distribution compared to the other POPs investigated in this study (Zeng et al., 2011; van Mourik et al., 2016). Interestingly, despite the high range of log K_{ow} values of PCNs (3.9–10.4) (NICNAS, 2002), their distributions were found to be the lowest in both sediment and crucian carp (lower than 0.01%). This result of the present study compares well with the result of a recently published study that reported that PCN residues and usage were scarce in South Korea (Kim et al., 2019). The emerging POPs were not detected in river water.

Aside from the distinct distribution of emerging POPs, the distribution of legacy POPs also varied according to the media. PFASs were overwhelmingly predominant (92.3%) in the river water (Fig. 1A), followed by OCPs (5.48%) ≫ PBDEs (1.23%) ≈ PCBs (0.57%) ≈ HBCDs (0.42%). The dominance of PFASs and OCPs (higher than 98%) in the river water seemed to be due to the much higher water solubility than that of other groups or major congeners of each POPs (PFOA: 3.4 g/L ≫ HCHs: 7.3 mg/L > PBDEs: 0.00087–0.04 mg/L, PCBs: 0.0027–0.42 ng/L, HBCDs: 0.002–0.03 mg/L) (PubChem; UNEP, 1999; Breitholtz et al., 2007; Gu et al., 2017; Lee et al., 2017). On the other hand, a dominance of PBDEs (3.49%), especially deca-BDE (3.37%) (Fig. S1), was observed in river sediment (Fig. 1B), which might be due to the high log K_{ow} value (9.97) (Mackay et al., 2006). These were followed by HBCDs (0.60%), OCPs (0.28%), PFASs (0.20%), and PCBs (0.02%), whose major congeners have a similar range of log K_{ow} values (HBCD: 5.62, dichloro-diphenyl-trichloroethanes (DDTs): 4.9–6.9, PFOS: 4.49, PCBs: 4.46–8.18) (Renner, 2002; Zhao et al., 2016; Demirtepe and Imamoglu, 2019) and were, in fact, the congeners with the highest concentrations in river sediment.

Distribution patterns similar to those of river sediment (Fig. 1B) were also observed for POPs in crucian carp (PFASs [3.13%] > HBCDs [0.65%] ≈ PCBs [0.28%] ≈ PBDEs [0.21%] ≈ OCPs [0.19%]) (Fig. 1C) because bioaccumulation of pollutants is also dependent on their partitioning, except for the case of PBDE. The low bioaccumulation of BDE209 was known, despite the dominance in the sediment, due to its relatively shorter half-life and lower bioavailability in fish species compared to BDE47 which was the most dominant congener in crucian carps (lake trout: 21–34 days for BDE209, 173–519 days for BDE47) (Stapleton et al., 2004; Thomas, 2005; Kim et al., 2015). Despite the low distribution of PFASs in river sediment (0.20%) (Fig. 1B), PFASs had the highest contribution among legacy POPs in crucian carp (3.13%, Fig. 1C), which is consistent with previously reported results (PFASs > PBDEs ≈ HBCDs) (Hloušková et al., 2013; Svihlikova et al., 2015). Compared to other lipid-soluble POPs, PFASs could have higher bioaccumulation potential in crucian carp due to their strong ability to bind to proteins, which occupy nearly 80% of the body components of crucian carp (Haukås et al., 2007).

Table 1
Concentration range (mean) of seven groups of POPs by comparison with previous studies conducted in the freshwater environment.

POPs	Location	Congeners	Sampling	Water (ng/L)	Sediment (ng/g dw[a])	Crucian carp (ng/g ww[b])	References
PBDEs *(legacy)*	South Korea	27	2018	0.06–0.37 (0.20)	1.59–5.57 (3.05)	0.001–1.80 (0.254) lw[c]: 0.11–87.2 (20.6)	This study
	South Korea	23	2008	4.5 ± 3.3	2,180 ± 5,273	–	Moon et al., 2012
	China	9	2009	–	0.15–2.4	lw: 14	Xu et al., 2009
	China	8	2006	23.8–25.0 (24.4)	–	lw: 46.6–853 (3 1 6)	Wu et al., 2008
PCBs *(legacy)*	South Korea	76	2018	0.04–0.21 (0.09)	0.01–0.05 (0.02)	0.02–1.36 (0.24) lw: 2.54–96.8 (22.3)	This study
	South Korea	61	2002	–	–	31.4	Moon et al., 2006
	China	10	2006	196–206 (2 0 4)	–	lw: 1,963–18,174 (8,338)	Wu et al., 2008
OCPs *(legacy)*	South Korea	23	2018	0.12–2.52 (0.89)	0.07–0.91 (0.25)	0.02–1.23 (0.23) lw: 0.78–171 (29.4)	This study
	China	10	2007	4.6 ± 1.2	14 ± 1.3	lw: 222 ± 38	Hu et al., 2010
	China	13	2005	136.9	32.9	14.7	Zhou et al., 2007
HBCDs *(legacy)*	South Korea	3	2018	ND[d]–0.19 (0.07)	0.03–1.79 (0.54)	ND–14.0 (0.79) lw: ND–330 (33.2)	This study
	South Korea	3	2015–2016	ND–0.34 (0.22)	0.15–25.7 (3.55)	0.38	Wang et al., 2018
	China	3	2009	9.5–82.4 (39.7)	0.07–31.6 (6.9)	lw: 17.5–154 (Mud carp)	He et al., 2013
	England	3	2008–2009	0.08–0.27	0.88–4.8	lw: 75–290	Harrad et al., 2009
PFASs *(legacy)*	South Korea	13	2018	1.07–44.7 (15.0)	0.11–0.35 (0.18)	0.39–22.9 (3.86) lw[c]: 8.57–2,164 (3 1 8)	This study
	Vietnam	13	2013–2015	ND[d]–107	ND–23.4	ND–26.2 (Fish species)	Lam et al., 2017
	Spain	21	2010	0.04–83.1	0.22–11.5	0.63–274 (Fish species)	Campo et al., 2016
	China	6	2008	1.70–73.5	0.06–0.64	ND–26.2 (1.13)	Shi et al., 2012
PCNs *(emerging)*	South Korea	41	2018	ND	0.002–0.01 (0.004)	0.22–4.84 (1.04) pg/g lw: 0.01–0.44 (0.15)	This study
	South Korea	41	2016–2017	ND	0.004–0.02	2.17 ± 0.68 pg/g	Kim et al., 2019
	US	38	2002–2003	–	28.9 ± 6.7	3.5 ± 3.2 (Lake trout)	Helm et al., 2008
	US	–	1996–1997	–	–	0.43–26	Kannan et al., 2000
SCCPs *(emerging)*	South Korea	24	2018	ND	27.4–146 (90.7)	ND–604 (1 2 3) lw: ND–20,568 (8,080)	This study
	China	–	2010	162–176	1.1–8.7 μg/g dw	lw: 25.2 μg/g	Zeng et al., 2011
	US	–	2000–2004	1.19 ± 0.43	–	123 ± 35 (Lake trout)	Houde et al., 2008
	Czech	–	2003–2004	–	ND–397	–	Přibylová et al., 2006

[a] Dry weight,
[b] Wet weight,
[c] Lipid weight,
[d] Not detected.

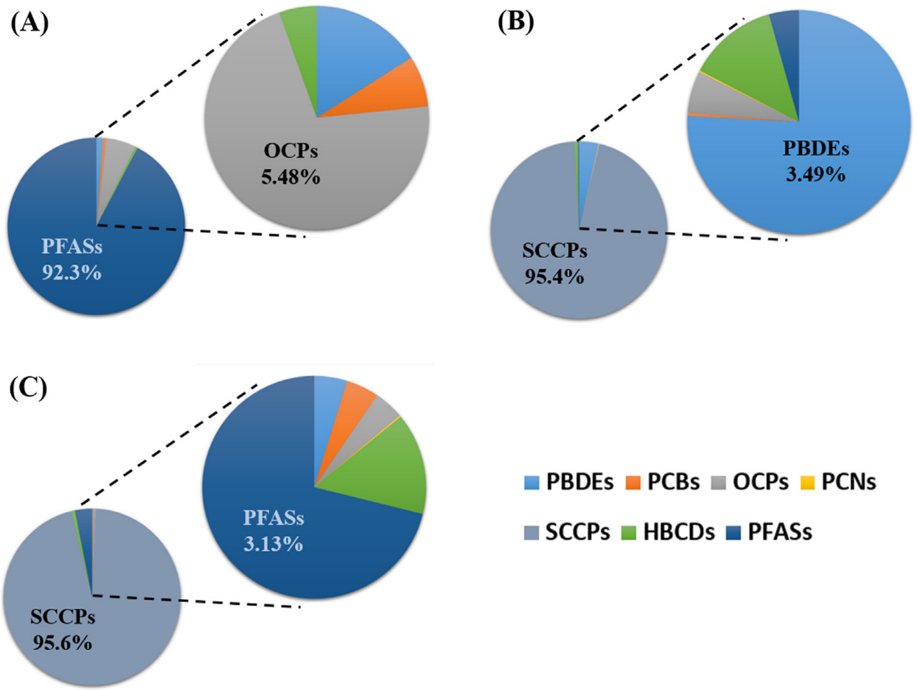

Fig. 1. Relative distribution of legacy and emerging POPs in river water (A), river sediment (B), and crucian carp (C).

3.2.2. The relationship between physicochemical properties and the fate of POPs

Efforts have been made in previous studies to assess the effects of physicochemical properties and compensate for the different distributions in both biotic and abiotic media by investigating the relationships between physicochemical properties and the fate of pollutants using a scatter plot of K_{ow}, BSAF, and BCF (deBruyn et al., 2009; Wang et al., 2019). As a result, it has been reported that the bioaccumulation of legacy POPs from both sediment (BSAF) and water (BCF) increased with increasing K_{ow} value, but the bioavailability of extremely hydrophobic compounds can be very low for sediment, which showed a decrease in log BSAF values similar to a parabolic relationship (Sun et al., 2017; Wang et al., 2019). However, only several congeners of one group of POPs were described, so there has been no comprehensive study that has investigated the relationship between K_{ow} and BCF/BSAF of several legacy and emerging POPs simultaneously. Therefore, in this present study, field-based BSAF and BCF values were calculated for major congeners of legacy and emerging POPs (BDE47, BDE100, CB52, CB99, CB153, DDTs, HCHs, CN42 α-HBCD, and PFOA), which had 100% detection frequency and the highest concentration in both biotic and abiotic media. Due to the limitation of quantification of each SCCP congener, we calculated the BSAF of SCCPs by using total concentration.

The scatter plot of K_{ow}, BSAF, and BCF is given in Fig. 2, and BSAF and BCF values are shown in Table S3. As is consistent with previous study results reported for each group of POPs (Wu et al., 2008; deBruyn et al., 2009), a parabolic relationship between log BSAF and log K_{ow} was observed for the seven groups of POPs (Fig. 2A). Interestingly, log BSAF values increased at log $K_{ow} < 7$ but then sharply decreased with increasing log K_{ow} (Wu et al., 2008). This tendency was also reported in previous results of each congener of PBDEs and PCBs (Sun et al., 2017; Wang et al., 2019), implying that this standard (log K_{ow}: 7) could apply not only for PBDE and PCB congeners but also for all legacy and emerging POPs. Fig. 2B shows a significant correlation between log K_{ow} and log BCF, indicating that the bioaccumulation of legacy POPs (ND for emerging POPs) can be increased with increasing hydrophobicity (Wang et al., 2019).

3.2.3. Similarity of each group of POPs in the freshwater system

To trace the similarity of each group of POPs in this study, a principal component analysis (PCA) was carried out, as shown in Fig. 3. All investigated media seemed to show a similar relationship in which the POP compounds clustered into four groups: one for PBDEs, PCBs, and OCPs, another for HBCDs and PFASs, and the two others for PCNs and SCCPs, respectively. PBDEs, PCBs, and OCPs are categorized as old legacy POPs, globally regulated since the 2000s (Lee et al., 2015), and their detection might have been due to the presence of their historical residues in the environment or uncommon use of their non-regulated congeners (Lee et al., 2015; Park et al., 2018). However, the contamination patterns of PFASs and HBCDs were quite different from those of the old legacy POPs because they are still used for industrial purposes (Lam et al., 2017; Park et al., 2018). Only PFOS and its salts have been regulated as POPs since 2009, and other PFAS compounds have been used continuously as alternatives; hence, these are still being detected in diverse environments due to their exceptional industrial uses in South Korea (Lam et al., 2017; Park et al., 2018). This tendency can also be observed in the case of HBCDs, which have been allowed for specific purposes such as in the production of expanded polystyrene (EPS) and extruded polystyrene (XPS) despite their ban in 2013 (Gu et al., 2017; Wang et al., 2018).

However, PCNs and SCCPs both had tendencies different from those of the other POPs in this study because they are emerging POPs. Even though PCNs can be released continuously from combustion and industrial processes as an emerging pollutant (Kim et al., 2019), it would be difficult to assess contamination patterns of PCNs by comparison with other POPs because the distribution of PCNs in all media (0.01% in sediment and crucian carp, 0% in water) of this study was significantly lower than those of other POPs in South Korea, as shown in Fig. 1, indicating that PCNs are not widely used and distributed in South Korea (Kim et al., 2019). On the contrary, loading routes of SCCPs in environmental media and biota might differ from those of the regulated POPs and less used PCNs due to their current usage and high production volume for industrial purposes (Sun et al., 2017). Especially, SCCPs, unlike other POPs, were greatly used as extreme pressure additives in metal-working fluids, indicating that differences in major usage can also be a significant factor influencing SCCP contamination routes (Zeng et al., 2016).

3.3. Growth-dependent accumulation and maternal transfer of POPs

To assess the growth-dependent accumulation of legacy and emerging POPs in crucian carp, the relationships between wet weight-based POPs concentration and growth factors (total length and body weight) were investigated, as given in Table 2. There were significant positive correlations between physical morphometrics and concentrations of legacy POPs, implying a tendency of POPs to accumulate in crucian carp as the fish grow, which is consistent with previous studies (Deribe et al., 2011; Lam et al., 2017; Wang et al., 2018).

Aside from the growth-dependent accumulation, we also evaluated gender difference simultaneously because sexually dimorphic growth tendencies are strongly related to maternal transfer (Nyholm et al., 2008; Sühring et al., 2015). As a result, unique growth-dependent accumulation potentials were observed when comparing the concentrations between 16 males and 30 females. Even though no statistical

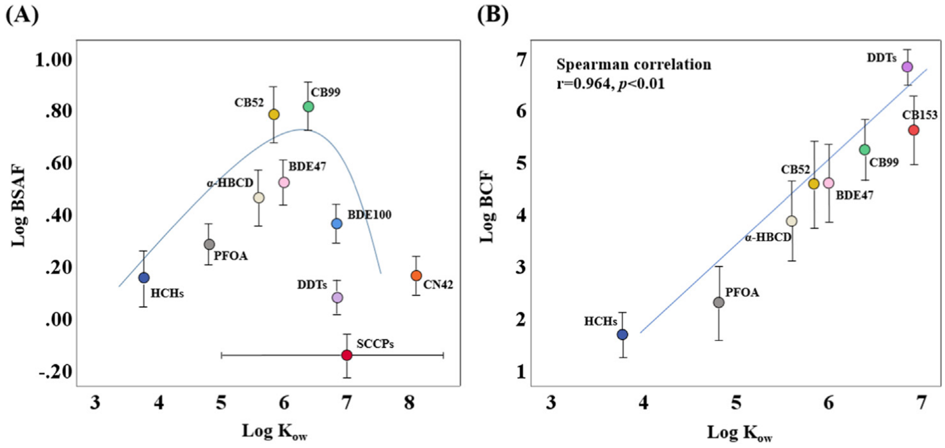

Fig. 2. Relationship between field-based BSAF (A) and BCF (B) with Log K_{ow} of the representative legacy POPs congeners.

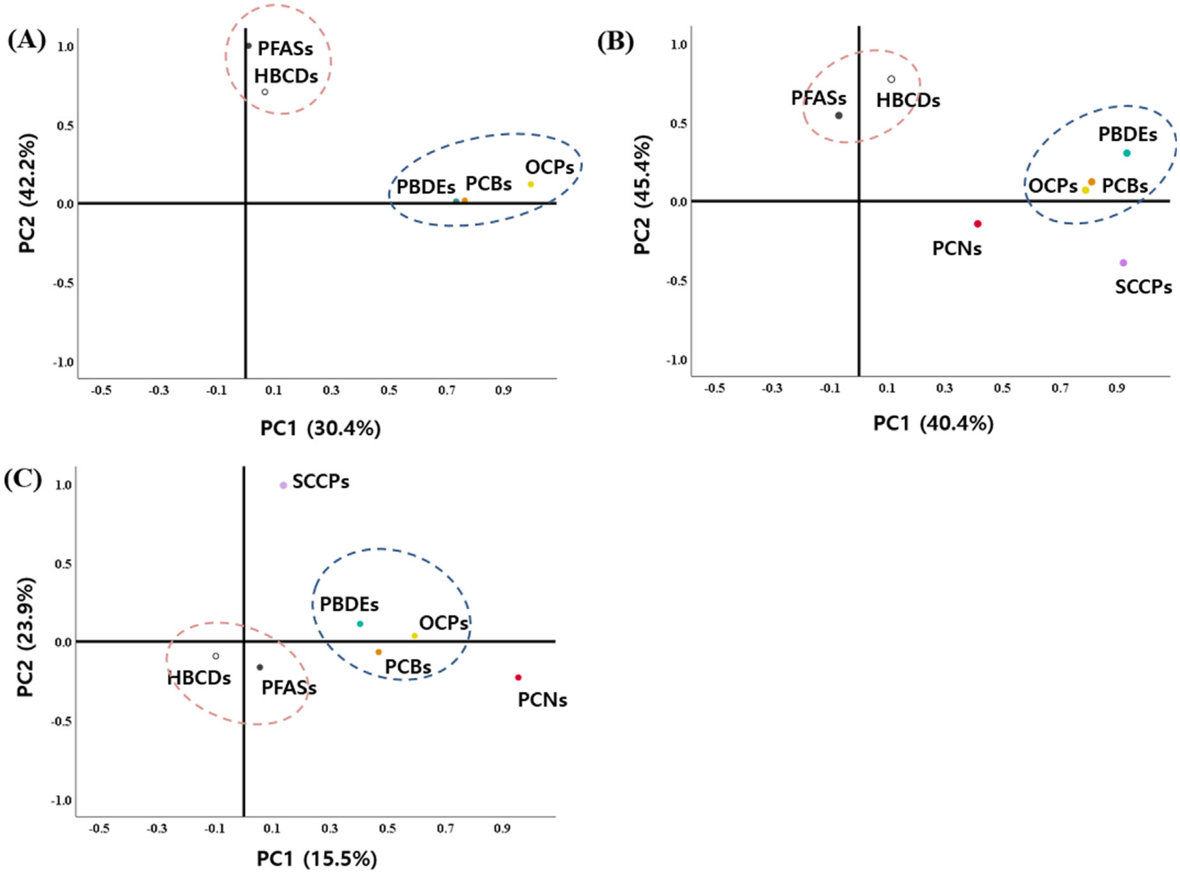

Fig. 3. Principal component analysis (PCA) of POPs in river water (A), sediment (B), and crucian carp (C).

Table 2
Relationship between POPs concentration and growth factors.

Growth	PBDEs	PCBs	OCPs	HBCDs	PFASs	PCNs	SCCPs
Total length (cm)	*0.393***	*0.372***	*0.449***	*0.311***	*0.624***	0.219	0.209
Body weight (g)	*0.412***	*0.410***	*0.392***	*0.333***	*0.534***	0.153	0.172

Spearman correlation, *: $p < 0.05$, **: $p < 0.01$.

Table 3
Mean concentration of POPs in males and females.

	ng/g ww						pg/g ww
Gender	PBDEs	PCBs	OCPs	HBCDs	PFASs	SCCPs	PCNs
Male	*0.477***	*0.414***	*0.354***	*1.76***	*7.18***	104	1.29
Female	0.139	0.154	0.165	0.268	1.90	126	0.887

Mann-Whitney *U* test, *: $p < 0.05$, **: $p < 0.01$.

difference was observed between the growth (total length and body weight) of males (31.0 cm and 405 g) and females (29.8 cm and 399 g) (Mann–Whitney *U* test, *p* greater than 0.05), significantly higher concentrations of legacy POPs (PBDEs, PCBs, OCPs, HBCDs, and PFASs) were observed in males than females, (Mann–Whitney *U* test, $p < 0.05$) as shown in Table 3. This phenomenon can be attributed to the possibility of decrement of POPs concentration in females due to maternal transfer as they attain reproductive maturity (Nyholm et al., 2008; Choo et al., 2018).

A more evident pattern of this phenomenon was observed when legacy POP concentrations of male and female crucian carp were compared before and after they reached reproductive maturity. In this present study, fish were divided into two groups by a specific growth point (approximately 27.4 cm of total length), which was believed to be near the maximum length of reproductive maturity (Balik et al., 2004; Choo et al., 2018). As shown in Fig. 4, although the male and female crucian carp did not show any statistically significant difference in POP concentrations prior to 27.4 cm growth length for all groups of POPs investigated in this study, legacy POP concentrations were significantly lower in females than males when their total length was 27.8–44.0 cm (Mann–Whitney *U* test, $p < 0.05$). Additionally, a statistically significant increase in legacy POPs concentration was observed when comparing male crucian carp species before and after they reached 27.4 cm, while a similar concentration was observed for all female crucian carp. This indicates that maternal transfer of PBDEs, PCBs, OCPs, HBCDs, and PFASs might play a significant role in bioaccumulation in crucian carp. However, unlike the case of legacy POPs, no significant correlation was observed between emerging POPs (PCN and SCCP) concentration and growth factors (Table 3), and there was also no maternal transfer tendency, which compares well with the results of a recently published study (Fig. 4) (van Mourik et al., 2016; Kim et al., 2019). Because the influence of morphological factors remain unclear as already pointed out by several recent studies (van Mourik et al., 2016; Labadie and Chevreuil, 2011; Kim et al., 2019), further lab- or large-scale monitoring research is required to improve the understanding of the biological factors of emerging POPs in biota.

4. Conclusion

This study presented the occurrence, relative distribution, and bioaccumulation of legacy (PBDEs, PCBs, OCPs, HBCDs, and PFASs) and emerging (PCNs and SCCPs) POPs in river sediment, water, and crucian carp of South Korea. Among the POPs, SCCPs showed the highest distribution in river sediment and crucian carp, while PFASs

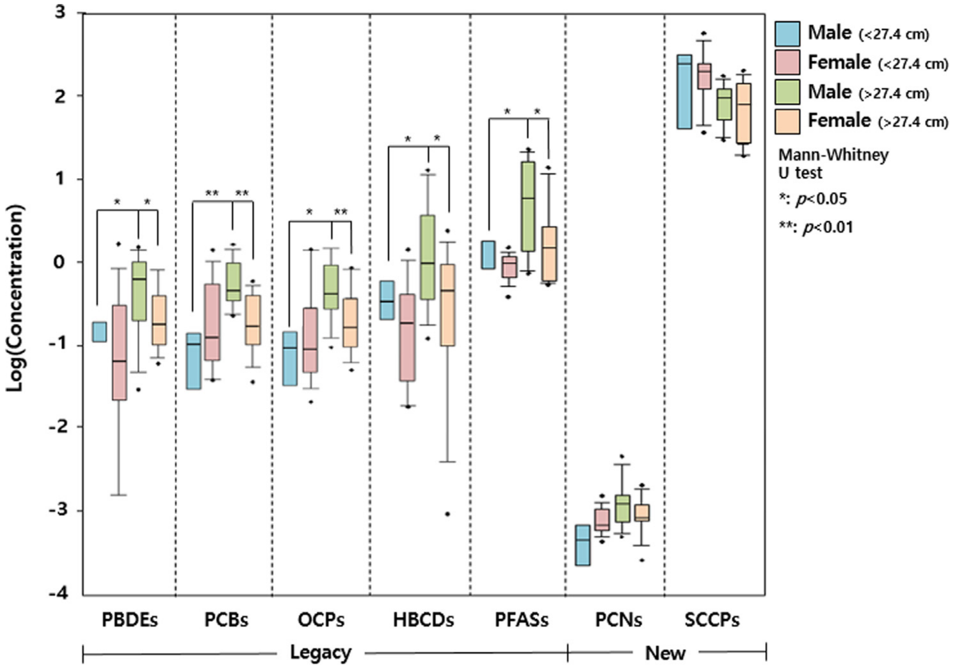

Fig. 4. Log concentration of legacy and emerging POPs in male and female crucian carp before and after maximum growth.

were predominant in river water. The distribution of legacy POPs seemed to have been influenced by their physicochemical characteristics, while emerging POPs showed quite different distribution patterns in all media. In addition, growth-dependent accumulation and maternal transfer were observed only for legacy POPs. In South Korea, even though continuous monitoring of legacy and emerging POPs has been conducted previously, it is difficult to evaluate the contamination status of all managed POPs because different biota species and targeted pollutants were investigated in each previous study. Therefore, the present study promises to be very useful for providing requisite knowledge on diverse bioaccumulation factors and different contamination patterns of legacy and emerging POPs through overall POPs monitoring in the freshwater system.

CRediT authorship contribution statement

Gyojin Choo: Investigation, Writing - original draft. **Wenting Wang:** Investigation, Validation. **Hyeon-Seo Cho:** Methodology, Validation. **Kyungtae Kim:** Project administration, Conceptualization. **Kyunghwa Park:** Project administration, Conceptualization. **Jeong-Eun Oh:** Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declared that there is no conflict of interest.

Acknowledgments

This work was supported by a grant from the National Institute of Environmental Research and Development Institute (NIER-SP2018-266) and Korea Environment Industry & Technology Institute (KEITI) through “The Chemical Accident Prevention Technology Development Project” funded by Korea Ministry of Environment (MOE).

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.105377.

References

Balik, İ., Özkök, R., Çubuk, H., Uysal, R., 2004. Investigation of some biological characteristics of the silver crucian carp, carassius gibelio (bloch 1782) population in lake eğirdir. Turk. J. Zool. 28, 19–28.

Breitholtz, M., Ricklund, N., Bengtsson, B.-E., Persson, N.J., 2007. Silica gel as a particulate carrier of poorly water-soluble substances in aquatic toxicity testing. Aquat. Toxicol. 82, 251–264. https://doi.org/10.1016/j.aquatox.2007.02.016.

Campo, J., Lorenzo, M., Pérez, F., Picó, Y., Farré, Marinel. la, Barceló, D., 2016. Analysis of the presence of perfluoroalkyl substances in water, sediment and biota of the Jucar River (E Spain). Sources, partitioning and relationships with water physical characteristics. Environ. Res. 147, 503–512. https://doi.org/10.1016/j.envres.2016.03.010.

Choo, G., Cho, H.-S., Park, K., Lee, J.-W., Kim, P., Oh, J.-E., 2018. Tissue-specific distribution and bioaccumulation potential of organophosphate flame retardants in crucian carp. Environ. Pollut. 239, 161–168. https://doi.org/10.1016/j.envpol.2018.03.104.

Choo, G., Kim, D.-H., Kim, U.-J., Lee, I.-S., Oh, J.-E., 2017. PBDEs and their structural analogues in marine environments: fate and expected formation mechanisms compared with diverse environments. J. Hazard. Mater. https://doi.org/10.1016/j.jhazmat.2017.09.026.

deBruyn, A.M.H., Meloche, L.M., Lowe, C.J., 2009. Patterns of bioaccumulation of polybrominated diphenyl ether and polychlorinated biphenyl congeners in marine mussels. Environ. Sci. Technol. 43, 3700–3704. https://doi.org/10.1021/es900472k.

Demirtepe, H., Imamoglu, I., 2019. Biostimulation enhanced the biotic degradation of hexabromocyclododecane in sediments. J. Soils Sediments. https://doi.org/10.1007/s11368-019-02280-z.

Deribe, E., Rosseland, B.O., Borgstrøm, R., Salbu, B., Gebremariam, Z., Dadebo, E., Norli, H.R., Eklo, O.M., 2011. Bioaccumulation of persistent organic pollutants (POPs) in fish species from Lake Koka, Ethiopia: the influence of lipid content and trophic position. Sci. Total Environ. 410–411, 136–145. https://doi.org/10.1016/j.scitotenv.2011.09.008.

Gu, S.-Y., Ekpeghere, K.I., Kim, H.-Y., Lee, I.-S., Kim, D.-H., Choo, G., Oh, J.-E., 2017. Brominated flame retardants in marine environment focused on aquaculture area: occurrence, source and bioaccumulation. Sci. Total Environ. 601–602, 1182–1191. https://doi.org/10.1016/j.scitotenv.2017.05.209.

Gui, D., Yu, R., He, X., Tu, Q., Chen, L., Wu, Y., 2014. Bioaccumulation and biomagnification of persistent organic pollutants in Indo-Pacific humpback dolphins (Sousa chinensis) from the Pearl River Estuary. China. Chemosphere 114, 106–113. https://doi.org/10.1016/j.chemosphere.2014.04.028.

Harrad, S., Abdallah, M.A.-E., Rose, N.L., Turner, S.D., Davidson, T.A., 2009. Current-use brominated flame retardants in water, sediment, and fish from english lakes. Environ. Sci. Technol. 43, 9077–9083. https://doi.org/10.1021/es902185u.

Haukås, M., Berger, U., Hop, H., Gulliksen, B., Gabrielsen, G.W., 2007. Bioaccumulation of per-and polyfluorinated alkyl substances (PFAS) in selected species from the Barents Sea food web. Environ. Pollut. 148, 360–371. https://doi.org/10.1016/j.envpol.2006.09.021.

He, M.-J., Luo, X.-J., Yu, L.-H., Wu, J.-P., Chen, S.-J., Mai, B.-X., 2013. Diasteroisomer and enantiomer-specific profiles of hexabromocyclododecane and tetrabromobisphenol A in an aquatic environment in a highly industrialized area, South China: Vertical profile, phase partition, and bioaccumulation. Environ. Pollut. 179, 105–110. https://doi.org/10.1016/j.envpol.2013.04.016.

Helm, P.A., Gewurtz, S.B., Whittle, D.M., Marvin, C.H., Fisk, A.T., Tomy, G.T., 2008. Occurrence and biomagnification of polychlorinated naphthalenes and non- and mono-ortho pcbs in lake ontario sediment and biota. Environ. Sci. Technol. 42, 1024–1031. https://doi.org/10.1021/es071819g.

Houde, M., Muir, D.C.G., Tomy, G.T., Whittle, D.M., Teixeira, C., Moore, S., 2008. Bioaccumulation and trophic magnification of short- and medium-chain chlorinated paraffins in food webs from lake ontario and lake michigan. Environ. Sci. Technol. 42, 3893–3899. https://doi.org/10.1021/es703184s.

Hu, G., Dai, J., Mai, B., Luo, X., Cao, H., Wang, J., Li, F., Xu, M., 2010. Concentrations and accumulation features of organochlorine pesticides in the baiyangdian lake freshwater food web of North China. Arch. Environ. Contam. Toxicol. 58, 700–710. https://doi.org/10.1007/s00244-009-9400-1.

Hloušková, V., Lanková, D., Kalachová, K., Hrádková, P., Poustka, J., Hajšlová, J., Pulkrabová, J., 2013. Occurrence of brominated flame retardants and perfluoroalkyl substances in fish from the Czech aquatic ecosystem. Sci. Total Environ. 461–462, 88–98. https://doi.org/10.1016/j.scitotenv.2013.04.081.

Iqbal, M. Legacy and emerging flame retardants (FRs) in the freshwater ecosystem: A review. 2017.

Jiang, W., Huang, T., Chen, H., Lian, L., Liang, X., Jia, C., Gao, H., Mao, X., Zhao, Y., Ma, J., 2018. Contamination of short-chain chlorinated paraffins to the biotic and abiotic environments in the Bohai Sea. Environ. Pollut. 233, 114–124. https://doi.org/10.1016/j.envpol.2017.10.034.

Kannan, K., Yamashita, N., Imagawa, T., Decoen, W., Khim, J.S., Day, R.M., Summer, C.L., Giesy, J.P., 2000. Polychlorinated naphthalenes and polychlorinated biphenyls in fishes from michigan waters including the great lakes. Environ. Sci. Technol. 34, 566–572. https://doi.org/10.1021/es990966e.

Kim, K.-W., Choo, G., Cho, H.-S., Lee, B.-C., Park, K., Oh, J.-E., 2019. The occurrence and distribution of polychlorinated naphthalenes (PCNs), focusing on tissue-specific bioaccumulation in crucian carp in South Korea. Sci. Total Environ. 665, 484–491. https://doi.org/10.1016/j.scitotenv.2019.01.238.

Kim, U.-J., Jo, H., Lee, I.-S., Joo, G.-J., Oh, J.-E., 2015. Investigation of bioaccumulation and biotransformation of polybrominated diphenyl ethers, hydroxylated and methoxylated derivatives in varying trophic level freshwater fishes. Chemosphere 137, 108–114. https://doi.org/10.1016/j.chemosphere.2015.05.104.

Lam, N.H., Cho, C.-R., Kannan, K., Cho, H.-S., 2017. A nationwide survey of perfluorinated alkyl substances in waters, sediment and biota collected from aquatic environment in Vietnam: Distributions and bioconcentration profiles. J. Hazard. Mater. Special Issue Emerg. Contamin. Eng. Nat. Environ. 323, 116–127. https://doi.org/10.1016/j.jhazmat.2016.04.010.

Labadie, Pierre, Chevreuil, Marc, 2011. Partitioning behaviour of perfluorinated alkyl contaminants between water, sediment and fish in the Orge River (nearby Paris, France). Environ. Pollut. 159 (2), 391–397. https://doi.org/10.1016/j.envpol.2010.10.039.

Lee, I.-S., Kang, H.-H., Kim, U.-J., Oh, J.-E., 2015. Brominated flame retardants in Korean river sediments, including changes in polybrominated diphenyl ether concentrations between 2006 and 2009. Chemosphere 126, 18–24. https://doi.org/10.1016/j.chemosphere.2015.01.004.

Lee, Y.-C., Wang, P.-Y., Lo, S.-L., Huang, C.P., 2017. Recovery of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) from dilute water solution by foam flotation. Sep. Purif. Technol. 173, 280–285. https://doi.org/10.1016/j.seppur.2016.09.012.

Mackay, D., Shiu, W.-Y., Ma, K.-C., Lee, S.C., 2006. Handbook of Physical-Chemical Properties and Environmental Fate for Organic Chemicals. CRC Press.

Malarvannan, G., Dirinck, E., Dirtu, A.C., Pereira-Fernandes, A., Neels, H., Jorens, P.G., Gaal, L.V., Blust, R., Covaci, A., 2013. Distribution of persistent organic pollutants in two different fat compartments from obese individuals. Environ. Int. 55, 33–42. https://doi.org/10.1016/j.envint.2013.02.012.

Moon, H., Kim, D.-H., Oh, J.-E., 2019. Dietary exposure to PCBs by seafood cooking method: A Korean study. Chemosphere 215, 775–782. https://doi.org/10.1016/j.chemosphere.2018.10.044.

Moon, H.-B., Choi, M., Yu, J., Jung, R.-H., Choi, H.-G., 2012. Contamination and potential sources of polybrominated diphenyl ethers (PBDEs) in water and sediment from the artificial Lake Shihwa, Korea. Chemosphere 88, 837–843. https://doi.org/10.1016/j.chemosphere.2012.03.091.

Moon, J.Y., Kim, Y.B., Lee, S.I., Song, H., Choi, K., Jeong, G.H., 2006. Distribution characteristics of polychlorinated biphenyls in crucian carp (Carassius auratus) from major rivers in Korea. Chemosphere 62, 430–439. https://doi.org/10.1016/j.chemosphere.2005.04.063.

NICNAS, 2002. Chlorinated naphthalenes. Chemical Assessment Report S48, National Industrial Chemicals Notification and Assessment Scheme.

Nyholm, Jenny Rattfelt, Norman, Anna, Norrgren, Leif, Haglund, Peter, Andersson, Patrik L., 2008. Maternal transfer of brominated flame retardants in zebrafish (Danio rerio). Chemosphere 73 (2), 203–208. https://doi.org/10.1016/j.chemosphere.2008.04.033.

Park, H., Choo, G., Kim, H., Oh, J.-E., 2018. Evaluation of the current contamination status of PFASs and OPFRs in South Korean tap water associated with its origin. Sci. Total Environ. 634, 1505–1512. https://doi.org/10.1016/j.scitotenv.2018.04.068.

Přibylová, P., Klánová, J., Holoubek, I., 2006. Screening of short- and medium-chain chlorinated paraffins in selected riverine sediments and sludge from the Czech Republic. Environ. Pollut. Soil and Sediment Remediation (SSR) 144, 248–254. https://doi.org/10.1016/j.envpol.2005.12.020.

PubChem. http://pubchem.ncbi.nlm.nih.gov.

Renner, 2002. The Kow controversy. Environ. Sci. Technol., 36 (21), pp. 410A–413A.

Shi, Y., Pan, Y., Wang, J., Cai, Y., 2012. Distribution of perfluorinated compounds in water, sediment, biota and floating plants in Baiyangdian Lake. China. J. Environ. Monit. 14, 636–642. https://doi.org/10.1039/C1EM10772K.

Stapleton, H.M., Letcher, R.J., Baker, J.E., 2004. Debromination of polybrominated diphenyl ether congeners BDE 99 and BDE 183 in the intestinal tract of the common carp (Cyprinus carpio). Environ. Sci. Technol. 38, 1054–1061. https://doi.org/10.1021/es0348804.

Sühring, R., Freese, M., Schneider, M., Schubert, S., Pohlmann, J.-D., Alaee, M., Wolschke, H., Hanel, R., Ebinghaus, R., Marohn, L., 2015. Maternal transfer of emerging brominated and chlorinated flame retardants in European eels. Sci. Total Environ. 530–531, 209–218. https://doi.org/10.1016/j.scitotenv.2015.05.094.

Sun, R., Luo, X., Tang, B., Chen, L., Liu, Y., Mai, B., 2017. Bioaccumulation of short chain chlorinated paraffins in a typical freshwater food web contaminated by e-waste in south china: Bioaccumulation factors, tissue distribution, and trophic transfer. Environ. Pollut. 222, 165–174. https://doi.org/10.1016/j.envpol.2016.12.060.

Svihlikova, V., Lankova, D., Poustka, J., Tomaniova, M., Hajslova, J., Pulkrabova, J., 2015. Perfluoroalkyl substances (PFASs) and other halogenated compounds in fish from the upper Labe River basin. Chemosphere, Per- and Polyfluorinated Alkyl substances (PFASs). Mater. Humans Environ. – Curr. Knowledge Sci. Gaps 129, 170–178. https://doi.org/10.1016/j.chemosphere.2014.09.096.

Thomas, A.M., 2005. Polybrominated diphenylether levels among united states residents: daily intake and risk of harm to the developing brain and reproductive organs. Integr. Environ. Assess. Manage. 1, 343–354.

UNEP C (1999) Guidelines for the identification of PCBs and materials containing PCBs, United Nations.

van Mourik, L.M., Gaus, C., Leonards, P.E.G., de Boer, J., 2016. Chlorinated paraffins in the environment: a review on their production, fate, levels and trends between 2010 and 2015. Chemosphere 155, 415–428. https://doi.org/10.1016/j.chemosphere.2016.04.037.

Wang, W., Choo, G., Cho, H.-S., Park, K., Shin, Y.-J., Oh, J.-E., 2018. The occurrence and distribution of hexabromocyclododecanes in freshwater systems, focusing on tissue-specific bioaccumulation in crucian carp. Sci. Total Environ. 635, 470–478. https://doi.org/10.1016/j.scitotenv.2018.03.262.

Wang, X., Zhong, W., Xiao, B., Liu, Q., Yang, L., Covaci, A., Zhu, L., 2019. Bioavailability and biomagnification of organophosphate esters in the food web of Taihu Lake, China: Impacts of chemical properties and metabolism. Environ. Int. 125, 25–32. https://doi.org/10.1016/j.envint.2019.01.018.

Wu, J.-P., Luo, X.-J., Zhang, Y., Luo, Y., Chen, S.-J., Mai, B.-X., Yang, Z.-Y., 2008. Bioaccumulation of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in wild aquatic species from an electronic waste (e-waste) recycling site in South China. Environ. Int. 34, 1109–1113. https://doi.org/10.1016/j.envint.2008.04.001.

Xu, J., Gao, Z., Xian, Q., Yu, H., Feng, J., 2009. Levels and distribution of polybrominated diphenyl ethers (PBDEs) in the freshwater environment surrounding a PBDE manufacturing plant in China. Environ. Pollut. 157, 1911–1916. https://doi.org/10.1016/j.envpol.2009.01.030.

Zeng, L., Wang, T., Wang, P., Liu, Q., Han, S., Yuan, B., Zhu, N., Wang, Y., Jiang, G., 2011. Distribution and trophic transfer of short-chain chlorinated paraffins in an aquatic ecosystem receiving effluents from a sewage treatment plant. Environ. Sci. Technol. 45, 5529–5535. https://doi.org/10.1021/es200895b.

Zeng, Y.-H., Luo, X.-J., Tang, B., Mai, B.-X., 2016. Habitat- and species-dependent accumulation of organohalogen pollutants in home-produced eggs from an electronic waste recycling site in South China: Levels, profiles, and human dietary exposure. Environ. Pollut. 216, 64–70. https://doi.org/10.1016/j.envpol.2016.05.039.

Zhao, P., Xia, X., Dong, J., Xia, N., Jiang, X., Li, Y., Zhu, Y., 2016. Short- and long-chain perfluoroalkyl substances in the water, suspended particulate matter, and surface sediment of a turbid river. Sci. Total Environ. 568, 57–65. https://doi.org/10.1016/j.scitotenv.2016.05.221.

Zhou, R., Zhu, L., Kong, Q., 2007. Persistent chlorinated pesticides in fish species from Qiantang River in East China. Chemosphere 68, 838–847. https://doi.org/10.1016/j.chemosphere.2007.02.021.