



## The distribution and accumulation of phosphate flame retardants (PFRs) in water environment



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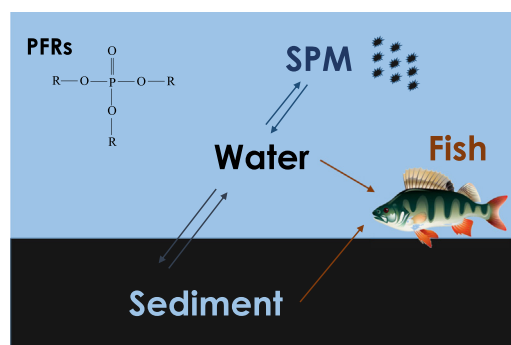
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### HIGHLIGHTS

- PFRs were measured in water, SPM, sediment, and fish samples.
- PFR levels in SPM and sediment were not influenced by organic carbon content.
- The chlorinated-PFRs were more abundant in sediment than water and SPM.
- The bioaccumulation of PFRs in fish was species-specific.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Phosphate flame retardants (PFRs) were measured in surface water ( $n = 11$ ), suspended particle matter (SPM,  $n = 11$ ), sediment ( $n = 11$ ), and fish samples ( $n = 26$ ) from the Pearl River Delta located in South China. Triethyl phosphate (TEP), tri(2-chloroethyl) phosphate (TCEP), tris(chloroisopropyl) phosphate (TCIPP), tri-*n*-butyl phosphate (TNBP), triphenyl phosphate (TPHP), and tricresyl phosphate (TMPP) were detected in more than half of surface water, SPM, and sediment samples. The median  $\Sigma$ PFR levels were 837 ng/L, 54.6 ng/g dry weight (dw), and 37.1 ng/g dw in surface water, SPM, and sediment samples, respectively. No significant correlations were found between the concentrations of most PFRs and organic carbon contents in SPM and sediment ( $p > 0.05$ ). In surface water samples, tris(2-butoxyethyl) phosphate (TBOEP, 27% of  $\Sigma$ PFRs) and TEP (23% of  $\Sigma$ PFRs) were the predominant chemicals, while TNBP (38% of  $\Sigma$ PFRs) and TCEP (32% of  $\Sigma$ PFRs) dominated in  $\Sigma$ PFRs in SPM samples, and TCEP (48% of  $\Sigma$ PFRs) and TCIPP (25% of  $\Sigma$ PFRs) dominated in  $\Sigma$ PFRs in sediment samples. The proportions of phenyl-PFRs and chlorinated-PFRs in  $\Sigma$ PFRs increased from surface water to SPM and sediment. The distribution ratios of PFRs between water and organic carbon in SPM (or observed  $K_{OC}$ ) were generally 2–3 orders of magnitude higher than the predicted  $K_{OC}$ . TNBP (nd–2.42 ng/g wet weight (ww)), TCEP (nd–4.96 ng/g ww), and TCIPP (nd–2.42 ng/g ww) were detected in 27%, 35%, and 23% of all fish samples, respectively. The log bioaccumulation factors (BAFs) ranged 2.56–2.78, 2.15–3.11, and 2.61–3.10 for TNBP, TCEP, and TCIPP, respectively. The biota-sediment accumulation factors (BSAFs) of TNBP, TCEP, and TCIPP were generally lower

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than 1 except for the BSAF of TCIPP in common carp. The results indicate the species-specific bioaccumulation of PFRs in fish species.

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## 1. Introduction

Phosphate flame retardants (PFRs) are additives widely applied in textiles, foams, plastics, and also electronic products (Van der Veen and de Boer, 2012; Wei et al., 2015). Some non-chlorinated PFRs are also used as plasticizers (Andresen et al., 2004). Polybrominated diphenyl ethers (PBDEs) used to be among the most commonly used FRs worldwide (de Wit, 2002). The Penta-BDE and Octa-BDE mixtures were banned because of their persistence, bioaccumulation, and toxicity in the environment and biota (UNEP, 2009). Deca-BDE was recently added in the list of persistent organic pollutants (POPs) by the Stockholm Convention (Stockholm Convention, 2017). Consequently, PFRs have been regarded as replacement of PBDEs. PFRs have been widely detected in water (Andresen et al., 2004; Cristale et al., 2013a, 2013b), sediment (Green et al., 2008; Kawagoshi et al., 1999; Zeng et al., 2014), indoor dust (Cao et al., 2014), wildlife (Brandsma et al., 2015; Giulivo et al., 2017), and human beings (Meeker et al., 2013). Some researchers have observed comparable or even higher concentrations of PFRs compared with PBDE concentrations in indoor dust (Cao et al., 2014; Zheng et al., 2015). Moreover, these flame retardants (FRs) are suspected to be toxicants (Meeker et al., 2013; Van der Veen and de Boer, 2012; WHO, 1998). Higher levels of tris(1,3-dichloroisopropyl) phosphate (TDCIPP) in house dust might be related with altered hormone levels and lower semen quality in males (Meeker and Stapleton, 2010). Several PFRs, including tri(2-chloroethyl) phosphate (TCEP), TDCIPP, tris(chloroisopropyl) phosphate (TCIPP), and tris(2-butoxyethyl) phosphate (TBOEP) were suspected to be carcinogenic (WHO, 1998). TDCIPP, TBOEP, and triphenyl phosphate (TPHP) exhibited *in vitro* estrogenic and anti-androgenic effect on human osteosarcoma cell line exposed to indoor dust extracts (Suzuki et al., 2013). Overall, PFRs could pose potential toxicity to organisms, which raised concern about the adverse effects of PFRs on wildlife and humans (Meeker et al., 2013; WHO, 1998).

The occurrence of PFRs in the water environment is of major concern due to their potential toxic and deleterious effects on biota. Many studies have detected PFRs in water, sediment, and fish worldwide (Van der Veen and de Boer, 2012; Wei et al., 2015). Nevertheless, little studies have investigated the migration and distribution of PFRs in water environment, such as the partition of PFRs in water and particulate matters, and the accumulation from water to organisms (Iqbal et al., 2017). Cao et al. (2017) estimated the partition of PFRs between particles and pore water of sediment using an equilibrium model. The targeted PFRs were predicted to largely vary in their distribution in pore water and particles (Cao et al., 2017). In previous studies, concentrations of PFRs were generally lower than those of persistent organic pollutants (POPs), such as PBDEs and polychlorinated biphenyls (PCBs) in fish species (Henríquez-Hernández et al., 2017). PFRs also had lower biota to sediment accumulation factors (BSAFs) than PBDEs (Giulivo et al., 2017). Brandsma et al. (2015) reported that TPHP, tricresyl phosphate (TMPP), and TDCIPP showed trophic dilution in an estuarine food web. The distribution and bioaccumulation of PFRs in water environment still warrants more specific data to verify.

The Pearl River Delta (PRD) is among the most developed and urbanized regions in China. Because of the extensive manufacturing industry and large populations, there is widespread use of large amounts of flame retardants and plasticizers such as PFRs in this region. A recent study reported PFR concentrations of 8.3 to 470 ng/g in sediments from the Pearl River Delta region (Tan et al., 2016). The e-waste recycling, industry activities, and sewage discharge were suspected as

sources of PFRs in PRD region (Tan et al., 2016). Zeng et al. (2014) reported that total concentrations of PFRs ranged from 96.7 µg/kg to 1312.9 µg/kg dry weight in wastewater treatment plant (WWTP) sludge in PRD. After the discharge of sewage, PFRs could diffuse in Pearl River and be absorbed to particles in water environment. PFRs have a wide range of organic carbon-water partition coefficient ( $K_{OC}$ ) values, indicating that the partition of PFRs between water and particles was highly variable. The different affinity of PFRs to suspended particles and sediments would influence the bioavailability and the subsequent toxicological effects of PFRs in aquatic organisms.

In the present study, we analyzed PFRs in paired surface water, suspended particulate matter (SPM), and sediment samples adjacent to the main WWTPs located in the Pearl River Delta. Several fish species were also collected to monitor PFRs during the sampling campaign. The compositions of PFRs in surface water, SPM, and sediment samples were explored to know the distributions of PFRs between water, SPM, and sediment in water environment. We also aimed to assess the bioaccumulation potential of PFRs in different fish species in comparison with previous studies.

## 2. Materials and methods

### 2.1. Sample collection

The surface water ( $n = 11$ ), SPM ( $n = 11$ ), and sediment ( $n = 11$ ) samples were collected from 11 sites, which were <1 km away from the outfalls of WWTPs located in Pearl River Delta (Fig. 1). The WWTPs mainly dealt with the domestic sewage from the biggest cities in PRD region, such as Guangzhou, Shenzhen, and Dongguan. The flux and sources of sewage were given in Table S1, Supplementary material. Surface water samples were collected at 0–1 m depth and stored in individual 4 L pre-cleaned glass bottles. After transported to the laboratory within 8 h, the SPM was collected by filtering water sample through the glass fiber filters (Whatman GF/F, 0.7 µm effective pore sizes, pre-combusted at 450 °C for 4 h). The filter was weighted to calculate the value of total suspended solids (TSS) in surface water. The filters were then placed in pre-cleaned glass dishes and wrapped with aluminum foil, stored under  $-20$  °C until further analysis. The filtered water was then stored at 5 °C until further analysis.

Fish samples ( $n = 26$ ) were collected from Site 4. The fish species included catfish (*Clarias batrachus*,  $n = 6$ ), tilapia (*Oreochromis mossambicus*,  $n = 5$ ), Common Carp (*Cyprinus carpio*,  $n = 2$ ), bream (*Parabramis pekinensis*,  $n = 3$ ), white semiknife-carp (*Hemiculter leucisculus*,  $n = 6$ ), and silver carp (*Hypophthalmichthys molitrix*,  $n = 4$ ). The sample information, feeding habit, and habitat of fish species were provided in Table S2, Supplementary material. The fishes were dissected and the muscle samples were collected. Fish muscle and sediment samples were weighted and lyophilized, and then stored under  $-20$  °C until further analysis.

### 2.2. Sample preparation and analysis

The surface water samples were extracted and purified within 24 h post-sampling. Each filtered water sample was pretreated by the automatic solid phase extractor (AQUA Trace@ASPE 799, SHIMADUZ-GL) after spiked with surrogate standard (100 ng of triphenyl phosphate- $d_{15}$  (TPHP- $d_{15}$ )). The cartridges (Oasis HLB, 6 mL/200 mg; Waters, Milford, MA, USA) were sequentially conditioned with 5 mL dichloromethane, 5 mL ethyl acetate, 10 mL methanol, and 10 mL ultrapure water.

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