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Q1 Distribution characteristics of poly- and perfluoroalkyl 2 substances in the Yangtze River Delta

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A B S T R A C T

In this work, a method was developed and optimized for the analysis of polyfluoroalkyl 17
 and/or perfluoroalkyl substances (PFASs) content in surface water and sediment samples 18
 with high instrumental response and good separation. Surface water and sediment samples 19
 were collected from the Yangtze River Delta (YRD) to analyze the distribution characteristics of 20
 perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkane sulfonic acids (PFASs), perfluoroalkyl 21
 phosphonic acids (PFPA), perfluoroalkyl phosphinic acids (PFPIAs), and polyfluoroalkyl 22
 phosphoric acid diesters (diPAPs). The results showed that the total concentrations of PFCAs 23
 and PFASs in YRD varied from 31 to 902 ng/L. PFCAs (≥ 11 carbons) and PFASs (≥ 10 carbons 24
 atoms) were not detected in any surface water samples. The mean concentrations of all 25
 PFCAs and PFASs in surface water from the sampling areas decreased in the following order: 26
 Yangtze river (191 ng/L) \approx Taihu lake (189 ng/L) $>$ Huangpu river (122 ng/L) \approx Qiantang river 27
 (120 ng/L) $>$ Jiaxing urban river (100 ng/L). Strong significant ($p < 0.05$) correlations between 28
 the concentrations of many of the compounds were found in the sampling areas, suggesting a 29
 common source for these compounds. Only perfluorooctanoic acid (PFOA) was observed in all 30
 sediment samples, at concentrations varying from 0.02 to 1.35 ng/g. Finally, detection rates of 31
 two diPAPs were only 8% and 10%, respectively and the concentration of diPAPs was two to 32
 three times lower compared to PFCAs and PFASs. 33

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43 44 Introduction

48 Polyfluoroalkyl and/or perfluoroalkyl substances (PFASs) are
 49 anthropogenic chemicals with a fluoroalkyl backbone (F (CF₂)_x)
 50 and a polar head group (i.e., sulfonic acid, carboxylic acid, or
 51 phosphonic acid). This particular molecular structure imparts
 52 oleophobic and hydrophobic properties to these chemicals
 53 (Kissa, 2001). PFASs have extensive applications in industrial
 54 and commercial products, including in non-stick, grease-
 55 proofing and surface treatment applications due to their high

surface activity and ability to repel water, oil, and stains. Their 56
 extensive applications and ability to resist biological and 57
 environmental degradation have led to the ubiquitous presence 58
 of perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane 59
 sulfonic acids (PFASs) in environmental samples, including 60
 surface water (De Silva et al., 2011; D'eon et al., 2009a; Hansen 61
 et al., 2002; Mak et al., 2009), groundwater (Moody et al., 2003; 62
 Murakami et al., 2009; Schultz et al., 2004), sea water (So et al., 63
 2004; Yamashita et al., 2005), sediments (Bečanová et al., 2016; 64
 Pan et al., 2014), soil (Li et al., 2010), air (Sinclair et al., 2007), and 65

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dust (Haug et al., 2011; Kubwabo et al., 2005; Shoeib et al., 2005), as well as in human sera (Hansen et al., 2001; Hölzer et al., 2008; Houde et al., 2006; Vestergren and Cousins, 2009) and animal sera and livers (Ahrens et al., 2009; Houde et al., 2006; Lau et al., 2007; Martin et al., 2003). Previous studies have assessed the distribution, transport, fate and sources of PFCAs and PFSAs, demonstrating the biotransformation of precursors into PFCAs and PFOS in microbial and soil systems (Russell et al., 2008; Wang et al., 2011), as well as *in vitro* (Martin et al., 2005; Nabb et al., 2007; Tomy et al., 2004; Benskin et al., 2009) and *in vivo* animal models (D'eon and Mabury, 2007, 2010; Fasano et al., 2009).

Recently, more types of PFASs have been detected in environmental samples, including perfluoroalkyl phosphonic acids (PFPA), perfluoroalkyl phosphinic acids (PFPIAs), polyfluoroalkyl phosphoric acid monoesters (monoPAPs), and polyfluoroalkyl phosphoric acid diesters (diPAPs), among others. It was reported that PFPA and PFPIAs were in the list of high production volume perfluoroalkyl acids (4500–227,000 kg/year) in 1998 and 2002 (Howard and Meylan, 2009). These compounds were commonly used as defoaming agents in pesticide formulations in USA (Heid et al., 1975) until their application was banned in 2008 (US EPA, 2006). The first report of PFPA and PFPIA detection in environmental samples was from Canada, where PFPA was observed in 80% of surface water samples at concentrations in the picogram to low nanogram per liter range and in six of the seven waste water treatment plants (WWTPs) effluent samples assessed (D'eon et al., 2009a). Perfluorooctyl phosphonic acid (C8-PFPA) was also detected at a concentration of 1 ng/L in Dutch surface water samples (Esparza et al., 2011). In lake trout collected from Lake Ontario, PFPIAs were identified at concentrations of one to two orders of magnitude lower than those of PFCAs and PFSAs (Guo et al., 2012). Human exposure to PFPIAs was confirmed in USA, where 6:6 PFPIA and 6:8 PFPIA were found in human sera at a concentration range of 4–38 ng/L (Lee and Mabury, 2011). In another study, PFPA (C6, C8) and PFPIAs (C6/C6, C6/C8, and C8/C8) were also detected in human sera in Germany and China (Yeung and Mabury, 2016).

Another class of PFASs, polyfluoroalkyl phosphoric acid esters (PAPs), are not only used as grease-proofing agents in food contact paper, but may also be found in cosmetics, hair and personal care products, floor waxes, paints and finishes, and cleaning fluids (US FDA, 2003). DiPAPs have been detected in Canadian WWTP sludge and paper fiber extracts at concentrations ranging from 47 ± 22 to 200 ± 130 ng/g and 34 ± 30 to 2200 ± 400 ng/g, respectively (D'eon et al., 2009b). Further, high concentrations of diPAPs (up to 7000 ng/g) were reported in dust samples collected from Canada, Faroe Islands, Sweden, Greece, Spain, Nepal, Japan, and Australia (De Silva et al., 2012; Eriksson and Karrman, 2015). Direct evidence of human exposure to diPAPs was obtained by the detection of diPAPs in human sera collected in USA in sub part-per-billion (ppb) to ppb levels (D'eon et al., 2009b; Lee and Mabury, 2011). Additionally, studies have demonstrated the degradation of diPAPs to PFCAs in WWTPs (Lee and Mabury, 2011) and their biotransformation in rats (D'eon and Mabury, 2007). Therefore, diPAPs are important both as a precursor to PFCAs and potentially as a fluorinated contaminant on their own. As PAPs are primarily used in food contact paper products, mono-,

di- and tri-substituted PAPs have been found in food packaging materials in the Swedish market, with up to nine congeners of diPAPs being detected in food samples (0.9 to 36 pg/g) (Gebbink et al., 2013). These results indicate that consumption of food packed in PAP-containing materials is an indirect source of human exposure to PFCAs. Nevertheless, compared to PFCAs and PFSAs, little is known about the environmental occurrence and fate of PFPA and diPAPs, particularly in China.

The Yangtze River Delta (YRD) region, with a population of 150 million people, has experienced rapid economic growth in the past three decades. It is one of the most economically dynamic and wealthy regions in China, with the highest degree of openness and innovation. The region includes several metropolises like Shanghai and the capital cities of Hangzhou, Suzhou, and Nanjing as well as medium-sized cities like Ningbo and Wuxi, among others. The chemical engineering, textile, and paper making industries are important components of the YRD industry, all of which are potential sources of PFASs. The Yangtze, Huangpu, and Qiantang rivers as well as the Taihu lake and Jiaying urban river, all of which are located in the YRD, are important drinking water sources for the region.

The consumption of PFAS contaminated drinking water or fish may pose a health risk to aquatic organisms, wildlife, and humans. Thus, the objectives of this study were to investigate the contamination profiles of 23 PFASs (11 PFCAs, 4 PFSAs, 3 PFPA, 3 PFPIAs and 2 diPAPs) in water and sediment samples collected from the middle and lower reaches of the Yangtze, Huangpu, and Qiantang rivers, Jiaying urban river and Gonghu region of Taihu lake; as well as to explore the factors influencing PFAS distribution between water and sediment.

1. Materials and methods

1.1. Chemicals and reagents

The target analytes included PFCAs (C4–C14), PFSAs (C4, C6, C8, and C10), PFPA (C6, C8, and C10), PFPIAs (C6/C6, C8/C8, and C6/C8), and diPAPs (6:2 and 8:2). The category, name and acronym of all the analytes are shown in Table 1. Mass-labeled PFASs standards include perfluoro-*n*- $^{13}\text{C}_4$]butanoic acid ($^{13}\text{C}_4$ -PFBA), perfluoro-*n*- $^{13}\text{C}_2$]hexanoic acid ($^{13}\text{C}_2$ -PFHxA), perfluoro-*n*- $^{13}\text{C}_4$]octanoic acid ($^{13}\text{C}_4$ -PFOA), perfluoro-*n*- $^{13}\text{C}_5$]nonanoic acid ($^{13}\text{C}_5$ -PFNA), perfluoro-*n*- $^{13}\text{C}_2$]decanoic acid ($^{13}\text{C}_2$ -PFDA), perfluoro-*n*- $^{13}\text{C}_2$]undecanoic acid ($^{13}\text{C}_2$ -PFUnDA), perfluoro-*n*- $^{13}\text{C}_2$]dodecanoic acid ($^{13}\text{C}_2$ -PFDoA), sodium perfluorohexane- $^{18}\text{O}_2$]sulfonate ($^{18}\text{O}_2$ -PFHxS), sodium perfluoro- $^{18}\text{O}_2$]octanesulfonate ($^{18}\text{O}_2$ -PFOS), sodium bis(1H,1H,2H,2H- $^{13}\text{C}_2$]perfluorooctyl)phosphate ($^{13}\text{C}_4$ -6:2 diPAP) and sodium bis(1H,1H,2H,2H- $^{13}\text{C}_2$]perfluorodecyl)phosphate ($^{13}\text{C}_4$ -8:2 diPAP). All native and mass labeled standards were purchased from Wellington Laboratories (Guelph, Canada). Liquid chromatogram-mass spectrum (LC-MS) grade methanol, acetonitrile, methyl-*tert*-butyl ether (MTBE), and ammonium acetate were purchased from Fisher Scientific (Fair Lawn, NJ, USA). Analytical grade sodium hydroxide (NaOH) was purchased from Sinopharm Chemical Reagent, Beijing, Co., Ltd., China. Tetra-butyl ammonium hydrogen sulfate (TBAS) was purchased from J.T. Baker (Phillipsburg, NJ, USA). Ottawa sand was purchased from Fisher Scientific (Pittsburgh, PA, USA). Oasis® weak anion

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