



Perfluorinated compounds in surface waters of Shanghai, China: Source analysis and risk assessment



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ABSTRACT

17 perfluorinated compounds (PFCs) were systematically investigated in the surface water from principal watersheds of Shanghai, China. 10 PFCs were above the detection limit (0.08–0.28 ng/L) in 39 surface water samples. The perfluorooctanoic acid (PFOA) and perfluorobutanesulfonate (PFBS) were the two dominant compounds with a median concentration 50.67 ng/L and 29.84 ng/L, respectively. Concentrations of perfluorooctanesulfonate (PFOS) were generally less than PFBS, which might result from the global phase-out of PFOS production and the use of PFBS as a substitute for PFOS-based products. There were three major polluted areas of PFOA along the Huangpu River. The PFOA concentration in groundwater samples collected from one of the three areas indicated that chemical industry might be the possible source. The perfluoroalkane sulfonates (PFASs) level had a spatial trend that indicated northwest had higher concentrations than the southeast. The distribution of PFCs was not much affected by atmospheric deposition. Mass loading analysis in the surface water revealed that the Huangpu River exhibited relatively large mass loading of total PFCs of 1742.43 kg/year to Yangtze River Estuary. The predominant of the PFC species was PFOA with 652.65 kg/year. The current concentrations of PFOA and PFOS were at middle level comparing to other studies in China and worldwide. Risk assessment of 6 PFCs showed that there is no risk to the aquatic organisms in Shanghai. PFOS and PFBS had low risk to the avian. Furthermore, the adults living in Shanghai were at low risk to exposure to PFCs through water consumption.

1. Introduction

Perfluorinated compounds (PFCs), such as perfluoroalkyl carboxylates (PFCAs) and perfluoroalkane sulfonates (PFASs), are synthetic chemicals that have been manufactured and commonly used in our daily life during the last half century (Lindstrom et al., 2011). Owing to their unique physicochemical properties such as the ability to reduce surface tension, the stability, and the hydrophobic and hydrophilic properties, PFCs were applied as nonstick coating in cookware and food packaging, water and stain repellants, lubricants, firefighting foams, and pesticide formulations (Route et al., 2014). Because of their widespread application and environmental persistence, they have resulted in global occurrence in the air (Ge et al., 2017), water (Kwadijk et al., 2010), soil (Chen et al., 2016), sediment (Yan et al., 2015), indoor dust (Shoeib et al., 2005), wild life (Surma et al., 2015) and even the polar ice sheet (MacInnis et al., 2017). Lots of toxicological studies have been conducted on PFCs, most of which were associated with many health risks, such as suppression of immunity (DeWitt et al., 2008; Corsini et al., 2014), disrupting thyroid functions (Ren et al.,

2015), neurotoxicity (Johansson et al., 2009; Viberg et al., 2013), and spermatogenesis influence (Louis et al., 2015). Considering their extreme persistence, ubiquitous distribution and high bioaccumulative potential (Olsen et al., 2007; Houde et al., 2011), the requirement of continual monitoring of PFCs is of great significance.

Among all the PFCs, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonates (PFOS) were the two most often observed and reported PFCs due to their abundant production and wide use (Gong et al., 2016; Liu et al., 2017). Continuous concerns over their production, use and potential hazards to humans led to restrictions and bans. In May 2009, PFOS and its salts, perfluorooctane sulfonyl fluoride (PFOSF), have been listed in Annex B of the Stockholm Convention (Munoz et al., 2015). Then shorter-chain PFCs (carbon chain length less than seven) were increasingly used since they have less toxic potency than the C8 PFCs (Buhrke et al., 2013; Z et al., 2015). Manufacturers in China began large scale production in 2003 at the advent of 3 M's 2001 global PFOS phase-out to accommodate both domestic and export demands (Lu et al., 2011). With mass production activities, PFCs were inevitably regarded as the significant contaminants in China.

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Aquatic system was considered as the most important way for PFCs when they transfer into the surroundings. Occurrences of PFCs in water have also been frequently reported worldwide (Saito et al., 2004; Loos et al., 2008; Tan et al., 2014; Zhang et al., 2015; Ahrens et al., 2016; Nguyen et al., 2016). These environmental monitoring attempts could be helpful to understand the source, distribution and environmental behavior of PFCs in the aquatic system. Generally, PFCs were transported into the aqueous environment via discharge from wastewater treatment plants or fluorochemical industries, domestic consumption and atmospheric deposition (Liu et al., 2017).

Shanghai is a typical plain crisscrossed by surface water that is very important to the development of the city and it is related to the quality of drinking water. Because most of the rivers are connected with each other, it could easily lead to cross-contamination. Meanwhile, previous studies showed that occurrence of PFCs was strongly correlated with population density and highly influenced by urban activities (Zhang et al., 2016). As a global financial center and a transport hub with one of the largest populations in the world, industrial and residential areas are complex and mixed in Shanghai. This urban structure could cause unexpected pollution to the environment. The existence of PFCs in water could potentially endanger aquatic organism, which might finally harm the human health. Until now, only a few surveys have addressed PFCs environmental contamination of surface water in Shanghai with scattered sampling sites (So et al., 2007; Lu et al., 2011; Z et al., 2015). These results indicated that aquatic system of Shanghai was heavily contaminated by PFCs. However, most of these researches were based on a large-span study, such as Yangtze River Delta or eastern China, which was not enough to illustrate the pollution characteristics and the source of PFCs in Shanghai.

In this paper, 39 surface water samples and 3 groundwater samples were collected from the major watersheds in Shanghai to characterize the source and risk assessment of PFCs. The sampling sites were at high spatial resolution which could display the pollution status and source analysis more detailed. The possible sources of PFCs in surface waters were identified, and the mass loadings were evaluated. Furthermore, the potential risks posed by PFCs to aquatic organisms, wildlife and adults living in Shanghai were evaluated.

2. Materials and methods

2.1. Chemicals and reagents

The detail descriptions on chemicals and their abbreviations were available in [Supporting information \(SI\)](#)

2.2. Site description and sampling

The biggest and uppermost river in Shanghai is Huangpu River, which is also one of the branches of the Yangtze River with 113 km long flowing through the city. Parts of the Huangpu River are used as drinking water reservoir areas. Due to the rapid development of industrialization and the growth of population, Shanghai is faced with serious environmental problems in the last few decades, especially in the aquatic systems. In order to comprehensively understand the occurrences and sources of PFCs in surface waters in Shanghai, samples were collected from principal watersheds including Huangpu, Suzhou, Yunzao, Chuanyang, Dianpu, Dazhi, Jinhui, Yexie, Maogang, Xietang, Taipu Rivers and Dianshan Lake. There was one sampling point at least before and after each junction. The sample sites were shown in the [Fig. 1](#). Every sampling site location was recorded by GPS (Garmin Ltd., USA). The detailed information was in [Table S1 in the SI](#).

Surface water samples were collected from November to December 2015. Grab water samples were drawn from the surface using a tinsplate bucket, from the middle of the river width at 0.5 m depths. Groundwater samples were collected from shallow aquifer from three 5–7 m wells in the area which was formerly used as chemical plants by



Fig. 1. The specific locations of the sampling sites in principal watersheds in Shanghai. (▲ Surface Water ▲ Groundwater).

Bailer's tube. Duplicated samples that were collected from each site, consisted of 1 L water in separated polyethylene terephthalate bottles. Before sample collection, all instruments were thoroughly pre-rinsed with methanol and ultrapure water at laboratory, and then rinsed with water sample prior to sample collection. The water samples were delivered to the laboratory immediately after collection, then stored at 4 °C for up to 1 week until analysis.

2.3. Analytical procedure

All water samples were previously filtered with rapid qualitative filter paper to remove large particles and then filtered with GF/F (Whatman, 0.7 μm, 47 mm) glass fiber filter, then the filtrates were concentrated by solid phase extraction (SPE) process. Replicate water samples were extracted for each sampling site. The SPE procedure was referenced from previous reported methods (Taniyasu et al., 2005, 2008; Pan et al., 2014) with a little modification. Briefly, each sample (250 mL) was firstly fortified with 5 ng (100 μL, 50 ng/mL) of an internal standard mixture (¹³C₄-PFOA, ¹³C₄-PFOS). SPE was performed with CNW PWAX Cartridges (6 mL, 150 mg) on a 12-position vacuum manifold (CNW, China). The cartridge was pre-conditioned by passing 4 mL 0.1% NH₄OH in methanol, 4 mL methanol and 4 mL ultrapure water in gravitational drops. Then, the sample was loaded onto the wet cartridges at approximately 2–3 drops per second. After loading, the cartridges were rinsed with 4 mL 25 mmol/L ammonium acetate buffer (pH = 6–7) and 4 mL methanol. The next step was to completely remove the remaining water for half an hour by the vacuum. After that, it was necessary to sequentially elute the cartridge by gravity with 4 mL 0.1% NH₄OH in methanol. The extract was evaporated to dryness under nitrogen before being reconstituted in 1 mL of methanol. Finally, the sample was filtered through a 0.45 μm nylon mesh filter into 1 mL polypropylene (PP) vial with polyethylene (PE) cap before injection.

The extracts were analyzed via a modified HPLC system interfaced to an Agilent 6460 A Triple Quadrupole (QQQ) mass spectrometer (Santa Clara, CA, USA) operated with electrospray ionization (ESI) in negative mode. Details of HPLC and mass spectrometer parameters for the instrumental analysis were reported in our former study (Wu et al., 2016).

2.4. Data analysis

Quality assurance and quality control were detailed in the [SI](#). All statistical analyses were performed using SPSS version 18.0 and Origin 8.0. Spatial distributions of PFCs were plotted on a contour map using Surfer 13.0 (Golden Software, Inc., USA). Probabilistic analysis (Monte

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