



Occurrence and Risk Assessment of per- and Polyfluoroalkyl Substances in Water Source Protection Area of Southeastern China

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In order to clarify the pollution characteristics and human health risks of PFASs pollutants in typical drinking water sources in Zhejiang Province, this study relies on ultra-performance liquid chromatography-mass spectrometry (UPLC-MS/MS) technology to analyze the pollution of 26 PFASs in 7 reservoirs in Zhejiang Province. The detected concentrations of PFASs were evaluated to further assess the human health risks. Total PFASs concentrations in the seven reservoirs ranged from 1.30 ng L⁻¹–24.90 ng L⁻¹. Among the 26 PFASs pollutants analyzed, PFOA and PFBA were the main PFASs pollutants, the detected concentrations of PFOA and PFBS ranging from 0.50 ng L⁻¹–13.70 ng L⁻¹ and 0 ng L⁻¹–1.70 ng L⁻¹, respectively. Then we evaluated 15 PFASs and calculated the results of the HQ value of the reproductive toxicity and hepatotoxicity of the total PFASs in this study ranged from 2.30 × 10⁻⁸ to 1.16 × 10⁻⁴ and 9 × 10⁻⁸ to 5.24 × 10⁻⁴ respectively, which were both lower than 0.01, indicating that there is no significant risk to the human body.

Keywords: detection, hazard quotient, Pafs, risk assessment, reservoir

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INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) are widely used in industrial and consumer products, including food packaging, metal plating materials, aqueous film-forming foams (AFFFs), textile coatings, and nonstick coatings, due to their unique amphiphilic nature and high chemical stability (strong C-F bonding energy) (Knutsen et al., 2019; Li et al., 2020). Moreover, many PFASs are also bioaccumulative and toxic (Lau et al., 2007). Many researchers have detected their presence in the aqueous environment, sediment, soil, and atmosphere (Ahrens et al., 2010; Li et al., 2011; Yang et al., 2011; Wang et al., 2015; Hu et al., 2019; Park et al., 2020; Xu et al., 2021), and they have even been reported in human blood and serum (Poothong et al., 2020). Perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFASs), such as perfluorooctanesulfonate (PFOS) and perfluorooctanoic acid (PFOA), have been found to be persistent in the environment and in living organisms and are difficult to remove by conventional drinking water treatment techniques (e.g., coagulation, filtration, and oxidation) (Rahman et al., 2014). PFASs can enter the human body through drinking water, which is considered to be one of the main exposure pathways for PFASs in the general population (Jian et al., 2018). High contamination (>100 ng L⁻¹) of PFASs in source water and drinking water has been found in many countries (Gebbink et al., 2017; Guelfo et al., 2018;

Kaboré et al., 2018; Liu et al., 2021). It has been shown that different PFASs show different toxicological profiles, including hepatotoxicity, reproductive and developmental toxicity, immune system toxicity, thyroid toxicity, neurotoxicity, cardiovascular toxicity, and endocrine toxicity (DeWitt, 2015).

Drinking water has been identified as a substantial source of PFAS exposure for many populations, particularly those living near contaminated sites (Hu et al., 2016; Banzhaf et al., 2017). Next to contaminated sites, drinking water has been reported to account for up to 75% of total PFAS exposure (Vestergren and Cousins, 2009; Hoffman et al., 2011). In recent years, several health-protective guidelines have been proposed, such as the United States Environmental Protection Agency (U.S. EPA) proposed a lifetime health advisory level for PFOS + PFOA of 70 ng L⁻¹ in drinking water in 2016 (U.S. EPA, 2016). In 2018, the Agency for Toxic Substances and Disease Registry (ATSDR) in the US further lowered the Minimum Risk Levels (MRLs) for PFOS and PFOA by approximately an order of magnitude compared to the reference dose (RfD) used by the U.S. EPA to develop the 2016 lifetime advisory (ATSDR, 2018). Despite industry has rapidly replaced PFOS and PFOA with shorter chain length PFASs and new chemicals due to these regulatory interventions, these chemicals are difficult to detect by using standard methods (Wang et al., 2017). Emerging evidence from animal experiments suggests some of these alternative PFASs can be equally hazardous (Gomis et al., 2018). Currently, most studies focus only on PFASs in rivers and water plants, and limited work has been done on PFASs in reservoirs. Given this, and the fact that reservoir water, as the front end of the drinking water supply chain, is an important part of ensuring water safety for the population, it is necessary to investigate the concentration levels and human health risks of PFASs contaminants in reservoir water.

In this study, seven reservoirs in a region of Zhejiang Province, China, were used as target areas, and 4–11 sampling sites were set up in each reservoir, for a total of 42 sampling sites, to investigate the distribution and concentrations of different PFASs contaminants in the region. An ultra-performance liquid chromatography-mass spectrometry (UPLC-MS/MS) was used to detect and analyze 26 PFASs pollutants, and the hazard quotient (HQ) modeling system was used to assess the human health risk of the target PFASs pollutants in the region. This work is expected to provide basic data and theoretical support for the potential risk of PFASs pollutant levels in urban reservoirs.

MATERIALS AND METHODS

Materials and Reagents

Analytical standards for 26 natural PFASs were purchased from Wellington Laboratories Inc. (Canada). The 26 natural PFASs include 18 PFCAs and eight PFASs. All stock solutions were prepared in methanol and stored at 4 C in polypropylene (PP) tubes. Other reagents included methanol for UPLC analysis, ≥ 99.9% (Sigma-Aldrich, USA), ammonium hydroxide (25%, ACOS Organics, USA), and ammonium acetate (CNW,

Germany). The solid phase extraction (SPE) column was purchased from Waters (6 ml, 150 mg, Oasis® WAX).

Sampling Campaign

The sampling activities in this study were conducted in June 2021 in the province of Zhejiang. A total of 42 water samples were collected in seven reservoirs from A to G. From four to eleven sampling sites were set up in each reservoir. Samples were collected from 0.5 to 1 m below the water surface using stainless steel buckets and stored in PP bottles (2 L). Samplers and collection containers were washed with methanol as well as the water column at the sampling sites, respectively, prior to sample collection. Water samples were transported to the laboratory at low temperature, pretreated within 2 days, and stored in cold storage at -20 C prior to pretreatment.

Sample Extraction

The protocol for the extraction of PFASs from water samples is based on ISO-25101. Briefly, the pH of unfiltered water samples (500 ml) is adjusted to 3 with acetic acid. The samples were then spiked with 5 ng of alternative standards using an Oasis® WAX SPE column. Extracted fractions were collected in PP medium tubes and then concentrated to 1 ml under a gentle nitrogen stream and prepared for injection into UPLC-MS/MS. Information on instrumental parameters is listed in **Supplementary Table S1**.

The main work of the extraction was divided into three parts: filtration, SPE, and nitrogen blowing concentration. 1) Measure 500 ml of water sample in a PP beaker and add 5 ng of internal standard. Using a circulating water vacuum pump and filtration device, the mixed solution was passed through a 0.45 μm filter membrane to separate the aqueous phase from the suspended particulate phase. In order to avoid the interference of the membrane and other organic components on the quantitative results, the membrane used in the experiment was preheated and dried to a constant weight in an electric blast dryer. 2) A WAX SPE column (6 ml, 150 mg) was used for the SPE. The solid-phase extraction process consisted of activation of the column, sample loading, solid-phase extraction, and elution of the target compounds. The SPE column was activated with 4 ml of 0.1% ammonia-methanol mixture, 4 ml of methanol solution and 4 ml of high purity water in turn. After activation of the column, the water sample was flowed through the column at a rate of six to eight ml min⁻¹. After all the target mixture flowed through the column, the column was washed with 4 ml of 25 mM ammonium acetate solution to clean any impurities other than the target compounds. After that, the SPE device and the column were dried under vacuum for 10 min using a vacuum pump, and the target compounds were eluted with 5 ml of methanol solution and 5 ml of 0.1% ammonia-methanol solution in turn, and the eluate was collected in a 15 ml centrifuge tube. 3) The eluate was concentrated by 12-well nitrogen blowing apparatus with 99.7% purity of nitrogen, and the water bath temperature was set at 40 C during the nitrogen blowing process. The auto-sampling vial was stored under refrigeration and protected from light.

TABLE 1 | Concentrations of PFASs in 7 reservoirs (unit: ng L⁻¹).

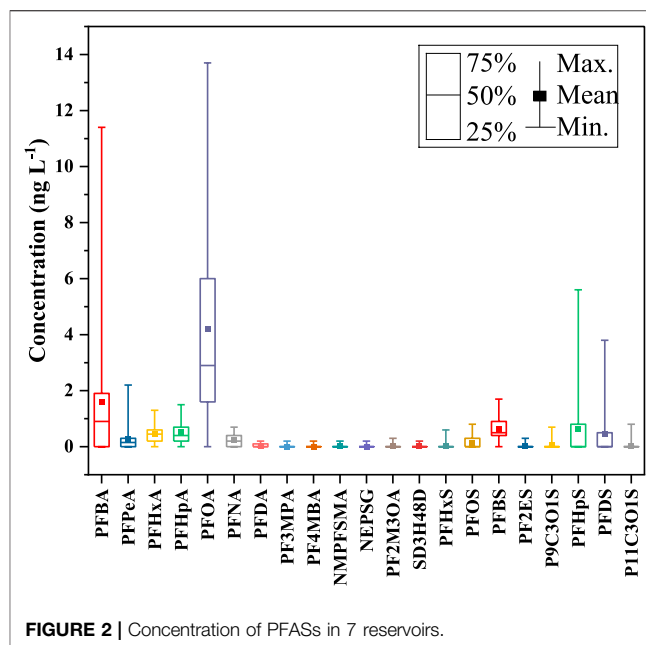
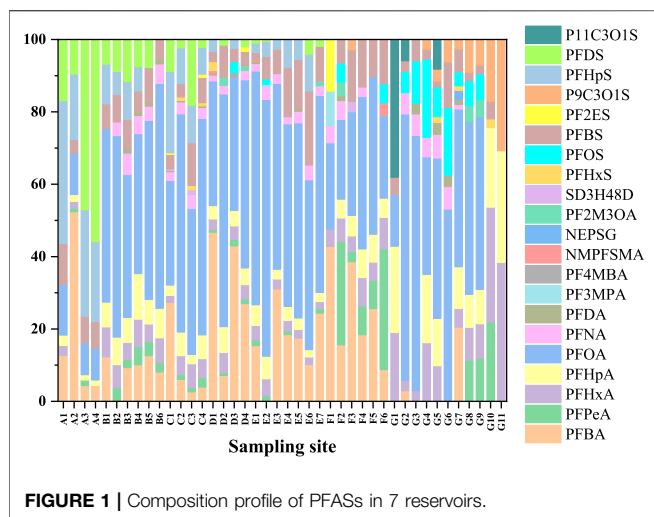
	A			B			C			D			E			F			G		
	Max	Min	Avg	Max	Min	Avg	Max	Min	Avg	Max	Min	Avg	Max	Min	Avg	Max	Min	Avg	Max	Min	Avg
PFBA	5.50	0.30	1.75	1.30	<LOQ	0.80	6.80	0.20	2.30	11.40	0.90	5.85	3.80	<LOQ	1.96	2.70	0.30	1.17	1.60	<LOQ	0.15
PFPeA	0.10	<LOQ	0.05	0.60	<LOQ	0.28	0.60	<LOQ	0.25	0.30	0.10	0.20	0.30	<LOQ	0.13	2.20	<LOQ	0.82	0.90	<LOQ	0.17
PFHxA	0.20	<LOQ	0.10	0.90	0.40	0.57	1.20	0.50	0.83	0.70	0.60	0.65	0.90	0.10	0.41	0.50	0.10	0.32	1.30	<LOQ	0.44
PFHpA	0.20	0.10	0.15	1.50	0.50	0.75	1.50	0.20	0.95	0.90	0.70	0.80	1.20	0.10	0.54	0.80	<LOQ	0.28	0.90	<LOQ	0.45
PFOA	1.20	0.60	0.85	5.10	3.40	4.27	13.70	3.10	9.23	8.40	6.00	7.60	13.70	2.30	7.56	9.10	0.50	1.47	3.40	<LOQ	1.66
PFNA	<LOQ	<LOQ	<LOQ	0.60	<LOQ	0.37	0.70	0.30	0.58	0.40	<LOQ	0.28	0.70	0.20	0.36	0.80	<LOQ	0.10	0.40	<LOQ	0.12
PFDA	<LOQ	<LOQ	<LOQ	0.10	<LOQ	0.07	0.10	0.10	0.10	0.10	<LOQ	0.03	0.10	<LOQ	0.03	0.10	<LOQ	0.02	0.20	<LOQ	0.05
PFUdA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFDDA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFTdA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFTaA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PF36DA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.50	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PF3MPA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.10	<LOQ	0.01	0.20	<LOQ	0.03	<LOQ	<LOQ	<LOQ
PF4MBA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.05	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
NMPFSMA	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.05	0.10	<LOQ	0.01	0.20	<LOQ	0.03	<LOQ	<LOQ	<LOQ
NEPSG	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.02
PF2M30A	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.30	<LOQ	0.13	0.20	<LOQ	0.03	0.30	<LOQ	0.05	0.20	<LOQ	0.04
SD3H48D	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.10	<LOQ	0.05	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.03	<LOQ	<LOQ	<LOQ
PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.13	0.60	<LOQ	0.15	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.10	<LOQ	0.03
PFOS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.50	<LOQ	0.13	0.30	<LOQ	0.04	0.40	<LOQ	0.12	0.80	<LOQ	0.31
PFBS	0.80	0.40	0.55	1.00	0.40	0.65	1.60	0.80	1.08	1.10	0.60	0.75	1.70	0.40	1.10	1.00	<LOQ	0.52	0.50	<LOQ	0.17
PF2ES	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.08	0.20	<LOQ	0.05	<LOQ	<LOQ	<LOQ	0.30	<LOQ	0.05	<LOQ	<LOQ	<LOQ
P9C3O1S	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	0.03	0.70	<LOQ	0.23
PFHpS	2.80	1.50	2.05	0.90	<LOQ	0.50	5.60	0.80	2.55	0.80	<LOQ	0.30	0.80	<LOQ	0.50	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
PFDS	3.80	1.00	2.30	1.00	<LOQ	0.65	2.20	0.50	1.15	0.30	<LOQ	0.13	0.30	<LOQ	0.11	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
P11C3O1S	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.80	<LOQ	0.14

Max.: maximum.

Min.: minimum.

Avg.: average.

LOQ: limit of quantification.



Quality Assurance and Quality Control

PP bottles and tubes, ultrapure water, methanol, and nitrogen were tested prior to sampling and no contamination was found in any of these blanks. Procedural blank samples and procedural spiked samples were performed for each set of extractions, and duplicate samples and archived blanks were performed for each sampling event. The limit of detection (LOD) was defined as the analyte peak required to give a signal-to-noise ratio (S/N) of 3:1, and the limit of quantification (LOQ) was defined as the analyte required to produce a S/N of 10:1, or as the lowest point of the calibration curve calculated to be within 30% of its actual value. Compound concentrations below the LOQ were reported as non-detected. Matrix spiked recoveries for water samples ranged from 75% to 117%. Proxy recoveries for water samples ranged from 81% to 115%. Internal calibration was used for quantification.

Risk Assessment

Since it is extremely difficult to remove PFASs by the current conventional water treatment process, this study assumes that PFASs in this reservoir can enter the drinking water network without loss and be absorbed into the body by humans. Based on this assumption, we selected the hazard quotient (HQ) model for risk assessment of 15 PFASs (data for other compounds of PFASs were not available). We adopted the assessment method of Borg (Borg et al., 2013) et al. for the hepatotoxicity as well as the reproductive toxicity of the target contaminants. The specific calculation equation is as follows:

$$HQ = \text{Exp}/\text{RfD}$$

$$\text{RfD} = \text{POD}/\text{Afs}$$

In the above formula, Exp is exposure concentration of PFASs; RfD is reference dose; POD is point of departure; Afs are assessment factors; HQ is the health quotient. When $HQ < 0.01$, very low risk; $0.01 < HQ < 0.1$, low risk; $0.1 < HQ < 1$, medium risk; $HQ > 1$, high risk (Lemly, 1996). POD is the PFASs serum/plasma concentration at the respective no observed adverse effect level (NOAEL), lowest observed adverse effect

level (LOAEL), or baseline dose (BMD). The data were collected from available studies. An Af of three was applied to calculate chronic toxic effects for hepatotoxicity and reproductive toxicity, respectively, according to the REACH guidelines for EU chemicals legislation (ECHA, 2010).

RESULTS AND DISCUSSIONS

Concentration and Composition Profile of Exposure

The maximum, minimum, and average concentrations of PFASs in a reservoir in a certain area of Zhejiang Province were shown in **Table 1**. In this study, total PFASs concentrations in the seven reservoirs ranged from 1.30 ng L^{-1} – 24.90 ng L^{-1} . Some PFASs in the reservoir have become common, and the detection rates of 7 pollutants in the 26 PFASs were above 50%. Among them, the detection rates of perfluorohexanoic acid (PFHxA) with 6 (C6) carbons, PFOA and perfluorobutane sulfonate (PFBS) with 8 (C8) carbons were more than 90%. As can be seen from **Table 1**, the detected concentrations of PFOA and PFBS ranged from 0.50 ng L^{-1} – 13.70 ng L^{-1} and 0 ng L^{-1} – 1.70 ng L^{-1} , respectively. In contrast, the detection concentrations of Perfluoro (3-methoxy)propionic acid (PF3MPA), Perfluoro (4-methoxy)butyric acid (PF4MBA), N-methyl perfluorooctane sulfonyl aminoacetic acid (NMPFSMA), N-Ethyl Perfluorooctane Sulfonyl Glycine (NEPSG), Sodium dodecafluoro-3H-4,8-dioxononate (SD3H48D) in PFCAs, and Potassium 11-chloroecosfluoro-3-oxaundecane-1-sulfonate (P11C3OLS) in PFSAs were lower, and the detection rates were in the range of 2%–8%, and the long-chain complete PFCAs (greater than or equal to 11 carbons) and Perfluoro-3,6-dioxaheptanoic acid (PF36DA) were not detected at all sampling points.

The composition and concentration of PFASs pollutants in various sampling points in this area of Zhejiang Province were shown in **Figures 1, 2**. As can be seen from **Figure 1**, among all the 26 PFASs pollutants detected, PFOA and Perfluorobutanoic acid (PFBA) were the main PFASs pollutants. The average contribution rate of PFOA with eight fluorinated carbon atoms to the overall PFASs was 39.42%. The composition and concentration of PFASs pollutants detected at each reservoir sampling point were shown in **Supplementary Figure S1–S14**. As shown in **Supplementary Figures S5, S9**, the highest PFOA concentrations was 13.7 ng L^{-1} sampled at C4 and E2, followed by C2 and E1, the concentrations of PFOA were 12.9 ng L^{-1} and 12.6 ng L^{-1} , respectively. In **Supplementary Figures S1, S11, S13**, lower concentrations of PFOA detected were 0.6 ng L^{-1} , 0.6 ng L^{-1} , 0.5 ng L^{-1} , and 0.3 ng L^{-1} sampled at A3, A4, F1, and G1, even no PFOA were detected at sampling points G10 and G11. In particular, the average contribution rate of PFOA in reservoir E was 56.48%, the average contribution rate of PFOA in reservoir A was 10.79%. Followed by the average contribution rate of PFBA in the seven reservoirs was 13.87%. The sampling points where the highest concentration of PFBA was 11.4 ng L^{-1} detected was D1 (**Supplementary Figure S7**), no PFBA were detected at sampling points B2, E2, and reservoir G except G2, G7 (**Supplementary Figure S3, S9, S13**). This shows that the concentrations of PFOA and PFBA varies greatly at different sampling points. There are many reasons for this, such as a degree of temporal variation in PFASs concentrations in the target reservoir, the use of a sewage treatment plant sludge as a soil amendment in the area, or the presence of fluoropolymer manufacturing plants in the area impacted the drinking water with PFOA or PFBA, etc. A more comprehensive survey of the area is needed to understand the details that are currently unknown to the authors.

This was similar to the study by Ding (Ding et al., 2018) et al. on PFASs in the seawater in the coastal waters of Dalian Bay, where PFOA is one of the main pollutants. Li (Li et al., 2016) et al. researched on PFASs occurrence in groundwater in rural areas in Liaoning peninsula located in the northeast of China, where PFOA and PFBA were the most frequently detected compounds. Wan (Wan et al., 2017) et al. found that the concentration range of total PFASs in seawater in the coastal waters of Shandong Peninsula was 23.69 ng L^{-1} – 148.48 ng L^{-1} , and PFOA, PFOS and PFHxA were the main PFASs in seawater in this area. In addition, Shao (Shao et al., 2016) et al. studied PFASs in the surface layer and low seawater of Shuangtaizi Estuary in Liaodong Bay and found that the total PFASs concentration was between 66.2 ng L^{-1} – 185 ng L^{-1} and 44.8 – 209 ng L^{-1} , respectively, and PFBA was the main short-chain pollutants. However, in previous studies, PFBS was the most abundant PFASs in the rivers of the Pearl River Delta (Liu et al., 2015), Wuhan, China (Zhou et al., 2017), and five major river basins in Italy (Valsecchi et al., 2015). 6:2 fluorotelomer alcohol (6:2 FTOH) and Perfluoroheptanoic acid (PFHpA) were detected as the most important PFASs in river water and sediment samples collected from a tributary of the Liu Xi River, a part of the Pearl River near Guangzhou (Si et al., 2021). The differences may be due to the different PFASs that were mainly used in different areas.

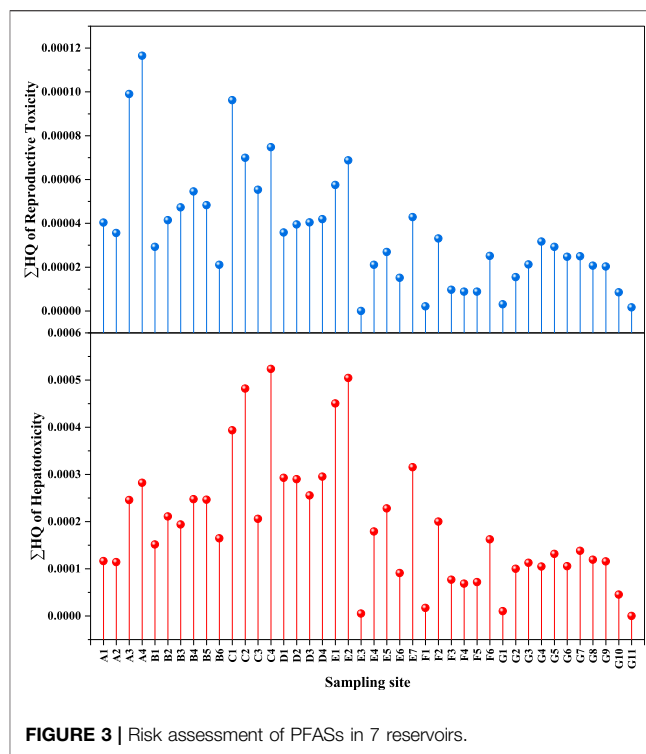


FIGURE 3 | Risk assessment of PFASs in 7 reservoirs.

As shown in **Figure 2**, among all the measured PFASs, PFCAs were the most important class of PFASs, with a total concentration ranged from 0.90 ng L^{-1} – 22.30 ng L^{-1} and the proportion ranged from 69.23% to 89.56%. PFOA and PFBA were the main PFASs pollutants with the average concentrations of 4.24 ng L^{-1} and 1.64 ng L^{-1} , respectively. Due to the limitation of long-chain PFASs, short-chain PFASs account for a relatively high proportion of total PFASs in aquatic environments, especially PFBA and PFBS are widely used as substitutes worldwide (Li et al., 2020). However, their adsorption to sediments and soil is limited (Zhao et al., 2016). In addition, the removal efficiency of short-chain PFASs is relatively limited even if advanced treatment processes such as granular activated carbon, ion exchange, membrane treatment are used in drinking water treatment (Zhao et al., 2016). Similarly, the results of Chen (Chen et al., 2017) et al. researched on PFASs in seawater in the Bohai Sea showed that the average concentration of PFOA was as high as 4.97 ng L^{-1} , which was the main PFASs pollutant. Liu (Liu et al., 2021) et al. found a survey of different enterprises in China in 2018 that extremely high concentrations of PFOA were mainly concentrated in polytetrafluoroethylene (PTFE)-producing provinces, such as Sichuan ($3,165 \text{ ng L}^{-1}$), Zhejiang (115.4 ng L^{-1}), Shanghai (78 ng L^{-1}), Jiangsu (61.4 ng L^{-1}), and Guangdong (53.4 ng L^{-1}). The patterns and concentrations of PFASs in drinking water can be affected due to the emission sources, especially fluorination plants, major local industrial users of PFASs, use of PFAS-containing firefighting foams (Hu et al., 2016; Mumtaz et al., 2019b; Li et al., 2019), the quality of drinking water treatment processes, and their precursor conversions. In this research, compared with PFCAs, PFASs accounted for a smaller proportion of total PFASs, and among the eight detected

PFASs, the average concentrations of PFBS and Perfluoroheptane sulfonic acid (PFHpS) were higher, the concentration levels were 0 ng L^{-1} – 1.6 ng L^{-1} and 0 ng L^{-1} – 5.6 ng L^{-1} , respectively.

Ecological Risk Assessment

This study adopts the Hazard Quotient (HQ) method to assess the level of potential health risk. HQ value is widely used to assess the potential health risk of different chemicals, including PFASs (Ludwicki et al., 2015; Habibullah-Al-Mamun et al., 2016; Mumtaz et al., 2019a). Due to the lack of toxicological data for some mixtures of PFAS in this assessment, toxicity data may not be completely accurate for all mixtures. However, some studies have shown that the toxicological endpoints evaluated were hepatotoxicity (hepatocellular hypertrophy, hepatocellular vacuolation, increased liver weight and liver-to-body ratio) and reproductive toxicity (reduced fetal/perinatal/neonatal viability, reduced body weight/body weight gain and litter) loss in the dams (Si et al., 2021). Reproductive toxicity and hepatotoxicity were considered to be the more common methods for assessing accumulation of PFASs. We evaluated 15 PFASs pollutants and the calculated results of the HQ value in this study were shown in **Figure 3**. It can be seen from **Figure 3** that the HQ values of the reproductive toxicity and hepatotoxicity of the total PFASs at different sampling points ranged from 2.30×10^{-8} to 1.16×10^{-4} and 9×10^{-8} to 5.24×10^{-4} respectively, which were low than 0.01, indicating that the potential health risk was very low. It was worth noting that the HQ values of them were much lower than 0.01, although PFOA and PFBA were the main PFASs pollutants. The average HQ values of reproductive toxicity and hepatotoxicity of PFOA were 6.02×10^{-4} and 5.75×10^{-3} , respectively, and the average HQ values of reproductive toxicity and hepatotoxicity of PFBA were 6.68×10^{-7} and 1.34×10^{-6} , respectively, indicating that PFOA and PFBA didn't pose the health risk to the target area. Some researchers believed that the detection of PFOS and PFOA in soil and groundwater near a fluorine chemical plant in China poses a much greater health risk than other fluorinated compounds (Wei et al., 2018; Xie et al., 2021). PFOS can suppress sheep red blood cell-specific immunoglobulin M production on mice, and cause hepatocellular hypertrophy in livers of rats (Peden-Adams et al., 2008; Butenhoff et al., 2012), even caused the death of newborn mice (Yahia et al., 2010). PFOA can derive stunted mammary epithelial growth on offspring of mice, and increased peroxisome proliferation and relative liver weights on rats (Perkins et al., 2004; Macon et al., 2011). The diversity of PFAS classes complicates the ecotoxicological study of PFAS. Although bioaccumulation of some persistent organic pollutants (POPs) is usually associated with lipid partition coefficients, PFAS are not exclusively lipid-related (De Silva et al., 2021). Bioaccumulation models suggest that both protein interactions and lipid partitioning are important parameters for accurate PFAS assessment (Ng and Hungerbühler, 2014; Dassuncao et al., 2018). Differences for specific physicochemical properties, such as chain length, can lead to different distribution of PFAS in biological tissues (Chen et al., 2021). In this study, although the assessed risks of all sampling points were low, long-term ecological effects and risks cannot be ignored due to the strong bioaccumulation of PFASs and their difficulty in degrading. In early 2020, China's Ministry of Ecology and Environment stated

that more POPs would be included in the national monitoring system. PFASs, particularly PFOS and PFOA, should be added to the candidate list (Zhang et al., 2021).

CONCLUSION

In this study, water samples from 7 reservoirs in a certain area of Zhejiang Province were detected, and 26 PFASs pollutants were selected for comprehensive evaluation. At the same time, the potential health risk assessment of the reservoir was carried out using the HQ method. Among the 26 PFASs pollutants, the detection rates of 7 pollutants ranged above 50%, and the detection rates of PFHxA, PFOA, and PFBS were all above 90%. The concentrations and detection rates of the mixtures tested show that PFOA and PFBA were the main PFASs pollutants, and the average contribution rates of PFOA and PFBA to the overall PFASs were 39.42% and 13.87%, respectively.

According to the potential health risk analysis, the HQ values of the reproductive toxicity and hepatotoxicity of the total PFASs at different sampling points were low than 0.01, indicating that there was no immediate risk in this area. When conducting water potential health risk assessment in the future, it is necessary to develop a comprehensive evaluation method for these compounds to comprehensively investigate the impacts and risks of PFASs pollutants on the water environment. In addition, PFASs pollutants have strong bioaccumulation and are difficult to degrade. Long-term healthy impacts and risks cannot be ignored. Strengthen the management of pollution sources and continue to pay attention to the situation of PFASs pollutants in the region.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

SH: Analysis and writing NR: Revision.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenvs.2022.913997/full#supplementary-material>

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The remaining author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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