




# Occurrence and Risk Assessment of Perfluorooctanoate (PFOA) and Perfluorooctane Sulfonate (PFOS) in Surface Water, Groundwater and Sediments of the Jin River Basin, Southeastern China

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## Abstract

Perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS) levels were determined in surface water, groundwater and sediments of the Jin River Basin, southeastern China. PFOA was detected in most of the samples, and its concentrations ranged from 0.53 to 8.77 ng/L, 0.26 to 15.1 ng/L and not detected (ND) to 23.9 ng/g in surface water, groundwater and sediments, respectively. Unlike PFOA, the detection frequency of PFOS was lower than 32%, and its concentrations ranged from ND to 2.56 ng/L, ND to 7.01 ng/L, ND to 11.1 ng/g in surface water, groundwater and sediments, respectively. The environmental risk assessment showed that PFOA could pose a high risk to surface water and groundwater, and both PFOA and PFOS posed a high risk to sediments. Moreover, the adults living in the Jin River Basin were at insignificant health risk to exposure to PFOA and PFOS through water consumption.

**Keywords** PFOA · PFOS · Spatial distribution · Risk assessment · Southeastern China

Per- and poly-fluorinated alkyl substances (PFASs) have been produced and used in different industrial and commercial fields for more than 50 years (Podder et al. 2021; Zhang et al. 2018). Due to their unique physicochemical properties such as the stability, the hydrophilic and hydrophobic properties and the ability to reduce the surface tension, PFASs have been widely applied as water and stain repellants, non-stick coatings for cookware, firefighting foams and pesticide formulations (Mumtaz et al. 2019). Widespread usage of PFASs over last several decades has resulted in their global occurrence in wastewater, surface water, groundwater, soils

and sediments (Chen et al. 2016; Xiao et al. 2021). Due to their ubiquitous distribution, bioaccumulation potential and threat to ecosystems and human health (Cai et al. 2021; Domingo and Nadal 2019; Li et al. 2020), PFASs contamination has been recognized as a global issue of increasing concern.

Perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS) were the two most frequently observed PFASs due to their mass production and wide use (Xiao et al. 2021). PFOA and PFOS have been detected in almost all environmental media, including surface water, groundwater, drinking water, coastal water, landfill leachates, sludge, sediments and soils at levels ranging from ng/L to µg/L or from ng/g to µg/g (Chen et al. 2016; Podder et al. 2021; Huang et al. 2021). Industrial wastewater, effluents discharged from wastewater treatment plants and landfill leachate are important sources for PFOA and PFOS (Xiao et al. 2022; Wang et al. 2019). According to toxicological studies, PFOA and PFOS could pose acute to chronic toxicity to freshwater fish and invertebrates, and cause liver and pancreatic tumors in laboratory animals (Lee et al. 2020; Cai et al. 2021). For the reasons given above, PFOA and PFOS were inevitably regarded as significant environmental contaminants in China and other countries.

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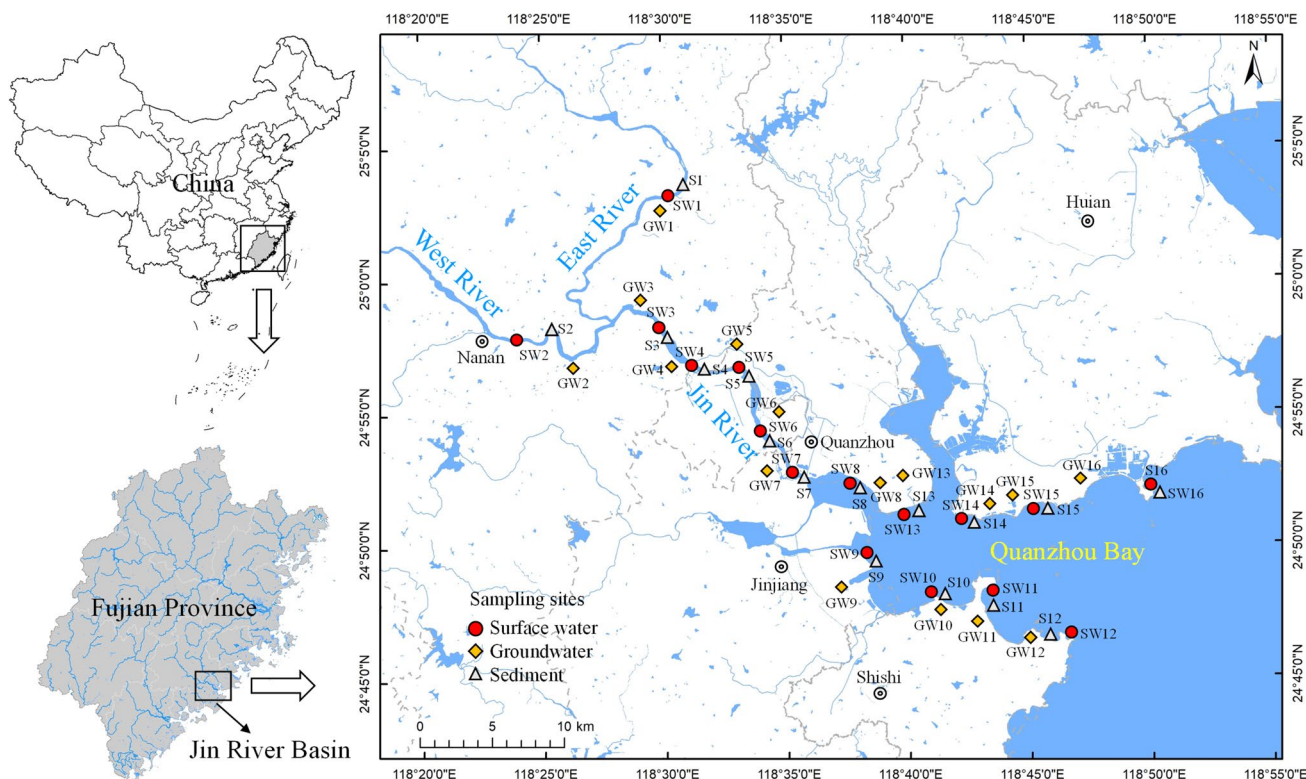
The Jin River Basin, covering a total area of 5629 km<sup>2</sup>, is located in Fujian Province, southeastern China. The average temperature ranges from 17 to 21 °C and the annual precipitation is about 1010 to 1756 mm. About 70% of the precipitation in the Basin is concentrated between June and September, leading to uneven annual runoff distribution in this area. The Jin River Basin performs as a major water head site for Quanzhou City. Electronic, textile and paper industries are widely distributed along the Jin River Basin, and many PFASs such as PFOA and PFOS are expected to be used in these industries (Chen et al. 2020). However, there has been little information about the distribution of PFASs in this area. To the best of our knowledge, there was only one recent publication reporting PFOA and PFOS levels in surface water of the Jin River, and the PFOA and PFOS concentrations ranged from 1.88 to 4.46 ng/L and from not detected (ND) to 1.49 ng/L, respectively (Qin et al. 2021). However, only three samples were collected in this study which might not be representative enough to reflect the pollution status. In addition, to date, no information about the PFOA and PFOS levels in groundwater and sediments along the Jin River basin is available.

In this study, a total of 16 surface water, 16 groundwater and 16 sediment samples were collected from the Jin River Basin to characterize the occurrence and spatial distribution

of PFOA and PFOS. Furthermore, the environmental and human health risks of the two compounds in this area were assessed.

### Materials and Methods

A total of 48 samples including 16 surface water (SW1–SW16), 16 groundwater (GW1–GW16) and 16 sediment (S1–S16) samples were collected along the Jin River Basin in June and July in 2020 (Fig. 1). Duplicate samples were collected from each site for surface water, groundwater and sediments. To cover the whole river basin especially the receiving environment for sewage, domestic and industrial wastes from urban and rural areas, the surface water or sediment sampling sites were evenly selected from both the Jin River and its tributaries (i.e., the West River and the East River). The distance between two adjacent sampling sites was kept at about 5 km. All the surface water and sediment samples were collected during low tide. In this case, the effect of dilution was minimal, which could better reflect the actual pollution situation in the coastal environment. The groundwater samples were collected from the existing drinking-water supply wells at depths of 3.5–13.4 m near the surface water sampling sites. All groundwater sampling sites



**Fig. 1** Sampling locations of surface water (SW1–SW16), groundwater (GW1–GW16) and sediment (S1–S16) samples along the Jin River Basin, southeastern China

were flushed for 10 min before collection. Water samples of 1 L were collected using a water sampler and stored in pre-cleaned polypropylene bottles. Surface sediment samples (1–10 cm, approximately 1000 g in weight) were collected using a stainless steel grab sampler and bagged with polyethylene ziplock bags. All the samples were immediately transported on ice in a cooler to laboratory. Water samples were stored at 4 °C. Sediment samples were stored at –18 °C.

PFOA and PFOS in surface water and groundwater samples were extracted following the previously validated methods (Xiao et al. 2021). Briefly, the water samples were firstly filtered through glass fiber membranes (GF/F, pore size 0.7 µm) obtained from Whatman (Maidstone, UK) to remove the very fine particles. After being spiked with internal standards ( $^{13}\text{C}_4$ -PFOA and  $^{13}\text{C}_4$ -PFOS, 10 ng each), 1 L of water was loaded (1.0 mL/min) to the HBL cartridge (200 mg, 6 mL Oasis, Waters) for solid phase extraction (SPE). After washing with 4 mL of acetate buffer (25 mmol/L, pH 4), the cartridge was dried under nitrogen steam for 60 min to remove the residual water. Then, the targets were eluted with 10 mL of methanol and evaporated under nitrogen steam to nearly dryness. Finally, the fractions were reconstituted with water to 1.0 mL and transferred to injection vials for further analysis.

The sediment samples were firstly dried in dark, and sieved (100 mesh) to remove the big particles. Extraction of the analytes from the sediment was carried out using a method similar to that described previously (Gao et al. 2015). A mass of 1.0 g of sample was placed in a 50 mL centrifugation tube and mixed with internal standards ( $^{13}\text{C}_4$ -PFOA and  $^{13}\text{C}_4$ -PFOS, 10 ng each), 2.0 mL of NaOH-methanol solution (0.2 mol/L) and 20 mL of methanol. After sonicated for 45 min, 0.2 mL of HCl solution (0.2 mol/L) was added and diluted with methanol to 30 mL. After centrifugation at 7000 rpm for 20 min, 2 mL of the supernatant was collected and diluted with water to 1.0 L for SPE. The SPE conditions for sediment samples were the same as for water samples.

Instrumental analysis of PFOA and PFOS was based on a previous work (Huang et al. 2021). The analytes were detected by liquid chromatography (LC) with a binary pump, a C18 column (100 mm × 4.6 mm, 2.6 µm particle size, Kinetex, Phenomenex), column oven (at room temperature) and a UV detector. The LC was coupled to a triple quadrupole mass spectrometer (LC-MS/MS, API 3200 Q TRAP, Applied Biosystems, USA). Mobile phase: A, water containing 2 mL of  $\text{NH}_4\text{Ac}$ ; B, Methanol. Gradient: 0–5 min, 50%–90% B; 5–9 min, 90% B; 9–9.5 min, 90%–50% B; 9.5–12 min 50% B. Other conditions: flow rate, 1.0 mL/min; injection volume, 20 µL. All the analytes were determined in negative multiple reaction monitoring (MRM). For each analyte, two ion transitions were applied, one for quantification (with higher signal response) and the other for confirmation

(Table S1). The electrospray ionization (ESI) source conditions were: ion source gas 1, 50 psi; gas 2, 60 psi; source temperature, 550 °C; ion spray voltage, –5500 V.

The physicochemical parameters [temperature, electric conductivity (EC), total dissolved solids (TDS), oxidation-reduction potential (ORP), pH and dissolved oxygen (DO)] of water samples were determined in situ by a portable multi-parameter water quality analyzer (HACH, SL1000) during sample collection. The organic matter (OM) of sediment samples was analyzed in a qualified laboratory (SUEZ Environmental Detection Technology (Guangzhou) Co., Ltd) according to Chinese standard methods within 2 weeks of sample collection.

The recovery of each spiked sample, limit of detection (LOD), limit of quantification (LOQ), field blank, matrix spike recovery, and duplicate samples collected from each site were measured. The LOD and LOQ were calculated based on a signal to noise ratio (S/N) of 3 and 10, respectively. All the analytical results lower than LOQ were reported as ND. PFOA and PFOS in blanks were well below the LOQ. The recoveries, LOD and LOQ for PFOA and PFOS were shown in Table S2.

For environmental risk assessment in water and sediments, risk quotient (RQ) was calculated by dividing the measured environmental concentration of PFOA and PFOS by predicted-no-effect concentration (PNEC) (Wang et al. 2019). The PNEC value of PFOA and PFOS in surface water and groundwater was selected from the lowest value of toxicological data or water quality criteria for the protection of aquatic organisms reported in literatures (i.e., 5.1 and 13 ng/L for PFOA and PFOS, respectively) (Garnick et al. 2021; Cordner et al. 2019). Sediment PNEC value was calculated by the equilibrium partitioning method (Huang et al. 2020). As a result, sediment PNEC value was derived from the above PNEC value in water and organic carbon normalized partitioning coefficient ( $\log K_{oc}$ , 2.11 and 2.68 L/kg for PFOA and PFOS, respectively) reported in literature (Higgins and Luthy 2006). Sediment PNEC was calculated for PFOA and PFOS at 0.020 and 0.19 ng/g, respectively. For health risk assessment in water, RQ was determined by dividing the exposure dose for adults by oral reference dose for non-carcinogenic risk (0.14 and 0.075 µg/kg bw/day for PFOA and PFOS, respectively) (Sun et al. 2018). The exposure dose was calculated using the exposure equation and parameters suggested by Sun et al. (2018). Risk levels were established based on the RQ values, i.e., high risk ( $\geq 1$ ), medium risk (0.1 to <1), low risk (0.01 to <0.1) and insignificant risk (<0.01) (Huang et al. 2020).

The difference in water quality parameters (EC, TDS, ORP, pH and DO) between surface water and groundwater was analyzed by Mann-Whitney test. Spearman's rank correlation analysis was adopted to study the relationship between PFOA levels and measured physicochemical parameters of

water (EC, TDS, ORP, pH and DO) and sediments (OM). The box plots and bar charts were plotted using SigmaPlot (version 14.0), and the statistical analysis were conducted by SPSS (version 22.0, IBM Inc., Chicago).

### Results and Discussion

The EC of surface water was  $24,639 \pm 22,025 \mu\text{S}/\text{cm}$ , which was significantly higher than that of groundwater ( $1284 \pm 1319 \mu\text{S}/\text{cm}$ ) (Fig. S1a). As for surface water, the EC values for SW7 to SW16 were all above the maximum permissible limit ( $1500 \mu\text{S}/\text{cm}$ ) (Table S3), as these sites were located in Quanzhou Bay. The groundwater samples collected along the northern coastal line (GW14-16) of Quanzhou Bay were more affected by saline intrusion, as reflected by higher EC values at these three sampling points (Table S4). Similar as EC, TDS, pH and DO in surface water exhibited higher levels than in groundwater, while the trend of ORP was the opposite (Fig. S1b–e).

The detection frequency of PFOA was 100% for both surface water and groundwater samples, and 93.8% for sediment samples (Table 1). The mean PFOA levels were  $3.41 \text{ ng}/\text{L}$  (ranging from  $0.53$  to  $8.77 \text{ ng}/\text{L}$ ) and  $5.67 \text{ ng}/\text{L}$  (ranging from  $0.26$  to  $15.1 \text{ ng}/\text{L}$ ) in surface water and groundwater of the Jin River Basin, respectively. The wastewater discharge near SW2 ( $8.77 \text{ ng}/\text{L}$ ) and SW5 ( $5.99 \text{ ng}/\text{L}$ ) might have led to relatively high levels of PFOA in surface water in these sampling points (Fig. 2). High concentrations

(>  $10 \text{ ng}/\text{L}$ ) of PFOA were detected in several groundwater samples, including GW7 ( $15.1 \text{ ng}/\text{L}$ ), GW9 ( $14.9 \text{ ng}/\text{L}$ ) and GW8 ( $12.0 \text{ ng}/\text{L}$ ). The PFOA levels in sediment samples ranged from ND to  $23.9 \text{ ng}/\text{g}$  (Table 1), and the highest level occurred at S4 (Fig. 2). Unlike PFOA, PFOS levels were relatively low, and it was detected in only 18.8%, 18.8%, and 31.3% of the surface water, groundwater and sediment samples, respectively (Table 1). The highest level of PFOS occurred at GW9 ( $7.01 \text{ ng}/\text{L}$ ) for water samples and S6 ( $11.1 \text{ ng}/\text{g}$ ) for sediment samples (Fig. 2).

There were a large number of factories including electronic, textile and paper industries and several wastewater treatment plants along the Jin River Basin, and PFOA and PFOS were discharged primarily from these point sources (Xiao et al. 2021; Qin et al. 2021). In addition, effluents from wastewater treatment plants receiving industrial as well as domestic discharges were also important sources of PFOA and PFOS (Chen et al. 2020; Podder et al. 2021). Some sampling points (SW2, SW5, GW7, GW8, GW9, S4 and S6) with relatively high levels of PFOA and PFOS were found close to several wastewater treatment plants and factories. Spatial analysis of PFOA and PFOS levels in this study together with the results obtained by other researchers indicated that emissions of wastes from manufacturing industries and effluent discharge from wastewater treatment plants could be important reasons for the high levels of PFOA and PFOS in these sampling points.

The PFOA and PFOS levels in surface water, groundwater and sediment samples collected from nearby regions

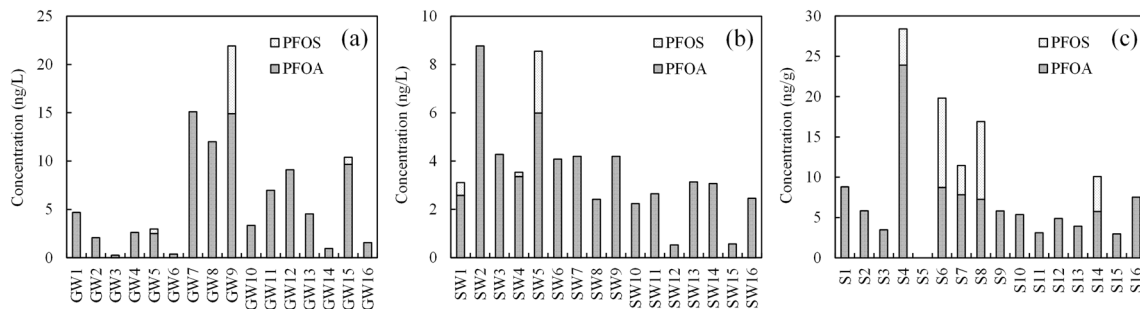
**Table 1** Concentrations of PFOA and PFOS in groundwater, surface water and sediments of the Jin River Basin

Chemical	Groundwater ( <i>n</i> = 16, ng/L)					Surface water ( <i>n</i> = 16, ng/L)					Sediment ( <i>n</i> = 16, ng/g)				
	Freq <sup>a</sup>	Max	Mean <sup>c</sup>	Median	Min	Freq	Max	Mean	Median	Min	Freq	Max	Mean	Median	Min
PFOA	100	15.1	5.67	3.94	0.26	100	8.77	3.41	3.11	0.53	93.8	23.9	6.66	5.79	ND
PFOS	18.8	7.01	0.58	ND <sup>b</sup>	ND	18.8	2.56	0.27	ND	ND	31.3	11.1	3.04	ND	ND

<sup>a</sup>Detection frequency (%)

<sup>b</sup>Not detected (below the LOQ)

<sup>c</sup>Half of LOQ was used for mean calculation whenever the level was below LOQ



**Fig. 2** Distribution of PFOA and PFOS in **a** groundwater, **b** surface water and **c** sediments of the Jin River Basin

**Table 2** Concentrations of PFOA and PFOS in different regions

Region	PFOA concentration	PFOS concentration	References
Surface water (ng/L)			
Jin River	2.65 (1.88–4.46)	1.00 (ND–1.49)	Qin et al. (2021)
Jin River	3.41 (0.53–8.77)	0.27 (ND–2.56)	This study
Min River	23.5 (17.4–28.0)	1.07 (ND–1.72)	Qin et al. (2021)
North River and West River	40.7 (0.125–1030)	0.50	Qin et al. (2021)
Longgang River and Pingshan River	41.5 (2.30–288.28)	175 (6.17–868)	Huang et al. (2021)
Pearl River	3.70 (0.71–8.70)	69.2 (16.8–144)	Zhang et al. (2013)
Pearl River	7.86 (0.11–26.5)	16.9 <sup>b</sup> (ND–321)	Pan et al. (2014)
Beibu Gulf	0.73 (0.16–1.46)	0.14 (0.06–0.21)	Xiao et al. (2021)
Groundwater (ng/L)			
Rural areas in eastern China	90.8 (7–175.2)	19.2 (< 0.5–37)	Chen et al. (2016)
Jin River Basin	5.67 (0.26–15.1)	0.58 (ND–7.01)	This study
Sediment (ng/g)			
Pearl River Delta	0.05–0.99	ND–11.4	Pan et al. (2014)
Beibu Gulf	0.11 (0.04–0.25)	0.10 (0.04–0.20)	Xiao et al. (2021)
Jin River	6.66 (ND <sup>a</sup> –23.9)	3.04 (ND–11.1)	This study

<sup>a</sup>Not detected (below the LOQ)<sup>b</sup>Median value when mean value was not available**Table 3** Correlation between different measured parameters

Parameter	EC	TDS	ORP	pH	DO	OM
TDS	1*					
ORP	−0.67*	−0.67*				
pH	0.49	0.49	−0.74*			
DO	0.18	0.18	−0.32	0.32		
PFOA level in water	0.48	0.48	−0.22	−0.1	−0.12	
PFOA level in sediments						−0.35

\*Indicated a significant Spearman's rank correlation ( $p < 0.05$ )

(i.e., southeastern China) were illustrated in Table 2. The PFOA and PFOS levels in this study were comparable with those in a recent study by Qin et al. (2021), which showed that PFOA and PFOS concentrations in surface water of the Jin River ranged from 1.88 to 4.46 ng/L and from ND to 1.49 ng/L, respectively. In their study, only three surface water samples were collected, and the comparability of PFOA and PFOS concentrations may be attributed to the similar sampling periods (rainy season of August 2018) with respect to the same river. The PFOA levels in the Jin River were much lower than those in Min River and several inland rivers (i.e., North River and West River, Longgang River and Pingshan River) (Qin et al. 2021; Huang et al. 2021) (Table 2). Likewise, PFOA and PFOS concentrations in groundwater of the Jin River Basin were observed to be lower than those of rural areas in eastern China (Chen et al. 2016) (Table 2). However, for sediment samples, the levels of PFOA and PFOS were higher than those detected in Pearl River Delta and Beibu Gulf (Pan et al. 2014; Xiao et al. 2021) (Table 2).

Spearman's rank correlation was applied since most of the dataset was not normally distributed. EC showed a significantly positive correlation with TDS, while ORP was negatively correlated with EC, TDS and pH (Table 3). The results of correlation analysis were in accordance with previous monitoring studies (Wang et al. 2018; Zhang et al. 2015). EC was directly correlated with TDS because both of them were used to describe salinity level.

As the detection frequency of PFOS was very low, the correlation between the level of PFOS and other parameters was not analyzed. The level of PFOA in water samples was not significantly correlated with any of the parameters, and PFOA in sediments was not significantly correlated with OM either (Table 3 and S5). This was different with the results from a previous study which reported significant correlation between PFASs concentrations and EC values in river water (Pan et al. 2014). Due to destabilized regional water or sediment environment under the impact of river plume and coastal current (Gao et al. 2015), the distribution of PFOA and PFOS in water-sediment system might not be

in equilibrium under these disturbances, thereby leading to inconsistent relationship between PFASs and EC. Some other studies have observed significantly positive correlation between OM and PFOA or PFOS levels in sediment samples (Gao et al. 2015; Lyu et al. 2019). However, it was also reported that no specific fraction of organic carbon could explain the variation in sorption of PFASs (Li et al. 2018; Oliver et al. 2020). Miao et al. (2017) observed that the sorption–desorption of PFOA was correlated with both OM and mineral composition, and retention and transport of PFOA in soils were controlled by both hydrophobic and electrostatic interactions. Due to the varied physicochemical properties of soils or sediments used in different studies, the relationship between OM in soils or sediments and the level of PFASs as well as the interactions between them are still under debate, which deserve further efforts.

The results of environmental risk assessment showed that the RQ values of PFOA in 12.5% of the surface water samples and 37.5% of the groundwater samples were higher than 1, indicating high risks of PFOA in the aqueous environment of the Jin River (Table S6). The risk levels in the present study were much higher than those reported in a previous study conducted at Beibu Gulf, China, which showed that PFOA posed minimal risks to the aqueous ecosystem since lower PFOA concentrations were detected in that region (Xiao et al. 2021). The RQ values of PFOS (0.007–0.54) in all surface water and groundwater samples were less than 1, indicating that PFOS posed insignificant to medium risks to the water environment. Similarly, the PFASs levels in the South China sea coastal regions did not pose any risk to aquatic organisms (Wang et al. 2019). Alarmingly, the RQ values of PFOA and PFOS in sediments in this study were all higher than 1, indicating that they were at a high-risk level in the Jin River Basin. This was in contrast to the results reported at the Pearl River Estuary in Southern China where PFOA and PFOS were unlikely to pose any risk to benthic organisms due to the low concentrations (Gao et al. 2015). The health risk assessment showed that the RQ values for PFOA and PFOS in water samples were all less than 0.01 (Tables S7 and S8), indicating that the adults living in the Jin River Basin were at insignificant risks to PFOA and PFOS through water consumption. Similar results were reported for adults living in Shanghai (Sun et al. 2018).

## Conclusions

The results obtained in this study showed that PFOA was detected in most of the samples collected from the Jin River Basin, and its levels ranged from 0.53 to 8.77 ng/L, 0.26 to 15.1 ng/L and ND to 23.9 ng/g in surface water, groundwater and sediments, respectively. Unlike PFOA, the detection rate of PFOS was low, and its levels ranged from ND to

2.56 ng/L, ND to 7.01 ng/L and ND to 11.1 ng/g in surface water, groundwater and sediments, respectively. In addition, the level of PFOA did not show any correlation with the measured physicochemical parameters of water (EC, TDS, ORP, pH and DO) and sediments (OM). The environmental risk assessment revealed that PFOA had a high risk to surface water and groundwater. Moreover, there was a high environmental risk for PFOA and PFOS in sediments. Furthermore, PFOA and PFOS were unlikely to pose health risks to the adults living in the Jin River Basin from water consumption.

**Supplementary Information** The online version of this article (<https://doi.org/10.1007/s00128-021-03435-w>) contains supplementary material, which is available to authorized users.

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**Author contributions** YL: Conceptualization, Writing-original draft, Visualization, Funding acquisition. YL: Methodology, Resources. GS: Investigation, Data curation. CL: Project administration. QH: Validation. LW: Formal analysis, Writing-review & editing, Supervision.

## Declarations

**Conflict of interest** The authors declare that they have no conflicts of interests.

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