

# Perfluorooctane sulfonate in water and sediment from the Yangtze Estuary and coastal waters: Occurrence, partitioning and diffusion behavior and risk assessment



Jing Chen <sup>a</sup>, Hong Huang <sup>a, b, \*</sup>, Ruijing Li <sup>b</sup>, Jinchen Yu <sup>c</sup>, Zhiming Hu <sup>a</sup>, Xu Zhao <sup>a</sup>, Yang Li <sup>a</sup>, Wenqian Wang <sup>a</sup>

<sup>a</sup> College of Marine Ecology and Environment, Shanghai Ocean University, Shanghai, China

<sup>b</sup> State Environmental Protection Key Laboratory of Coastal Ecosystem, Dalian, China

<sup>c</sup> East China Sea Fisheries Research Institute, Chinese Academy of Fishery Sciences, Shanghai, China

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## ABSTRACT

As an emerging environmental contaminant, perfluorooctane sulfonate (PFOS) has attracted increasing attention. This study investigated the occurrence, partitioning and diffusion behavior, and risk assessment of PFOS in 19 water and sediment samples collected from the Yangtze Estuary and coastal waters in March 2022. The concentration of PFOS is 0.053–0.95 ng L<sup>-1</sup> in water and 0.031–0.18 ng g<sup>-1</sup> (dw) in sediments, demonstrating a transparent concentration gradient from inshore to offshore sites. The fugacity fraction model results showed that, except for a few sites, the diffusion trend of PFOS was from sediment into the water. Correlation and response coefficient (RC) analyses revealed a significant relationship between sediment organic carbon (TOC) and PFOS partitioning. According to the sediments-water partitioning coefficient K<sub>d</sub>, in estuarine areas, seawater salinity tended to allocate PFOS to sediments. Ecological risk assessment findings revealed that PFOS posed a low to moderate risk in sediment rather than a problem in water.

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## 1. Introduction

Perfluorooctane sulfonate (PFOS) is a significant perfluorinated surfactant [1]. All perfluorooctane sulfonyl fluoride (POSF) and perfluorinated substances base on POSF can degrade to PFOS, and both are crucial precursors to various other perfluorinated compounds [2]. Since the 1960s, when the electrochemical fluorination reaction method was used to produce PFOS and other perfluorinated compounds, thousands of perfluorinated organic compound series products containing sulfonyl groups have been developed, produced, and widely used [3,4]. PFOS is widely used in textiles, leather products, furniture, carpets, foam fire extinguishing agents, floor polishing agents, pesticides, and detergents due to their hydrophobic and oil-repellent properties [1]. Therefore, PFOS was found in waters [5–8], atmosphere [9,10], sediments [11–13],

and soils [12,14], as well as in the vast and desolate ocean [15], the Arctic and Antarctic [16], and the Tibetan Plateau [17]. Moreover, PFOS was discovered in human blood, breast milk, and liver [18,19], posing significant environmental and biological risks that can no longer be ignored. As the general public and scientific community become more aware of perfluorinated compounds' environmental consequences and biological toxicity, numerous national and international regulations have been developed to regulate their usage [20]; for example, PFOS was included in the Stockholm Convention in 2009 [21]. PFOS remains the most prevalent perfluoroalkyl compound in the marine environment [15,22–25].

Because of river input and atmospheric deposition, the marine environment is considered the final sink of PFOS [26,27]. According to a global study of PFOS in oceans, coastal water has PFOS concentrations as high as 192 ng L<sup>-1</sup>, roughly three to four orders of magnitude more than open ocean water [5]. Moreover, PFOS has been frequently detected in seawater, marine sediments, and marine organisms [28]. Knowing how PFOS is transported and transformed in the aquatic environment and estimating the danger to aquatic life and people exposed to pollution are critical [29].

\* Corresponding author. College of Marine Ecology and Environment, Shanghai Ocean University, Shanghai, China.

E-mail address: [hhuang@shou.edu.cn](mailto:hhuang@shou.edu.cn) (H. Huang).

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Furthermore, the partitioning of PFOS between water and sediment needs to be understood. Studies have shown that the partitioning of perfluorinated compounds in sediments and water is related to salinity [30], total TOC content (TOC) [31,32], pH [31,33], ionic strength, sediment properties and other factors [34], and the correlation of all factors need to be further confirmed. The partitioning of PFOS varies significantly in different water-sediment systems. Therefore, additional field study data on the dispersal and partitioning behavior of PFOS sediments and aquatic species are required better to understand the fate of PFOS in the aquatic environment. These data are also essential for understanding the migration trend of PFOS in seawater.

China's longest river, the Yangtze, flows through various towns before discharging into the East China Sea near Shanghai. With the rapid economic and social development, the rapid increase in population, and the acceleration of urbanization in the Yangtze River Basin, a large amount of domestic and production sewage has been imported into the estuary and exported offshore [23], the leading causes of water pollution in the Yangtze Estuary are saline water intrusion and organic pollutants. It has been reported that the annual load of PFOS in the Yangtze River Basin is 0.88 tons [35]. Estuaries are a significant area for transporting PFOS to the open sea [36,37], where the environmental conditions differ from freshwater [38]. Previous research on the Yangtze Estuary had mainly concentrated on traditional pollutants like ammonia nitrogen and heavy metals, as well as a large number of new pollutants (like organophosphate esters), organochlorine pesticides, tetracycline antibiotics, and polychlorinated biphenyls. More study is still required to fully comprehend the partitioning and fate of PFOS in this location. Investigating PFOS contamination in the Yangtze Estuary and coastal waters is therefore necessary.

This research aimed to look into the concentrations of PFOS contamination and partitioning between water and sediments in the Yangtze Estuary and coastal waters. To this end, PFOS concentrations in waters and sediments of this area were measured, diffusion trends of PFOS sediments and water were studied, the effects of total TOC content and salinity on the partitioning of PFOS sediments and water were assessed, and the risk of the Yangtze River estuary was evaluated.

## 2. Material and methods

### 2.1. Chemicals and reagents

Methanol was purchased from Fisher Scientific (HPLC grade, UK), acetonitrile from ACS (HPLC grade, USA), and ammonium acetate and ammonia water from Amphorus (LCMS grade and HPLC grade, respectively, China). The PFOS internal standard was sourced from Wellington Laboratories (Canada), and Watsons supplied the Milli-Q water used throughout the experiment.

### 2.2. Sample collection

The NSF Shiptime Sharing Project cruise collected overlying water and surface sediment samples from 19 stations in the Yangtze Estuary and the adjacent East China Sea (Fig. 1.) in March 2022 (the dry season of the Yangtze River). These sampling sites can be divided into three zones by salinity: freshwater zone (K1–K5), mixed zone (K6, K8–K11, K17), and seawater zone (K7, K12–K16, K18–K19).

Before being collected in stainless steel drums, water samples are pre-cleaned with methanol and mg of water. Water samples are kept in polypropylene (PP, 4 °C) bottles. Sediment sample collection

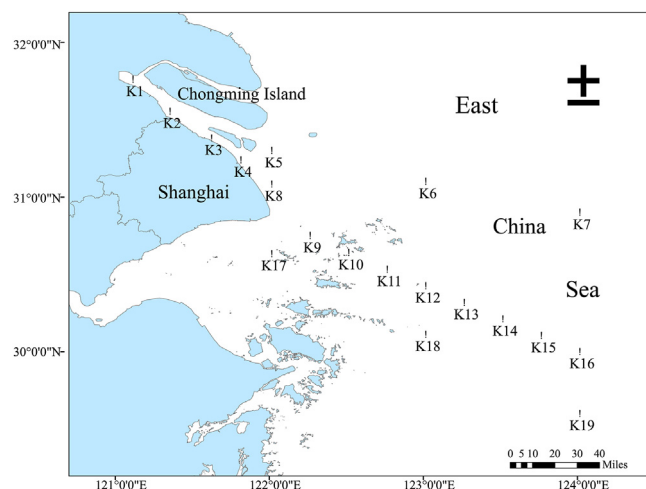


Fig. 1. Schematic graph of the sampling stations in Yangtze Estuary and coastal waters, China.

took place in the same location. Grab-type mud harvesters were used to gather sediment samples, which were then kept in polypropylene bags.

### 2.3. Sample preparation and analysis

A 0.45  $\mu\text{m}$  glass filter membrane was used to filter a 500 mL water sample, and an internal reference solution was added. A solid phase extraction column was installed with Styrene-divinylbenzene copolymer on the solid phase extraction apparatus as the filler or an equivalent column efficiency. Subsequently, 4 mL of a 0.1% ammonia/methanol solution, 4 mL of methanol, and 4 mL of ultrapure water were added. An average of one drop per second of the inner sample has flowed through the solid phase extraction column. The solid phase extraction column was cleaned with ammonium acetate buffer (4 mL 25  $\text{mmol L}^{-1}$ ). After being vacuum freeze-dried, the solid phase extraction column was washed with 4 mL of methanol and 4 mL of an ammonia/methanol solution. The eluent was then collected in a 15 mL polypropylene centrifuge tube. High-purity nitrogen concentrated the collected eluent and blow-dried it to almost dryness. The eluent was stored in a 1.5 mL brown injection vial through a 0.22  $\mu\text{m}$  nylon organic filter membrane to prevent blocking of Mass spectrometers and kept at 4 °C for LC-MS/MS analysis after 1 mL of the first mobile phase was redissolved.

A 10 mL acetonitrile solution was added, and  $10 \pm 0.1\text{g}$  of the damp sediment sample was deposited in a 50 mL centrifuge tube after being precisely weighed and undergoing a 30-min ultrasonic cleaning procedure. The samples were then put into syringes, filtered through 0.22  $\mu\text{m}$ , and kept in a 1.5 mL brown injection bottle at 4 °C for LC-MS/MS analysis after being centrifuged for 5 min at 4000 RPM.

### 2.4. Quality assurance and quality control

The standard solution was prepared into 0.01, 0.05, 0.1, 1.0, 2.0, 5.0, 10, 20, and 50  $\text{g L}^{-1}$  standard solutions, which were then mixed with the internal standard solution. For every 20 sample analyses, one blank sample, one parallel sample, one blank markup, and one sample markup were tested.

The recoveries of the substrates were calculated by introducing 10 ng and 100 ng standard compounds to a 500 mL water sample

( $n = 19$ ) and a 10 g sediment sample ( $n = 19$ ). Sediment samples recovered 71–102% (SD, 8.24%), and water samples recovered 87.5–105% (SD, 4.72%). Repeated samples' relative standard deviation was under 10%, and both methods meet the precision and accuracy requirements.

## 2.5. Calculation method

Coefficient of variation ( $C_v$ ) is used to characterize the spatial dispersion degree of organic pollutants [39], to analyze the variability of their content, as shown in Equation (1):

$$C_v = s/\mu \quad (1)$$

When  $C_v < 0.1$ , the variation is weak; When  $C_v$  is between 0.1 and 1, the variation is moderate. When  $C_v > 1$ , there is a significant variation [40].

The diffusion of contaminants between environmental media can be described by the escaping tendency in a particular medium, which Lewis defines as fugacity  $f$  in Pa. Contaminants always “escape” from environmental media with high fugacity to adjacent environmental media with low fugacity [41]. Therefore, the fugacity ratio can be used to illustrate the diffusion and migration direction of compounds:

$$f_s/f_w = C_{ps}/f_{oc}K_{ow}C_w \quad (2)$$

Where  $f_s$  and  $f_w$  are the fugacities in the sediments and waters, respectively,  $\rho_s$  is the density of the sediments ( $1500 \text{ kg m}^{-3}$ ), and  $K_{ow}$  is the octanol-water partitioning coefficient, obtained from the ECOSAR model software V 2.0 of Environmental Protection Agency (EPA).

When  $f_s/f_w > 1$ , it indicates that the contaminant is diffused from sediment to seawater. When  $f_s/f_w = 1$ , the contaminant is in equilibrium at the water-sediment interface. When  $f_s/f_w < 1$ , it indicates that the contaminant is diffused from seawater to sediment [40].

Equation (2) can be obtained from Equation (1), and fugacity fraction  $ff$  can be calculated:

$$ff = f_s / (f_s + f_w) = (f_s/f_w) / (f_s/f_w + 1) \quad (3)$$

When  $ff > 0.5$ , the diffusion trend is sediment to seawater. When  $ff = 0.5$ , the contaminant is in equilibrium at the water-sediment interface. When  $ff < 0.5$ , the diffusion trend is seawater to sediment. Both  $f_s/f_w$  and  $ff$  can be used as the basis to determine the diffusion direction of contaminants at the sediment-water interface. However, the variation range of the  $f_s/f_w$  ratio may be several orders of magnitude span, and the fugacity fraction is between 0 and 1, so it is convenient for comparison and statistical analysis.

An essential factor of the fugacity model is uncertainty [42]. The uncertainty of  $ff$  is affected by  $C_w$ ,  $C_s$ ,  $\rho_s$ , octanol-water partitioning coefficient ( $K_{ow}$ ), and  $f_{oc}$ . The error can be ignored because the variation of  $f_{oc}$  and  $\rho_s$  is slight [43]. Therefore, the uncertainty of  $ff$  can be calculated by Equation (3):

$$U_{ff} = \sqrt{ff \times (RSD^2(C_s) + RSD^2(C_w) + RSD^2(K_{ow}))} \quad (4)$$

Where  $RSD(C_s)$ ,  $RSD(K_{ow})$ , and  $RSD(C_w)$  are the relative standard deviations of  $C_s$ ,  $K_{ow}$ , and  $C_w$ , respectively.  $RSD$  is calculated from Equation (1), where  $s$  and  $\mu$  represent relative standard deviation and mean value, respectively.

## 3. Results and discussion

### 3.1. Concentration and partitioning of PFOS in water and sediment

PFOS concentration and comparison of water and sediment samples were given in Table 1 and Fig. 2. The detected PFOS concentration ranged from 0.053 to 0.95  $\text{ng L}^{-1}$ , with samples collected at K4 having the highest concentrations (0.95  $\text{ng L}^{-1}$ ). The K2 sample also contained a relatively high concentration of PFOS (0.70  $\text{ng L}^{-1}$ ). The sub- $\text{ng L}^{-1}$  concentration of PFOS detected in this study were significantly lower than those in the coastal waters of South Korea (4.11–450  $\text{ng L}^{-1}$ ), Japanese waters of Tokyo Bay (0.88–9.0  $\text{ng L}^{-1}$ ), coastal areas of Vietnam (40.2  $\text{ng L}^{-1}$ ), San Francisco Bay of the United States (42.3  $\text{ng L}^{-1}$ ), and waters of Shandong Peninsula, Bohai Bay of China (0.951–14.3  $\text{ng L}^{-1}$ ) and Liaodong Bay (<LOD–50.4  $\text{ng L}^{-1}$ ), the higher concentration of PFOS pollution in these areas is due to the discharge of nearby pollution sources. The coastal areas of Thailand (<LOD–5  $\text{ng L}^{-1}$ ), Baltic Seas (0.09–9  $\text{ng L}^{-1}$ ), Hong Kong (0.09–3.1  $\text{ng L}^{-1}$ ), and Dalian Wan (0.05–3.14  $\text{ng L}^{-1}$ ) had similar low concentrations of PFOS. The concentration of PFOS in a subtropical Gulf (0.06–0.21  $\text{ng L}^{-1}$ ) in the South China Sea was similar (see Table 2). Finally, the concentration of PFOS in the Yangtze Estuary and coastal waters was relatively low, and the spatial coefficient of variation was 0.50, corresponding to moderate variation intensity in seawater.

The range of PFOS concentrations found in sediment samples was 0.031–0.18  $\text{ng g}^{-1}$ , with sample K5 having the most significant concentrations (Table 1). Its concentration was similar to that of PFOS in sediments of South Korea (0.01–0.48  $\text{ng g}^{-1}$ ), Dalian Bay (0.03–0.17  $\text{ng g}^{-1}$ ), and a subtropical bay of the South China Sea (0.04–0.20  $\text{ng g}^{-1}$ ), and was a lower contaminant concentration than that of Tokyo Bay (0.13–1.4  $\text{ng g}^{-1}$ ), San Francisco Bay (<LOD–3.07  $\text{ng g}^{-1}$ ), Baltic seas (0.02–2.4  $\text{ng g}^{-1}$ ), German Bight, North Sea (0.023–5.4  $\text{ng g}^{-1}$ ), Shandong peninsula (China) (0.21–5.25  $\text{ng g}^{-1}$ ), and Bohai Bay (0.682–5.50  $\text{ng g}^{-1}$ ) (see Table 2). The amount of PFOS in the sediments of the Yangtze Estuary stayed low, and the spatial variation intensity was 0.96, which denoted a medium level of variation. However, compared to PFOS variation intensity in marine water, PFOS variation intensity in sediments was close to strong, indicating that PFOS has a more significant spatial dispersion in sediments.

We collected data on PFOS concentration in the waters of the

**Table 1**

Water salinity, PFOS in water and sediment samples, TOC content in sediment, and fugacity fraction  $ff$ .

Site	$C_w$ ( $\text{ng L}^{-1}$ )	$C_s$ ( $\text{ng g}^{-1}$ )	Salinity	TOC ( $\text{mg g}^{-1}$ )	$ff$
K1	0.51	0.031	0.135	6.69	0.3069
K2	0.70	0.15	0.136	12.2	0.4535
K3	0.57	0.036	0.140	6.75	0.3124
K4	0.95	0.053	1.58	6.80	0.2842
K5	0.47	0.18	0.197	8.55	0.6795
K6	0.14	0.093	28.3	12.6	0.7216
K7	0.10	0.076	33.6	7.41	0.8342
K8	0.23	0.12	16.9	11.4	0.6781
K9	0.22	0.15	28.0	14.0	0.7045
K10	0.19	0.13	29.5	14.6	0.6936
K11	0.20	0.14	28.4	16.2	0.6670
K12	0.12	0.092	33.8	11.3	0.7663
K13	0.07	0.055	33.9	7.18	0.8377
K14	0.057	0.044	34.7	7.92	0.8255
K15	0.076	0.059	34.2	6.56	0.8517
K16	0.053	0.039	33.6	6.62	0.8435
K17	0.19	0.13	28.8	12.6	0.7210
K18	0.089	0.067	34.0	7.46	0.8303
K19	0.081	0.063	34.1	7.68	0.8309

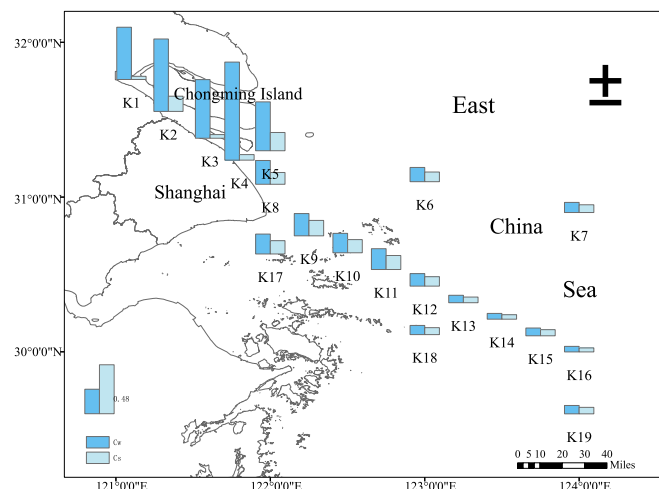


Fig. 2. Concentrations of PFOS in water and sediment of this work.

Yangtze River Estuary over the years, and compared with the results of the samples collected in 2021, the concentration of PFOS in the water body showed an overall decreasing trend over time. In 2003, the concentration of PFOS ranged from 0.1 to 37.8 ng L<sup>-1</sup> [6]; in 2005, it was <LOD-14 ng L<sup>-1</sup> [52]; in 2013, it was 0.02–4.47 ng L<sup>-1</sup> [62] and <LOD-3.93 ng L<sup>-1</sup> [35], and the concentration range in this study was 0.053–0.95 ng L<sup>-1</sup>. Due to manufacturers' voluntary phase-outs, associated policy limitations by the Chinese government, and the fact that PFOS was relatively stable in the environment, decreased source release may be the primary cause of the observed low concentration [63]. However, the risks still cannot be ignored. The minor trend in the concentration of PFOS in sediments over time scales is related to the relative stability of sediment properties.

Table 2  
Comparison with other countries and regions.

Locations	Water concentration (ng L <sup>-1</sup> )	Sediment concentration (ng g <sup>-1</sup> dw)	Refs.
South Korea	4.11–450	–	[14]
	–	0.01–0.48	[44]
Tokyo Bay (Japan)	0.88–9.0	0.13–1.4	[45]
Chao Phraya River and Bangpakong River (Thailand)	<LOD-5	–	[46]
Coastal areas in Vietnam	40.2	–	[47]
San Francisco Bay	42.3	–	[48]
	–	<LOD-3.07	[11]
German Bight, North Sea	–	0.023–5.4	[49]
Baltic seas	0.09–9	–	[50]
	–	0.02–2.4	[51]
Pearl River Delta China Sea (including South China Sea)	0.02–12	–	[52]
	–	0.86	[53]
East China Sea	–	32.4	[54]
Shandong peninsula (China)	4.2–25.4	0.21–5.25	[55]
Hong Kong	0.09–3.1	–	[56]
Dalian Bay	0.05–3.14	0.03–0.17	[57]
Bohai Bay	0.951–14.3	0.682–5.50	[58]
Liaodong Bay	<LOD-50.4	–	[59]
a subtropical Gulf, South China Sea	0.06–0.21	0.04–0.20	[26]
Laizhou Bay (China)	–	0.02–1.6	[60]
Along the Yangtze River	0.1–37.8	–	[6]
	<LOD-14	–	[52]
	–	<LOD -0.46	[61]
	0.02–4.47	–	[62]
	<LOD -3.93	<LOD-0.59	[35]
	–	<0.03–0.20	[9]
Yangtze Estuary	0.053–0.95	0.031–0.18	This study

### 3.2. Diffusion trend of PFOS at the water-sediment interface

Equation (2) can be used to determine the diffusivity *ff* value of PFOS between water and sediment at each site. When *ff* = 0.5, the relative standard deviation *Uff* is 0.18, so the equilibrium range of fugacity *ff* is 0.32–0.68. When the *ff* value exceeds 0.68, contaminants are likely moving from sediments to water bodies. In the water-sand interface, the pollutants typically balance when *ff* is between 0.32 and 0.68. When *ff* is less than 0.32, it indicates that contaminants frequently migrate from water to sediment. The findings demonstrated that in the K1, K3, and K4 sites, PFOS spread from sediment to water (Fig. 3). These three stations are in freshwater regions and are near the ocean. Contaminants diffuse from the water body to the sediment because fresh water discharged into the estuary brings a large number of contaminants into the Yangtze Estuary, and there was a high concentration of pollutants in the water body before adequate mixing, adsorption, and dispersion into the sediment. According to the findings, terrigenous pollution—where sewage discharge and surface runoff play significant roles—is the predominant type of pollution in the Yangtze Estuary area. The PFOS *ff* values at K2, K5, K8, and K11 ranged between 0.32 and 0.68, indicating that the water-sediment interface was in dynamic equilibrium. PFOS appeared to enter the water from sediment at the other nine sites, where *ff* values were greater than 0.68. PFOS adsorbed more in the sediments at these locations, which could be due to the local influence of different ocean currents on sediment deposition, such as the Zhejiang-Fujian coastal current, Kuroshio current, and Yangtze River diluted water [64].

When PFOS diffuses from sediments to water, PFOS may be a potential secondary release source to the water. As previously reported, some studies have described sediment as a secondary source. The prevailing consensus is that surface sediments are important sinks or potential secondary sources of PFOS [65]. Environmental behavior surveys in Lake Tyrifjorden indicated that hydrophobic precursors of PFOS had been transformed in sediments and are a significant contributor to PFAA buildup in top

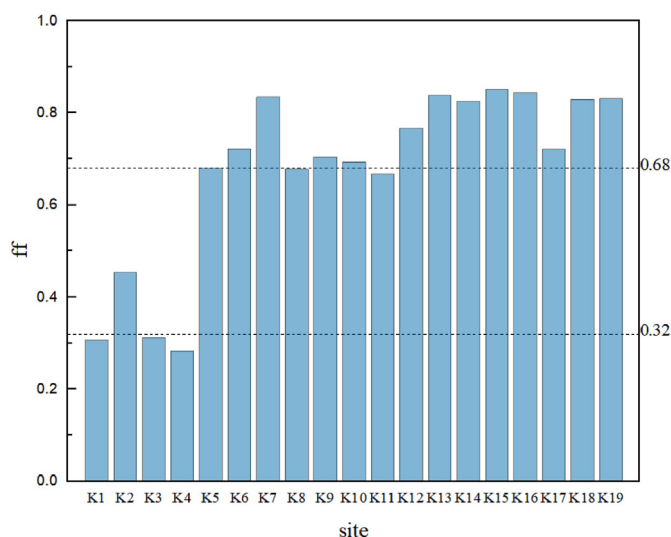


Fig. 3. Fugacity fractions (ff) of PFOS in the Yangtze Estuary and coastal waters.

predator fish [66]. Similarly, the dispersal of persistent organic pollutants (PAHs) in water-sediment showed the same tendency, and the PAHs concentrations in pore water were influenced by the sediment environment [67].

### 3.3. Effect of TOC on the partitioning behavior of PFOS

The total TOC in the Yangtze Estuary ranged from 6.56 to 16.23 mg g<sup>-1</sup>, with an average value of 9.68 mg g<sup>-1</sup>. Pearson correlation analysis indicated that TOC and PFOS content in sediments were significantly positively correlated (R<sup>2</sup> = 0.76, p < 0.01). Similar to the findings of this research, which found that the adsorption capacity of anionic perfluorinated surfactants on the sediment was positively correlated with the TOC content in the sediment [33]. Similarly, PFOS content was positively correlated with TOC in sediments from the East China Sea [68], Bohai Bay [32], and the North and Baltic Sea [51].

When the physicochemical properties of PFOS are constant, the fugacity fraction ff values of PFOS is significantly different when TOC content changes within a specific range. When the minimum, maximum and average ff were used, the ff values were 0.69, 0.25, and 0.45, respectively. Therefore, the variation of ff would affect the partitioning and diffusion of PFOS between water and sediment. The response coefficient (RC) can be used to describe the degree of influence of TOC content on PFOS partitioning behavior in sediment water [69]. The calculation formula is as follows:

$$RC = (ff_{W(OC)min} - ff_{W(OC)max}) / ff_{W(OC)mean} \quad (5)$$

ff<sub>W(OC)min</sub>, ff<sub>W(OC)max</sub>, and ff<sub>W(OC)mean</sub> are fugacity fraction values of minimum, maximum, and average organic matter content, respectively. Moreover, their values are respectively 0.79, 0.61, and 0.32. The larger the RC is, the more the TOC content affects the pollutant diffusion process [39].

The impact of TOC content on the pollutant diffusion process increases with the response rate (RC), which measures this effect. In this study, the RC calculated result is 0.26, which is the intermediate-range RC value (0.0002–0.42) of perfluoroalkyl substances with different chain lengths in the literature, indicating that the content of organic carbon greatly influences the enrichment and diffusion of PFOS in water-sediment. This result was similar to perfluorinated compounds in German Bay, where TOC strongly

influenced the adsorption of PFOS in coastal areas with homogeneous sediments [49]. The PFOS adsorption to marine sediments in saltwater was potent and nonlinear (10 times greater than that in freshwater), and the hydrophobic interaction was crucial to the adsorption process [70]. The TOC in sediments encouraged the buildup of PFOS, which affected the diffusion behavior between seawater and sediments.

### 3.4. Effect of salinity on the partitioning behavior of PFOS

One might utilize the partitioning coefficient K<sub>d</sub> to assess how organic contaminants in the aquatic environment partitioning. Salinity and several organic contaminants' partitioning coefficients have been shown to significantly positively correlated [71]. The behavior and outcome of organic pollutants in the environment would depend on how they are partitioned between water and sediment. K<sub>d</sub> is calculated using the following formula:

$$K_d = C_s / C_w \quad (6)$$

K<sub>d</sub> values in this study ranged from 0.055 to 0.78 L g<sup>-1</sup>, with K5 and K17 serving as the lowest and highest points, respectively. The present study showed a higher value of K<sub>d</sub> for PFOS than in the Llobregat river basin (0.02 L g<sup>-1</sup>) in Spain [72]. However, it agreed well with the reported values from the coast of New South Wales (0.0038–0.103 L g<sup>-1</sup>) in Australia [73], Bohai coastal watersheds (0.05 L g<sup>-1</sup>) in China [74], Tokyo Bay (0.13 L g<sup>-1</sup>) in Japan [7] and Taihu Lake (0.04–0.71 L g<sup>-1</sup>) in China [75]. K<sub>d</sub> values, except for K1–K5, showed a positive correlation with salinity, as shown in Fig. 4. With increased seawater salinity, the partitioning coefficient K<sub>d</sub> rose. The adsorption of PFOS on sediments would be aided by increased salinity, and a similar phenomenon has been confirmed in Bohai Bay [32]. Therefore, the adsorption and resolution of PFOS between sediments and seawater were strongly influenced by salinity, which can be explained by the salt-out effect and the Ca-bridging sorption mechanism [30]. Some studies also suggested that hydrophobic and electrostatic interactions were crucial for PFOS sorption onto sediment [76]. Some scholars believed that in salinity < 10, the relationship between freshwater sediments and PFOS partitioning was fragile, and the K<sub>d</sub> value was not correlated with PFOS partitioning [77]. This finding also explains why K<sub>d</sub> and

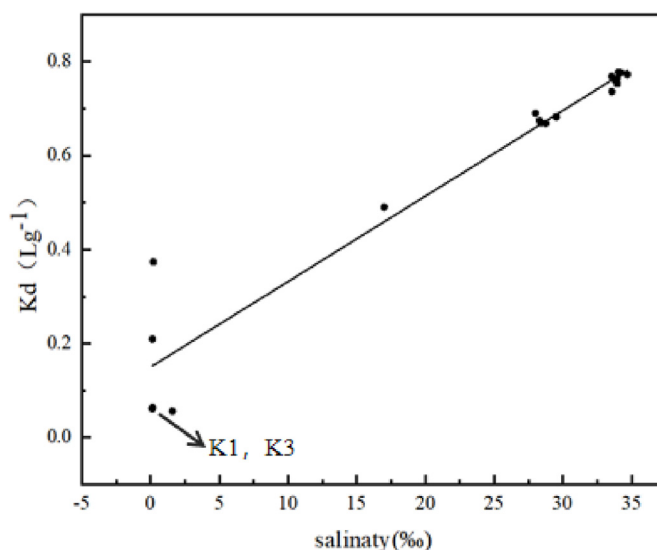


Fig. 4. Relationship between salinity and partitioning coefficient (K<sub>d</sub>) of PFOS between water and sediment.

salinity at the K1–K5 stations in our analysis only has a minor connection. Salinity at the K1–K5 station was only 0–1, the sediment was currently freshwater, close to the source of pollution, and the concentration partitioning was irregular.

There were also some varied conclusions when comparing the correlation between salinity and the concentration of PFOS contaminant in different areas. In all the sampling points, K6, K9, and K17 were located on the same cross-section, and the salinity range is between 28 and 29. K7, K13, and K18 lied on the same cross-section farther offshore, with salinity ranges of 33–34. We could see that the salinity of the water was constant across a cross-section and rises with distance from the shore on different parts. As illustrated in Fig. 5, the concentration of PFOS in the cross-section of K6, K9, and K17 with lower salinity was higher than that in the cross-section with higher salinity. The concentration of PFOS in both water and sediment decreased with the increase in salinity. This phenomenon was at odds with the reported finding that PFOS concentration in water decreases with an increase in salinity, whereas PFOS concentration in sediment increases with an increase in salinity [78]. This phenomenon may be caused by the dilution of far sea water in the study area, which decreased pollutant concentration in both water and sediment.

### 3.5. Preliminary environmental risk assessment of PFOS

Persistent organic pollutants are increasingly being seen in coastal fish species, and ingesting them has been shown to have harmful physiological consequences on people [79] and other living things [80]. Due to their hydrophobic, oleophobic, bioaccumulative, and environmental persistence, PFOS can build up in sediments, where it can then be consumed by benthic organisms and passed on to animals with greater nutrient levels, with non-negligible effects on ecosystems, benthic organisms, and so far as to harm human health [77].

To get the risk entropy (RQ) of a single perfluorinated molecule, use the Equation below [25]:

$$RQ = MEC / PNEC \quad (7)$$

MEC and PNEC represent measured ambient concentrations and expected no-effect concentrations, respectively. For seawater, the PENC threshold for PFOS derived by Qi et al. (2011) was between 0.61 and 6.66  $\mu\text{g L}^{-1}$  [81]. Furthermore, the European Commission

set an environmental quality standard (EQS) of 0.21  $\mu\text{g L}^{-1}$  for PFOS in seawater to protect aquatic pelagic organisms in 2013 [82]. The PENC values in seawater were minimized (0.21  $\mu\text{g L}^{-1}$ ) to provide the most sensitive risk assessment. When toxicity data is unavailable, sediment PNEC can be calculated using the equilibrium partitioning method [83]. Considering that the environmental conditions of the Changjiang Estuary were similar to Laizhou Bay, we chose the PENC in Laizhou Bay (0.49  $\text{ng g}^{-1}$ ) as the PENC reference for the sediments [60].

Risk assessment is represented by RQ values, which are divided into four levels: unlikely to pose a risk ( $RQ < 0.01$ ), low risk ( $0.01 < RQ < 0.1$ ), medium risk ( $0.1 < RQ < 1$ ), and high risk ( $RQ > 1$ ) [84].

In order to evaluate the environmental and ecological dangers in this area, the Yangtze Estuary's PFOS RQ values were established. The concentration of PFOS in sediments along the Yangtze Estuary can be considered safe with little risk to nearby aquatic life, according to the RQ values of PFOS in water ( $2.52 \times 10^{-4}$ – $4.54 \times 10^{-3}$ ), which were substantially lower than 0.01. For RQ in sediments (0.063–0.36), 79% of sampling sites were higher than 0.1 but less than 1 (medium risk), and 21% were higher than 0.01 but less than 0.1 (low risk). This result indicated that PFOS in sediments has a particular risk to the aquatic environment, which should be paid attention to, and long-term monitoring should be carried out. Moreover, the accumulation of PFOS in biota and its migration up the food chain should not be ignored, and some studies have shown that PFOS has a biological amplification phenomenon in the food web [85].

## 4. Conclusions

According to the findings, PFOS was found in the water and sediment of the Yangtze Estuary and coastal waters. PFOS concentrations in water and sediment are 0.053–0.95  $\text{ng L}^{-1}$  and 0.031–0.18  $\text{ng g}^{-1}$ , indicating apparent enrichment in environmental media. The concentration gradient of PFOS in water and sediments was pronounced from inshore to offshore stations, and the primary source of these two compounds was freshwater flowing into the Yangtze Estuary. PFOS reached equilibrium at the water-sediment interface and diffused from sediments to seawater, implying that PFOS in sediments could be a secondary source in the water environment. TOC and PFOS concentration in sediments had a significant correlation ( $R^2 = 0.76$ ,  $p < 0.01$ ), and seawater salinity affected PFOS adsorption in sediments, both of which had significant effects on the spatial partitioning and fate of PFOS in the water-sediment system. In addition, preliminary environmental risk assessments indicated that since PFOS in sediments may present a low to moderate risk at sampling points, more research is needed on its ecological risks and negative impacts. The results of this study will help us better understand how PFOS behaves in the ecosystem and where it ends up in water and sediment. They will also help control emerging contaminants in estuarine areas.

### Credit authorship statement

Jing Chen: Conceptualization, Analyzing the data, Original Draft writing; Hong Huang: Supervision, Study Design, Writing-Review & Editing, Funding acquisition; Jinchen Yu: Field investigation; Ruijing Li, Zhiming Hu, Xu Zhao, Yang Li, and Wenqian Wang: sample analysis and data interpretation. All authors read, edited, and approved the submitted version of the article.

### Declaration of competing interest

The authors declare no competing financial interest.

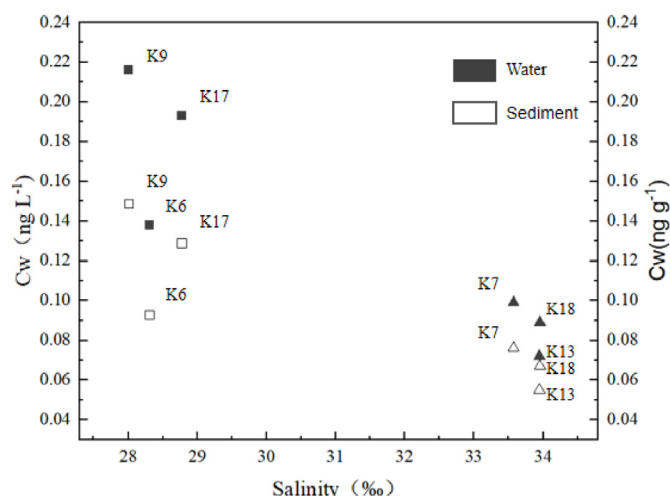


Fig. 5. Concentrations of PFOS in water and sediment at different salinity.

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