Perfluorooctane Sulfonate (PFOS) Distribution in the Water and Sediment of the Yellow River Estuary, China

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Abstract The distribution of perfluorooctane sulfonate (PFOS) was investigated in the water and sediment of the Yellow River Estuary (YRE) in the total of 15 water and sediment samples from the Yellow River Estuary (YRE), China. The results indicated that the concentrations of PFOS in the water and sediment samples averaged 157.5 ng/L and 198.8 ng/g, and ranged from 82.30 to 261.8 ng/L and 75.48 to 457.0 ng/g, respectively. The concentration of PFOS in the sediment column increased from 45.32 to 379.98 ng/g with the decrease of the sampling depth, which showed that the increased PFOS pollution in the sediment appeared in over recent years in this region. The distribution coefficient (K_d) of PFOS between water and sediment linearly increased from 0.37 to 4.80 L/g as the salinity (S‰) increased from 0.18 to 4.47. Correlations analysis revealed that the K_d was significantly and positively correlated to the contents of total organic carbon (TOC) and clay of the sediment and salinity. From the results of this study, salinity was an important parameter in controlling the sediment-water interactions and the fate or transport of PFOS in the aquatic environment. The estuary was an important sink for PFOS, which suggested that PFOS might be carried with the river water and transported for long distances before it reached to the sea and largely scavenged to the sediment in the estuaries due to the change in salinity.

Key words: Perfluorooctane sulfonate (PFOS), Distribution, Salinity, Water, Sediment, Estuary

1. Introduction

Perfluorooctane sulfonate ($PFOS^-$) has been a focus of concern because of its persistence in the environment (Giesy and Kannan 2002), retention in human body (Olsen et al. 2007), and

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toxicities (Giesy and Kannan 2002; Hekster et al. 2003; Ankley et al. 2004). Previous studies have shown that PFOS is ubiquitously detected in the environment, even in remote arctic areas (Giesy and Kannan 2001; Hekster et al. 2003). Perfluorooctane sulfonate is chemically and thermally stable, and is considered to be the terminal degradation product of many perfluorochemicals (PFCs) (Development 2002). Because of the potential bioaccumulation and the possible negative impact on the total environment and ultimately humans (Beach et al. 2006), it is urgent to investigate the environmental fate of PFOS. Several direct and indirect sources of POFS-emission to the environment have been reported, such as manufacturing processes in the polymer and electronics industries (Prevendouros et al. 2006), in the degradation of volatile precursors (Ellis et al. 2004; Dinglasan et al. 2004; Lange 2000), or in the release of treated waste waters (Becker et al. 2008; Boulanger et al. 2005; Schultz et al. 2006a; Sinclair and Kannan 2006).

Sediment and the deep oceans are the two sinks of PFOSs (Prevendouros et al. 2006). Some studies have observed PFOS in sediments and sludge (Higgins et al. 2005; Sinclair and Kannan 2006). Field data from rivers in America and Japan showed that the concentrations of PFOS in water were detectable (Hansen et al. 2002; Senthilkumar et al. 2007). Martin et al. (2004) suggested that sediment was the major sources of perfluoroalkyl introduction into the food web in Lake Ontario. Consequently, this puts forward an important issue on the bioavailability of sediment-bound PFOS, whether sediments serve as long-term sources of PFOS to the biosphere. Therefore, understanding the distribution mechanisms of PFOS in the water-sediment system is very important in aquatic environment. Unfortunately, most of the earlier studies have focused on water and biological matrices; little information is available on PFC concentrations in sediment. However, river is an important pathway for the transport of PFOS from lands to oceans. The partitioning of PFOS between water and sediment is crucial for understanding the transport/transform mechanism of PFOS in the environment.

As the second long river in China, Yellow River flows into the Bohai Sea in Dongying, Shandong Province in China. The Yellow River Estuary (YRE) area is one populous area with many kinds of human activities. Organic pollutants and saltwater are the major causes to water pollution in these areas (Hui et al. 2009). Seawater intrudes into the YRE in low flow season, which changes the salinity under different hydrological conditions. To study the salinity effect on

PFOS distribution in the YRE may reveal important mechanism in understanding the distribution and transport mechanisms of PFOS. However, detailed spatial distribution and partitioning mechanisms of PFOS in the water-sediment system of the Yellow River Estuary have not been elucidated.

The purpose of this study was to comprehensively elucidate the spatial distribution and partitioning of PFOS in the water and sediment of the YRE in China. The effect of salinity on the distribution of PFOS between the water and sediment in the estuary was determined. The distribution coefficient (K_d) of PFOS between water and sediment is drawn to analyze the effects of salinity on the distribution and fate of PFOS. Effects of environmental parameter such as sediment compositions including sediment organic matter and inorganic fractions are also studied.

2. Materials and Methods

2.1 Sediment and water sampling

Water and sediment samples were collected in April 2011 from the YRE in China (Fig. 1). Water samples were collected from each location using a stainless steel bucket, precleaned using methanol, Milli-Q water, and the water from specific location. The water samples were characterized using water quality survey instrument (556 Handheld Multiparameter, YSI, USA) and stored in polypropylene (PP) bottles at 4 ± 2°C before extraction. Sediment samples were collected from 0 to 10 cm depth at the same sampling locations, using cable operated sediment samplers (Van Veen grabs, Eijkelkamp). A sediment column of 50 cm downward from the sediment surface at the site Y14 was collected, and it was divided into five sections at each 10cm depth from sediment surface. These sediment samples were placed in polypropylene bags and transported in icebox at -4 ± 2°C to the laboratory. Before analysis, the sediment samples were air-dried, sieved with 0.25 mm stainless mesh, and dried at 103°C for 4 h to eliminate moisture.

Figure 1 appears here

2.2 Chemical analysis

In this study, water and sediment samples were prepared and analyzed as described previously (Pan and You 2010). Briefly, for determination of total concentrations of PFOS in water, about 100 mL aliquots of the samples were filtered through 0.22 μm fiberglass membranes before extraction to remove sediments and biota. All the water samples were extracted by solid phase extraction (GracePure^{TM} SPE C-18-MAX, 6 mL, 500 mg, 30 pk) (Grace, Illinois, USA). The SPE cartridge was first preconditioned by passing through 10 mL of methanol and 10 mL of 1% acetic acid at a rate of two drops per second in sequence. Depending on the concentrations of PFOS in the water samples, 200 mL, 500 mL or 1 L filtered water was passed through the preconditioned cartridge at a rate of one drop per second. It was then rinsed with 10 mL Milli-Qwater. The cartridges were dried under vacuum for at least two hours in order to remove the water in the cartridge completely to maintain the concentration efficiency in later processes. The target analyte was eluted by 5 mL methanol at a rate of one drop per second and collected in 1:1 (v/v) methanol/acetone-washed 10 mL PP tube. The eluent was dried under a stream of high purity nitrogen and fixed to 5 mL or 2 mL by adding methanol.

Freeze-dried sediment samples of about 1.0 g was extracted by adding 7.5 mL of 1% acetic acid, treated using ultrasonic at 60^{o}C for 15 min, and then centrifuged at 3000 rpm for 10 min. The supernatant was collected in a precleaned 50 mL PP bottle, and the residual sediment was followed by further treatment. 1.7 mL mixture of 9:1 (v/v) methanol and 1% acetic acid was added to the tube, processed using ultrasonic at 60^{o}C for 15 min, and then centrifuged at 3000 rpm for 10 min. The supernatant was also collected and added to the PP bottle mentioned above. These operations were repeated three times. In order to enrich the analyte and remove the acetic acid, salts, and potential matrix interferences, the extracted solution was loaded to SPE cartridges. All the prepared solutions were stored at 4^{o}C prior to PFOS analysis. Extracted solution was transferred to 100 mL polypropylene insert with polymer feet in 2 mL HPLC/GC auto sampler vial.

The PFOS concentrations of water and sediment samples were analyzed by High Performance Liquid Chromatography/Hybrid Quadrupole Time-of-Flight Mass Spectrometer (HPLC/Q-ToF MS). Separation of the analyte was performed by an Alliance 2695 (Waters, USA), interfaced with a quadrupole time-of-flight mass spectrometer (Micromass, Manchester, UK) with a Z-spray ESI source working in negative mode. A 10 mL aliquot of extract was injected into a reversed-phase C18 column (2.1 mm×150 mm, packed with particle size of 5 mm) (Waters, USA) with 10 mmol/L ammonium acetate and methanol in 55:45 (v/v) as the mobile phases at a flow rate of 0.2 mL/min. The mass tune employed the optimized parameters described by Guo et al.

(2008). The mass spectrometer parameters were optimized to transmit the [M¯] ion for PFOS and MPFOS using atmospheric pressure ionization, operated in the electrospray negative ion mode. PFOS and MPFOS ion was monitored using selected reaction monitoring for quantitative determination.

2.3 Quality assurance and control

The precision and accuracy of the extraction and the analytical procedures were determined by spike and recovery experiments. The method blank was checked by analyzing one blank sample for each sampling, and the analytes were not detected in these samples. The results showed that the preparation and analytical procedure did not introduce any impurities. The method recovery test was conducted by adding 10-100 ng/L standard solution. The recovery rates were in the range of 96.4-98.2%, which meant that there was little loss of PFOS in the extract procedure. Standard solutions containing PFOS and MPFOS analyte were used for calibration. The standard addition experiment was performed by adding 2-50 ng/L standard PFOS to the samples. Method recovery rate ranged from 93.26 to 112.82%, and relative standard deviation (RSD) ranged from 0.53 to 11.33%. These results showed acceptable stability of the samples over the course of analysis. The limits of quantification (LOQ, signal to noise ratio = 13.23) for sediments were 0.05 ng/g for sediment (dry weight), and 0.50 ng/L for river water. The detection limit of the instrument used for the standard PFOS solution was 0.2 μg/L and was 0.5 μg/L for water and sediment samples respectively.

3. Results and discussion

3.1 Water and sediment characterization

A selection of general physical and chemical characteristics of the water and sediment are shown in Table 1. The pH values of the studied water samples were in the range of 6.46-8.33, with an average value of 7.55, displaying an alkaline character. The salinity (S‰) of the water was in the range of 0.18-4.47‰. The amount of total organic carbon (TOC) in sediment ranged from 1.15 to 6.95%. The percentages of clay and silt were in the ranges of 7.49-20.53% and 28.89-58.62%, respectively, with mean values of 13.13 and 41.23%.

Table 1 appears here

3.2 Concentrations of PFOS in water and sediment of the YRE

The PFOS concentrations in the water samples ranged from 82.3 ng/L at site Y9 to 261.8 ng/L at site Y1, with a mean value of 157.5 ng/L (Fig. 2). In this study, the PFOS concentrations in water at the sites Y1-Y5 are generally higher than other water samples. Whereas the PFOS concentrations in water at the sites Y6-Y9 is relatively low. There are big industrial bases (Dongying and Hekou) in south of the sites Y1-Y5 and north of the sites Y10-Y14 but a rural setting and large-scale farms along the sites Y6-Y9. Therefore, the urban runoff, industrial and municipal wastewater effluents might be large contributors to the contamination of PFOS at sites Y1-Y5 and Y10-Y14. Moreover, the PFOS concentrations at the sites Y1-Y5 are obviously higher than that at the sites Y10-Y14, that likely owing to the rapid urbanization and industrialization in the region around the sites Y1-Y5 and high silt content in the river sediment where the sites Y10-Y14 lies. The highest concentration of PFOS in water was observed at the site Y2 (261.8 ng/L). The site Y2 is near the Lijun County, significant amount of domestic sewerage from the nearby town may be large contributors to the high content of PFOS at this site. However, the PFOS concentration in water sample of the site Y15 was lower than other samples, which was because this site is near the mouth of Yellow River and terrestrial input of PFOS is diminished. Therefore, the observed high value of PFOS in water samples of the YRE may be induced by industrial and municipal influences discharged from the regions around the river mouth.

Figure 2 appear here

Previous study reported that the highest concentration of PFOS in the water of the Yangtze River Estuary was 703 ng/L (Pan and You 2010), which was much higher than that of the site Y1. Whereas the mean value of PFOS concentration in the water samples of the YRE (n=4) was 213.3 ng/L (Pan and You 2010), which was higher than that in the YRE (167.3 ng/L). Except the high value at site Y1, the PFOS concentrations detected for other sites were comparable with values reported in sixteen Japanese rivers (0.2-157ng/L) (Satio et al. 2003) and in Great Lakes (21-70 ng/L) (Boulanger et al. 2004). The PFOS concentrations investigated in this study were much greater than that detected in Huangpu River (0.62-14ng/g) (So et al. 2007). There was previous study reported that domestic sewage contained high levels of PFOS (Hekster and Voogt 2003).

As shown in Fig. 2, the concentrations of PFOS in the sediment of the YRE were in the range of 75.48-456.98 ng/g, with an average of 198.81 ng/g. The spatial distribution of PFOS in the sediment didn't show the tendency as that of in the water. The sample from the site Y15 had the highest concentration of PFOS, which was higher than the reported concentrations of PFOS in sediments or soils for other regions of the world (Hekster and Voogt 2003; Higgins et al. 2005; Voogt and Roon 2005). Pan and You (2010) reported that the PFOS concentrations in the sediments of the YRE was in the range of 72.9-536.7 ng/g, with an average value of 236.2 ng/g. Higgins et al. (2005) observed that the PFOS concentrations in domestic sludge from USA were in the range of 14.4-2610 ng/g. Gou et al. (2008) the PFOS concentrations in sewage sludge from sixteen cities of China were in the range of 278-5383 ng/g.

The concentrations of PFOS in the sediment column from the site Y14 were shown in Fig. 3. The PFOS concentrations ranged from 45.32 to 379.98 ng/g with decreasing in the sampling depth. Suspended matters in water of the YRE deposited at a general rate of 1-2 cm per year (Hu et al. 2011). Therefore, the result of this study showed that the increased PFOS pollution in the sediments appeared in over recent years in this region.

Figure 3 appears here

3.4 Distribution of PFOS between water and sediment of the YRE

The distribution coefficient (K_d) of PFOS between water and sediment was calculated by the formula presented below:

$K_d=C_s/C_w$

where C_s and C_w were the concentrations of PFOS in sediment sample (ng/g) and in water sample (ng/L), respectively.

In this study, the value of K_d ranged from 0.37 to 4.80 L/g. Correlation analysis showed that K_d was significantly and positively correlated to the salinity (Pearson Correlation=0.9476, n=15) (Fig.4a). You et al. (2010) observed that PFOS could be increasingly sorbed by the suspended particles and the sediment as the salinity of overlying water increased. Development (2002) observed that potassium salt of PFOS has a solubility of 25 mg/L in filtered seawater, but 370 mg/L in freshwater. Some previous studies also found that the K_d of some organic pollutants was positively correlated with salinity (Brunk et al. 1997; Luo et al. 2008).

Figure 4 appears here

From the results of this study, at high salinity area, the concentrations of PFOS were high in sediment but relatively low in water. In contrast, at low salinity area, the concentrations of PFOS were low in sediment by high in water. For example, at low salinity of site Y2, the PFOS concentration was high in water but relatively low in the sediment. In contrast, at a higher salinity of site Y13, the PFOS concentrations in water were low but relatively high in the sediment. Therefore, the results of this study might confirm that salinity had an important effect on distribution of PFOS between the water and sediment; and PFOS could be largely scavenged into the sediment in the estuary area due to the increased sorption as salinity increased.

Because of the specificity of estuarine environment, an important consideration for water quality management is necessary (Brunk et al. 1997). The results from this study suggested that the affinity of the sediment for PFOS would be higher during salt intrusion, and PFOS might be carried with the river water and transported for long distances before it reached to the sea and largely scavenged to the sediment in the estuaries due to the dramatic change in salinity.

The correlation analysis between K_d and TOC, clay were shown in Fig. 4(b,c). The correlation analysis between K_d and silt, sand were not shown in this study. The correlation coefficients between K_d and TOC, clay, silt, and sand were 0.90 and 0.26, 0.001, and -0.05, which suggested that the enrichment of PFOS on the sediment was significantly and positively correlated to the contents of TOC. This result is similar with that described by Higgins and Luthy (2006), but different from observed by Becher et al. (2008). From the result of this study, the TOC content of the sediment contributed more to the sorption of PFOS than other constituents of sediment mentioned in this study.

4. Conclusions

This study showed that PFOS was detectable in all water and sediment samples of the YRE. The concentrations of PFOS in the water and sediment samples ranged from 82.3 to 261.8 ng/L and 75.48 to 456.98 ng/g, respectively. In water samples, the high concentrations of PFOS were observed in the area with high levels of industrial activities. But, for sediment samples, the high concentration of PFOS was mainly observed near the estuarine area. Salinity had an important effect on the transport and distribution of PFOS in aquatic environment. At low salinity area, the concentration of PFOS was high in water but relatively low in the sediment. In contrast, at high salinity area, the concentration of PFOS in water was low but relatively high in the sediment. As the salinity (S‰) increased from 0.18 to 4.47, the distribution coefficient (K_d) between sediment and water linearity increased from 0.37 to 4.80 L/g. The concentrations of PFOS in the sediment column increased from 45.32 to 379.98 ng/g with the sampling depth decreasing from 40-50 cm to 0-10 cm, which showed an enrichment trend of PFOS into the sediment over the recent years. The results of this study suggest that PFOS can be largely scavenged to the sediment in estuary due to the dramatic change in salinity.

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Table 1 Summary of sample characteristics and concentration of PFOS in water and sediment samples

Sampling Sites	Water sample		Sediment sample			
	pH	Salinity (‰)	TOC (%)	Clay (%)	Silt (%)	Sand (%)
Y1	7.87	0.31	1.94	10.34	28.89	60.77
Y2	8.11	0.15	1.54	11.68	35.69	52.63
Y3	8.32	0.25	1.15	18.98	42.57	38.45
Y4	7.93	0.58	2.15	10.35	39.85	49.8
Y5	8.33	0.42	1.64	9.65	52.36	37.99
Y6	7.69	1.12	3.65	10.85	44.35	44.8
Y7	7.85	1.34	2.64	12.35	36.05	51.6
Y8	7.49	1.42	3.25	16.48	42.68	40.84
Y9	7.48	1.82	3.42	7.49	58.62	33.89
Y10	7.38	2.35	3.92	13.65	42.53	43.82
Y11	7.18	3.25	4.04	15.58	35.51	48.91
Y12	7.11	2.96	3.45	10.08	41.75	48.17
Y13	7.03	3.75	5.35	13.35	42.53	44.12
Y14	6.96	3.98	6.62	20.53	33.56	45.91
Y15	6.46	4.57	6.35	17.58	41.58	40.84
Ave	7.55	1.88	3.41	13.26	41.23	45.50
Min	6.46	0.15	1.15	7.49	28.89	33.89
Max	8.33	4.57	6.62	20.53	58.62	60.77

Ave: average content

Min: the minimum value

Max: the maximum value

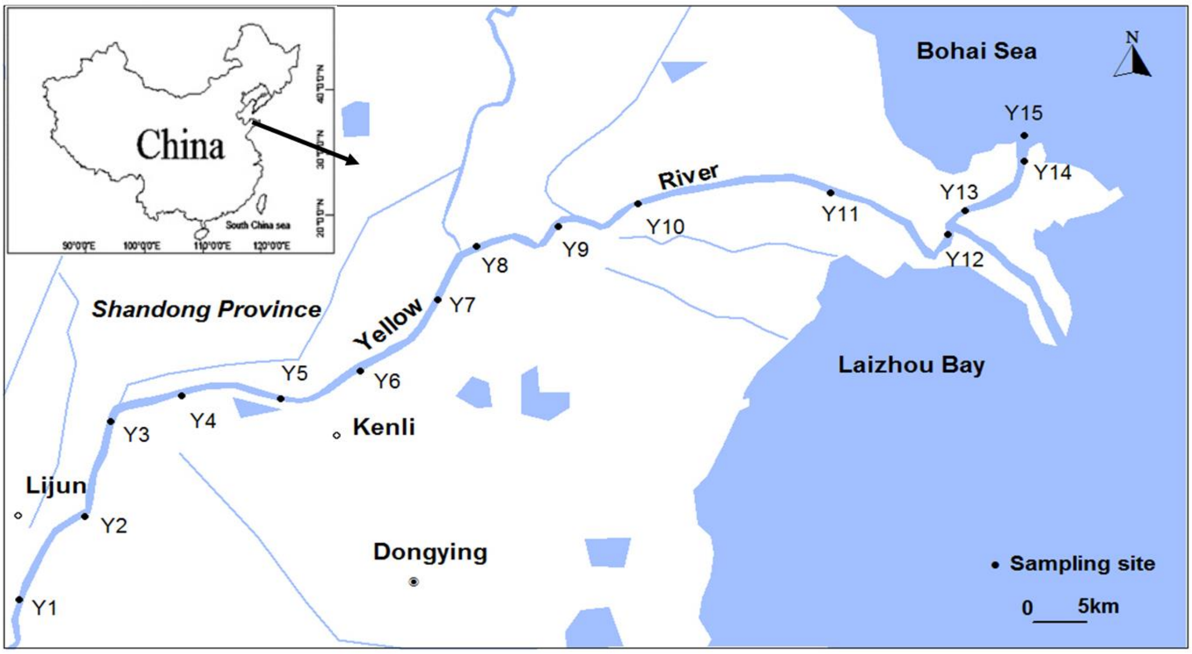
Figure Legends:

Fig. 1 Schematic graph of the sampling sites in the Yellow River Estuary.

Fig. 2 Concentrations of PFOS in the water and sediment of the Yellow River Estuary.

Fig. 3 Average concentrations of PFOS at different sampling depth in sediment column from the site Y14.

Fig. 4 Correlations between distribution coefficient (K_d) of PFOS and salinity (S‰), TOC, clay (a) K_d vs. salinity (S‰), (b) K_d vs. TOC, (c) K_d vs. clay.



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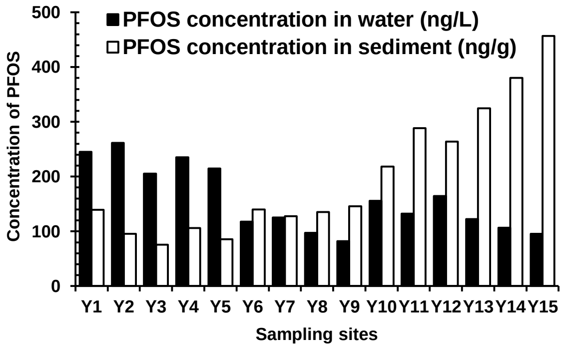

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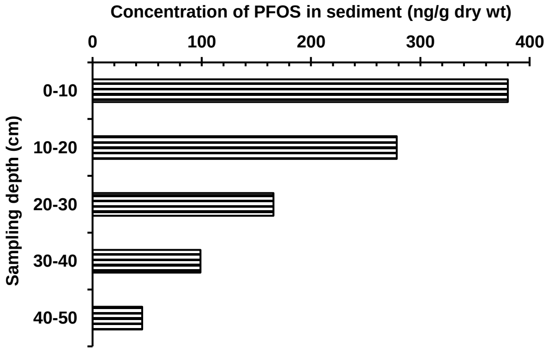

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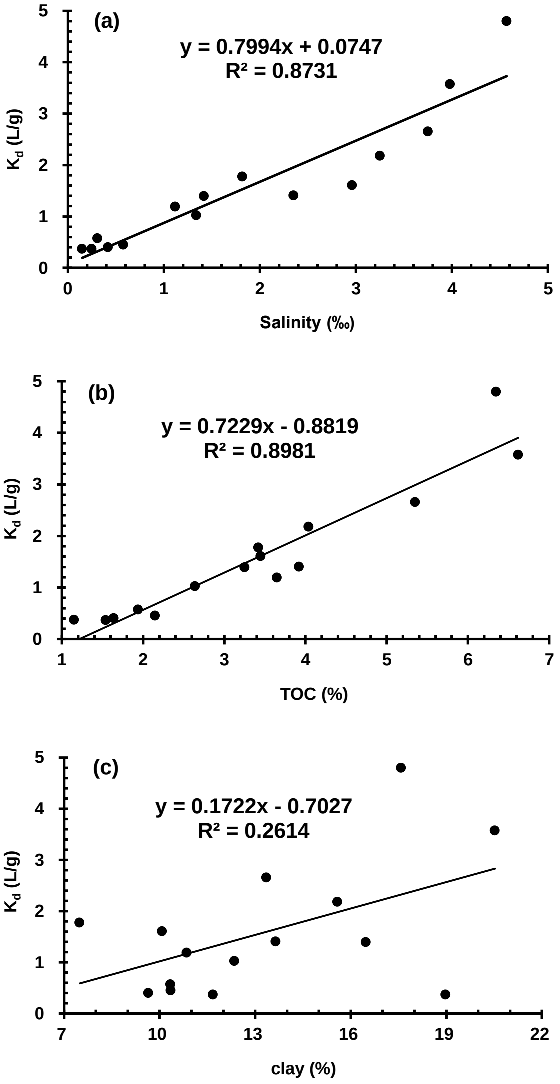

Fig. 4 Correlations between distribution coefficient (K_d) of PFOS and salinity (S‰), TOC, clay (a) K_d vs. salinity (S‰), (b) K_d vs. TOC, (c) K_d vs. clay.