



Adsorption of perfluorooctane sulfonate on soils: Effects of soil characteristics and phosphate competition



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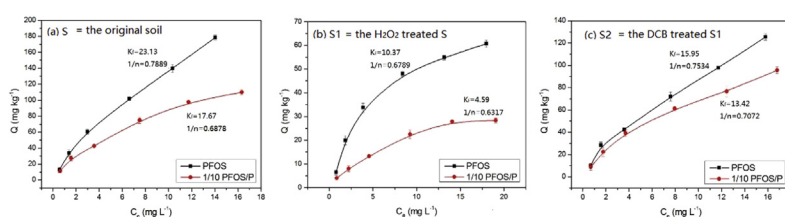
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HIGHLIGHTS

- We examined effects of soil characteristics and phosphate on soil PFOS adsorption.
- SOM in soils promoted sorption of PFOS; ferric oxides had the opposite effect.
- Batch sorption data gave nonlinear PFOS adsorption isotherms for each tested soil.
- In ferric oxide-rich soil, PO_4 is a stronger adsorption site competitor than PFOS.
- In soil with a high SOM content, PO_4 is a weaker competitor than PFOS.

GRAPHICAL ABSTRACT



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ABSTRACT

Perfluorooctane sulfonate (PFOS) is an emerging contaminant, whose presence has been detected in different compartments of the environment in many countries. In this study, the effects of soil characteristics and phosphate competition on the adsorption of PFOS on soils were investigated. Results from batch sorption experiments showed that all the adsorption isotherms of PFOS on three tested soils were nonlinear. In experiments without the addition of phosphate (P) to the soil solution, the Freundlich sorption affinity (K_f) of PFOS on S (original soil), S1 (soil from which soil organic matter (SOM) had been removed), and S2 (soil from which both SOM and ferric oxides had been removed) were 23.13, 10.37 and 15.95, respectively. The results suggested that a high amount of SOM in soil can increase the sorption affinity of PFOS on soils and that a greater amount of ferric oxides can reduce it. The addition of P in the soil solution reduced the K_f of PFOS on S, S1, and S2 by approximately 25%, 50%, and 15%, respectively. For the binary system of PFOS and P, soil with higher ferric oxide content showed greater K_f reduction after P addition; whereas soil with higher SOM content showed less K_f reduction. Our results suggest that for soils dominated by ferric oxides, P is a more effective competitor than PFOS for the adsorption sites in the binary system; whereas in soils containing more SOM, P is a weak competitor.

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1. Introduction

Recently, considerable attention has been focused on the environmental pollution caused by perfluorooctane sulfonate (PFOS), due to its high bioaccumulation, wide distribution, and persistence in natural environments (Beach et al., 2006). PFOS has been widely used in industry, e.g., in the manufacture of paper, textiles, and leather, and in semi-conductors and fire-fighting foam (Tsai et al., 2002; Zareitalabad et al., 2013). The PFOS produced by factories has mostly been discharged into the environment (Prevedouros et al., 2006; Lau et al., 2009; Lindstrom et al., 2011). PFOS is less likely to evaporate into the surrounding atmosphere because of its low vapor pressure (Kaiser et al., 2005). Moreover, PFOS mostly exists as an ion (Hu et al., 2011). Therefore, PFOS commonly persists in water, sediment, and soil. Since it has both hydrophilic and hydrophobic properties, PFOS shows a higher solubility in water (570 mg L^{-1}) than traditional hydrophobic pollutants (Rayne and Forest, 2009; Liou et al., 2010). Accordingly, the transport of PFOS in soil and aquatic systems is considered an important process in controlling its environmental distribution and fate.

Sorption is an important process for the transport, distribution, and fate of chemicals in the soil–aquatic environment (You et al., 2010), and recently several studies on the sorption behavior of PFOS in soils have been reported (Higgins and Luthy, 2006; Johnson et al., 2007; You et al., 2010; Ahrens et al., 2011). Soil comprises a complicated matrix of organic and inorganic components that play different roles in the adsorption of PFOS (Sullivan and Mabury, 2001). Higgins and Luthy (2006) have studied the sorption of PFOS on soils of varying total organic carbon (TOC) and iron oxide contents, and have demonstrated that the TOC of soils is the dominant parameter associated with adsorption, whereas clay fractions make a relatively minor contribution to this process. Johnson et al. (2007) have reported the sorption of PFOS on sand, clay, and iron oxide (in which organic carbon is not present), and have indicated that surface electrostatic interaction is the main sorption mechanism for PFOS. However, previous studies have also found that in soils of Germany and Shanghai the adsorption capacity of PFOS is not correlated with soil TOC (Becker et al., 2008; Li et al., 2010). Despite the findings of these previous studies, the retention properties of PFOS in soils of different compositions have yet to be well established, which may limit our understanding of PFOS behavior in soils.

The effect of phosphate (P) on PFOS adsorption at the water–soil interface is also unclear, and thus represents a factor warranting further investigation. P is a well-known pollutant and a primary factor limiting eutrophication of surface waters. P can convey negative charges to the soil particle surfaces, thereby changing the adsorption capacity of pollutants in soils (Wang et al., 2010). Wang and Xing (2002) have reported that the adsorption of P on iron oxides may enhance cation adsorption. They found that the adsorption of Cd on goethite apparently increased after P addition, and attributed this response to an increase in the surface negative charges of goethite and a decrease in electrostatic potential near the solid surface. Moreover, several researchers have demonstrated that P can compete with organic matter such as glyphosate in soils (Gimsing et al., 2004; Gimsing and Borggaard, 2007). PFOS and P are likely to co-exist in soils, and it can be conjectured that the sorption behavior of PFOS in soils is strongly modified by P. Nevertheless, to the best of our knowledge, none of the previous studies have investigated the effect of P competition on PFOS adsorption in soils. Accordingly, to gain a better understanding of the fate of PFOS in soils, the present study was designed to investigate the adsorption of PFOS in soils of different characteristics, and to examine the effect of P addition on PFOS adsorption.

2. Materials and methods

2.1. Sorbents and chemicals

PFOS (>98% purity), KH_2PO_4 (>98% purity), and ammonium acetate (>99% purity) were purchased from Sigma-Aldrich Co. (St. Louis, MO). Surface soils (0–20 cm) were sampled from the Taihu area, Jiangsu Province, China. Soil samples were air-dried and passed through a 0.2-mm-pore-size sieve. To characterize the effect of soil organic matter (SOM) on the sorption of PFOS, the H_2O_2 treatment method (Hyeong and Capuano, 2000) was used to remove SOM at room temperature (298 K). The fractions remaining in the residual soil after SOM removal were mainly inorganic components such as silicate and metal oxides (Mikutta et al., 2005). Having removed SOM, the dithionite–citrate–bicarbonate (DCB) treatment method was used to remove free ferric (hydro) oxides from the residual soil (Bigham et al., 1978; McCarty et al., 1998). However, it should be noted here that although we used relatively mild treatments to remove SOM and free ferric oxides, soil mineral surfaces may still have been altered to a certain degree (Tong et al., 2015).

The pH values of all soil samples were measured with an electrode using a soil:solution ratio of 1:2.5. SOM was measured by dichromate oxidation and DCB Fe was determined using the DCB digestion method (Sparks, 1996). The pipette method was used to obtain particle size distribution (Soil Conservation Service, 1972).

2.2. Adsorption experiments

All the adsorption experiments were conducted using the batch method (You et al., 2010) at room temperature (298 K) in 50-mL polypropylene (pp) centrifuge tubes. All the experiments were carried out at the natural pH (6.5) of the soil. We performed each experiment three times in order to obtain reproducible results. We used a solid (soil) concentration of 40 g L^{-1} . To obtain accurate results without degradation or retention in vessels during sample preparation and shaking, control samples containing PFOS solution without soil were used.

2.2.1. PFOS adsorption experiments

PFOS has been used in certain specialized industries, including the semiconductor, metal plating, and textile industries (Fujii et al., 2007; Paul et al., 2009). The wastewaters discharged by these industries contain high PFOS concentrations. For example, Lin et al. (2009) reported that approximately 200 ton of wastewater containing 12.566 mg/L of PFOS was generated daily from a semiconductor plant in Chinese Taiwan. On the basis of this figure, the initial PFOS concentration adopted in the present study was $1\text{--}20 \text{ mg L}^{-1}$.

For the experiments, 1 g of each of three soil samples [S (original soil), S1 (after removing SOM from S), and S2 (after removing ferric oxides from S1)] was initially added to 20 mL of distilled water and left standing for 24 h to ensure soil hydration. The PFOS solution was then added to the pp centrifuge tubes to give an initial concentration ranging from 1 mg L^{-1} to 20 mg L^{-1} , and the volumes of all the mix solutions were adjusted to 25 mL. The pp centrifuge tubes were shaken in a 2D-shaker at 250 rpm for 24 h, as this has been reported to be the sorption equilibrium time of PFOS (Pan et al., 2009). After this equilibration step, the mixtures were centrifuged at 9000 rpm for 30 min. The resulting supernatants were then collected for analysis. The concentration of PFOS in the supernatant was measured using a Waters Acquity ultra-performance liquid chromatography–tandem mass spectrometry system (UPLC/MS/MS) equipped with a $50 \times 2.1 \text{ mm}$ Waters BEH C18 column ($1.7 \mu\text{m}$ particle size) and tandem quadrupole mass spectrometers (Milford,

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