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Occurrence and fate of perfluoroalkyl substances in marine sediments from the Chinese Bohai Sea, Yellow Sea, and East China Sea

Yan Gao [a], Jianjie Fu [a], Lixi Zeng [b], An Li [c], Huijuan Li [a], Nali Zhu [a], Runzeng Liu [a], Aifeng Liu [a], Yawei Wang [a, *], Guibin Jiang [a]

[a] State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100085, China
[b] School of Chemistry and Chemical Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
[c] School of Public Health, University of Illinois at Chicago, Chicago, USA

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ABSTRACT

In this study, 166 surface sediments and 3 sediment cores from the Bohai Sea (BS), Yellow Sea (YS) and East China Sea (ECS) in China were collected to investigate the spatial and temporal distributions and the transport of PFASs. PFASs concentrations in the surface sediments ranged from below detection limit (<LOD) to 2.78 ng g^{-1} with an average value of 0.55 ng g^{-1} on a dry weight basis (dw). A general decreasing trend of PFASs from the coast areas to the open sea was found. Multivariate regression analysis indicated pH and longitude were the major factors influencing surficial distributions of PFASs in the sampling areas ($R^2 = 0.29$, $p < 0.01$). Total PFASs concentrations in the sediment cores ranged from <LOD to 1.65 ng g^{-1} dw, with an increasing trend from the lower to the upper layers, corresponding well to the increasing production and usage in China in recent years.

1. Introduction

Per- and polyfluoroalkyl Substances (PFASs), such as perfluorocarboxylates (PFCAs) and perfluorosulfonates (PFSAs), have been widely used in the past six decades as additives in surface coatings of carpets, leather, papers, and textiles, as well as in fire-fighting foams, pesticides, paints, etc. Their excellent thermal and chemical stability renders PFASs highly persistent in the environment, evidenced by their ubiquitous global detection (Cai et al., 2012b) and accumulations in human and wildlife (Kelly et al., 2009; Li et al., 2013). In 2009, perfluorooctanesulfonate (PFOS) and its salts were listed in the Stockholm Convention as Persistent Organic Pollutants (POPs) (Loi et al., 2011).

Production and usage of PFASs in China has increased rapidly in recent years. A large proportion of the total production output in China was from coastal provinces/cities, including Zhejiang, Fujian, Guangdong, Jiangsu, Liaoning and Shanghai (Chen et al., 2009), adjacent to the Bohai Sea (BS), Yellow Sea (YS), East China Sea (ECS), and South China Sea (SCS). Recently, PFASs have been detected in the coastal marine environment of China. Chen et al. investigated PFASs in the water and sediment from coastal northern BS and found sediment was an important sink for perfluoroundecanoic acid (Chen et al., 2011). Pan et al. found low PFASs concentrations in mollusks from the coastal waters in the BS (Pan et al., 2010). The investigation on the PFASs in coastal waters of Hong Kong, South China indicated that PFASs in the sampling areas were strongly influenced by discharge from the Pearl River (So et al., 2004). Cai et al. considered the Yangtze River as an important source of PFASs found in the coastal waters of southern China (Cai et al., 2012a).

In spite of the ubiquitous existence of PFASs in the global environment, the mechanisms of long range transport of the PFASs and their precursors across continents and on global scales remain unclear. Relatively volatile perfluoroalkyl precursors such as fluorotelomer alcohols (FTOHs), perfluoroalkyl sulfonamides (FASAs) and perfluoroalkylsulfonamidoethanols (FASEs) were transported via the atmosphere and subject to photolytic degradation in the air (Ellis et al., 2004, 2003), whereas the ionisable PFASs such as PFCAs and PFSAs are predominantly transported via the ocean current (Prevedouros et al., 2006).

This work is a systematic survey of PFASs in the sediments of the BS, YS and ECS of China. The primary objective of this work was to depict the spatial and temporal distribution patterns of PFASs in the three sea regions of China. The large number of samples allowed us to examine the factors influencing the accumulation of PFASs in

* Corresponding author.
E-mail address: ywwang@rcees.ac.cn (Y. Wang).

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marine sediments. The large geographical coverage enabled us to gain insights into the anthropogenic impacts, the transport pathways, and the environmental fate of PFASs in the sampling areas.

2. Materials and methods

2.1. Sample collection

The entire study area encloses the BS, YS and ECS, extending 2000 km from south to north and totaling about 1,230,000 km^2 water surface (Fig. 1). The major mud areas and the dominant ocean currents are shown in Fig. S1. The BS is a semi-enclosed shallow (mean depth 26 m) marginal sea receiving waters from more than 40 rivers in northern China. The YS is located between Chinese mainland to the west and Korean Peninsula to the east, with an average depth of 44 m. ECS is mostly on a continental shelf with water depth <200 m, and its water flow is strongly influenced by the freshwater input from the Yangtze River and the saline Kuroshio Current (Yanao and Matsuno, 2013). The YS and ECS constitute the western margin of the North Pacific Ocean.

Sediment sampling was carried out onboard of *R/V Dong Fang Hong 2* in 2011 and 2012. The sampling locations are shown in Fig. 1, and additional information is summarized in Table S1. In all, 166 surface sediments were collected using a stainless steel box sampler and transferred to polyethylene bags. Three sediment cores were collected. Core-1 and Core-2 were collected in the ECS in 2011, and Core-3 was in the YS in 2012, all from the mud areas of the shelf. The sediment cores were sectioned onboard into 1 cm intervals using a stainless steel cutter. A total of 111 core slices were obtained. All samples were kept in a shipboard refrigerator immediately after sampling, and subsequently freeze-dried and ground after being transported to the laboratory. All the samples were stored frozen at −20 °C until analysis.

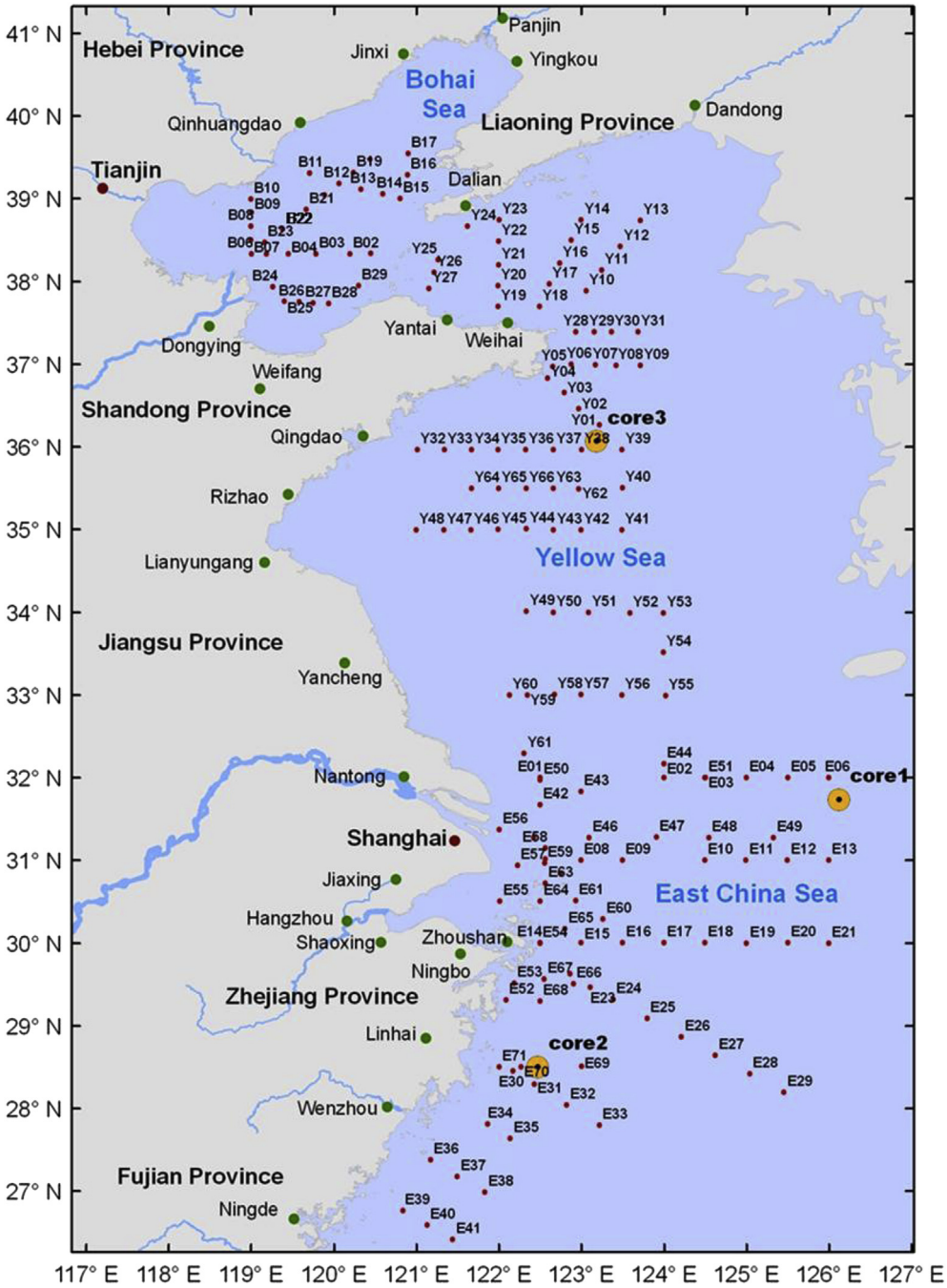

Fig. 1. Study area and sampling sites of surface sediments (small red dots) and sediment cores (large orange dots). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)