



Influence of human activities and organic matters on occurrence of polybrominated diphenyl ethers in marine sediment core: A case study in the Southern Yellow Sea, China



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HIGHLIGHTS

- \sum_7 PBDEs and BDE-209 showed sharp increase since the 1990s.
- Increasing human activities were responsible for these increases.
- Core analysis indicated debromination of highly brominated congeners.
- TOC-dependent deposition of PBDEs.
- Marine organic matter drove PBDE deposition in sediment.

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ABSTRACT

The Southern Yellow Sea (SYS) is an important reservoir of anthropogenic organic contaminants, such as polybrominated diphenyl ethers (PBDEs). To reconstruct the historical records of PBDEs and examine their relationships with the human activities and organic matters, a ^{210}Pb -dated sediment core was collected from the central mud area in the SYS. The concentrations of tri- to hepta-BDEs (\sum_7 PBDEs) and BDE-209 ranged from 9.8 to 99.8 pg g^{-1} d.w. and from 12.1 to 855.4 pg g^{-1} d.w., respectively, both displaying the increasing trends from the bottom to the surface. More importantly, there was a faster increase for PBDEs since the 1990s, especially for BDE-209, which responded well with the rapid economic growth, and the increases of urbanization and industrialization in the local areas of the SYS. The analogously vertical patterns and significant relationships between PBDEs and total organic carbon (TOC) implied the TOC-dependent deposition of PBDEs in the core. Furthermore, multiple biomarker-based proxies of terrestrial organic matter (TOM) and marine organic matter (MOM) were introduced to systematically investigate the different effects of TOM and MOM on PBDE deposition in the SYS. The similarly down-core profiles and significant correlations were found between PBDEs and the MOM proxies (sum of rassicasterol, dinosterol and C_{37} alkenones ($\sum A + B + D$)) and marine TOC as well as the branched and isoprenoid tetraether (BIT), but not for TOM proxies ($\sum \text{C}_{27} + \text{C}_{29} + \text{C}_{31}$ *n*-alkanes, terrestrial and marine biomarker ratio (TMBR) and terrestrial TOC), indicating that MOM was an important factor driving PBDE deposition in the sediment core from the SYS.

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1. Introduction

As a typical group of brominated flame retardants, polybrominated diphenyl ethers (PBDEs) have been extensively used in the electrical and electronic equipments, textiles, plastics, building materials and furnishings for more than five decades (Besis and

Samara, 2012; Ma et al., 2012). Generally, PBDEs are commercially produced and used in three technical formulations: Penta- (over 70% of BDE-47 and BDE-99), Octa- (over 40% of BDE-183) and Deca- (over 98% of BDE-209) BDEs (Alaee et al., 2003). The global production of commercial PBDEs was estimated to be 40,000 tons in 1992, while it reached 67,000 tons in 2001 (Li et al., 2016a). In China, the estimated domestic production of Deca-BDE, the predominantly commercial PBDE mixture, had increased from 10,000 tons in 2000–30,000 tons in 2005 (Chen et al., 2007). In addition,

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China have long been suspected of receiving and disposing the electronic wastes (e-wastes) imported legally and illegally from the developed countries, and approximate 1.5–3.3 million tons are annually imported to China (Zhou and Xu, 2012). PBDEs are dissolved in the polymers without chemical bonding, and can thus be easily released to the environment during the production, usage and disposal of these PBDE-containing products. High levels of PBDEs had been widely documented in air (Guo et al., 2015; Wang et al., 2014), water (Qiu et al., 2010a; Xiong et al., 2015), soils (Wu et al., 2015), sediments (Lv et al., 2015; Wang et al., 2016), organisms (Nie et al., 2015; Sun et al., 2015) and even human tissues (Chen et al., 2014). Environmental and human health concerns led to the banning on production and usage of Penta- and Octa-BDEs in 2004. Shortly afterwards, some factories in the USA and Europe voluntarily phased out Deca-BDE manufacturing in 2009 due to the high levels of BDE-209 in various environmental matrixes. However, Deca-BDE is still produced and used in the Asian countries, such as China.

PBDEs can be transported from the terrestrial emission sources to marine environments via riverine inputs, surface runoff of contaminated soils, and atmospheric deposition (Wang et al., 2015a, 2016). Due to the high hydrophobicity, PBDEs tend to strongly partition to particle matters, and finally settle down from water column and bury in the marine sediments, which are regarded as one of the long-term reservoirs for PBDEs (Li et al., 2012; Zhao et al., 2011). Therefore, the undisturbed and well-dated sediment core can be a good proxy to reconstruct the historical records of PBDEs inputs into marine environments, elucidate the influence of human activities (e.g. economic growth, urbanization, industrialization) on PBDE pollution, and evaluate the effectiveness of regulation and management measures on PBDE emission. As we known, organic matters (OMs) are an important constituent of the marine sediments, mainly consisting of terrestrial organic matter (TOM) from terrestrial inputs and marine organic matter (MOM) from marine primary production (Xing et al., 2011a, 2011b). Many studies have documented that sedimentation TOM played an important role on the accumulation and distribution of organic pollutants in marine environments, especially for the distal mud areas (Fan et al., 2014; Zhao et al., 2013). However, there is very limited information regarding the discrepancies in effects of TOM and MOM on the accumulation of organic pollutants in the marine sediments. Thus, the studies on insight into the comprehensive roles of TOM and MOM on the sedimentation of organic pollutants in the marine sediments are urgently needed.

Southern Yellow Sea (SYS), located between the Chinese mainland and the Korean Peninsula, is a shallow shelf sea of the northwest Pacific, with a mean water depth of 44 m and a maximum depth of 140 m (Xing et al., 2011a). With the rapid economic development, high urbanization and industrialization in the coastal areas (mainly Shandong and Jiangsu provinces) in recent decades, large quantities of anthropogenic pollutants are discharged into the SYS. Besides, the biggest manufacturing base of brominated flame retardants is located in Weifang of Shandong province in China (Pan et al., 2010). Inevitably, a mass of PBDEs are believed to be inputted into the SYS. For the SYS, most of the sediments are directly and indirectly derived from the Yellow River and Yangtze River through long-range sediment dispersal processes (Hu et al., 2013; Lim et al., 2007). Furthermore, the SYS is in the pathway of Asian continental outflow to the northwest Pacific driven by the East Asian monsoon (Zeng et al., 2013), and thus receive large amounts of atmospheric particles. On the whole, the SYS is an important receptor of land-based OMs and associated organic pollutants via riverine input, surface runoff and atmospheric deposition (Duan et al., 2013; Hu et al., 2013). However, to date, few data on historical deposition of PBDEs and relevant

influencing factors are available in the SYS. Consequently, the aims of the present study were to (1) investigate the chronology of PBDE inputs into the SYS as recorded in the dated sediment core; (2) reveal the impacts of human activities (e.g. economic growth, urbanization and industrialization) on the PBDE pollution; (3) comprehensively explore the different effects of TOM and MOM on PBDE deposition by using multiple biomarker-based proxies in the sediment core.

2. Materials and methods

2.1. Sample collection

To reconstruct the pollution histories of PBDEs in the SYS, one sediment core (Core N02, 122°38'24"E and 36°3'N) was collected from the central mud area by using a gravity corer (Qiangdao Orson, China) on board of the R/V "Dong Fang Hong 2" in 2011 (Fig. 1), with the length of 20 cm. The core was immediately sliced into 1 cm intervals using a stainless steel cutter on board. All samples were wrapped in aluminum foil and stored at –20 °C until analysis. According to the previous study, the sediment core was dominated by clayey silt, and the vertical profiles of grain composition showed a slight variation, indicating a stable depositional environment (Wang, 2014). In addition, the core was dated by using ²¹⁰Pb dating technique, and the average sedimentary rate was 0.19 cm yr⁻¹ (Wang, 2014). Thus, the sediment core N02 of 20 cm covered about 100 years of sedimentation records.

2.2. Chemicals and reagents

A standard mixture of BDE-28, 47, 99, 100, 153, 154, 183 and 209 were selected for quantitative analysis. PCB-209 and 2,4,5,6-tetrachloro-m-xylene (TCmX) were added as surrogate standards, and BDE-77 was used as internal standard for the quantification of the targets. All of the standards were purchased from AccuStandard, Inc (USA). The neutral silica gel (80–100 mesh) was heated at 180 °C for 3 h, and sodium sulfate was baked at 450 °C for 4 h, which were all stored in sealed containers after cooling. All solvents used in the sample analysis, including n-hexane, methanol and isoctane, were of UPLC grade, which were also purchased from AccuStandard, Inc (USA).

2.3. Sample analysis

The sample extraction, cleanup and instrumental analysis had been reported in our previous studies (Wang et al., 2015b, 2016). Briefly, about 10 g of freeze-dried sediment of each sample was processed in an ultrasonic bath with 2.0 M NaOH-methanol solution for 30 min to remove some impurities, such as organochlorine pesticides. Then, distilled water and n-hexane, in turn, were added, and the organic phase of extracts was separated from the mixture. The organic phase was concentrated in a rotary evaporator, and purified in a multilayer silica gel column packed with neutral silica gel (5% water deactivated), sulfuric acid silica (44% sulfuric acid, w/w) and anhydrous sodium sulfate from the bottom to top. The PBDE fraction was eluted by n-hexane, and then concentrated, dried up under a gentle stream of nitrogen and finally redissolved in isoctane with a known amount of BDE-77. Identification and quantification of PBDEs were performed on a Shimadzu 2010plus gas chromatograph coupled with an electron capture detector. A DB-5 capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness) and a shorter one (15 m × 0.25 mm i.d. × 0.10 μm film thickness) were used for the separation of BDE-209 and other low brominated congeners, respectively.

The multiple biomarkers of TOM and MOM had been previously

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