



Insights into spatially and temporally co-occurring polybrominated diphenyl ethers in sediments of the East China Sea



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HIGHLIGHTS

- PBDEs were ubiquitous in sediments from 24 sampling stations in the East China Sea.
- Potential sources of PBDE congeners were traced using co-occurrence analysis.
- Network was used in the co-occurrence analysis of PBDE congeners for the first time.
- Network visualized the correlations of co-occurring congeners and sampling sites.

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ABSTRACT

In this study, twenty-four core sediment samples were collected from monitoring stations in the East China Sea (ECS) to investigate the spatial and temporal distributions of polybrominated diphenyl ethers (PBDEs) and their potential sources. In surface sediment samples (the top 0–3 cm segment), the concentrations of Σ_8 PBDEs (sum of BDE15, 28, 47, 99, 100, 153, 154 and 183) and BDE209 were between 0.03 and 1.26 ng g⁻¹ d.w., and 0.20 and 3.15 ng g⁻¹ d.w., respectively. The PBDEs concentrations were decreased generally from the shore towards the sea, indicating that PBDEs are mainly derived from anthropogenic sources. As the predominant congener, BDE209 was primarily distributed in the nearshore regions, suggesting that it might be derived from runoff and sewage. In contrast, higher levels of BDE99 and BDE47 were detected in the offshore samples, and the concentrations of BDE47 were higher than those of BDE99. All of their concentrations were poorly correlated with TOC, suggesting BDE47 and BDE99 might be transferred mainly along with the atmosphere. In most sediment cores, the Σ_8 PBDEs and BDE209 concentrations increased from the bottom (18–30 cm), reached their maximum concentrations in the middle (6–15 cm) or upper (0–6 cm) sections, and leveled off near the surface (0–3 cm). This trend is consistent with the phasing-out of PBDEs and the strict management of recycling e-waste. This is the first study for network used to analyze the distribution properties of pollutant congeners in environment, which facilitates the visualization of cluster results among multivariable data and helps in tracing the potential sources of pollutants.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been produced and applied extensively as additive brominated flame retardants (BFRs) in various consumer products, such as plastics, textiles and electronic equipment, in recent decades. Because they are not chemically bound to the products, PBDEs are easily released into the environment during application and disposal processes.

In addition, PBDEs have been detected ubiquitously in various abiotic (sediment, soil, water and atmosphere) and biotic (human hair, milk and blood) mediums due to their persistence, bioaccumulation and long-distance transport (de Wit, 2002; Darnerud, 2003). Thus, PBDEs have received extensive attention from researchers around the world (Sjödin et al., 2003; Wang et al., 2007). It should be noticed that PBDEs released in the environment could be bioaccumulated in the biota and magnified through the food chain, thus threaten human health (Schuhmacher et al., 2013). Specifically, PBDEs may cause neural development deficits, disrupt thyroid hormones and are potential carcinogens

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for humans (Darnerud et al., 2001), and some low brominated PBDEs may even result in subtle developmental, immunological and endocrinological effects on children (Herbstman et al., 2010; Gascon et al., 2011; Hertz-Picciotto et al., 2011). Therefore, the distribution levels and compositions of PBDEs in environments could be carefully inspected to estimate their environmental and ecological security.

With the rapid development of electrical and electronic industries in recent decades, China has become a major manufacturer and consumer of BFRs (Mai et al., 2005). The consumption of deca-BDE technical mixture, which is the predominant BFRs additive used in China, amounted to 30000 tons in 2005 (Zou et al., 2007). In addition, the import and disposal of electronic waste (e-waste) has also increased the widespread occurrence of PBDEs in China. Furthermore, crude recycling and dismantling techniques have aggravated the potential release of PBDEs and their pollution in environments (Chen et al., 2009). Considering the geographic position of the coastal East China Sea (ECS), which receives pollutants from the Yangtze River and is adjacent to regions with highly development urban areas with intensive industrial activities, the levels and distribution properties of PBDEs in this district should draw more attention.

In general, sediment is considered as a major sink for organic pollutants and plays an important role in the distributions and transformations of these contaminants. The various contaminants imported via runoff from land, discharge from industrial and wastewater treatment plants and deposition from the atmosphere tend to be trapped into sediments, accumulate in organisms and transfer along the food chain, eventually pose a threat to human health. Additionally, these trapped pollutants could interact with compounds in sediments and be degraded by the resident microbes (Gerecke et al., 2005), resulting in more complex contamination. Because lower brominated PBDE congeners are more toxic than deca-BDE (Darnerud, 2003), investigations of the composite and concentration levels of PBDEs in sediments might be helpful for illustrating the biotransformation of PBDEs and the risk assessment of the polluted sediment.

Considering the position specialty and environment quality of the ECS, PBDEs in the marine core sediments were quantified to determine their spatial and temporal distribution trends of the PBDE congeners. Additionally, network analysis was applied to gain insights regarding the co-occurrences and associations of PBDE congeners in the horizontal and vertical directions to facilitate the exploration of co-occurring complex data patterns (Liu et al., 2014). This is the first study to apply network for co-occurring correlations analysis among organic pollutants and co-occurring congeners. The co-occurrence results can be used to distinguish the pollution trends of PBDEs and the correlations of PBDE congeners in ECS region, and to propose their potential transformation and migration processes. Therefore, this study verified the feasibility of using network in the field of environmental chemistry to analyze spatial and temporal monitoring data and to further visualize the correlations among co-occurring items from complex systems.

2. Materials and methods

2.1. Sample collection

Firstly, 24 sediment cores (from depths of approximately 30 cm below the seawater) were collected at monitoring stations in the East China Sea (26°23'N–30°00'N, 120°47'E–124°00'E) during October and November, 2013. The sampling locations are summarized in Table S1 and are labeled in Fig. 1. All of the core sediment samples were collected using a box-core sampler and sliced at 3 cm intervals along the length of the onboard. The top 0–3 cm

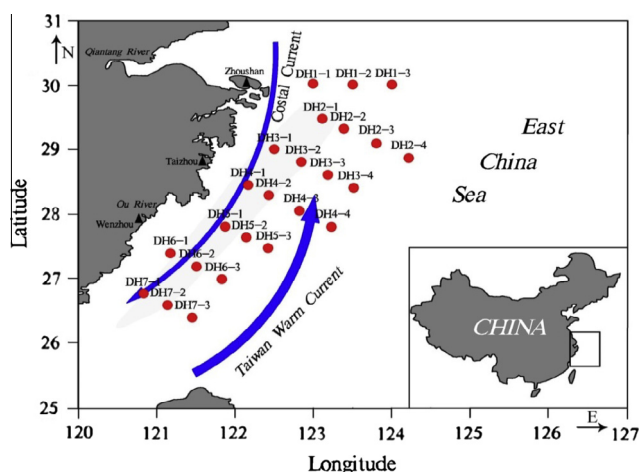


Fig. 1. The location map of 24 sampling stations (marked with red node) in the East China Sea. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

layer was regarded as the surface sediment sample at each site. All of the samples were wrapped in aluminum foil and transported on ice to the laboratory, and stored at $-20\text{ }^{\circ}\text{C}$ until analysis.

2.2. Standard materials

A standard mixture (BDE15, 28, 47, 99, 100, 153, 154, 183 and 209) was used for quantification that was purchased from Accu-Standard (New Haven, CT, USA). The ^{13}C -PCB208, ^{13}C -PCB141 and ^{13}C -BDE209 were obtained from Wellington Laboratories Terra Chem Inc (Shawnee Mission, KS). The ^{13}C -PCB208 was used as an internal standard, and ^{13}C -PCB141 and ^{13}C -BDE209 were added as surrogates. All of the chemicals used during the experimental and instrumental processes, including the acetone, n-hexane, dichloromethane and methylbenzene, were analytical grade and purchased from Shanghai Chemical Company (Shanghai, China). Neutral silica gel (100–200 mesh) and alumina (80–100 mesh) were extracted using a mixture of dichloromethane and acetone (1:1, v:v) for 72 h before being heated to $180\text{ }^{\circ}\text{C}$ and $120\text{ }^{\circ}\text{C}$ for 12 h, respectively. Sodium sulfate was boiled at $650\text{ }^{\circ}\text{C}$ for 4 h prior to use.

2.3. Analysis procedure

Freeze-dried samples (approximately 10 g) were spiked with 24 ng of the surrogate standard and extracted for 8 h using 100 mL of an acetone/hexane (1:1, v:v) mixture in a Soxhlet apparatus (Buchi, Switzerland). During the extraction process, activated copper was added to the sediment samples to remove elemental sulfur. The extract was concentrated using a rotary evaporator and purified using a silica gel column before eluting with methylene chloride/hexane (1:1, v:v). Finally, each extract was concentrated to 1 mL using a nitrogen blowing instrument.

The concentrations of the PBDE congeners were determined using an Agilent 7890A gas chromatograph coupled with a 5975C mass spectrometer with negative chemical ionization (GC-NCI-MS) and in the selected ion monitoring (SIM) mode. Helium and methane were used as the carrier and reagent gases, respectively, at a flow rate of 1.5 mL min^{-1} . A DB-5MS capillary column (30 m \times 0.25 mm i.d. \times 0.25 μm film thickness) was used to identify the PBDE congeners, except for BDE209. Furthermore, BDE209 was detected using a DB-5HT (15 \times 0.25 mm i.d., 0.1 μm film thickness) capillary column. For all of the PBDE congeners,

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