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Levels and distribution of methoxylated and hydroxylated polybrominated diphenyl ethers in plant and soil samples surrounding a seafood processing factory and a seafood market

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) along with hydroxylated polybrominated diphenyl ethers (OH-PBDEs) and methoxylated polybrominated diphenyl ethers (MeO-PBDEs) were found in plant and soil samples collected surrounding a seafood processing factory and a seafood market in China. The profiles of MeO-PBDE congeners were different between seafood processing factory and seafood market. The detection frequency and concentration of 6-OH-BDE-47 were lower than that of MeO-PBDEs. Near seafood processing factory, a decreasing trend of analyte concentrations in plants was found downstream the river where factory wastewater was discharged. Concentrations of ΣMeO-PBDEs in plant and soil samples showed difference as root > soil > leaf. However, at seafood market, the concentrations of ΣMeO-PBDEs were much higher in leaves than those in soil. The concentration of ΣMeO-PBDEs in leaves showed a remarkable difference between *Calystegia soldanella* (Linn.) R. Br. and *Setaira viridis* (L.) Beauv.
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1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been used as flame retardants in many manufactured items and have received great attention due to their ubiquitous environmental distribution and bioaccumulation potential (Hites, 2004). Recently, focus has shifted to structural analogues of PBDEs, such as hydroxylated (OH) and methoxylated (MeO) PBDEs. OH-PBDEs are structurally similar to the thyroid hormone thyroxin (T4). The toxicity of OH-PBDEs has been investigated and was considered more potent than that of PBDEs. Effects of OH-PBDEs on organisms include oxidative phosphorylation disruption, neurotoxicity and thyroid disruptions (Canton et al., 2008; Harju et al., 2007; Meerts et al., 2001; Mercado-Feliciano and Bigsby, 2008). Relative to OH-PBDEs, MeO-PBDEs were reported to have a greater effect on mRNA abundance of steroidogenic enzymes in the H295R cell line (He et al., 2008).

The marine environment is considered as the richest source of biogenic organohalogens (Gribble, 2003). Up to now, OH-PBDEs and MeO-PBDEs were mainly detected in marine organisms such as algae, mussels and fish (Malmvarn et al., 2005; Teuten et al., 2005; Unson et al., 1994). There has been a considerable interest in determining the sources and relationships among PBDEs, OH-PBDEs and MeO-PBDEs. 6-MeO-BDE-47, 2′-MeO-BDE-68, 6-OH-BDE-47, and 2′-OH-BDE-68 were the dominant MeO-PBDEs and OH-PBDEs found in marine organisms (Kelly et al., 2008; McKinney et al., 2006; Verreault et al., 2005; Wan et al., 2009). These *ortho*-substituted OH-PBDEs and MeO-PBDEs have been structurally identified and confirmed as natural compounds (Malmvarn et al., 2005, 2008). The *meta*- and *para*-substituted OH-PBDEs have been reported to be biotransformation products produced during PBDE exposure (Malmberg et al., 2005; Marsh et al., 2006; Qiu et al., 2007). Recently, Wan et al. demonstrated the interconversion of OH-PBDE and MeO-PBDE in Japanese medaka (Wan et al., 2010).

The objectives of this work were to investigate the presence and distribution of OH-PBDEs and MeO-PBDEs in plants lived surrounding a seafood processing factory and a seafood market in Longkou, Shandong province, China. Plants play an important role in the terrestrial ecosystem. The uptake and metabolism of organic contaminants in the environment frequently occurred in plants and represent the first step of the food chain (Collins et al., 2006). Plant uptake of PBDEs was already observed (Tian et al., 2012), however, there have been few studies on the fate of OH-PBDEs and MeO-PBDEs in environmental plants. Seafood processing factories and seafood markets were important places where marine organisms were gathered in land and consumed by human. It was assumed in a previous work that the seafood factories discharge and the

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consumption of marine products by coastal residents were the possible major sources for the detected OH-PBDEs in sewage sludge of coastal cities (Sun et al., 2013). In Seafood processing factories, fish was treated in different ways. Head, viscera and skin of fish are generally the waste of filleting process. In seafood markets, a large amount of marine products were traded and also much waste was produced. The OH- and MeO-PBDEs existed in fish and mollusk could be released and transferred to the surrounding environment such as atmosphere, water and soil. Seafood processing factories and seafood markets were possible sources for OH- and MeO-PBDE in terrestrial environment. It is important to evaluate the potential exposure risks of OH- and MeO-PBDEs for the environment and human health.

2. Experimental section

2.1. Materials, standards and reagents

Commercial standards of MeO-PBDEs (4-MeO-BDE-42, 4′-MeO-BDE-49, 3-MeO-BDE-47, 5-MeO-BDE-47, 6-MeO-BDE-47, 2′-MeO-BDE-68, 6-MeO-BDE-85, 5′-MeO-BDE-99, and 6′-MeO-BDE-99) and OH-PBDEs (4-OH-BDE-42, 4′-OH-BDE-49, 3-OH-BDE-47, 5-OH-BDE-47, 6-OH-BDE-47, 2′-OH-BDE-68, 6-OH-BDE-85, 5′-OH-BDE-99, and 6′-OH-BDE-99) were purchased from AccuStandard (New Haven, CT, USA). Several PBDEs congeners which were potential precursors of above MeO- and OH-PBDEs including BDE-28, 47, 66, 68, 99 (AccuStandard, New Haven, CT, USA) were analyzed simultaneously to explore the possible sources of OH-PBDEs and MeO-PBDEs. Surrogate standards (BDE-75 and ^{13}C-6-OH-BDE-47) were purchased from Wellington (Guelph, ON, Canada). Working solutions of PBDEs (in hexane), MeO-PBDEs (in hexane), and OH-PBDEs (in acetonitrile) were prepared at 1 μg/mL for quantitative analysis. Silica gel (100–200 mesh size) was obtained from Merck (Darmstadt, Germany). Deionized water (18.2 MΩ) was obtained from ultrapure water purification system (Barnstead International, Dubuque, USA). Acetonitrile (HPLC grade), methyl *tert*-butyl ether (MTBE) (HPLC grade), 2-propanol (HPLC grade), acetone (pesticide grade), hexane (pesticide grade) and dichloromethane (DCM) (pesticide grade) were purchased from J. T. Baker (Phillipsburg, NJ, USA). All other chemicals and reagents used were of analytical reagent grade or higher purity.

2.2. Sampling site description and sample collection

The sampling map and sites are shown in Fig. 1. Samples were collected around seafood processing factory (Fig. 1A) and seafood market (Fig. 1B), which are located in Longkou, Shandong province, in eastern China. The seafood processing factory started in 1998. The approximate annual amount of seafood products was 80–100 tons. The seafood market started in 1997. The annual amount of traded seafood was about 2200 tons. When sampling, grass which was similar in size and growth age was selected as sample. At each sampling site, one sample consisted of 3–5 plant individuals. Sampling sites A1, A2, A3, A4 and A5 were located in an area (grey color) inside the seafood processing factory, where head, viscera and skin of fish were discarded. Site A6 was located in a flower bed and did not contact seafood or any waste directly. The wastewater generated during fish processing was discharged into a river in the north of the factory through a sewage pipe. Site A7 was located at the end of sewage pipe where the effluent entered the riverway. Site A8, A9, A10, A11 was selected downstream along the riverside at specific intervals of about 10 m. Site A12 was a control sampling site which was 40 m upstream of site A7. Another control sampling site was site A13 which was one hundred meters away from the factory. Grass samples (*Capsella bursa-pastoris*) (expressed as Cb) was sampled at site A1, A4–A13 and *Erigeron annuus* (L.) Pers. (expressed as Ea) was sampled at site A2 and A3. The grass samples were divided into roots and leaves. The relevant rhizosphere soil was also collected. A fish sample, hairtail (*Coiliaspp*) which consisted of 5 individuals was collected inside the factory and wastewater from sewage pipe was collected at site A7.

Around seafood market, eight sampling sites (B1–B8) were selected in different directions around the market. Grass samples, *Setaira viridis* (L.) Beauv (expressed as Sv) were sampled in all sampling sites. In site B4, another plant, *Calystegia soldanella* (Linn.) R. Br. (expressed as Cs), was also sampled to study the interspecies variability. Site B10 was a control sampling site far away in the other side of a river which was located western of the market. Leaves and soil samples were obtained in all sampling sites except for site B9, in the middle of the market, where have no grass but a kind of mollusk, *Crassostrea talienwhanensis*, was collected. In total, 22 leaf samples, 13 root samples, 22 soil samples, one wastewater sample, one fish sample, and one mollusk sample were collected. Before sampling, all the containers were pre-cleaned with acetone. All solid samples were packed in aluminum foil and sealed in Ziplock bags. Biological samples were stored in an ice-box after collection and kept at −20 °C until analysis. The wastewater sample was collected using glass bottles to avoid adsorption, stored at 4 °C during transportation and analyzed as soon as possible after back to the laboratory.

2.3. Sample preparation

The sample preparation procedures for PBDEs, MeO-PBDEs and OH-PBDEs were modified from a previous developed method (Sun et al., 2013). In brief, solid samples were freeze-dried and then homogenized. Soils were sieved through a stainless steel 75-mesh sieve. The sieve was rinsed by acetone between samples to minimize cross-contamination. A suitable amount of the samples (800 mL for water and 2.0 g for solid samples) were spiked with BDE-75 and ^{13}C-6-OH-BDE-47. Wastewater sample was prefiltrated through a 0.45 μm filter membrane and extracted twice with 40 mL of hexane/MTBE (1:1; v/v) after the addition of 4 mL of 2-propanol. The organic extracts were combined and evaporated to dryness. The solid samples were extracted three times in an ultrasonic bath with 10 mL of hexane/MTBE (1:1; v/v) after the addition of 2 mL of 2-propanol. The extracts were combined and dried under a gentle flow of high-purity nitrogen. The dried residues were then dissolved in 30 mL of DCM. Acidified silica gel (10 g, 44% H_2SO_4 acidified) was added to remove any lipids in the extract and the sulfuric acid residue was eliminated through an anhydrous Na_2SO_4 column. The extract was thereafter concentrated by rotary evaporation to ~2 mL and fractioned on a silica (5 g, deactivated with 5% water) column. The column was first preconditioned by 30 mL of hexane and eluted by 60 mL of 20% DCM in hexane, and 70 mL of DCM in sequence. PBDEs and MeO-PBDEs were eluted in the first fraction and concentrated to a volume of 100 μL prior to GC–ECNI–MS analysis. OH-PBDEs were eluted in the second fraction and concentrated and solvent exchanged to 100 μL of acetonitrile for subsequent LC–MS/MS determination.

2.4. Instrumental analysis

Analysis of PBDEs, MeO-PBDEs and OH-PBDEs was performed based on a previously developed method (Sun et al., 2013). For PBDEs and MeO-PBDEs analysis,

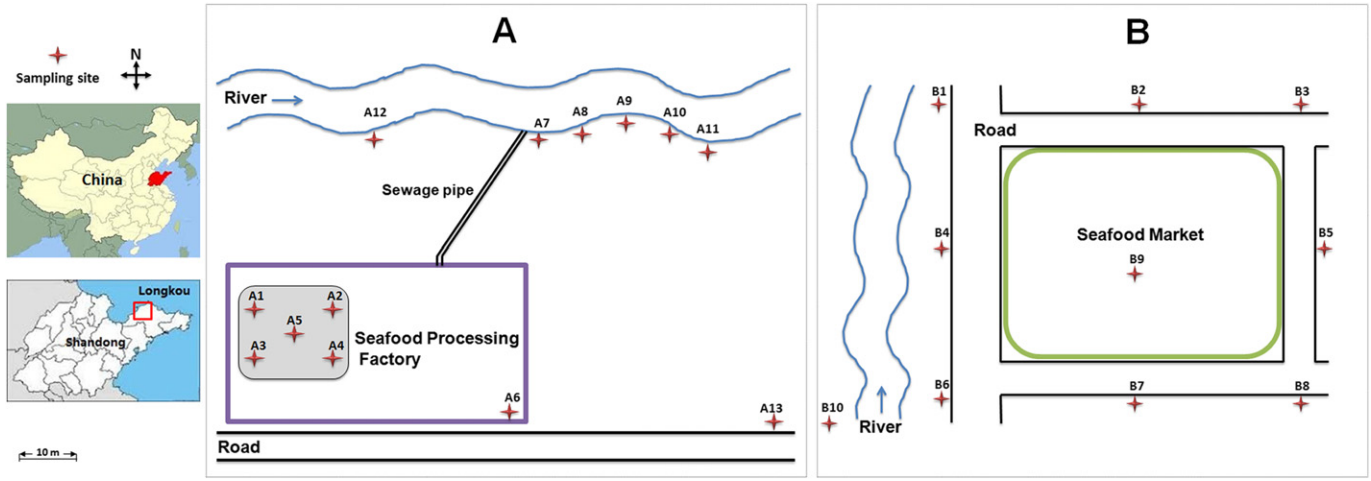

Fig. 1. Sampling locations around the seafood processing factory (A) and seafood market (B).

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