



Distribution and partitioning of polybrominated diphenyl ethers in sediments from the Pearl River Delta and Guiyu, South China[☆]

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) were investigated by GC–NCI–MS in sediments collected from the Pearl River Delta (PRD) and Guiyu town, South China. The concentrations of \sum_{39} PBDEs and BDE 209 were in the ranges of 0.31–38.9 ng g⁻¹ and 12.2–488 ng g⁻¹ in the PRD, and 2.57–21,207 ng g⁻¹ and 7.02–66,573 ng g⁻¹ in Guiyu, respectively. The levels of PBDEs in Dongjiang River (DJ), Zhujiang River (ZJ), and Beijiang River (BJ), and Guiyu (GY) followed the order: GY > DJ > ZJ > BJ. The very high PBDE concentration (87,779 ng g⁻¹) was detected at G1 sediment in Guiyu compared with those in sediments from other regions around the world. The PBDE mixtures detected were mainly comprised of penta-, octa-, and deca-BDEs, in which deca-BDE was the dominant constituent. The abundant congeners, excluding BDE-209, were BDE-47, BDE-99, and BDE-183, suggesting the diverse use of commercial products containing these congeners in this region. The concentrations of major congeners were significantly correlated with total organic carbon (TOC) contents ($p < .01$). A good regression between the logarithmic TOC-normalized BDE average concentrations and their log K_{ow} confirmed that the sorption of PBDEs on sediment organic matter governed their spatial distribution, transport, and fate in the sediments. Furthermore, risk quotients (RQs) derived from concentrations of PBDEs in sediments from our study may pose high ecological risks to exposure of benthic organisms.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a group of additive flame-retardants, which have been widely utilized in many types of polymers such as plastics, paints, electrical components, textiles, foam, rubber, and other casing materials. In previous reports, PBDEs were found in many commercial and household products (Chen et al., 2009; Cheng et al., 2014; Hale et al., 2002; Ran et al., 2013). The commercial PBDE mixtures contain penta-BDE, octa-BDE, and deca-BDE, among which the penta- and octa-products include different BDE congeners, while deca-product mainly consists of BDE-209 (Hites, 2004; La Guardia et al., 2006). It was estimated that the worldwide demand for these compounds in 2001 was approximately 70,000 tons, among which 49%, 37%, and 12% were used in North America, Asia, and Europe, respectively (de Wit, 2002; Hites, 2004). However, PBDEs have attracted considerable

concerns in recent years due to their long persistence, bio-accumulation, increasing detectable frequency, and potential toxic effects on every level of living organisms including humans and wildlife (Gorgy et al., 2010; Macias-Zamora et al., 2016; Tombesi et al., 2017; Wang et al., 2015b).

Currently, e-waste recycling is considered to be a significant source of released PBDEs to the environment especially in developing countries. E-wastes are generated from the fast growing disposal of electronic products throughout the world, most of which are transported to developing countries and dismantled. It was reported by United Nations Environment Programme (UNEP) in 2005 that approximately 80% of the computer e-wastes were exported to Asia, 90% of which enters into China through illegal imports (UNEP, 2005). Usually, during e-waste recycling, valuable metals from electronics are recovered by primitive techniques, while other parts such as large amounts of plastic are either burned or subsequently dumped in the wide fields (Luo et al., 2011), leading to the release of a wide range of hazardous chemicals such as PBDEs into the surrounding environment. It has been reported that extremely high levels of PBDEs were found in combusted

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residues, soils, sediments, water, plants, aquatic species, and even human serum surrounding the typical e-waste sites in China and other countries (Cheng et al., 2014; Leung et al., 2007, 2010; Nnorom and Osibanjo, 2008). Consequently, the concentrations of PBDEs in the environment matrix surrounding the e-waste sites were found to be 2–3 magnitudes higher than those found in environment further far away from e-waste sites (Deng et al., 2007). Zhao et al. (2009) used *Cinnamomum camphora* leaves as biomonitors to investigate the diffusion of PBDEs from an e-waste recycling area to the surrounding regions, demonstrating that the contamination of PBDEs extended to a radius of at least 74 km. The above facts indicate that PBDE congeners emitted could easily transfer in different environmental compartments, enter the food chain transfer, and bioaccumulate in various trophic organisms. Therefore, the investigation on distribution and partitioning of PBDEs in soils, waters, and sediments could provide valuable information for further understanding their exposure risks and fates in contaminated environment (Besis and Samara, 2012; Leung et al., 2010; Macias-Zamora et al., 2016; Zhang et al., 2010).

The PRD is one of the most economically prosperous regions in China with an area of 41,700 km² and 41 million inhabitants. It is the largest production base for computers, electronic parts, and communication instruments. It is also the largest e-waste recycling processing site in China. Moreover, as it is located in the northern subtropical zone, frequent rainfalls during the year are abundant, which could facilitate the transport of different contaminants including PBDEs to aquatic environments. In addition, Guiyu town (GY) having a total area of 52 km² and a population of 150,000 is a well-known e-waste recycling site as 80% of families and nearly 10,000 migrants living in this town are engaged in e-waste recycling works. However, the dismantling methods to manage the electronic wastes are primitive, with little safety measures and attempts (gloves and breathing masks, etc) taken to control the exposure to complex chemicals presented in the e-wastes (Deng et al., 2006; Zhao et al., 2006), which pose a threat to the health of local residents and workers.

The present investigation was conducted to characterize the spatial distribution and partitioning of PBDEs in sediments from the PRD and Guiyu town in order to provide a better understanding the contamination extents, potential risks, and fates of PBDEs in the investigated region.

2. Materials and methods

2.1. Study area and sample collection

Three major tributaries of the Pearl River system were selected for sampling (<http://www.sciencedirect.com/science/article/pii/S0304389411007254> Fig. 1). Dongjiang River (DJ) and Zhujiang River (ZJ) are located in the northeast of the PRD. DJ serves as the drinking water source of Dongguan, Shenzhen, and Hong Kong. It runs across the city of Dongguan, where manufacturing and processing industries are well developed. ZJ flows through Guangzhou which is the biggest and highly urbanized city with a population of 12 million and various industries in South China. These two rivers join at the Shizhiyang Waterway and flow into the Pearl River Estuary (PRE) through the Humen outlet. On the other hand, Beijiang River (BJ), located in the southwest of the PRD, runs across less developed areas and flows into the estuary via the Jiaomen, Hongqimen, and Hengmen outlets. In this study, fifteen surface sediments (0–5 cm) were collected from DJ, ZJ, and BJ (Fig. 1) in July 2006 using a stainless steel grab sampler. Because the average sedimentation rates were 1.33 cm yr⁻¹ in the Zhujiang River (Zhang et al., 2002), the top 5-cm layer of sediments was expected to represent modern inputs. In order to investigate the contaminated

levels of PBDEs in point-source sites, five surface sediments were also collected from the e-waste site (GY) following the same procedures in February 2007 (Fig. 1). The sediment samples were kept at –20 °C until further analysis.

2.2. Chemicals

Mixed standards of 39 PBDE congeners (including BDE-1, -2, -3, -7, -8, -10, -11, -12, -13, -15, -17, -25, -28, -30, -32, -33, -35, -37, -47, -49, -66, -71, -75, -77, -85, -99, -100, -116, -118, -119, -126, -138, -153, -154, -155, -166, -181, -183, -190) and individual standard of BDE-209, were purchased from Accustandards (New Haven, CT). Two surrogate standards of ¹³C-PCB-141 and PCB-209 and also ¹³C-PCB-208 as an internal standard were obtained from Cambridge Isotope Laboratories (Andover, MA), Ultra Scientific (North Kingstown, RI) and Cambridge Isotope Laboratories (Andover, MA), respectively.

2.3. Samples pretreatment and analytical procedure

Analysis of PBDE congeners was performed through multi-step procedure including soxhlet extraction, chromatographic column purification, and GC-NCI-MS analysis following the method described previously (Zou et al., 2007). First, about 10 g freeze-dried and grinded samples were spiked with two surrogate standards (PCB-209 and ¹³C-PCB-141), extracted with acetone and hexane mixture (1:1 v:v) for 48 h by using Soxhlet extractors, and activated copper was added for desulphurization during the extraction. Then, the extracts were concentrated, cleaned, and consequently fractionated on a 1 cm i.d. silica/alumina column. The purified column was packed with neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), 25% sodium hydroxide silica (5 cm), neutral silica gel (2 cm, 3% deactivated), 50% sulfuric acid silica (8 cm), and anhydrous sodium sulfate (1 cm). The PBDE mixture was eluted with 30 ml of hexane and 60 ml of hexane:methylene chloride (1:1), and the final extract volume was reduced to 500 µL under a gentle N₂ stream. Prior to instrumental analysis, ¹³C-PCB-208 was added to each of the extracts as the internal standard, and finally the extracts were analyzed by GC-NCI-MS using an HP-5MS capillary column (30 m × 0.25 mm i.d. with 0.25 µm film thickness) (J&W Scientific Corp., CA, USA) and a DB-5MS capillary column (15 m × 0.25 mm i.d. with 0.25 µm film thickness) to separate 39 PBDE congeners and BDE-209, respectively.

2.4. TOC measurement

Measurement of TOC was accomplished according to previously reported method (Ran et al., 2007). Three grams of dried sediment sample were treated with 1M HCl to remove inorganic carbon and followed by washing three times with de-ionized water and drying overnight at 60 °C. Then, the total organic carbon was measured with an elemental analyzer (VarioEL III Elementar, Germany). Acetanilide was used as an external standard.

2.5. Ecological risk assessments of PBDEs in sediments

The risk quotient (RQ) approach is a widely used method in risk assessment studies on heavy metals and organic pollutants (Khairy et al., 2009; Wang et al., 2015a). It provides a quantitative estimate of the hazard associated with a single chemical or element (Environment Canada. Canadian Environmental Protection Act, 2013). In this study, the RQ values were determined by dividing the measured concentrations of PBDE congeners in the sediments by their respective sediment quality guidelines. Prior to the RQ

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