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Bioaccumulation and biomagnification of halogenated organic pollutants in mangrove biota from the Pearl River Estuary, South China

Yu-Xin Sun [a], Zai-Wang Zhang [a,d], Xiang-Rong Xu [a,*], Yong-Xia Hu [a,d], Xiao-Jun Luo [b], Ming-Gang Cai [c], Bi-Xian Mai [b]

[a] Key Laboratory of Tropical Marine Bio-resources and Ecology, Guangdong Provincial Key Laboratory of Applied Marine Biology, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China
[b] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[c] College of Ocean and Earth Sciences, Xiamen University, Xiamen 361005, China
[d] University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

Four biota species were collected from mangrove ecosystems of the Pearl River Estuary to investigate the bioaccumulation and biomagnification of polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), polybrominated diphenyl ethers (PBDEs), dechlorane plus (DP), and decabromodiphenyl ethane (DBDPE). Concentrations of ΣPCBs, ΣDDTs, ΣPBDEs, DP, DBDPE and *anti*-Cl_{11}-DP (the dechlorination product of *anti*-DP) in mangrove biota ranged from 32.1–466, 153–3819, 3.88–59.8, 0.18–6.88, not detected (nd)–30.6 and nd–2.65 ng/g lipid weight, respectively. Daggertooth pike conger (*Muraenesox cinereus*) had higher concentrations of contaminants than the other three biota species. Significant positive relationship between *anti*-Cl_{11}-DP and *anti*-DP levels was observed in mangrove biota. DDTs were the predominant HOPs in all biota species, followed by PCBs and PBDEs. All the target compounds exhibited biomagnification, with biomagnification factors greater than 1 in the studied feeding relationships. Food web magnification was found for ΣPCBs, ΣDDTs, ΣPBDEs and DP, with trophic magnification factors of 2.76, 2.61, 2.20 and 2.31, respectively.

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1. Introduction

Halogenated organic pollutants (HOPs), such as polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), polybrominated diphenyl ethers (PBDEs), dechlorane plus (DP), and decabromodiphenyl ethane (DBDPE), have been of great concern due to their persistence, bioaccumulation and potential toxicity to wildlife and human. PCBs were used primarily as dielectric and coolant fluids in capacitors, transformers and electric fluids (Xing et al., 2005). DDT was extensively used as an agricultural insecticide during 1940–1970s (van den Berg, 2009). PCBs and DDT were banned and added to the list of the 12 initial persistent organic pollutants (POPs) under the Stockholm Convention in 2001. PBDEs (including Penta-, Octa- and Deca-BDE commercial formulations), DP and DBDPE are important additive flame retardants widely used in electronics, textiles, thermoplastics, polyurethane foams and building materials (Alaee et al., 2003; Covaci et al., 2011; Sverko et al., 2011). Among them, Penta- and Octa-BDE technical mixtures have been added to the list of emerging POPs by the Stockholm Convention in 2009 and Deca-BDE technical mixtures have been phased-out in Europe and America, while Deca-BDE, DP and DBDPE are still widely used in China (Sverko et al., 2011; Zhang et al., 2015). Therefore, more studies are still needed to understand the environmental behaviors of these contaminants.

Mangrove ecosystems, the intertidal wetlands along the tropical and subtropical coastlines, are unique transitional ecosystems between terrestrial and marine environments and have important ecological services and economic value, such as prevention of flood and shoreline erosion, tourism and fisheries (Lewis et al., 2011). Although their functions have received great attention, mangroves are considered as one of the most threatened marine ecosystems due to natural and anthropogenic activities including deforestation, hurricane, insect infestations and chemical pollution (Lewis et al., 2011; Bayen, 2012). Mangrove sediments can act as sinks for varieties of man-made pollutants (Vane et al., 2009; Zhang et al., 2014). To date, several studies have been conducted on PCBs, DDTs and PBDEs contamination in mangrove sediments (Zheng et al., 2000; de Souza et al., 2008; Zhu et al., 2014; Zhang

* Corresponding author.
E-mail address: xuxr@scsio.ac.cn (X.-R. Xu).

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et al., 2015), however, little information on bioaccumulation and biomagnification of these HOPs, especially for PBDEs, DP and DBDPE, in mangrove ecosystems, could be found in literature.

The Pearl River Delta (PRD), as one of the most prosperous regions in China, has become a hotspot for HOPs contamination due to rapid industrialization and urbanization, as well as agricultural development. The Pearl River Estuary (PRE) has acted as a major reservoir for HOPs discharged from municipal sewage, industrial waste, and upstream runoff in the PRD, which may pose adverse effects to mangrove ecosystems in the coastal areas (Fu et al., 2003; Zhang et al., 2014). Although the contamination of PCBs, DDTs, PBDEs and DBDPE has been found in mangrove sediments from the PRE (Zheng et al., 2000; Tam and Yao, 2002; Zhu et al., 2014; Zhang et al., 2015), data on the occurrence of these contaminants in mangrove biota, which might be exposed to elevated levels of HOPs, is scarce. Therefore, an investigation on bioaccumulation and biomagnification of HOPs is essential to gain a better understanding of their environmental behaviors in mangrove ecosystems.

In the present study, four biota species from the Qi'ao Island Mangrove Nature Reserve of Zhuhai in the PRE were collected and analyzed for PCBs, DDTs, PBDEs, DP and DBDPE. The objectives of this study were to explore the levels and composition profiles of these HOPs in mangrove biota in the PRE and investigate the bioaccumulation and biomagnification of these HOPs in mangrove food chains. To the best of our knowledge, this is the first study to investigate the bioaccumulation and biomagnification of DP and DBDPE in mangrove ecosystems.

2. Materials and methods

2.1. Sample collection

A total of 22 biota samples were collected from the Qi'ao Island Mangrove Nature Reserve of Zhuhai in the PRE of South China in November 2012 (Fig. 1). There are 33 mangrove plant species in this reserve with an area of 51 km^2. Mangrove biota species, including 5 mud crabs (*Scylla serrata*, MC), 6 Chinese black sleepers (*Bostrichthys sinensis*, CBS), 6 red eelgobys (*Odontamblyopus rubicundus*, RE), and 5 daggertooth pike congers (*Muraenesox cinereus*, DPC), were captured by a fishing net. Detailed information on the feeding habits of each species is given in Table S1 (Supplementary material). All samples were kept in a refrigerator at −20 °C and immediately transferred to the laboratory. Biota samples were cleaned with deionized water. Muscle tissue was excised from each species and stored at −20 °C until chemical analysis.

2.2. Sample extraction and cleanup

The extraction and cleanup for HOPs in mangrove biota samples followed the procedures described by Sun et al. (2014a). Briefly, muscle tissue was freeze-dried, grounded into fine powder, mixed with anhydrous sodium sulfate, spiked with surrogate standards (PCB 30, 65 and 204, $^{13}C_{12}$-BDE 209, BDE 77, 181 and 205) and then Soxhlet extracted with 200 mL acetone/hexane (*v*/*v* = 1:1) for 48 h. One aliquot of the extract was used for the gravimetric determination of lipid content. Another extract was purified by a gel permeation chromatographic column packed with 40 g SX-3 Bio-Beads (Bio-Rad Laboratories, Hercules, CA) and further cleaned up on a multilayer column filled with 1 cm anhydrous sodium sulfate, 8 cm acidified silica, and 8 cm neutral silica from top to bottom. The extract was eluted with 30 mL hexane/dichloromethane (*v*/*v* = 1:1) and the eluate was concentrated to near dryness under a gentle nitrogen flow and reconstituted in 100 μL of isooctane. Internal standards (PCB 24, 82 and 198, $^{13}C_{12}$-PCB 208, 4-Fluoro-BDE 67, 3-Fluoro-BDE 153, BDE 118 and 128) were spiked before instrumental analysis.

2.3. Instrumental analysis

Nineteen PCB congeners (PCB 28, 52, 60, 66, 87, 99, 101, 107, 110, 118, 128, 136, 138, 141, 153, 164, 170, 174, 180, 183, 187 and 209), DDT and its metabolites (*p*,*p*′-DDM, *p*,*p*′-DDE, *o*,*p*′-DDE, *p*,*p*′-DDMU, *p*,*p*′-DDD, *o*,*p*′-DDD, *p*,*p*′-DDT, *o*,*p*′-DDT) were analyzed by an Agilent 7890 gas chromatograph (GC) coupled with an Agilent 5975C mass spectrometer (MS) using electron impact in the selective ion-monitoring (SIM) mode and separated by a DB-5MS (60 m × 0.25 mm × 0.25 μm, J&W Scientific) capillary column. BDE 28, 47, 66, 99, 100, 153, 154, and 183, *anti*- and *syn*-DP, *anti*-Cl_{11}-DP were quantified by an Agilent 6890 GC coupled with 5975 MS with electron capture negative ionization (ECNI) in the SIM mode and separated by a DB-XLB (30 m × 0.25 mm × 0.25 μm, J&W Scientific) capillary column. BDE 202 BDE 209 and DBDPE were analyzed by a Shimadzu model 2010 GC coupled with a model QP2010 MS (Shimadzu, Japan) using ECNI in the SIM mode and separated by a DB-5HT (15 m × 0.25 mm × 0.10 μm, J&W Scientific) capillary column. More information on instrumental analysis can be found elsewhere (Luo et al., 2009).

2.4. Stable isotope analysis and trophic level (TL) determination

The subsamples of muscle tissue were lyophilized and grounded into ultra-fine powder for stable nitrogen isotope analysis. Approximately 0.8 mg of the grounded samples was weighed in tin capsule and analyzed by a Flash EA 112 series elemental analyzer interfaced with a Finigan MAF ConFlo 111 isotope ratio mass spectrometer. The stable nitrogen isotope composition is expressed as δX (‰), with $\delta X = (R_{sample}/R_{standard} - 1) \times 1000$, where δX is $\delta^{15}N$ and R is the corresponding ratio of $^{15}N/^{14}N$. The precision for this technique is 0.5‰ (2 SD) for $\delta^{15}N$.

TLs of the biota species were calculated according to the following equation (Loi et al., 2011):

$$TL_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{zooplankton})/3.8$$

where $TL_{consumer}$ and $\delta^{15}N_{consumer}$ are the TL and stable nitrogen isotope abundance of the consumer, respectively; $\delta^{15}N_{zooplankton}$ is the stable nitrogen isotope abundance of the zooplankton with an average of 5.376‰; 3.8 is the enrichment factor.

2.5. Biomagnification factor (BMF) and trophic magnification factor (TMF)

BMF was calculated as the ratio of the lipid normalized HOPs concentrations in the predator and prey species using the following equation:

$$BMF = C_{predator}/C_{prey}$$

where $C_{predator}$ and C_{prey} are the chemical concentrations in the predator and prey species, respectively.

The trophic transfer of HOPs through a food chain is based on the relationship between TL and HOPs concentrations:

$$\ln C = a + b \times TL$$

where *C* is the concentrations of HOPs, *a* is the *y*-intercept (constant), *b* is the slope of the regression of ln*C* against TL. TMFs for each contaminant were calculated from the slope using the following equation:

$$TMF = e^{b}$$

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