



Concentrations and distributions of polybrominated diphenyl ethers and novel brominated flame retardants in tree bark and human hair from Yunnan Province, China



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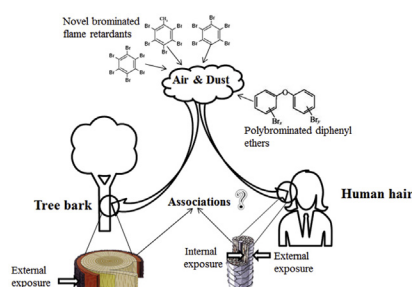
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HIGHLIGHTS

- Associations between BFR concentrations in tree bark and hair samples from inhabitants of the same areas were found.
- BDE-209 was found to be the source of lower BDE congeners.
- External exposure was found to be the main factor for BDE-209 and PBT concentration in hair.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 21 January 2016

Received in revised form

27 March 2016

Accepted 28 March 2016

Available online 6 April 2016

Handling Editor: Myrto Petreas

Keywords:

Polybrominated diphenyl ethers
Novel brominated flame retardants
Tree bark
Human hair

ABSTRACT

The concentrations and distributions of polybrominated diphenyl ethers (PBDEs) and novel brominated flame retardants (NBFRs) in tree bark and hair samples from the same area in Yunnan Province, China, were determined. The total PBDE and NBFR concentrations in the tree bark samples were 3.8 ng/g lipid weight to 91 ng/g lipid weight and 0.23 ng/g lipid weight to 5.0 ng/g lipid weight, respectively. The total PBDE and NBFR concentrations in the hair samples were 2.1 ng/g dry weight to 14 ng/g dry weight and 0.083 ng/g dry weight to 0.29 ng/g dry weight, respectively. Decabromodiphenyl ether had similar distributions in the tree bark and hair samples, but other PBDE congeners and NBFRs had different distributions in the tree bark and hair samples. External exposure was found to be mainly responsible for the total PBDE and pentabromotoluene concentrations in hair, but both external and internal exposure were responsible for the pentabromophenyl and hexabromobenzene concentrations in hair.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) are synthetic

halogenated organic compounds that have been widely used in commercial and household products, such as electronics, furniture, and textiles (Shaw and Kannan, 2009). Three main commercial PBDEs mixtures, namely Penta-BDE, Octa-BDE, and Deca-BDE, have been used. Octa-BDE has never been produced in China, and production of Penta-BDE in China ceased in 2004, however in China, Deca-BDE is currently being produced and is used without restriction (Ni et al., 2013). It has previously been reported that PBDE

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exposures may cause thyroid hormone disruption and neuro-developmental harm (Zota et al., 2013). PBDEs have been forbidden gradually around the world because of their potential toxicity (Kang et al., 2011). Restrictions on the production and use of 'traditional' brominated flame retardants have led to novel brominated flame retardants (NBFRs), including pentabromophenyl (PBBz), pentabromotoluene (PBT), 2,3,5,6-tetrabromo-*p*-xylene, and hexabromobenzene (HBB), to be widely used as alternatives of PBDEs (Xian et al., 2008; Qiu et al., 2010). NBFRs have been detected in air (Venier et al., 2012), sediment (Olukunle and Okonkwo, 2015), water (Cristale et al., 2013), biological samples (Wang et al., 2014), and other media. Wang et al. (2014) analyzed brominated flame retardants (BFRs) in pooled serum samples from inhabitants of the south coast of Laizhou Bay (Shandong Province, China), and found that the BFR concentrations and distributions in the samples from different age groups were closely related to historic BFR production and usage volumes in China. This suggested that humans are gradually being exposed to higher concentrations of BFRs as more Deca-BDE and NBFRs are being produced and used (Wang et al., 2014). BFRs are found at relatively high concentrations in cities and electronic waste recycling areas in China (Zhao et al., 2008b; Wen et al., 2009; Hu et al., 2011), but the concentrations and distributions of BFRs, particularly NBFRs, in remote parts of China are not well understood.

Tree bark has a relatively high lipid content, is widespread, and is conveniently sampled, so it is often used to monitor persistent organic pollutants (POPs) in the environment (Hermanson and Johnson, 2007; Salamova and Hites, 2010; Peverly et al., 2015). The surface of bark is very porous, and the absence of metabolic processes in bark means that inorganic and organic substances present in bark are almost inert, so bark is a useful passive bio-indicator for the presence of airborne pollutants (Samecka-Cymerman et al., 2006). It has been concluded in some studies that the aerial parts of trees (i.e., leaves and bark) have large capacities for accumulating POPs from the atmosphere and that trees take up negligible amounts of POPs from soil unless the soil is very contaminated (Tarcau et al., 2013). POP concentrations in bark and the atmosphere therefore tend to positively correlate, and bark can be used to study the long distance migration of POPs and to identify potential sources of POPs (Fu et al., 2014; Peverly et al., 2015).

Hair is a keratinous matrix consisting of 88% protein and 3%–4% lipid, and POP concentrations in hair can be determined readily (Poon et al., 2014). It is convenient and harmless to sample human hair (Poon et al., 2014). Hair can be exposed to BFRs through two routes, external exposure (deposition from air and dust) and internal exposure (through exchange between blood and the root of the hair follicle) (Covaci et al., 2002). Hair is therefore often used to identify the main pathways through which humans are exposed to POPs (Zheng et al., 2011; Tang et al., 2013; Krol et al., 2014). However, hair has been used to monitor BFRs, and particularly NBFRs, in very few previous studies. Associations between BFR concentrations in tree bark samples and hair samples from inhabitants of the same areas have not been assessed before.

Kaiyuan City is in the southeastern part of Yunnan Province, and is in the Hani-Yi Autonomous Prefecture of Honghe. The 'great western development strategy' and the 'belt and road initiative' have led to the economy in Kaiyuan City developing rapidly in recent years, and the population of Kaiyuan City continues to increase. In 2013, Kaiyuan City had 310200 inhabitants and a gross domestic product of 1.4×10^{10} yuan (Li, 2014). The risk of humans being exposed to BFRs increases as the population increases and industry becomes more developed, but the current state of BFR pollution in Kaiyuan City is not well understood.

The aims of this study were (1) to determine the BFR concentrations in tree bark and human hair from Yunnan Province,

Southwest China, (2) to attempt to identify possible BFR sources in the study area, and (3) to identify similarities in and differences between the BFR distributions in bark and hair from the same area and to identify the possible pathways through which BFRs migrate to tree bark and human hair.

2. Materials and methods

2.1. Sampling

Thirteen sampling sites in Kaiyuan City were selected. The sampling sites were (in order of increasing altitude) at Niluojiang (labeled NLJ), Tonglingcun (TLC), Shiyancun (SYC), Lujiao (LJ), Lebaichong (LBC), Zhonghe-Ying (ZHY), Bajiazai (BJZ), Shantaipu (STP), Chenggan (CG), Hulutang (HLT), Deguo (DG), Dama-zhe (DMZ), and Beige (BG). Detailed sampling information is given in Table 1, and the locations of the sampling sites are shown in Fig. 1.

All of the samples were collected in January 2014. At this time, the meteorological conditions were relatively stable, and the temperature was relatively low, which were beneficial to the collection and transport of our samples. The bark samples collected and analyzed in this study are the same as those reported in our previous study (He et al., 2014). Bark was chiseled from around the trunks, at approximately 1.5 m (the average height of human respiration) above the ground, of three pine trees at each site. The mean trunk diameter at the sample height was 35 cm, to ensure the tree bark had fully absorbed pollutants from the air. The hair samples collected and analyzed in this study are the same as those reported in our previous study (Zhou et al., 2015). Hair samples were collected from people living within 10 km of where the tree bark samples were collected. Each pooled hair sample (non-dyed) was collected from 10 people (5 male and 5 female) aged from 20 to 40, each person provided a hair of 5 g. The sampling site was on the occipital area, and hair segments were cut with 1–5 cm from hair root. In total, 13 pooled bark samples and 13 pooled hair samples were collected. Each sample was wrapped in aluminum foil, sealed in a plastic bag, and stored at 5 °C for transport to the laboratory. In the laboratory, the samples were stored at –20 °C.

2.2. Chemicals

Pesticide grade acetone, dichloromethane, and *n*-hexane were obtained from J.T. Baker (Phillipsburg, USA), high purity nitrogen was obtained from Cheng Wei Xin (Beijing, China), and ultra-pure water was produced using a Milli-Q system (EMD Millipore, Billerica, MA, USA). Analytical grade anhydrous sodium sulfate was baked at 450 °C for 5 h before use. Silica gel (100–200 mesh; Merck, Darmstadt, Germany) activated at 105 °C for 12 h, then cooled and deactivated by adding 3% of the sorbent weight of deionized water.

Individual PBDE standards (BDE-28, BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-206, and BDE-209) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). A PBT standard was purchased from AccuStandard (New Haven, CT, USA). A HBB standard was purchased from Dr. Ehrenstorfer (Augsburg, Germany). 2,3,5,6-Tetrabromo-*p*-xylene and PBBz standards were purchased from Wellington Laboratories (Guelph, Canada). Internal standards ($^{13}\text{C}_{12}$ -labeled BDE-139, $^{13}\text{C}_{12}$ -labeled BDE-209, and $^{13}\text{C}_6$ -labeled HBB) were purchased from Cambridge Isotope Laboratories.

2.3. Sample preparation

Preparation of bark samples. Each bark sample ($n = 13$) was dried, then each raw bark sample (10.0000 g) was spiked with internal standards (2.5 ng $^{13}\text{C}_{12}$ -labeled BDE-139, 25 ng $^{13}\text{C}_{12}$ -

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