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Brominated flame retardants (BFRs): A review on environmental contamination in China

Gang Yu [a], Qingwei Bu [a, b, *], Zhiguo Cao [c], Xinming Du [a], Jing Xia [a], Min Wu [a], Jun Huang [a, **]

[a] School of Environment, State Key Joint Laboratory of Environmental Simulation and Pollution Control, Beijing Key Laboratory for Emerging Organic Contaminants Control, Tsinghua University, Beijing 100084, China
[b] School of Chemical & Environmental Engineering, China University of Mining & Technology-Beijing, Beijing 100083, China
[c] School of Environment, Henan Normal University, Key Laboratory of Yellow River and Huai River Water Environment and Pollution Control, Ministry of Education, Henan Key Laboratory for Environmental Pollution Control, Xinxiang, Henan 453007, China

HIGHLIGHTS

- Contamination of major BFRs in China were reviewed comprehensively.
- High levels of PBDEs were observed around e-waste dismantling areas.
- Decreasing trend of PBDEs in human milk was observed in some regions.
- Indoor dust was highlighted due to the possible high human exposure.
- High exposure of residents and workers in e-waste dismantling areas was indicated.

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ABSTRACT

Brominated flame retardants (BFRs) which were detected extensively in environmental and biota samples worldwide, have raised significant concerns during past decades for their persistence, bioaccumulation and potential toxicity to ecological environment and human health. In this paper, we have compiled and reviewed existing literature on the contamination status of BFRs in abiotic and biotic environments in China, including polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane, tetrabromobisphenol A and new BFRs. Temporal trends were also summarized and evaluated. Based on this review, it has been concluded that (1) high concentrations of PBDEs were generally related to the e-waste disposal processing, while the spatial distribution pattern of other BFRs was not necessarily in accordance with this; (2) extremely high concentrations of BFRs in indoor dust emphasized the importance of indoor contamination to human body burdens, while more work need to be done to confirm its contribution; (3) PBDEs in electronics dismantling workers were higher compared to the general population, indicating the occupational exposure should be of particular concern; (4) more data are now becoming available for BFRs in aquatic and terrestrial organisms not previously studied, while studies that consider the occurrence of BFRs in organisms of different trophic levels are still of urgent need for evaluating the fate of BFRs in the food web; and (5) limited data showed a decreasing trend for PBDEs, while more data on time trends of BFR contamination in various matrices and locations are still needed before the impact of regulation of BFRs can be assessed.

1. Introduction

Brominated flame retardants (BFRs) are chemicals widely incorporated as additives in consumer products such as electronics, vehicles, polyurethane foams, textiles to reduce the likelihood of ignition of the flammable materials (de Wit, 2002), and include diverse chemical classes of compounds. There is raised concern

* Corresponding author.
** Corresponding author.
E-mail addresses: yg-den@tsinghua.edu.cn (G. Yu), qingwei.bu@cumtb.edu.cn (Q. Bu), wq11ab@163.com (Z. Cao), dxm13@mails.tsinghua.edu.cn (X. Du), xiaj14@mails.tsinghua.edu.cn (J. Xia), wum10@mails.tsinghua.edu.cn (M. Wu), huangjun@tsinghua.edu.cn (J. Huang).

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worldwide on BFRs during past decades as they have detected extensively in the environment, even in locations far from where they were produced or used, as well as in bodies of human and other organisms (Chen et al., 2011; de Wit et al., 2006; de Wit et al., 2010; Goutte et al., 2013). Increasing concern due to their persistence, bioaccumulation and potential toxic effects on the environment and human health (Stieger et al., 2014) has promoted some international organizations and European countries to ban some of them following the precautionary principle.

China produces and consumes a large quantity of products containing BFRs. As the development of economy in China, consumption of commodities containing BFRs (e.g. electronics, clothes and furniture) has been growing rapidly. Moreover, China especially Southern and Southeastern China ‘digests’ a large quantity of electronic waste (e-waste) from developed countries that formed new sources for BFRs. Guiyu and Taizhou have become famous worldwide as the e-waste dismantling has been developed in industrial scale there (Sepúlveda et al., 2010; Wong et al., 2007). Thus numerous investigations on BFRs in various environmental matrices in China have been conducted during the past decade (Luo et al., 2007; Wang et al., 2007a, 2007b; Wong et al., 2007). There is, however, a lack of a systematic investigation that provides a good overview of the environmental contamination of BFRs in different parts of China.

After the first review on BFRs by de Wit (2002), many influential later reviews have been published, such as Alaee et al. (2003), Birnabaum and Staskal (2004), Law et al. (2006), Tanabe et al. (2008), Law et al. (2008), Yogui and Sericano (2009), Daso et al. (2010), de Wit et al. (2010), Marvin et al. (2011), Besis and Samara (2012) and Law et al. (2014). As for China, Wang et al. (2007a) has reviewed part of the situation of BFR contamination in the view of considering the whole East Asia about a decade ago, while a detailed and up-to-date status of BFR contamination in China during the last decade has not been clearly summarized and evaluated. It is the intention of this review to provide an overall contamination status of BFRs in China by reviewing and evaluating a series of papers. Be specific, this review starts with a brief general overview of production and consumption of flame retardants (FRs) in China. The main part of this review is devoted to contamination status of some BFRs in the abiotic and biotic environments in China. Also temporal trend of some BFRs in human milk and some elements concerned was discussed.

Many BFRs are produced and used with varying chemical properties. In this paper, we focus on the most important ones that are and/or were commercially available in the Chinese market, including three main groups of “typical” or “conventional” BFRs (i.e. polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and tetrabromobisphenol A) and four “new” BFRs (NBFRs) (i.e. 2-ethylhexyl 2,3,4,5-tetrabromobenzoate, 1,2-bis(2,4,6-tribromophenoxy)ethane, bis (2-ethylhexyl) tetrabromophthalate and decabromodiphenyl ethane). Details on physio-chemical properties of these chemicals are listed in Table 1.

2. Overview of production and consumption of FRs

Limited information is publicly available for the production of BFRs in China now. Here the production and consumption of FRs were summarized. Literature review showed that progress of the FR industry in China could date back to the late 1970s and early 1980s. The industries dealing with construction, electrical/electronic components, and transportation are the three of greatest importance. According to the statistic in 2009, there were over 100 factories of various sizes that manufacture FRs in China. In recent years, the annual growth rate of production of FRs has reached around 15% between 1999 and 2007 and the production of the halogenated aliphatic phosphates in China has the highest output in the world (Chen and Wang, 2010). In 2001, FRs produced in China were dominated by chlorinated FRs, with production volume of 112,500 tonnes, in addition to which phosphorus, brominated and inorganic FRs contributed production volumes of 9000, 10,500 and 18,000 tonnes, respectively (Liang, 2003). In 2004, FR consumptions were 564,000, 487,000, 160,000 and 269,000 tonnes for United States of American (USA), European Union, Japan and Asia (except Japan), respectively. BFR consumptions in the four regions were 66,000, 56,000, 50,000 and 140,000 tonnes year^{-1}, respectively (Jiang, 2006), with Asia consuming around 60% BFRs among these four regions (Fig. 1a). In 2006, FR production volume was 260,000 tonnes in China and they were mainly manufactured in Shandong and Jiangsu provinces (Zhang and Lu, 2011). From Fig. 1b, it can be found that in 2006 the most widely produced BFRs were deca-BDE (36,000 tonnes), accounting for around 45% of the total BFRs. Other widely produced BFRs included TBBPA (18,000 tonnes), DBBPE (11,000 tonnes), octa-BDE (8000 tonnes) and HBCDs (7500 tonnes) (Jiang, 2006). Phosphorus FRs commonly produced in China were phosphate, phosphonate and phosphite ester, nevertheless, data on output of them is unavailable (Li et al., 2007).

3. Occurrence of BFRs in the abiotic environment

3.1. Sediments

3.1.1. PBDEs

Many BFRs could accumulate in sediments due to their hydrophobic properties (Table 1). Thus sediments may serve as the sink and source of BFRs in the aquatic environment. Concentrations of PBDEs in sediments were determined in most of the major river systems and several local lakes in China. The detailed data are shown in Table S1-1. Among the nine PBDEs, BDE-209 was detected at high concentrations most frequently in China.

For each individual PBDEs, the highest concentrations were all found in local rivers near the e-waste dismantling area in Guiyu, Guangdong. Fig. 2a shows the distribution of concentrations for BED-209 in sediments from the e-waste dismantling and the rest regions. From the figure it can be found that levels of BDE-209 in e-waste dismantling regions (e.g. Taizhou, Qingyuan) were approximately two orders of magnitude higher than those in non e-waste disposal regions. In contrast to these hotspots of BFR contamination, PBDEs were extensively detected in the major river systems in China but at relatively lower concentrations ranging from a few to several hundreds ng g^{-1}. In general, concentrations of each individual PBDEs in Yangtze River were mostly lower than those in Pearl River (e.g. BDE-209, up to 2500 ng g^{-1} (He et al., 2012)). In different lakes and rivers, PBDE levels were relatively higher in southeastern regions with rapid industrial development speeds and high economic levels, such as Guangdong, Jiangsu, and Zhejiang. By contrast, in western areas such as Sichuan and Yunnan, and northern areas such as Anhui and Jilin, the detected concentrations were two or three orders of magnitude lower (Table S1-1).

PBDE concentrations are comparable to those from some of the other countries or regions, such as Belgium (BDE-209, 315–8410 ng g^{-1}) (Covaci et al., 2005), Japan (BDE-209, ND–11,600 ng g^{-1}) (Japan, 1991), Netherland (BDE-47, 0.30–7.10 ng g^{-1}; BDE-99, ND–5.50 ng g^{-1}; BDE-209, ND–510 ng g^{-1}) (de Boer et al., 2000), Sweden (BDE-209, ND–16,000 ng g^{-1}) (Sellström et al., 1998), UK (BDE-47, 0.15–368 ng g^{-1}; BDE-99, 0.30–898 ng g^{-1}; BDE-209, 34.0–1800 ng g^{-1}) (Allchin et al., 1999) and USA (BDE-209, 4.00–242 ng g^{-1}) (Song et al., 2004, 2005a, 2005b). However, concentrations of some PBDEs in sediments in China may be

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