



Review

Extent and mechanisms of brominated flame retardant emissions from waste soft furnishings and fabrics: A critical review



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ABSTRACT

Use of brominated flame retardants (BFRs) in soft furnishings has occurred for over thirty years with the phase out of polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) only relatively recently begun. As products treated with BFRs reach the end of their lifecycle they enter the waste stream, thereby constituting an important and increasing reservoir of these chemicals. This review highlights the dearth of data on the extent and potential mechanisms of BFR emissions from waste soft furnishings. However, insights into what may occur are provided by scrutiny of the larger (though still incomplete) database related to BFR emissions from electronic waste (e-waste). In many countries, municipal landfills have historically been the primary disposal method of waste consumer products and therefore represent a substantial reservoir of BFRs. Published data for BFR emissions to both air and water from landfill and other waste disposal routes are collated, presented and reviewed. Reported concentrations of PBDEs in landfill leachate range considerably from $<1 \text{ ng L}^{-1}$ to $133,000 \text{ ng } \Sigma\text{PBDE L}^{-1}$. In addition to direct migration of BFRs from waste materials; there is evidence that some higher brominated flame retardants are able to undergo degradation and debromination during waste treatment, that in some instances may lead to the formation of more toxic and bioavailable compounds. We propose that waste soft furnishings be treated with the same concern as e-waste, given its potential as a reservoir and source of environmental contamination with BFRs.

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1. Introduction

Brominated flame retardants (BFRs) are a group of synthetic chemical additives that have been applied to a wide range of consumer products such as: building materials, electronics and electrical goods, textiles and furnishings, to meet and comply with fire safety codes, standards

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and regulations. Currently there are more than 75 different commercially-recognised BFRs. Moreover, BFRs have historically been the most widely used organic flame retardants (FRs) due to their low cost and high efficacy (Covaci et al., 2008). The five classes of BFRs that have found most widespread use are tetrabromobisphenol-A (TBBP-A), hexabromocyclododecane (HBCD), and three technical mixtures of polybrominated diphenyl ethers (PBDEs): penta-BDE, octa-BDE, and deca-BDE (Leonards et al., 2008). In an environmental context, BFRs are highly resistant to degradation whether through chemical, biological, or photolytic processes and are therefore capable of long range transport, bioaccumulation in human and animal tissues and potential trophic magnification. As a result, widespread contamination of air, water, soil, sediment, and terrestrial and marine biota by BFRs has been widely documented (Harrad et al., 2009b; Harrad et al., 2010; Law, 2010; Law et al., 2006; Muir and de Wit, 2010).

These considerations are exacerbated by evidence of the toxicity of some BFRs. HBCD has been identified as an endocrine disrupting chemical (EDC) that induces enzymes and alters thyroid homeostasis with potential to cause adverse effects in humans at relatively low exposure levels (Darnerud, 2003, 2008; van der Ven et al., 2006, 2009; Yamada-Okabe et al., 2005). With respect to PBDEs, acute toxicity studies have suggested that they can be hepatotoxic at relatively high doses (Bruchajzer et al., 2011; Zhou et al., 2001). Moreover, several studies have indicated that penta- and octa-BDE mixtures, as well as several of the major individual congeners present in wildlife and people, can alter liver enzymes, disrupt thyroid homeostasis, and are associated with early onset of puberty, reduced fertility, and impaired neurological development (Viberg et al., 2007). This latter effect is illustrated by two cohort studies of in utero and child PBDE exposure in New York City and California, in which prenatal PBDE exposure (as indicated by cord blood PBDE levels) was associated with lower mental and physical development test scores for children aged 1, 4, and 6 years (New York City) and poorer attention, fine motor coordination, and cognition for children aged 5 and 7 years (California) (Eskenzazi et al., 2013; Herbstman et al., 2010). In general, the lower brominated congeners are more acutely toxic than the higher ones (Darnerud, 2003). Decabromodiphenyl ether (BDE-209) is relatively less persistent and bioaccumulative than the lower brominated congeners and has been shown to have very low acute toxicity when given by oral, inhalation and dermal routes (Kelly et al., 2008; Tomy et al., 2008; Wu et al., 2009). Moreover, BDE-209 does not appear to be genotoxic, is not teratogenic, and does not seem to cause developmental toxicity (Hardy et al., 2009; Wang et al., 2010). Nonetheless, work by Johansson et al. (2008) suggests that BDE-209 can be as potent as the lower brominated PBDEs in causing developmental neurotoxic defects. In contrast, TBBP-A is relatively less persistent and bioaccumulative than most other BFRs (Domínguez et al., 2011). Furthermore, it is not acutely toxic (WHO, 1995) and the toxicity and teratogenicity of TBBP-A appeared low in experimental in vivo studies (Darnerud, 2003). However, TBBP-A is a potential EDC, acting as a thyroid hormone and oestrogen agonist (Kitamura et al., 2002, 2005; Meerts et al., 2000), and additionally has been reported to be immunotoxic as a result of its in vitro inhibition of a key T-cell receptor (Mariussen and Fonnum, 2003).

Human exposure to BFRs occurs predominantly via indoor dust and food ingestion (mainly oily fish, meat, dairy products, and the neonatal sources of human milk and placental transfer), with a minor contribution from inhalation (Abdallah and Harrad, 2009; Covaci et al., 2009; Frederiksen et al., 2009). Concentrations of BFRs are higher in indoor air than outdoor air; this is likely due to the usage and slow release of BFRs from consumer products and building materials (Abdallah et al., 2008; Sjödin et al., 2001; Takigami et al., 2007; Toms et al., 2011).

These concerns about their adverse environmental impacts have meant that production of the penta- and octa-BDE commercial mixtures ceased in the EU and North America in 2004, and their use in all applications in 2006. Likewise, deca-BDE was restricted severely in the EU in 2008, with US manufacturers voluntarily committing to phase it out

from most uses in the USA by the end of 2012, and to end all uses by 31 December 2013 (BSEF, 2014). As a further reflection of concern about their use, the penta- and octa-BDE commercial mixtures (tetra-, penta-, hexa- and hepta-BDEs) and HBCD are listed as persistent organic pollutants (POPs) by the United Nations Environment Programme (UNEP) under the Stockholm Convention on Persistent Organic Pollutants, while deca-BDE is under active consideration for listing (UNEP, 2014).

1.1. BFR production and applications

In 2005, the total annual usage of BFRs was estimated to be about 311,000 tonnes, 130,000 tonnes of which was TBBP-A (Harju et al., 2008). HBCD annual production was estimated at up to 28,000 tonnes (9000 to 15,000 tonnes in China, 13,426 tonnes in Europe and the US) (UNEP, 2011). Total historic production of PBDEs (including deca-BDE) has been estimated to amount to 1.3 million to 1.5 million tonnes between 1970 and 2005 (UNEP, 2010a). In 1999, it was reported that North America represented 50.6% of total PBDE consumption; 97.5% of penta-BDE (8290 tonnes), 35.9% (1375 tonnes) of octa-BDE, and 44.3% (24,300 tonnes) of deca-BDE (Hale et al., 2003). In Asia, 53% (2030 tonnes) of octa-BDE and 42% (23,038 tonnes) of global deca-BDE was used, but no penta-BDE was produced in 1999 (Hale et al., 2003). Europe accounted for the remaining global demand of 2.5% (213 tonnes) penta-BDE, 12% (460 tonnes) octa-BDE and 13.7% (7515 tonnes) deca-BDE (Hale et al., 2003).

There are essentially two modes via which BFRs are incorporated into polymers. One is referred to as additive; in this instance the BFR is added to the molten polymer. This “additive” mode means that the BFR is not tightly bound to the polymer and therefore its migration to the environment is relatively facile. The alternative approach is where the BFR is covalently-bound to the polymer via reaction. Release to the environment of BFRs incorporated via this “reactive” process is comparatively restricted (Law, 2010). A caveat however, is that during the process of incorporation into the treated product, a proportion of the reactive BFR used may not polymerise, and this residual fraction has potential to be released to the environment (de Wit, 2002). TBBP-A is primarily used as a reactive FR in epoxy, polycarbonate and phenolic resins in printed circuit boards (58%), but has also been used as an additive BFR in the manufacture of acrylonitrile–butadiene–styrene (ABS) resins (18%), and was “possibly” used as an additive in high impact polystyrene (HIPS) (BSEF, 2009; EFRA, 2007). Concentrations of TBBP-A commonly found in these applications are between 10% and 20% by weight, depending on the polymer (Covaci et al., 2009). Moreover, during the 1990s, TBBP-A was increasingly used as a replacement for octa-BDE in ABS polymers (Watson et al., 2010).

The available data indicates that in general PBDEs (all congeners) were added at concentrations between ~3 and 30% by weight (WHO, 1994). The exact concentration applied was determined by several considerations such as the degree of flame retardancy required, the efficacy of the FR selected, whether it is applied in conjunction with a synergist (e.g. antimony oxide), physical attributes of the end product (e.g. colour, density and stability), and its specific application (European Chemicals Bureau, 2000).

In Europe approximately 95% of penta-BDE was used in flexible polyurethane foam (PUF), mainly used for furniture upholstery and automotive applications (European Chemicals Bureau, 2000). The UNEP initially reported treated PUF to usually contain 10–18% by weight of penta-BDE, however they have subsequently revised these estimates to be around 3–5% by weight for upholstery, cushions, mattresses, and carpet padding (UNEP, 2006; UNEP, 2010a). A similar concentration range of between 6 and 18% by weight of penta-BDE in PUF was provided in the *Plastics Additives Handbook* (Zweifel, 2001; cited in *European Commission*, 2011). The remaining minor uses of penta-BDE (totalling 5%) include: textiles, printed circuit boards, insulation foam, cable sheets, conveyor belts, lacquers,

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