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Review

Ecotoxicity and biodegradability of new brominated flame retardants: A review

M. Ezechiáš^{a,b}, S. Covino^a, T. Cajthaml^{a,b,*}^a Laboratory of Environmental Biotechnology, Institute of Microbiology ASCR, v.v.i., Vídeňská 1083, 142 20 Prague, Czech Republic^b Institute for Environmental Studies, Faculty of Science, Charles University in Prague, Albertov 6, 128 43 Prague, Czech Republic

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ABSTRACT

Brominated flame retardants (BFRs) have been routinely used as additives in a number of consumer products for several decades in order to reduce the risk of fire accidents. Concerns about the massive use of these substances have increased due to their possible toxicity, endocrine disrupting properties and occurrence in almost all the environmental compartments, including humans and wildlife organisms. Several conventional BFRs (e.g. polybrominated diphenylethers (PBDE)) have been included in the list of Persistent Organic Pollutants and their use has been restricted because of their established toxicity and environmental persistence. Over the past few years, these compounds have been replaced with “new” BFRs (NBFRs). Despite the fact that NBFRs are different chemical molecules than traditional BFRs, most of physical–chemical properties (e.g. aromatic moiety, halogen substitution, lipophilic character) are common to both groups; therefore, their fate in the environment is potentially similar to the banned BFRs. Therefore, this article has been compiled to summarize the published scientific data regarding the biodegradability of the most widely used NBFRs, a key factor in their potential persistency in the environment, and their ecotoxicological effects on humans and test organisms. The data reviewed here document that the mechanisms through NBFRs exhibit their ecotoxicity and the processes leading to their biotransformation in the environment are still poorly understood. Thus emphasis is placed on the need for further research in these areas is therefore emphasized, in order to avoid the massive use of further potentially harmful and recalcitrant substances of anthropogenic origin.

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* Corresponding author at: Laboratory of Environmental Biotechnology, Institute of Microbiology ASCR, v.v.i., Vídeňská 1083, 142 20 Prague, Czech Republic. Tel.: +420 241062498; fax: +420 241062384.

E-mail address: cajthaml@biomed.cas.cz (T. Cajthaml).

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1. Introduction

To date, a large number of brominated flame retardants (BFRs) have been produced and introduced into the market. These substances are incorporated as additives in materials used in common household items such as TVs, electronics, polyurethane foams, textiles etc. (de Wit, 2002) in order to reduce the likelihood of ignition of these materials and/or decrease their rate of combustion; however, BFRs can increase the toxicity of fires through the release of byproducts such as brominated and chlorinated dioxins and furans during combustion. The incorporation of organohalogens can increase levels of carbon monoxide, irritant gases, and soot, the main factors resulting in fire deaths and injuries in fires. Therefore the anticipated improvement in consumer safety is highly disputable (DiGangi et al., 2010; Shaw et al., 2010). Some BFRs are not chemically bound to a polymeric matrix, leading to the risk of diffusion into the environment.

Three main groups of BFRs, namely polybrominated diphenylethers (PBDE), hexabromocyclododecane (HBCD) and tetrabromobisphenol A (TBBPA) are considered “typical” or “conventional” flame retardants, since they were introduced into the market in the past. A number of scientific articles have dealt with the occurrence of these BFRs in the environment (Haneke, 2002a; Shaw and Kannan, 2009; Shaw et al., 2009; de Wit et al., 2010; Kefeni et al., 2011; Nyholm et al., 2013; Gorga et al., 2013), their biodegradation (Vrkoslavová et al., 2011; Eljarrat et al., 2011; Lal et al., 2010), and their accumulation in human and animal tissues (Hites, 2004; Inoue et al., 2006; Sjödin et al., 2003) and their toxicological properties (Birnbaum and Staskal, 2004; Viberg et al., 2004; Zhou et al., 2001) also including their non-specific aquatic baseline toxicity (Stieger et al., in press; Mayer and Reichenberg, 2006). The problem associated with their biodegradation in the environment is linked with the fact that they often content an aromatic moiety. Aromatic compounds are usually chemically resistant due to delocalization of energy, moreover, it has been suggested that the environmental persistence of conjugated compounds is due to the dense clouds of PI-electrons

on both the sides of the ring structures, which greatly reduces their susceptibility to nucleophilic attack (Johnsen et al., 2005). The resistance is even greater when the structures are substituted with so-called “xenophores” e.g., halogens (Alexander, 1994). The problem is even more pronounced in the presence of bromine atoms, which are somewhat larger than other substituents and limit the potential for microbial intracellular transformation processes (Cajthaml and Svobodová, 2012).

The described negative effects of these BFRs (see below) prompted the production and commercialization of alternative organobromines, which are commonly used at the present time. It is the intention of this paper to review recent findings on the ecotoxicity and environmental risk associated to these newer and alternative compounds with aromatic structure, as well as their biodegradability.

2. Legislative regulations on BFRs in the European Union

Since a number of BFRs have been found to be persistent in the environment, legislative steps have been taken to discontinue their production. The European Union (EU) performed risk assessment for penta- and octabrominated diphenyl ethers according to the REACH Regulation (Registration, Evaluation and Authorization of Chemicals; Regulation (EC) No. 1907/2006). It was subsequently agreed to prohibit their sale and use in the EU. Since 2004, Directive 2003/11/EC had prohibited trading and use of PentaBDE and OctaBDE in products containing more than 0.1% of these substances by weight. Waste products containing more than 0.25% PentaBDE were classified as hazardous waste. With reference to the Directive on Waste Electrical and Electronic Equipment (2002/96/EC – also known as WEEE) and the Directive restricting the use of certain hazardous substances in electrical and electronic equipment (2002/95/EC – known as RoHS), an agreement was reached to put an end to the use of BFRs in electrical and electronic equipment by 1 July 2006. Directive 2002/95/EC was followed

Table 1
Properties of NBFR reviewed in this paper. The abbreviation system was adopted from Bergman et al. (2012).

Name	Abbreviation	CAS	Solubility	Melting point (°C)	Log K_{ow}	Log K_{oc}
1,2-Bis(2,4,6-tribromophenoxy)ethane	BTBPE	37853-59-1	0.72 µg/L ^a	350 ^a	3.55 ^a	6.1 ^d
Decabromodiphenylethane	DBDPE	84852-53-9	2.1×10^{-7} g/L ^b	333–349 ^b	11.1 ^b	13.2 ^e
Bis(2-ethylhexyl) tetrabromophthalate	BEH-TEBP	26040-51-7	< 0.05 µg/L ^a	–27 ^a	10.2 ^a	7.4 ^d
Tetrabromophthalic anhydride	TEBP-Anh	632-79-1	241 mg/L ^a	298.8 ^a	1.98 ^a	3.43 ^g
2,3,4,5,6-Pentabromoethylbenzene	PBEB	85-22-3	3.5×10^{-4} g/L ^b	138 ^b	6.4 ^b	4.86 ^g
2,3,4,5,6-Pentabromotoluene	PBT	87-83-2	7.8×10^{-4} g/L ^b	280–289 ^b	5.87 ^b	4.57 ^g
Hexabromobenzene	HBB	87-82-1	1.1×10^{-7} g/L ^b	326–327 ^b	6.07 ^b	4.56 ^g
2-Allyloxy-1,3,5-tribromobenzene	TBP-AE	3278-89-5	0.02 g/L ^b	Not available	5.4 ^c	4.08 ^g
2,4,6-Tribromophenol	TBP	118-79-6	50 mg/L ^a	95.5 ^a	3.27 ^a	2.98 ^g
Decabromobiphenyl	BB209	13654-09-6	$1.24 \times 10^{-10-11}$ g/L ^b	380–386 ^b	12.63 ^f	
Tetrabromobisphenol A bis(dibromopropyl ether)	TBBPA-BDBPE	21850-44-2	0.144 µg/L ^a	113.4 ^a	10.3 ^e	12.9 ^e

^a Data from Registered substances database, European Chemicals Agency (ECHA).

^b Data from Covaci et al. (2011).

^c Data from Bergman et al. (2012).

^d Data from La Guardia et al. (2012).

^e Data from Nyholm et al. (2013).

^f Data from Doucette and Andren (1987).

^g Data from Harju et al. (2009).

^h Data from Hazardous Substances Data Bank (HSDB), U.S. National Library of Medicine.

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