International Journal of Hygiene and Environmental Health xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

International Journal of Hygiene and Environmental Health

journal homepage: www.elsevier.com/locate/ijheh

Review

Brominated flame retardants – Exposure and risk assessment for the general population

H. Fromme[a,d,*], G. Becher[b,c], B. Hilger[a], W. Völkel[a]

[a] Bavarian Health and Food Safety Authority, Department of Chemical Safety and Toxicology, D-80538 Munich, Germany
[b] Division of Environmental Medicine, Norwegian Institute of Public Health, Oslo, Norway
[c] Department of Chemistry, University of Oslo, P.O. Box 1033 Blindern, 0315 Oslo, Norway
[d] Institute and Outpatient Clinic for Occupational, Social and Environmental Medicine, Ludwig-Maximilians-University, Ziemssenstrasse 1, D-80336 Munich, Germany

ARTICLE INFO

Article history:
Received 24 April 2015
Received in revised form 10 August 2015
Accepted 12 August 2015

Keywords:
PBDE
HBCD
Flame retardants
Exposure
Biomonitoring
Indoor air
House dust

ABSTRACT

Brominated flame retardants (BFRs) are a large group of different substances used in numerous products to prevent fire hazards. Some of them are persistent in the environment, accumulate in the food chain and are of toxicological concern, while for others current data are limited. Meanwhile, BFRs have been found in many environmental media, foods, and biota including humans.

This review presents recent findings obtained from monitoring data in environmental media relevant for human exposure, as well as dietary exposure. In this context, concentrations in indoor and ambient air and in house dust are outlined. Furthermore, we summarize human biomonitoring data on BFR levels in blood and breast milk. Current estimates of the overall exposure of the general population using different relevant subsets are also addressed. All of these data are discussed in relation to currently available toxicological reference values used for risk assessment purposes.

Obviously, the exposure of the general population varies considerably in different parts of the world and even within countries. Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) show the highest intake during infancy. While the highest intake for BDE 47 for all groups was observed in the US, the total BDE 209 and HBCD intake was highest in the UK. For HBCD and all PBDEs except BDE 209, diet accounts for a large proportion of the total intake during infancy in all countries. With regard to toddlers and adults, the contribution of diet to total intake is high in Germany and the UK, while in the US, the high concentrations of PBDE in dust resulted in a notably smaller proportion of the intake being attributed to diet.

Contents

* Corresponding author at: Bavarian Health and Food Safety Authority, Department of Chemical Safety and Toxicology, D-80538 Munich, Germany.
E-mail address: hermann.fromme@lgl.bayern.de (H. Fromme).

http://dx.doi.org/10.1016/j.ijheh.2015.08.004
1438-4639/

1. Introduction

Fire hazards are serious health threats, especially in highly industrialized countries with a high population density. Different techniques have been used to reduce hazards, such as raising the ignition temperature of polymers, reducing the burning rate, prevent flame spreading or reducing smoke generation (Morose, 2006). Currently, four main groups of chemicals are used in large quantities: (a) Inorganic salts (e.g., antimony trioxide, aluminum hydroxide and borate), (b) phosphorus compounds (e.g., organophosphates, halophosphates, phosphine oxides and red phosphorus), (c) nitrogen-based compounds (e.g., melamine and melamine derivatives) and (d) halogenated organic compounds (e.g., chlorinated paraffins and alicyclic compounds, and brominated aromatic compounds). The focus of this paper is the large group of brominated flame retardants (BFRs). The main representatives of this group were chosen according to their environmental occurrence and the availability of exposure data. Specifically, the following substances were included: Polybrominated diphenyl ethers (PBDEs), which are a class of substances with 209 theoretical congeners depending on number of bromine atoms and the substitution pattern, 1,2,5,6,9,10-hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA), 1,2-bis-(2,4,6-tribromophenoxy)ethane (BTBPE), 2-ethyl-1-hexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), and bis(2-ethyl-1-hexyl)tetrabromophthalate (BE-TEBP). The chemical structures are given in Fig. 1 with abbreviations according to Bergman et al. (2012).

While PBDEs have been used as additive flame retardants in electronic devices such as computers and TV sets as well as in upholstery and carpets, HBCD is primarily used in polystyrene foams and upholstered textiles, and TBBPA is applied as a reactive flame retardant in epoxy resins and polycarbonate polyesters (NICNAS, 2007; De Wit et al., 2006; EFSA, 2011a,b). Only limited information is available on the current global market volume, but approximately 311,000 metric tons of BFRs were used worldwide in 2005, which is about 21% of the total consumption of flame retardants (Harju et al., 2008).

Since August 2006, the use and import of electric and electronic products containing more than 0.1% by weight of technical penta- and octaBDE in homogeneous materials has been banned in the European Union (EU, 2003). Following a decision made by the European Court of Justice, these restrictions have also been valid for decaBDE since July 2008 (ECJ, 2008). Moreover, HBCD was listed in annex XIV of REACH as a substance subject to authorization, and restrictions on penta- and octaBDE were provided in annex XVII. In the US, 10 states have banned penta- and octaBDE, and 3 have enacted limited bans on decaBDE so far. US companies phased out the production and use of penta- and octaBDE at the end of 2004 and had decided to voluntarily cease the production and use of decaBDE by the end of 2012 (US-EPA, 2015). In Canada, the manufacture of PBDEs (tetra- to decaBDE congeners) is prohibited, and additionally, the use, sale, offer for sale and import of some PBDEs (TetraBDE, PentaBDE and HexaBDE congeners) is also prohibited (EC, 2015). Finally under the Stockholm Convention, tetra- to heptaBDEs have been listed for elimination in 2009 and HBCD in 2013 and the proposed listing of decaBDE is presently discussed. Overall, these restrictions led to a shift in the market with an increased use of alternative BFRs to substitute for the discontinued PBDEs.

Numerous studies have revealed that PBDEs and HBCD are persistent, bioaccumulative and globally distributed in the environment and have been found in different environmental media and in biota (e.g., Law et al., 2008; Frederiksen et al., 2009; Daso et al., 2010; Covaci et al., 2011; Besis and Samara, 2012). For most of the other BFRs, only limited data on production and use, as well as their recent state of exposure, are available. Further, toxicological data are still limited and very often restricted to technical mixtures of PBDEs and HBCD (NICNAS, 2007; US-EPA, 2010; EC/HC, 2011; EFSA, 2011a,b; Kefeni et al., 2011). Overall, the main targets of PBDEs and HBCD are the liver and thyroid hormone homeostasis. In addition, the reproductive and nervous system with regard to effects on neurodevelopment have been identified as critical endpoints.

Meanwhile, different reviews are published summarizing data on environmental and dietary exposure, and toxicological properties of BFRs, mainly for PBDEs (Domingo, 2004, 2012; Domingo et al., 2008; Frederiksen et al., 2009. Toms et al., 2011; Kefeni et al., 2011; Besis and Samara, 2012; Kim et al., 2014; Colnot et al., 2014; Lyche et al., 2015; Linares et al., 2015).

The aim of this review is to summarize the current data available for those environmental media that contribute to human exposure to PBDE and non-PBDE brominated flame retardants (N_{PBDE}BFRs). For this purpose, we used the results of different Medline and Web of Science inquiries to obtain an overview of the current scientific literature. We also included papers presented at conferences, reports from governmental, scientific and other institutions, and, where possible, unpublished reports and other gray literature. In this context, BFR concentrations in indoor and outdoor air, house dust, and food and breast milk are summarized, as well as data on the body burden of humans. It should be noted that the comparison of such global data should be done with caution due to different methodologies employed in the investigations, such as for sampling, analytical measurements including lipid determinations and reporting, i.e., number of congeners considered for summing, treatment of values below LOQ (upper bound: non detects (ND) = LOQ; medium bound: ND = ½ LOQ; lower bound: ND = 0), reporting median or arithmetic and geometric means. Current estimates of