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Review article

Children's exposure to brominated flame retardants in indoor environments - A review

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ABSTRACT

The aim of this review is to present up-to-date research on children's exposure to brominated flame retardants (BFRs) in indoor environments. Large geographical variations were observed for all BFRs [polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD), tetrabromobisphenol A (TBBPA)], with the highest concentrations of PBDEs measured in North America (BDE-47) and Europe (BDE-209), where higher concentrations of PBDEs are present in dust from houses, daycare centers and primary schools. In Asia the highest PBDE concentrations were measured in China, near e-waste recycling areas. In the Middle East, Australia and Africa BFR levels were low in most indoor spaces. Asian countries also have the highest concentrations of TBBPA and HBCDD, followed by European countries. Fewer studies have been conducted measuring novel and emerging BFRs (NBFRs or EBFRs), of which decabromodiphenylethane (DBDPE) has the highest concentration in indoor environments, especially in China. The vast majority of children's exposure studies have been conducted in houses, sampling either dust or air, and considerably fewer in schools, daycare centers, cars and public facilities, despite BFR levels being comparable to (or sometimes even higher than) house dust. Relatively fewer studies focused on children's tissues such as serum, and only two studied exposure via mouthing toys. Alternative noninvasive sampling matrices that may act as surrogates for exposure to BFRs such as handwipes and silicone wristbands have recently started to gain momentum, because of the ease of sampling, faster collection time and better correlations to serum than house dust. Feces sampling is another promising alternative to children's serum that warrants further research. While many studies have associated different indoor environment characteristics, there is a knowledge gap on the association between children's behaviour and activity patterns and their exposure to BFRs, as well as data on infant exposure to BFRs via baby products. Results from the studies showed that dust ingestion was the dominant exposure pathway for most studied BFRs compared to indoor air inhalation and dermal contact, especially for infants and toddlers who have higher exposures than older children.

1. Introduction

Brominated flame retardants (BFRs) are chemicals applied to various materials in order to reduce their flammability. BFRs are used in a wide range of products such as clothes, children's toys, carpets, mattresses, housing and wiring of TV sets, computers and mobile phones, as well as in electrical kitchen appliances, upholstery and textiles, building materials and many plastic products (Frederiksen et al., 2009). The most commonly used BFRs are polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA). Following studies reporting widespread environmental occurrence and adverse effects both in vitro and in vivo, some of these BFRs were subsequently banned or voluntarily withdrawn (such as penta- and octa-BDE formulations) (Hoh et al., 2005), while the deca-BDE mixture has been banned in the European Union and parts of the United States (Kalantzi and Siskos, 2011). These bans led to a market shift towards the use of alternative BFRs as substitutes for the discontinued compounds. These substitute flame retardants have been referred to in different publications as “alternative”, “novel”, “emerging” or “non PBDE” flame retardants and have been reported in a

Abbreviations: BFRs, brominated flame retardants; FRs, flame retardants; PBBs, polybrominated biphenyls; PBDEs, polybrominated diphenyl ethers; HBCDD, hexabromocyclododecane; TBBPA, tetrabromobisphenol A; NBFRs, novel brominated flame retardants; EBFRs, emerging brominated flame retardants; DBDPE, decabromodiphenylethane; BTBPE, 1,2-bis(2,4,6-tribromophenoxy) ethane; TBBPA-BDBPE, tetrabromobisphenol A bis(2,3-dibromopropyl) ether; EH-TBB, 2-ethylhexyl-2,3,4,5-tetrabromobenzoate; BEH-TEBP, bis(2-ethylhexyl) tetrabromophthalate; HBB, hexabromobenzene; TBP-AE, 2,4,6-tribromophenyl allyl ether; PBT, pentabromotoluene; TBP-DBPE, 2,4,6-tribromophenyl 2,3-dibromopropyl ether; PBEB, pentabromoethylbenzene; TBCT, 2,3,4,5-tetrabromo-6-chloromethylbenzene; PBB-Acr, pentabromobenzyl acrylate; OBTMPI, octabromotrimethylphenyl indane; α- and β-DBE-DBCH, 4-(1,2-dibromoethyl)-1,2-dibromocyclohexane and 1-(1,2-dibromoethyl)-3,4-dibromocyclohexane; TBCO, 1,2,5,6-tetrabromocyclooctane

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variety of environmental media as well as in humans (Ali et al., 2011; Harju et al., 2008).

BFRs are lipophilic, persistent and bioaccumulative compounds with a tendency to accumulate in the fatty tissues of organisms. They are globally distributed in the environment, having been found in different environmental media and in biota such as air, water, soil, fish, birds and mammals, including humans (Kalantzi and Siskos, 2011). BFRs have been associated with thyroid disorders, diabetes, reproductive health problems, cancers and neurobehavioural and developmental disorders (Kim et al., 2014; Lyche et al., 2015). Studies have shown that neonatal exposure to PBDEs can cause neurobehavioural defects, including changes in locomotor activity, cognitive effects, spontaneous behaviour, and cholinergic susceptibility (Chao et al., 2014a). PBDE exposure may disrupt thyroid hormone functions, such as the binding with thyroid hormone receptors and transporter proteins (i.e., transthyretin), and interfere with thyroid homeostasis (Chao et al., 2014a).

The main routes of human exposure are diet, inhalation, dust ingestion and dermal absorption. House dust in particular is a significant medium for exposure to BFRs and is the subject of growing concern in recent years (Harrad et al., 2010). Studies have found high levels of BFRs in house dust from different regions of the world (Jiang et al., 2014; Harrad et al., 2008c; Wilford et al., 2005), as a result of BFR emissions from household products and absorption by house dust, which may represent the most important transfer pathway for HBCDD (Rauert et al., 2016). The indoor environment has been recognized as a significant source of BFRs, which may be released indoors from product use (Covaci et al., 2011). Their behaviour depends on different mass transfer phenomena occurring naturally or as a result of human activities, ultimately determining their fate indoors (Liagkouridis et al., 2014).

In children exposure to BFRs may occur through ingestion of dust, air inhalation, dermal absorption from indoor dust, toy chewing, hand-to-mouth behaviour and dietary intake. Children have a decreased metabolic capacity to detoxify and eliminate environmental contaminants, making them more susceptible to these contaminants (Landrigan et al., 2004). Studies over the last decade have highlighted many of these exposure pathways in children in indoor environments. In Europe, higher concentrations of PBDEs in dust are present in homes, daycare centers and primary school classrooms (Harrad et al., 2010, 2008a; Sjödin et al., 2008). In Asia the highest concentrations of PBDEs in dust have been measured in China, near e-waste recycling areas (Jiang et al., 2014). In the rest of the world BFR dust levels are low in most indoor environments (Ali et al., 2013; Hassan and Shoeib, 2015; Sjödin et al., 2008). With regards to TBBPA and HBCDD, Asia has the highest concentrations (Takigami et al., 2009), followed by Europe with the highest concentrations in dust from UK daycare centers and primary schools (Harrad et al., 2010). Relatively fewer studies have measured NBFRs, with decabromodiphenylethane (DBDPE) being the most abundant in indoor environments (Zheng et al., 2011, 2015). It is worth noting that most published reviews summarizing data on human exposure to BFRs are mainly concentrated on PBDEs, which are the most studied of all BFRs.

The present review focused on the following BFRs, based on their environmental occurrence and children's exposure data availability: PBDEs, HBCDD, TBBPA, DBDPE, 1,2-Bis(2,4,6-tribromophenoxy) ethane (BTBPE), tetrabromobisphenol A bis (2,3-dibromopropyl)ether (TBBPA-BDBPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP), and hexabromobenzene (HBB). All chemical abbreviations referred to in this study are as described in Bergman et al. (2012). The aim of this study is to compile and present data on children's exposure and levels of the aforementioned BFRs in indoor environments and identify research gaps.

2. Methodology

2.1. Inclusion and exclusion criteria

This review aims to incorporate all the studies that have been carried out and are available in the scientific literature regarding children's indoor exposure to BFRs. The following inclusion criteria were considered: 1) the studies must be published in a scientific journal, 2) they must refer to children's exposure to brominated flame retardants specifically and not occupational exposure (i.e., studies of children working at waste disposal or e-waste sites were not considered), 3) they must be in English, and 4) they must refer to children exposed through direct non-dietary contact with brominated flame retardants (as such, studies referring to breast milk and breastfeeding alone were not included). Studies that only sampled adults, but estimated daily intakes for children or toddlers based on dust measurements were included. Infant studies were only considered if the pathway of exposure was non-dietary. Studies reporting dust or indoor air BFR concentrations in environments other than schools, kindergartens and daycare centers were included only if they had intakes estimated for infants, toddlers or children. Studies reporting BFR serum and hair concentrations in children were not considered unless they included matching dust samples or were correlated with dust or house determinants.

2.2. Search strategy description

Literature searches were carried out in PubMed, Scopus, Web of Science and Science Direct covering studies published from 2002 to 2017 using the following keywords (and their combinations): a) regarding FRs: "BFRs" (or "brominated flame retardants"), "polybrominated diphenyl ethers" (or "PBDEs"), "HBCDD" (or HBCD), "TBBPA", "emerging brominated flame retardants" (or "EBFRs") and "NBFRs" (or "novel brominated flame retardants"), b) regarding microenvironments and children: "daycare", "classroom", "indoor environment", "school", "dust", "indoor air", "exposure", "children", "toddlers" and "infants". The last search was conducted on April 1st 2017.

Caution should be exercised when comparing data from different sources worldwide because of differences in the number of congeners used for the sum of PBDEs (which is why this study considered individual congeners and not just the sum of congeners), different children's age groups, analytical protocols, sampling, as well as treatment of concentrations below the limit of quantification (reported in our study as < dl, less than detection limit, where the concentrations were less than the method's detection limit). Temporal comparisons should be done with caution, as several BFRs (such as penta-, octa- and deca-BDE) were either banned or restricted during the sampling years of the included studies.

3. Polybrominated diphenyl ethers (PBDEs)

PBDEs are a class of chemicals with 209 congeners depending on the number of bromine atoms and the substitution pattern (Fromme et al., 2016). They have been used as additive FRs in electronics, uholstery and carpets and are produced at three degrees of bromination: penta- (penta-BDE), octa- (octa-BDE) and decabromodiphenyl ether (deca-BDE) (Kalantzi and Siskos, 2011). This study included the congeners that had the highest concentrations and the highest detection rates in samples from various indoor microenvironments in which children and toddlers spend the majority of their time (houses, schools, day care centers and cars): BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-183, and BDE-209. PBDE data are divided into two tables based on the type of matrix that has been studied: dust (the majority of studies) (Table 1) and all other matrices (indoor air, serum, handwipes, product surfaces, children's toys and baby products) (Table 2).