Chemosphere 189 (2017) 186–197

Contents lists available at ScienceDirect

Chemosphere

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

Predictors of human PBDE body burdens for a UK cohort

L. Bramwell [a, *], S. Harrad [b], M. Abou-Elwafa Abdallah [b], C. Rauert [b], M. Rose [c], A. Fernandes [c], T. Pless-Mulloli [a]

[a] Newcastle University, Institute of Health and Society, Baddiley-Clark Building, Richardson Road, Newcastle upon Tyne, UK
[b] University of Birmingham, School of Geography, Earth & Environmental Sciences, Birmingham, UK
[c] Fera Science, Sand Hutton, York, UK

HIGHLIGHTS

- We report intake and body burdens of tri-hepta BDEs and BDE-209 for 20 UK adults.
- Diet was the major source of tri-hepta BDEs, meat associated with higher exposure.
- Dust was the major source of BDE-209, more frequent dusting reduced exposure.
- Health concerns are indicated for infants with high PBDE intake from dust and diet.

ARTICLE INFO

Article history:
Received 19 June 2017
Received in revised form
11 August 2017
Accepted 12 August 2017
Available online 22 August 2017

Handling Editor: J. de Boer

Keywords:
PBDE
Serum
Breast-milk
Indoor dust
Diet
Exposure

ABSTRACT

Human exposure to polybrominated diphenyl ethers (PBDEs) was investigated in a cohort of 20 UK adults along with their anthropometric covariates and relevant properties such as room surveys, lifestyle, diet and activity details. Selected PBDE congeners were measured in matched samples of indoor dust ($n = 41$), vehicles ($n = 8$), duplicate diet ($n = 24$), serum ($n = 24$) and breast milk ($n = 6$).

Combined exposure estimates via dust and diet revealed total PBDE intakes of 104 to 1,440 pg kg^{-1} bw d^{-1} for $\Sigma BDEs_{3-7}$ and 1,170 to 17,000 pg kg^{-1} bw d^{-1} for BDE-209. These adult intakes are well within health reference doses suggested by the European Food Safety Authority (EFSA) and the US EPA. Diet was the primary source of intake of BDE_{3-7} congeners for the majority of the cohort, with dust the primary source of BDE-209. Primary sources of PBDE exposure vary between countries and regions with differing fire prevention regulations. Estimated infant exposures (ages 1.5–4.5 years) showed that BDE-99 intake for one of the households did not meet EFSA's recommended margin of exposure, a further two households had borderline PBDE exposures for high level dust and diet intake.

Males and those having a lower body fat mass had higher serum BDE-153. Higher meat consumption was significantly correlated with higher $BDEs_{3-7}$ in serum. A reduction in dietary $BDEs_{3-7}$ would therefore result in the greatest reduction in BDE-99 exposure. Rooms containing PUF sofas or armchairs over 20 years old had more $BDEs_{3-7}$ in their dust, and rooms with carpets or rugs of that age had higher dust BDE-209. Dusting rooms more frequently resulted in significantly lower concentrations of all major congeners in their dust. Correlation between BDE-209 body burden and dust or diet exposure was limited by its low bioaccessibility. Although vehicle dust contained the highest concentrations of $BDEs_{3-7}$ and BDE-209, serum $BDEs_{3-7}$ correlated most strongly with bedroom dust.

1. Introduction

UK residents are still exposed to a class of potentially harmful brominated flame retardants, polybrominated diphenyl ethers (PBDEs), even though European Union regulations restricting their manufacture, use and importation came into force in 2004 and 2008. Since the 1970s PBDEs have been incorporated into fabrics, foam cushioning and plastics used in everyday items such as vehicles, soft furnishings and electronics. PBDEs slow the rate of ignition and fire growth in petroleum based polymers and resins. PBDEs are not chemically bonded to these materials and are emitted into indoor dust and air through use and volatilisation

* Corresponding author.
E-mail address: lindsay.bramwell@ncl.ac.uk (L. Bramwell).

http://dx.doi.org/10.1016/j.chemosphere.2017.08.062
0045-6535/

(Rauert and Harrad, 2015; Sjödin et al., 2003). They can then move into the wider environment where they have been found in sewage sludge, soils and river and lake sediments (Allchin et al., 1999; De Boer et al., 2003; Eljarrat et al., 2008; Harrad et al., 2009). They are persistent organic pollutants as defined by the United Nations Environment Programme's Stockholm Convention and have an environmental half-life of several years. They can travel long distances in the atmosphere and are lipophilic, concentrating in animal and marine fats. These qualities and their wide usage have led them to permeate environments and food chains around the world (Fromme et al., 2016).

A systematic review of human health consequences of exposure to PBDEs concluded health effects may include thyroid disorders, reproductive health effects, and neurobehavioral and developmental disorders (Kim et al., 2014). Evidence of these effects has been seen in animal and *in vitro* research, where the mechanism appears to be altered hormone regulation (endocrine disruption) (Linares et al., 2015; Marchesini et al., 2008; Meerts et al., 2000; Viberg et al., 2006). Exposure during key developmental stages in infancy is most damaging as this is the time when altered hormone regulation will have the greatest impact. Recent estimates of the economic cost of just the intelligence quotient (IQ) points loss and intellectual disability due to PBDE exposure was $266 billion in the USA and $12·6 billion in the EU (Attina et al., 2016). These figures must be balanced against amounts saved due to fire prevention resulting from furnishing flammability standards e.g. £140 million annual savings in the UK estimated by prevention of death, injury and damage to property as a result of Furniture and Furnishings Fire Safety Regulations (1988) that require use of flame retardant chemicals. (BIS, 2009). PBDEs were only one group of flame retardant chemicals from the several BFR groups commonly used to meet such regulations.

In 2004, use of two commercial PBDE products, Penta-BDE and Octa-BDE, were restricted within the EU (European Council Directive 2003/11/EC) and voluntarily phased out in the USA. In 2009, they were added to the Stockholm Convention list of POPs for elimination. Penta-BDE had been primarily used in polyurethane foam (PUF) in soft furnishings, vehicles and printed circuit boards, in greatest amounts in the USA. Furnishings could contain one to four percent Penta-BDE to comply with fire safety regulations (Hammel et al., 2017). The Octa-BDE commercial product has been produced and used less widely than Penta-BDE. Its major use has been in acrylonitrile-butadiene-styrene (ABS) plastics, such as electronics and resin casings of office equipment. The Deca-BDE commercial product has been added to furnishing textiles, and in high impact polystyrene (HIPS) for cables, sockets, mobile phones, fridges and TV housings.

Concentrations of BDE-209 are higher in UK indoor dusts than in dusts from mainland Europe (Frederiksen et al., 2009; Harrad et al., 2008b) as a result of the UK's more stringent fire safety regulations (Furniture and Furnishings Fire Safety Regulations, 1988/1989, 1993 and 2010). Deca-BDE has been restricted from use in electrical and electronic equipment in the EU since 2008 and was added to Annex A of the Stockholm Convention list of POPs in 2017. Both diet and contact with indoor dust constitute important exposure pathways for PBDEs (Abdallah and Harrad, 2014). Foods from higher up the food chain, of animal origin, with a higher fat content (i.e. fish), meat and dairy have higher PBDE concentrations (EFSA, 2011). PBDEs will be circulating in our food chains for many years to come (Harrad and Diamond, 2006), and will be re-circulated back into homes as a result of plastics recycling (Samsonek and Puype, 2013).

Whether dust or diet is the primary exposure source for an individual depends on a number of factors; loading of PBDE in dust or food items and the amounts ingested, whether and when PBDE technical products have been phased out in that country and on the age of the individual (Bramwell et al., 2016). PBDE intake via ingestion and inhalation of dust is the major exposure route for young children in the USA that have frequent hand to mouth behaviours and spend lots of time on floors and carpets (Stapleton et al., 2012). Foetal exposure in the womb and transfer of PBDEs from mother to child during breastfeeding are key exposures for children during important developmental periods. For countries outside of the US and Canada, the largest contribution to tri-hepta BDE body burden is thought to be from diet, especially in regions where Penta-BDE use has been restricted for longer. Dust is likely to be most important contributor to exposure to higher brominated congeners in all regions (Sahlström et al., 2015).

The aim of this study was to determine the major dust and diet sources of PBDEs for a north east England cohort and to consider any potential health risks. The six specific objectives were: (a) to measure PBDE concentrations in dust from homes, work places and vehicles, (b) to calculate relative intake of PBDE via dust in the microenvironments, (c) to evaluate the relative importance of PBDE exposure via indoor dust versus dietary PBDE exposure, (d) to compare intake estimates with reference health values, (e) to investigate relationships between matched environmental and biomonitoring data, and (f) to determine the most effective means of reducing PBDE exposure for the cohort.

2. Materials and methods

We used a cross sectional and subjective sampling strategy to provide a snap shot of PBDE exposures and body burdens for individuals with expected high, average and low exposures. By comparing individuals with expected divergent exposures, we aimed to reveal the factors influencing body burdens.

2.1. Volunteer recruitment

We targeted individuals with a range of occupations and diets; such as workers in electronics, soft furnishings, transport, office workers, outdoor workers, oily fish eaters, omnivores and vegetarians. In 2010/11, following ethical approval for the study, volunteers over 18 years of age and with six months or more of domestic and occupational stability were recruited via local authorities, universities, businesses, hospitals, playgroups and breastfeeding groups. A short pre-screening questionnaire was used to identify volunteers that could provide the optimum range of exposures. 79 couples completed the pre-screening questionnaires, 10 couples were invited, and agreed, to participate in the full study week. Further description of the cohort is provided in the Supplementary Information. Volunteers gave written informed consent prior to participation.

2.2. Timing of sample collection

Participants undertook a ‘sampling week’ during which they completed an exposure and food frequency questionnaire (FFQ), food and activity diaries, room surveys including contents, usage and cleaning information and they were asked not to vacuum or dust their home. We adapted the validated WHO-IARC EPIC semi-quantitative dietary questionnaire for the study. On the seventh day of their sampling week, participants collected their duplicate diet samples (DD), and the researcher visited that evening to collect the DD samples, home and vehicle dust samples, questionnaires and surveys. The participants then fasted until their blood sample collection appointment the following morning where anthropometric measurements were also taken. Two couples repeated the