Predictors of human PBDE body burden for a UK cohort

Bramwell L[1], Harrad S[3], Rose M[2], Fernandes A[2], Abdallah M[3]F, Rauert C[3], Pless-Mulloli T[1]

[1]Newcastle University, Institute of Health and Society, Baddiley-Clark Building, Richardson Road, Newcastle upon Tyne, UK; [2]Fera Science Ltd, Sand Hutton, York, UK; [3] University of Birmingham, School of Geography, Earth & Environmental Sciences, Birmingham, UK

Introduction

PBDEs are a group of additive flame retardants that were widely used in the late 20th Century until their association with negative human health effects became apparent. Whether dust or diet is the primary exposure source for PBDEs differs between individuals and over time. Exposures may be related to occupational exposure, regional use and regulation of PBDE and diet type. In a recent systematic review of associations between human exposure to PBDEs via diet and indoor dust, and internal dose, we concluded three key factors influenced correlations between external PBDE exposure and human body burden: 1) half-life of individual congeners in the human body; 2) proximity and interaction between PBDE source and study subject; and 3) time of study relative to phase out of PBDE technical products[1].

Penta, Octa and Deca are the three commercial mixes of PBDEs. Production and use of Penta and Octa BDE were restricted from use in the EU in 2004[2]. Use of Deca BDE in electronics and electrical goods ceased in 2008[3]. PBDEs display a range of half-lives in humans with a general trend of shorter half-lives for the higher brominated compounds. Specifically, while estimates of human half-lives for Deca (BDE-209) are just a few days, for the main congeners of the Penta BDE mixture they are around two to four years[4,5]. The origins of human body burdens of PBDEs can be expected to change over time away from indoor dust towards diet as household goods containing PBDEs, such as soft furnishings and electronics, are replaced with items that contain alternative flame retardents[6]. The aim of this study was to determine the major sources of various dietary and indoor environment PBDE exposures for individuals in a UK cohort, with a view to making recommendations to reduce exposure.

Materials and methods

Participants were selected using a screening questionnaire to represent a range of anticipated low, medium and high PBDE exposures by considering diet type, occupation and urban and rural locations. The study was approved by the Durham and Tees Valley Research Ethics Committee.

In 2011, 10 co-habiting volunteer couples each completed a study week with food frequency and lifestyle questionnaires, food and activity diaries and room contents surveys to evaluate their PBDE exposure sources. Study participants also provided a 24 hour duplicate diet sample, samples of dust from homes (living areas and bedrooms), workplaces and vehicles, 60 mL blood sample, and 50+ mL breast milk samples where possible (see Figure 1). Serum, milk and duplicate diet samples were extracted and analysed for PBDEs at Fera, York, UK, using previously described methods[7]. Dust samples were analysed at the University of Birmingham, UK, again using previously described methods[8]. The following PBDE congeners were measured: BDE17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 183 and 209.

Results and discussion

We measured PBDEs from the Penta commercial mix in all of the samples collected with ranges of 0.78-12.8 ng/g lw in serum, 1.33-21.0 ng/g lw in breast milk, 0.1-1.4 ng/g lw in the duplicate diet samples, 2,230-3,760 ng/g dust in indoor dusts and 88.1-677 ng/g dust in vehicles. Deca-BDE was measured above the limit of detection in 17% of serum samples, 83% of breast milks, 63% of diet samples and 100% of dusts. Concentrations of the Deca-BDEs had ranges <1.13-19.8 ng/g lw in serum, <0.19-1.04 ng/g lw in breast milk, <0.001-3.13 ng/g lw in duplicate diets, 806-65,500 ng/g in indoor dusts and 315-137,000 in vehicles. Further descriptive statistics on PBDE congener concentrations in the matched matrices are provided in Table 1.

Individuals' estimated daily PBDE exposure via dust ranged from 14 to 1,000 pg/kg bw/d for Σtri–hepta BDEs, and 280-to 15,900 pg/kg bw/d of BDE-209 using an average adult dust intake scenario of 20 mg d^{-1}. Combined exposure estimates via dust and diet revealed total PBDE intakes of 104 to 1,440 pg/ kg bw/d for Σtri–hepta BDEs and 1,170 to 17,000 pg/kg bw/d for BDE-209. These adult intakes were well within health reference doses suggested by the European Food Safety Authority (EFSA) and the US EPA. However, estimated infant exposures (ages 1.5 to 4.5 years) indicated that BDE-99 intake (using average dust and diet intakes) for one of the households did not meet EFSAs recommended margin of exposure and another two households were borderline for BDE-99 for high level dust and diet intake.

Diet was found to be the major exposure source for Penta-mix BDEs and indoor dust the major source of BDE-209. Individuals' exposures showed great diversity within the cohort. Participants that ate above the group median of meat portions per week had higher serum Penta-mix BDE concentrations. Participants with BMI above the group median tended to have lower Penta-mix BDE concentrations.

Room contents that were indicated as key PBDE sources were: soft furnishings manufactured during the 1980s and 1990s and newer soft furnishings labelled as meeting the TB117 fire safety regulations from the USA. More frequent cleaning was associated with lower PBDEs in dust and body burden for participants where exposure was expected to be at the high end of the cohort.

Acknowledgements
This study was funded by the UK's Natural Environment Research Council (NERC): CASE studentship NE/F014139/1, the Institute of Health and Society, Newcastle University, UK, the Food and Environment Research Agency (Fera) and University of Birmingham, UK. Thanks also to study participants.

References:

1. Bramwell L, Glinianaia SV, Rankin J, Rose M, Fernandes A, Harrad S, Pless-Mulloli T. (2016) *Environ Int.* doi 10.1016/j.envint.2016.02.017
2. EU (2004) Amendment to Directive 76/769/EEC
3. EU (2008) Regulation of Hazardous Substances (RoHS)
4. Thuresson K, Hoglund P, Hagmar L, Sjodin A, Bergman A, Jakobsson K. (2006) *Environ. Health Perspect.* 114(2): 176-181.
5. Geyer HJG, Schramm K-W, Darnerud PO, Aune M, Feicht EA, Fried KW. (2004) *Organohalogen Compd*. 66: 3867–3872.
6. Harrad S and Diamond M. (2006) *Atmos. Environ.* 40(6): 1187-1188.
7. Fernandes A, White S, D'Silva K, Rose M, (2004) *Talanta* 63(5): 1147-1155
8. Harrad S, Ibarra C, Abdallah MA, Boon R, Neels H, Covaci A. (2008) *Environ. Int.* 34(8): 1170-1175

Table 1: PBDE Concentrations in different matrices

PBDEs	Matrix	Detection rate (%)	Median (MB)	Average (MB)	Range (MB)	P90 (MB)
BDE-47	Serum (n=24) (ng/g lw)	63	1.21	1.28	0.36-3.35	2.13
	Milk (n=6) (ng/g lw)	100	1.92	3.75	0.32-13.1	8.31
	24 hr duplicate diet (n=24) (ng/g lw)	100	0.10	0.18	0.04-0.86	0.31
	Indoor dusts (n=32) (ng/ g dust)	100	22.4	129	4.93--983	246
	Vehicles (n=8) (ng/ g dust)	100	23	38.5	15.8-105	74.1
BDE-99	Serum (n=24) (ng/g lw)	75	0.79	1.22	0.35 -5.61	2.54
	Milk (n=6) (ng/g lw)	100	0.88	1.18	0.12-3.74	2.39
	24 hr duplicate diet (n=24) (ng/g lw)	100	0.10	0.14	0.03-0.44	0.30
	Indoor dusts (n=32) (ng/ g dust)	100	36	230	7.63-2,000	270
	Vehicles (n=8) (ng/ g dust)	100	43.5	87.9	18.3-344	185
BDE-153	Serum (n=24) (ng/g lw)	100	0.37	0.67	0.12-4.05	1.03
	Milk (n=6) (ng/g lw)	100	1.01	1.1	0.70-1.68	1.53
	24 hr duplicate diet (n=24) (ng/g lw)	88	0.02	0.03	0.01-0.10	0.05
	Indoor dusts (n=32) (ng/ g dust)	100	18.5	36.7	3.52-170	69.5
	Vehicles (n=8) (ng/ g dust)	100	16.3	27.8	1.44-117	50.9
BDE-183	Serum (n=24) (ng/g lw)	67	0.05	0.09	0.02-0.33	0.18
	Milk (n=6) (ng/g lw)	100	0.05	0.07	0.02-0.23	0.15
	24 hr duplicate diet (n=24) (ng/g lw)	96	0.01	0.02	0.003-0.08	0.03
	Indoor dusts (n=32) (ng/ g dust)	100	7.7	9.16	1.98-22.4	17
	Vehicles (n=8) (ng/ g dust)	100	6.55	52.34	2.11-367	124
BDE-209	Serum (n=24) (ng/g lw)	17	1.73	2.81	<1.13-19.8	4.62
	Milk (n=6) (ng/g lw)	83	0.52	0.58	<0.19-1.04	1.02
	24 hr duplicate diet (n=24) (ng/g lw)	63	0.73	0.85	<0.001-3.13	1.52
	Indoor dusts (n=32) (ng/ g dust)	100	4,480	14,920	806-65,500	51,900
	Vehicles (n=8) (ng/ g dust)	100	19,000	41,000	315-137,000	120,000
∑tri-hepta BDE	Serum (n=24) (ng/g lw)	100	3.07	4.14	0.78-12.6	8.07
	Milk (n=6) (ng/g lw)	100	4.80	7.47	1.33-21.0	14.7
	24 hr duplicate diet (n=24) (ng/g lw)	100	0.30	0.50	0.10-1.40	1.06
	Indoor dusts (n=32) (ng/ g dust)	100	137	499	2,230-3,760	736
	Vehicles (n=8) (ng/ g dust)	100	179	251	88.1-677	504