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PBDEs and PBBs in human serum and breast milk from cohabiting UK couples

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HIGHLIGHTS

- UK body burdens of tri to deca PBDE and PBB are reported.
- Paired male and female serum, and female serum and breast milk were measured.
- These are the first UK serum data since EU restrictions on PBDEs.
- A small reduction in median $\sum PBDE_{3\text{-}7}$ UK serum levels are indicated.
- Estimated infant intakes are amongst the highest in the EU for BDEs-47 and -99.

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ABSTRACT

Concentrations of PBDEs and PBBs were measured in matched blood and breast milk samples from 10 UK couples collected in 2011–12. These data are the first measurements in human serum from the UK since the 2004 EU ban on all uses of the penta-and octa-BDE formulations and the 2008 ban on the use of the deca-BDE formulation in some applications. Serum ∑PBDE tri-hepta concentrations ranging from 1.0 to 16 ng g^{-1} lipid weight, with median 4.0 ng g^{-1} lw were measured. Breast milk ∑PBDE tri-hepta concentrations ranged from 1.3 to 21 ng g^{-1} lw, with median 5.7 ng g^{-1} lw. Couples had similar serum congener concentrations unless one of them frequently stayed away from home for work (different diet and dust exposures) or one had occupational exposure to foams and furnishings or electronics. BB-153 were measured above LOD in 40% of sera and 100% of breast milks samples, with median concentrations of 0.04 and 0.06, and maximums of 0.91 and 0.79 ng g^{-1} lw respectively. Concentrations in this study indicated a modest decrease from pre-ban levels reported for the UK. BDE-209 was detected above the limit of detection (LOD) in 15% of sera and 83% of breast milks, with ranges <1.2–20 and <0.2–1.0 ng g^{-1} lw respectively. Average daily infant intakes were estimated at 17, 5, 5 and 3 ng kg^{-1} bw for BDE-47,-99,-153 and -209 respectively, all well below relevant US-EPA threshold reference dose values (RfDs).

1. Introduction

Polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls (PBBs) are classes of flame retardant that have been used to meet fire safety regulations for fabrics, furnishings, electronics and vehicles since the 1970s. PBDEs are additive flame retardants that are mixed into plastics or foam, or sprayed onto fabrics, without forming chemical bonds with the material. During the use and lifetime of the product, PBDEs may migrate from the material (Sjödin et al., 2003). PBDEs are now ubiquitous in indoor air and dusts (Harrad et al., 2010) and transfer into the wider environment and food chains (Harrad and Diamond, 2006).

PBBs are another group of brominated flame retardants similar in structure, use, manufacture, contamination pathways and toxicological impact to PCBs. Production in the USA ceased following the Michigan Firemaster incident of 1973 where PBB was accidentally introduced into animal feed. Their use as flame retardants in textiles was banned in the EU where they have not been used or manufactured since 1996.

In 1999, Merionyté et al. noted a marked increase in concentrations of PBDEs in the breast milk of Swedish women with levels doubling approximately every four years (Meironyte et al., 1999).This triggered a global interest in human body burden and

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exposure to PBDEs and investigations into their persistence in humans and the environment. Inhalation and ingestion of indoor dust and food are potential pathways of exposure to PBDEs. Mother to child transfer of PBDEs occurs during breast feeding (Guvenius et al., 2003; Carrizo et al., 2007). Potential adverse human health effects of PBDE exposure and body burden are reproductive toxicity, neurotoxicity and immune effects (Darnerud et al., 2001; Meeker et al., 2009; Gascon et al., 2012; Eskenazi et al., 2013). Evidence of PBDE concentrations in indoor dusts and air in the UK has grown in recent years (Santillo et al., 2003; Harrad et al., 2004, 2006, 2008a,b,c; Harrad and Hunter, 2006; Sjödin et al., 2008a; Harrad and Abdallah, 2011) but few data are available regarding resultant UK human body burdens. It is widely accepted that PBDEs have long half-lives in humans but, even for the same congener, estimates of these values vary widely. There is a general trend of shorter half-lives for the more brominated compounds. Penta- and octa-BDE were banned from use in the EU in 2004 (EU, 2004) with use of deca-BDE in electronics and electrical goods also banned in 2008 (EU, 2008).

1.1. Aim

The aims of this study were to investigate (a) levels of PBDE and PBBs in the sera of co-habiting UK couples, and (b) paired sera and breast milk samples for nursing female partners. Measurements were compared with previous UK data collected prior to the implementation of EU restrictions on the manufacture and use of PBDEs (EU, 2004, 2008). Concentrations of PBDEs in human milk were used to derive estimated infant intakes of tri- to deca-PBDEs.

2. Materials and methods

Paired serum and breast milk samples were obtained from volunteer couples living in the north east of England as part of a wider in-depth study into potential human exposure sources and uptake of PBDE and emerging brominated contaminants. The wider study matches indoor dust and 24 h duplicate diet samples with these serum and breast milks as well as room surveys, diaries and exposure questionnaires, the results of which will be reported later. Sampling was undertaken between April 2011 and February 2012. Ten couples took part in the study and women from six of the couples provided breast milk samples.

2.1. Volunteer recruitment

We aimed to recruit individuals with a range of occupations and diets to reflect low, medium and high exposure to PBDEs, such as workers in electronics, soft furnishings or transport and outdoor workers, oily fish eaters and vegetarians. A short pre-screening questionnaire identified volunteers that would provide the optimum range of exposures. Recruitment was via local universities, local authorities, hospitals, playgroups and breast feeding groups. 79 couples completed the pre-screening questionnaires in 2011. 10 couples were invited, and agreed, to participate in the full study week. Of these 10 couples, two repeated the sampling week as a validation of the method. Inclusion in the study required the participants to be over 18 years of age and to have six months or more of domestic and occupational stability. Volunteers gave written informed consent prior to participation. Ethical approval for the study was provided by the NHS National Research Ethics Committee North East, Durham and Tees Valley, the Newcastle upon Tyne Hospitals NHS Foundation Trust, Newcastle University's Research Ethics Committee and the Food and Environment Research Agency's Ethics Committee.

2.2. Sample collection

Volunteers visited the Clinical Research Facility (CRF) at the Royal Victoria Infirmary at Newcastle to provide 60 mL fasted blood, sampled in the morning of the 8th day of the wider study. Physical measurements, such as body mass index (BMI), were also recorded at the CRF. Where a breast milk sample was provided, $\geqslant$50 mL milk was collected by the volunteer by either pump or manual expression up to 12 h before and 24 h after provision of the blood sample. Blood samples were collected in 6 $\times$ 10 mL red-top vacutainers, allowed to coagulate for 20 min, then centrifuged at 1000 rpm to separate the serum which was frozen to -18 °C and stored at the CRF laboratory until analysis. Breast milk samples were collected in Nalgene bottles and stored at -18 °C until transport with the sera samples to the Food and Environment Research Agency (Fera) Laboratories, York, UK for analysis. A set of unused sampling equipment was collected as field blanks in case sample results indicated a potential contamination source, however this was not required. Two couples repeated the sampling week, with sampling points 6.5 and 7.5 months apart. This provided a longitudinal element to the study.

2.3. Laboratory analyses

Details of the methods used for sample preparation, extraction, clean up and analysis of PBDEs and PBBs by high resolution GC MS are described elsewhere (Fernandes et al., 2004 and Fernandes et al., 2008). The performance characteristics of the methodology, including QA parameters such as limits of detection (LODs), precision, linear range of measurement, recoveries etc. have been reported earlier (Fernandes et al., 2008). Further confidence in the data is provided by regular and successful participation in inter-comparison schemes such as POPs in Food 2012 (Bruun Bremnes et al., 2012). The following congeners were measured: BDEs-17, -28, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -153, -154, -183 and -209 and PBBs -49, -52, -80, -101, -153 and -209. Lipid determination was carried out by West Yorkshire Analytical Services, Leeds, using the ISO17025 accredited Werner-Schmidt method; acid hydrolysis and solvent extraction.

2.4. Data analysis

Data were normalised to ng g^{-1} lipid for serum and breast milk to enable comparison between matrices and with previous studies. Descriptive statistics were calculated for both lower bound (LB) and upper bound (UB) data, where concentrations <LOD are treated as 0 and the LOD respectively.

2.5. Statistical analysis

Due to the small sample size statistical analyses were mostly descriptive. Statistical analyses were carried out using lower bound data, in keeping with previous UK serum data (Thomas et al., 2006). The analyses used IBM SPSS Statistics for Windows, Version 21.0. Armonk, NY: IBM Corp. Spearman's correlation coefficients were determined between individual and sum congeners and age, BMI, waist-hip ratio, total months breast feeding and parity. The Mann Whitney U test was used to determine any serum differences between males and females, urban and rural inhabitants and different diets.

2.6. Infant intake estimations

Nursing infants' intakes of PBDEs were estimated by multiplying age-appropriate estimated mean daily lipid intakes per kg bodyweight by the lipid weight concentrations of PBDE congeners

for each breast milk sample. The EFSA mean daily intake scenarios used were for a three months old infant (body weight 6.1 kg) with average intake of 800 mL and higher intake 1200 mL. Whole weight PBDE and PBB concentrations were used for the calculations.

2.7. Risk assessment

Estimated BDE-47, -99 and -153 exposures were compared with the corresponding threshold reference dose (RfD) suggested by the US-EPA to determine whether the PBDE infant intakes estimated for this study might be associated with any toxicological endpoints. RfDs are an estimate of oral daily human exposure to a chemical at no-observed-adverse, non-carcinogenic effects levels (NOAEL). For these PBDEs the endpoints considered by the US-EPA are neurodevelopmental toxicological effects. The daily exposures considered were BDE-47 (100 ng kg^{-1}) (US-EPA, 2006b), BDE-99 (100 ng kg^{-1}) (US-EPA, 2006a) and BDE-153 (200 ng kg^{-1}) (US-EPA, 2006c).

3. Results

Serum ∑PBDE tri-hepta concentrations ranged from 1.0 to 16 ng g^{-1} lipid weight (lw), with a median concentration of 4.0 ng g^{-1} lw. BDE-209 was detected above the LOD in 15% of samples, with a maximum of 19.8 ng g^{-1} lw. Breast milk ∑PBDE tri-hepta concentrations ranged from 1.3 to 21 ng g^{-1} lw, with a median concentration of 5.7 ng g^{-1} lw. BDE-209 was detected in 83% of breast milk samples with a maximum of 1.04 ng g^{-1} lw. BDE-47 was usually the most abundant congener present in sera in this study, making up 40% (median) of the ∑PBDE tri-hepta. This was followed by BDE-99 (20%), BDE-153 (9%), BDE-66 (6.5%) then BDE-100 (3.7%) and BDE-49 (2.9%). In samples where BDE-209 was measured above LOD (15% detection rate) this congener comprised 40% (median) of the ∑PBDE. Results for PBDE congener concentrations in individual serum samples from couples 1 to 10 are presented in Fig. 1. Repeat samples for couples 1 and 2 are also depicted. A summary of the serum data (not including repeat samples for couples 1 and 2) and breast milk data is presented in Table 1, including sums of all BDEs measured (∑PBDE), all BDEs measured excluding BDE-209 (∑PBDE tri-hepta), BDE-47, -99, -100, -153, -154 and -183 ($\sum PBDE_{(6)}$), BDEs-47, -99 and -153 ($\sum PBDE_{(3)}$). Individual concentrations are provided in supplementary data (Tables S-1 and S-2). BB-153 was the only PBB detected above the LOD in sera in this study (40% detection rate). BB-153 was detected in 100% of breast milk samples and BB-101 in one sample. PBB measurements are included in Table 1 and both supplementary data Tables S1 and S2.

A significant negative association was found between ∑PBDE in serum and age (Spearman's rho $r = -0.55$, $p = 0.01$), but the association with breast milk and age was not significant ($r = 0.09$, $p = 0.87$). The associations between ∑PBDE in serum and BMI ($r = -0.03$, $p = 0.89$) or body fat mass ($r = -0.07$, $p = 0.78$) were not found to be significant and neither were ∑PBDE in breast milk and BMI ($r = 0.09$, $p = 0.87$). For both congeners BDE-49 and BDE-66 in breast milk, the same significant positive association was noted with BMI ($r = 0.845$, $p = 0.03$). Significant negative associations were found for individual congeners in serum with waist-hip ratio (WHR); BDE-28 ($r = -0.46$, $p = 0.04$); BDE-153 ($r = -0.47$, $p = 0.04$). Parity was found to have a significant negative association with ∑PBDE in females' serum ($r = -0.67$, $p = 0.04$) and serum BDE-153 ($r = -0.69$, $p = 0.03$). Associations between ∑PBDE in breast milk and parity ($r = -0.29$, $p = 0.57$) or total months breastfeeding ($r = -0.26$, $p = 0.62$) were weaker. However, significant negative associations were found between total months breastfeeding and serum BDE-49 ($r = -0.67$, $p = 0.04$). Lipid normalised ∑PBDE data indicated that males had higher PBDE body burdens than their corresponding female partners (7/10 cases). The median concentration of ∑PBDE tri-hepta in males (4.6 ng g^{-1} lipid) was higher than that for females (3.5 ng g^{-1} lipid). However, the difference was only significant for BDE-153 ($p = 0.03$), with a weaker association with BDE-209 ($p = 0.7$). A weak association was found between sera ∑PBDE and having a home in an urban or rural environment ($p = 0.32$). For the two couples who provided two serum samples, differences between PBDE or PBB congener concentrations from the first and second samples were not generally significant, except for increases in BDE-28 ($p = 0.02$) and BDE-47 ($p = 0.04$).

4. Discussion

This 2011/12 study documents UK serum and breast milk data for PBDEs and PBBs. A modest decrease in UK serum PBDE concentrations since the EU bans was found. Compared to earlier studies, participants were recruited from as wide a pool of socio-economic class, occupation, diet and location as possible, with great focus on the detail of information collected for each individual. The small number of participants and the focus on breastfeeding mothers means, however, that results are not representative of all UK residents' exposures.

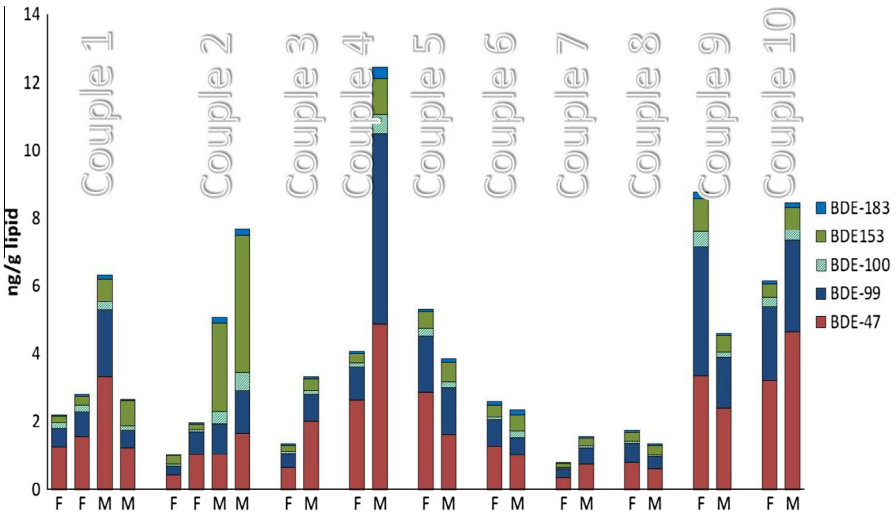

Fig. 1. Individual UK serum PBDE concentrations (ng g^{-1} lw) for congeners with detection rate ⩾50%: BDE-47, -99, -100, -153, -183.

Table 1
Summary of selected[a] PBDE and PBB body burden concentrations for all study participants (ng g^{-1} lw).

Congener	Serum (n = 20)					Breast milk (n = 6)				
	Median		Range		% > LOD	Median		Range		% > LOD
	LB[h]	UB[i]	Min	Max		LB[h]	UB[i]	Min	Max	
BDE-28	0.04	0.12	<0.03	0.55	55	0.09.	0.091.92	0.02	0.31	100
BDE-47	0.63	1.582	<0.36	4.87	60	1.92	1.92	0.32	13.09	100
BDE-49	0.06	0.12	<0.03	0.65	60	0.03	0.03	<0.02	<0.11	67
BDE-66	0.05	0.26	<0.03	0.79	55	0.03	0.03	<0.03	0.13	67
BDE-85	0.02	0.05	<0.02	0.26	50	0.04	0.04	<0.01	0.35	83
BDE-99	0.79	0.79	<0.26	5.61	70	0.88	0.88	0.12	3.74	100
BDE-100	0.15	0.15	<0.03	0.57	80	0.64	0.64	0.07	2.19	100
BDE-153	0.37	0.37	0.12	4.05	100	1.01	1.01	0.70	1.68	100
BDE-138	<LOD	0.07	0.03	0.19	5	0.02	0.02	<0.01	0.04	67
BDE-154	<LOD	0.06	<0.02	0.38	35	0.07	0.07	0.01	0.18	100
BDE-183	0.05	0.07	<0.03	0.33	60	0.05	0.05	0.02	0.23	100
BDE-209	<LOD	3.27	<1.24	19.80	15	0.52	0.54	<0.20	1.04	83
BB-153	<LOD	0.04	<0.01	0.91	40	0.08	0.08	0.06	0.79	100
$\sum$PBDEs[b]	2.41	8.32	2.25	35.41		5.59	5.67	1.28	22.02	
$\sum PBDE_{tri\text{-}hepta}$[c]	2.41	4.03	1.01	15.61		4.76	4.84	1.28	21.03	
$\sum PBDE_{(6)}$[d]	2.23	3.12	0.80	12.82		4.59	4.59	1.25	20.09	
$\sum PBDE_{(3)}$[e]	1.98	2.85	0.73	11.53		3.88	3.88	1.14	5.89	
$\sum PBB_{tri\text{-}hepta}$[f]	<LOD	0.37	<0.04	1.60		0.08	0.17	0.06	0.81	
$\sum$PBB[g]	<LOD	0.74	<0.09	2.27		0.08	0.22	0.06	0.86	
Sample fat %	0.43	0.43	0.25	0.93		2.61	2.61	0.97	4.56	

[a] Where % > LOD is over 15% for either serum or breast milk (BDE-17, -71, -77, -119, -126, and PBB-15, -49, -52, -80, -101, 209 measured but not reported here).
[b] Sum of all PBDE congeners measured.
[c] Sum of all PBDE congeners measured except BDE-209.
[d] Sum of BDE-47, -99, -100, -153, -154 and -183.
[e] Sum of BDE-47, -99 and -153.
[f] Sum of all PBB congeners measured except BB-209.
[g] Sum of all PBB congeners measured.
[h] Lower bound data.
[i] Upper bound data.

The finding of a significant negative association of $\sum$PBDE in serum and age was in line with some previous studies with greater age range of adult participants (Fromme et al., 2009; Garí and Grimalt, 2013; Shi et al., 2013). A non-significant negative association was found with PBDE in serum and BMI in keeping with Jain who found PBDE-153 and PBB-153 to be negatively associated, (Jain, 2013) and in contrast to Fitzgerald et al. who found a significant positive association (Fitzgerald et al., 2010). The present finding that serum BDE-153 was significantly higher for males was in contrast to the two earlier studies of cohabiting couples where gender differences were found not to be significant (Gomara et al., 2007; Fromme et al., 2009).

To the authors' knowledge, the only previous published UK PBDE study is provided by Thomas et al., from samples collected in 2003. This study had 154 participants from thirteen locations across the mainland, including some from the general area of this study. Thomas et al. suggested that their 154 volunteers were not representative of the general UK population as they had a higher proportion of women (68%) and vegetarians (12%) and that sampling on weekdays might select against people in work. They noted that the majority of mothers in their cohort breastfed, which was also found in this study. This study had only one vegetarian (5%). The median age was 37 years (range 26–43), Thomas et al.'s being 41 (range 22–80). The median BMI was 26 (range 22–33) for this study, Thomas et al.'s being 23 (18–43). Technological advances have reduced the LOD for analyses since 2003, most significantly for BDE-209. By considering ratios of data provided by Thomas et al. and the equivalents from this study, indicators of potential changes in UK body burdens were gauged. The data and ratios are presented in Table 2. All median congener concentrations for this study were lower than those reported by Thomas et al. except for BDE-99, where the measurements were very similar. It is likely that significant reductions would take time to manifest due to the long life of PBDE-containing items such as household and office soft furnishings and electronics. The maximum concentrations for individual congeners in 2003 were considerably higher (~50 times) than those in our study (2011/12). The absence of similar high maximum values in this study may indicate a downward shift

Table 2
Comparison of serum PBDE content between UK studies (ng g^{-1} lw).

	Thomas et al. (2006) 2003 data			This study, 2011 data			Ratio medians 2003/2011
	Median LB (ng g^{-1} lw)	Range (ng g^{-1} lw)	% Detects	Median LB (ng g^{-1} lw)	Range (ng g^{-1} lw)	% Detects	
BDE 28	<0.14	<0.14–10	27	0.04	<0.03–0.55	55	
BDE 47	0.82	<0.30–180	68	0.63	<0.36–4.87	60	1.26
BDE 99	0.76	<0.16–150	41	0.79	<0.26–5.61	70	0.96
BDE 100	<0.16	<0.17–390	92	0.15	<0.03–0.57	80	
BDE 153	1.7	<0.26–87	99	0.37	0.12–4.05	100	4.72
BDE 154	0.6	<0.15–4.4	86	<LOD	<0.12–0.38	35	5.45
BDE 183	0.3	<0.14–1.8	55	0.05	<0.03–0.33	60	2.73
BDE 209	<15	<15–240	7	<LOD	<1.24–19.8	15	
$\sum$PBDE	5.6	0.63–420	100	2.408	2.25–35.41	100	3.13
% Fat				0.426	0.25–0.93		

in the distribution of exposure within the UK population, but a more representative study is needed to fully evaluate this hypothesis.

Medians and ranges for BDE-47, -99, -100 and -153 in sera for this study are presented alongside examples from Europe, Asia and North America in Fig. 2. These examples are for studies that include data for men and women together, and where occupational exposure was not targeted. Human body burdens in North America are approximately one to two orders of magnitude higher than those found elsewhere in the developed world. This may be explained by the history of flame retardant regulations in the USA and California in particular (Shaw et al., 2010). The UK has more stringent flame retardant furnishings regulations than mainland Europe (EFRA, 2011) and the highest reported indoor dust BDE-209 concentrations in Europe (Harrad et al., 2008a). However, there is currently no evidence to suggest that penta- BDE use has been any higher in the UK than in mainland Europe. Median results for this study and previous UK figures indicate that BDE-209 and ∑BDEtri to hepta levels sit at the mid to lower end of European data (Thomas et al., 2006; Gomara et al., 2007; Antignac et al., 2009; Frederiksen et al., 2009; Fromme et al., 2009; Roosens et al., 2009). Maximum UK values reported by Thomas et al. are similar to those from the USA, indicating that some UK individuals have had higher PBDE exposure.

To the authors' knowledge, data presented here are the first UK PBB data for both human sera and breast milk. Previous studies with such PBB measurements have often been focused around the Michigan incident and its legacy. General population exposures were sought for comparison with the present results and are shown in Table 3. The median serum BB-153 concentration of 0.04 ng g^{-1} lw (range < 0.01–0.9) for this study, is almost two

Table 3
Median concentrations of BB-153 (ng g^{-1} lw) in human serum samples from different countries.

Location	Year	Number	BB-153	Reference	Note
UK	2011–12	20	0.04 (<0.01–0.9)	This study	(Range)
China	2009–10	21	0.024	Yang et al. (2013)	Geometric mean (gm)
China*	2009–10	35	0.52	Yang et al. (2013)	e waste dismantlers (gm)
Greenland	2002–4	99	1.2	Lenters et al. (2013)	gm
Poland and Ukraine	2002–4	200	<LOQ	Lenters et al. (2013)	
Northwest USA	2004	2062	3.3 (1.4–5.5)	Sjödin et al. (2008b)	NHANES

* Occupationally exposed cohort.

Table 4
Comparison of median (range) PBDE measurements in UK breast milks (ng g^{-1} lw).

Study	Number	Year	Sum PBDE tri-hepta	BDE-209
This study	6	2011–12	5.7 (1.3–21.0)	0.5 (0.2–1.0)
Abdallah and Harrad (2014)	28	2009	5.0 (0.2–26.1)[a]	0.3 (0.1–0.9)
D'Silva (2005)	10 Pooled groups	2000–2	6.4 (0.7–19.3)	
Kalantzi et al. (2004)	54	2001–3	6.6 (0.3–69.0)	

[a] tri-hexa

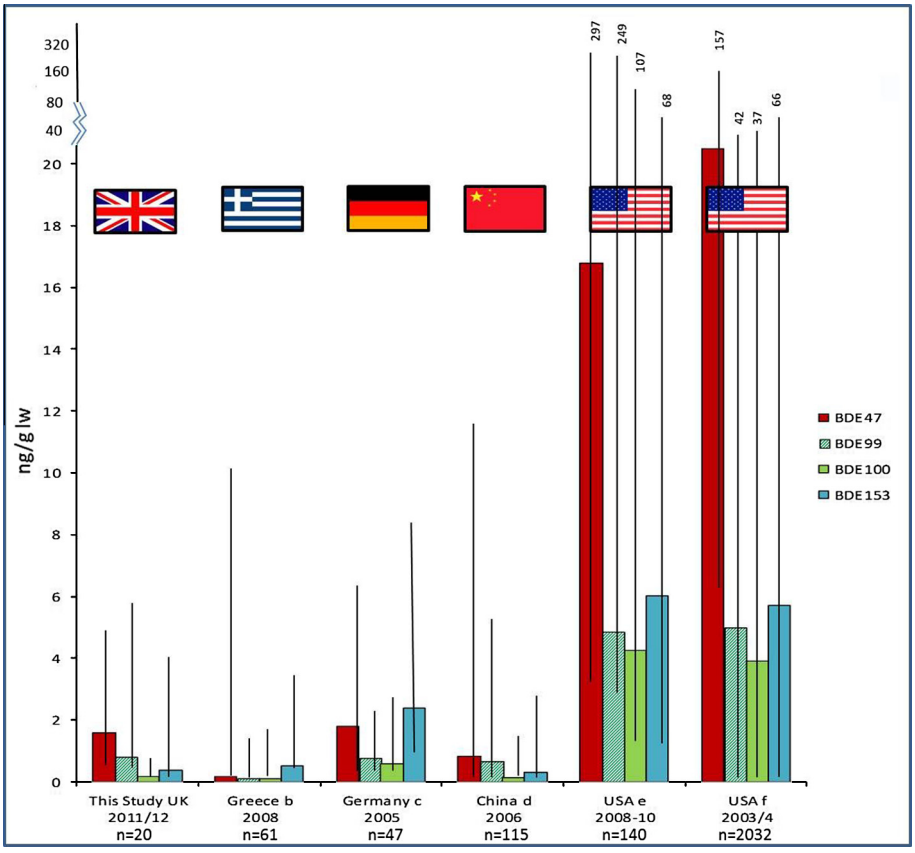

Fig. 2. Median levels and ranges (minimum–maximum) of major PBDE congener concentrations in serum for this and some previous studies of general populations. [b]Kalantzi et al. (2011), [c]Fromme et al. (2009), [d]Zhu et al. (2009), [e]Buttke et al. (2013), [f]Sjödin et al. (2008b).

Table 5
Comparison of mean daily infant intake estimations (ng kg^{-1} bw).

	Average daily consumption scenario 800 mL				High daily consumption scenario 1200 mL			
	BDE 47	BDE 99	BDE153	BDE 209	BDE 47	BDE 99	BDE153	BDE209
UK, This study (n = 6) 2012[a]	17	5	5	3	26	8	8	4
EU, EFSA (24 EU studies)[b]	0.6–14	<0.1–5	0.5–11	1.0–13	1.0–21	<0.1–8	0.7–17	1.0–20
China, Shi et al. (n = 103) 2013[c]				64	41	6	13	1109
NZ, Coakley et al. (n = 33) 2013[d]	11	3	3					
UK, Abdallah and Harrad (n = 28) 2009[e]	19.3	4.2	6.5	1.8				
UK, Kalantzi et al. (n = 54) 2003[b]	14	4	6					

[a] Intakes are calculated for the scenario of a three month old infant weighing 6.1 kg using whole weight breast milk data.
[b] EFSA (2011) Intakes are calculated for the scenario of a three month old infant weighing 6.1 kg and assuming breast milk to contain 3.5% lipid.
[c] Shi et al. (2013) estimated daily maximums and a mean of 64 for BDE-209.
[d] Coakley et al. (2013) estimated daily median exposures.
[e] Abdallah and Harrad (2014) estimated the mean exposure for a 1 month old infant weighing 4.14 kg and consuming 24.4 g lipid day^{-1}.

orders of magnitude below those found in the North American and Inuit studies.

PBDE concentrations in breast milk samples for this study were very similar to the two previous UK studies and current Birmingham UK study (Abdallah and Harrad, 2014). Data for comparison are presented in Table 4. A slight reduction in the median $\sum PBDE_{3\text{-}7}$ may be indicated by the two most recent studies. BDE-209 was measured in 28 breast milk samples collected from women in Birmingham, UK in 2009 with a median concentration of 0.25 and a range <0.06–0.92 ng g^{-1} lw (71% > LOQ) (Abdallah and Harrad, 2014). These are broadly consistent with those reported in this study, where the median concentration of BDE-209 was 0.54 ng g^{-1} lw (range < 0.20–1.04 ng g^{-1} lw).

Although a significant negative association was found with $\sum$PBDE in females' serum and parity, the association with breast milk was negative but not significant. Of the six women who provided breast milk samples for this study, three were primiparus and three were feeding their second child. Parity has been reported to have a decreasing association with $\sum$PBDE levels in breast milk (Kang et al., 2010). For all women in the study, total breastfeeding time prior to blood and breast milk sampling for this study ranged from 0 to 60 months, with a median of 10 months. Although a significant negative association was found between total months breastfeeding and BDEs-49 and -153 in females' serum, the negative associations between PBDEs or PBBs in breast milk and breast feeding were not found to be significant. Significant associations were found between BDE-49 and BDE-66 in breast milk and BMI in contrast with earlier studies that found no significant association between BMI and $\sum$PBDE in breast milk (Chao et al., 2010; Thomsen et al., 2010). The positive association found between age and BDE-153 in breast milk was not significant, although earlier studies have reported that the association is significant (Koh et al., 2010; Lignell et al., 2011) and increased PBDE levels in breast milk with age has been reported (Chao et al., 2010). The breast feeding mothers' median age in this study (35 years, range 27–39) was older than earlier studies by Koh et al. (30.5 years), Lignell et al. (mean 28.7, range 19–41), Chao et al. (30.1, range 22–42), and Kalantzi et al. (range 24–34). In a study of UK breast milk sampled in 2003, the participants were from Lancaster (a small city in rural north-west UK) and London (Kalantzi et al., 2004). Kalantzi et al.'s London participants were found to have higher PBDE body burdens suggesting higher concentrations in individuals residing in larger cities. This study also noted a positive association between breast milk $\sum$PBDE and urban living, although the difference was not found to be significant.

4.1. Infant intake via breast milk

The infant intake estimations in this study used the average (800 mL) and high (1200 mL) daily consumption scenarios used by the European Food Safety Authority (EFSA, 2011), and are presented in Table 5. Whole weight breast milk PBDE concentrations were used for calculations. EFSA intakes are calculated for the scenario of a three month old infant weighing 6.1 kg and assuming breast milk to contain 3.5% lipid. Lipid content in breast milk samples for this study ranged from 0.97% to 4.56% (median 2.61%). Estimated mean daily infant exposures of BDE-47,-99, -153 and -209 for earlier UK studies and some comparable data from the Europe, China and New Zealand are also presented in Table 4. The estimates in this study are very similar to the most recent UK estimates (Abdallah and Harrad, 2014). BDE-47 and -99 estimates are towards the top end of the EFSA EU data whilst BDE-153 and -209 are more central. The infant intake estimations for BDE-47, -99 and -153 in this study were compared with corresponding US-EPA RfDs. Maximum values for high infant consumption for this study were well within US-EPA guidelines. A NOAEL range of 18.8–41.4 ng kg^{-1} bw for BDE-99 has also been proposed (Bakker et al., 2008). The maximum estimate of daily exposure to BDE-99 (20 ng kg^{-1} bw) in this study is at the low end of this NOAEL. This limited assessment indicates that BDE-47, -99 and -153 in UK breast milk are unlikely to raise health concerns.

5. Conclusions

Evidence of current UK body burdens of PBDEs and PBBs is reported. Although the study is limited in size, it was found that the EU penta- and octa-BDE bans have yet to translate into substantial reductions in internal exposure of the UK population. Little or no reduction in breast milk levels since 2003 has been found.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.chemosphere.2014.03.060.

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