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Polybrominated diphenyl ethers and polychlorinated biphenyls in dust from cars, homes, and offices in Lagos, Nigeria

Stuart Harrad [a, *], Mohamed Abou-Elwafa Abdallah [a, b], Temilola Oluseyi [c]

[a] School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, UK
[b] Department of Analytical Chemistry, Faculty of Pharmacy, Assiut University, 71526 Assiut, Egypt
[c] Department of Chemistry, Faculty of Science, University of Lagos, Lagos, Nigeria

HIGHLIGHTS

- PBDEs and PCBs measured in Nigerian car, home & office dust.
- Penta-BDE levels in 2 cars amongst highest ever reported.
- Only second ever report of PCBs in cars.
- PCB 180 in Nigerian house dust at high end of global range.
- Levels of all target PCBs in Nigerian offices exceed those in cars.

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were measured in dust from 16 cars, 12 homes, and 18 offices in Lagos, Nigeria. These represent the first and second reports respectively of contamination of Nigerian indoor dust with these contaminants, and the second report on PCBs in car dust worldwide. Concentrations of BDE-47 and BDE-99 in two car dust samples (9300 and 3700 ng g^{-1} for BDE-47 and 4200 and 19,000 ng g^{-1} for BDE-99), are amongst the highest ever reported in car dust. ANOVA comparison with Canada, New Zealand, the UK, and the USA; reveals concentrations of BDEs-28, 49, 47, 66, 100, 99, 154, and 153 in Nigerian house dust, to be significantly lower than in Canada and the USA, with those of BDE-49 and 154 significantly lower than in New Zealand and the UK. Concentrations of BDE-209 in Nigeria were significantly lower than concentrations in the UK and the USA; while concentrations of PCB-180 were significantly greater than those in New Zealand, the UK, and the USA. Median concentrations of PCBs in cars were substantially higher than in the only previous study (in Kuwait and Pakistan). While median concentrations of PBDEs in cars generally exceeded those in homes, this was significant only for BDEs-49, 154, and 197, with concentrations in cars significantly greater than those in offices for BDEs-49 and 154. Contrastingly, concentrations of all target PCBs in offices exceeded significantly those in cars. This study underlines the truly global distribution of indoor contamination with PBDEs and PCBs.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are chemicals added to a wide range of consumer products (electrical and electronic equipment, textiles, polyurethane and polystyrene foams) to meet flame retardancy standards set by various jurisdictions worldwide (Alaee et al., 2003). Since these chemicals are used additively in most applications – i.e. they are not covalently bound to the products to which they are added – they can transfer from such products into the environment. An extensive body of evidence exists concerning the presence of PBDEs in indoor air (Allen et al., 2007; Harrad et al., 2004; Newton et al., 2015) and indoor dust (Harrad et al., 2008a, b; Jones-Otazo et al., 2005; Stapleton et al., 2005). Evidence of their persistence and capacity for bio-accumulation, coupled with concerns about their adverse health effects (Birnbaum and Staskal, 2004), have led to widespread bans and restrictions on the manufacture and use of both the Penta- and Octa-BDE mixtures and their listing under the Stockholm

* Corresponding author.
E-mail address: S.J.Harrad@bham.ac.uk (S. Harrad).

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Convention on Persistent Organic Pollutants (POPs) (UNEP, 2007). Moreover, manufacture and use of Deca-BDE has been progressively restricted and it is currently under consideration for listing under the Stockholm Convention (UNEP, 2013).

Human exposure to PBDEs occurs via the diet, and via inhalation of (primarily indoor) air, as well as ingestion of indoor dust (Harrad et al., 2004, 2006; Jones-Otazo et al., 2005; Lorber, 2008). The relative significance of each pathway varies considerably according to factors such as: geographical location (dust ingestion appears more important in North America than elsewhere (Harrad et al., 2008b)), age (dust ingestion is considered of greater magnitude for young children than adults (Jones-Otazo et al., 2005)), and the physicochemical properties of a given PBDE congener (exposure to decabromodiphenyl ether (BDE-209) is dominated by dust ingestion as a consequence of its very low vapour pressure and comparatively low capacity for bioaccumulation).

To date, the vast majority of exposure assessments conducted for PBDEs, have been conducted in East Asia (China, Korea, and Japan), Europe, and North America (Besis and Samara, 2012; Harrad et al., 2010). While data is emerging for other regions (including Egypt (Hassan and Shoeib, 2015), Kuwait (Ali et al., 2013; Gevao et al., 2006), and South Africa (Kefeni and Okonkwo, 2012; Abafe and Martincigh, 2015)), to our knowledge only two previous studies exist concerning the presence of PBDEs in indoor dust in Nigerian car and house dust respectively (Olukunle et al., 2015a, b).

Another class of POPs listed under the Stockholm Convention are polychlorinated biphenyls (PCBs). Despite almost universal cessation of their manufacture and new use in the late 1970s, their extensive use in applications such as plasticisers in building sealants and dielectric fluids in capacitors and transformers (Harrad et al., 1994), coupled with their persistence, means that they maintain a discernible environmental presence even today. Their substantial use in indoor applications is manifested by numerous reports of elevated concentrations of PCBs in indoor environments (Currado and Harrad, 1998; Harrad et al., 2006; Herrick et al., 2004; Kohler et al., 2005). Given their comparatively higher vapour pressures than PBDEs, far fewer data exist about concentrations of PCBs in indoor dust than for PBDEs. However, a previous study by our group suggests that although inhalation is the principal indoor exposure pathway under a typical dust ingestion scenario, exposure via dust ingestion exceeds that from either inhalation or diet for a small proportion of North American toddlers (Harrad et al., 2009). To our knowledge, this study constitutes the first measurements of PCBs in indoor dust in Nigeria. Moreover, PCBs in car dust have only been reported in one previous study conducted in Kuwait and Pakistan (Ali et al., 2013).

Against this background, this study seeks to corroborate the recently reported presence of PBDEs in indoor dust from various microenvironments in Makurdi, Benue State, Nigeria (Olukunle et al., 2015a, b), and to provide the first data on concentrations of PCBs in Nigerian indoor dust. We examine exposure in homes, offices and cars because they are oft-frequented environments. Moreover, we provide only the second such report worldwide on concentrations of PCBs in cars. We place our data for Nigerian indoor dust in an international context by comparing the levels found with those reported previously elsewhere. While the Nigerian economy is growing, it is not yet at the level of countries in North America and the EU for example, and thus our overarching hypothesis was that concentrations of PBDEs in Nigeria would be lower than those in more developed countries. However, it has been suggested that import of older electrical and electronic equipment may be an important source of BFRs like PBDEs in countries such as Nigeria (Nnorom and Osibanjo, 2008). As similar considerations may apply to PCBs, we tested our hypothesis by determining concentrations of PBDEs and PCBs in samples of settled dust from 16 cars, 12 homes, and 18 offices in Lagos, Nigeria.

2. Materials and methods

2.1. Sample collection

Dust samples were collected from 16 private cars, 18 offices, and the living areas of 12 houses in Lagos, Nigeria (the largest city in Nigeria) between September and October 2014. House and office dust samples were collected using a vacuum cleaner, according to a standardised method (Harrad et al., 2008b). Briefly, 1 m^2 of carpeted floor was vacuumed for 2 min, while for bare floors, 4 m^2 surface was vacuumed for 4 min. Dust was retained using 25 μm pore size nylon sample socks (Allied Filter Fabric Pty Ltd, Australia) mounted in the furniture attachment tube of the vacuum cleaner. In cars, dust was sampled from the dashboard, seats, and the floor in the passenger cabin, as well as in the boot. After sampling, socks were closed with a twist tie, sealed in a plastic bag and stored at −20 °C until transportation via courier to the University of Birmingham for sieving and analysis. Before sampling, the furniture attachment and the vacuum tubing were cleaned thoroughly using an isopropanol-impregnated disposable wipe. At the time of sample collection, information on potential influences on BFR contamination was recorded. In homes and offices, this comprised the number and type of putative sources like electronic devices, foam-filled furniture and floor material; while in cars, the vehicle manufacturer and age was recorded. Prior to analysis, all dust samples were passed through a pre-cleaned, n-hexane rinsed 500 μm mesh testing sieve (UKGE Limited, UK), covered with the lid and shaken for 2–4 min. Sieved samples were stored in clean, n-hexane rinsed glass jars and stored at 4 °C until analysis.

2.2. Sample extraction

Accurately weighted aliquots of dust (~0.15 g) were loaded into pre-cleaned 66 mL cells containing 1.5 g Florisil and Hydromatrix (Varian Inc., UK) to fill the void volume of the cells, and spiked with internal (surrogate) standards (15 ng of each of BDE 77, BDE 128 and 30 ng of $^{13}C_{12}$-BDE 209) prior to pressurised liquid extraction (ASE 350, Dionex, Hemel Hempstead, UK) using hexane:dichloromethane (1:9, *v/v*) at 90 °C and 1500 psi. The heating time was 5 min, static time 4 min, purge time 90 s, flush volume 50%, with three static cycles (Harrad and Abdallah, 2011).

2.3. Clean up

The crude extracts were concentrated to 0.5 mL using a Zymark Turbovap® II then purified by loading onto SPE cartridges filled with 8 g of pre-cleaned acidified silica (44% concentrated sulfuric acid, *w/w*). The analytes were eluted with 25 mL of hexane:dichloromethane (1:1, *v/v*). The eluate was evaporated to dryness under a gentle stream of nitrogen then reconstituted in 100 μL of isooctane containing 2.5 ng of $^{13}C_{12}$-BDE 100 used as recovery determination (syringe) standard for QA/QC purposes.

2.4. Instrumental analysis

Target PBDEs (BDEs 17, 28, 49, 66, 100, 99, 85, 154, 153, 183, 197, 203, 196, 208, 207, 206, and 209) were quantified using a TRACE™ 1310 Gas Chromatograph coupled to ISQ™ single quadrupole mass spectrometer (ThermoScientific, Austin, TX, USA) operated in negative chemical ionisation mode. Chromatographic resolution of PBDEs was achieved on a HP5-MS capillary column (15 m × 0.25 mm × 0.1 μm; Agilent, CA, USA) according to a previously reported method (Harrad et al., 2008b). PCB analysis was

conducted in accordance with our previous study of PCBs in indoor dust (Harrad et al., 2009) using an Agilent 5975C GC-MSD. In this study, our target PCBs were the ICES (International Council for the Exploration of the Seas) 6 indicator congeners 28, 52, 1010, 138, 153, and 180.

2.5. Quality assurance/Quality control

Recoveries of the internal standards used for PBDE analysis in individual samples ranged from 71 to 104%. Those for PCB analysis fell in the range 71–102%. Method blanks (n = 10) consisting of sodium sulfate were found to contain concentrations of target PBDEs and PCBs no greater than 5% of the concentrations found in the corresponding samples. Our data are thus not corrected for blank concentrations. For target compounds that were detected in the blanks. The limit of detection (LOD) was calculated as average blank level + 3*standard deviation of the blanks. For compounds that were not detected in the blanks. LOD was calculated as the sample level that produces a signal to noise ratio of 3:1.

2.6. Statistical analysis

Statistical analysis of our data was performed using IBM SPSS for Mac software (v. 22.0.0.0). For the purposes of statistical evaluation, all concentrations below the limit of detection (LOD) were assigned a value of f × LOD, where f is the fractional detection frequency of the contaminant in samples from that microenvironment in this study (Roosens et al, 2009). The distribution of our concentration data for PBDEs and PCBs were tested separately for each microenvironment using the Shapiro–Wilk test. This - combined with visual inspection of frequency plots – indicated that while our PCB data displayed a normal distribution, those for PBDEs were log-normally skewed ($P < 0.05$), therefore all PBDE data were log-transformed prior to comparison of means via ANOVA, as well as regression analysis. A P value < 0.05 was used as the level indicating statistical significance.

3. Results and discussion

3.1. Concentrations of PBDEs and PCBs in Nigerian house dust

Statistical summaries of concentrations of selected PBDEs and PCBs in samples analysed in this study are provided as Tables 1 and 2 respectively, with concentrations of all target contaminants in each individual sample supplied as supplementary material. Each target PCB was detected in every sample. By comparison, detection frequency varied between PBDE congeners. BDEs 47, 99, 100, 183, and 209 were detected in nearly all samples, with detection frequencies for other congeners ranging from 17% for BDE-17 in house and office dust to 88% for BDEs-197, 203, and 196 in car dust.

3.2. Do concentrations of PBDEs and PCBs vary between microenvironment categories?

Significant differences in concentrations of PBDEs in dust samples from different microenvironment categories have been reported (Harrad et al., 2008a, 2010). While similar differences have not been reported for PCBs, this may possibly be due to the far more limited database on these contaminants in indoor dust. We therefore hypothesised that significant differences would exist in concentrations of PBDEs and PCBs in cars, homes, and offices in this study. To test this hypothesis, we subjected our data to ANOVA. This analysis revealed the following significant differences between concentrations of target contaminants in different microenvironments. For BDEs-49 and -154, concentrations in car dust exceeded significantly those in both homes and offices; while for BDE-197, concentrations in cars were significantly greater than those in homes. In addition, concentrations of each individual target PCB in office dust were significantly higher than those in car dust; with concentrations of PCBs in car dust also exceeded significantly by those in house dust. No other significant differences were observed.

3.3. How do concentrations of PBDEs and PCBs in this study compare with previous studies?

Particularly noteworthy, are the highly elevated concentrations of Penta-BDE congeners in 2 car dust samples. At 9300 and 3700 ng g^{-1} for BDE-47 and 4200 and 19,000 ng g^{-1} for BDE-99, these concentrations approach the maximum values reported anywhere in car dust of 30,000 ng g^{-1} and 63,000 ng g^{-1} (Batterman et al., 2009). These two samples came from vehicles that were respectively 14 and 11 years old at the time of sampling. Both were Japanese marques that to our knowledge were manufactured in Japan. Further discussion of the relationship between PBDE concentrations and potential influencing factors follows in section 3.4.

Median concentrations detected in dust from cars, homes, and offices in this study are compared with those from selected other studies in Tables 3 and 4 for PBDEs and PCBs respectively. Median concentrations of PBDEs in both house and office dust in this study are lower than those reported in the only other studies conducted in Nigeria (Olukunle et al., 2015a, b). This difference may be due to small sample numbers in these studies, and perhaps also due to the different regions sampled (Makurdi, Benue State versus Lagos) but may also be explained by the different sampling method employed in the previous Nigerian studies (Olukunle et al., 2015a, b), in which the dust samples analysed comprised of a mix of floor dust and dust sampled from product surfaces. We have shown elsewhere that PBDE transfer from products to dust via direct product:dust contact is substantial (Rauert and Harrad, 2015), and thus dust sampled from product surfaces would likely contain higher concentrations than dust taken from the floor. Elsewhere in the African region, our concentrations of PBDEs in house and office dust are lower than those reported recently for South Africa (Abafe and Martincigh, 2015), but exceed those in earlier South African studies (Kefeni and Okwonkwo, 2012; Kefeni et al., 2014), and those reported elsewhere for Egypt and Iraq (Al-Omran and Harrad, in press; Hassan and Shoeib, 2015).

For PCBs, concentrations in this study are lower than those reported for South African homes and offices (Abafe and Martincigh, 2015), but exceed substantially those in Kuwaiti and Pakistani homes (Ali et al., 2013), and also those in Hong Kong offices (Kang et al., 2013). To the best of our knowledge, there is only one other study of PCBs in car dust, and concentrations of all target congeners in this study exceed substantially those reported for cars in Kuwait and Pakistan (Ali et al., 2013).

In addition, we used ANOVA to test whether concentrations of both PBDEs and PCBs in house dust in this study were significantly different to those reported previously by our research group using identical sampling and analytical procedures in house dust from Canada, New Zealand, the UK, and the USA (Harrad et al., 2008b, 2009). As New Zealand dust samples were only analysed for PCBs and Penta-BDE congeners, our comparison here for Octa- and Deca-BDE components does not include New Zealand.

This ANOVA comparison revealed concentrations of BDEs 28, 49, 47, 66, 100, 99, 154, and 153 to be significantly lower in Nigerian house dust than in dust from Canadian and USA homes. This is consistent with the well-documented greater use of the Penta-BDE product in North America. In addition, concentrations of BDEs-49 and 154 in New Zealand and UK dust exceeded significantly those

Table 1
Statistical Summary[a] of Concentrations (ng g^{-1}) of Selected PBDEs in Dust from Nigerian Cars, Homes, and Offices.

Compound	Microenvironment	Detection %	Minimum	Median	Average	Maximum
BDE-28	Cars	81	<0.04	1.1	7.0	65
	Homes	83	<0.04	0.48	0.75	3.1
	Offices	83	<0.04	1.1	4.7	65
BDE-47	Cars	88	<0.05	28	900	9300
	Homes	100	2.2	8.0	13	50
	Offices	100	2.8	14	20	100
BDE-49	Cars	81	<0.02	3.0	39	440
	Homes	33	<0.02	<0.02	0.46	2.6
	Offices	67	<0.03	1.0	1.1	4.2
BDE-100	Cars	94	<0.03	12	330	3100
	Homes	92	<0.03	4.0	8.4	44
	Offices	94	<0.03	4.2	7.3	24
BDE-99	Cars	88	<0.04	49	1700	19,000
	Homes	100	1.5	14	31	170
	Offices	94	<0.04	18	28	110
BDE -154	Cars	69	<0.04	3.6	450	3500
	Homes	42	<0.04	<0.04	1.7	7.9
	Offices	22	<0.04	<0.04	0.97	6.4
BDE − 153	Cars	69	<0.05	9.0	720	6000
	Homes	92	<0.05	3.9	5.3	22
	Offices	72	<0.05	3.7	5.4	24
BDE − 183	Cars	88	<0.04	8.8	83	480
	Homes	100	2.9	18	26	90
	Offices	100	4.3	26	66	350
BDE − 197	Cars	88	<0.06	7.7	19	97
	Homes	58	<0.06	1.8	2.1	8.4
	Offices	72	<0.06	4.5	8.4	39
BDE − 203	Cars	88	<0.06	4.7	12	76
	Homes	83	<0.06	1.6	1.9	7.9
	Offices	67	<0.06	2.8	3.8	13
BDE − 206	Cars	75	<0.08	61	300	3100
	Homes	58	<0.08	15	19	61
	Offices	72	<0.08	52	56	180
BDE − 208	Cars	63	<0.08	22	84	870
	Homes	42	<0.08	<0.08	6.1	23
	Offices	56	<0.08	9.2	15	88
BDE − 209	Cars	81	<0.11	780	10,000	52,000
	Homes	100	77	390	420	940
	Offices	89	<0.11	930	1200	4900

[a] For the purposes of statistical evaluation, all concentrations below the limit of detection (LOD) were assigned a value of f × LOD, where f is the fractional detection frequency of the contaminant in samples from that microenvironment.

Table 2
Statistical summary[a] of concentrations (ng g^{-1}) of PCBs detected in dust from Nigerian cars, homes, and offices.

Compound	Microenvironment	Minimum	Median	Average	Maximum
PCB-28	Cars	0.24	1.9	1.9	4.0
	Homes	1.5	3.8	3.9	7.8
	Offices	0.74	5.2	4.8	11
	Cars	0.08	2.1	2.3	5.9
PCB-52	Homes	0.80	4.2	4.5	8.6
	Offices	0.54	4.4	4.6	11
	Cars	0.14	0.72	1.3	5.5
PCB-101	Homes	0.37	2.7	3.7	9.4
	Offices	2.6	6.2	8.7	24
	Cars	0.08	1.6	2.5	9.1
PCB-153	Homes	0.22	5.8	7.3	22
	Offices	3.8	6.5	10	23
	Cars	0.40	1.4	1.9	6.7
PCB-138	Homes	0.31	6.0	7.5	22
	Offices	2.3	5.1	7.3	26
	Cars	0.46	1.9	2.3	5.7
PCB-180	Homes	4.4	10	10	24
	Offices	1.9	14	14	34

[a] For the purposes of statistical evaluation, all concentrations below the limit of detection (LOD) were assigned a value of f × LOD, where f is the fractional detection frequency of the contaminant in samples from that microenvironment.

in house dust from Nigeria in this study. While concentrations of the Octa-BDE marker congener BDE-183 in this study were statistically indistinguishable from those in the other countries examined; concentrations of the Deca-BDE indicator (BDE-209) were significantly lower in this study than in house dust from both the UK and the USA.

Similar comparison for PCBs, showed concentrations in this study to be statistically indistinguishable from those in Canadian, New Zealand, UK, and USA house dust, with the exception of PCB-180. For this congener, concentrations in this study exceeded significantly those in New Zealand, the UK, and the USA. As PCB-180 was most prevalent in the highly chlorinated commercial formulations such as Aroclor 1260 manufactured by Monsanto, this may indicate more extensive application of this product in Nigeria.

3.4. What influences concentrations of PBDEs in indoor dust samples?

We examined our data on concentrations of PBDEs and potential factors influencing these concentrations. For car dust, we noted no correlation between vehicle age (which ranged between 3 and 29 years, with an average of 11.6 years) and log-transformed concentration of any target PBDE. Moreover, there was no apparent relationship between PBDE concentrations and the car marque (5 Honda, 4 Toyota, 2 Nissan, and 1 each of Geely, Kia, Lexus, Mercedes, and Rover). For example, the highest BDE-99 concentration (19,000 ng g^{-1}) was observed in an 11 year old vehicle, while the

Table 3
Comparison of median concentrations (ng g^{-1}) of selected PBDEs detected in dust in this study with selected previous reports.

Sampling year-Country (Reference)	n	BDE 28	BDE 47	BDE 100	BDE 99	BDE 154	BDE 153	BDE 183	BDE 208	BDE 207	BDE 206	BDE 209
Cars												
2013 – Nigeria (this study)	16	1.1	28	12	49	3.6	9.0	8.8	22	42	61	780
2014 – Nigeria (Olukunle et al., 2015b)	12	n.a.	68	17	14	19	16	25	n.a.	n.a.	n.a.	122
2013 – Egypt (Hassan and Shoeib, 2015)	5	1.2	5.7	4.8	23	3.6	16	5.8	n.a.	n.a.	n.a.	1540
2009 – UK (Harrad and Abdallah, 2011)	14	n.a.	100	17	130	10	14	6	4100	3700	4800	190,000
2006/07 – USA (Batterman et al., 2009)	12	13	1800	790	2600	120	77	73	580	490	250	3100
Homes												
2013 – Nigeria (this study)	12	0.48	8.0	4.0	14	<0.042	3.9	18	<0.081	13	15	390
2012 – Nigeria (Olukunle et al., 2015a)	10	n.a.	47	43	46	45	50	30	n.a.	n.a.	n.a.	139
2012 – South Africa (Abafe and Martincigh, 2015)	10	n.a.	156	37	507	97	65	45	n.a.	n.a.	n.a.	1550
2010/11 - South Africa (Kefeni et al., 2014)	31	n.a.	2.6	<0.13	2.6	<LOD	<LOD	n.a.	n.a.	n.a.	n.a.	<1.8
2013 – Iraq (Al-Omran and Harrad, 2015)	18	<0.10	3.6	0.60	6.7	0.61	0.54	7.5	n.a.	n.a.	n.a.	610
2013 – Egypt (Hassan and Shoeib, 2015)	17	0.34	1.7	0.37	2.7	0.38	6.3	1.1	n.a.	n.a.	n.a.	40
2006 – UK (Harrad et al., 2008b)	28	0.53	13	4.2	23	3.3	5.2	13	n.a.	n.a.	n.a.	2800
2006 – USA (Harrad et al., 2008b)	20	14	410	160	820	89	110	16	n.a.	n.a.	n.a.	1300
Offices												
2013 – Nigeria (this study)	18	1.1	14	4.2	18	<0.042	3.7	26	9.2	24	52	930
2012 – Nigeria (Olukunle et al., 2015a)	11	n.a.	47	51	54	59	67	72	n.a.	n.a.	n.a.	140
2013 – Egypt (Hassan and Shoeib, 2015)	9	0.39	2.3	0.60	7.1	0.83	33	2.3	n.a.	n.a.	n.a.	366
2012 – South Africa (Abafe and Martincigh, 2015)	11	n.a.	119	<0.16	148	51	88	75	n.a.	n.a.	n.a.	324
2010 – South Africa (Kefeni and Okonkwo, 2012)	16	n.a.	44	n.a.	77	n.a	<0.5	n.a.	n.a.	n.a.	n.a.	<1.2
2006/07 – UK (Harrad et al., 2008a)	18	<0.5	23	3.2	65	5.1	8.7	8.3	n.a.	n.a.	n.a.	6200
2006/07 – USA (Batterman et al., 2010)	10	3.0	978	399	1760	78	48	30	n.a.	n.a.	n.a.	1.0

n.a. = not available.

Table 4
Comparison of median concentrations (ng g^{-1}) of selected PCBs detected in dust in this study with selected previous reports.

Sampling year – Country (reference)	n	PCB 28	PCB 52	PCB 101	PCB 153	PCB 138	PCB 180
Cars							
2013 – Nigeria (this study)	16	1.9	2.1	0.72	1.6	1.4	1.9
2011 – Kuwait (Ali et al., 2013)	15	n.a.	n.a.	<0.2	<0.2	<0.1	0.2
2011 – Pakistan (Ali et al., 2013)	15	n.a.	n.a.	<0.2	0.3	0.6	0.3
Homes							
2013 – Nigeria (this study)	12	3.8	4.1	2.7	5.8	6.0	10
2012 – South Africa (Abafe and Martincigh, 2015)	10	10.9	n.a.	n.a.	150	n.a.	585
2006 – Canada (Harrad et al., 2009)	10	7.3	7.2	8.8	9.9	9.5	6.8
2011 – Kuwait (Ali et al., 2013)	15	n.a.	n.a.	<0.2	<0.2	<0.1	0.4
2006 – New Zealand (Harrad et al., 2009)	20	2.3	1.4	1.6	1.4	1.8	1.3
2011 – Pakistan (Ali et al., 2013)	15	n.a.	n.a.	<0.2	<0.2	<0.1	0.3
2006 – UK (Harrad et al., 2009)	20	3.4	1.8	1.2	1.2	1.1	0.89
2006 – USA (Harrad et al., 2009)	20	5.1	6.2	8.7	7.1	6.5	2.6
2011 – USA (Dodson et al., 2012)	16	n.a.	n.a.	n.a.	9.5	n.a.	8.5
Offices							
2013 – Nigeria (this study)	18	5.2	4.4	6.2	6.5	5.1	14
2012 – South Africa (Abafe and Martincigh, 2015)	11	28	n.a.	n.a.	136	n.a	812
2009 – Hong Kong (Kang et al., 2013)	20	7.8	1.9	1.1	4.3	2.2	2.3

n.a. = not available.

same congener was not detected in a 13 year old car of the same marque. This observation is consistent with previous observations that the year of vehicle manufacture is not the only factor influencing concentrations of PBDEs in cars (Hazrati and Harrad, 2006).

We next conducted multiple linear regression analysis of our data for office and house dust using automatic linear modelling with log-transformed PBDE concentrations as the dependent variable and the numbers of: PUF-containing furniture, TVs, PCs, printers/copiers, microwaves, and fridge/freezers as independent variables. In homes, we also incorporated whether the home was air conditioned or naturally ventilated as an independent variable – all offices were air-conditioned. Office and house dust data were examined separately. Results of this automatic linear modelling showed no significant relationships, except that concentrations of Σtri-through hexa-BDEs (a proxy for the Penta-BDE formulation) were significantly ($p = 0.04$) positively correlated with the number of TVs in offices. While we have no information on the age of the TVs in the offices studied, this is not inconsistent with the reported past use of the Penta-BDE formulation in printed circuit boards (Betts, 2006).

3.5. Human exposure implications

Tables 5–7 give illustrative estimates of human exposure to (respectively) BDE-99, BDE-209, and ΣICES-6 PCBs that would arise from ingestion of the dust samples measured in this study. In common with the vast majority of other studies, we have conservatively assumed 100% absorption of intake and used average adult and toddler dust ingestion figures of 20 and 50 mg day^{-1}, and high dust ingestion figures for adults and toddlers of 50 and 200 mg day^{-1} (Jones-Otazo et al., 2005). We have then estimated exposure under various dust ingestion scenarios for homes, offices, and cars separately, assuming ingested dust is contaminated at the 5th percentile, median, and 95th percentile concentrations in our dust samples from each microenvironment category. Overall dust ingestion exposure estimates are then calculated taking into

Table 5
Overall exposure of Nigerian adults and toddlers (ng day^{-1}) to BDE-99 via ingestion of dust and percentages arising from ingestion of dust in cars, homes, and offices.

	Exposure in offices		Exposure in homes		Exposure in cars		Overall exposure		RfD[b]
	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	
Adult 5th percentile	0.01 (17.3)	0.03	0.06 (76)	0.15	0.01 (6.7)	0.01	0.08	0.20	7000
Adult median	0.08 (25.8)	0.21	0.20 (61.5)	0.50	0.04 (12.6)	0.10	0.33	0.82	7000
Adult 95th percentile	0.39 (4.5)	0.96	1.5 (17.2)	3.7	6.7 (78.3)	17	8.6	22	7000
Toddler 5th percentile	–	–	0.20	0.80	0.01 (6.2)	0.05	0.21	0.85	2000
Toddler median	–	–	0.67	2.67	0.10 (13.4)	0.41	0.77	3.1	2000
Toddler 95th percentile	–	–	4.9	20	17 (77.3)	67	22	87	2000

Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust ingestion rate scenarios.
[a] Assuming mean and high dust ingestion rates of 20 and 50 mg day^{-1} for adults and 50 and 200 mg day^{-1} for toddlers, that dust ingestion is pro rata to estimated time spent in each microenvironment category: for adults 72% homes, 23.8% offices, 4.2% cars; for toddlers 95.8% homes, 4.2% cars; and that concentration in dust is either 5th, 50th, or 95th percentile for that microenvironment.
[b] RfD is USEPA reference dose for BDE-99 (100 ng (kg body weight) day^{-1}), converted to ng day^{-1} assuming 70 kg and 20 kg body weight for adults and toddlers respectively.

Table 6
Overall exposure of Nigerian adults and toddlers (ng day^{-1}) to BDE-209 via ingestion of dust and percentages arising from ingestion of dust in cars, homes, and offices.

	Exposure in offices		Exposure in homes		Exposure in cars		Overall exposure		RfD[b]
	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	
Adult 5th percentile	<0.01 (0)	<0.01	1.29 (100)	3.2	<0.01 (0)	<0.01	1.3	3.2	490,000
Adult median	4.4 (41.4)	11	5.6 (52.5)	14	0.66 (6.2)	1.7	11	27	490,000
Adult 95th percentile	17 (23.5)	43	13 (17.9)	32	43 (58.6)	110	73	180	490,000
Toddler 5th percentile	–	–	4.3 (100)	17	<0.01 (0)	<0.01	4.3	17	140,000
Toddler median	–	–	19 (91.9)	75	1.7 (8.1)	6.6	20	81	140,000
Toddler 95th percentile	–	–	43 (9.2)	170	110 (71.2)	430	150	600	140,000

Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust ingestion rate scenarios.
[a] Assuming mean and high dust ingestion rates of 20 and 50 mg day^{-1} for adults and 50 and 200 mg day^{-1} for toddlers, that dust ingestion is pro rata to estimated time spent in each microenvironment category: for adults 72% homes, 23.8% offices, 4.2% cars; for toddlers 95.8% homes, 4.2% cars; and that concentration in dust is either 5th, 50th, or 95th percentile for that microenvironment.
[b] RfD is USEPA reference dose for BDE-209 (7000 ng (kg body weight) day^{-1}), converted to ng day^{-1} assuming 70 kg and 20 kg body weight for adults and toddlers respectively.

Table 7
Overall exposure of Nigerian adults and toddlers (ng day^{-1}) to ΣICES-6 PCBs via ingestion of dust and percentages arising from ingestion of dust in cars, homes, and offices.

	Exposure in offices		Exposure in homes		Exposure in cars		Overall exposure	
	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion[a]	High dust ingestion[a]	Mean dust ingestion	High dust ingestion
Adult 5th percentile	0.09 (25.6)	0.24	0.27 (73.7)	0.68	<0.01 (0.7)	0.01	0.37	0.92
Adult median	0.20 (28.4)	0.50	0.49 (70.2)	1.2	0.01 (1.4)	0.02	0.70	1.8
Adult 95th percentile	0.42 (31.6)	1.0	0.88 (66.7)	2.2	0.02 (1.8)	0.06	1.3	3.3
Toddler 5th percentile	–	–	0.90 (99.3)	3.6	0.01 (0.7)	0.02	0.91	3.6
Toddler median	–	–	1.6 (98.5)	6.5	0.02 (1.5)	0.10	1.7	6.6
Toddler 95th percentile	–	–	2.9 (98)	12	0.06 (2)	0.23	3.0	12

Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust ingestion rate scenarios.
[a] Assuming mean and high dust ingestion rates of 20 and 50 mg day^{-1} for adults and 50 and 200 mg day^{-1} for toddlers; that dust ingestion is pro rata to estimated time spent in each microenvironment category: for adults 72% homes, 23.8% offices, 4.2% cars; for toddlers 95.8% homes, 4.2% cars; and that concentration in dust is either 5th, 50th, or 95th percentile for that microenvironment.

account ingestion of dust in each of the relevant microenvironments. Dust ingestion is assumed to occur pro-rata to typical activity patterns (i.e. for adults: 72% home, 23.8% office, 4.2% car, for toddlers: 95.8% home and 4.2% car).

In summary, based on our data, Nigerian adult exposure via dust ingestion ranges between 0.08 and 22 ng day^{-1}, 1.3 and 180 ng day^{-1}, and 0.37 and 3.3 ng day^{-1} for BDE-99, BDE-209, and ΣICES-6 PCBs respectively. The equivalent exposure estimates for Nigerian toddlers are 0.21–87 ng day^{-1}, 4.3–600 ng day^{-1}, and 0.91 and 12 ng day^{-1}. While to our knowledge there are no health based

limit values (HBLVs) against which we can compare our estimate of exposure to ΣICES-6 PCBs, such values do exist for BDEs-99 and -209. For BDE-99, the relevant value is the USEPA's reference dose RfD of 100 ng kg body weight^{-1} day^{-1} (USEPA, 2008a), while for BDE-209 the USEPA RfD is 7000 ng kg body weight^{-1} day^{-1} (USEPA, 2008b). Assuming a child weight of 20 kg, our worst-case exposure estimate (high-end for toddlers) for BDEs-99 and -209 are 4.35 and 30 ng kg body weight^{-1} day^{-1} respectively. While we have not considered exposure via other pathways such as inhalation, diet, and dermal contact with dust and BFR-treated materials, there is a clearly substantial margin of safety between apparent exposure of the Nigerian population to these PBDEs and the USEPA RfD values. As a caveat to this reassuring message, we highlight the existence of a HBLV (albeit of no legislative standing) for BDE-99 proposed by researchers from the Netherlands of 0.23–0.30 ng kg body weight^{-1} day^{-1} for which impaired spermatogenesis is the end point of concern (Bakker et al., 2008). Our estimates of exposure of a 20 kg child arising from ingestion of dust contaminated with BDE-99 at the 95th percentile concentration at both the average (1.1 ng kg body weight^{-1} day^{-1}) and high ingestion rates (4.35 ng kg body weight^{-1} day^{-1}), both exceed this HBLV. Continued monitoring of human exposure to PBDEs would therefore appear warranted.

4. Conclusions

This study shows both PBDEs and PCBs to be ubiquitous in Nigerian cars, homes, and offices. While concentrations are in the main at the lower end of those reported globally, concentrations of PCB 180 in Nigerian homes are significantly higher than those recorded previously for New Zealand, the UK, and the USA. Moreover, concentrations of Penta-BDE congeners in 2 car dust samples are amongst the highest ever reported. Overall, this study underlines the truly global distribution of indoor contamination with PBDEs and PCBs, and the continuing need for action to reduce and eventually eliminate this potential public health hazard.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2015.12.045.

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