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

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1	POLYBROMINATED DIPHENYL ETHERS AND
2	POLYCHLORINATED BIPHENYLS IN DUST FROM
3	CARS, HOMES, AND OFFICES IN LAGOS, NIGERIA
4	
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#### 16 ABSTRACT

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were 17 18 measured in dust from 16 cars, 12 homes, and 18 offices in Lagos, Nigeria. These represent 19 the first and second reports respectively of contamination of Nigerian indoor dust with these 20 contaminants, and the second report on PCBs in car dust worldwide. Concentrations of BDE-47 and BDE-99 in two car dust samples (9,300 and 3,700 ng  $g^{-1}$  for BDE-47 and 4,200 and 21 19,000 ng g<sup>-1</sup> for BDE-99), are amongst the highest ever reported in car dust. ANOVA 22 comparison with Canada, New Zealand, the UK, and the USA; reveals concentrations of 23 24 BDEs-28, 49, 47, 66, 100, 99, 154, and 153 in Nigerian house dust, to be significantly lower 25 than in Canada and the USA, with those of BDE-49 and 154 significantly lower than in New 26 Zealand and the UK. Concentrations of BDE-209 in Nigeria were significantly lower than 27 concentrations in the UK and the USA; while concentrations of PCB-180 were significantly 28 greater than those in New Zealand, the UK, and the USA. Median concentrations of PCBs in 29 cars were substantially higher than in the only previous study (in Kuwait and Pakistan). 30 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 31 than those in offices for BDEs-49 and 154. Contrastingly, concentrations of all target PCBs 32 in offices exceeded significantly those in cars. This study underlines the truly global 33 distribution of indoor contamination with PBDEs and PCBs. 34

## 36 KEYWORDS

- 37 POPs
- 38 BFRs
- 39 PCBs
- 40 Africa
- 41 Nigeria
- 42 Indoor dust

### 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

#### 44 **1. INTRODUCTION**

45 Polybrominated diphenyl ethers (PBDEs) are chemicals added to a wide range of consumer 46 products (electrical and electronic equipment, textiles, polyurethane and polystyrene foams) 47 to meet flame retardancy standards set by various jurisdictions worldwide (Alaee et al, 2003). 48 Since these chemicals are used additively in most applications - i.e. they are not covalently 49 bound to the products to which they are added - they can transfer from such products into the 50 environment. An extensive body of evidence exists concerning the presence of PBDEs in 51 indoor air (Allen et al, 2007; Harrad et al, 2004; Newton et al, 2015) and indoor dust (Harrad 52 et al, 2008a,b; Jones-Otazo et al, 2005; Stapleton et al, 2005). Evidence of their persistence 53 and capacity for bioaccumulation, coupled with concerns about their adverse health effects 54 (Birnbaum and Staskal, 2004), have led to widespread bans and restrictions on the 55 manufacture and use of both the Penta- and Octa-BDE mixtures and their listing under the 56 Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP, 2007). Moreover, 57 manufacture and use of Deca-BDE has been progressively restricted and it is currently under 58 consideration for listing under the Stockholm Convention (UNEP, 2013).

59

60 Human exposure to PBDEs occurs via the diet, and via inhalation of (primarily indoor) air, as 61 well as ingestion of indoor dust (Harrad et al, 2004; 2006; Jones-Otazo et al, 2005; Lorber, 62 2008). The relative significance of each pathway varies considerably according to factors 63 such as: geographical location (dust ingestion appears more important in North America than 64 elsewhere (Harrad et al, 2008b)), age (dust ingestion is considered of greater magnitude for 65 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 66 67 dust ingestion as a consequence of its very low vapour pressure and comparatively low 68 capacity for bioaccumulation).

69

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 Okwonkwo, 2012; Obafe 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).

78 Another class of POPs listed under the Stockholm Convention are polychlorinated biphenyls 79 (PCBs). Despite almost universal cessation of their manufacture and new use in the late 80 1970s, their extensive use in applications such as plasticisers in building sealants and 81 dielectric fluids in capacitors and transformers (Harrad et al, 1994), coupled with their 82 persistence, means that they maintain a discernible environmental presence even today. Their 83 substantial use in indoor applications is manifested by numerous reports of elevated 84 concentrations of PCBs in indoor environments (Currado and Harrad, 1998; Harrad et al, 85 2006; Herrick et al, 2004; Kohler et al, 2005). Given their comparatively higher vapour 86 pressures than PBDEs, far fewer data exist about concentrations of PCBs in indoor dust than 87 for PBDEs. However, a previous study by our group suggests that although inhalation is the 88 principal indoor exposure pathway under a typical dust ingestion scenario, exposure via dust 89 ingestion exceeds that from either inhalation or diet for a small proportion of North American 90 toddlers (Harrad et al, 2009). To our knowledge, this study constitutes the first measurements 91 of PCBs in indoor dust in Nigeria. Moreover, PCBs in car dust have only been reported in 92 one previous study conducted in Kuwait and Pakistan (Ali et al, 2013).

93

94 Against this background, this study seeks to corroborate the recently reported presence of 95 PBDEs in indoor dust from various microenvironments in Makurdi, Benue State, Nigeria 96 (Olukunle et al, 2015a,b), and to provide the first data on concentrations of PCBs in Nigerian 97 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 98 99 concentrations of PCBs in cars. We place our data for Nigerian indoor dust in an international 100 context by comparing the levels found with those reported previously elsewhere. While the 101 Nigerian economy is growing, it is not yet at the level of countries in North America and the 102 EU for example, and thus our overarching hypothesis was that concentrations of PBDEs in 103 Nigeria would be lower than those in more developed countries. However, it has been 104 suggested that import of older electrical and electronic equipment may be an important 105 source of BFRs like PBDEs in countries such as Nigeria (Nnorom and Osibanjo, 2008). As 106 similar considerations may apply to PCBs, we tested our hypothesis by determining 107 concentrations of PBDEs and PCBs in samples of settled dust from 16 cars, 12 homes, and 18 108 offices in Lagos, Nigeria.

109

#### 110 2. MATERIALS AND METHODS

111 **2.1 Sample collection** 

112 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. 113 114 House and office dust samples were collected using a vacuum cleaner, according to a standardised method (Harrad et al, 2008b). Briefly, 1 m<sup>2</sup> of carpeted floor was vacuumed for 115 2 min, while for bare floors, 4  $m^2$  surface was vacuumed for 4 min. Dust was retained using 116 25 µm pore size nylon sample socks (Allied Filter Fabric Pty Ltd, Australia) mounted in the 117 118 furniture attachment tube of the vacuum cleaner. In cars, dust was sampled from the 119 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 120 transportation via courier to the University of Birmingham for sieving and analysis. Before 121 122 sampling, the furniture attachment and the vacuum tubing were cleaned thoroughly using an 123 isopropanol-impregnated disposable wipe. At the time of sample collection, information on 124 potential influences on BFR contamination was recorded. In homes and offices, this 125 comprised the number and type of putative sources like electronic devices, foam-filled 126 furniture and floor material; while in cars, the vehicle manufacturer and age was recorded. 127 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. 128 Sieved samples were stored in clean, n-hexane rinsed glass jars and stored at 4 °C until 129 130 analysis.

131

#### 132 **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 minutes, static time 4 min, purge time 90 s, flush volume 50 %, with three static cycles (Harrad and Abdallah, 2011).

140

#### 141 **2.3 Clean up**

142 The crude extracts were concentrated to 0.5 mL using a Zymark Turbovap® II then purified 143 by loading onto SPE cartridges filled with 8 g of pre-cleaned acidified silica (44% 144 concentrated sulfuric acid, w/w). The analytes were eluted with 25 mL of 145 hexane:dichloromethane (1:1, v/v). The eluate was evaporated to dryness under a gentle

- 146 stream of nitrogen then reconstituted in 100  $\mu$ L of isooctane containing 2.5 ng of <sup>13</sup>C<sub>12</sub>-BDE
- 147 100 used as recovery determination (syringe) standard for QA/QC purposes.
- 148

#### 149 2.4 Instrumental analysis

150 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<sup>TM</sup> 1310 Gas Chromatograph coupled to ISQ<sup>TM</sup> 151 152 single quadrupole mass spectrometer (ThermoScientific, Austin, TX, USA) operated in 153 negative chemical ionisation mode. Chromatographic resolution of PBDEs was achieved on a 154 HP5-MS capillary column (15 m x 0.25 mm x 0.1 µm; Agilent, CA, USA) according to a 155 previously reported method (Harrad et al., 2008b). PCB analysis was conducted in 156 accordance with our previous study of PCBs in indoor dust (Harrad et al, 2009) using an 157 Agilent 5975C GC-MSD. In this study, our target PCBs were the ICES (International Council 158 for the Exploration of the Seas) 6 indicator congeners 28, 52, 1010, 138, 153, and 180.

159

#### 160 2.5 Quality Assurance/Quality Control

161 Recoveries of the internal standards used for PBDE analysis in individual samples ranged 162 from 71 to 104 %. Those for PCB analysis fell in the range 71 to 102 %. Method blanks (n =163 10) consisting of sodium sulfate were found to contain concentrations of target PBDEs and 164 PCBs no greater than 5 % of the concentrations found in the corresponding samples. Our data 165 are thus not corrected for blank concentrations. For target compounds that were detected in 166 the blanks. The limit of detection (LOD) was calculated as average blank level + 3\*standard 167 deviation of the blanks. For compounds that were not detected in the blanks. LOD was 168 calculated as the sample level that produces a signal to noise ratio of 3:1.

169

#### 170 **2.6 Statistical analysis**

171 Statistical analysis of our data was performed using IBM SPSS for Mac software (v. 172 22.0.0.0). For the purposes of statistical evaluation, all concentrations below the limit of detection (LOD) were assigned a value of f x LOD, where f is the fractional detection 173 174 frequency of the contaminant in samples from that microenvironment in this study (Roosens 175 et al, 2009). The distribution of our concentration data for PBDEs and PCBs were tested 176 separately for each microenvironment using the Shapiro-Wilk test. This - combined with 177 visual inspection of frequency plots – indicated that while our PCB data displayed a normal 178 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.</li>
- 181

#### 182 **3. RESULTS AND DISCUSSION**

#### 183 **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.

191

#### 192 **3.2** Do concentrations of PBDEs and PCBs vary between microenvironment categories?

193 Significant differences in concentrations of PBDEs in dust samples from different 194 microenvironment categories have been reported (Harrad et al, 2008a; 2010). While similar 195 differences have not been reported for PCBs, this may possibly be due to the far more limited 196 database on these contaminants in indoor dust. We therefore hypothesised that significant 197 differences would exist in concentrations of PBDEs and PCBs in cars, homes, and offices in 198 this study. To test this hypothesis, we subjected our data to ANOVA. This analysis revealed 199 the following significant differences between concentrations of target contaminants in 200 different microenvironments. For BDEs-49 and -154, concentrations in car dust exceeded 201 significantly those in both homes and offices; while for BDE-197, concentrations in cars 202 were significantly greater than those in homes. In addition, concentrations of each individual 203 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 204 205 differences were observed.

206

# 3.3 How do concentrations of PBDEs and PCBs in this study compare with previousstudies?

209 Particularly noteworthy, are the highly elevated concentrations of Penta-BDE congeners in 2 210 car dust samples. At 9,300 and 3,700 ng  $g^{-1}$  for BDE-47 and 4,200 and 19,000 ng  $g^{-1}$  for 211 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.

217

218 Median concentrations detected in dust from cars, homes, and offices in this study are 219 compared with those from selected other studies in Tables 3 and 4 for PBDEs and PCBs 220 respectively. Median concentrations of PBDEs in both house and office dust in this study are 221 lower than those reported in the only other studies conducted in Nigeria (Olukunle et al, 222 2015a,b). This difference may be due to small sample numbers in these studies, and perhaps 223 also due to the different regions sampled (Makurdi, Benue State versus Lagos) but may also 224 be explained by the different sampling method employed in the previous Nigerian studies 225 (Olukunle et al, 2015a,b), in which the dust samples analysed comprised of a mix of floor 226 dust and dust sampled from product surfaces. We have shown elsewhere that PBDE transfer 227 from products to dust via direct product: dust contact is substantial (Rauert and Harrad, 2015), 228 and thus dust sampled from product surfaces would likely contain higher concentrations than 229 dust taken from the floor. Elsewhere in the African region, our concentrations of PBDEs in 230 house and office dust are lower than those reported recently for South Africa (Abafe and 231 Martincigh, 2015), but exceed those in earlier South African studies (Kefeni and Okwonkwo, 232 2012; Kefeni et al, 2014), and those reported elsewhere for Egypt and Iraq (Al-Omran and 233 Harrad, in press; Hassan and Shoeib, 2015).

234

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).

241

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 246 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.
- 248

249 This ANOVA comparison revealed concentrations of BDEs 28, 49, 47, 66, 100, 99, 154, and 250 153 to be significantly lower in Nigerian house dust than in dust from Canadian and USA 251 homes. This is consistent with the well-documented greater use of the Penta-BDE product in 252 North America. In addition, concentrations of BDEs-49 and 154 in New Zealand and UK 253 dust exceeded significantly those in house dust from Nigeria in this study. While 254 concentrations of the Octa-BDE marker congener BDE-183 in this study were statistically 255 indistinguishable from those in the other countries examined; concentrations of the Deca-256 BDE indicator (BDE-209) were significantly lower in this study than in house dust from both 257 the UK and the USA.

258

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.

265

#### 266 **3.4 What influences concentrations of PBDEs in indoor dust samples?**

267 We examined our data on concentrations of PBDEs and potential factors influencing these 268 concentrations. For car dust, we noted no correlation between vehicle age (which ranged 269 between 3 and 29 years, with an average of 11.6 years) and log-transformed concentration of 270 any target PBDE. Moreover, there was no apparent relationship between PBDE 271 concentrations and the car margue (5 Honda, 4 Toyota, 2 Nissan, and 1 each of Geely, Kia, 272 Lexus, Mercedes, and Rover). For example, the highest BDE-99 concentration (19,000 ng g 273 <sup>1</sup>) was observed in an 11 year old vehicle, while the same congener was not detected in a 13 274 year old car of the same marque. This observation is consistent with previous observations 275 that the year of vehicle manufacture is not the only factor influencing concentrations of 276 PBDEs in cars (Hazrati and Harrad, 2006).

277

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 280 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 281 282 whether the home was air conditioned or naturally ventilated as an independent variable – all 283 offices were air-conditioned. Office and house dust data were examined separately. Results 284 of this automatic linear modelling showed no significant relationships, except that 285 concentrations of  $\Sigma$ tri-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 286 no information on the age of the TVs in the offices studied, this is not inconsistent with the 287 288 reported past use of the Penta-BDE formulation in printed circuit boards (Betts, 2006).

289

#### 290 **3.5 Human exposure implications**

291 Tables 5, 6, and 7 give illustrative estimates of human exposure to (respectively) BDE-99, BDE-209, and  $\Sigma$ ICES-6 PCBs that would arise from ingestion of the dust samples measured 292 293 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 294 of 20 and 50 mg day<sup>-1</sup>, and high dust ingestion figures for adults and toddlers of 50 and 200 295 mg day<sup>-1</sup> (Jones-Otazo et al., 2005). We have then estimated exposure under various dust 296 297 ingestion scenarios for homes, offices, and cars separately, assuming ingested dust is contaminated at the 5<sup>th</sup> percentile, median, and 95<sup>th</sup> percentile concentrations in our dust 298 samples from each microenvironment category. Overall dust ingestion exposure estimates are 299 300 then calculated taking into account ingestion of dust in each of the relevant 301 microenvironments. Dust ingestion is assumed to occur pro-rata to typical activity patterns 302 (i.e. for adults: 72% home, 23.8% office, 4.2% car, for toddlers: 95.8% home and 4.2% car).

303

304 In summary, based on our data, Nigerian adult exposure via dust ingestion ranges between 0.08 and 22 ng day<sup>-1</sup>, 1.3 and 180 ng day<sup>-1</sup>, and 0.37 and 3.3 ng day<sup>-1</sup> for BDE-99, BDE-209, 305 and  $\Sigma$ ICES-6 PCBs respectively. The equivalent exposure estimates for Nigerian toddlers are 306 0.21 to 87 ng day<sup>-1</sup>, 4.3 to 600 ng day<sup>-1</sup>, and 0.91 and 12 ng day<sup>-1</sup>. While to our knowledge 307 308 there are no health based limit values (HBLVs) against which we can compare our estimate of exposure to  $\Sigma$ ICES-6 PCBs, such values do exist for BDEs-99 and -209. For BDE-99, the 309 relevant value is the USEPA's reference dose RfD of 100 ng kg body weight<sup>-1</sup> day<sup>-1</sup> (USEPA, 310 2008a), while for BDE-209 the USEPA RfD is 7,000 ng kg body weight<sup>-1</sup> day<sup>-1</sup> (USEPA, 311 2008b). Assuming a child weight of 20 kg, our worst-case exposure estimate (high-end for 312

toddlers) for BDEs-99 and -209 are 4.35 and 30 ng kg body weight<sup>-1</sup> day<sup>-1</sup> respectively. 313 While we have not considered exposure via other pathways such as inhalation, diet, and 314 315 dermal contact with dust and BFR-treated materials, there is a clearly substantial margin of 316 safety between apparent exposure of the Nigerian population to these PBDEs and the USEPA 317 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 318 of 0.23-0.30 ng kg body weight<sup>-1</sup> day<sup>-1</sup> for which impaired spermatogenesis is the end point 319 of concern (Bakker et al, 2008). Our estimates of exposure of a 20 kg child arising from 320 ingestion of dust contaminated with BDE-99 at the 95<sup>th</sup> percentile concentration at both the 321 average (1.1 ng kg body weight<sup>-1</sup> day<sup>-1</sup>) and high ingestion rates (4.35 ng kg body weight<sup>-1</sup> 322 323 day<sup>-1</sup>), both exceed this HBLV. Continued monitoring of human exposure to PBDEs would 324 therefore appear warranted.

325

#### 326 4. CONCLUSIONS

327 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, 328 329 concentrations of PCB 180 in Nigerian homes are significantly higher than those recorded 330 previously for New Zealand, the UK, and the USA. Moreover, concentrations of Penta-BDE 331 congeners in 2 car dust samples are amongst the highest ever reported. Overall, this study 332 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 333 334 hazard.

335

#### 336 ACKNOWLEDGEMENTS

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339

#### 340 APPENDIX A. SUPPLEMENTARY MATERIAL

Tables of concentrations of individual PBDEs and PCBs in all samples analysed, as well as
information on factors potentially influencing concentrations. Supplementary data associated
with this article can be found, in the online version, at

344

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Table 1: Statistical Summary<sup>a</sup> of Concentrations (ng g<sup>-1</sup>) of Selected PBDEs in Dust
from Nigerian Cars, Homes, and Offices.

Compound	Microenvironment	Detection %	Minimum	Median	Average	Maximun
BDE -28	Cars	81	< 0.04	1.1	7.0	65
DDE -28	Homes	83	< 0.04	0.48	0.75	3.1
	Offices	83	< 0.04	1.1	4.7	65
	Cars	88	< 0.05	28	900	9300
BDE- 47	Homes	100	2.2	8.0	13	50
	Offices	100	2.8	14	20	100
	Cars	81	< 0.02	3.0	39	440
BDE-49	Homes	33	< 0.02	< 0.02	0.46	2.6
	Offices	67	< 0.03	1.0	1.1	4.2
	Cars	94	< 0.03	12	330	3100
BDE -100	Homes	92	< 0.03	4.0	8.4	44
	Offices	94	< 0.03	4.2	7.3	24
	Cars	88	< 0.04	49	1700	19000
BDE -99	Homes	100	1.5	14	31	170
	Offices	94	< 0.04	18	28	110
	Cars	69	< 0.04	3.6	450	3500
BDE -154	Homes	42	< 0.04	< 0.04	1.7	7.9
	Offices	22	< 0.04	< 0.04	0.97	6.4
	Cars	69	< 0.05	9.0	720	6000
BDE – 153	Homes	92	< 0.05	3.9	5.3	22
	Offices	72	< 0.05	3.7	5.4	24
	Cars	88	< 0.04	8.8	83	480
BDE – 183	Homes	100	2.9	18	26	90
	Offices	100	4.3	26	66	350
	Cars	88	< 0.06	7.7	19	97
BDE – 197	Homes	58	< 0.06	1.8	2.1	8.4
	Offices	72	< 0.06	4.5	8.4	39
	Cars	88	< 0.06	4.7	12	76
BDE – 203	Homes	83	< 0.06	1.6	1.9	7.9
	Offices	67	< 0.06	2.8	3.8	13
	Cars	75	< 0.08	61	300	3100
BDE – 206	Homes	58	< 0.08	15	19	61
	Offices	72	< 0.08	52	56	180
	Cars	63	< 0.08	22	84	870
BDE - 208	Homes	42	< 0.08	< 0.08	6.1	23
-	Offices	56	< 0.08	9.2	15	88
	Cars	81	< 0.11	780	10000	52000
BDE - 209	Homes	100	<0.11 77	390	420	940
	Offices	89	< 0.11	930	1200	4900

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- 481
- <sup>a</sup>For the purposes of statistical evaluation, all concentrations below the limit of detection (LOD) were assigned a value of f x LOD, where f is the fractional detection frequency of the 482
- 483 contaminant in samples from that microenvironment

485 **Table 2: Statistical summary**<sup>a</sup> of concentrations (ng g<sup>-1</sup>) of PCBs detected in dust from

486 Nigerian cars, homes, and offices

487

Compound	Microenvironment	Minimum	Median	Average	Maximum
	Cars	0.24	1.9	1.9	4.0
PCB-28	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

488

491 contaminant in samples from that microenvironment

<sup>489 &</sup>lt;sup>a</sup>For the purposes of statistical evaluation, all concentrations below the limit of detection

<sup>490 (</sup>LOD) were assigned a value of f x LOD, where f is the fractional detection frequency of the

Sampling year-Country (Reference)	n	<b>BDE 28</b>	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.	1,540
2009 - UK (Harrad and Abdallah, 2011)	14	n.a.	100	17	130	10	14	6	4,100	3,700	4,800	190,000
2006/07- USA (Batterman et al, 2009)	12	13	1,800	790	2,600	120	77	73	580	490	250	3,100
							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 (Olunkunle 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.	1,550
2010/11 - South Africa (Kefeni et al, 2014)	31	n.a.	2.6	< 0.13	2.6	<lod< td=""><td><lod< td=""><td>n.a.</td><td>n.a.</td><td>n.a.</td><td>n.a.</td><td>&lt;1.8</td></lod<></td></lod<>	<lod< td=""><td>n.a.</td><td>n.a.</td><td>n.a.</td><td>n.a.</td><td>&lt;1.8</td></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.	2,800
2006 - USA (Harrad et al, 2008b)	20	14	410	160	820	89	110	16	n.a.	n.a.	n.a.	1,300
							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 Okwonkwo, 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.	6,200
2006/07 - USA (Batterman et al, 2010)	10	3.0	978	399	1,760	78	48	30	n.a.	n.a.	n.a.	1.0

Table 3: Comparison of median concentrations (ng g<sup>-1</sup>) of selected PBDEs detected in dust in this study with selected previous reports.

494 n.a. = not available

San	pling year - Country (Reference)	n	PCB 28	PCB 52	PCB 101	PCB 153	PCB 138	PCE 180
	· · · · · · · · · · · · · · · · · · ·			Cars				
201	3 – Nigeria (this study)	1 6	1.9	2.1	0.72	1.6	1.4	1.9
2011 -	- Kuwait (Ali et al, 2013)	1 5	n.a.	n.a.	< 0.2	< 0.2	< 0.1	0.2
2011 -	Pakistan (Ali et al, 2013)	1 5	n.a.	n.a.	<0.2	0.3	0.6	0.3
				Hom es				
201	3 – Nigeria (this study)	1 2	3.8	4.1	2.7	5.8	6.0	10
	South Africa (Abafe and Martincigh, 2015)	1 0	10.9	n.a.	n.a.	150	n.a.	585
2006 – 0	Canada (Harrad et al, 2009)	1 0	7.3	7.2	8.8	9.9	9.5	6.8
2011 -	- Kuwait (Ali et al, 2013)	1 5	n.a.	n.a.	<0.2	<0.2	< 0.1	0.4
2006 - 1	New Zealand (Harrad et al, 2009)	2 0	2.3	1.4	1.6	1.4	1.8	1.3
2011 -	Pakistan (Ali et al, 2013)	1 5	n.a.	n.a.	< 0.2	< 0.2	< 0.1	0.3
2006 -	UK (Harrad et al, 2009)	2 0	3.4	1.8	1.2	1.2	1.1	0.89
2006 -	USA (Harrad et al, 2009)	2 0	5.1	6.2	8.7	7.1	6.5	2.6
2011 -	USA (Dodson et al, 2012)	1 6	n.a.	n.a.	n.a.	9.5	n.a.	8.5
				Offic				
201	3 – Nigeria (this study)	1 8	5.2	<b>es</b> 4.4	6.2	6.5	5.1	14
	South Africa (Abafe and Martincigh, 2015)	1 1	28	n.a.	n.a.	136	n.a	812
	Hong Kong (Kang et al, 2013)	2 0	7.8	1.9	1.1	4.3	2.2	2.3

Table 4: Comparison of median concentrations (ng g<sup>-1</sup>) of selected PCBs detected in
 dust in this study with selected previous reports.

497 n.a. = not available

	Exposure in offices		Exposure in homes		Exposur	e in cars	Overall o		
	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	RfD <sup>b</sup>						
Adult 5 <sup>th</sup> percentile	0.01 (17.3)	0.03	0.06 (76)	0.15	0.01 (6.7)	0.01	0.08	0.20	7,000
Adult median	0.08 (25.8)	0.21	0.20 (61.5)	0.50	0.04 (12.6)	0.10	0.33	0.82	7,000
Adult 95 <sup>th</sup> percentile	0.39 (4.5)	0.96	1.5 (17.2)	3.7	6.7 (78.3)	17	8.6	22	7,000
Toddler 5 <sup>th</sup> percentile	-	-	0.20	0.80	0.01 (6.2)	0.05	0.21	0.85	2,000
Toddler median	-	-	0.67	2.67	0.10 (13.4)	0.41	0.77	3.1	2,000
Toddler 95 <sup>th</sup> percentile	-	-	4.9	20	17 (77.3)	67	22	87	2,000

Table 5: Overall exposure of Nigerian adults and toddlers (ng day<sup>-1</sup>) to BDE-99 via ingestion of dust and percentages arising from
 ingestion of dust in cars, homes, and offices

<sup>a</sup>assuming mean and high dust ingestion rates of 20 and 50 mg day<sup>-1</sup> for adults and 50 and 200 mg day<sup>-1</sup> 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 5<sup>th</sup>, 50<sup>th</sup>, or 95<sup>th</sup> percentile for that microenvironment

503 Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust 504 ingestion rate scenarios

<sup>505</sup> <sup>b</sup>RfD is USEPA reference dose for BDE-99 (100 ng (kg body weight) day<sup>-1</sup>), converted to ng day<sup>-1</sup> assuming 70 kg and 20 kg body weight for

adults and toddlers respectively.

	Exposure	in offices	Exposure	in homes	Exposur	e in cars	Overall		
	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	<b>RfD</b> <sup>b</sup>
Adult 5 <sup>th</sup> 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 95 <sup>th</sup> percentile	17 (23.5	43	13 (17.9)	32	43 (58.6)	110	73	180	490,000
Toddler 5 <sup>th</sup> 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 95 <sup>th</sup> percentile	-	-	43 (9.2)	170	110 (71.2)	430	150	600	140,000

Table 6: Overall exposure of Nigerian adults and toddlers (ng day<sup>-1</sup>) to BDE-209 via ingestion of dust and percentages arising from
 ingestion of dust in cars, homes, and offices

<sup>a</sup>assuming mean and high dust ingestion rates of 20 and 50 mg day<sup>-1</sup> for adults and 50 and 200 mg day<sup>-1</sup> 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,

512 4.2 % cars; and that concentration in dust is either  $5^{\text{th}}$ ,  $50^{\text{th}}$ , or  $95^{\text{th}}$  percentile for that microenvironment

513 Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust 514 ingestion rate scenarios

<sup>b</sup>RfD is USEPA reference dose for BDE-209 (7,000 ng (kg body weight) day<sup>-1</sup>), converted to ng day<sup>-1</sup> assuming 70 kg and 20 kg body weight for  $\frac{1}{2}$ 

516 adults and toddlers respectively.

517

	Exposure in offices		Exposure	in homes	Exposur	e in cars	Overall exposure		
	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion <sup>a</sup>	High dust ingestion <sup>a</sup>	Mean dust ingestion	High dust ingestion	
Adult 5 <sup>th</sup> 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 95 <sup>th</sup> percentile	0.42 (31.6)	1.0	0.88 (66.7)	2.2	0.02 (1.8)	0.06	1.3	3.3	
Toddler 5 <sup>th</sup> 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 95 <sup>th</sup> percentile	-	-	2.9 (98)	12	0.06 (2)	0.23	3.0	12	

Table 7: Overall exposure of Nigerian adults and toddlers (ng day<sup>-1</sup>) to ΣICES-6 PCBs via ingestion of dust and percentages arising 519 520 from ingestion of dust in cars, homes, and offices

<sup>a</sup>assuming mean and high dust ingestion rates of 20 and 50 mg day<sup>-1</sup> for adults and 50 and 200 mg day<sup>-1</sup> for toddlers; that dust ingestion is pro 521

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 5<sup>th</sup>, 50<sup>th</sup>, or 95<sup>th</sup> percentile for that microenvironment 522

523

Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust 524 ingestion rate scenarios 525

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