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

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*DOI:*

[10.1016/j.chemosphere.2015.12.045](https://doi.org/10.1016/j.chemosphere.2015.12.045)

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*Document Version*

Peer reviewed version

*Citation for published version (Harvard):*

Harrad, S, Abdallah, MA-E & Oluseyi, T 2016, 'Polybrominated diphenyl ethers and polychlorinated biphenyls in dust from cars, homes, and offices in Lagos, Nigeria', *Chemosphere*, vol. 146, pp. 346-353.  
<https://doi.org/10.1016/j.chemosphere.2015.12.045>

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Checked Jan 2016

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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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15

16 **ABSTRACT**

17 Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were  
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-  
21 47 and BDE-99 in two car dust samples (9,300 and 3,700 ng g<sup>-1</sup> for BDE-47 and 4,200 and  
22 19,000 ng g<sup>-1</sup> for BDE-99), are amongst the highest ever reported in car dust. ANOVA  
23 comparison with Canada, New Zealand, the UK, and the USA; reveals concentrations of  
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  
31 significant only for BDEs-49, 154, and 197, with concentrations in cars significantly greater  
32 than those in offices for BDEs-49 and 154. Contrastingly, concentrations of all target PCBs  
33 in offices exceeded significantly those in cars. This study underlines the truly global  
34 distribution of indoor contamination with PBDEs and PCBs.

35

36 **KEYWORDS**

37 POPs

38 BFRs

39 PCBs

40 Africa

41 Nigeria

42 Indoor dust

43

## **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  
66 a given PBDE congener (exposure to decabromodiphenyl ether (BDE-209) is dominated by  
67 dust ingestion as a consequence of its very low vapour pressure and comparatively low  
68 capacity for bioaccumulation).

69  
70 To date, the vast majority of exposure assessments conducted for PBDEs, have been  
71 conducted in East Asia (China, Korea, and Japan), Europe, and North America (Besis and  
72 Samara, 2012; Harrad et al, 2010). While data is emerging for other regions (including Egypt  
73 (Hassan and Shoeib, 2015), Kuwait (Ali et al, 2013; Gevao et al, 2006), and South Africa  
74 (Kefeni and Okwonkwo, 2012; Obafe and Martincigh, 2015)), to our knowledge only two  
75 previous studies exist concerning the presence of PBDEs in indoor dust in Nigerian car and  
76 house dust respectively (Olukunle et al, 2015a,b).

77

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  
98 environments. Moreover, we provide only the second such report worldwide on  
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  
113 houses in Lagos, Nigeria (the largest city in Nigeria) between September and October 2014.  
114 House and office dust samples were collected using a vacuum cleaner, according to a  
115 standardised method (Harrad et al, 2008b). Briefly, 1 m<sup>2</sup> of carpeted floor was vacuumed for  
116 2 min, while for bare floors, 4 m<sup>2</sup> surface was vacuumed for 4 min. Dust was retained using  
117 25 µm pore size nylon sample socks (Allied Filter Fabric Pty Ltd, Australia) mounted in the  
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,  
120 socks were closed with a twist tie, sealed in a plastic bag and stored at -20 °C until  
121 transportation via courier to the University of Birmingham for sieving and analysis. Before  
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  
128 µm mesh testing sieve (UKGE Limited, UK), covered with the lid and shaken for 2-4 min.  
129 Sieved samples were stored in clean, n-hexane rinsed glass jars and stored at 4 °C until  
130 analysis.

131

## 132 **2.2 Sample extraction**

133 Accurately weighted aliquots of dust (~0.15 g) were loaded into pre-cleaned 66 mL cells  
134 containing 1.5 g Florisil and Hydromatrix (Varian Inc., UK) to fill the void volume of the  
135 cells, and spiked with internal (surrogate) standards (15 ng of each of BDE 77, BDE 128 and  
136 30 ng of <sup>13</sup>C<sub>12</sub>-BDE 209) prior to pressurised liquid extraction (ASE 350, Dionex, Hemel  
137 Hempstead, UK) using hexane:dichloromethane (1:9, v/v) at 90 °C and 1500 psi. The heating  
138 time was 5 minutes, static time 4 min, purge time 90 s, flush volume 50 %, with three static  
139 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  $^{13}\text{C}_{12}$ -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,  
151 206, and 209) were quantified using a TRACE™ 1310 Gas Chromatograph coupled to ISQ™  
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  $\mu$ m; 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  
173 detection (LOD) were assigned a value of  $f \times \text{LOD}$ , where  $f$  is the fractional detection  
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

179 were log-transformed prior to comparison of means via ANOVA, as well as regression  
180 analysis. A P value <0.05 was used as the level indicating statistical significance.

181

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

#### 183 **3.1 Concentrations of PBDEs and PCBs in Nigerian house dust**

184 Statistical summaries of concentrations of selected PBDEs and PCBs in samples analysed in  
185 this study are provided as Tables 1 and 2 respectively, with concentrations of all target  
186 contaminants in each individual sample supplied as supplementary material. Each target PCB  
187 was detected in every sample. By comparison, detection frequency varied between PBDE  
188 congeners. BDEs 47, 99, 100, 183, and 209 were detected in nearly all samples, with  
189 detection frequencies for other congeners ranging from 17 % for BDE-17 in house and office  
190 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  
204 of PCBs in car dust also exceeded significantly by those in house dust. No other significant  
205 differences were observed.

206

#### 207 **3.3 How do concentrations of PBDEs and PCBs in this study compare with previous 208 studies?**

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<sup>-1</sup> for BDE-47 and 4,200 and 19,000 ng g<sup>-1</sup> for  
211 BDE-99, these concentrations approach the maximum values reported anywhere in car dust  
212 of 30,000 ng g<sup>-1</sup> and 63,000 ng g<sup>-1</sup> (Batterman et al, 2009). These two samples came from

213 vehicles that were respectively 14 and 11 years old at the time of sampling. Both were  
214 Japanese marques that to our knowledge were manufactured in Japan. Further discussion of  
215 the relationship between PBDE concentrations and potential influencing factors follows in  
216 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  
235 For PCBs, concentrations in this study are lower than those reported for South African homes  
236 and offices (Abafe and Martincigh, 2015), but exceed substantially those in Kuwaiti and  
237 Pakistani homes (Ali et al, 2013), and also those in Hong Kong offices (Kang et al, 2013). To  
238 the best of our knowledge, there is only one other study of PCBs in car dust, and  
239 concentrations of all target congeners in this study exceed substantially those reported for  
240 cars in Kuwait and Pakistan (Ali et al, 2013).

241  
242 In addition, we used ANOVA to test whether concentrations of both PBDEs and PCBs in  
243 house dust in this study were significantly different to those reported previously by our  
244 research group using identical sampling and analytical procedures in house dust from  
245 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  
247 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

259 Similar comparison for PCBs, showed concentrations in this study to be statistically  
260 indistinguishable from those in Canadian, New Zealand, UK, and USA house dust, with the  
261 exception of PCB-180. For this congener, concentrations in this study exceeded significantly  
262 those in New Zealand, the UK, and the USA. As PCB-180 was most prevalent in the highly  
263 chlorinated commercial formulations such as Aroclor 1260 manufactured by Monsanto, this  
264 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 marque (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<sup>-1</sup>)  
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

278 We next conducted multiple linear regression analysis of our data for office and house dust  
279 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,  
281 microwaves, and fridge/freezers as independent variables. In homes, we also incorporated  
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  
286 significantly ( $p=0.04$ ) positively correlated with the number of TVs in offices. While we have  
287 no information on the age of the TVs in the offices studied, this is not inconsistent with the  
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,  
292 BDE-209, and  $\Sigma$ ICES-6 PCBs that would arise from ingestion of the dust samples measured  
293 in this study. In common with the vast majority of other studies, we have conservatively  
294 assumed 100% absorption of intake and used average adult and toddler dust ingestion figures  
295 of 20 and 50  $\text{mg day}^{-1}$ , and high dust ingestion figures for adults and toddlers of 50 and 200  
296  $\text{mg day}^{-1}$  (Jones-Otazo et al., 2005). We have then estimated exposure under various dust  
297 ingestion scenarios for homes, offices, and cars separately, assuming ingested dust is  
298 contaminated at the 5<sup>th</sup> percentile, median, and 95<sup>th</sup> percentile concentrations in our dust  
299 samples from each microenvironment category. Overall dust ingestion exposure estimates are  
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  
305 0.08 and 22  $\text{ng day}^{-1}$ , 1.3 and 180  $\text{ng day}^{-1}$ , and 0.37 and 3.3  $\text{ng day}^{-1}$  for BDE-99, BDE-209,  
306 and  $\Sigma$ ICES-6 PCBs respectively. The equivalent exposure estimates for Nigerian toddlers are  
307 0.21 to 87  $\text{ng day}^{-1}$ , 4.3 to 600  $\text{ng day}^{-1}$ , and 0.91 and 12  $\text{ng day}^{-1}$ . While to our knowledge  
308 there are no health based limit values (HBLVs) against which we can compare our estimate  
309 of exposure to  $\Sigma$ ICES-6 PCBs, such values do exist for BDEs-99 and -209. For BDE-99, the  
310 relevant value is the USEPA's reference dose RfD of 100  $\text{ng kg body weight}^{-1} \text{day}^{-1}$  (USEPA,  
311 2008a), while for BDE-209 the USEPA RfD is 7,000  $\text{ng kg body weight}^{-1} \text{day}^{-1}$  (USEPA,  
312 2008b). Assuming a child weight of 20 kg, our worst-case exposure estimate (high-end for

313 toddlers) for BDEs-99 and -209 are 4.35 and 30 ng kg body weight<sup>-1</sup> day<sup>-1</sup> respectively.  
314 While we have not considered exposure via other pathways such as inhalation, diet, and  
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  
318 (albeit of no legislative standing) for BDE-99 proposed by researchers from the Netherlands  
319 of 0.23-0.30 ng kg body weight<sup>-1</sup> day<sup>-1</sup> for which impaired spermatogenesis is the end point  
320 of concern (Bakker et al, 2008). Our estimates of exposure of a 20 kg child arising from  
321 ingestion of dust contaminated with BDE-99 at the 95<sup>th</sup> percentile concentration at both the  
322 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>  
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  
328 offices. While concentrations are in the main at the lower end of those reported globally,  
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  
333 the continuing need for action to reduce and eventually eliminate this potential public health  
334 hazard.

335

#### 336 **ACKNOWLEDGEMENTS**

337 The authors express their thanks to all the dust donors. Temilola Oluseyi acknowledges  
338 gratefully funding from the Commonwealth Scholarship Commission.

339

#### 340 **APPENDIX A. SUPPLEMENTARY MATERIAL**

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

344

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

<b>Compound</b>	<b>Microenvironment</b>	<b>Detection %</b>	<b>Minimum</b>	<b>Median</b>	<b>Average</b>	<b>Maximum</b>
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	19000
	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	10000	52000
	Homes	100	77	390	420	940
	Offices	89	<0.11	930	1200	4900

481 <sup>a</sup>For the purposes of statistical evaluation, all concentrations below the limit of detection  
482 (LOD) were assigned a value of  $f \times \text{LOD}$ , where  $f$  is the fractional detection frequency of the  
483 contaminant in samples from that microenvironment  
484

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
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
PCB-52	Cars	0.08	2.1	2.3	5.9
	Homes	0.80	4.2	4.5	8.6
	Offices	0.54	4.4	4.6	11
PCB-101	Cars	0.14	0.72	1.3	5.5
	Homes	0.37	2.7	3.7	9.4
	Offices	2.6	6.2	8.7	24
PCB-153	Cars	0.08	1.6	2.5	9.1
	Homes	0.22	5.8	7.3	22
	Offices	3.8	6.5	10	23
PCB-138	Cars	0.40	1.4	1.9	6.7
	Homes	0.31	6.0	7.5	22
	Offices	2.3	5.1	7.3	26
PCB-180	Cars	0.46	1.9	2.3	5.7
	Homes	4.4	10	10	24
	Offices	1.9	14	14	34

488  
 489 <sup>a</sup>For the purposes of statistical evaluation, all concentrations below the limit of detection  
 490 (LOD) were assigned a value of f x LOD, where f is the fractional detection frequency of the  
 491 contaminant in samples from that microenvironment

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**Table 3: Comparison of median concentrations (ng g<sup>-1</sup>) 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
<b>Cars</b>												
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
<b>Homes</b>												
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	<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
<b>Offices</b>												
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

494

n.a. = not available

495 **Table 4: Comparison of median concentrations (ng g<sup>-1</sup>) of selected PCBs detected in**  
 496 **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
<b>Cars</b>							
2013 – 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
<b>Homes</b>							
2013 – Nigeria (this study)	1 2	3.8	4.1	2.7	5.8	6.0	10
2012 - South Africa (Abafe and Martincigh, 2015)	1 0	10.9	n.a.	n.a.	150	n.a.	585
2006 – 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 - 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
<b>Offices</b>							
2013 – Nigeria (this study)	1 8	5.2	4.4	6.2	6.5	5.1	14
2012 - South Africa (Abafe and Martincigh, 2015)	1 1	28	n.a.	n.a.	136	n.a.	812
2009 - Hong Kong (Kang et al, 2013)	2 0	7.8	1.9	1.1	4.3	2.2	2.3

497 n.a. = not available

498 **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**  
 499 **ingestion of dust in cars, homes, and offices**

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

500 <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  
 501 rata to estimated time spent in each microenvironment category: for adults 72 % homes, 23.8 % offices, 4.2 % cars; for toddlers 95.8% homes,  
 502 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

505 <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  
 506 adults and toddlers respectively.  
 507



508 **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**  
 509 **ingestion of dust in cars, homes, and offices**

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

510 <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  
 511 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<sup>th</sup>, 50<sup>th</sup>, or 95<sup>th</sup> 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

515 <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  
 516 adults and toddlers respectively.

517  
 518

519 **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**  
 520 **from ingestion of dust in cars, homes, and offices**

	Exposure in offices		Exposure in homes		Exposure 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
<b>Adult 5<sup>th</sup> percentile</b>	0.09 (25.6)	0.24	0.27 (73.7)	0.68	<0.01 (0.7)	0.01	0.37	0.92
<b>Adult median</b>	0.20 (28.4)	0.50	0.49 (70.2)	1.2	0.01 (1.4)	0.02	0.70	1.8
<b>Adult 95<sup>th</sup> percentile</b>	0.42 (31.6)	1.0	0.88 (66.7)	2.2	0.02 (1.8)	0.06	1.3	3.3
<b>Toddler 5<sup>th</sup> percentile</b>	-	-	0.90 (99.3)	3.6	0.01 (0.7)	0.02	0.91	3.6
<b>Toddler median</b>	-	-	1.6 (98.5)	6.5	0.02 (1.5)	0.10	1.7	6.6
<b>Toddler 95<sup>th</sup> percentile</b>	-	-	2.9 (98)	12	0.06 (2)	0.23	3.0	12

521 <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  
 522 rata to estimated time spent in each microenvironment category: for adults 72 % homes, 23.8 % offices, 4.2 % cars; for toddlers 95.8% homes,  
 523 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  
 524 Figures in parentheses denote percentage of overall exposure received in that microenvironment category; note percentage is same for both dust  
 525 ingestion rate scenarios

**Supplementary Material**

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