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## Concentrations of halogenated flame retardants and polychlorinated biphenyls in house dust from Lagos, Nigeria†

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Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and hexabromocyclododecane (HBCDD) are regulated under the Stockholm Convention of the United Nations' Environment Programme; with similar concerns emerging about alternative halogenated flame retardants (alt-HFRs), the use of which is increasing as replacements for PBDEs and HBCDD. While the presence in indoor dust of PCBs, PBDEs, and HBCDDs has been reported previously in a few African locations including Lagos, Nigeria, we are unaware of similar data for alt-HFRs. The present study thus aimed to provide the first information on alt-HFRs in indoor dust in sub-Saharan Africa, and to evaluate the impact of restrictions on the use of PBDEs, HBCDD, and PCBs on their concentrations in house dust in Lagos, Nigeria. Concentrations of  $\sum_8$ PBDEs,  $\sum$ HBCDDs,  $\sum_7$ alt-HFRs, and  $\sum_8$ PCBs in 15 samples of dust from homes in Lagos, Nigeria were found to be: 43–810 (median = 300) ng g<sup>-1</sup>, <dl – 66 (median = <dl) ng g<sup>-1</sup>, 32–2600 (median = 320) ng g<sup>-1</sup> and 3.8–61 (median = 18) ng g<sup>-1</sup> respectively. The dominant PBDE was BDE-209, its replacement decabromodiphenyl ethane (DBDPE) was the predominant alt-HFR, while PCB-138 displayed the highest concentration of the 8 PCBs targeted. Likely due to their higher vapour pressures, concentrations of the non-arochlor PCB 11, as well as those of PCB 28, and PBDE 28 were below detection limits. Concentrations of PBDEs and PCBs reported are generally below those reported previously for Lagos, Nigeria; suggesting restrictions on their manufacture and use have been effective. In contrast, while concentrations of BDE-209 in this study were lower than in one previous study in Lagos, they exceeded those in another; implying that the more recent restrictions on the deca-BDE product have yet to be fully effective. The evidence presented here of concentrations of alt-HFRs in Nigerian house dust provide a valuable benchmark against which future trends in their concentrations may be evaluated.

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### Environmental significance

Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) found extensive past use as flame retardants, with polychlorinated biphenyls (PCBs) also used widely, leading to substantial environmental contamination and human exposure. Alongside toxicity concerns, this resulted in bans on their manufacture. Despite this, emissions continue from products remaining in use, with new concerns raised about exposure to non-regulated, structurally similar alternative halogenated flame retardants (alt-HFRs). This study augments the sparse database on PBDEs, HBCDD, and PCBs in African house dust and provides the first information on alt-HFRs in indoor dust from sub-Saharan Africa. Encouraging declines in PCBs and some PBDEs indicate a beneficial impact of past restrictions; and we provide a benchmark against which future trends in alt-HFRs may be evaluated.

## 1. Introduction

While polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs) like polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) are not known to have

been produced in Africa;<sup>1</sup> due to *inter alia* their use in imported goods, their presence is widely reported in diverse environmental compartments across the African continent.<sup>2</sup>

Both 'legacy' brominated flame retardants (BFRs) like PBDEs and HBCDD, as well as alternative halogenated flame retardants (alt-HFRs) were or are applied principally as additives in a wide range of consumer products including electronics, fabrics, and furniture foams.<sup>3–5</sup> As a consequence of concerns over their toxicity, environmental persistence, capacity for bioaccumulation, and potential for long range atmospheric transport; both PBDEs and HBCDD are listed as persistent organic

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pollutants (POPs) under the Stockholm Convention of the United Nations Environment programme.<sup>6</sup>

Alt-HFRs encompass a range of chemicals that are potential replacements for the regulated PBDEs and HBCDD. Those targeted in this study are: decabromodiphenyl ethane (DBDPE), dechlorane plus (DP), hexabromobenzene (HBBz), pentabromobenzene (PBBz), pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT).<sup>3</sup> These are targeted here due to concerns over their toxicity, persistence and bioaccumulation potential.<sup>7,8</sup> For example, DBDPE, a potential BDE 209 replacement, was highlighted as bioaccumulative<sup>9</sup> and a potential thyroid hormone disruptor.<sup>10</sup> Moreover, the USEPA has highlighted DBDPE to display similar developmental toxicity to that of decaBDE.<sup>11</sup> This is of concern as the EPA's reference dose for decaBDE (7 µg per kg per body weight per day) is based on its neurodevelopmental toxicity.<sup>12</sup>

Up until widespread bans on their manufacture and new use in the late 1970s-early 1980s; the commercial mixtures of PCBs produced under different trade names such as arochlor, were applied in many jurisdictions as dielectric fluids in electrical transformers and capacitors, alongside other applications in: adhesives, paints, building sealants, fire proofing agents, inks, and paints.<sup>1,13</sup> Furthermore, recent studies have reported substantial concentrations of congeners such as PCB 11, which are not present in technical commercial mixtures like aroclors. Sources of PCB 11 and other so-called non-arochlor PCBs include diarylide dyes and degradation of higher chlorinated PCB congeners.<sup>14</sup> Current knowledge of the toxicity of PCB 11 is limited, but the arochlor PCBs have been regulated since 2001 under the UNEP Stockholm Convention on POPs.<sup>6</sup>

Ingestion of domestic dust is an established pathway of human exposure to organic contaminants, particularly for toddlers due to their greater hand-to-mouth behaviour compared to adults combined with their low body weight. There are several studies targeting PBDEs,<sup>15-19</sup> HBCDDs,<sup>17,20</sup> and PCBs<sup>15,16,21</sup> in domestic dust from a number of locations in the African continent. However, data on concentrations of alt-HFRs in African domestic dust are scarce. Against this backdrop, this study's objectives were to evaluate temporal trends in concentrations of PBDEs and PCBs in house dust from Lagos, Nigeria by comparing data for samples collected in 2020 with those reported previously for samples collected in 2014,<sup>16,20</sup> and to provide baseline data for concentrations of alt-HFRs in the same dust samples.

## 2. Materials and methods

### 2.1 Sampling procedures

One convenience sample of indoor dust was collected from each of 15 homes in Lagos, Nigeria, which is one of the most industrialised and urbanised cities in Africa with population of about 24 million. Samples were collected with a hand brush comprising synthetic polyester bristles.<sup>22</sup> Dust samples, excluding small stones and heavy materials, were collected from the most-frequented parts of the living room floor, wrapped carefully in hexane-washed aluminium foil, and sealed in a zip lock bag before they were transferred to the laboratory. Samples

were then stored at ~20 °C before shipping to the University of Birmingham for chemical analysis. On receipt at Birmingham, the dust samples were each passed through a hexane – pre-cleaned 500 µm mesh testing sieve (UKGE Limited, UK) prior to extraction and analysis.

### 2.2 Chemical reagents and standards

All solvents used in this study were of HPLC grade or higher, obtained from Fisher Scientific (Loughborough, UK). Individual stock standard solutions of natives, internal standards, and recovery determination standards (PBDEs – 28, 47, 99, 100, 153, 154, 183, 209, 77, 128, <sup>13</sup>C-BDE-100, <sup>13</sup>C-BDE-209,  $\alpha$ -HBCDD,  $\beta$ -HBCDD,  $\gamma$ -HBCDD, <sup>13</sup>C- $\alpha$ -HBCDD, <sup>13</sup>C- $\beta$ -HBCDD, <sup>13</sup>C- $\gamma$ -HBCDD, d<sub>18</sub>- $\gamma$ -HBCDD, DBDPE, *anti*-DP, *syn*-DP, HBBz, PBBz, PBEB, and PBT) were purchased from Wellington Laboratories (Guelph, ON, Canada). PCBs – 11, 14, 28, 29, 34, 52, 62, 101, 118, 119, 129, 138, 147, 153, 173, and 180 were bought from Greyhound Chromatography (Birkenhead, Merseyside, UK). Pesticide grade florisil (60–100 mesh) and concentrated sulfuric acid, were purchased from Acros Organics (Geel, Belgium) and Sigma-Aldrich (St. Louis, MA, US) respectively.

### 2.3 Sample extraction and cleanup

Approximately 100 mg (accurately weighed) of dust was spiked with an internal standard mixture comprising: BDE-77, BDE-128, <sup>13</sup>C-BDE-209; <sup>13</sup>C- $\alpha$ -HBCDD, <sup>13</sup>C- $\beta$ -HBCDD, <sup>13</sup>C- $\gamma$ -HBCDD; PCB-14, PCB-34, PCB-62, PCB-119, PCB-147, and PCB-173. The spiked dust sample was extracted with 5 mL hexane followed by 1 min vortexing, prior to 15 min sonication, followed by 5 min centrifugation at 3500 rpm. This procedure was repeated twice and the supernatants combined in a 100 mL TurboVap® tube (Drage *et al.*, 2020). The crude extract was concentrated to ~2 mL hexane using a TurboVap® evaporator before acid digestion with ~2 mL concentrated sulfuric acid overnight.<sup>23</sup> After decanting the hexane layer, the cleaned extract was transferred with hexane washings and applied to a 9 mm i.d. chromatographic column containing 1 g florisil topped with 1 cm anhydrous sodium sulphate, pre-washed with 5 mL hexane, before extract elution with 15 mL hexane. The resulting eluate was concentrated to incipient dryness using a TurboVap® before reconstitution in 100 µL toluene containing 20 ng PCB-29 and -129, and 50 ng <sup>13</sup>C-BDE-100 and d<sub>18</sub>- $\gamma$ -HBCDD as recovery determination (or syringe) standards (RDS).<sup>22,23</sup>

### 2.4 Instrumental analysis

Target BFRs were analysed as previously described.<sup>23</sup> Briefly, 1 µL of the extract was injected onto a TRACE™ 1310 Gas Chromatograph/ISQ™ single quadrupole mass spectrometer (Thermo Fisher Scientific, Austin, TX, USA) operated in electron ionisation, selected ion monitoring (EI-SIM) mode and fitted with a Restek Rxi-5Sil MS chromatographic column (15 m × 0.25 mm × 0.25 µm film thickness). Analysis of target PCBs was performed on GC Agilent HP6850/5975 MS. One µL of the final sample extract was injected onto a Restek Rxi-5Sil MS column (30 m × 0.25 mm × 0.25 µm film thickness). The carrier gas



was pure helium at a flow rate of  $1.5 \text{ mL min}^{-1}$  for both BFRs and PCBs, with the column oven temperature programmes and  $m/z$  ions monitored provided in Tables S1 and S2a.†

After analysis of BFRs and PCBs was complete, the extract was reconstituted in methanol and HBCDDs measured using a dual pump Shimadzu LC-20AB prominence liquid chromatograph (Shimadzu, Kyoto, Japan) interfaced with a Sciex API 2000 triple quadrupole mass spectrometer (Applied Biosystems, Foster City, CA). Target HBCDD diastereomers were separated on an Agilent Pursuit XRS3 C18 reversed phase analytical column ( $150 \text{ mm} \times 2 \text{ mm i.d.}$ ,  $3 \mu\text{m}$  particle size). Full details of the ions monitored and the mobile phase elution gradient employed for HBCDD analysis are provided as ESI in Table S2b and Fig. S1.†<sup>20,24</sup>

## 2.5 Quality assurance/quality control

All glassware were thoroughly cleaned with tap water, then detergent solution, further with Milli Q water and a solvent mixture of dichloromethane : hexane (1 : 1), before finally baked for  $\sim 3$  hours at  $425 \text{ }^\circ\text{C}$  before use. Aliquots ( $n = 10$ ) of the Standard Reference Material NIST SRM 2585 (organics in indoor dust) were analysed to evaluate method accuracy and precision, with the concentrations obtained within acceptable values (Table S3b†). A procedural blank in which the sample was substituted with anhydrous sodium sulfate was analysed for every 5 samples. Only BDE-47 was found in the blank samples at concentrations below 5% of those detected in samples in all cases. Limits of quantifications (LOQs) were  $0.19\text{--}1.5 \text{ ng g}^{-1}$ ,  $2.4\text{--}11 \text{ ng g}^{-1}$ ,  $0.10\text{--}54 \text{ ng g}^{-1}$ , and  $0.19\text{--}0.57 \text{ ng g}^{-1}$  for BDEs, HBCDDs, alt-HFRs, and PCBs respectively. Average recoveries of internal standards were 71–110% (BFRs), 75–76% (HBCDD); and 56–89% (PCBs) (Table S4c†).

## 2.6 Statistical analysis

Box plot and Pearson correlations were performed in Microsoft Excel 365, while one-way ANOVA tests were performed in IBM SPSS Statistics 26. For statistical analysis purposes, non-detect values were replaced by the fractional detection frequency ( $df$ )  $\times$  LOQ. A confidence interval of  $\geq 95\%$  was taken to denote statistical significance.

## 2.7 Estimation of exposure

Exposures to BDEs, NBFRs, and PCBs *via* dust ingestion were estimated using the following algorithm (eqn (1)):<sup>21,25,26</sup>

$$\text{Estimated daily intake (EDI) (ng per kg bw per day)} = [C_H \times F_H \times I_R] / \text{body weight} \quad (1)$$

where  $C_H$  is the concentration of target contaminant in dust ( $\text{ng g}^{-1}$ ),  $F$  = fractional time spent at home,  $I_R$  = ingestion rate for dust ( $\text{mg per day}$ ). Average fractional time spent by adults and toddlers at home were assumed to be 0.58 (58%) and 0.92 (92%) respectively, while adult and toddler body weights were assumed to be 60 and 10 kg respectively.<sup>27,28</sup> Exposure *via* house dust ingestion was calculated under eight scenarios: (a) assuming typical dust ingestion rates of 20 mg per day for adults

and 50 mg per day for toddlers<sup>28</sup> and that ingested dust contained each contaminant at the 5<sup>th</sup> percentile, arithmetic mean, median, and 95<sup>th</sup> percentile concentration; and (b) assuming high dust ingestion rates of 50 mg per day for adults and 200 mg per day for toddlers<sup>28</sup> and that ingested dust contained each contaminant at the 5<sup>th</sup> percentile, arithmetic mean, median, and 95<sup>th</sup> percentile concentration.

# 3. Results

## 3.1 Concentrations of the target chemicals in domestic indoor dust from Lagos, Nigeria

### 3.1.1 Concentrations of PBDEs and HBCDDs in dust from Nigerian homes.

The concentrations of PBDEs and HBCDD measured in indoor dust in this study are summarised in Fig. 1 and Table 1 respectively. Those of  $\sum_8\text{BDEs}$  ranged from  $43 \text{ ng g}^{-1}$  to  $810 \text{ ng g}^{-1}$  average =  $310 \text{ ng g}^{-1}$ . As shown in Fig. 1, BDE-209 was the dominant PBDE, comprising on average 87% of  $\sum_8\text{BDEs}$ , with the relative abundance being: BDE-209 (average concentration =  $290 \text{ ng g}^{-1}$ ) > BDE-99 (average concentration =  $6.6 \text{ ng g}^{-1}$ ) > BDE-47 (average concentration =  $5.8 \text{ ng g}^{-1}$ ) > BDE-183 (average concentration =  $4.5 \text{ ng g}^{-1}$ ) > BDE-153 (average concentration =  $2.4 \text{ ng g}^{-1}$ ) > BDE-154 (average concentration =  $1.6 \text{ ng g}^{-1}$ ) > BDE-100 (average concentration =  $1.3 \text{ ng g}^{-1}$ ). BDE-28 was not detected in any sample ( $< 0.78 \text{ ng g}^{-1}$ ). This dominance of BDE-209 – which has been reported elsewhere<sup>16,23,29,30</sup> – suggests decaBDE is the principal PBDE formulation used in the products in the Nigerian homes sampled. Interestingly, while the detection frequency for BDE-209 and BDE-47 was 93% and that for BDE-99, 86%;

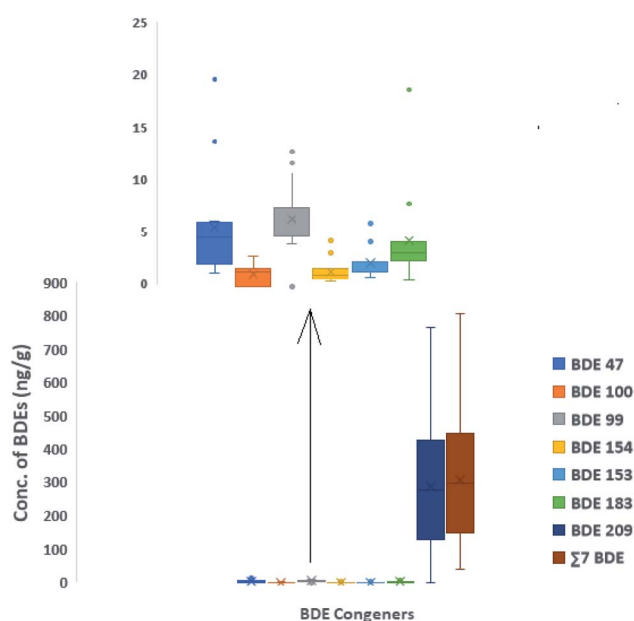


Fig. 1 Concentrations of target BDE congeners in Lagos home settled dust. \* – the boxes indicate the 25<sup>th</sup> and 75<sup>th</sup> percentiles while the bars and the “x” in the box indicate the median and mean concentration respectively. The whiskers indicate the maximum and minimum concentrations, and the ‘dots’ show the outliers.





Table 1 Concentrations of HBCDD in Lagos home dust

	Concentration (ng g <sup>-1</sup> )			
	$\alpha$ -HBCDD	$\beta$ -HBCDD	$\gamma$ -HBCDD	$\Sigma$ HBCDD
Mean	5.7	<11	5.1	11
Median	<5.3	<11	<2.4	<dl
Range	<5.3–40	<11	<2.4–19	<dl – 66

congeners commonly associated with the octaBDE formulation – *i.e.* BDE-153, 154, and 183 were detected in all samples (Table S5a†).

Concentrations of  $\Sigma$ HBCDD range from <2.4 to 66 ng g<sup>-1</sup> (mean = 11, median = <2.4) (Table 1), with HBCDD detected in only 4 samples. Although HBCDD has some minor applications in consumer products such as electronics and furniture fabric covers, its predominant application is as an additive to polystyrene building thermal insulation foam.<sup>24,31</sup> Hence, the low concentrations of HBCDD detected here are unsurprising given the tropical climate of Lagos. With respect to the diastereomer pattern,  $\gamma$ -HBCDD was observed in four samples,  $\alpha$ -HBCDD was observed in two with  $\beta$ -HBCDD not detected in any samples.

**3.1.2 Concentration of alt-HFRs in dust from Nigerian homes.** Concentrations of alt-HFRs are presented in Fig. 2 and S7b.† Those of  $\Sigma$ alt-HFRs are in the range 32–2600 ng g<sup>-1</sup> (mean = 560, median = 320). DBDPE was the dominant alt-HFR targeted, on average comprising 68% of  $\Sigma$ alt-HFRs (Table S7c†). Average concentrations of the alt-HFRs targeted are: DBDPE (480 ng g<sup>-1</sup>) > *anti*-DP (36 ng g<sup>-1</sup>) > PBEB (27 ng g<sup>-1</sup>) >

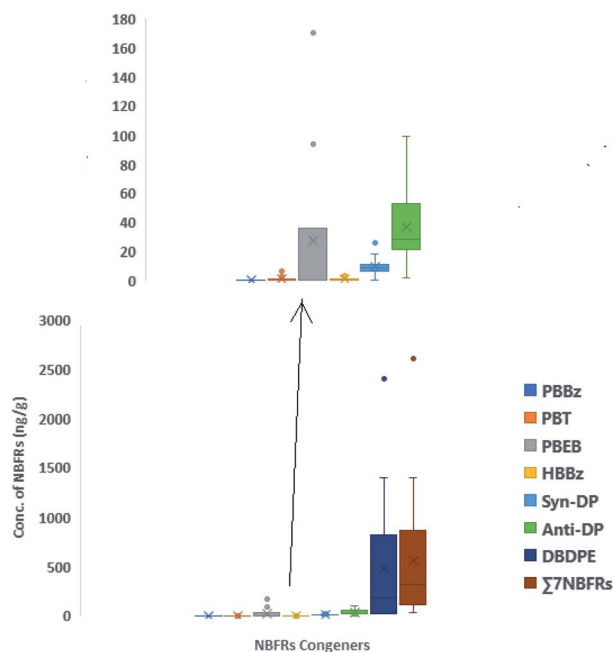


Fig. 2 Concentrations of target alt-HFRs in Lagos home settled dust. \* – the boxes indicate the 25<sup>th</sup> and 75<sup>th</sup> percentiles while the bars and the “x” in the box indicate the median and mean concentration respectively. The whiskers indicate the maximum and minimum concentrations, and the ‘dots’ show the outliers.

*syn*-DP (9.1 ng g<sup>-1</sup>) > PBT (0.96 ng g<sup>-1</sup>) > HBBz (0.93 ng g<sup>-1</sup>) > PBBz (0.1 ng g<sup>-1</sup>). PBBz was only found only in one sample while *syn*-DP and *anti*-DP display the highest detection frequencies of 93% and 87% respectively. The target alt-HFRs are all classified by the USEPA as low production volume chemicals, with the exceptions of DBDPE and DP which are classified as high production.<sup>3,32</sup> This is consistent with the relative abundance in this study of both DBDPE and *anti*-DP. The fractional contribution of *anti*-DP to  $\Sigma$ DP ( $f_{anti-DP}$ ) fell in the range 0.26–0.96 (mean = 0.76, median = 0.79). Similar wide variations have been documented previously.<sup>33</sup>

### 3.1.3 Concentration of PCBs in dust from Nigerian homes.

Concentrations of target PCBs detected in indoor dust in this study are summarised in Fig. 3 and S8b,† with those of  $\Sigma_7$ PCBs ranging from 3.8 ng g<sup>-1</sup> to 61 ng g<sup>-1</sup> (mean = 22 ng g<sup>-1</sup>, median = 18) ng g<sup>-1</sup>. Likely reflecting their comparatively high vapour pressures, neither PCB-28 or the non-arochlor PCB-11 were detected in any sample. Of the remaining target congeners, only PCBs 138 and 180 were detected in all samples, while average concentrations were in the order: PCB-138 (9.0 ng g<sup>-1</sup>) > PCB-180 (4.5 ng g<sup>-1</sup>) > PCB-153 (3.7 ng g<sup>-1</sup>) > PCB-52 (2.8 ng g<sup>-1</sup>) > PCB-118 (1.0 ng g<sup>-1</sup>) > PCB-101 (0.55 ng g<sup>-1</sup>).

## 3.2 Comparison of concentrations of target contaminants in dust from Nigerian homes with similar reports elsewhere

**3.2.1 PBDEs and HBCDD.** Table 2 compares the concentrations of PBDEs and HBCDD detected in this study with those from selected other studies in home dust. Concentrations in our study were lower than those reported previously for Nigeria,<sup>16,19,20</sup> with the exception that concentrations of BDE-209 in this study exceeded those reported.<sup>19</sup> This may suggest that restrictions on the manufacture and use of the penta- and octa-BDE formulations have lowered concentrations of associated PBDE congeners like BDE-47, BDE-99, BDE-153, BDE-154, and BDE-183. However, concentrations of BDE-209 have declined from the levels reported in Harrad *et al.*,<sup>16</sup> but not compared to those reported by Olukunle *et al.*<sup>19</sup> This is consistent with the

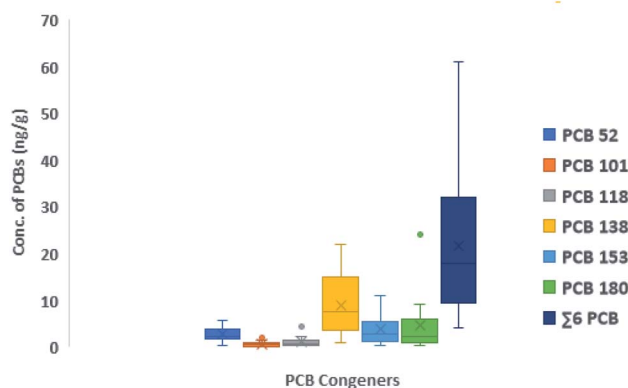


Fig. 3 Concentrations of target PCB congeners in Lagos home settled dust. \* – the boxes indicate the 25<sup>th</sup> and 75<sup>th</sup> percentiles while the bars and the “x” in the box indicate the median and mean concentration respectively. The whiskers indicate the maximum and minimum concentrations, and the ‘dots’ show the outliers.



Table 2 Comparison of concentrations of PBDEs and HBCDDs in domestic indoor dust from this study with those reported elsewhere<sup>a</sup>

Location	Sampling year(s) (sample number)	Concentration (ng g <sup>-1</sup> ) – range (mean, median)									
		BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	∑ <sub>8</sub> PBDEs	∑ HBCDD
Lagos, Nigeria (this study)	2019 (n = 15)	<0.79	<1.5–20 (5.8, 4.8)	<0.19–13 (6.6, 6.9)	<0.21–3.1 (1.3, 1.5)	1–6.2 (2.4, 1.9)	0.64–4.6 (1.6, 1.2)	0.71–19 (4.5, 3.4)	<0.16–770 (290, 280)	43–810 (310, 300)	<2.4–66 (11, <2.4)
Lagos, Nigeria <sup>16</sup>	2014 (n = 12)	<0.04–3.1 (1, 0.5)	2.2–50 (13, 8)	1.5–31 (17, 14)	<0.03–44 (8, 4)	<0.04–22 (5, 4)	<0.04–3500 (450, 4)	2.9–90 (26, 18)	77–940 (420, 390)	—	—
Lagos, Nigeria <sup>20</sup>	2014 (n = 10)	—	—	—	—	—	—	—	—	—	41–1900 (659, 394)
Makurdi, Nigeria <sup>19</sup>	2012 (n = 10)	—	26–51 (43, 47)	0.24–62 (99, 46)	0.1–56 (39, 43)	0.03–74 (39, 50)	0.03–72 (35, 35)	38.8–85 (62, 30)	79–202 (141, 139)	#45	—
Cairo, Egypt <sup>17</sup>	2013 (n = 17)	0.15–4 (0.7, 0.3)	0.34–380 (29, 1.7)	0.53–510 (35, 2.7)	0.07–98 (7.2, 0.4)	0.54–200 (28, 6.3)	<0.04–7.9 (4.9, 0.4)	0.27–5.2 (1.7, 1.1)	2.2–590 (130, 40)	5.0–1900 (250, 57)	(21, 6.2)
Pretoria, South Africa <sup>18</sup>	2010–2011 (n = 31)	—	#2.6	2.6	<0.13	<0.13	<0.13	—	<1.8	—	—
Durban, South Africa <sup>5</sup>	2012 (n = 10)	1.1–30 (15, 12)	56–470 (180, 160)	150–1100 (540, 510)	<dI – 150 (47, 37)	34–180 (78, 65)	<dI – 130 (76, 97)	5.6–110 (44, 45)	59–2200 (730, 660)	690–3900 (1700, 1600)	—
Birmingham, UK <sup>23</sup>	2020 (n = 14)	<0.1	<0.1–37 (6, 0.93)	<0.1–61 (11, 2.9)	<0.1–6.9 (1.5, 0.29)	<2.0	<2.0	<2.0	21–18 000 (4800, 1600)	21–18 000 (4800, 1700)	76–570 000 (460 000, 280)
Annecy, France <sup>20</sup>	2008 (n = 9)	—	—	—	—	—	—	—	—	—	360–1900 (1200, 1400)
Australia <sup>34</sup>	2009–2011 (n = 30)	<dI – 10) 0.7	(2.6–390) 37	(2.9–170) 57	(0.52–71) 9.1	<dI – 60) 6.4	<dI – 32) 5.1	(0.53–44) 4.6	<dI – 82 000) *420	—	—
Ireland <sup>30</sup>	2016–2017 (n = 26)	—	0.6–240 (26, 8)	<0.2–500 (45, 13)	—	—	—	<0.3–33 (4, 1)	140–650 000 (58 000, 1300)	—	1.3–430 000 (2900, 490)
Stockholm, Sweden <sup>35</sup>	2006 (n = 10)	<0.1–5.6 (#1.3)	<0.5–230 (42)	<1–140 (52)	—	0.61–23 (6.6)	—	<0.7–49 (12)	53–4000 (510)	—	15–990 (100)
Beijing, China <sup>3</sup>	2014 (n = 30)	0.34–3.8 (1.1, 0.68)	0.67–4.2 (1.6, 1.3)	1.2–25 (6.3, 4.9)	<dI – 1.9 (0.37, 0.44)	0.27–3.5 (1.4, 0.89)	0.25–1.6 (0.70, 0.53)	0.21–3.7 (0.84, 0.37)	69–410 (190, 150)	90–430 (200, 160)	110–390 (253, 258)
Toronto, Canada <sup>29</sup>	2006 (n = 10)	1.4–20 (6.6, 4.1)	47–20 (300, 140)	80–1800 (510, 330)	14–420 (120, 65)	9.4–260 (71, 43)	6.2–280 (69, 39)	7–30 (13, 9)	290–1100 (670, 560)	160–3600 (1100, 620)	—

<sup>a</sup> dI – detection limit, # – median, \* – mean.

Table 3 Comparison of concentrations of alt-HFRs in domestic indoor dust from this study with those reported elsewhere<sup>a</sup>

Location	Sampling period (sample number)	Concentrations (ng g <sup>-1</sup> ) – range (mean, median)									
		PBBz	PBT	PBEB	HBBz	syn-DP	anti-DP	DBDPE			
Lagos, Nigeria (this study)	2019 (n = 15)	<0.7–0.89 (0.10, <0.7)	<0.59–6.3 (0.96, <0.59)	<0.71–170 (27, <0.71)	<0.76–3.5 (0.93, <0.76)	<0.1–26 (9.1, 8.7)	<2.4–99 (36, 28)	<54–2400 (480, 180)			
Birmingham, UK <sup>23</sup>	2020 (n = 14)	—	—	—	—	—	—	<50–7500 (1500, 660)			
Ireland <sup>30</sup>	2016–2017 (n = 29)	—	—	—	—	—	—	410–460 000 (39 000, 4200)			
Melbourne, Australia <sup>26</sup>	2016 (n = 24)	—	<dl – 8.0	<dl	<dl – 4.6 (1.0, 0.71)	—	—	<dl – 9000 (2000, 1600)			
Birmingham, UK <sup>36</sup>	2013–2015 (n = 30)	—	<0.01–90 (7.1, 1.8)	<0.01–21 (2.3, 0.78)	—	—	—	<1.2–2300 (240, 41)			
Alexandria, Egypt <sup>37</sup>	2014 (n = 12)	<dl – 0.20 (0.10, 0.08)	<dl–0.08 (<dl, <dl)	<dl – 0.10 (<dl, <dl)	<dl – 0.70 (0.30, 0.29)	—	—	—			
Beijing, China <sup>13</sup>	2014 (n = 30)	—	<dl – 4.9 (1.1, 0.72)	<dl – 2.1 (0.53, 0.20)	0.10–5.4 (2.0, 1.4)	—	—	220–3000 (770, 560)			
Basrah, Iraq <sup>38</sup>	2013 (n = 18)	—	—	<0.1–0.55 (0.14, <0.1)	—	—	—	33–270 (130, 130)			
Oslo, Norway <sup>39</sup>	2013–2014 (n = 60)	—	<5.2–250	<0.37–12	<0.21–110	—	—	11–430			
Cairo, Egypt <sup>17</sup>	2013 (n = 17)	—	—	—	0.1*	0.3*	0.01*	—			
Kuwait <sup>21</sup>	2011 (n = 15)	—	—	—	0.2–3.6 (1.4, 1.2)	—	—	40–2200 (510, 220)			
Antwerp, Belgium <sup>21</sup>	2008 (n = 39)	—	—	—	—	—	—	55–2100 (300, 150)			
Hanoi, Vietnamese <sup>41</sup>	2008 (n = 6)	—	—	—	—	—	—	17–150 (median = 40)			
Vancouver, Canada <sup>33</sup>	2007–2008 (n = 116)	—	—	<0.1–4.1 (**0.5)	<0.02–130 (6.8, 3.7)	<0.70–170 (11, 0.9)	<0.70–170 (11, 4)	—			

<sup>a</sup> dl – detection limit, \* – median, \*\* – mean.

Table 4 Comparison of concentrations of PCBs in domestic indoor dust from this study with those reported elsewhere<sup>a</sup>

Location	Sampling year (sample number)	Concentration (ng g <sup>-1</sup> ) - range (mean, median)							
		PCB-28	PCB-52	PCB-101	PCB 118	PCB-138	PCB-153	PCB-180	∑ <sub>7</sub> PCBs
Lagos, Nigeria (this study)	2019 (n = 15)	<0.31	<0.49–7.6 (2.8, 2.1)	<0.32–1.9 (0.55, 0.51)	0.59–4.2 (1.0, 0.62)	0.82–22 (9.0, 7.5)	<0.46–11 (3.7, 2.8)	0.27–24 (4.5, 2.2)	3.8–61 (22, 18)
Lagos, Nigeria <sup>16</sup>	2014 (n = 12)	1.5–7.8 (3.8, 3.9)	0.8–8.6 (4.2, 4.5)	0.4–9.4 (2.7, 3.7)	—	0.3–22 (6.0, 7.5)	0.2–22 (5.8, 1.3)	4.4–24 (10, 10)	—
Durban, South Africa <sup>15</sup>	2012 (n = 10)	16, 11	—	—	—	—	173, 150	702, 585	—
Kuwait <sup>21</sup>	2011 (n = 15)	—	—	<0.2–835 (63, <0.2)	<0.2–560 (35, <0.2)	<0.1–435 (28, <0.1)	<0.2–310 (20, <0.2)	<0.1–105 (7, 0.4)	—
Faisalabad, Pakistan <sup>21</sup>	2011 (n = 15)	—	—	<0.2	<0.2–0.8 (<0.2, 0.2)	<0.1–2.2 (0.5, <0.1)	<0.1–4.0 (0.8, 0.4)	<0.1–11 (1.2, 0.3)	—
Texas, US <sup>28</sup>	2006 (n = 20)	*1.6–37 (9.5, 5.1)	1.7–28 (7.9, 6.2)	1.9–29 (10, 8.7)	1.8–44 (10, 5.5)	1.1–31 (8.6, 6.5)	1–22 (8.4, 7.1)	0.7–20 (4.5, 2.6)	—
Birmingham, UK <sup>28</sup>	2006 (n = 20)	*0.5–39 (6.3, 3.9)	0.3–53 (5.6, 1.8)	0.1–73 (6.1, 1.2)	0.06–56 (4.3, 0.92)	0.1–50 (4.1, 1.1)	0.1–32 (3.3, 1.2)	0.1–8.1 (1.8, 0.89)	—
Toronto, Canada <sup>28</sup>	2006 (n = 10)	*3.5–29 (10, 7.3)	3.4–37 (12, 7.2)	1.9–60 (15, 8.8)	1.1–55 (13, 8.7)	1.9–60 (15, 8.8)	1–49 (12, 9.5)	0.9–36 (11, 9.9)	—
Wellington, New Zealand <sup>28</sup>	2006 (n = 20)	*0.8–11 (3.3, 2.3)	0.4–13 (2.6, 1.4)	0.4–21 (3.1, 1.6)	<dl – 14 (1.9, 0.95)	0.4–21 (3.1, 1.6)	0.3–11 (2.8, 1.8)	0.4–12 (2.7, 1.4)	—
Hanoi, Vietnamese <sup>41</sup>	2008 (n = 6)	0.18–0.59 (**0.49)	0.17–2.7 (0.40)	0.35–3.3 (0.93)	0.38–8.4 (0.64)	0.09–1.2 (0.36)	0.62–18 (1.2)	0.18–2.3 (0.31)	—
Kuwait <sup>40</sup>	n = 15 (2011)	—	—	<0.2–840 (63, <0.2)	<0.2–560 (35, <0.2)	<0.2–310 (20, <0.2)	<0.1–440 (28, <0.1)	0.1–110 (7, 0.4)	—
Canada <sup>42</sup>	n = 16 (2013)	—	—	—	—	—	—	—	<dl – 521 (69.1, <dl)
Czech Republic <sup>42</sup>	n = 28 (2013)	—	—	—	—	—	—	—	11–360 (79, 75)

<sup>a</sup> dl – detection limit, \* PCBs – 28 + 31, \*\* – median.

more recent restrictions on the manufacture and use of the deca-BDE commercial product. In general, comparing with data obtained further afield in other locations in Africa and elsewhere, concentrations in our study are very much at the low end of those reported. Overall, the available data suggest that PBDE and HBCDD use in Nigerian homes is lower than in many other countries.

**3.2.2 Alt-HFRs.** To our knowledge, this is the first report of concentrations of alt-HFRs in indoor dust in sub-Saharan Africa. We nonetheless compare our data for these contaminants with those reported in other locations (Table 3). In

keeping with studies elsewhere, DBDPE is the dominant alt-BFR in our study, and in general, concentrations of our other target alt-HFRs fall within the range reported previously in other locations.

**3.2.3 PCBs.** Concentrations detected in this study together with those from selected similar studies are summarised in Table 4. Concentrations of individual PCBs are a little lower but within the range reported previously for homes in Lagos, Nigeria.<sup>16</sup> In general, concentrations in this study are at the lower end of those reported previously elsewhere, possibly reflecting a general temporal decline in PCB contamination as

Table 5 Estimated exposure of toddlers to BFRs and PCBs (ng per kg bw per day)<sup>a</sup>

Compounds	Toddlers							
	Average dust ingestion				High dust ingestion			
	5 <sup>th</sup>	Mean	Median	95 <sup>th</sup>	5 <sup>th</sup>	Mean	Median	95 <sup>th</sup>
BDE-209	0.22	1.3	1.3	2.8	0.87	5.3	5.0	11
DBDPE	0.13	2.2	0.82	7.74	0.53	8.8	3.2	31
∑HBCDD	<0.0006	0.0052	<0.0006	0.028	0.024	0.20	0.024	1.0
∑alt-HFRs	0.15	2.5	1.4	8.1	0.61	10	5.8	32
∑PCBs	0.024	0.098	0.083	0.25	0.097	0.38	0.33	0.98
∑PBDEs	0.36	1.4	1.3	2.9	1.4	5.7	5.4	11

<sup>a</sup> ∑alt-HFRs = DBDPE + anti-DP + syn-DP + PBEB + PBT + HBBz + PBBz, ∑BDEs = BDE 47 + BDE 99 + BDE 100 + BDE 153 + BDE 154 + BDE 183 + BDE 209, ∑PCBs = PCB 52 + PCB 101 + PCB 118 + PCB 138 + PCB 153 + PCB 180.





Table 6 Estimated exposure of adults to BFRs and PCBs (ng per kg bw per day)<sup>a</sup>

Compounds	Adults							
	Average dust ingestion			High dust ingestion				
	5 <sup>th</sup>	Mean	Median	95 <sup>th</sup>	5 <sup>th</sup>	Mean	Median	95 <sup>th</sup>
BDE-209	0.0093	0.056	0.053	0.12	0.023	0.14	0.13	0.29
DBDPE	0.0056	0.093	0.035	0.33	0.014	0.23	0.087	0.82
$\sum$ HBCDD	<0.0003	0.0021	<0.0003	0.011	<0.0006	0.0052	<0.0006	0.028
$\sum$ alt-HFRs	0.0065	0.11	0.061	0.34	0.016	0.27	0.15	0.86
$\sum$ PCBs	0.001	0.004	0.004	0.01	0.003	0.01	0.009	0.026
$\sum$ PBDEs	0.015	0.060	0.057	0.12	0.038	0.15	0.14	0.30

<sup>a</sup>  $\sum$ alt-HFRs = DBDPE + anti-DP + syn-DP + PBEB + PBT + HBBz + PBBz,  $\sum$ BDEs = BDE 47 + BDE 99 + BDE 100 + BDE 153 + BDE 154 + BDE 183 + BDE 209,  $\sum$ PCBs = PCB 52 + PCB 101 + PCB 118 + PCB 138 + PCB 153 + PCB 180.

samples reported previously were collected 6–13 years before those in this study. Interestingly, concentrations of PCB 28, 153, and 180 detected in 10 house dust samples collected in 2012 in Durban, South Africa<sup>15</sup> exceed substantially (by up to two orders of magnitude) those found in Lagos in our study. Notwithstanding likely temporal declines in PCB contamination, this suggests lower PCB use in Nigeria than South Africa.

### 3.3 Implication of exposure risks of BFRs and PCBs in Lagos home settled dust

Estimated exposures to our target compounds *via* ingestion of indoor dust were calculated for a range of scenarios as outlined in Section 2.6. The resulting estimates for toddlers and adults are summarised in Tables 5 and 6 respectively. In general, the order of exposures for both age groups is:  $\sum$ alt-HFRs >  $\sum$ PBDEs >  $\sum$ PCBs > HBCDD. DBDPE and BDE-209 dominate estimated exposures for  $\sum$ alt-HFRs and  $\sum$ PBDEs respectively.

The USEPA<sup>43</sup> have published reference dose (RfD) values for: BDE 209 (7000 ng per kg per day), BDE 47 (100 ng per kg per day), and BDE 99 (100 ng per kg per day). These RfD values substantially exceeded the exposure levels observed in this study.

## 4. Conclusion

This study reports concentrations of BFRs and PCBs in floor dust samples from homes in Lagos, Nigeria. The general order of concentrations of our target chemicals was:  $\sum$ alt-HFRs >  $\sum$ BDEs >  $\sum$ PCBs > HBCDD. DBDPE and BDE 209 were the dominant alt-HFR and PBDE respectively. The concentrations of PBDEs and PCBs reported are below those reported previously for house dust from Lagos, Nigeria, suggesting that restrictions on the manufacture and use of such chemicals have had beneficial effects. However, while concentrations of BDE-209 in this study were lower than in one previous study in Lagos, they exceeded those in another; this suggests that the more recent restrictions on the deca-BDE product have yet to lead to a consistent reduction in environmental concentrations of BDE-209. To our knowledge, we report the first evidence of the presence of alt-HFRs in house dust from Africa; these data will provide a valuable benchmark against which possible

future trends in concentrations of these replacements for banned BFRs may be evaluated.

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## Author contributions

The manuscript was written through the contributions of all authors. All authors have given approval to the final version of the manuscript. OEA and WS designed and performed the experiments, drafted, and revised the manuscript. SH designed the experiments, drafted, revised, and edited the manuscript. MAA drafted, revised, and edited the manuscript. OA and RA revised, and edited the manuscript.

## Conflicts of interest

The authors declare no conflicts of interest.

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