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Folarin, Bilikis Temitope; Abdallah, Mohamed Abou-Elwafa; Oluseyi, Temilola; Olayinka, Kehinde; Harrad, Stuart

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CONCENTRATIONS OF POLYCHLORINATED BIPHENYLS IN SOIL AND INDOOR DUST ASSOCIATED WITH ELECTRICITY GENERATION FACILITIES IN LAGOS, NIGERIA

Bilikis Temitope Folarin\textsuperscript{1, 2}, Mohamed Abou-Elwafa Abdallah\textsuperscript{2}, Temilola Oluseyi\textsuperscript{1}, Kehinde Olayinka\textsuperscript{1}, Stuart Harrad\textsuperscript{2}* 

\textsuperscript{1}Department of Chemistry, University of Lagos, Lagos, Nigeria

\textsuperscript{2}School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham, United Kingdom

*Corresponding author

S.J.Harrad@bham.ac.uk
Abstract

Concentrations of 7 indicator polychlorinated biphenyls (PCBs) were measured in dust and soil samples from 12 power stations collected over the two major seasons of the Nigerian climate. Median ΣPCB₇ concentrations in soil ranged from 2 ng/g for power station A to 220 ng/g for power station I; while those in dust ranged from 21 ng/g for power station L to 2,200 ng/g for power station I. For individual congeners, median PCB concentrations ranged from 3.8 ng/g for PCB 101 to 52 ng/g for PCB 180 in dust, and <0.07 ng/g for PCB 28 to 5.9 ng/g for PCB 153 in soil. The type of power station activity exerted a significant influence on concentrations of ΣPCB₇ in dust and soil (generation > transmission > distribution). Congener patterns in dust and soil samples were compared using principal component analysis (PCA) with those in transformer oil samples from 3 of the power stations studied and with common PCB mixtures (Aroclors). This revealed congener patterns in soil were more closely related to that in the transformer oil than dust. Congener patterns in most samples were similar to Aroclor 1260. Concentrations of PCBs in soil samples close to the transformers significantly exceeded those in soil sampled further away.

Keywords

PCBs; power generation plants; Africa; transformers

1. Introduction
Polychlorinated biphenyls (PCBs) are a class of industrial chemicals listed as persistent organic pollutants (POPs) under the Stockholm convention (UNEP, 2009). Prior to their restriction, PCB mixtures had been used extensively as dielectric fluids in electrical appliances and power generating plants as well as additives in many other non-electrical products in everyday use (Xing et al., 2011; Breivik et al., 2002).

The large-scale production and usage of PCBs, has resulted in widespread contamination of the environment (Harrad et al., 1994). Efforts to gradually eliminate the presence of these toxic, ubiquitous chemicals, led to ban on their production (Robinson, 2009; Koppe and Keys, 2002). However, over four decades post-restriction, PCBs remain in circulation within the global environment, due to their persistent nature (Gouin et al., 2005). Moreover, PCBs continue to be released into the environment, for example through spillage from transformers during maintenance and day-to-day operations in power stations (Tue et al., 2016; Xing et al., 2011).

The soil is a primary recipient of many outdoor contaminants (Domotorova et al., 2012). PCBs have been detected in soil around power plants (generating, transmitting and distribution transformers) and other e-waste sites in China (Tang et al., 2010; Shen et al., 2008). Meanwhile, reported sources of PCBs in indoor environments include paints, building sealants and adhesives. PCB fumes can be released from hot electrical gadgets, wires and cables, since PCBs have been used as components of these products because of their excellent insulating properties (Xing et al., 2011; Kohler et al., 2005).

The high $K_{OW}$ values of PCBs increases their tendency to bind to dust and soil (Kuusisto et al., 2007). Indoor dust has been reported as a major sink of PCBs and other pollutants in indoor environments (Wang et al., 2015; Whitehead et al., 2011), leading to exposure via inhalation, ingestion and/or dermal uptake (Harrad et al., 2006; Jones-Otazo et al., 2005; Freels et al., 2007).
To the best of our knowledge, there are only two reports of PCB concentrations in indoor dust in Africa. Firstly, Abafe et al., 2015 measured concentrations of three PCB congeners (PCB 28, 153, and 180) in indoor dust samples from ten homes, eleven offices, and thirteen computer laboratories in Durban, South Africa. The second report measured six indicator PCBs in dust samples from three microenvironment categories (homes, cars and offices) in Nigeria (Harrad et al., 2016). The present study measures concentrations of PCBs in dust and soil from the vicinity of power stations in Nigeria to test our hypothesis that such stations are a source of PCBs to the surrounding environment. This work reports for the first time, the concentrations of PCBs in dust samples from power stations in Nigeria and provides only the second report on PCBs in Nigerian dust.

Concentrations of PCBs in soil from non-African countries have been extensively reported. Vane et al., 2014 (UK); and Jiang et al., 2011 (China), reported concentrations of PCBs in soil from urban areas, while Melynk et al., 2015 (Poland) reported PCBs in landfill soil. Concentrations of PCBs in industrial and agricultural soils were also compared in Pakistan by Syed et al., 2013. The few reports on PCBs in African soil are from South Africa (Batterman et al., 2009); Kenya (Sun et al., 2016); Ghana (Tue et al., 2016) and Nigeria (Alani et al., 2013). All these African reports are centred around PCBs in soil from urban areas, except for Tue et al., 2016 that measured dioxin-like PCBs and other dioxin related compounds released from open burning of e-waste in Ghana. The current study is the first investigation of PCBs in soil in the vicinity of power stations in Nigeria.

Against this backdrop, this paper reports the concentrations of PCBs in 48 office dust and 48 soil samples collected within and in the vicinity of 12 power stations in Lagos, Nigeria (6 power distribution stations, 5 power transmission stations, and 1 former generation station within Lagos, Nigeria). These data are used to: (i) test the hypothesis that power stations in Nigeria constitute a source of PCBs to the surrounding environment; (ii) evaluate the effect of
seasonal variation (wet and dry seasons) on concentrations of PCBs in soil and dust samples; and (iii) compare the pattern of PCBs in transformer oil samples with dust and soil samples.

2. Materials and Methods

2.1 Sample collection

A total of 48 dust samples and 48 soil samples were collected from twelve power stations in Lagos, Nigeria in the months of June 2015 (24 dust samples and 24 soil samples) and December 2015 (24 dust samples and 24 soil samples). Surface dusts were collected from two offices within each power station while soil samples were collected from two points for each power station. The first point was 1-3 m and the second sampling point 50-100 m away from the transformer plants. Collection of top soil (0 - 5 cm depth) was done using a hand trowel, which was washed and pre-cleaned with hexane before each sampling. Following collection, each sample was passed through a 2 mm sieve, wrapped in aluminium foil and stored at −20 °C in individual zip lock bags.

Dust samples were collected from elevated surfaces (e.g. shelves and air conditioning panels, electrical control panels etc) using a paintbrush which had been washed and pre-cleaned with hexane before each sampling, and wrapped in aluminium foil. Similar to soils, dust samples were kept in individual zip lock bags and stored at −20 °C. Both soil and dust samples were transported to the University of Birmingham for analysis. At Birmingham, the dust samples were sieved with a 500 µm sieve, using procedures similar to Harrad et al. (2016). In Birmingham, both soil and dust samples were stored at 4 °C prior to analysis.

Transformer oil samples were collected from three of the studied power stations. Two were used transformer oil while the third one was a fresh unused oil. Oil sample 1 was collected from Site I and J, sites that are linked - one being a former generation station (I) and the other
a transmission station (J). The other used oil was from site H, while the unused oil was from site K.

2.2 Chemicals and materials

Individual indicator PCB standards (#s 28, 52, 101, 118, 138, 153, 180), and 5 native internal standards (ISs) (PCB 34, 62, 119, 131 and 173) were purchased from LGC standards, UK. These native PCB congeners are absent in environmental samples and have been previously reported to be suitable for use as internal standards (Ayris et al., 1997). Recovery determination standard (RDS), PCB 129 was from QMX laboratories, UK; HPLC grade solvents (DCM, n-Hexane) were from Fisher Scientific, UK. Analytical grade reagents purchased from Sigma Aldrich, UK were: anhydrous sodium sulfate, which was baked at 450 °C for 4 hours before use and florisil (60-100 mesh) activated at 120 °C for 2 hours.

2.3 Sample extraction and clean up

Accurately weighed dust (approx. 200 mg) and soil (between 1-3 g) samples were carefully introduced into glass extraction tubes that had prior to use been washed, oven dried for several hours, and thoroughly rinsed with extraction solvent (1:1 (v/v) hexane: DCM). A known amount of internal standard was added prior to addition of 2 mL and 5 mL extraction solvent (1:1 (v/v) hexane: DCM) for dust and soil respectively. The mixture was vortexted for about 1 minute before ultrasonic extraction for 15 minutes and 30 minutes for dust and soil samples respectively. The supernatant was separated from the solute by centrifugation at 3500 rpm for 5 mins. The extraction was repeated twice for each sample using fresh solvent for each repeat extraction. Extracts were combined and blown down to dryness under a gentle stream of nitrogen.
Concentrated crude extracts were reconstituted in 2 mL hexane and shaken with 2 mL concentrated H$_2$SO$_4$, to eliminate lipids. The organic layer was removed and concentrated to about 1 mL before passing through a bed of florisil, pre-conditioned with 6 mL hexane, and the PCBs on the florisil column were eluted with 20 mL hexane. The eluate was reduced under N$_2$ to incipient dryness and reconstituted in 150 µL recovery determination standard in iso-octane. While for the majority of samples, this relatively simple clean-up was sufficient; for some samples, DMSO clean up was introduced before florisil clean up to eliminate aliphatic hydrocarbons which were major interferences by introducing 2 mL DMSO to 1 mL acid cleaned extract (in hexane). In such instances, the mixture was vortexed for 1 minute and then centrifuged at 3500 rpm for 2 minutes to facilitate removal of the top hexane aliphatic-containing layer. To the DMSO aromatic-containing layer, 1 mL water was added, and the mixture vortexed to break down the DMSO charge transfer complex. The aqueous DMSO mixture was then back extracted with 4 $\times$ 1 mL hexane. These hexane back extracts were combined and blown down ready for florisil clean up as described above.

For transformer oil samples, 50 µL (approx. 40 mg) of oil was diluted with 1 mL hexane and cleaned up using a sequential combination of sulfuric acid, DMSO, and florisil clean up methods as described above.

2.4 Instrumental Analysis

Target PCBs were quantified in calibration standard and sample extracts on an Agilent 5975C GC-MSD, fitted with a RestekRXi-PAH capillary column (40 m × 180 µm × 0.01 µm) using the GC and MS parameters stated in the supplementary material (Table S1). The GC inlet temperature was set to 280 ºC, injection was performed in splitless mode. A carrier gas (Helium) flow of 0.5 mL/min was employed. The initial oven temperature was set at 140 ºC and held for 2 mins, ramped at 5 ºC/min to 215 ºC, held for 5 mins and thereafter ramped at 2
°C/min to 245 °C. The temperatures of the quadrupole and ion source were 150 °C and 230 °C respectively.

The target PCBs monitored belong to 5 PCB homologues. To quantify target PCBs (PCB # - 28, 52, 101, 118, 138, 153, and 180), internal standard quantification was employed. PCB # - 34, 62, 119, 131, and 173 were used as internal standards for the tri CB, tetra-CB, penta-CB, hexa-CB, and hepta-CB respectively. The MS was programmed in the selected ion monitoring (SIM) mode, with the two most abundant ions monitored for each PCB homologue. The masses of the 2 ions monitored and their percentage abundance are presented in Table S2 (supplementary material).

2.5 QA/QC

Two standard reference materials (SRM 2585, NIST organics in indoor dust and CRM 481, PCBs in industrial soil) were analysed for method validation. As a measure of precision, 6 replicate analysis of PCBs in SRM 2585 and CRM 481, extracted and cleaned-up as described above, gave a RSD ranging from 4 % to 13 % and 5 % to 10 % for dust and soil reference materials respectively. Concentrations of PCBs in these reference materials obtained using our methods were deemed acceptably similar to the certified values (Tables S3 and S4). Mean recoveries of internal standards in SRM2585 ranged from 80 % to 94 % and are provided in table S5. Analyses of method blanks were carried out once after every 12 samples. The method blanks were treated in a similar way as the samples but sodium sulfate was extracted in place of dust/soil under study. Results obtained were not corrected for blank concentrations since almost all the target PCBs were absent from blanks, and even where present in blanks, concentrations were less than 5 % of the concentration found in the corresponding sample. The limit of detection was obtained as the concentration equivalent to a signal to noise ratio of 3:1. LOD values obtained for dust were: 0.09 ng/g, 0.08 ng/g, 0.11 ng/g, 0.11 ng/g, 0.11 ng/g, 0.11 ng/g, and 0.08 ng/g for PCB 28, 52, 101, 118, 138 and 180.
respectively. The corresponding LOD values for soil were: 0.07 ng/g, 0.06 ng/g, 0.07 ng/g, 0.06 ng/g, 0.07 ng/g, 0.07 ng/g, and 0.05 ng/g respectively.

2.6 Data treatment

The results were analysed statistically using Minitab 18. Given the distribution of our data revealed by visual inspection, non-parametric methods were used to test for significant differences in concentrations of PCBs in different sample groups. Specifically, a Wilcoxon test was performed to test: (a) differences in concentrations of PCBs in soil samples taken at two distances from each power station, and (b) the influence of season on concentrations of PCBs in our soil samples. In addition, a Kruskal-Wallis test was used to test if different categories of power station activity exerted a significant influence on the concentration of PCBs in our soil samples. Finally, principal component analysis (PCA) was used to compare PCB congener patterns in samples with those in transformer oil and Aroclor mixtures.

3. Results and Discussion

3.1 Differences in concentration of PCBs in soil samples from point 1 and 2.

We hypothesised that soil samples taken closer to the power station facility would have higher concentrations of PCBs compared to samples acquired further away. Table 1 shows the \( \Sigma \text{PCB}_7 \) concentrations in samples taken from both points at each power station studied. A Wilcoxon test revealed that concentrations of \( \Sigma \text{PCB}_7 \) closer to the power station (point 1 samples) were significantly higher (\( p=0.013 \)) than those in samples taken further away (point 2 samples). Similar significantly (\( p<0.05 \)) elevated concentrations in point 1 samples were observed for all individual PCB congeners. This increment in PCB contamination of soil
close to power generation facilities, suggests strongly that such activities have constituted a substantial source of PCBs to their local environments.

Power station I (PS I), the former generation station, was the only site that had high concentrations of target PCBs in both point 1 as well as point 2 samples (Table 1). The high concentrations of PCBs in the point 2 soil from PS I could be due to emissions arising from the conveying of transformers in and out of the power station since the power station is now used for transformer repair purposes.

3.2 Influence of power station activity category on PCB concentrations in dust and soil

The 12 power stations studied comprised 6 distribution sub-stations, 5 transmission stations and 1 former generation station, now used for repair of transformers. A Kruskal Wallis test was performed to test whether the type of activity (transmission, distribution, or transformer dismantling (station I)) impacted on the concentrations of PCBs in soil and dust. On the basis of power station activity, results revealed a statistically significant difference (P<0.05) in concentrations of total PCBs in dust and soil-between the three categories. For dust, the mean ΣPCB₇ concentration for generation station samples was 2241.8 ng/g; in transmission station samples (n=5) the mean ΣPCB₇ concentration was 314.5 ng/g, while concentrations in the distribution station samples (n=6; mean ΣPCB₇ = 122.8 ng/g) were the lowest. For soil, the mean ΣPCB₇ concentration for generation station samples was 197.7 ng/g; in transmission station samples (n=5) the mean ΣPCB₇ concentration was 92.6 ng/g, while distribution station samples (n=6; mean ΣPCB₇ = 38.3 ng/g) had the lowest concentrations. The persistent nature of PCBs was confirmed from the results obtained for PCBs in soil samples around power generation transformers from PS I that has been non-functional for over a decade, with residual concentrations very close to being the highest in soil samples from power stations in this study. These high levels could be residual concentration from day to day operations (before the ban of PCBs) when the power station was fully functional.
3.3 Congener pattern in dust, soil and transformer oil samples relative to PCB commercial mixtures

PCBs were mostly used as mixtures, manufactured under various trade names by different producers. One of the most popular of these PCB mixtures were known as Aroclors. Principal component analysis (PCA) was conducted to identify which of the Aroclors have a congener pattern similar to those in samples from this study. It was observed that many of the soil samples, some of the dust samples, as well as transformer oil samples have positive component scores in PC 1, similar to Aroclor 1260. PC 1 could explain 39% of the congener variations within the data set and was driven positively by PCB 153 > 101 > 118 > 138, with eigenvectors of 0.490, 0.446, 0.435, 0.279 respectively. PC 1 was driven negatively by PCB 52 > 28 > 180 with eigenvectors of -0.360, -0.310, -0.262 respectively.

Aroclor 1260 had positive component scores in PC 2, together with some of the dust and soil samples. The oil from PS I and J also displayed positive PC 2 scores. PC 2, which explained an additional 24.8% of the variation in congener patterns in the samples, was driven towards the positive direction substantially by PCB 180, with an eigenvector of 0.512, and also by PCB 138 and 153, with eigenvectors of 0.399 and 0.142 respectively. PCB 28 made the greatest negative contribution to PC 2, with an eigenvector of -0.427; while PCB 101, 52 and 118 also contributed negatively to PC 2, with eigenvectors of -0.375, -0.36, and -0.326 respectively.

Worthy of note are the soil (L-SOIL) and dust (L-DUST) samples from PS L (Figure 1), these samples from power station L both had the highest negative scores in PC 2, of all the soil and dust samples. Also, of all the soil and dust samples, L-SOIL contributed most negatively to scores of PC 1 while L-DUST contributed most positively to the scores of PC 1.

It was observed that Aroclor 1242 displayed a very similar PC1 score to L-SOIL. Moreover,
Aroclors 1242 and 1016 displayed PC2 scores similar to those for both soil and dust from PS L (Figure 1).

3.4 Effect of seasonal variation on concentrations of target PCBs in dust and soil

Wilcoxon test employed to test the effect of seasonal variation revealed statistically indistinguishable concentrations of individual target PCBs in dust samples for the month of June and December. A similar absence of seasonal variation was observed when comparing PCB concentrations in soil in June with those from the same locations in December.

3.5 Comparison of concentrations obtained in soil and dust with relevant reports

Concentrations of PCBs in dust from our Nigerian power station offices exceed those reported for PCBs in dust from Nigerian cars, homes and office (Harrad et al, 2016). They also exceed those in other previous studies, (Tue et al., 2013; Tan et al., 2007; Ali et al., 2012) reported PCBs in Vietnamese homes around e-waste sites; Singaporean, Kuwaiti, and Pakistani homes respectively; but are lower than those reported by Abafe and Martincigh, 2015 and Zheng et al., 2015 for South African offices and homes around e-waste handling sites respectively. Worthy of note is the similar congener profile in our report to the other two reports on PCBs in dust from African countries (Harrad et al., 2016; Abafe and Martincigh, 2015) (Table 2). This may imply that the PCB formulations used in Nigeria and South Africa are similarly dominated by higher chlorinated congeners (Harrad et al., 2009) – such as Aroclor 1260.

With respect to soil, concentrations of both individual congeners and ΣPCB7 in our study exceed those in other studies from locations not directly impacted by power station activities (Table 3). This is further evidence of the potential importance of power stations as sources of local contamination with PCBs, and further study appears warranted.
4. Conclusion

The concentrations of PCBs in samples of soil taken 1-3 m distant from Nigerian power stations exceed significantly those detected in soil samples taken 50-100 m from the same facilities. The type of activity conducted at each power station exerted a significant effect on the concentrations of PCBs in soil and dust samples with the distribution stations having the lowest concentrations.

Irrespective of the power station activity, the PCB congener pattern in soil was observed to be more similar to that in transformer oil taken from some of our power stations than that in dust samples. We thus infer that concentrations of PCBs in soils in this study were substantially influenced by emissions from transformer oil, while our dust samples were influenced by alternative sources such as control panels present inside power stations. The evidence presented here supports the hypothesis that activities at power stations have contributed significantly to PCB contamination of the surrounding environment.

Acknowledgements

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Soil samples (yellow triangles); dust samples (purple rhombuses); oil from power station I&J (red circle); oil from power station H (blue triangle); oil from power station K (green square)

Figure 1: Score plot for principal component 1 versus principal component 2 for soil, indoor dust, transformer oil and Aroclors
Table 1: Concentrations of $\Sigma$PCB$_7$ (ng/g) in soil from both sampling points at each site

<table>
<thead>
<tr>
<th>Power station (type)</th>
<th>June</th>
<th>December</th>
<th>Mean Point 1</th>
<th>Mean Point 2</th>
<th>Mean $\Sigma$PCB$^7$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>point 1</td>
<td>point 2</td>
<td>point 1</td>
<td>point 2</td>
<td></td>
</tr>
<tr>
<td>A (distribution)</td>
<td>3.5</td>
<td>2.4</td>
<td>0.4</td>
<td>1.2</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>1.8</td>
<td></td>
<td></td>
<td>1.9</td>
</tr>
<tr>
<td>B (transmission)</td>
<td>40.9</td>
<td>1.6</td>
<td>16.8</td>
<td>0.5</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>1.1</td>
<td>1.1</td>
<td></td>
<td></td>
<td>15</td>
</tr>
<tr>
<td>C (distribution)</td>
<td>27.4</td>
<td>17.4</td>
<td>10.8</td>
<td>2.8</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>10</td>
<td></td>
<td></td>
<td>14.6</td>
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<tr>
<td>D (distribution)</td>
<td>50</td>
<td>26.6</td>
<td>310</td>
<td>4.6</td>
<td>180</td>
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<tr>
<td></td>
<td>16</td>
<td>16</td>
<td></td>
<td></td>
<td>96.6</td>
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<tr>
<td>E (transmission)</td>
<td>320</td>
<td>10.7</td>
<td>71.3</td>
<td>5</td>
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<td></td>
<td>7.9</td>
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<td>101.7</td>
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<td>F (distribution)</td>
<td>148.4</td>
<td>51.4</td>
<td>43.3</td>
<td>26.3</td>
<td>96</td>
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<td>G (distribution)</td>
<td>74.6</td>
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<td>H (transmission)</td>
<td>162.2</td>
<td>18</td>
<td>254.9</td>
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<td>16</td>
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<td>I (generation)</td>
<td>220.1</td>
<td>211.1</td>
<td>125.7</td>
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<td>220</td>
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<td>197.7</td>
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<tr>
<td>J (transmission)</td>
<td>317.6</td>
<td>57.3</td>
<td>422.4</td>
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<td>K (distribution)</td>
<td>33</td>
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<td>L (transmission)</td>
<td>83.2</td>
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<td>4.3</td>
<td>45</td>
</tr>
<tr>
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<td>26.4</td>
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### Table 2: Comparison of median (mean) concentrations (ng/g) of 7 indicator PCBs in dust from this study with those from selected other studies

<table>
<thead>
<tr>
<th>COUNTRY (REF.)</th>
<th>MICROENVIRONMENT</th>
<th>N</th>
<th>PCB 28</th>
<th>PCB 52</th>
<th>PCB 101</th>
<th>PCB 118</th>
<th>PCB 138</th>
<th>PCB 153</th>
<th>PCB 180</th>
</tr>
</thead>
<tbody>
<tr>
<td>NIGERIA (This study)</td>
<td>POWER STATION OFFICES</td>
<td>4</td>
<td>8.9 (18.3)</td>
<td>16.3 (22.4)</td>
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**Notes:** NA - NOT APPLICABLE (Congeners were not analysed); NS - NOT SPECIFIED (Congeners were analysed but concentrations were not reported).
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Table 3: Comparison of median (mean) concentrations (ng/g) of PCBs in soil from this study with those from selected other studies.
Supplementary Material

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