Occurrence, Compositional Profiles and Health Risks of Polybrominated Diphenyl Ethers (PBDEs) in Leachate and Groundwater in and around Dumpsites in Lagos State, Nigeria

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Research Article

Keywords: Occurrence, Distribution, PBDEs, Groundwater, Dumpsites, Lagos State

Posted Date: July 28th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-749279/v1

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Abstract

Groundwater from the communities adjoining Abule Egba (ABE), Epe (EPE), Ikorodu (IKR), Olushosun (OSH) and Solous (SOL) dumpsites in Lagos State, Nigeria were collected and analyzed for Polybrominated diphenyl ethers. Water samples were extracted using liquid-liquid extraction followed by Gas Chromatography/Mass Spectroscopy analysis using already validated standard method. PBDEs concentrations ranged from 7.1 ± 10.1 (IKR) to 78 ± 109 μgL^{-1} (ABE) for BDE 28; 5.4 ± 0.3 (OSH) to 31.5 ± 27.3 μgL^{-1} (ABE) for BDE 47; 17 ± 24 (ABE) to 183 ± 226 μgL^{-1} for BDE 100; 11.7 ± 16.5 (ABE) to 174 ± 217 μgL^{-1}(EPE) for BDE 99; 117 ± 166 (ABE) to 2034 ± 2819 µg L^{-1} (EPE) for BDE 183; 296 ± 392 (IKR) to 4283 ± 1278 µg L^{-1} (EPE) for BDE 209. BDE 153 and BDE 154 were not detected in all the water samples while BDE 28 was only detected in ABE and IKR water samples. The BDE 209 was the most dominant congener in all the water samples. The cumulative HI values for children through ingestion route ranged from 4.95 E+00 (SOL) to 4.53 E+01 (EPE) while in adult, it ranged from 4.25 E+00 (SOL) to 3.88 E+01 (EPE). This study confirmed the presence of PBDEs at elevated concentration in groundwater. Since there is no regular supply of pipe-borne water in the study area, residents consume the contaminated groundwater and may therefore be exposed to endocrine disrupting chemicals. Government should provide pie-borne water for the populace and discourage people from living close to the dumpsites.

Keywords: Occurrence; Distribution; PBDEs; Groundwater; Dumpsites; Lagos State

1.0 Introduction

Open dumps and non-engineered landfill with inadequate or poor facilities for leachate and gas control are major sources of solid waste management in many developing countries, Nigeria not an exception (Kanmani and Gandhimathi, 2013; Ancic et al., 2020). Dumpsites are known to receive various kinds of domestic and industrial materials that often contains endocrine disrupting compounds (EDCs). Poor solid waste management is a major route of EDCs into the environment (Jiang et al., 2014). Dumpsites leachates is a heterogeneous mixture of persistent organic compounds and inorganic elements that are formed from interaction between excess water percolating through layers of waste in dumpsites (Kapelewska et al., 2016). Leachates in unlined dumpsites have been reported to contaminate groundwater (Nevondo et al., 2019; Harrad et al., 2020). The acute toxicity

of leachate from selected dumpsites has been reported worldwide (Olujimi et al., 2016; Ancic et al., 2020). Among the EDCs that has been reported in dumpsites are Polybrominated Diphenyl Ethers (PBDEs). PBDEs are flame retardants which are produced in large quantities all over the world. They are therefore ubiquitous compounds in the environment. PBDEs are widely added as flame retardants to consumer products such as TV sets, computer circuit boards, plastics, polyurethane foams, textiles, etc (O'Driscoll et al., 2016). PBDEs are chemical contaminants of global concern because they are persistent, bioaccumulate, biomagnify and therefore highly toxic (Zhang et al., 2014). They therefore pose a risk by causing adverse effects to animals, human health and the environment (Zhang et al., 2014; Harrad et al., 2020).

The strategic location of Lagos State in Nigeria and Africa has assisted in the rapid urbanization and industrialization of the city. Fifty percent of Nigeria's industrial activities are in Lagos area, thus, accounting for the mass movement of people from far and near, which produce thousands of tons of municipal solid waste (MSW) (Oresanya, 2000). Industrial expansion of the city with consequent increase in the number of inhabitants have resulted in generation and release of untreated and poorly managed solid wastes (Olukoju, 2018). Like many other developing nations commercial capitals, the MSW composition of Lagos varies widely due to socio-economic conditions, seasons and many other factors. In addition, most of the surrounding areas of the dumpsites in Lagos State are fully built and therefore people live next door to dumpsite. Since there is no regular supply of pipe borne water, residents of such areas rely on well water for drinking. However, there have been several complaints on poor management of solid waste at different dumpsites within Lagos State due foul air and groundwater contamination and subsequent impact on health of people living close to these dumpsites (Olukoju, 2018). Therefore, the main objective of this study was to assess the level and health risk of PBDEs in groundwater around selected dumpsites in Lagos State.

2.0 Materials and methods

2.1 Chemicals and reagents

Certified standard solutions of 9 PBDE (BDE-28, BDE-47, BDE-100, BDE-99, BDE-154, BDE153, BDE-183 and BDE-209), surrogate standard standards (BDE-50 and BDE-172) and internal standards (BDE 118 and 128) were purchased from Wellington Laboratories (Guelph, Ontario,

Canada) while silica gel (100 to 200 mesh), sodium sulfate (purity 99.9 %), and HPLC grade solvents: acetone, hexane and dichloromethane were purchased from Sigma Aldrich (Pty) Ltd.

2.2 Study Areas/Sampling: Landfill/Dumpsite Management in Lagos State is a unit under Waste Management Services. This study covers 4 active dumpsites and 1 decommissioned dumpsite. The name and description of dumpsites investigated is presented in Table 1 and the map of Lagos State showing the sampling points is depicted in Figure 1. Prior to sample collection, the Winchester amber bottles used for sampling were washed with soapy water and rinsed with clean tap water, soaked overnight in 10 % nitric acid (Sigma Aldrich, South Africa). A total of sixty (60) water samples were collected from the communities of the five dumpsites (Abule Egba (ABE), Epe (EPE), Ikorodu (IKR), Olusosun (OSH) and Solous (SOL) dumpsites). Water samples were either collected from hand dug wells. The depth and water level of groundwater sources were measured using a dip-meter attached to the water drawer. The hand dug wells are all shallower than 18m, with the majority less than 10m deep. Samples were collected within 500-meter distance from the dumpsite except for EPE dumpsite where samples were collected from an hand dug well within the dumpsite as there was no houses around. The well was the main source of water for the workers, food vendor and scavengers working at the dumpsite. Samples from adjourning communities for each of the dumpsite were pooled into four to obtain a representative sample. The samples were acidified with few drops of nitric acid to pH 2 to prevent biological activities, stored in ice chest and transported to the laboratory. In the laboratory, samples were kept in the freezer and samples were extracted before seven days and analyzed before 40 days after collection.

2.3 Preparation of water samples

500 mL of water sample was measured into pre-cleaned one (1) L separatory funnel. The samples were then spiked with 20 µL of 500 μgL^{-1} of surrogate standards (BDE 50 and BDE 172). 40 mL of double re-distilled dichloromethane was added, and the mixture was shaken for 3 minutes with intermittent venting. The sample was allowed to stand for about 15 minutes to allow for phase separation. The organic phase was then filtered over 15 g anhydrous sodium sulphate. The extraction and separation were repeated two more times and the organic phase combined. The organic phase was transferred into a clean beaker and the separating funnel was rinsed with 10 mL of

126 dichloromethane. The beaker containing the extract was covered with baked aluminum foil,
127 perforated, placed in a fume cupboard for air drying. The extract was dried to about 2 mL prior to
128 sample cleanup. The extract was thereafter cleaned up on a column packed with 5 g pre-extracted and
129 activated silica gel. The adsorbent was preconditioned with solvent prior to introduction of the extract
130 to the column. Exactly 20 mL of n-hexane was used as eluting solvent. The eluent was collected into
131 a clean 100 mL flask and concentrated to about 1 mL under gentle nitrogen. The extracts were
132 analyzed using GC/MS.

133 **2.4 Gas Chromatography–Mass Spectrometry analysis**

134 The quantitative analyses of all the congeners were determined on an ultra-trace 2010 Shimadzu GC
135 equipped with QP 2010 ultra-mass spectrometer operated in Electron Ionization (EI) mode. The
136 chromatographic separation of the PBDEs congeners were obtained using ZB-5 MS (15 m, .25 mm
137 I.D, 0.25 µm film thickness, J & W Scientific Folsom, CA, USA) capillary column. Due to thermal
138 instability of BDE 209, shorter column was used (i.e. 15m column instead of 30m). Helium gas was
139 used on carrier gas at a flow rate of 1.2ml/min, and the temperature program was set as follows; 90°
140 for 2 min, increased at 320°C at 15°C/min and held for 7min. The temperature of the GC inlet, transfer
141 line and ionization source and quadrupole were set at 290°C, 300° and 150°. To eliminate
142 interferences of co-extractants and to enhance sensitivity of the instrument, MS acquisition was
143 carried out in selected low monitoring SIM mode. For the SIM mode, a target ion and two reference
144 ions were selected for each of the target congeners for their identification and quantification. The
145 quantification of all the targeted congeners was calculated using external calibration on the basis of
146 the peak area. Five points calibration level was carried out 25,50,100, 250 and 500 $ngmL^{-1}$ to all the
147 PBDEs congeners. The linearity (R^2) of the calibration plot for each target analyte was greater than
148 0.995 except for PBDE-209 which was 0.978.

149 **2.5 Quality Assurance and Control**

150 Silica gel used for clean-up were pre-extracted with dichloromethane (DCM) before use. All solvents
151 were of analytical grade. Each sample was extracted thrice. A procedural blank and solvent blanks
152 (PBDE congeners spiked into sample) were analyzed simultaneously with the samples to check for
153 interferences and contamination for each batch of 12 samples. Three criteria were also used to ensure
154 the correct identification of the target compounds: (a) The GC (gas chromatography) retention times

matched those of the authentic standards within ±0.1 min and (b) the signal-to-noise ratio was greater than 3:1. The recoveries for BDE-100, 154, 153, 183, and 209 were 60–107% while for BDE-28, 47, and 99, the recoveries were 62–88%. For all the target compounds, the relative standard deviations of duplicate samples were less than 14%.

2.6. Health Risk Assessment.

Health risk of PBDEs in groundwater is necessary in this study because of reported cases of skin diseases e.g. eczema and acne by residents around Solous dumpsite. Adults and children were considered separately in the risk considering their exposure regimes and durations. Only two routes of exposures were considered i.e. (i) ingestion though drinking and (ii) dermal absorption through domestic chores. Methods as described by (Fatoki et al., 2010; Olujimi et al., 2015; Olujimi et al., 2017), and United States Environmental Protection Agency— US EPA (2013) were adopted with slight modification for health risk assessment (Tables 1 and 2). The human health risk was computed as the average concentration of PBDEs exposed to human on daily basis over a specific exposure time. The average daily exposure concentration is referred to as average daily dose (ADD) and it was computed using Eq. 1 as follows:

$$ADD_{ing} = \frac{C_{mean} \times IR \times ED \times F_c}{BW \times AT} \qquad \text{Equation 1}$$

C_{mean} is the concentration of PDBEEs in the groundwater samples; IR is the average daily consumption rate; ED is the exposure period (in years); Fc is the fraction contaminated; BW is the average bodyweight; AT (mg/ kg day) is the average lifetime of exposure. Carcinogenic risk assessment due to lifetime exposure was assessed using equation 2.

$$LADD_{ingestion} = ADD_{ingestion} \times ({}^{ED}/_{Lft}) \qquad \text{Equation 2}$$

Dermal absorption dose (DAD) was assessed using equation 3:

$$DAD_{dermal} = \frac{C_{mean} \times SA \times SL \times ABS \times EF \times ED}{BW \times AT} \qquad \text{Equation 3}$$

SA is skin surface area; SL is skin adherence factor = 0.7 mg $cm^{-2}day^{-1}$; ABS is dermal absorption factor; EF is exposure frequency. Body weight 70 kg (adult), 15 kg (children) Lifetime 70 years (adult), 6 years (children) Exposure period 35 years (adult), 6 years (children) Drinking water 365 events/year; 2 L/event (adult), 1 L/event (children); 100 % fraction contaminated Dermal absorption 365 events/year; 12 min/event, 6 min/event; 3300 cm^2 skin surface (adult), 2800 cm^2 skin surface (children).

The risk of cancer development due to PBDEs exposure was estimated based on the assumption that polluted water is the main source of water for domestic chores for the people. For chemicals that have potential for causing cancer, risk was calculated using equation 4.

$$Cancer\ Risk = \beta \times LADD \qquad \text{Equation 4}$$

where Risk ingestion is the potential risk due to ingestion of contaminated water; LADD is lifetime average daily dose; β is oral potency factor/slope.

3.0 Results and Discussion

3.1 PBDEs Concentration in water

The concentrations of PBDEs in groundwater from the five dumpsites and surroundings in Lagos State is presented in Table 2 while Table 3 compares data from elsewhere for referencing purposes. All the BDE congeners were detected in all samples except for BDE 153 and 154. BDE-153 production was banned in the 1970s, this study considered the congener to assess its persistence and availability. BDE 28 was conspicuously absent in samples from SOL, OSH and IKR. BDE 47, 100, 99, 183 and 209 were significantly different across the dumpsites. BDEs 28 and 47 had the highest concentrations of 78 μgL^{-1} and 32 μgL^{-1} at ABE dumpsite while BDEs 100, 99, 183 and 209 had highest concentrations of 183 μgL^{-1}, 174 μgL^{-1}, 2034 μgL^{-1}at EPE dumpsite. The lowest concentrations of BDEs 100, 99 and 183 concentrations were recorded at ABE dumpsite while the lowest concentrations of BDEs 28 and 209 were recorded at IKR dumpsite.

The distribution pattern of the BDE congeners in water samples from and around dumpsite showed that BDE 28 exhibited the following; SOL = OSH = EPE < IKR < ABE; while BDE 47 showed that OSH < SOL < IKR < ABE; BDE 100 also exhibited that ABE < SOL < IKR < OSH < EPE; BDE 99 showed that ABE < SOL < IKR < OSH < EPE; BDE 183 showed that ABE < SOL < IKR < OHS < EPE and BDE 209 showed that IKR < SOL < OSH < ABE < EPE. It is important to note that the distribution pattern of BDEs 99, 100 and 183 are similar for all the dumpsites. It can be inferred from this distribution pattern that the sources of these congeners to the dumpsites were from similar products that were subsequently leached into the groundwater. Similarly, the distribution pattern for Σ_8PBDES was EPE < ABE < OSH < IKR < SOL with Epe having the highest Σ_8PBDES of 6687.6 μgL^{-1} in water sample. EPE being the youngest of all the dumpsites in the state, the justification for the high concentration obtained may be attributed to the burning of contraband goods such as milk, tyres, body creams among others at the site. Burning of goods/waste usually increase the release and availability of PBDEs into the environment (Jiang et al., 2014). Water around SOL dumpsite had the least concentration of total PBDEs. This can be linked to the type of waste received at the site. At IKR, aside from municipal waste received, ash medical waste is dumped at the site.

The compositional profile of PBDEs congeners in water samples around the dumpsites is shown in Figures 2 and 3(A-D). BDE 209 was the most abundant congener in the water samples around ABE, SOL and EPE dumpsites which accounted for 92 %, 65 % and 64 %, respectively. The high levels of BDE-209 may be due to deposits/leachate from the used and improper disposal of deca-BDE formulation such as textiles in upholstery, electronic circuit boards, mattresses, etc at the site. Water samples around IKR and OSH dumpsites had BDE 183 as the most abundant congener contributing 58 % and 52 % respectively and BDE 209 as the next dominant congener accounting for 27 % and 37 % respectively. The significant level of BDE-183 in IKR and OSH showed deposition of contaminants from acrylonitrile-butadiene-styrene (ABS) which were present in computers and housing appliances (Sutton et al., 2014). This may be due to improper disposal and burning of electronic gadget, foams in upholsteries etc which eventually releases these congeners. EPE, OSH and SOL recorded the least contribution of BDE 47 which accounted for 0.2, 0.3 and 0.9 % respectively while ABE had 0.3 % of BDE 99 and IKR had 0.6 % of BDE 28 as their lowest congeners (Figure 3A-D). The dominance of BDE 209 reflects the change from the general global use of the commercial penta- and octa-BDE formulations to deca-BDE, as well as high sorption and persistence

of BDE 209. The variation in the most abundant congeners in groundwater samples around each dumpsite may be due to the differences in the types and volume of waste deposited at various each site.

The concentration of BDE congeners in all the groundwater samples were higher when compared to previous studies (Streets et al., 2006; Wurl et al., 2006). Also, previous work on Predecelle River (France), Aire River (UK), electronic industrial water (China) and groundwater in Ireland (Harrad et al., 2020) had reported varying concentrations of ΣPBDEs but these are comparatively lower to the results obtained in this study (Labadie et al., 2010; Cristale et al., 2013; Jiang et al., 2014; Olutona et al., 2017; Harrad et al., 2020). Essumang et al. (2018) reported BDE 100 as the most abundant congener, followed by BDE-99. BDE 209 was the dominating congeners in most of the previous studies. The Regulation of the Ministry of Environment (2011) specifies the admissible level of PBDEs to be 0.5 ng L^{-1} as the year-average concentration in homogeneous water and 0.2 ngL^{-1} for homogeneous parts of coastal and flowing waters. The Federal Water Quality Guidelines for PBDE congeners are BDE-47 (24 ngL^{-1}) BDE-99 (3.9 ngL^{-1}), BDE-100 (0.23 ngL^{-1}) (Wollenberger et al., 2005) and BDE-153 (120 ngL^{-1}) (Environmental Canada, 2013). The mean levels recorded for the various congeners in this study were often higher by a 1000-fold except for BDE 153 and BDE 154 that were not detected in all samples. Thus, an indication that groundwater in adjourning dumpsites in Lagos State is highly polluted with respect to PBDEs. Figure 3 depicts the compositional distribution of the different BDE congeners. This clearly gives the similarities in waste composition received at each of the dumpsites and or anthropogenic activities at each site leading to the leaching of each and or group of the congener. Although ABE dumpsite was decommissioned, but still receives waste from nearby market and residential houses around it.

3.2 Health Risk Assessment

Non-cancer and cancer risk

Human health risk may arise due to ingestion or dermal contact. The non-carcinogenic effect (HQ) and cancer risk for ingestion and dermal is presented in Tables 4 and 5 (See Supplementary Table for

detail calculation). The HQ index was higher than the threshold values of Federal Environmental Quality Guidelines for PBDEs in Canada (FEQG). This clearly indicates that the water was not safe for domestic use. For the ingestion route, the non-cancer risk for BDE 28 exceeded the acceptable (safe) limit at ABE dumpsite while BDE 47 was below the safe limits for all the sites. BDEs 99 and 100 were only safe at ABE and SOL while BDEs 183 and 209 exceeded the safe limit for all the dumpsites. The cumulative HI values for children through ingestion route ranged from 4.95 E+00 (SOL) to 4.53 E+01 (EPE) while in adult, it ranged from 4.25 E+00 (SOL) to 3.88 E+01 (EPE). For dermal route, the non-cancer risk for children ranged from 5.6 E+02 (SOL) to 1.64 E+04 (EPE). The non-cancer risk for children showed that dermal route poses higher risk. The cumulative non-cancer risk showed that EPE dumpsite was more contaminated and poses the highest risk of water use. Additionally, the values of the congeners reported in this study confirmed incidences of groundwater pollution around SOL dumpsite that warranted this study. For examples, residents around SOL dumpsite had reported skin diseases e.g. skin rashes, blisters which when busted gave a yellowish foul-smelling discharge following the continuous use of polluted groundwater. Although EPE being the youngest of all the dumpsites was located outskirt of the metropolis was 16000 times higher than the acceptable limit. The dumpsite seems not to pose any problem now; however, action is required for the future impact.

Cancer risk was only determined for BDE 209 due to availability of slope factor for this congener. Cancer risk for ingestion route in children ranged from 6 E-07 to 9 E-06 and for dermal route, it ranged from 9.6 E-06 to 7.4 E-06. In adult, cancer risk due to ingestion ranged from 1.91 E-07 (IKR) to 2.76 E-06 (EPE) while dermal route ranged from 2.34 E-04 (IKR) to 3.39 E-03 (EPE). Like non-cancer risk, the main route for possible cancer risk development was via the dermal route most especially for adult. For the ingestion route, the cancer risk was within the acceptable limit of 1 E-05 (WHO, 2003) while both children and adult were more susceptible to cancer risk via dermal as the estimated values were higher that WHO prescribed value.

4.0 Conclusion

Findings from this study showed that PBDEs were present at very elevated concentrations in the groundwater around the dumpsites. Researchers have shown that low-level exposure to PBDEs

causes neurotoxin damage, cancer and other health related issues. Since there is no regular supply of pipe-borne water in the study area, people rely on well water for drinking. Therefore, many people who depend on groundwater as their major water supply will be exposed to these endocrine disrupting chemicals. Government should provide pipe borne water for the populace and discourage people from living close to dumpsites.

Acknowledgements

The research team expresses profound gratitude for the funding support from Lagos State Government through the Lagos State Research and Development Council (LRSC). We also thank the Directorate of Grants Management, Federal University of Agriculture, Abeokuta for professional coordination/administration of the grant.

Conflicts of Interest

The authors unanimously declare that there is no conflict of interest from the inception to the end of this research.

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Figures

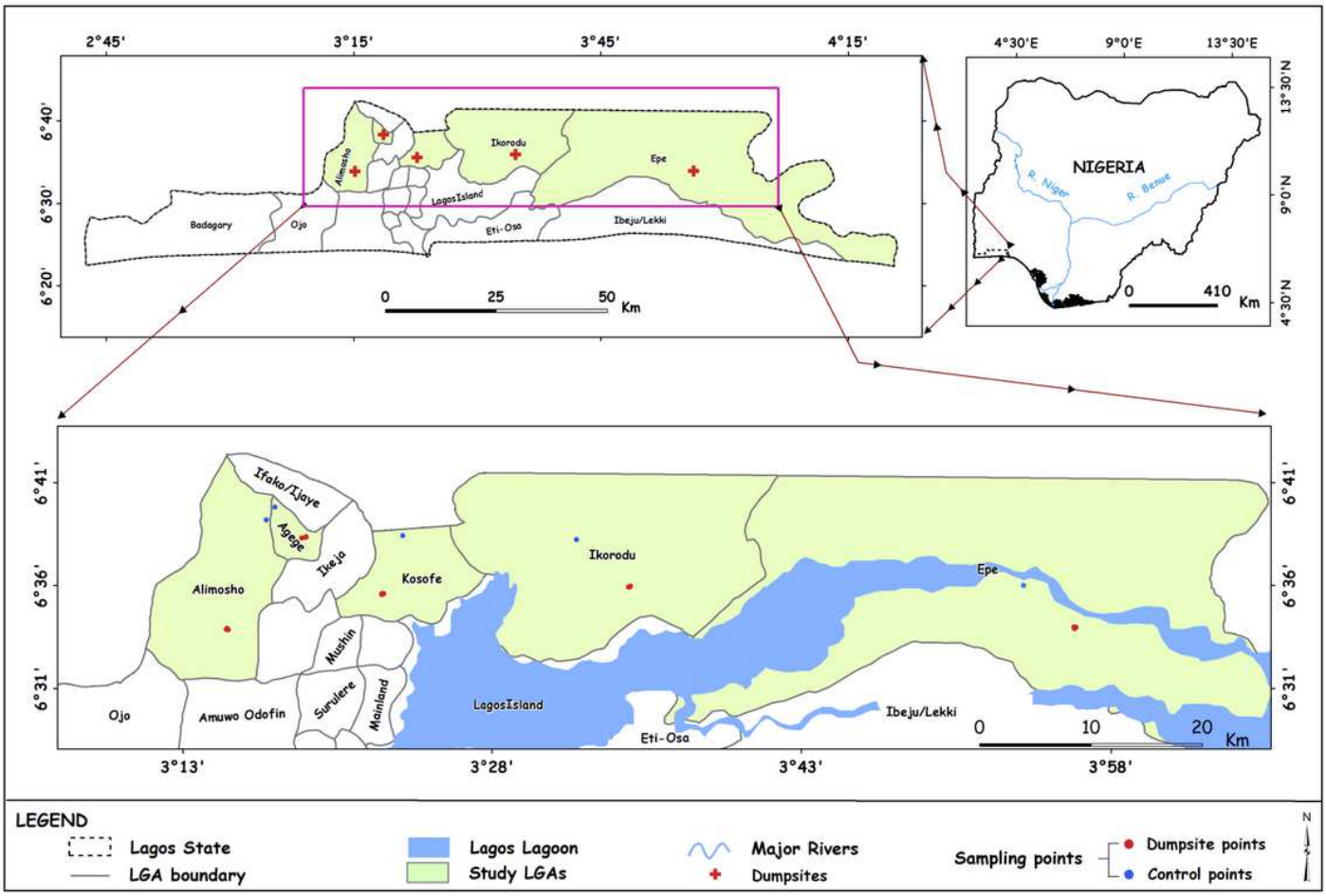

Figure 1

Map of Lagos State, Nigeria showing the dumpsites and sampling points Note: The location of Lagos State in Nigeria is shown at the top right while the location of the sample areas in Lagos State is shown at the top left.

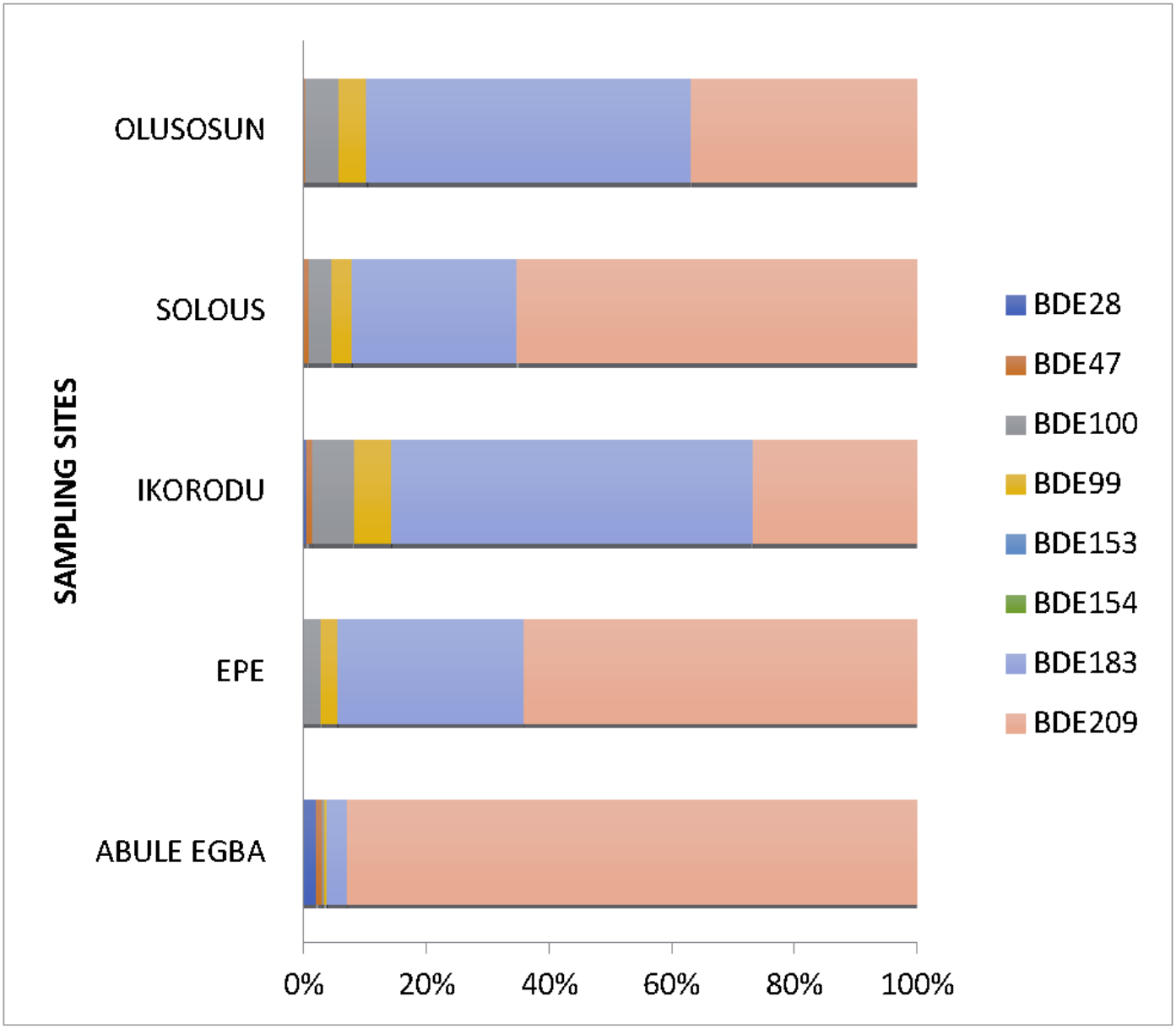

Figure 2

Compositional profile of PBDEs congeners in water around the dumpsites

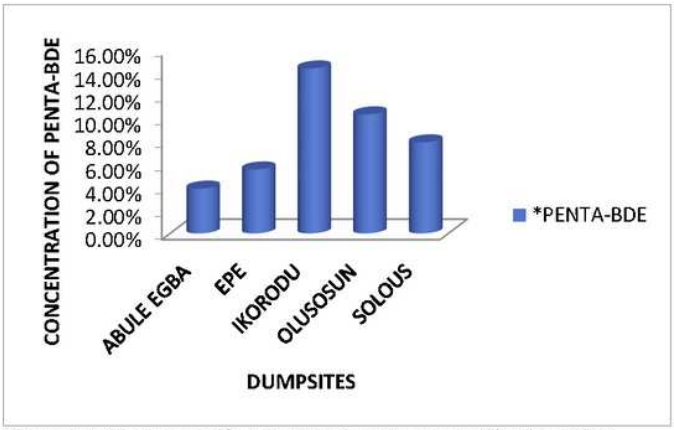

Figure 3A: Percentage of penta-BDE in water around the dumpsites
BDE= Addition of BDE-28, 47, 99 and 100

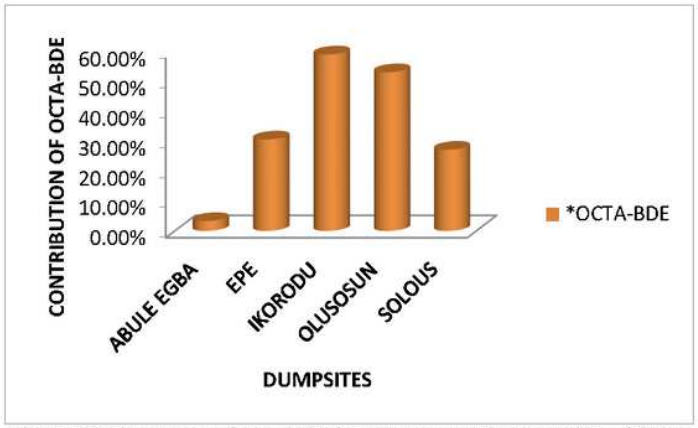

Figure 3B: Percentage of octa-BDE in water around the dumpsite *Penta-
*Octa-BDE= Addition of BDE-153, 154 and 183

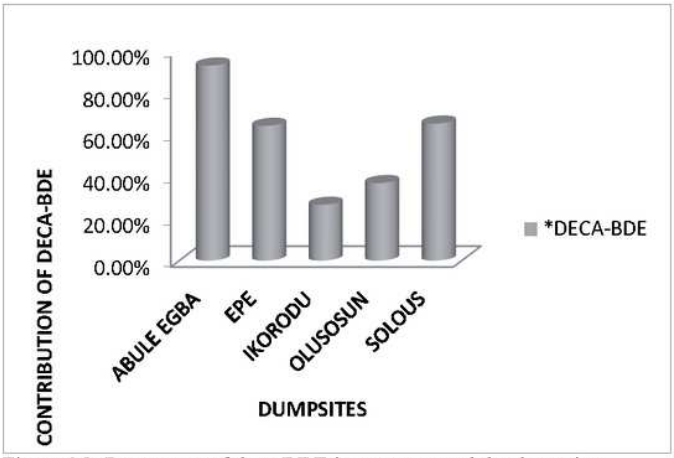

Figure 3C: Percentage of deca-BDE in water around the dumpsites
*Deca-BDE= BDE-209

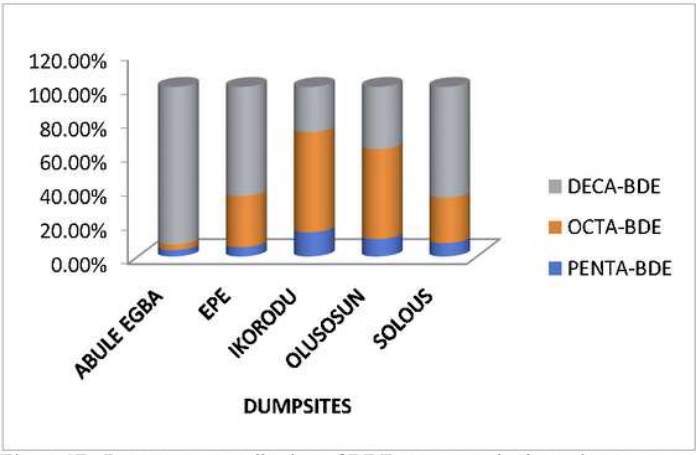

Figure 3D: Percentage contribution of BDE congeners in dumpsites water

Figure 3

See image above for figure legend

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Tables.pdf
- PBDEsSupplementaryTable1.xlsx