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Carcinogenicity of dioxin-like polychlorinated biphenyls in transformer soil in vicinity of University of Port Harcourt, Choba, Nigeria

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ABSTRACT

Polychlorinated Biphenyls (PCBs) in the vicinity of transformers soils at main campus of university of Port Harcourt, Choba, Nigeria was monitored. Evaluation was done for both total PCBs (Aroclor) and congener's form using Gas Chromatography at four designated sites; A, B, C, and D with geographical co-ordinates for site A-Donald Ekong Library, latitude 4˚54', 32''N and longitude 6˚55:05''E, site B-Senate building with latitude 4˚54'14''N and longitude 6˚55', 23''N, site C-Transformer close to music department with latitude 4˚54', 01''N and longitude 6˚55; 56''E and finally site D-Gana-Ma Lecturers residential quarters with latitude 4˚54'.23'' N and longitude 6˚55', 74''E. All the sites are polluted with PCBs that exceeds the maximum limit of 2.0 mg/kg as per United States Toxic Substances Control Act (TSCA). The order of total PCBs was site D site > site A > site B > site C, which also corresponds with the order of sites carcinogenicity of dioxin-like PCBs, calculated as Total toxicity Equivalence concentration (TTEC). The TTEC for site A, B, C and D corresponds to 0.000012, 0.000035, 0.0000185 and 0.00039 (mg/kg), respectively, which exceeded the method B clean up levels for 2, 3, 7, 8-tetrachloro dibenzo-p-dioxin levels of 1.3×10^{-5} mg/kg and need of massive cleanup for carcinogenic dioxin-like PCBs. We also found out there is high loadings of PCBs congeners with little or no biodegradability across the four sites. To mitigate the known human health risks posed by PCBs toxicity, non-PCBs transformers should be replaced with current ones and extensive soil remediation is needed to clean up the PCBs to avoid negative impact.

Capsule Summary: The PCBs concentrations at various sites (Port Harcourt, Nigeria) were monitored, PCBs and dioxin-like PCBs were beyond the permissible limits, which are carcinogenic and needs to be remediated to avoid negative impact.

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INTRODUCTION

Polychlorinated Biphenyls (PCBs) are chemicals (usually man-made) formed as result of attachment of one or more chlorine atoms to a Biphenyl ring structure. The number and position of chlorine attachment to the biphenyl ring structure varies and resulting to 209 different PCBs formations called Congeners (Waid, 1986a; Bedard et al., 1987b; Quensen et al., 1990; Mousa et al., 1996; WHO, 1993; WHO, 2003; USEPA, 2016; Eromisele and Iwuoha, 2018). First synthesized in 1864 and its commercial production started in 1929 as

congeners and aroclor. The application and usage of PCBs skyrocketed due to its favorable physical and chemical reactivity, high electrical resistance, high thermal stability and insulating properties as well as high stability under high stress and pressure (Eromosele and Iwuoha, 2018; Roberta, 2013).

All these adorable properties, most especially its chemical stability under extreme condition of heat, pressure and electricity means PCBs persists in the environment. The bio-accumulation potentials in living organisms and toxicity of PCBs were later noted and these became very serious environmental concerns for its continuous usage and application worldwide (Hayat et al., 2019; Liu et al., 2019; Monfort et al., 2019; Xu et al., 2019; Yurdakul et al., 2019; Zhao et al., 2016). Some PCBs congeners and aroclors dioxin-like toxicity have same toxicity as toxicity due to polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzo-furan (PCDFs) while other show non-dioxin-like toxicity. Due to the potential carcinogenicity of PCBs, its production were banned starting from the United State in 1977 (Eromosele and Iwuoha, 2018; Roberta, 2013).
PCBs application/usage as transformer fluids and capacitors containing PCBs in electrical transformers are one of the major leaks/source of PCBs in the environment and once released to the environment, it will bio-accumulate and biomagnify up the food chain, persists in the environment due to its poor degradability and its transported and recycled within the ecosystem (ATSDR, 2000; Safe, 2007).

Our focus on dioxin-like PCBs in soil is due to reports of its acute toxicity like chloracine, teratogenicity, endocrine disruption, endometriosis, immunotoxicity, and tumor promotion and carcinogenicity. (IARC, 1997; ATSDR, 1998; Bornbaun and Cummings, 2002), and more over PCBs are strongly absorbed soil. Generally PCB congeners could be subdivided into coplanar, the mono-ortho- substituted PCBs and non-dioxin-like toxicological effect as enumerated above. Both dioxin-like and non-dioxin-like PCBs as well have been noted to elicit broad spectrum of toxic and deadly responses in humans, laboratory animals and wildlife (Exner, 1982; Safe and Hutzinger, 1987; Crine, 1988; ATSDR, 2000).

The aim of present research was to determine the total concentration of PCBs in soils around transformer sites in the University of Port Harcourt. The 2006 toxicity equivalence factor (TEF) was applied in evaluation of the carcinogenic risks of the dioxin-like PCBs in the environmental matrix. Due to lack of research in this area in Niger-delta, it will help not only form baseline in further evaluation, but enable appropriate authorities to take actions to safeguard environment and reduce exposure risks to humans.

MATERIAL AND METHODS

Sample collection and sampling sites

Top soil samples matrix were collected randomly from four transformers areas within the University of Port Harcourt (i. e samples A, B, C and D respectively for site A, B, C and D) by scooping the top soil at a depth of approximately 2 cm each near the transformers. The geographical co-ordinates of the sites are: Site A-Donald Ekong Library, latitude 4°54, 32''N and longitude 6°55:05''E, site B- Senate building with latitude 4°54'14N and longitude 6°55; 23''N, site C- transformer close to music department with latitude 4°54; 01''N and longitude 6°55; 56''E and finally site C- GanaMa Lecturers residential quarters with latitude 4°54; 23''N and longitude 6°55; 74''E.

Soil samples were collected in amber glass bottles and were stored in an available refrigerator until they were analyzed. Reagents and apparatus used include: stock standard solution, methylene chloride, anhydrous sodium sulphate, GC-FID system, weighing balance, ultrasonic sonicator, 2ml amber screw cap vials, glass wares and extraction bottles.

Analytical procedure

USEPA 8083A was used for determining the concentration of PCBs in the soil matrix using GC-FID-7890A. The clean amber bottles were rinsed with distilled water to eliminate any form of cross contamination or interferences to the samples. Method 3550 for using ultrasonic extraction was used. 10gram each of well homogenized samples were weighed into three different solvents rinsed extraction bottles and labelled site A1, A2 and A3 respectively. 50ml of the extraction solvent i. e methylene chloride was added to the respective samples in induction bottles labeled A1, A2 and A3 respectively.

The respective samples were sonicated for about 30 min 70 °C. The respective extracts were filtered through a glass funnel with glass wool and anhydrous sodium sulphate. The respective extracts were allowed to concentrate to 1ml in a fume wood and stored in a Teflon-lined screw cap for GC analysis. The vial bottles were placed on the injection chamber of the GC and µL was injected into the GC-FID set-up and the result was calculated from the calibration using CHEM 32 software.

RESULTS AND DISCUSSION

Tables (1 to 4) reflect concentration (mg/kg) of PCBs in the respective congener forms along with respective total concentrations (mg/kg) of PCBs. The detected congeners are fifteen in each site. Where, Site A, B, C and are Transformer at Donald Ekong Library Complex, Transformer at the senate Building, Transformer close to the music department and Transformer at Gana-ama staff quarters, respectively.

The total concentration of PCBs in tables 1, 2, 3 and 4 are troubling i.e. 45.561 mg/kg, 18.757 mg/kg, 14.568 mg/kg and 93.04 mg/kg, respectively. Under the toxic substance control act (TSCA), which regulates the PCBs in the environment, stipulates that any soil or sediments containing PCBs ≥ 2 mg/kg that are spilled after 1978 from a source ≥ 50 mg/kg or a source unauthorized for use are

regulated as PCB remediation waste (Cook and USEPA, 2012; Davila, et al., 1993).

Therefore, from the results obtained, all the sites contain PCBs ≥ 2 mg/kg and are in need of PCBs remediation. The decreasing levels in the toxicity of PCBs in the sites is Site D > Site A > Site B > Site C. The total concentration of detected PCBs in Site D is about two times the total concentration in Site A, five times the total concentration in site B and six times the total PCBs concentration in Site C. The total concentration of PCBs in Site A is about 2.4 times the total concentration of PCBs in Site B. A closer look at all the tables will give information about the biodegradability and persistence of the PCBs congeners in the environment of these sites.

Generally, in the entire table, one can observe that the concentration of PCBs congener increases with higher chlorine content of the PCB congener. Apart from site A, other sites i.e. (B, C and D) have decachlorobiphenyls congener having the highest concentration of 4.297 mg/kg, 3.974 mg/kg and 13.79 mg/kg, respectively. This implies

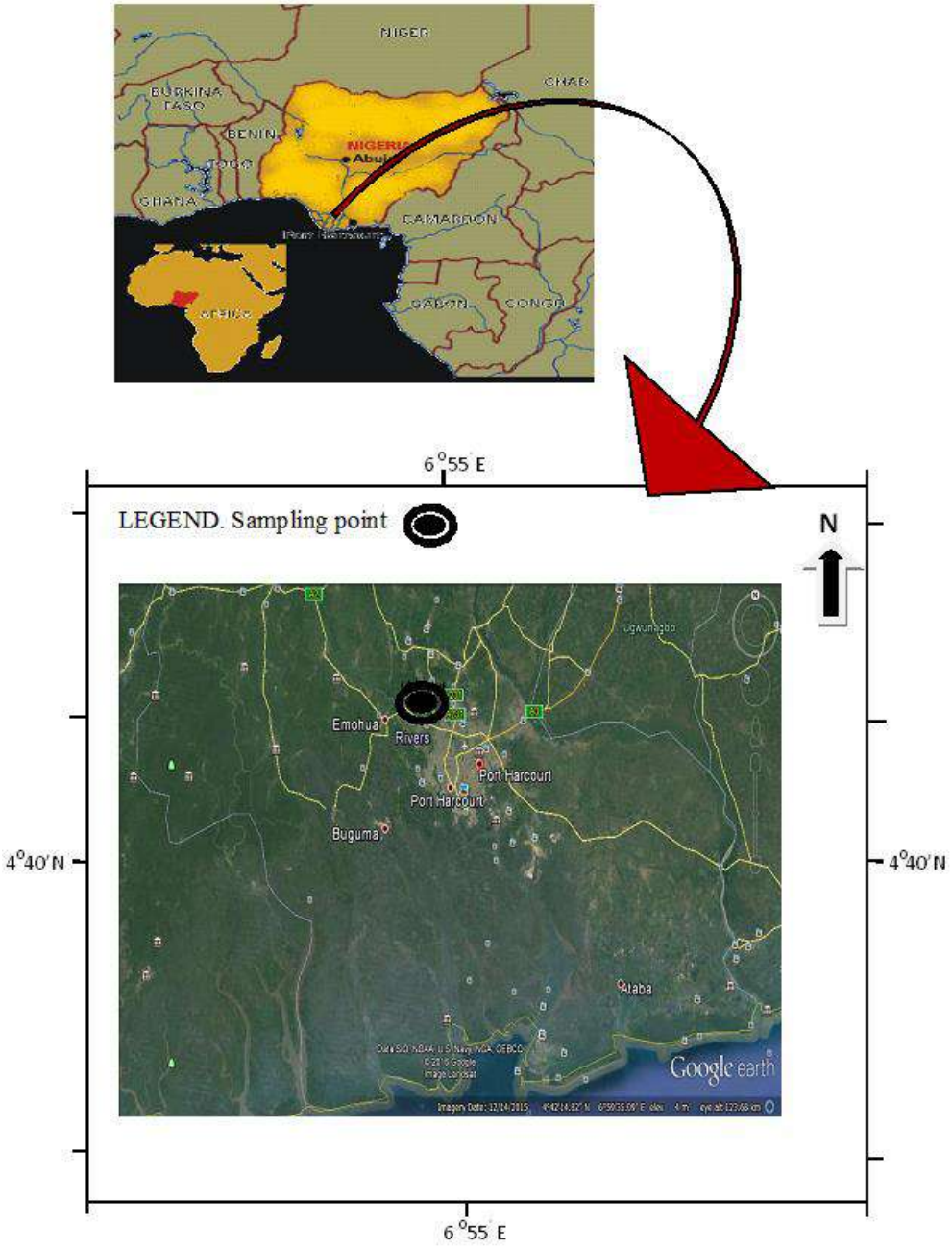

Fig. 1: Map of Port Harcourt, Nigeria indicating sampling point (Port Harcourt) (Google Earth, 2019)

Table 1: Concentration of PCBs in congener at site A

Nos	Detected PCBs	mg/kg
1	4,4'-Dichlorobiphenyl	0.0802
2	2,3,4,4',5-Pentachlorobiphenyl	1.133
3	2,2',3,3',4,5-Hexachlorobiphenyl	1.719
4	2,2',3,4,4'5-Hexachlorobiphenyl	2.795
5	2,2',4,4',5,5'-Hexachlorobiphenyl	1.881
6	2,2',3,3',4,4',6-Heptachlorobiphenyl	2.004
7	2,2',3,4,4',5',6-Heptachlorobiphenyl	3.084
8	2,2',3,4,5,5',6-Heptachlorobiphenyl	1.042
9	2,3,3',4,4',5,5'-Heptachlorobiphenyl	2.896
10	2,3,3',4,4',5',6-Heptachlorobiphenyl	8.792
11	2,2',3,3',4,5',6,6'-Octachlorobiphenyl	5.841
12	2,2',3,3',4,5,5',6,-Octachlorobiphenyl	1.573
13	2,2',3,4,4',5,5',6,-Octachlorobiphenyl	4.379
14	2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	4.193
15	Decachlorobiphenyl	4.220
	Total	45.561

Table 2: Concentration of PCBs in congener at site B

Nos	Detected PCBs	mg/kg
1	4,4'-Dichlorobiphenyl	0.0608
2	2,3,4,4',5-Pentachlorobiphenyl	0.642
3	2,2',3,3',4,5-Hexachlorobiphenyl	0.419
4	2,2',3,4,4'5-Hexachlorobiphenyl	0.399
5	2,2',4,4',5,5'-Hexachlorobiphenyl	1.408
6	2,2',3,3',4,4',6-Heptachlorobiphenyl	1.061
7	2,2',3,4,4',5',6-Heptachlorobiphenyl	1.185
8	2,2',3,4,5,5',6-Heptachlorobiphenyl	0.713
9	2,3,3',4,4',5,5'-Heptachlorobiphenyl	0.536
10	2,3,3',4,4',5',6-Heptachlorobiphenyl	0.842
11	2,2',3,3',4,5',6,6'-Octachlorobiphenyl	1.586
12	2,2',3,3',4,5,5',6,-Octachlorobiphenyl	0.113
13	2,2',3,4,4',5,5',6,-Octachlorobiphenyl	3.538
14	2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	1.950
15	Decachlorobiphenyl	4.297
	Total	18.757

the most stable, persistent or lack of biodegradability PCBs congener are in site D followed by site A, followed by site B and C, respectively when the congeners of octachlorobiphenyls and nonachlorobiphenyls are added together in each of the sites. The lack of biodegradability of PCBs compounds implies high bioaccumulation of PCBs in the environment and as a result, it possesses great environmental and serious health risks to human and living organisms.

The works (Walkowiak et al., 2001; ATSDR, 2000; Sweirenga, 19900 showed in-vitro studies that PCBs, especially with higher chlorine content are biopersistent and active through the Ah receptor mechanism. Their studies further demonstrated various toxicological features, carcinogenicity, tumor promoter-type effects, endocrine disruption, teratogenicity and possible impairment of psycho-development in human and animals as well. Estimation of accurate toxicity and assessing the carcinogenic risk for mixtures of PCBs is best done with use of the congener forms (Cook and USEPA, 2012; Davila et al., 1993) rather than the total PCBs (Aroclors) using their Toxicity Equivalent Factor (TEF). The Department of Ecology in 2007 noted that the contribution of dioxin-like PCB is important in evaluation of risks related to PCBs release and in the development of PCBs cleanups level, because any evaluation of Aroclors or total PCBs would only lead to misinformation on the environmental matrix.

To establish and determine compliance with cleanup and remediation levels for environmental matrix like soil for PCBs under the Model Toxic Control Act (MTCA) cleanup regulation, the soil matrix is considered a single hazardous substance and the target cancer risk level of one in one million (10^{-6}) is used in estimating cleanup levels under method B. The 2006 Toxicity Equivalent Factor (Table 5) is used to evaluate the carcinogenicity of 12 dioxin-like PCBs. The Cancer Potency Factor of 2, 3, 7, 8-TCDD (tetrachlorodibenzo-p-dioxin) with a method B cleanup level 13 ppt (13 ng/kg) or 13 x 10^{-6} mg/kg =1.3 x 10^{-5} mg/kg is used as shown in Eq. 1.

$$TTEC/TEQ = \Sigma C_n \times TEF_n \quad (1)$$

Where, C_n = Concentration of individual congener in the soil mixture, TEF_n = Toxic Equivalent Factor of the individual congener, TTEC = Total Toxicity Equivalent Concentration and TEQ = Toxicity Equivalence. The 2006 Van Der Berg TEF is used in this work because it is the most update relative to the 1998 TEF for dioxin-like PCBs (Table 5). The PCB congeners that are affected in the whole fifteen detected PCB congeners are 2,3,4,4',5 pentachlorobiphenyl (114) and 2, 3, 3', 4, 4', 5, 5'-peptachlorobiphenyl pentachlorobiphenyl (189). Applying their respective TEF 2006 value and computing using Eq. 1, the TTEC for all sites could be calculated. For site A, the TTEC is 0.00012 mg/kg, For site B, the TTEC is 0.000035 mg/kg, For site C, the TTEC is 0.0000185 mg/kg and For site D, the TTEC is 0.00039 mg/kg.

Again the values for the sites exceeded the methods B cleanup levels for 2, 3, 7, 8-tetrachloro-dibenzo-p-dioxin, i.e., 1.3 x 10^{-5} mg/kg and therefore are needed for massive clean-up for carcinogenic dioxin-like PCBs. The carcinogenicity followed similar trend for the Aroclor trend i.e. the concentration of carcinogenic dioxin-like PCBs decreases in the sites accordingly site D > Site A > site B > site C. In view of the aforementioned toxicological symptoms of Dioxin-like and non-Dioxin-like PCBs, i.e. tumor promoter-type effects, endocrine disruption, teratogenicity, carcinogenic effect and given its little

Table 3: Concentration of PCBs in congener at site C

Nos	Detected PCBs	mg/kg
1	4,4'-Dichlorobiphenyl	0.133
2	2,3,4,4',5-Pentachlorobiphenyl	0.333
3	2,2',3,3',4,5-Hexachlorobiphenyl	0.158
4	2,2',3,4,4'5-Hexachlorobiphenyl	0.385
5	2,2',4,4',5,5'-Hexachlorobiphenyl	0.441
6	2,2',3,3',4,4',6-Heptachlorobiphenyl	0.285
7	2,2',3,4,4',5',6-Heptachlorobiphenyl	0.888
8	2,2',3,4,5,5',6-Heptachlorobiphenyl	0.368
9	2,3,3',4,4',5,5'-Heptachlorobiphenyl	0.287
10	2,3,3',4,4',5',6-Heptachlorobiphenyl	1.608
11	2,2',3,3',4,5',6,6'-Octachlorobiphenyl	0.982
12	2,2',3,3',4,5,5',6,-Octachlorobiphenyl	0.093
13	2,2',3,4,4',5,5',6,-Octachlorobiphenyl	3.393
14	2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	1.231
15	Decachlorobiphenyl	3.974
	Total	14.568

Table 4: Concentration of PCBs in congener at site D

No	Detected PCBs	mg/kg
1	4,4'-Dichlorobiphenyl	0.866
2	2,3,4,4',5-Pentachlorobiphenyl	8.390
3	2,2',3,3',4,5-Hexachlorobiphenyl	5.300
4	2,2',3,4,4'5-Hexachlorobiphenyl	5.415
5	2,2',4,4',5,5'-Hexachlorobiphenyl	4.693
6	2,2',3,3',4,4',6-Heptachlorobiphenyl	6.947
7	2,2',3,4,4',5',6-Heptachlorobiphenyl	4.639
8	2,2',3,4,5,5',6-Heptachlorobiphenyl	4.395
9	2,3,3',4,4',5,5'-Heptachlorobiphenyl	4.775
10	2,3,3',4,4',5',6-Heptachlorobiphenyl	4.111
11	2,2',3,3',4,5',6,6'-Octachlorobiphenyl	10.261
12	2,2',3,3',4,5,5',6,-Octachlorobiphenyl	3.499
13	2,2',3,4,4',5,5',6,-Octachlorobiphenyl	7.976
14	2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	7.977
15	Decachlorobiphenyl	13.79
	Total	93.04

biodegradability. It is pertinent for the relevant authorities to remediate and cleanup the transformer locations to avoid further direct human contact and toxicity. It is imperative for these now confirmed PCB transformers to be replaced by non-PCB releasing transformers. The concentrations of PCBs in the soil at various sites indicated that their age is a contributing factor to the accumulation of PCBs in the soil. The earlier installed transformers showed higher concentration of PCBs than the later installed ones.

It is quite remarkable that this research findings in Choba, Port Harcourt clearly contrast earlier works of Eromosele and Iwuoha (2016) in Rumuagholu's area of Port Harcourt metro-polis, where PCBs (Aroclor) toxicity in the soil around transformer areas were not established and therefore in no need of remediation in area under evaluation using relevant approaches (Gomes et al., 2015; Hayat et al., 2019; Huang et al., 2018; Li et al., 2018; Monfort et al., 2019; Needham and Ghosh, 2019; Terzaghi et al., 2018; Terzaghi et al., 2019; Viisimaa et al., 2013; Xu et al., 2019).

CONCLUSIONS

The result of total PCBs concentration in soils around the transformers at all the four sites exceeded the maximum required baseline of 2 mg/kg for cleanup. Calculations of the toxicity (Carcinogenicity) of detected Dioxin-Like PCBs confirms the initial trend of the toxicity of the sites, whereby Site D > Site A > Site B > Site C. The results indicates presence of PCBs congeners that lack biodegradability and are therefore more bioaccumulating in living tissues due to higher concentrations of PCBs congeners with higher chlorine content in the biphenyl ring. Vast clean-ups and remediation of all the soil sites of PCBs to get rid of biopersistent or bioaccumulative PCBs and if necessary, replace all the transformers with modern ones is strongly recommended.

Table 5: Changes in TEFs values for human risk assessment of Dioxin-like PCBs

Nos	PCB Nos	TEF[a]	TEF[b]
1	PCB 77	0.0001	0.0001
2	PCB 81	0.0001	0.0003
3	PCB 126	0.1	0.1
4	PCB 169	0.01	0.03
5	PCB 105	0.0001	0.00003
6	PCB 114	0.0005	0.00003
7	PCB 118	0.0001	0.00003
8	PCB 123	0.0001	0.00003
9	PCB 156	0.0005	0.00003
10	PCB 157	0.0005	0.00003
11	PCB 167	0.0001	0.00003
12	PCB 189	0.0001	0.00003

PCB = Polychlorinated Biphenyl
a = Van der Berg et al. (1998); b = Van der Berg et al. (2006)

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