Human Health Risk Assessment of The Levels of Dioxin-Like Polychlorinated Biphenyls (PCBs) in Soils From Mechanic Workshops Within Nekede Mechanic Village, Imo State, Nigeria

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Abstract

Dioxin-like polychlorinated biphenyls (DL-PCBs) are a class of persistent toxic substances with a high potential to accumulate in the soil as an organic pollutant and bioaccumulate in humans. The study determined the level and human health risk of twelve (12) DL-PCBs in soils from mechanic workshops within Nekede mechanic village (NMV), Imo State. In this study, soil samples (0–15 cm depth) were collected at four different sampling points within Nekede mechanic village. At each sampling point, composite samples consisting of 10 sub-samples were collected with pre-cleaned pet bottles using soil auger and were labeled SA, SB, SC, SD. Control sample was taken 500 metres away from the mechanic village where there was no form of auto-mechanic activities. Sample analysis was conducted using Agilent GC 6890N coupled with Agilent MS 5970B to determine the levels of DL-PCBs. The data were analyzed using SPSS version 23. The results revealed that the mean concentration of 12 DL-PCB congeners (Σ12PCBs) in soil ranged from 0.31–16.31 μg kg^{-1}, which was much higher than that in the control sample (0.006–0.087 μg kg^{-1}). PCB-126 was the major contributor (58.99%) while the least contributor was PCB-157 (0.0040%) of the Σ12 DL-PCBs TEQ. The estimated LCR (adults 1.29×10^{1}, children 4.42×10^{1}) and HQs (adults 6.68×10^{2}, children 2.29×10^{3}) were above the acceptable risk limit (LCR = 10^{-4}, HQ = 1); indicating a very high adverse effect of DL-PCBs on humans especially children.

1. Introduction

The impact of wastes from auto-mechanic workshops on health and the environment has been an issue in developing countries over the years (Nwachukwu and Huan, 2010; Okoro et al., 2013; Mahmood *et al.*, 2014; Al-Wabel *et al.*, 2016). Auto-mechanic workshops are sources of environmental pollution through which persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), Hexachlorobenzene and polyaromatic hydrocarbons (PAHs) are introduced into the environment through soil and groundwater at levels that are considered to be above their threshold limit (Bentum, 2012; Ibe *et al.*, 2021). Polychlorinated Biphenyls has been in existence since 1864 although they were first used globally in 1929 (Bentum, 2012; Zhang, 2013). Even though the use of PCBs has been heavily restricted and are no longer manufactured, they can still be found in the environment. The presence of PCBs in the environment could bioaccumulate within the food chain, due to their high affinity for organic materials (Scheter et al., 2006). Besides, they have been found in human tissues, blood, and breast milk, and are introduced via the consumption of meat, fish, and dairy products (Van den Berg et al., 2006). Also, due to the toxic characteristics of PCBs in the soil, they have been linked to chronic effects in humans including immune system damage, decreased pulmonary function, bronchitis, and interferences with hormones leading to carcinogenic and non-carcinogenic health risks to the populace (Scheter et al., 2006). The International Agency for Research on Cancer (IARC) and the Environmental Protection Agency (EPA) classify PCBs as probable human carcinogens. The National Toxicology Program (NTP) concluded that PCBs are reasonably likely to cause cancer in humans (Van den Berg et al., 2006; USEPA, 2007). Moreover, the effect of PCBs on human health is dependent upon the concentrations of PCBs, type of chlorinated homology and the extent of exposure (Wang et al., 2012; Kumar *et al.*, 2014; Al-Wabel *et al.*, 2016; Enyoh and Isiuku, 2020; Eze et al., 2020a; Eze et al., 2021).

In Nigeria, it has been established that industrialization and urbanization have led to the migration of humans from rural to urban areas in search of greener pastures, and this has led to an increase in the means of transporting people from one point to another (Okoro et al., 2013; Duru *et al.*, 2021). One common mode of transportation is the use of vehicles and in effect, this has led to an upsurge in the number of vehicles available and used in urban areas. At some point, these vehicles require servicing, repairing and replacing of damaged parts; which are carried out in auto-mechanic workshops by auto-mechanic artisans (Nkwoada and Amakom, 2018; Duru *et al.*, 2021; Ibe *et al.*, 2021). In most cities in Nigeria, vehicular repair locations often referred to as "auto-mechanic villages" are allocated for auto-mechanic works in other to prevent traffic congestion on the highways (Duru et al., 2017; Duru et al., 2019; Ibe *et al.*, 2021). The auto-mechanic villages are mainly sited in neighbourhoods of cities and adjourning towns (Duru et al., 2019; Ibe *et al.*, 2021). In such areas, the spills from lubricants, gasoline and diesel oil constitute the major contaminants of the soil and groundwater within the area (Nkwoada and Amakom, 2018). Additionally, malfunctioning vehicular parts are discarded and abandoned on the surface of the soil, which leads to increased soil and groundwater contamination through the leaching of persistent organic pollutants such as PCBs, PAHs and potentially toxic elements (heavy metals) (Bentum, 2012; Kumar *et al.*, 2014; Duru *et al.*, 2020; Ibe *et al.*, 2021). The need for this study became necessary owing to the paucity of PCB data in the study area. Many studies have been carried out by Nigerian researchers on the impact of auto-mechanic activities on surface soil and groundwater quality concerning potentially toxic elements (Mahmood *et al.*, 2014; Al-Wabel *et al.*, 2016; Duru et al., 2017; Duru et al., 2019; Duru *et al.*, 2021; Ibe *et al.*, 2021). There are few documented studies by Nigerian researchers on persistent organic pollutants (Okoro et al., 2013; Nwoko *et al.*, 2017; Nkwoada and Amakom, 2018; Ibe *et al.*, 2021). However, there was no reference to dioxin-like PCBs in the present study, and this study is aimed at filling the knowledge gap. This is the first report on levels and human health risk assessment of dioxin-like PCBs in soils from Nekede mechanic village, Imo State, Nigeria.

2. Materials And Methods

2.1. Study area

The Nekede mechanic village (NMV) is situated in Nekede, Owerri West Local Government Area of Imo State, and located geographically on longitude 7°2'15" – 7°3'00"E and latitude 5°27'80" – 5°28'00"N (Fig. 1). It is about 130 acres and was set up in the year 1983 to repair and maintain automobile vehicles used in the transportation of humans, animals and goods (Onweremadu and Duruigbo, 2007; Nwachukwu et al., 2010; Nwoko *et al.*, 2017). The soils of this area are drained by Nworie and Otamiri Rivers and underlain by the sandy Benin formation of Miocene to recent age, and consists of coastal plain sands (about 0.05–2.0 mm in size) with minor clay beds; this type of soil has good drainage and is well aerated, causing it to dry out quickly (Onweremadu and Duruigbo, 2007; Isiuku and Enyoh, 2020; Duru *et al.*, 2021). The topography of NMV is relatively a level ground but has been sculpted by erosion, forming deep gullies towards the Otamiri River. The climate of the area is tropical rainy, with two distinct seasons, i.e. rainy season and dry season with mean annual rainfall (2500–4000 mm) and daytime temperature that ranges from 18 to 34°C (Nduka et al., 2013; Nwoko *et al.*, 2017; Eze et al., 2020a; Duru *et al.*, 2021).

2.2. Samples collection and Pre-treatment

Soil samples (0–15 cm depth) were collected at four different sampling points within Nekede mechanic village. At each sampling point, composite samples consisting of 10 sub-samples were collected with pre-cleaned pet bottles using soil auger and were labeled SA, SB, SC, SD. Samples from the same point were air-dried, debris removed, and then homogenized to form a representative sample. A control sample was taken 500 metres away from the mechanic village where there was no form of auto-mechanic activities. The samples were immediately transported to the laboratory for analysis. The coordinates of the sampling points were referenced with the Garmin GPSMAP 76, a handheld global positioning system (GPS) unit, and was used in the map generation (Fig. 1). The soil samples were collected in June 2021. In all, a total of forty sub-samples were collected for this study.

2.3 PCBs extraction and clean-up

Analysis of the twelve dioxin-like PCB congeners (PCB-77, PCB-81, PCB-105, PCB-114, PCB-118, PCB-123, PCB-126, PCB-156, PCB-157, PCB-167, PCB-169, and PCB-189) in soil samples was conducted using Agilent GC 6890N coupled with Agilent MS 5970B. 10.0 g of the sample was mixed with 30 ml of acetone/hexane mixture (1:1 v/v) and ultrasonicated at 50°C for 30 min. The contents were filtered through a 0.45 μm pore size filter and the sonication process repeated three times with fresh portions of the acetone/hexane mixture. The extracts were then combined and concentrated to 2 ml with a rotatory vacuum evaporator and then purified with a multi-layer silica gel/alumina glass column. The multi-layer column was then loaded from bottom to top with neutral silica gel (4 g, 5% deactivated), neutral alumina (2.0 g, 6% deactivated) and anhydrous sodium sulphate (5.0 g). The PCBs were finally eluted with 20 ml of the acetone/hexane mixture (1:1 v/v) and concentrated to approximately 1 ml under a gentle stream of high purity nitrogen gas.

2.4 Quality assurance and quality control (QA/QC)

All analytical processes were strictly monitored using quality assurance and control measures in addition to using high purity analytical reagents and solvents. The instruments were checked with calibration standards. The analytical method validation was performed by procedure blanks, triplicate analysis and the analysis of certified reference materials (CRM). The method limit of detection (LoD), repeatability, reproducibility, accuracy, and precision were determined for each organic contaminant of the soil sample. The limits of quantification ranged from 0.004 to 0.006 μg kg^{-1}, depending on the specific congeners. Internal standards were added to the samples before extraction and recoveries and ranged from 90.2–106% which were very good according to Chinese standard HJ 743–2015. For quantification, calibration curves were constructed for PCB congeners using a series level of concentrations that cover the dynamic range in which the targeted compounds are expected to be present.

2.5 Data analysis

Data analyses were performed using statistical software package SPSS version 23.0. Test of significance for variability in the distribution of PCB congeners among different sampling points was carried out using one-way analysis of variance (ANOVA).

2.6 Calculation of toxicity equivalent quotient (TEQ)

In this study, the procedure described by the World Health Organization (Van den Berg et al., 2006) was adopted for risk assessment of the dioxin-like PCBs (DL-PCBs) contamination in soils from mechanic workshops within Nekede mechanic village, Imo State, Nigeria. The toxicity equivalence factors (TEF) with reference to 2,3,7,8,-TCDD and toxicity equivalent quotient (TEQ) were used to expedite the monitoring and toxicity risk assessment of the DL-PCBs (Table 1). The TEQ values of PCBs were calculated by multiplying the detected concentration of individual analytes with their corresponding toxicity equivalence factors provided by the World Health Organization (Van den Berg et al., 2006). The TEQ was calculated using Eq. 1.

$TEQ = (C_i \times TEF_i)$ *(1)*

Where C_i is the individual dioxin-like PCBs concentration (μg kg^{-1}) in the soil sample, and

TEF_i is the TEQ factor assigned to individual dioxin-like PCBs.

Table 1
TEF, CSF and RfD values for human health risk assessment

PCB Congener	TEF	Oral Cancer Slope Factor (CSF)	Oral Reference Dose (RfD)	Reference
PCB-77	1×10^{-4}	1.5×10^{5}	1.0×10^{-5}	Van den Berg et al., 2006
PCB-81	3×10^{-4}	1.5×10^{5}	3.3×10^{-6}	Van den Berg et al., 2006
PCB-105	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-114	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-118	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-123	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-126	1×10^{-1}	1.5×10^{5}	1.0×10^{-8}	Van den Berg et al., 2006
PCB-156	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-157	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-167	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006
PCB-169	3×10^{-2}	1.5×10^{5}	3.3×10^{-8}	Van den Berg et al., 2006
PCB-189	3×10^{-5}	1.5×10^{5}	3.3×10^{-5}	Van den Berg et al., 2006

2.7 Health risk assessment

Humans (adults and children) may be exposed to PCB contaminated soils through different intake routes. As a result of the carcinogenicity of PCBs and intake of contaminated soil through ingestion, adults and children may get exposure to PCBs. Therefore, DL-PCBs exposure assessment was carried out by estimating the lifetime average daily dose (LADD) followed by lifetime cancer risk (LCR) and non-cancer risk (hazard quotient, HQ) for adults and children. In this study, soil ingestion was considered as the main exposure route of life-long intake of PCB contaminants in humans. Lifetime cancer risk (LCR) and hazard quotient (HQ) were assessed by calculating the lifetime average daily dose (LADD) of the DL-PCBs. Equations (2,3,4) were used for estimating LADD, LCR and HQ respectively.

$$\text{LADD } (\text{mg kg}^{-1}\ \text{d}^{-1}) = (C_s \times IR \times CF \times EF \times ED) / (BW \times AT) \quad (2)$$

$$\text{Lifetime Cancer Risk (LCR)} = \text{LADD} \times \text{Cancer Slope Factor}_{oral}\ (CSF_{oral}) \quad (3)$$

$$\text{Hazard Quotient}_{oral}\ (HQ_{oral}) = \text{LADD}/\text{RfD} \quad (4)$$

Where, C_s is the pollutant concentration in soil (µg kg^{-1}), IR is the soil ingestion rate (mg kg^{-1} day^{-1}), CF is the unit conversion factor (10^{-6} mg kg^{-1}), EF is exposure frequency (days/year), ED is the lifetime exposure duration (years), BW is the body weight (kg), and AT is the averaging time for carcinogens (EF×ED days). CSF_{oral} is an oral cancer slope factor (mg kg^{-1} day^{-1}). RfD is the reference dose of individual DL-PCBs (mg kg^{-1} day^{-1}). The exposure parameters for human health risk assessment are outlined in Table 2.

Table 2
Exposure parameters for human health risk assessment

Symbol	Parameter	Unit	Estimate	References
C_s	Concentration	µg kg^{-1}	-	Kumar *et al.*, 2014
IR	Soil ingestion rate	mg day^{-1}	Adult = 100 Children = 200	Kumar *et al.*, 2014
EF	Exposure frequency	days/year	365 days/year	Kumar *et al.*, 2014
AT= (EF×ED)	Lifetime	days	Adult = 25,550 Children = 4382	Kumar *et al.*, 2014
ED	Exposure Duration	years	Adult = 70 Children = 12	Kumar *et al.*, 2014
BW	Body Weight	kg	Adult = 60 Children = 35	Kumar *et al.*, 2014

3 Results And Discussions

3.1 Concentration of dioxin-like polychlorinated biphenyls (DL-PCBs) in soils

The concentrations (μg kg^{-1}) of the dioxin-like PCB congeners in soil samples from Nekede mechanic village are represented in Table 3. Twelve DL-PCBs were detected in all the samples, which showed no significant differences ($p > 0.05$). The mean concentration of 12 DL-PCB congeners (Σ12PCBs) in soil ranged from 0.31–16.31 μg kg^{-1}, which was much higher than that in the control sample (0.006–0.087 μg kg^{-1}). The highest Σ12PCBs was observed in sample SC while the lowest was observed in sample SB. This was attributed to poor management in the handling of lubricants, gasoline and diesel oil, and malfunctioning vehicular parts discarded on the surface of the soil containing DL-PCBs. In comparison to other studies (Wang et al., 2012; Kumar *et al.*, 2014; Al-Wabel *et al.*, 2016; Kim et al., 2017), the mean concentration of 12 DL-PCB congeners reported in this study was higher. Hyun et al., (2010) noted that soil samples collected within a research station have elevated concentrations of chlorinated homologues compared to other samples, which implies that local pollution activities influence the concentrations in that area.

Table 3

Concentrations (μg kg^{-1}) of dioxin-like PCB congeners in soil samples

DL-PCB congeners	Sample SA	Sample SB	Sample SC	Sample SD	Mean	Minimum	Maximum	Std. Dev.
PCB-77	1.78	2.05	1.99	2.50	2.08	1.78	2.50	0.30298515
PCB-81	6.09	5.49	6.66	5.98	6.055	5.49	6.66	0.4803124
PCB-105	5.07	5.45	4.99	5.01	5.13	4.99	5.45	0.21602469
PCB-114	2.31	2.48	3.05	2.32	2.54	2.31	3.05	0.34880749
PCB-118	16.13	15.50	16.31	15.79	15.9325	15.5	16.31	0.36003472
PCB-123	1.24	1.77	1.01	0.89	1.2275	0.89	1.77	0.38973281
PCB-126	2.00	2.32	2.88	2.23	2.3575	2.00	2.88	0.37348583
PCB-156	6.67	5.56	6.08	6.99	6.325	5.56	6.99	0.63416612
PCB-157	0.66	0.31	0.75	0.44	0.54	0.31	0.75	0.20116328
PCB-167	1.54	1.01	1.86	1.94	1.5875	1.01	1.94	0.42200908
PCB-169	5.09	5.82	5.55	4.97	5.3575	4.97	5.82	0.39693618
PCB-189	2.86	2.09	2.22	2.69	2.465	2.09	2.86	0.36846529
Sum of DL-PCBs	51.44	49.85	53.35	51.75	51.5975	49.85	53.35	1.43446564

3.2 Toxicity equivalent quotient (TEQ)

The toxicity equivalent quotients (TEQ) for 12 DL-PCBs at the different sampling points (Fig. 3) were calculated according to the World Health Organization toxic equivalency factors (WHO-TEF) (WHO, 2001; Van den Berg et al., 2006). The 12 DL-PCBs ranged from 1.62×10^{-5} to 2.36×10^{-1} μg kg^{-1}. PCB-126 accounted for 58.99% of the

Σ12 DL-PCBs TEQ, and the other major contributors were PCB-169 (40.24%) and PCB-81 (0.45%). The least contributor was PCB-157 (0.0040%). According to (WHO, 2001; Li et al., 2010; Wang et al., 2012) PCB-126 is the most toxic DL-PCB with relatively higher TEF values compared to the other dioxin-like PCB congeners. In addition, Li and Su (2011) considered PCB-126 as a major contributing factor for the TEQ. It is of importance to note that the Σ12 DL-PCBs TEQ values exceeded the Canadian soil quality guidelines of dioxin (0.004 μg TEQ kg^{-1}). Furthermore, Ma et al., (2009) reported TEQ for humans and mammals which are lower than that of the current study.

3.3 Human health risk of dioxin-like polychlorinated biphenyls (DL-PCBs)

The estimated LADD of DL-PCBs (Table 4) for adults and children ranged from 9.0×10^{-7} μg kg^{-1} to 2.66×10^{-7} μg kg^{-1} and 3.08×10^{-6} μg kg^{-1} to 9.10×10^{-5} μg kg^{-1} respectively. These estimated intakes for adults and children within NMV were higher than the recommended tolerable daily intake (1.0×10^{-4} μg kg^{-1}) set by the agency for toxic substances and disease registry (ATSDR, 2000). The total lifetime cancer risks (adults 1.29×10^{1}, children 4.42×10^{1}) and hazard quotients (adults 6.68×10^{2}, children 2.29×10^{3}) for 12 DL-PCBs were compared with recently conducted studies. Al-Wabel *et al.*, (2016) reported hazard quotients and lifetime cancer risks lower than that of the current study. Also, the estimated LCR and HQs were above the acceptable risk limit (LCR = 10^{-4}, HQ = 1); indicating a very high adverse effect of DL-PCBs on humans.

Table 4
Estimated lifetime average daily dose (LADD), lifetime cancer risk (LCR) and hazard quotient (HQ) for adults and children due to DL-PCBs exposure.

Dioxin-like PCB congener	**Adult**	**Children**	**Adult**	**Children**	**Adult**	**Children**
	LADD		**LCR**		**HQ**	
PCB-77	3.47×10^{-6}	1.19×10^{-5}	0.521	1.785	0.347	1.190
PCB-81	1.01×10^{-5}	3.46×10^{-5}	1.515	5.190	3.061	10.485
PCB-105	8.55×10^{-6}	2.93×10^{-5}	1.283	4.395	0.259	0.888
PCB-114	4.23×10^{-6}	1.45×10^{-5}	0.635	2.175	0.128	0.439
PCB-118	2.66×10^{-5}	9.10×10^{-5}	3.990	13.650	0.806	2.758
PCB-123	2.05×10^{-6}	7.01×10^{-6}	0.308	1.052	0.062	0.212
PCB-126	3.93×10^{-6}	1.35×10^{-5}	0.589	2.025	393.0	1350.0
PCB-156	1.05×10^{-5}	3.61×10^{-5}	1.575	5.415	0.318	1.094
PCB-157	9.0×10^{-7}	3.08×10^{-6}	0.135	0.462	0.027	0.093
PCB-167	2.65×10^{-6}	9.07×10^{-6}	0.398	1.361	0.080	0.275
PCB-169	8.93×10^{-6}	3.06×10^{-5}	1.339	4.590	270.61	927.27
PCB-189	4.10×10^{-6}	1.41×10^{-5}	0.615	2.115	0.124	0.427
Sum of dioxin-like PCB congeners	8.60×10^{-5}	2.95×10^{-4}	12.903	44.215	668.822	2295.131

Conclusion

Dioxin-like PCBs were found in all the soil samples in this study. The mean concentration of 12 DL-PCB congeners (Σ12PCBs) in soil ranged from 0.31–16.31 µg kg^{-1}. The highest Σ12PCBs was observed in sample SC while the lowest was observed in sample SB. PCB-126 was the major contributor (58.99%) while PCB-157 was the least contributor (0.0040%) of the Σ12 DL-PCBs TEQ. Worthy of note is that the Σ12 DL-PCBs TEQ values exceeded the recommended guidelines for soils for the protection of human and environmental health. Furthermore, the total lifetime cancer risks (adults 1.29×10^{1}, children 4.42×10^{1}) and hazard quotients (adults 6.68×10^{2}, children 2.29×10^{3}) for 12 DL-PCBs were higher than the recommended tolerable limits; indicating a very high harmful effect of DL-PCBs in humans. This quantitative evidence demonstrates the need for remediation of soils within mechanic villages to protect residents, especially children from persistent organic pollutants such as DL-PCBs in the environment.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

All data and materials are available.

Competing interests

The authors declare that they have no competing interests.

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Authors' contribution

VCE contributed to writing the original draft of the manuscript. ARN contributed to methodology and investigation. CED assisted in supervision and validation. BOI helped in conceptualization, methodology and statistical data analysis. FCI assisted in conceptualization and visualization. JOO contributed to supervision and validation. CEE contributed to statistical data analysis, review and editing. MOM helped in investigation, review and editing.

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Figures

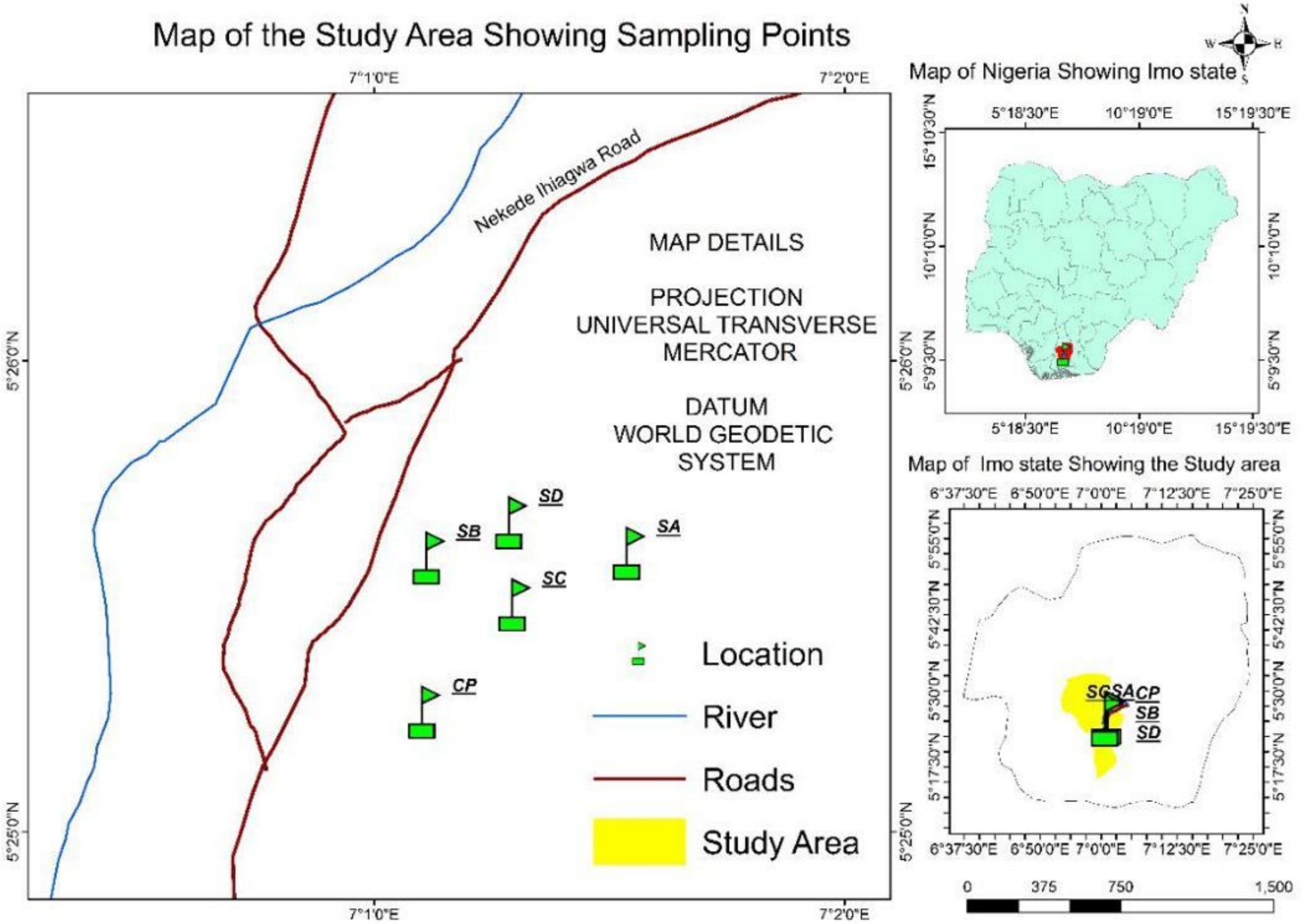
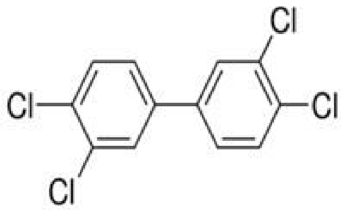


Figure 1

Map of study area showing the sampling points



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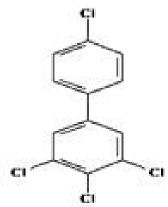

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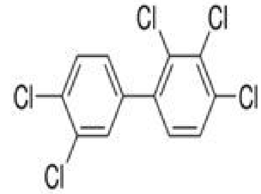

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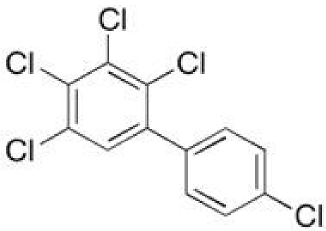

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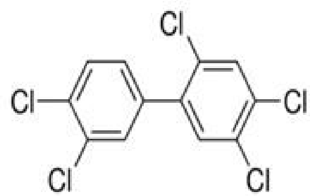

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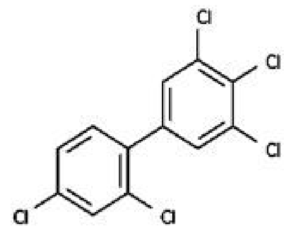

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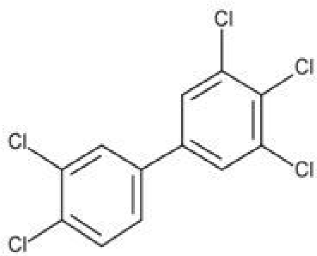

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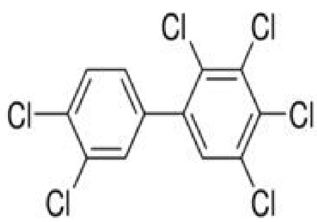

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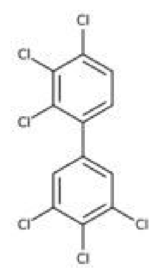

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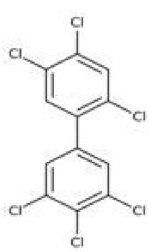

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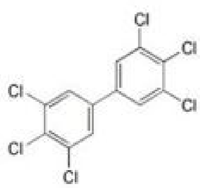

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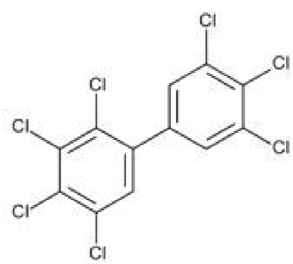

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Figure 2

Chemical structure of 12 dioxin-like polychlorinated biphenyls

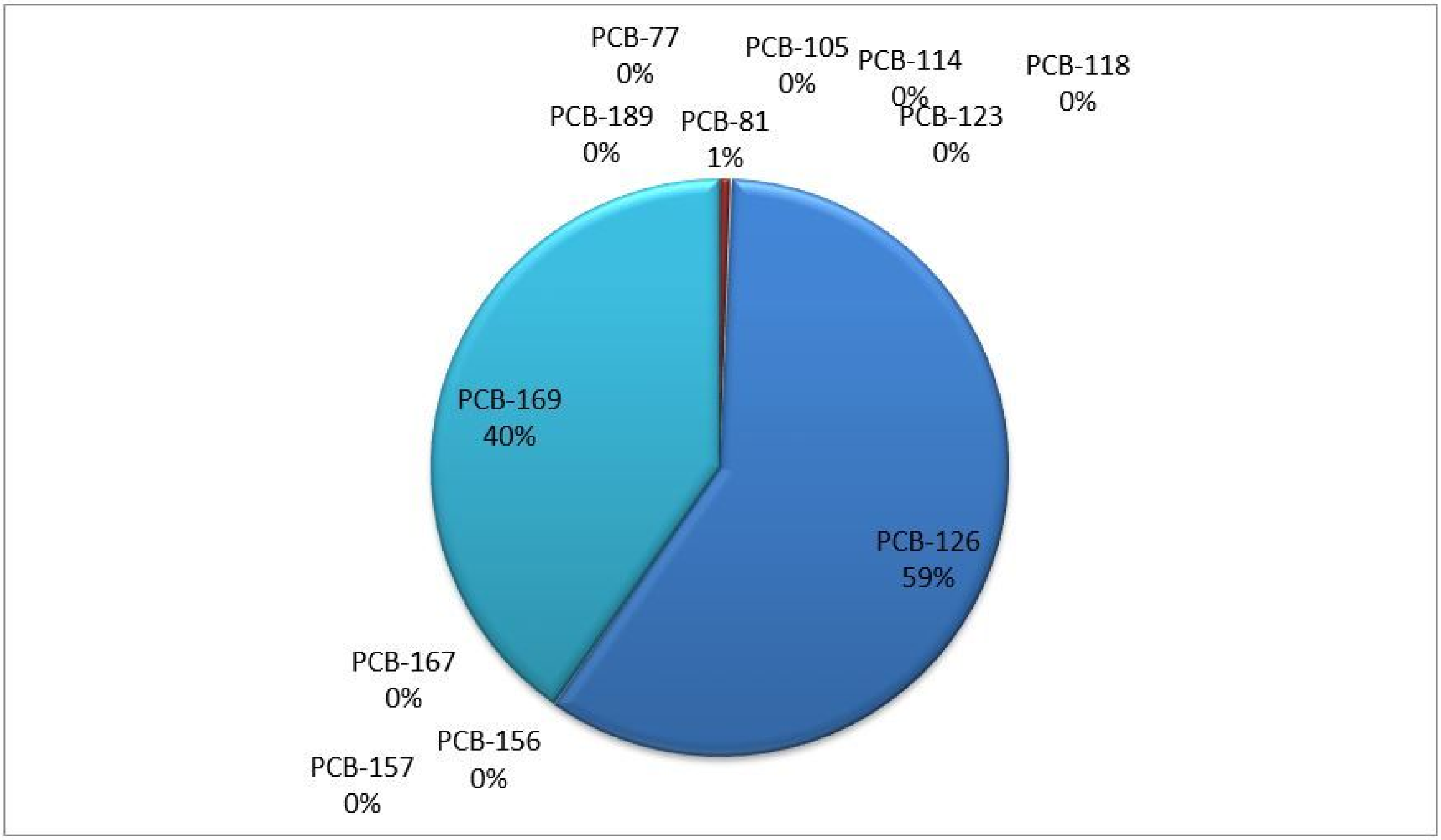

Figure 3

Toxicity equivalent quotients for 12 DL-PCBs at the different sampling points