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Evaluation of Polychlorinated Biphenyls in Sediments from Surface Waters of Igodan, Okunmo, Lebi, Idepe and OAUSTECH in Okitipupa, Ondo State Nigeria

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ABSTRACT: The presence of PCBs in water bodies is an emerging issue for water quality. This is because, once PCBs reach a lake or river, they concentrate in the sediments by attaching to the organic matter. Therefore, the objective of this paper is to evaluate the level of polychlorinated biphenyls (PCBs) in sediment from surface waters of Igodan, Okunmo, Lebi, Idepe and Oaustech in Okitipupa, Ondo State Nigeria using appropriate standard methods. Data obtained reveals that in Igodan the concentration of PCBs in sediment w.as PCB 44 (5.42±0.00), PCB 18 (3.04±0.00), PCB 28 (3.27±0.07) and PCB 170 (6.60±0.00). Okunmo PCBs concentration in sediment was PCB 44 (0.67±0.00), PCB 18 (0.03±0.00), PCB 28 (0.32±0.00) and PCB 170 (0.18±0.00). Lebi PCBs concentration in Sediment was PCB 44 (0.82±0.00), PCB 18 (3.96±0.00), PCB 28 (1.12±0.00) and PCB 170 (1.08±0.00). Except for PCB 170 (0.33±0.02), the PCB concentration in sediment from the Idepe and the OAUSTECH were both below the detection limit (BDL). Correlation between Physicochemical Parameters and PCBs homologs shows a negative correlation for the relationship of EC (Electrical Conductivity), TOC (Total Organic Carbon) and pH at (-0.283), (-0.262) and (-0.246) respectively. According to multivariate investigation, releases from nearby factories, deterioration of electrical cables and transformers, waste from university laboratories and facilities, as well as diesel residues from lister engines could be associated with the elevated levels of PCBs in sediments collected from the five sampled sites in Ondo State. TriPCBs > tetraPCBs were in this order in which the median concentrations of PCB homologues were found in sediments from the surface water.

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Like other developing nations, Nigeria has a significant market for used, refurbished, and second-hand electrical and electronic equipment. These used electronics quickly become e-waste. E-waste management and disposal has been a serious issue in Nigeria, and this might increase PCB concentrations in the environment. Nigeria has not set up any detailed national regulations for tracking and managing PCB releases into various environmental matrices. Given this context, it is necessary to compile data on PCB concentrations and occurrences in the Nigerian environment. With a focus on the effects of urbanization, industrial growth, and agricultural practices on the patterns of dispersion, monitoring PCBs in river sediments provides information on the geographic features of the area. Moreover, it

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demonstrates the efficacy of any controls implemented as well as the response and levels of compliance to prohibited compounds (Irerhievwie *et al*, 2020). A significant group of synthetic organic chemicals that are persistent and widely distributed because of long-distance air travel and deposition are polychlorinated biphenyls (PCBs). In the food chain, PCBs have the potential to biomagnify, bioconcentrate, and bioaccumulate. They are extremely harmful to both people and wildlife. Cancer, reproductive diseases, neurological and endocrine and immune system disturbance have all been linked to exposure to PCBs (Baqar *et al.*, 2017). The Stockholm Agreement, which was signed on May 22, 2001, classified PCBs like usual POPs (Liu *et al.*, 2017). They have been extensively utilised in a variety of industrial products, including printing inks, metallic coating compounds, electrical capacitors, paints, transformers, and others (Baqar *et al.,* 2017; Duan *et al.,* 2013; Iwegbue *et al.*, 2019; Va den Borg *et al.,* 2006; Schecter *et al.,*2006). PCBs are no longer permitted to be used but due to their persistence and legacy usage, such as discharges from outdated transformers and electronic devices, PCBs can still be discovered in environmental matrices, biotas, and human tissues (Iwegbue *et al.*, 2019). They are organic persistent pollutants, meaning that their effects linger long after use. They are frequently employed and use in dielectric fluids for things like transformers, capacitors, coolants, and other uses in agriculture. In all parts of the world, they have been discovered in a wide range of environmental compartments, as well as biota (fish and other benthic creatures), soil, water, air, sediment, plants, and animal tissue. Their claimed half-lives in sediment and soil might be anywhere between months and years. The majority of them are trapped in sediments that act as environmental reservoirs because of their extremely poor solubility in water and low volatility. For instance, electrical transformers that contain PCBs have a life expectancy of 30 years or longer (Wang and Zhong, 2011). Its physicochemical characteristics, such as thermal stability, low flammability, and high permittivity, have made them particularly suited for industrial uses during the past 50 years. These characteristics have also contributed to their persistence, accumulation, and biomagnification across food chain in the environment. They can build up to significant levels in top-of-the-food-chain species, which are present in many areas of the environment. The first step in lowering the amount of pollutants in the environment is to identify their source, their emissions, and their leftovers. PCBs are only man-made, and they are present in every environmental component (Kumar *et al.,* 2013). Due to their long-distance travel, bioaccumulation, persistence, hydrophobicity, and poisonous properties, they constitute a global health problem. PCBs are adsorbed onto particulate matter in an aquatic environment, where they go through sedimentation processes and then accumulate in sediments. Due to its large surface area, sediment has a great ability to retain PCBs and may act as a second source of PCBs for the water body above it due to physical or mechanical turbulence or changes in the geochemical makeup of the sediments (Sakan *et al.*, 2017). Thus, sediment has a major impact on how PCBs are disposed of and cycled globally. Moreover, interplay between sediment, edible fish and benthic invertebrates allow PCBs to reach the human food chain. A few of the sources where PCBs can enter the environment include the discharge of industrial and municipal wastewater, weathering of asphalt, various uses, plasticizers, leaching from landfills, and hydraulic fluids and lubricants. There are also salvage yards in San Antonio, tar roofing materials, auto crushing, repair work, used oil, tar paper, scrap metal recycling, and tar paper. (Ssebugere *et al*., 2013; Zhang *et al.*, 2014). Therefore, the objective of this paper is to evaluate the level of polychlorinated biphenyls (PCBs) in sediments from surface waters of Igodan, Okunmo, Lebi, Idepe and OAUSTECH in Okitipupa, Ondo State, Nigeria.

MATERIALS AND METHODS

Study area: Nigeria's Ikale-speaking region in Ondo State includes the Local Government Area of Okitipupa. Olusegun Agagu University of Science and Technology is a university in Okitipupa Township, which also houses the offices of the Okitipupa Local Government (OAUSTECH). It has a total population of about 316,100 people at the 2016 census, it has an area of 803Km^2. Okitipupa is of economical relevance in Ondo State Nigeria due to vast palm oil production activities, average fishing activities and lumbering activities. Rivers are highly polluted due to chemical and biological inputs in the river, and this could result in concentration of PCBs. Okitipupa is a palm oil producing area which makes it very polluted due to waste of palm oil products into river bodies. Okitipupa over the years has been lacking power supply with abandoned electric cables, transformers and some other electrical mechanics. The sampling locations are shown in Figure 1

Sample collection and storage: Twenty -five Sediment samples were taken from Five (5) surface water in Okitipupa, Ondo State Nigeria. The sample locations are Oaustech, Igodan, Okunmo, Idepe, Lebi. With the help of a stainless Van-Veen grab, sediment was taken as previously described (Okunola *et al.,* 2012). The sediments from each study site were wrapped in aluminium foil and placed in clean polythene bags before being transported to the laboratory and pooled to form a composite sample for each study site. Sediments were air dried and ground to powder using a laboratory mortar and pestle, then sieved through a 2 mm sieve and stored at -20 degrees Celsius until further analysis (Okunola *et al.,* 2012).

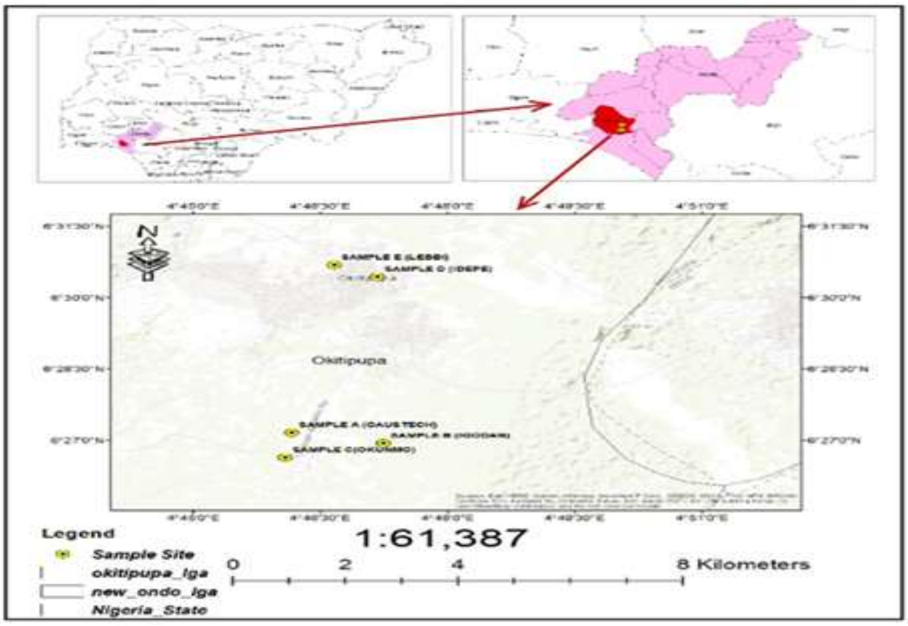

Fig. 1: Showing Sample Locations

Determination of polychlorinated biphenyls (PCBs): GC-ECD Agilent 7820A, was used to determine the concentrations of 28 PCB congeners in the sediment samples. Individual PCB congeners had recovery rates between 87% and 100%. This technique was also described in detail by Igbo *et al*. (2018) and Osuala *et al.* (2019).

Physicochemical Parameters: Standard techniques were used to measure physicochemical characteristics such pH, electrical conductivity, and total organic carbon. (Radojevic and Bashkin, 1996).

Statistical analysis: SPSS version 26 was used to conduct data analyses. Data were submitted to a two-way analysis of variance (ANOVA), and the Duncan Multiple Range test was used to distinguish between significant means (p 0.05). The mean and standard error were used to express the results.

RESULTS AND DISCUSSION

The mean concentrations of polychlorinated biphenyls (PCBs)(µg/kg) from the Five sample areas are shown in Table 1. The highest recorded level of PCBs congeners occurred in Igodan River Sediment. The most dominant indicator PCBs in Igodan River was PCB-170 (6.60 µg/kg) and the lowest in concentration in Igodan River was PCB-18 (3.04 µg/kg). In Okitipupa Ondo State PCB-170 was observed to have the highest level of 6.60 µg/kg in Igodan with the lowest level of 0.33 µg/kg recorded for CB-170 in Oaustech. High PCB concentrations found in sediment has been reported (Wang and Zhong, 2011), suggesting that because of their hydrophobic properties and propensity to accumulate in sediments, persistent organic pollutants are strongly particle associated (Ashraf 2017; Gomez-Gutierrez *et al.,* 2007). Because of their endurance, hydrophobic nature, and poisonous qualities, PCBs are dangerous compounds. PCBs accumulate in sediments because of their persistence and hydrophobicity, where they are likely to be preserved for many years. As a result, sediments serve as an essential reservoir for these chemicals (Quesada *et al.,*2014; Ediagbonya *et al.,* 2022ab; Ediagbonya *et al.,*2020; Ediagbonya *et al.,*2019; Ediagbonya *et al.,*2018ab). Moreover, these chemicals produce a variety of problems with human health, involving neurotoxicity, dermatological, and respiratory illnesses (Motano *et al.,*2022; Ivanscu,2015). Consequently, PCB tests for sediment samples should be pursued further by developing analytical methodologies. The greatest concentration values, with a high degree of chlorination dependent on concentration and amount of organic waste material supplied, and a longer persistence value, are observed for those organic waste materials that supply the highest amounts of PCBs.

Table 1: PCBS congeners and concentrations in sediment from five surface water

Congeners	Igodan	Okunmo	Lebi	Idepe	Oaustech	P
PCB 44	5.42±0.00	0.67±0.00	0.82±0.00	BDL	BDL	<0.001
PCB 18	3.04±0.00	0.03±0.00	3.96±0.00	BDL	BDL	<0.001
PCB 28	3.27±0.07	0.32±0.00	1.12±0.00	BDL	BDL	<0.001
PCB 170	6.60±0.00	0.18±0.00	1.08±0.00	BDL	0.33±0.02	<0.001
Total PCBs	18.33±0.07	1.19±0.00	6.98±0.01	BDL	0.33±0.02	<0.001

The concentrations of ∑- PCBs were in the following decreasing order in Okitipupa river Sample Site: Igodan River > Lebi River > > Okunmo River > Oaustech River > and Idepe River with PCBs Below Detection Limit. Generally, relatively highest PCBs burden was recorded in Igodan River (18.33µg/kg±0.07) than other four sample sites. This was attributed to a higher persistent waste products from oil palm factory, more rain-washed pollutants from abandoned electric cables and transformers in Igodan Community than in Okunmo, Oaustech Campus, Lebi Community & Idepe Community. In general, variation in topographic features, source inputs, flow velocity, tidal action, sediment geochemical parameters, source inputs, and the interaction between PCB transport behaviours, horizontal distribution, and adsorption can be used to explain variations in PCB concentrations in sediments (Barhoumi *et al.,* 2014; Yang *et al.,* 2011; Cui *et al.,* 2016). Igodan and Lebi river sediments in this study contain more PCBs than sediments from the other rivers, on average. The highest levels of PCB170, PCB44, and PCB28 were found in the Igodan, with 6.60 g/kg± 0.00, 5.42 g/kg ±0.00, and 3.27 g/kg ±0.07, respectively, while the highest level of PCB18 was found in the Lebi River at 3.96 g/kg ±0.00. Okunmo River had the lowest PCB170 concentration (0.18 g/kg ±0.00). For PCB44, PCB28, PCB170, and Total PCBs, statistically significant differences were found, indicating significant spatial variation in these PCBs. The physicochemical properties of PCBs determined in sediment samples collected from Five Okitipupa Rivers are reported in Table 2.

Table 2: The mean concentration of physicochemical parameters in sediments from the five sampled areas.

SITE	pH	EC	TOC
OAUSTECH	6.37±0.06	96.67±1.15	1.94±0.02
Igodan	6.48±0.21	67.37±0.55	1.74±0.03
Okunmo	6.41±0.25.	57.33±0.58	1.04±0.01
Idepe	6.45±0.13	74.67±0.58	1.52±0.02
Lebi	6.43±0.06	86.67±2.52	1.60±0.10

The physicochemical parameters measured across the five sampling locations is of different variations. The pH values ranged between 6.37 and 6.48 while EC and TOC values ranged between 57.33 – 96.67 and 1.04 – 1.94 respectively. Okunmo recorded the lowest values of physicochemical parameters while Oaustech River recorded the highest values of physicochemical parameters. The pH, electrical conductivity (EC) and total organic carbon (TOC) of the sediments were determined following standard procedures. The sediments from Oaustech showed higher electrical conductivity values than those from Okunmo, Idepe, Igodan and Lebi. The high electrical conductivity of Oautech is not surprising because these areas are strongly influenced by massive University activities like waste of chemicals from laboratories, university farm activities and waste from university confectionery into the water body. Except for one sample, the distribution of particle sizes reveals that river systems' sediments are generally sandy in character, with a clay component of less than 3%. (Irerhievwie *et al.,* 2020). Increased TOC concentration in sediment can lead to increased bioaccumulation and bioactivity in the water system. The greatest TOC content was found in the Oaustech. The Oaustech sediment absorbed a high volume of effluent from university operations, which is rich in organic materials. Our findings demonstrated that anthropogenic activities are the primary contributors of sediment TOC concentration and can have a significant impact on carbon cycling processes in coastal sediments (Arfaeinia *et al.,* 2017; Ekanem *et al.,* 2019). The pollution accumulates in aquatic creatures after being deposited in sediments. Notwithstanding the fact that PCB manufacture was outlawed in Slovakia in 1984, pollution of bed sediments remains a source of persistent contamination. Because of their high bioaccumulation potential, polychlorinated biphenyls (PCBs) are one of the most harmful contaminants. Polychlorinated biphenyls (PCBs) are prevalent in contaminated area and can be seen in both biotic (from plants to humans) and abiotic (soil, air, water, and sediments) (De Boer *et al.,* 2001). Organic carbon from university activities and industry operations in Igodan and Idepe may have an impact on the movement and dispersion of these pollutants across the Okitipupa rivers (Kim *et al.*, 2009; Vives *et al.,* 2007). Table 3 revealed that tetraPCBs and triPCBs had a substantial positive connection. This implies that a rise in triPCBs will be followed by a rise in tetraPCBs and vice versa.

Table 3: Pearson correlation coefficients for the relationship between the PCB homologs.

	triPCB	TetraPCB
triPCB	1	.679
tetraPCB	.679	1

Light PCB congeners, particularly tetra- and tri- PCBs, dominated PCB composition in sediments from three locations, although OAUSTECH and Idepe Rivers had 0% Tri- and Tetra-PCBs, respectively. Since tri-CBs and tetra-CBs could move downhill, this could be explained by the leaching and deposition of light PCB congeners, which would result in a significant portion of light PCB homologues moving deeper into the sediment or soil (Mai *et al.,* 2002). The simpler breakdown of PCB congeners with no more than three chlorines may be the reason why tri-PCBs made up a lower percentage than other congeners (Bopp, 1986). However, the proportion of low-chlorinated PCBs (tri-CBs and tetra-CBs) increased with depth, according to Zhang *et al.,* (2014). Planar (non-*ortho*) PCB and PCDD/F aryl hydrocarbon (Ah) receptor binding explains why PCBs may augment the genotoxic activity of comparable chemicals, resulting in more severe toxic consequences (Boalt *et al.,* 2014). The author also highlighted their lipophilic properties have the ability to bioaccumulate, especially in lipid-rich organs and tissues, which leads to their putative association with carcinogenesis in living creatures. Aquatic species rapidly acquire them and bioaccumulate them throughout the aquatic food chain. PCBs are often found at higher concentrations in adipose tissue, liver, breast milk and skin. Breast milk contains 90% to 100% of the dose of tetra and higher chlorinated congeners that nursing newborns absorb (Wang and Zhong, 2011). Table 4 Shows negative correlation was recorded for the relationship of pH, EC and TOC at -0.246, -0.283 and -0.262 respectively. However negative correlations show's that the sources are different. The presence of PCBs in sediments is heavily controlled by physicochemical parameters such as pH, Electrical Conductivity (EC), and Total Organic Content (TOC). This has a significant impact on the distribution and mobility of PCBs throughout the polluted environment.

Table 4: Correlation of the Physicochemical Parameters between the PCBs homologs.

	pH	EC	TOC	Total PCBs
pH	1	-0.123	-0.051	-0.246
EC	-0.123	1	.784	-0.283
TOC	-0.051	.784	1	-0.262
Total PCBs	-0.246	-0.283	-0.262	1

Correlation is significant at 0.05 level (2 tailed) -Correlation is significant at 0.01 level (2 tailed)

Table 4 displays the results of physicochemical property measurements. The statistical correlation study performed to explore the association between total PCB concentrations and TOC, as shown in Table 4, revealed a strong negative correlation (r = -0.262). The pH of the sediment was adversely connected with total PCB (r = -0.246), while the EC of the sediment was negatively correlated with total PCB (r =- 0.283). The link between sediment PCB and TOC concentration is noticed. Linear regression associations were discovered between PCB and TOC concentrations (with R values of 1 and -0.262 respectively). These connections suggest that organic carbon may have an impact on the movement and distribution of contaminants in Okitipupa rivers (Vives *et al*., 2007). They allow for the tracking of environmental changes at the local and regional levels, and they can provide a wealth of information on the causes of such changes. Since dangerous material concentrations in sediments are several times greater than in water, chemical examination of sediments may indicate the existence of such chemicals and changes in their concentrations even at low levels of environmental pollution. Polychlorinated biphenyls (PCBs) have been widely employed in electrical appliances, heat transfer systems, and hydraulic fluids. It has been shown that the physicochemical characteristics of sediments, as well as TOC, and clay concentration, significantly affect the fate and transit of PCBs in sediments (Yang *et al.,* 2011; Gao *et al.,* 2013). The lack of a positive relationship between TOC and PCB concentrations in sediments suggests that TOC is not the only factor affecting PCB fate in river sediments (Mai *et al.,* 2005). Additionally, it implies that the influence of TOC and particle size on the fate and dispersion of PCBs in these river systems was obscured by other factors like hydrodynamic setting, source intake, and degradation. PCB concentrations in Okitipupa five sampled area are compared to those found in other locations throughout is shown Table 5. Because of the disparities in the number of samples and congeners examined, the extraction, procedures for instrumental analysis used, and other factors, such a comparison is sometimes challenging. This does not, however, negate the necessity to determine the worldwide prevalence and level of PCBs concentrations in sediments. PCB concentrations were higher in the Igodan than those from other river systems in Africa, such as the Benin River in Nigeria and Monastir Bay in Tunisia (Nouira et al. 2013, Ezemonye 2005). PCB levels are greater in sediments from other parts of the world than in the examined rivers in Okitipupa Ondo State, Nigeria, such as the Chenab in Pakistan (Equani *et al.,* 2012), Tam Giang Lagoon in Vietnam, and the Missouri River in the United States (Frugnani *et al.* 2007; Echola *et* *al.*, 2008). Meanwhile Lake Qarum in Egypt Africa has higher PCBs in sediment than Sediments in study area. Furthermore, the high detection of PCBs in sediments from other regions of the world compared to Nigeria can be linked to drastic migration of PCBs into river bodies from heavy Industrial operations, factories etc. It is observed that PCBs River sediments from some of African regions were lower than most other regions in the world.

Table 5: Comparison of PCBs concentration in sediments from the five sampled rivers in Okitipupa with those reported from other part of the world.

Country/ Location	River	Conc Range (μgKg^{-1})	Reference
Nigeria	Igodan River	18.33±0.07	This Study
Nigeria	Okunmo River	1.19±0.00	This Study
Nigeria	Lebi River	6.98±0.01	This Study
Nigeria	Idepe River	BDL	This Study
Nigeria	Oaustech River	0.33±0.02	This Study
United Kingdom	Thames River	0.12-27.4	Lu *et al.* (2017)
United States	Missouri River	11.0-250	Echols *et al.* (2008)
China	Lianjiang River	4.7-743	Leung *et al.* (2006)
Egypt	Lake Qarum	1.48-137.2	Barak *et al.* (2013)
Nigeria	Benin River	0.35-15.2	Ezemonye (2005)
Indian	Yamuna River	0.21-21.16	Kumar *et al.* (2013)
Cuba	Cienfuegos River	2.50-15.49	Tolosa *et al.* (2010)
Romania	Bahlui River	3-26	Neamtu *et al.* (2009)
Pakistan	Chenab	9.33-144.3	Equani *et al.* (2012)
Tunisia	Monastir Bay	1.1-9.3	Nouira *et al.* (2013)
Vietnam	T am Giang Lagoon	2.03-24.7	Frignani *et al.* (2007)

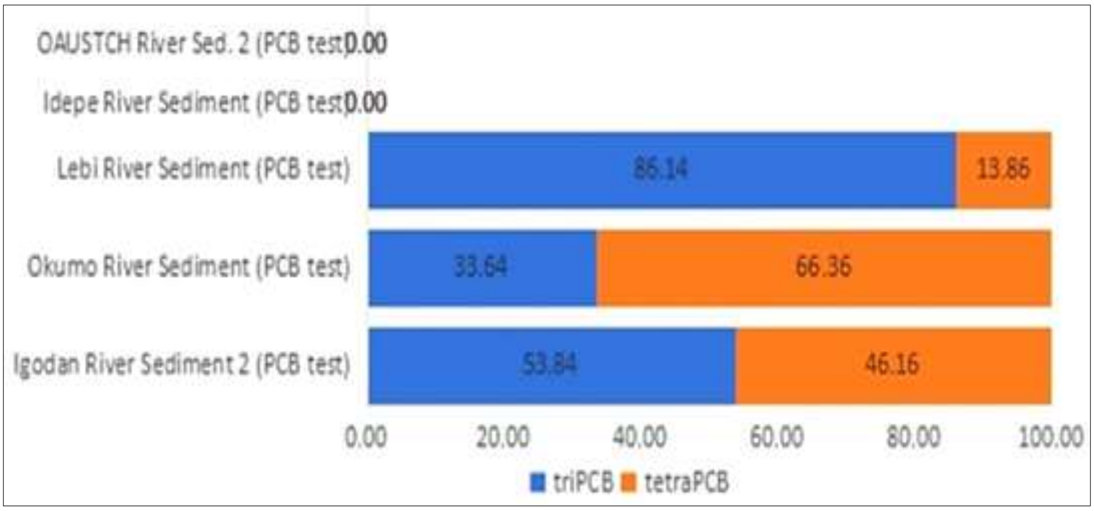

Fig. 2: Distribution of the PCBs Homologous in Sediments

The order of median concentrations of PCB homologues in sediments was Tri-PCBs > Tetra-PCBs. The ∑-Tri-PCBs dominated (86.14%) in Lebi samples with (13.86% Tetra PCBs). Okunmo samples were dominated by ∑-tetra PCBs accounting for about 66.36%. Igodan Samples accounted for about (53.84% dominated Tri-PCBs & 46.16% Tetra-P to CBs). Oaustech and Idepe samples had 0% Tri-PCBs and Tetra-PCBs respectively. Tri-PCBs are thought to predominate in Lebi consequent upon their widespread use in electrical goods (Ren *et al.*, 2007; Zhang *et al.*, 2014). In addition to being widely utilised in transformers and capacitors, tri-PCBs are also employed as paint additives (Li *et al.,* 2014). Nigeria, unlike numerous other nations throughout the world, has not totally phased out the usage of equipment using PCBs. Thus, the dispersion of PCBs is controlled by release from outdated electrical equipment as well as non-point sources such air deposition and surface runoff (Gao *et al.,* 2013). Polychlorinated biphenyls (PCBs) have been produced industrially for many years, and their inappropriate disposal has contaminated numerous places. PCBs were used extensively and unrestrictedly in industry, which led to their ubiquity as pollutants around the globe. The distribution of congeners according to chlorine substitution varied from one site to another. The PCB homologue distribution showed that the relative composition of PCB congeners was clearly dominated by Tri-PCBs. Tri-PCBs > Tetra-PCBs was the order in which the PCB homologue distribution patterns were found in the soil. Moreover, the limited water solubility and poor migratory ability of highly chlorinated PCBs in these sediments may contribute to their low detection frequencies (Liu *et al.,* 2017). Contrarily, after being released into the environment, lighter PCB homologues like di-PCBs, tri-PCBs, and tetra-PCBs are vulnerable to microbial breakdown and volatilization (Zhang *et al.,* 2003; Eqani *et al.,* 2012; Baqar *et al.,* 2017).

Conclusion: Okunmo and Oaustech had low levels of PCBs. The highest levels of PCBs were recorded at Igodan. Excessive burning of charcoal and wood for domestic uses by Igodan residents must have caused it to have the highest PCBs among the five rivers. Also leaks from damaged transformers and electrical cables could be a major cause. It is possible that the source of PCBs emissions and migration pathways into Okitipupa surface water are as a result of water erosion from rain or wind movement. The PCB concentrations in the sediments are within the permissible limits. Tri-PCBs > tetra-PCBs were the order of the homologue distribution in the sediments from the surface water that were sampled.

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