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Occurrence of a Polychlorinated Biphenyl (PCB) Congener in Surface Water, Sediments and Blackchin Tilapia (*Sarotherodon melanotheron*) from Ologe Lagoon, Nigeria

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ABSTRACT: Polychlorinated biphenyls (PCBs) are persistent organochlorine chemicals that are toxic to aquatic organisms and humans. PCBs levels were assessed in surface water, sediments and fish from Ologe Lagoon, a major water body receiving treated effluents from Agbara Industrial Estate, using Gas Chromatograph-Electron Capture Detector (GC-ECD). 2,4,4'-Trichlorobiphenyl (2,4,4'-TCB) was the only PCB congener detected in the assessed matrices. Sediment 2,4,4'-TCB levels ranged from 0.0033±0.00333 to 0.0430 ± 0.00351ng/g. There was a significant ($p < 0.05$) seasonal variation in the levels in sediments and surface water. The level in surface water from Zone 3 was above USEPA's safe limit (0.0005 mg/L) in the rainy season. There was no significant ($p > 0.05$) seasonal difference in the mean concentrations of 2,4,4'-TCB in fish, and the levels were lower than WHO's safe limit (0.2 mg/kg). Based on the negligible to low levels of 2,4,4'-TCB in *S. melanotheron* inhabiting the lagoon, the fish was considered safe for consumption. Levels of 2,4,4'-TCB found in the sediments and surface water in this study indicate that levels may change with season, therefore, we recommend that the concentrations of the compound are regularly monitored in order to timely avert toxic levels of bioavailable 2,4,4'-TCB in the water body.

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Polychlorinated biphenyls (PCBs) are synthetic organochlorine chemicals that were used as coolants and lubricants in transformers, generators, and electrical capacitors because of their electrical insulating properties, low burning capacity and chemical inertness (Necibi and Mzoughi, 2017). They were also used in the production of plasticizers in rubber and polyvinyl chloride plastics (Erickson and Kaley, 2011). They were historically produced in the US and Europe (Gioia *et al.*, 2013). Efforts have been made in those regions to significantly reduce the emissions of PCBs from their sources, however, high concentrations of these chemicals have been found in non-producing regions such as Africa (Gioia *et al.*, 2013). Gioia *et al.*, (2011) attributed these high levels to the atmospheric deposition of PCBs. Importation of fairly used and old electrical equipment from developed countries has also contributed to the increasing levels of the chemicals in the regions (Gioia *et al.*, 2013). The high demand for electronic equipment and disposal of these products as e-wastes are regarded as important sources of PCBs in aquatic environments in Nigeria (Obaje, 2013). Aquatic ecosystems are usually the final destination of most wastes (Amiard-Triquet, 2015). Due to the persistent nature of PCBs, those that end up in water and sediments can be taken up or bioaccumulated by aquatic organisms (Okoh, 2015). Biomagnification of PCBs along the food chain ultimately increases the levels of PCBs in aquatic organisms such as fish at higher levels of the food chain (Mackay and Fraser, 2000). Dietary exposure of PCBs to humans has been noted as an important route of exposure (Moon and Ok, 2006). Therefore, the monitoring of PCBs in aquatic foods such as fish is of importance because of the several effects of PCBs that have been observed over the years. The biological effects include carcinogenicity, immune suppression, neurobehavioural effects, hypothyroidism, infertility and reproductive system disorders, cardiovascular disease and elevated serum lipids, hypertension, diabetes, liver disease, asthma, arthritis, and low birth weight (Breivik *et al.*, 2007). The Ogun - Lagos industrial axis has many sprawling industrial estates among which is the Agbara Industrial Estate. This estate hosts several large production plants which discharge their effluents, a possible source of PCBs, into the central sewage treatment plant which finally empties into Ologe Lagoon. The purpose of this study was to determine the levels of PCBs in environmental

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media (water and sediments) and fish of Ologe Lagoon.

MATERIALS AND METHODS

Study Area: Ologe Lagoon is a brackish coastal water body that covers about 6354.71 km^2 and 285 km in area and perimeter respectively, and it lies in close proximity to Agbara Industrial Estate (Oluwatosin *et al.,* 2008). Ologe Lagoon receives wastewater from the estate. The industrial estate has an integrated waste treatment plant that treats wastewater before discharging them into Ologe Lagoon via River Owo (Okogwu, 2006).

Sample Collection: Triplicate samples were collected from three zones of the lagoon (Fig. 1). Sampling was done in the rainy (May, 2017) and dry seasons (November, 2017). Water samples were collected using a 2.5 L amber glass bottle. Before analysis, samples were filtered through 0.45µm fibreglass filters to remove sand and debris. Sediments were also collected *in situ* using a fabricated 30 kg Van Veen Grab sampler. The sediments were subsequently retrieved, scooped into aluminium foil, wrapped and appropriately labelled. Fishes were collected using set nets. They were wrapped in aluminium foil, labelled and placed in coolers with ice packs until reaching the laboratory. Analyses began within 24 hours after collection. *S. melanotheron* was selected for this study based on its ecological and economic importance.

Analytical Procedure for Polychlorinated Biphenyls (PCBs) extraction and quantification: The extraction of the water samples was done according to the method of USEPA (2006a.) Briefly, 40 ml of dichloromethane (DCM) was used to extract 200 ml of the water sample twice. The extract was filtered through a funnel containing glass wool and 1 g of anhydrous Na_2SO_4 into an Erlenmeyer flask and then washed with 10 ml of DCM. The combined sample extracts were concentrated to about 5 ml using a rotary evaporator. The concentrated extract was dissolved in 40 ml of n-hexane and further concentrated to 1 ml. The extract was solvent exchanged to n-hexane. The extract was transferred into a Florisil column twice pre-washed with 20 ml of n-hexane. Elution was done with 50 ml of DCM/hexane (vol/vol ratio). The eluate was concentrated on a rotary evaporator until the volume reached 3 ml and then further reduced to 1 ml under a stream of nitrogen and solvent exchanged to n-hexane. Extraction of sediment and fish samples was done in accordance with the methods described by Adeogun *et al.*, (2016). Analyses were performed using a gas chromatograph (Agilent 7890A, Agilent Technologies, Palo Alto, CA, USA) with electron capture detection (ECD), and the results were confirmed by GC/MS (mass spectrometry) analysis. The GC-ECD column was an HP-5 fused silica column of 30 m length, 0.32 mm inner diameter and 0.25 µm film thickness.

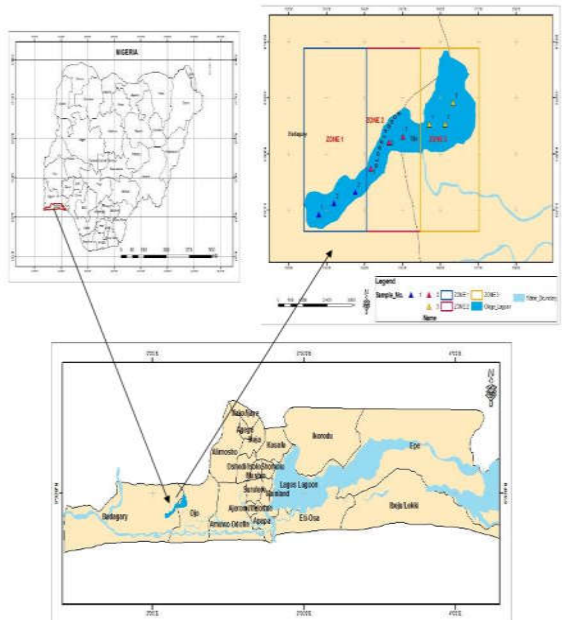

Fig 1: Map of Ologe Lagoon, Lagos, Nigeria (Adapted from Umulor *et al.,* 2018)

Nitrogen was used as the carrier gas. The injector temperature was set at 280 and 320°C for the detector, the oven temperature was set at 70°C for a minute and increased to 170 ºC at 40 ºC/min, increasing at 3 ºC/min to 195 ºC (holding this temperature for 3 min), increasing 0.5 ºC/min to 210 ºC (holding for 5 min) and increasing 20 ºC/min to 300 ºC with a final hold time of 10 min (Combi *et al.*, 2013). A procedural blank was included with each set of samples. The recoveries for the compounds ranged from 80 to 110%.

Biota-Sediment Accumulation Factor (BSAF): The biota-sediment accumulation factor (BSAF) was calculated as a ratio of the relative levels of PCBs in the fish and sediment samples. The BSAF is commonly used to support remedial decisions, and BSAF methodologies are clearly defined by USEPA (2006b).

$$\mathbf{BSAF} = \frac{\text{mean concentration of PCBs in fish}}{\text{mean concentration of PCBs in sediment}}$$

Bioaccumulation Factor (BAF)

The bioaccumulation Factor (BAF) was calculated as a ratio of PCBs in the fish and surface water (USEPA, 2003).

$$\text{BAF} = \frac{\text{mean concentration of PCBs in fish}}{\text{mean concentration of PCBs in water}}$$

Data Analysis: Data analyses were performed using SPSS version 20. Data were subjected to Two-way analysis of variance (ANOVA), and significant means ($p < 0.05$) were separated using Duncan Multiple Range test. Results were expressed as Mean ± Standard Error.

RESULTS AND DISCUSSION

Polychlorinated Biphenyls in the Environmental Media and Fish: Ologe Lagoon, a major 'receptacle' for industrial effluents from over 20 factories in Agbara Industrial Estate, was assessed to determine the levels of PCBs in sediments, water and fish from the water body. The results revealed that 2,4,4'-Trichlorobiphenyl (2,4,4'-TCB) also known as PCB 28 was the only detected PCB congener in the lagoon.

Sediments: The results from the sediment sample analysis indicated that the mean concentrations of 2,4,4'-TCB in sediments from the three zones ranged from 0.0290±0.00586 to 0.0430 ± 0.00351 ng/g during the rainy season and 0.0033±0.00333 to 0.0100±0.00577 ng/g during the dry season (Figure 2). There was no significant ($p > 0.05$) difference in the mean concentrations of 2,4,4'-TCB in sediments across the three zones. The mean concentration of 2,4,4'-TCB recorded in the rainy season was significantly ($p < 0.05$) higher than the concentrations recorded in the dry season. Fu and Wu (2006) assessed the seasonal variation of the distribution of PCBs in sediments and biota in a PCB-contaminated estuary and observed that the concentration of PCBs in the sediments increased in the rainy season. They attributed the elevation in the concentration to the surface runoff of PCBs previously buried in the surface soil into the sediments. In other parts of Nigeria, PCBs levels in sediments from Choba River (0.1701 $\mu g g^{-1}$) (Archibong *et al.*, 2017), Ona River (589 - 1353.6 µg/kg) and Ogun River (322.9-2002.6 µg/kg) (Adeogun *et al.*, 2016) were much higher than those in the present study. In comparison to studies conducted in other African countries and countries outside Africa, levels of the PCB compound in sediments in the present study were lower than most of the levels reported by Kampire *et al.*, (2017) (1.60 to 3.06 ng g-1; South Africa), Verhaert *et al.*, (2013) (<50–1400 pg g-1 dw; Congo), Barakat *et al.*, (2013) (1480–137,200 pg g^{-1}; Egypt), de Souza *et al.*, (2008) (18–184 pg g-1 dw; Brazil), Tombesi *et al.*, (2017) (0.61 to 17.6 ng g^{-1}; Argentina) Odabasi *et al.*, (2017) (2.7–2450 µg kg^{-1}dw; Turkey), Li *et al.*, (2007) (0.03 – 13.99 ng/g; China), and Kim *et al.*, (2018) (18.1–136.8 µg/kg; USA). These differences could lie on the specific industrial activities and sources of PCBs that release the chemicals into the various water bodies. Although the concentrations in sediments in the present study were relatively low, Obanya *et al.*, (2018) asserted that chronic exposure to low concentrations of toxicants plays a greater role in toxicology than acute exposure.

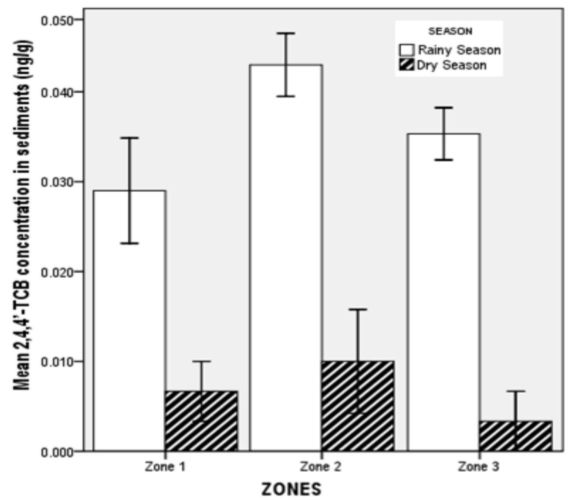

Fig 2: Concentrations of 2,4,4'-TCB in Sediments

Surface Water: The results obtained from the lagoon surface water analysis indicated that 0.00133± 0.000667mg/L of 2,4,4'-TCB was detected in surface water from Zone 3 during the rainy season (Figure 3). The PCB compound was not detected in surface water in the dry season (Figure 3). The variation between seasons was significant ($p < 0.05$). The 2,4,4'-TCB value in water from Zone 3 during the rainy season was higher than the USEPA PCBs maximum limit for water (0.0005 mg/L) (USEPA, 2009). Organic contaminants are spread in the environment through water and air, however, they are expected to move to the sediments because organic contaminants are hydrophobic (Beyer and Biziuk, 2009). Similar studies in Nigeria (Ethiope, Benin and Warri Rivers (Ezemonye, 2005a; 2005b); Choba River (Archibong *et al.*, 2017) had concentrations higher than the USEPA PCBs limit for water. Zone 3 is the closest zone to the point source of industrial effluents; this could explain the higher level of the detected PCB congener in relation to USEPA's safe limit. In the rainy season, the heavy rains tend to flush in more effluents from different sources into water bodies thus, the increase in the levels of various contaminants in water (Gao *et al.*, 2013). Several studies have shown that PCBs in aquatic environments are toxic to inhabiting organisms at significant levels. The swimming velocity of Japanese medaka (*Oryzias Latipes*) exposed to 1-25 µg/g of PCBs decreased in a dose-dependent manner (Nakayama *et al..,* 2005).

Lerner *et al.,* (2007) reported that juvenile Atlantic salmon (*Salmo salar*) exposed to 1 and 10 µg/L of PCBs during smolting exhibited a dose-dependent reduction in preference for seawater. They also found that 10 µg/L of PCBs exposed juvenile *S. salar* exhibited a 50% decrease in gill Na+,K+-ATPase activity and a 10% decrease in plasma chloride levels in freshwater. Plasma triiodothyronine was significantly reduced in *S. salar* exposed to PCBs at 1 and 10 µg/L (Lerner *et al.,* 2007). Gonzalez *et al.,* (2016) observed that larval zebrafish (*Danio rerio*) treated with 2-10 ppm exhibited enhanced thigmotaxis (edge preference).

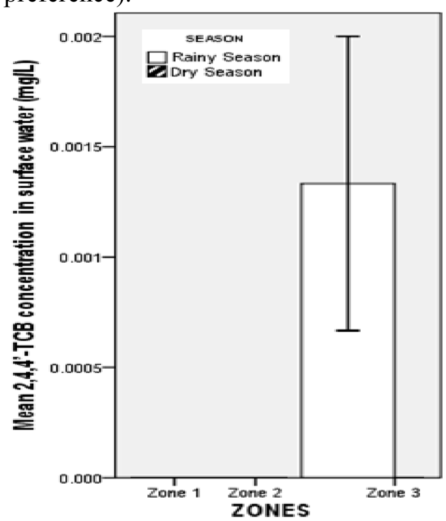

Fig 3: Concentrations of 2,4,4'-TCB in Surface Water

Bioaccumulation of 2,4,4'-TCB in the tissues of Blackchin Tilapia (S. melanotheron) inhabiting Ologe Lagoon**:** The mean concentrations of 2,4,4'-TCB in fish ranged from 0.0370±0.01739 to 0.0533±0.00667 ng/g during the rainy season and 0.0367±0.00882 to 0.0467±0.01764 ng/g during the dry season (Figure 4). There was no significant ($p > 0.05$) difference in the mean concentrations of 2,4,4'-TCB in fish across the three zones. Seasonal variation did not significantly ($p > 0.05$) affect the concentrations of 2,4,4'-TCB in fish. The mean concentrations of 2,4,4'-TCB in fish were lower than the WHO's safe limit (0.2 mg/kg) (WHO and FAO, 2011). The levels of 2,4,4'-TCB in the fish were low when compared to the levels reported in other parts of Nigeria and other countries (Table 1). The biota to sediment accumulation factor (BSAF) values of 2,4,4'-TCB in fish from the three zones ranged from 1.09 to 1.51 during the rainy season whereas during the dry season the values ranged from 0.48 to 14.15 (Table 2). The bioaccumulation factor (BAF) value of fish samples from zone 3 during the rainy season was 4.0075×10^{-5} (Table 2).

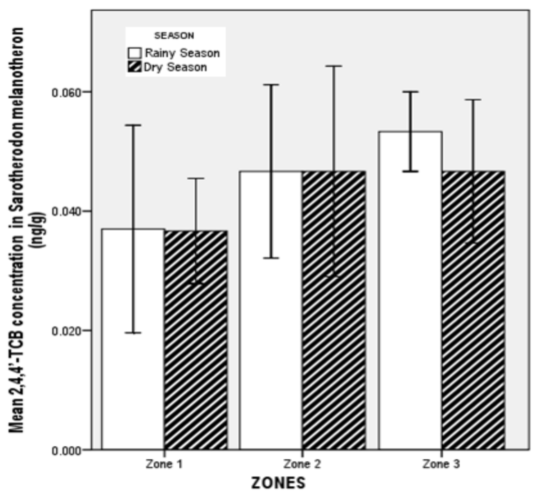

Fig 4: Concentrations of 2,4,4'-TCB in *S. melanotheron*

Table 1: PCBs levels in fish from other water bodies

Country	Levels	References
Ologe Lagoon, Nigeria	0.0367 - 0.0533 ng/g	Present study
Lagos Lagoon, Nigeria	0.4228 to 2.94 ppm	(Adeyemi *et al.,* 2009; Igbo, 2012)
Choba River, Nigeria	0.2683 µg/g	(Archibong *et al.*, 2017)
Ogun River, Nigeria	248.1 - 2433.6 µg/kg	(Adeogun *et al.*, 2016)
Ona River, Nigeria	421.7 - 2191.7 µg/kg	(Adeogun *et al.*, 2016)
Lake Tanganyika in Burundi	24,300–77,700 pg g^{-1} lw	(Manirakiza *et al.*, 2002)
Lake Burullus in Egypt	3320–72,060 pg g^{-1}	(Said *et al.*, 2008)
Lake Victoria, Uganda	41 to 670 pg g-1 lw	(Ssebugere *et al.*, 2014)
Pearl River Delta in China	5150 to 226,000 pg g^{-1} lw	(Nie *et al.*, 2006)
Hong Kong	0.028-6.3 ng·g^{-1}	(Su *et al.*, 2018)
Manoa Stream in Hawai'i (USA)	51,900–89,420 pg g^{-1} lw	(Yang *et al.*, 2008)

Table 2: Relative Bioaccumulation of 2,4,4'-TCB in the tissues of Black Chin Tilapia (*S. melanotheron*) inhabiting Ologe Lagoon in the rainy and dry seasons

	Sampling Stations					
	Rainy Season			Dry Season		
	1	2	3	1	2	3
BSAF	1.28	1.09	1.51	5.48	0.48	14.15
BAF	-	-	4.0075×10^{-5}	-	-	-

BSAF- Biota to sediment accumulation factor (Fish : sediment); BAF- Bioaccumulation factor (Fish : surface water)

Results from the bioaccumulation studies suggest that 2,4,4'-TCB was more bioavailable in the sediments in the lagoon than water. Studies have shown that sediments act as a sink for organic contaminants (Kumar and Singh, 2013; Pozo *et al.*, 2014). These chemicals, due to their hydrophobicity and lipophilic characteristic adhere to sediments, thereby, making them bioavailable for organisms (Alkhatib and Weigand, 2002). The spatial difference in the biota-sediment accumulation of 2,4,4'-TCB in the fish could be attributed to the varying distances of the zones from the source of industrial effluents and the behaviour of the fish as noted by Mourier *et al.*, (2014).

Conclusion: Data from this study suggest that *S. melanotheron* could be safe for consumption with respect to PCBs contamination as the concentrations recorded in all the fish samples were consistently below WHO's safe limit. Conversely, the concentration of the compound in water from one of the zones in the rainy season was above USEPA's safe limit. This indicates that despite the negligible to low levels of the compound found in the environmental media of Ologe Lagoon, regular monitoring of the compound in the water body should not be neglected. This ensures that potentially toxic levels of the compound in Ologe Lagoon are forestalled.

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