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ASSESSMENT AND RISK OF POLYCHLORINATED BIPHENYLS IN *Chrysichthys nigrodigitatus, Cynoglossus senegaliensis* AND *Pseudolithus elongatus* TO CONSUMERS: A STUDY OF LAGOS LAGOON

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Abstract: The study investigated the mean concentrations of polychlorinated biphenyls (PCBs) in three (3) marine fishes specie; *Chrysichthys nigrodigitatus, Cynoglossus senegaliensis* and *Pseudolithus elongatus* (total of 112 fishes) collected from the Lagos Lagoon as well as the calculated risks probably associated with the consumption of the PCBs in the selected fishes. Analysis of PCBs congeners was detected using a gas chromatograph (Model 5890, PerkinElmer, MA, USA) equipped with nickel-63 electron capture detector (GC-μECD). The mean ∑PCBs in the fishes varied from 8.97 ± 5.62 to 16.28 ± 5.05 mg/kg ww. The non-carcinogenic hazard quotient (HQ) of dioxin-like congener exceeded the threshold for children and adult while the cancer risk values ranged from 1.71×10^{-4} to 3.11×10^{-4} which were below USEPA acceptable risk level (10^{-4}). The risk assessment showed that the exposed population is at risk of non-cancer adverse health effects and lifetime cancer. Therefore, measures should be initiated to prevent subsequent exposure of humans to PCBs through consumption of these selected fish species and continuous monitoring of these edible fish species.
Keywords: Gas chromatograph, organic chlorine, marine fishes, chlorination, *polychlorinated biphenyls*

Introduction

Polychlorinated biphenyls (PCBs) as defined by Ahmed *et al*. (2016) are a class of artificial organic compounds that are produced by the direct chlorination of the biphenyl ring system. The authors asserted that PCBs are among the most widespread and resistant chemicals in the world. Polychlorinated biphenyls were used as coolants and lubricants in transformers, capacitors, and other electrical equipment such as generators because of their general chemical inertness, insulating capacity, heat stability, and low burning capacity (Wong *et al*., 2007). Polychlorinated biphenyls are used as components (Plasticizers) utilized in the production of adhesives, casting agents, cements, de-dusting agents, paints, stabilizing additives in flexible polyvinyl chloride coatings of electrical cables and electronic components, pesticide extenders, reactive flame retardants, sealants for caulking, waterproofing compounds, wood floor finishes, and waterproofing compounds (Thomson and Rose, 2011). PCBs in the gaseous state partition among air, water, soil and sediments as they are transported through several pathways including deposition and evaporation (Gdaniec-Pietryka *et al*., 2007). Organic compounds within the water column can accumulate in living organisms either by uptake from the environment over time (bioaccumulation) or along the food chain (biomagnification) since these compounds are not very soluble in water but are readily soluble in fatty tissues (Mackay and Fraser, 2000). Thus, the concentrations of PCB is greater in organisms (birds, predatory fishes and mammals) over time, that are higher in the food chain. The degree of solubility is mainly dependent on the number and position of chlorine atoms on the PCB molecule (Beyer and Biziuk, 2009).

The Lagos Lagoon is a major recipient of wastewater from industries and municipal areas; run-offs from dumpsites and agricultural lands. The wastewater and run-offs contain organic chemicals that could include PCBs. These chemicals have the potential to accumulate in the tissues of aquatic organisms such as fish.

Fish is a major source of dietary protein for children and adults in Nigeria (Food and Agriculture Organization, 2011). Despite the ban on the production of PCBs in the Stockholm Convention on Persistent Organic Pollutants in the year 1979, varying concentrations of PCBs in fish have been reported in Nigeria (Adeyemi *et al*., 2009; Igbo, 2012; Adeogun *et al*., 2016). As regards these reported concentrations, the need to assess the health risks associated with the consumption of PCBs contaminated fishes inhabiting the Lagos Lagoon is, therefore imperative. To the best of our knowledge, the non-cancer and cancer health risks associated with the intake of PCBs through consumption of some fishes from the Lagos Lagoon are not yet known.

The objectives of this study were to investigate the levels of polychlorinated biphenyls (PCBs) in three marine fish species from the Lagos Lagoon and the calculated associated risks due to the consumption of these species.

Materials and Methods

Sample collection

One hundred and twelve (112) locally consumed fishes such as Silver catfish (*Chrysichthys nigrodigitatus* Lacepede, 1803, Sole-fish (*Cynoglossus senegaliensis* Kaup, 1858) and Croaker (*Pseudolithus elongatus* Bowdich, 1825) were selected randomly from the designated study area (Table 1). These fishes were collected from the Lagos Lagoon by using set nets over a three (3) month period (August to October in 2016). This is because of their abundance in the lagoon and mostly consumed by man. They were wrapped in aluminum foils, labeled accordingly, and kept in coolers that had temperatures of ≤ - 20 before they were taken to the laboratory for analysis.

Table 1: Study Stations and their Coordinates

S/N	Stations	Location	Coordinates
1	Station 1	Iddo	N 06º 28' 18.1" E 003º 23' .053'
2	Station 2	Commodore	N 06º 25' 22.6" E 003º 24 .392'
3	Station 3	Tarkwa Bay	N 06º 24' 31.2" E 003º 23 .372'

Estimation of condition (k) factor of experimental fish

The condition factor (k) of the fish samples was calculated with the formula below

$$k = \frac{100 \times W}{L^3}$$

W - wet weight of fish L - total length (Fulton, 1902).

Extraction of fish samples for analysis of PCBs

The fish species were dissected and the muscle tissues harvested. Anhydrous sodium sulphate (Na_2SO_4) was used to grind five grams (5 g) of the muscle tissue for homogenization. In a cold extraction mode, extraction was done with dichloromethane, which was later evaporated on a rotary evaporator (Anyakora *et al*., 2005).

Analysis of fish samples for PCBs concentration

Identification and quantification of PCB congeners

The gas chromatograph (GC) conditions used were those outlined by USEPA (1996). A gas chromatograph (Model 5890, PerkinElmer, MA, USA) equipped with nickel-63 electron capture detector (GC-μECD) was used for the identification and quantification of PCB congeners. A low polar HP–5 column (30 m × 0.32 mm i.d, 0.25 mm film thickness was used. Nitrogen was used as a carrier gas at a flow rate of 40 ml/s. data were processed with an HP 3396 integrator (Hewlett-Packard, California, USA). The operating guidelines were as follows: injector temperature was set at 250 and 300°C for the identifier, the oven temperature was programmed at 150°C at first (with 5 min hold) and increased to 300°C at a warming rate of 4°C/min to give the analysis period of 34 min. (Anyakora *et al*., 2005).

Quality control

Spiked blank, reagent blank and appropriate standard reference materials were included with each set of sample to confirm the quality of the analytical method and corresponding analytical results. There were no target compounds detected in the routine blank and solvent blank. The recoveries for the PCB congeners, fluctuating between 80 and 110%, were determined by adding known amounts of PCB standards to samples before extraction.

Estimated daily intake (EDI) and lifetime daily average daily dose (LADD)

Estimated Daily Intake (EDI) was calculated by the formula stated by USEPA (2000).

EDI (mg/kg/d) = Mean dioxin-like PCB concentration × food consumption/body weight.

An individual consumes 9 kg of fish in Nigeria, and FAO has set the body weight of children (1-11 years) at 30 kg and adults at 70 kg. The EDI was compared to USEPA reference dose (RfD) (2×10^{-5} mg/kg/d), which represents the maximum exposure limit below which it is improbable for sensitive populations to experience adverse health effects (USEPA, 2000).

LADD was calculated by using the formula stated in 'Risk Assessment Guidance for Superfund' by USEPA (1989).

Hazard quotient (HQ)

Human health risk with regard to non-carcinogenic hazard quotient (HQ) as a result of consumption of dioxin-like (DL) PCB contaminated fish muscle was estimated for the three fish species using tissue-screening levels.

The non-carcinogenic HQ was estimated using the following equation:

$$HQ = \frac{EDI}{RfD} \quad \text{(USEPA, 2000)}$$

The non-carcinogenic screening levels are based on a target HQ of 1, and HQ higher than 1 is considered a health risk (USEPA, 2000).

Cancer risk

Cancer risk is used to estimate the possibility of developing cancer in the whole life of an individual due to dietary exposure to PCBs in the fish (USEPA, 2005). Cancer slope factor (CSF) as presented by USEPA (2012) is 2 mg/kg/d. The cancer risk of PCBs in the fish is calculated by dividing the LADD of PCBs in each fish species by the CSF (USEPA, 2005).

Statistical analysis

Data analysis was done using Statistical Package for Social Sciences (SPSS) version 20.0. Data collected for the PCB concentrations were subjected to one-way Analysis of Variance (ANOVA) and Duncan's Multiple Range Test (DMR) to determine mean-variance among samples.

Results and Discussion

Estimation of condition (k) factor of experimental fish

The weight and k factors of the fish species significantly differed from one another. Among these species, only *C. senegaliensis* was significantly ($P < 0.05$) different in total length. The k values for all the fish species examined were less than 2.0 (Table 2). The energy reserves in fish are indicated by Fulton's condition factor (k) and the highest value of 1.31 ± 0.48 in *P. elongatus* indicated that these species were in a better condition than *C. senegaliensis* and *C. nigrodigitatus*. *Cynoglossus senegaliensis* and *C. nigrodigitatus* were in poor conditions having values less than 1. It has been reported that fishes with low k values utilize more energy for survival than growth due to the stress in their environment (Adeogun *et al*., 2013).

Table 2: Mean Weight, total lengths and condition factors of the fish

Fish species	Total length (cm)	Weight (g)	K-factor
C. nigrodigitatus	22.47 ± 1.51^a	51.91 ± 0.97^a	0.48 ± 0.10^a
C. senegaliensis	40.37 ± 1.68^b	144.73 ± 2.17^b	0.16 ± 0.03^b
P. elongatus	18.47 ± 0.26^a	81.97 ± 1.20^c	1.31 ± 0.07^c

Different letters (a, b & c) showed significant difference

Total PCBs levels in fish

There was no significant ($P > 0.05$) difference among the mean concentrations of ∑PCBs in the three (3) species. The concentration trends of ∑PCBs in the fishes were 16.28 ± 5.05 mg/kg wet weight (*C. nigrodigitatus*) > 12.50 ± 3.48 mg/kg wet weight (*C. senegaliensis*) > 8.97 ± 5.62 mg/kg wet weight (*P. elongates*) (Fig. 1). The mean concentrations of ∑PCBs in the fish species exceeded the WHO maximum limit (0.2 mg/kg) (WHO and FAO, 2011). Previous studies have documented that diet serves as a major route of uptake of PCBs in fish (Doi *et al*., 2000; Carlson and Hites, 2005; Nichols *et al*., 2001). This was illustrated by feeding trout to environmentally relevant concentrations of PCBs in its diet. The authors found that over 90% of the concentration in the diet was taken up by the organism. *C. nigrodigitatus* is an omnivorous fish that feeds on seeds, insects, bivalves, and detritus (Abowei and Ezekiel, 2013). *Pseudolithus elongatus* feeds on fish and shrimps (Oribhabor and Ogbeibu, 2012) while *C. senegaliensis* feeds on molluscs, shrimps, crabs, and fish (Daniel, 2015). Shrimps (Jaward *et al*., 2012), molluscs (Orbea *et al*., 2002), insects (Paine *et al*., 1993), and crabs (Magalhães *et al*., 2012) have all been reported to accumulate PCBs. This means that organism have the ability to accumulate PCBs in various concentrations. The diet of *C. nigrodigitatus* which comprised several aquatic organisms predisposes it to various sources of PCBs. This was evident in this study in that the level of PCBs in *C. nigrodigitatus* was highest when compared to the other species. The concentration of the total PCBs in *C. nigrodigitatus* was higher than WHO maximum limit of 0.2 mg/kg. This corroborates the study of Adeyemi *et al*. (2009) that recorded a value higher than 0.2 mg/kg in *C. nigrodigitatus. P. elongatus* and *C. senegaliensis* also had concentrations higher than WHO maximum limit (0.2 mg/kg). High concentrations of PCBs in fish recorded globally include 10.14 mg/kg ww in fish from Georges/Cooks Rivers and Sidney Harbor, Australia (Roach and Runcie, 1998); 15 mg/kg ww in fish from an estuary in Argentina (Colombo *et al*., 2007); 5 mg/kg ww in fish from lakes in Canada (Bhavsar *et al*., 2007); 108.752 mg/kg ww in fish from Slovakia (Šalgovičová and Zmetáková, 2006). This indicated that the concentrations recorded in this study were at the higher end of the global range.

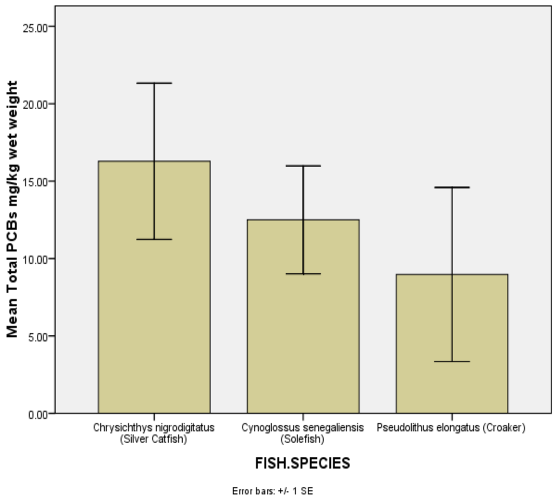

Fig. 1: Mean concentrations of ∑PCBs in fish

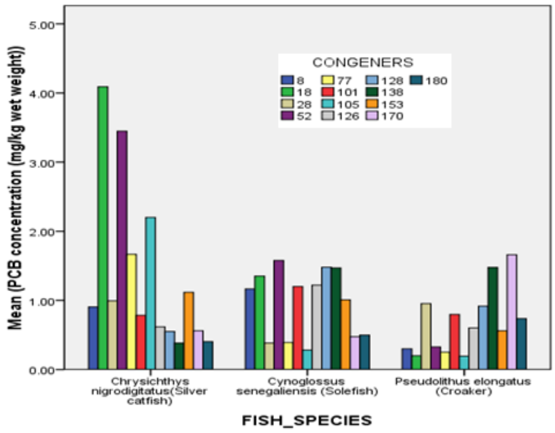

Fig. 2: Distribution of PCB congeners in fish

PCBs congeners in fish

The predominant congeners in *C. nigrodigitatus, C. senegaliensis, and P. elongatus* were 18, 52, and 170, respectively (Fig. 2). *Pseudolithus. elongatus* had the highest mean concentrations of PCB congeners 138, 170 and 180 (1.48 ± 1.12, 1.66 ± 1.66 and 0.74 ± 0.67 mg/kg wet weight, respectively. *Cynoglossus senegaliensis* had the highest mean concentrations of PCB congeners 8, 101, 126 and 128 (1.17 ± 1.17, 1.20 ± 0.33, 1.22 ± 0.13 and 1.48 ± 0.59 mg/kg wet weight, respectively) (Fig. 3). *Chrysichthys nigrodigitatus* presented the highest mean concentrations of PCB congeners 18, 28, 52, 77, 105 and 153 (4.09 ± 2.90, 0.99 ± 0.99, 3.44 ± 3.13, 1.67 ± 0.95, 2.20 ± 1.24 and 1.12 ± 1.12 mg/kg wet weight, respectively) (Fig. 3). There was no significant ($P > 0.05$) difference among the mean concentrations of the PCB congeners in the three fish species.

The sum of PCBs 28, 52, 101, 138, 153 and 180 was used to evaluate the human exposure to non-dioxin like PCB (NDL-PCB) (EC, 2011). The sums of the six (6) congeners in all the fish species in this study were above the European Commission maximum level (0.075 mg/kg wet weight). This revealed that these fish species were not suitable for human consumption.

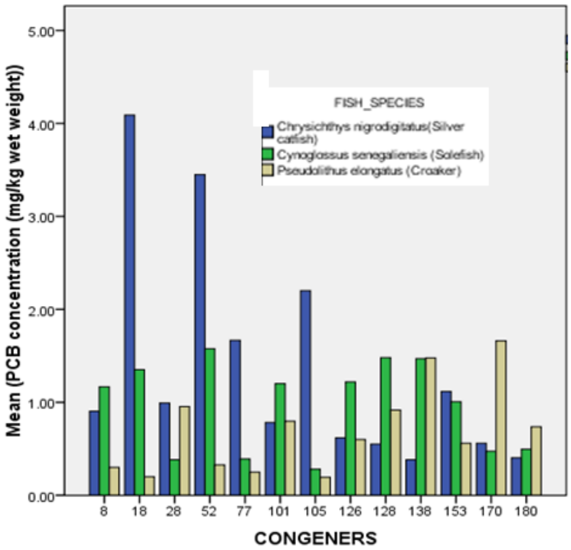

Fig. 3: Comparison of congeners among species

Table 3: Estimated daily intake of PCBs and lifetime daily dose through consumption of fish

Fish species	EDI (mg/kg/d), children 1–11 years	EDI (mg/kg/d), Adults
C. nigrodigitatus	1.37×10^{-3}	5.88×10^{-4}
C. senegaliensis	9.62×10^{-4}	4.12×10^{-4}
P. elongatus	6.16×10^{-4}	2.64×10^{-4}

EDI: Estimated Daily Intake based on calculation

Daily dietary intake

The average daily intake of DL PCB by a child through per-capita consumption of 9 kg of *C. nigrodigitatus, C. senegaliensis and P. elongatus* flesh per year were estimated to be 1.37×10^{-3}, 9.62×10^{-4}, and 6.16×10^{-4} mg/kg/d, respectively (Table 3).

The average daily intake of DL PCB by an adult through per-capita consumption of 9 kg of *C. nigrodigitatus, C. senegaliensis and P. elongatus* flesh per year were estimated to be respectively 5.88×10^{-4}, 4.12×10^{-4}, and 2.64×10^{-4} mg/kg/d (Table 3). The EDI values of DL-PCB in all the fish species were generally higher in children than in adults. The values were above USEPA reference dose (2×10^{-5} mg/kg/d) for children and adults. The dietary daily intake values of DL-PCB through the investigated fish species showed that *C. nigrodigitatus* exposed humans to a higher concentration of DL-PCB in a day than the other species. The daily intake values of the compound were consistently higher in children. This is similar to the study by Adeogun *et al*. (2016) who investigated the EDI of DL PCBs through *Clarias gariepinus* and found that children consumed higher concentration of the compound in a day than adults. USEPA (2000) has stipulated that the oral reference dose for the daily intake of PCBs through fish is 2×10^{-5} mg/kg/d and the value was used to calculate non-carcinogenic risk of consuming PCB contaminated fish. In that regard, the high values of HQ in this study showed that humans that consume these fish species from the study area could be predisposed to non-carcinogenic risks as presented by USEPA (2000).

Furthermore, it was observed that *C. nigrodigitatus* had higher non-cancer and cancer risk values than the other species and it has been established that fish with high lipid content accumulates high concentrations of chemicals and consequently has a high health risk to individuals that consume it. Thus further studies on the correlation between lipid content, and non-cancer and cancer health risks associated with PCBs contamination in fish should be carried out.

Hazard quotient (HQ)

The non-carcinogenic hazard quotient (HQ) for humans from intake of contaminated fish for PCB 77 for children aged 1–11 years and were 68.5, 48.1 and 30.8 for *C. nigrodigitatus, C. senegaliensis and P. elongatus* respectively while values for adults were 29.4, 20.6, 13.2 for *C. nigrodigitatus, C. senegaliensis and P. elongates,* respectively (Table 4). The HQ values of DL-PCB in all the fish species were generally higher in children than in adults. HQ values for all fish species were greater than 1 for both children and adults. A high likelihood of non-carcinogenic health risks for communities fishing from these water bodies (USEPA, 2000b). The elevated HQ obtained in the fish species showed that anthropogenic activities appreciably increased the potential of non-carcinogenic hazards to humans who consume fish from these water bodies (Morra *et al.,* 2006).

Table 4: Non-carcinogenic hazard quotient (HQ) for humans from exposure to polychlorinated biphenyl through ingestion of the three fish species

Fish species	Hazard quotient (HQ), children 1–11 years	Hazard quotient (HQ), adult
C. nigrodigitatus	68.5	29.4
C. senegaliensis	48.1	20.6
P. elongatus	30.8	13.2

Table 5: Calculated cancer risks for humans from exposure to polychlorinated biphenyls through fish consumption

Fish species	LADD (mg/kg/d) ($\times10^{-4}$), adults	Cancer risk ($\times10^{-4}$)
C. nigrodigitatus	6.21	3.11
Cynoglossus senegaliensis	4.77	2.39
Pseudolithus elongatus	3.42	1.71

LADD = Lifetime Daily Average Daily Dose; *C. nigrodigitatus* = *Chrysichthys nigrodigitatus;* ***C. senegaliensis*** = *Cynoglossus senegaliensis;* ***P. elongatus*** = *Pseudolithus elongatus*

Cancer risk

The LADD values of PCBs in the fish species ranged from 3.42×10^{-4} to 6.21×10^{-4} mg/kg/d (Table 5). The cancer risks of consuming fish from the study area are presented in Table 5. The values ranged from 1.71×10^{-4} to 3.11×10^{-4}. All the values were above USEPA acceptable risk level (10^{-4}) (USEPA 2005). The calculated LADD revealed that humans were exposed to higher concentration of PCBs if they consume *C. nigrodigitatus* in their lifetime than the other two species. However, the concentrations of PCBs in the fish species do not pose significant cancer risks to humans because the values were below USEPA acceptable risk level (10^{-4}). In a recent study in Choba River, Rivers State, Archibon *et al.* (2017) obtained values that ranged from 2.8 x 10^{-7} to 4.6 x 10^{-5} in *Chrysichthys nigrodigitatus* (catfish) and *Liza falcipinnis* (mullet) which were lower than the values obtained in this study though, the values were generally below USEPA acceptable limit.

Conclusion

It has been established that the selected fish species collected from the Lagos Lagoon are contaminated with PCBs which bioaccumulated and biomagnified. This finding recommends further studies that will incorporate organisms lower in the food chain to determine the possible biomagnification of PCBs along the food chain in Lagos Lagoon. The risk studies revealed that the exposed population is at risk of non-cancer adverse health effects and lifetime cancer. Due to these risks, there is a need for an urgent action by relevant environmental and public health agencies to implement strategies to prevent subsequent exposure of humans to PCBs through consumption of these fish species, and eliminate sources of PCBs pollution in the water body such as the Lagos lagoon which in turn leads to the sustainable management of the pollution.

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Conflict of Interest

Authors declare that there are no conflicts of interest

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