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A Pilot Investigation on the Residual Polychlorinated Biphenyls (PCBs) in Imported Frozen Atlantic Mackerel at Selected Markets in Benin Metropolis, Nigeria, West Africa

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Abstract

Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) that have gained worldwide recognition due to their deleterious effects on both wildlife and man. The paucity of ecotoxicological data regarding the PCB content of fish from markets in Benin City, Nigeria, West Africa, warranted this research. Accordingly, the residual levels of PCBs in samples of frozen Atlantic Mackerel (*Scomber scombrus*) of mean total length 24.68 ± 1.04 cm and mean weight 197.06 ± 0.84 g, purchased from selected markets were determined by gas chromatographic (GC) technique in order to ascertain their suitability for human consumption. The summary statistics for PCBs in *S. scombrus* ranged from 0.022 mg/kg (PCB 156) to 1.903 mg/kg (PCB 126) while the summary statistics for PCBs in *S. scombrus* by market ranged from 0.008 mg/kg (PCB 156) at Ekiedo market to 2.03 mg/kg (PCB 126) also at Ekiedo market with no significant differences (P>0.05) in the mean concentrations of PCBs in fish between markets. The estimated daily intake (EDI) values for PCBs ranged from 0.00088 for PCB 156 to 0.076 for PCB 126 while the toxic quotient (TQ) values ranged from 0.011 for PCB 156 to 0.95 for PCB 126. The total PCB content in *S. scombrus* by market ranged from 2.358 mg/kg at Ekioba market to 3.202 mg/kg at Edaiken market while a toxic equivalency (TEQ) value of 0.191 was recorded with PCB 126 presenting the highest sub-TEQ value of 0.1903. The fishes are considered safe for human consumption as the quantified PCB content fell below International threshold levels. It was advocated that *S. scombrus* be rigorously spot-checked by relevant enforcement agencies at the point of entry in order to ensure that episodic hazards associated with the consumption of PCBs via fish are kept to the barest minimum.

Keywords: Polychlorinated biphenyls, Toxic quotient, Toxic equivalency

1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) made up of the biphenyl molecule that has been chlorinated several times over. These compounds or congeners have been linked to cancers, congenital defects, liver damage, cardiovascular diseases, asthma, impaired immune systems, alteration of sex hormones, neurobehavioural and developmental deficits and cognitive problems in man as well as the inducement of multiple impairments in living beings (Inomata, 2009; Center for Health Environment and Justice, 2019). Some popular trade names for different mixtures of PCBs include Aroclor, Pyranol, Pyroclor, Phenochlor, Pyralene, Clophen, Elaol, Kanechlor, Santotherm, Fenchlor, Apirolio and Sovol (World Health Organization, 2008). It has been reported by the New York State Department of Environmental Conservation (2004), that people who live near hazardous waste sites may be exposed to PCBs by consuming contaminated fish, inhaling PCB-laden air or by drinking contaminated water. Furthermore, the United States Environmental Protection Agency (2004), has observed that contamination of the environment by PCBs can occur via maintenance operations, decontamination operations, transport operations, draining and refilling operations, contamination of waste oil, drainage systems, stormwater systems, discharge points, sumps, and areas adjacent to surface waters, railroad transformers, lack of spill containment provisions in work pits, wear of equipment in service (e.g., valves, gaskets and fittings), cracked or damaged transformer bushings, improper storage of PCB-containing and/or PCB-contaminated equipment, improper disposal of defective PCB-containing or PCB-contaminated equipment.

Although the large-scale production of PCBs ceased decades ago, their recalcitrant nature contributes to their persistence in the environment. They are known to have significant environmental stability owing to their thermo-resistance and can only be destroyed entirely at temperatures reaching 1000° C. Such pollutants have been reported to constitute environmental refectories because they are not usually eliminated from ecosystems via natural degradation (Oyakhilome *et al.*, 2019). Persistent organic pollutants have generally been reported to threaten the biodiversity and life of aquatic species (Tufail *et al.*, 2019). The marine environment is the ultimate endpoint and repository for PCBs. For example, approximately over 100,000 tonnes of PCBs have been deposited in the North Atlantic alone, which is why this ocean has become more contaminated by PCBs than any other ecosystem, and therefore marine species cherished as food by man need to be screened for PCBs (Dookie, 2019; Farsani *et al.*, 2019). Furthermore, fish consumption by man has been reported to be the major route of exposure to PCBs (the United States Public Health Service/Agency for Toxic Substances and Disease Registry/United States

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Department of Health and Human Services/United States Environmental Protection Agency, 2000). Fish can become contaminated with PCBs as a result of migration from packaging materials into fish and direct contamination by industrial accidents (World Health Organization, 2008). Benin City, the administrative headquarters of Edo State, Nigeria, is awash with fish markets that sell the Atlantic Mackerel *Scomber scrombrus* (Linnaeus, 1758) in its frozen form. The fish comes into the country via importation from European countries such as Norway (Wangboje *et al.*, 2017). This marine species is of commercial importance; hence its choice for this research. Available ecotoxicological data on chemical contaminants in fish from markets in Benin City have shown that there is a dearth of information on the PCB content in fish from such markets. It is on this premise that this research becomes relevant in order to fill an existing gap in knowledge and to further guide potential consumers of the product.

2. Materials and Methods

2.1 The study area

The study was conducted in Benin City, Edo State, Nigeria, gridlocked within Latitude 6º 20' 00'' N and Longitude 5º 37' 20'' E (Fig. 1). Extensive details of the study area have been published by Wangboje *et al*. (2017). Four markets were purposely chosen for the study based on their strategic locations and the all-year availability of *S. scrombrus*. They include Edaiken market which is located along the busy Uselu-Ugbowo road of the city, Ekiedo market which is situated at the intersection of New Lagos and Mission roads of the city, Ekioba market which is situated at the Ring road axis, and Ekiuwa market which is located off Television (TV) road.

2.2 Collection of fish samples

Fishes were purchased from the aforementioned markets for the research between July and December 2019. They were placed in septic polythene bags, sealed with selotape® and transported to the laboratory in a Thermolineo® icebox within 24 hours.

Fig. 1. Map of Benin City, Edo State, Nigeria

2.3 Laboratory procedures

The identities of the fish species were confirmed using the electronic version of the Food and Agriculture Organization of the United Nations (FAO) species identification sheet (FAO, 2018). Total length (cm) measurements were taken using a measuring board while weight (g) of fish samples were measured using an electronic scale (Mettler® PM4800 Delta Range). The mean total length was 24.68 ± 1.04 cm, while the mean weight was 197.06 ± 0.84 g (n=24). All reagents and chemicals used were of analytical grade (BDH, Poole, England and Sigma, USA). All glassware was soaked in detergent and then rinsed alternately with running tap water and distilled water. Ten grammes of muscle tissue was excised from the flanks with a stainless-steel lancet and ground with anhydrous sodium sulphate until a completely dry homogenate was obtained. The extraction of PCB in fish tissue was performed according to standard procedures (USEPA, 1996). The extract was concentrated to 2 ml with a rotary evaporator (Rotovap) at 40°C. The concentrated extract was thereafter used for clean-up and for gravimetric lipid determination. Clean-up of extracts was done in line with the method by Kampire *et al.*, (2015) while a Perkin model 5890 gas chromatograph equipped with Ni 63 electron capture detector was used for quantification of PCBs. The quality control was performed by regular analyses of procedural blanks and blind duplicate samples along with a random injection of standards and solvent blanks.

2.4 Estimation of Daily Intake (EDI) of PCBs by Man

The EDI was calculated based on the method by Anyakora *et al.* (2008) and Wangboje and Okpobo (2019).

EDI= 40g/person/day * (CPCB) mg/kg/1000g/kg = X mg/person/day.

Where:

40g/person/day = Estimated average fish consumption in Niger Delta, Nigeria.

(CPCB) = PCB concentration in fish.

Toxicity/Hazard Quotient (TQ) for PCBs

The Toxicity/Hazard Quotient (TQ) for chemical contaminants is a comparison of the measured concentration of site-related chemical contaminants in ecological matrices with specific health-based criteria (Newstead *et al*., 2002).

$$TQ = \frac{\text{Concentration of PCB in fish sample}}{\text{Health based criteria}}$$

2.5 Calculation of Toxic Equivalency (TEQ) for PCBs

The toxic equivalency (TEQ) for PCBs is expressed as follows:-

$TEQ = \Sigma\, Ti * TEF$

Where: TEQ = Toxic equivalency

Ti = PCB concentration in organism

TEF = Toxic equivalency factor.

PCB 114 and PCB 156 have TEF values of 0.0005 while PCB 126 and PCB 169 have TEF values of 0.1 and 0.01, respectively (New York State Department of Environmental Conservation, 2004; Commission Regulation, 2008).

2.6 Statistical Methods

GENSTAT® computer software (Version 12.1 for Windows) was used for statistical analysis. Data were subjected to Analysis of variance (ANOVA) to determine significant differences between mean values of PCBs while significance means ($P<0.05$) were separated with Duncan multiple range tests (DMRT). Microsoft® Excel (for Windows 2010) is used for all graphical presentations.

3. Results and Discussion

As presented in Table 1, the summary statistics for PCBs in *S. scombrus* ranged from 0.022 mg/kg (PCB 156) to 1.903 mg/kg (PCB 126) while the summary statistics for PCBs in *S. scombrus* by market ranged from 0.008 mg/kg (PCB 156) at Ekiedo market to 2.03 mg/kg (PCB 126) also at Ekiedo market with no significant difference ($P>0.05$) in the mean concentrations of PCBs in fish between markets (Table 2). As shown in Table 3, the summary statistics for PCBs in *S. scombrus* by month ranged from 0.0025 mg/kg (PCB 156) in September to 2.07 mg/kg (PCB 126) also in September with significant differences ($P<0.05$) in the mean concentrations of PCB 114 and PCB 138 in *S. scombrus* between months. The estimated daily intake (EDI) values in mg/person/day for PCBs, ranged from 0.00088 for PCB 156 to 0.076 for PCB 126 as shown in Fig. 2 while the toxic quotient (TQ) values ranged from 0.011 for PCB 156 to 0.95 for PCB 126 as presented in Fig. 3. Fig. 4 shows the total PCB content in *S. scombrus* by market which ranged from 2.358 mg/kg at Ekioba market to 3.202 mg/kg at Edaiken market while a TEQ value of 0.191 was calculated with PCB 126 presenting the highest sub-TEQ value of 0.1903 as shown in Fig. 5. The quota of toxic equivalency (TEQ) for PCBs in *S. scombrus* peaked at 99.63% for PCB 126 as presented in Fig. 6.

A total of 209 PCB congeners have been reported to exist in the environment (Kampire *et al.*, 2015). However, only 6, namely PCB 114, PCB 126, PCB 138, PCB 153, PCB 156 and PCB 169, were observed to be present in *S. scombrus* in this research. Similarly, Adeyemi *et al.*, (2009) identified 8 PCB congeners in the muscle of *Tilapia zillii* (red-belly tilapia), *Ethmalosa fimbriata* (bonga shad) and *Chrysichthys nigrodigitatus* (silver catfish) harvested from the Lagos Lagoon in Nigeria. Common to both researches were PCB 138, PCB 153 and PCB 156. Although the aforementioned PCBs have been presented using their congener identification, it is instructive to note that each congener also has an alternative IUPAC name, based on the attachment and arrangement of the available

Table 1. Summary statistics for PCBs (mg/kg) in *S. scombrus*

PCB congener	Mean	Minimum	Maximum	Threshold*
PCB 114	0.086	0	0.98	2
PCB 126	1.903	1.33	2.25	2
PCB 138	0.495	0.01	1.83	2
PCB 153	0.109	0	0.51	2
PCB 156	0.022	0	0.1	2
PCB 169	0.068	0.01	0.2	2

*Food and Agriculture Organization of United Nations (1983) and Agency for Toxic Substances and Disease Registry (2016).

Table 2. Summary statistics for PCBs (mg/kg) in *S. scombrus* by market

Market	PCB 114	PCB 126	PCB 138	PCB 153	PCB 156	PCB 169
Ekiedo	0.058[a]	2.03[a]	0.55[a]	0.13[a]	0.008[a]	0.05[a]
Ekioba	0.057[a]	1.702[a]	0.42[a]	0.087[a]	0.027[a]	0.065[a]
Edaiken	0.22[a]	2.0[a]	0.68[a]	0.18[a]	0.035[a]	0.087[a]
Ekiosa	0.012[a]	1.89[a]	0.33[a]	0.04[a]	0.02[a]	0.07[a]

Means with similar superscripts are not significantly different ($P>0.05$). Vertical comparisons only.

Table 3. Summary statistics for PCBs (mg/kg) in *S. scombrus* by months

Month	PCB 114	PCB 126	PCB 138	PCB 153	PCB 156	PCB 169
June	0.03[b]	1.93[a]	0.163[b]	0.09[a]	0.033[a]	0.098[a]
July	0.048[b]	1.65[a]	0.10[b]	0.075[a]	0.01[a]	0.083[a]
August	0.028[b]	1.76[a]	0.97[a]	0.10[a]	0.018[a]	0.045[a]
September	0.07[b]	2.07[a]	0.23[b]	0.15[a]	0.0025[a]	0.058[a]
October	0.038[b]	1.74[a]	1.23[a]	0.11[a]	0.020[a]	0.050[a]
November	0.31[a]	2.0[a]	0.27[b]	0.14[a]	0.053[a]	0.075[a]

Means with similar superscripts are not significantly different ($P>0.05$). Vertical comparisons only.

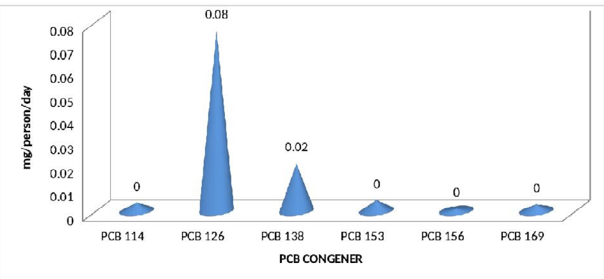

Fig. 2. Estimated daily intake (EDI) values for PCBs

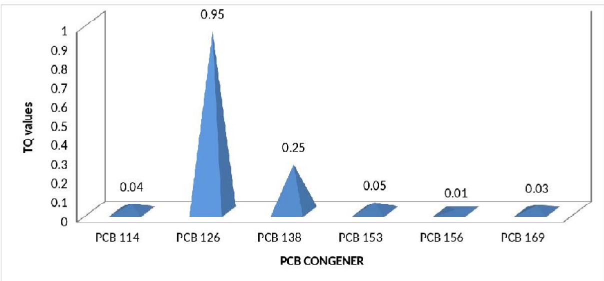

Fig. 3. Toxic quotient (TQ) values for PCBs

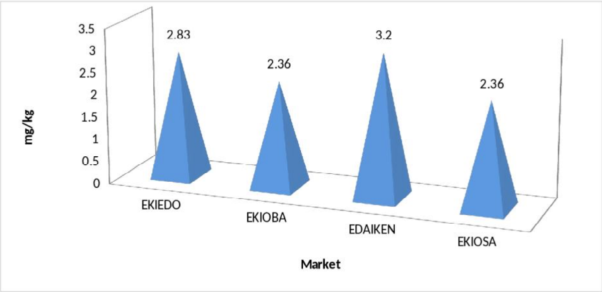

Fig. 4. Total PCB content in *S. scombrus* by market

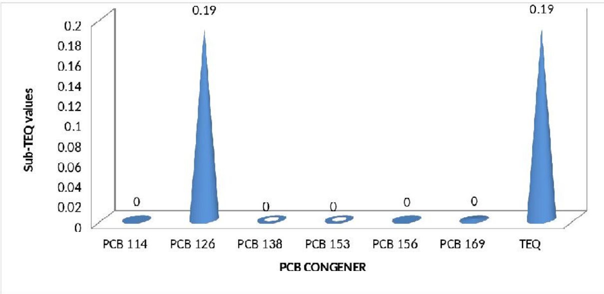

Fig. 5. Toxic equivalency (TEQ) values for PCBs in *S. scombrus*

and associated chlorine atoms. For example, PCB 114 is also known as 2,3,4,4',5-Pentachlorobiphenyl, while PCB 126 is known as 3,3',4,4',5-Pentachlorobiphenyl. PCB 138 and PCB 153 are known as 2,2',3,4,4',5'-Hexachlorobiphenyl and 2,2',4,4',5,5'-Hexachlorobiphenyl, respectively while PCB 156 and PCB 169 are known as 2,3,3',4,4',5-Hexachlorobiphenyl and 3,3',4,4',5,5'-Hexachlorobiphenyl, respectively (New York State Department of Environmental Conservation, 2004). The PCB rank profile in *S. scombrus* was PCB 126> PCB 138 > PCB 114 > PCB 169 > PCB 153 > PCB 156. This observed profile is suggestive of the dominance of PCB 126 over the other congeners in *S. scombrus*.

The PCB content in fish could clearly be defined by the PCB content of the ambient water medium in which such fish thrives, which could further influence the extent of subsequent bioaccumulation. Based on this premise, it could be posited that the aforementioned PCBs are probably dominant in the home marine water from where the fish species were harvested. Furthermore, it has been reported that fish due to relatively low activity of the mono-oxygenase enzymes has a small capacity to metabolize persistent, lipophilic and bioaccumulative environmental pollutants such as PCBs (Falandysz, 2000). The lipophilic and hydrophobic properties of PCBs may also contribute to their presence in the tissues of aquatic animals, including fish (Voorspoels *et al.,* 2004; Filipkowska, 2013). It was observed that there are no significant differences (P>0.05) in the mean concentrations of the PCB congeners in *S. scombrus* between markets suggesting that these fishes may have come from a common source or supplier. However, on the other hand, there are significant differences (P<0.05) in the mean concentrations of PCB 114 and PCB 138 in *S. scombrus* between months indicating the possibility of temporal variation. The estimated daily intake (EDI) values for PCBs revealed that PCB 126 and PCB 156 have the highest and lowest values, respectively. This finding can be attributed to the fact that PCB 126 had the highest mean concentration in *S. scombrus* while the opposite is the case for PCB 156. Wangboje and Okpobo (2019) observed that the EDI ranking and consequent pattern is usually defined by the concentrations of chemical contaminants in fish samples. According to the Agency for Toxic Substances and Disease Registry (2016), the World Health Organization (WHO) and the Food and Agriculture Organization of the United Nations (FAO), have recommended that the daily intake of PCBs should not exceed 6µg/kg per day. The toxic quotient (TQ) values for PCBs in *S. scombrus* were observed to be generally below unity although PCB 126 had a TQ score of 0.95, which is very close to unity. The TQ values are used in evaluating non-carcinogenic risk. Accordingly, the PCBs in *S. scombrus* do not present a carcinogenic risk to the consuming public, at the moment.

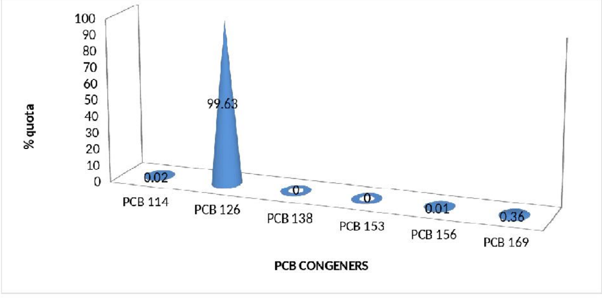

Fig. 6. Quota of toxic equivalency (TEQ) for PCBs in *S. scombrus*

The total PCB content (TPCB) in fish by market revealed that Ekioba and Edaiken markets have the lowest and highest TPCB values, respectively, giving an indication that ideally *S. scombrus* should be purchased from Ekioba market owing to the relative lower PCB burden in such fish. The TEQ calculated in this research is 0.191 with PCB 126, accounting for 99.63% of this value while PCB 156 accounted for 0.0057% on the opposite end of the scale. This finding further reveals the potency of PCB 126, which must be closely monitored in order to curtail human health risk in future. To buttress this, PCB 126 has been recognized to be an endocrine disruptor and causes hepatic fat accumulation (Boucher *et al.*, 2015). It has also been observed to be the most potent dioxin-like PCB, which disrupts energy metabolism (Zhang *et al.*, 2012). It is pertinent to note that presently, toxic equivalency factors (TEF) for PCB 138 and PCB 153, which are required for the computation of TEQ values, are yet to be established and published. The mean concentrations of PCBs in *S. scombrus* did exceed the threshold of 2.0 mg/kg as provided for fish by the Food and Agriculture Organization of the United Nations (1983) and Agency for Toxic Substances and Disease Registry (2016). This observation is thus in agreement with the interpretation of the TQ values obtained in this research. It can thus be said that *S. scombrus* purchased from the aforementioned markets is relatively safe for human consumption with regard to PCB content. The research has successfully provided the PCB profile of *S. scombrus* that is readily available in markets in Benin metropolis, Nigeria. Although a clean bill can be given to this fish species regarding PCB contamination and human consumption, attention must be paid to PCB 126 in particular as it has the potential to be of human risk in future. Being an imported species, *S. scombrus* must be spot-checked by relevant enforcement agencies at the point of entry in order to curtail episodic hazards associated with the consumption of PCBs via fish. Information from this research is expected to serve as a reference point for future studies as baseline data for PCBs in *S. scombrus* have been provided.

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