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Health risk assessment of polychlorinated biphenyls in a fish species (*Clarias gariepinus*) from southwestern rivers, Nigeria†

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The catfish (*Clarias gariepinus*) is commonly eaten in Nigeria, especially in the southwestern region. In this study, the levels of polychlorinated biphenyls (PCBs) in the muscles of *Clarias gariepinus* from six major rivers in the area were measured using gas chromatography with an electron capture detector. The PCB concentration ranged from 4.63 to 21.96 mg kg^{-1} in the dry season and from 5.26 to 23.52 mg kg^{-1} in the wet season. There were significant differences in PCB concentrations between the two seasons. The $\sum$PCB concentrations at any location were above the Food and Drug Administration tolerance level of 2.0 mg kg^{-1} and other regulatory limits. The study found that chlorinated PCB congeners with a high octanol–water partition coefficient (K_{ow}) dominated the congener profiles. The most dominant congener was #101, comprising 12.3% to 17.8% of the total PCB concentration. The study also found that the levels of non-carcinogenic hazard quotient (HQ) and hazard index (HI) were below 1, suggesting no non-carcinogenic health risks from consuming *Clarias gariepinus*. However, the cumulative cancer risks ($\sum$CR) fall within the low CR classification (10^{-6}–10^{-4}) by USEPA for all age categories. Also, the toxic equivalent quantity range was comparatively and significantly higher than the estimated non-carcinogenic screening values, suggesting potential health concerns. The study concluded that regular and continuous consumption of *Clarias gariepinus* as a significant portion of the diet may expose humans to unacceptable PCB concentrations due to residual environmental concentrations rather than a recent introduction.

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Environmental significance

This study highlights critical environmental issues linked to the contamination of aquatic ecosystems with polychlorinated biphenyls (PCBs). PCBs, being highly persistent and bioaccumulative, pose a long-term threat to biodiversity and human health through bio-magnification in the food chain. The research findings emphasize the vulnerability of aquatic environments, especially in regions with poor waste management practices, to industrial, agricultural, and domestic pollutants. The elevated PCB levels in *Clarias gariepinus*, a staple fish species in Nigeria, underline the urgent need for stricter regulation of waste discharge into water bodies. The results serve as a baseline for identifying hotspots of PCB contamination and encourage proactive monitoring of aquatic ecosystems. Concentrating on the effects of PCBs, the prevention of indiscriminate waste disposal and the preservation of local water resources to safeguard public health and preserve ecological balance are promoted by this study. The study also highlights the possible health risks of eating contaminated fish, underscoring the significance of protecting aquatic biodiversity for future generations.

1. Introduction

Polychlorinated biphenyls (PCBs) are synthetic organochlorine chemicals that were utilized as coolants and lubricants in transformers, generators, and electrical capacitors due to their electrical insulating qualities, low burning capacity, and chemical inertness.[1] They were also utilized to make plasticizers for rubber and polyvinyl chloride polymers.[2] Historically, they were made in the United States and Europe, and consequent to their highly toxic nature, the US government banned their production in 1977.[3] Going by the persistence in the environment, PCBs were included in the United Nations Environmental

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Programme Stockholm Convention (2001) list of persistent organic pollutants (POPs) and later in 2004 by the Organization for Economic Co-operation and Development.[4] However, despite the ban, PCB-containing electrical transformers and capacitors in use at that time were approved to remain in place or "grandfathered" into continued use.[5] The spread continues because of slow degradation and high environmental persistence at dangerous levels. High levels of these chemicals have been discovered in non-producing regions such as Africa,[6] and this has been linked to PCB atmospheric deposition. In Nigeria, the importation of used and old electrical equipment from developed nations, including electrical transformers and oils, during the 1970s and 1980s contributed to the presence of PCBs in the environment, particularly in the energy production sector. Additionally, run-offs from dumpsites where wastes from various sectors—including plasticizers, paints, adhesives, lubricants, and capacitors—are indiscriminately discarded have also been recognized as significant local sources of PCBs in Nigerian aquatic habitats and many other parts of the world.[7]

Generally, PCBs and various organic pollutants enter the aquatic environment and become major sinks and sources in biological systems, including sediment, affecting the entire ecosystem.[8,9] Most of these pollutants accumulate in organisms, reaching levels several times higher than in the water. The behavior and distribution of PCBs in aquatic systems depend significantly on the octanol–water partitioning coefficient (K_{ow}).[10,11] The K_{ow} values are often used as a measure of the hydrophobic nature to estimate bioconcentration factors (accumulation after uptake from water), toxicity, and aquatic solubility.[12,13] This is based on the assumption that the uptake of an organic substance is driven by its hydrophobicity.

Previous and most recent studies have identified fish as one suitable bioindicator of contaminants, including PCBs.[14,15] Going by the feeding nature of some fish species including *Clarias gariepinus*, they can be exposed to such contaminated sediments, particularly during sediment remobilization. People can be indirectly exposed to this toxic chemical by consuming contaminated fish. In a recent study, the primary route of PCB absorption in man has been observed to be dietary exposure *via* the ingestion of aquatic animals, particularly fish.[16] The accumulation of poorly metabolized compounds in fish can thus indicate the level of pollution in an aquatic system.[17] The Environmental Protection Agency (EPA) rates PCBs as "probable human carcinogens" since they cause cancer in laboratory animals. In addition, the biological effects can cause cardiovascular disease, immune system suppression, neuro-behavioral changes, hypertension, hypothyroidism, infertility, and disorders of the reproductive system, among others.[1]

One of the most widely distributed African freshwater omnivorous fish is the African sharp-tooth catfish *Clarias gariepinus. Clarias gariepinus* and others like *Heterobranchus dorsalis*, *Clarias buthupogon*, *Clarias anguillaris*, and *Clarias nigrodigitatus* are the most common species found in Nigeria.[18] Due to its hardiness and excellent growth rate, the *gariepinus* species is a common and significant cultured species, highly valued in Nigeria as a staple food for local communities and an ideal species for aquaculture. *Clarias gariepinus* is commonly used for pollution monitoring and is regarded as having high ecological significance. Therefore, it is a suitable organism for monitoring organic pollutants in aquatic environments.[19] The relatively high-fat content of catfish meat allows fat-soluble environmental pollutants, including PCBs, to accumulate steadily.[20] Several studies in Nigeria have investigated the levels of various toxicants in *Clarias* sp.[18,21–25] Similarly, some previous studies on catfish and other fishes in Nigeria reported wide PCB concentration ranges including 0.064–4.254 mg kg^{-1} in Wupa river, Nigeria,[24] 0.00444–0.00501 mg kg^{-1} in Otamiri River,[26] and 1.64–16.4 mg kg^{-1} and 0.56 mg kg^{-1} in Ogun and Ona rivers, respectively in Nigeria.[27] In some other fishes, a range of 0.795–1.830 mg kg^{-1} and 0.557–1.688 mg kg^{-1} had been reported.[28,29] These concentration ranges suggest that catfish consumption may be a significant source of PCB exposure.[30,31]

In several ecotoxicological studies, fish have been used to monitor several pollutants, including PCBs.[19,32–34] Generally, the use of biological groups such as phytoplankton, invertebrates, and fish for pollution monitoring is a vital and rapidly growing field.[17,25,35,36] In a recent report from the location investigated in this study, concentrations of PCBs were detected in sediments at levels of concern.[37] In Nigeria and most African nations, information on PCB distribution across major rivers, coupled with congener-specific data, is limited. In addition, there is very scarce data on non-ortho congeners. Non-ortho PCBs (such as PCBs 77, 126, and 169) are considered particularly toxic due to their structural similarity to dioxins and are often referred to as coplanar PCBs. A better understanding of factors increasing PCB body burden in fish will help identify and provide solutions for exposure through subsistence fishing and other potential sources. Therefore, the objectives of this study are (i) to provide reliable data on PCBs in the *Clarias gariepinus* species with regard to tissue concentration from selected rivers within Southwestern Nigeria, (ii) to quantify the daily dietary intake of PCBs, and (iii) to evaluate the potential health risks associated with fish consumption from the study locations.

2. Materials and methods

2.1. The study area

The locations and rivers examined (Fig. 1) have been detailed elsewhere.[9,36–42] In summary, the six major rivers [Ibese River: IBR, Ogbese River: OGBR, Ona River: ONR, Asejire River: ASR, Ogun River: OGR, and the Ocean housing the Third Mainland Bridge (TMBO), Lagos] studied in this research are highly significant within the southwestern geopolitical zone in Nigeria (Fig. 1). Two of the rivers (TMBO and IBR) are situated in the city of Lagos State, a densely populated industrial and economic city in Nigeria. The remaining rivers are found in other states, where they serve as major water sources for agriculture, including fishing and irrigation, as well as for domestic activities.[39,40] One of the rivers (ASR) has a dam on it that provides water for the waterworks in Oyo State, Nigeria, for processing potable water for the state. The areas surrounding these rivers are typically home to various industries, including food production, beverages, agriculture, and pharmaceuticals. Unfortunately, untreated effluents from these industries are discharged

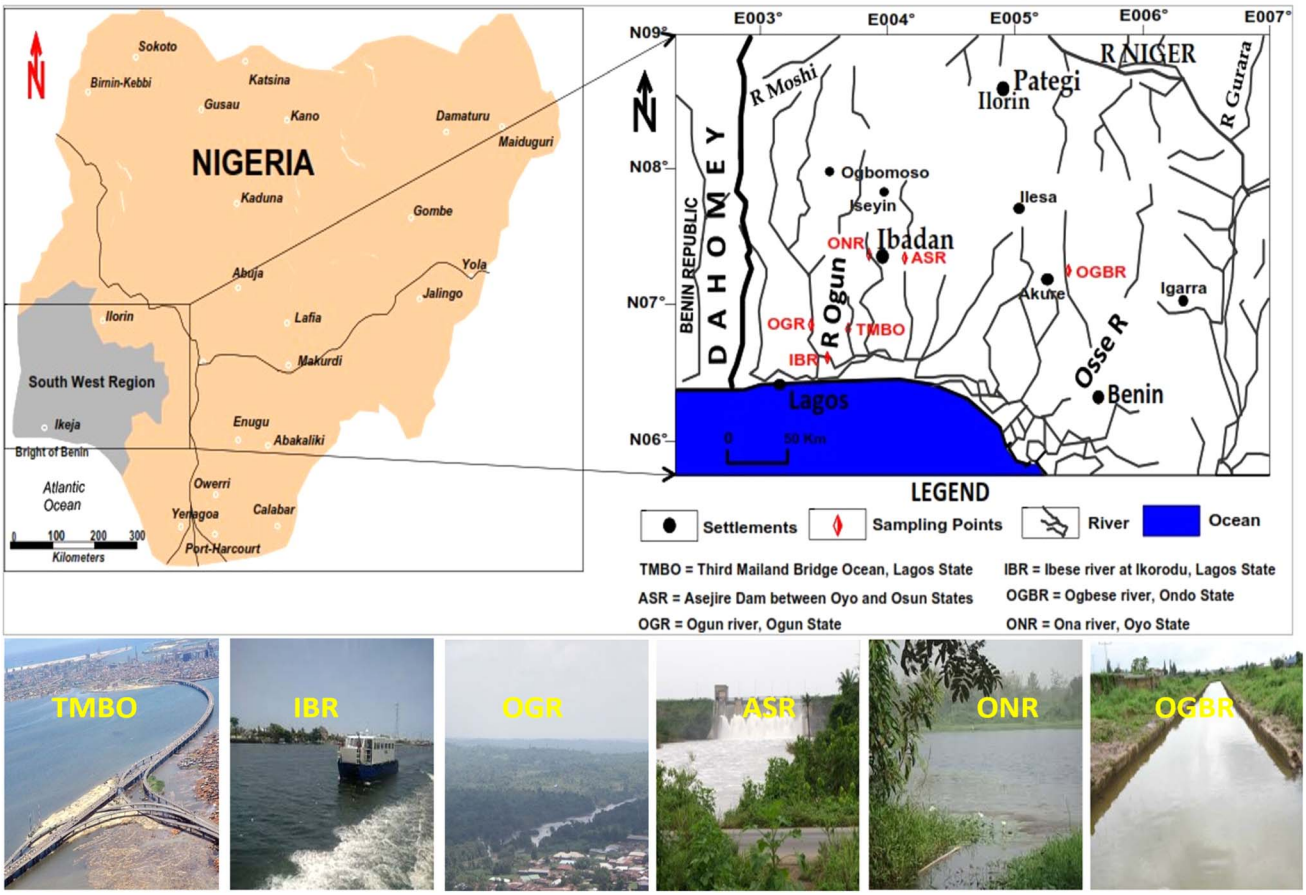

Fig. 1 Area map of studied locations. The top left is the geographical map of Nigeria displaying the Southwestern zone. The top right is the GIS map of the rivers investigated. The bottom part contains photographs of the rivers studied.

directly into the river through a network of canals. Additionally, drains are also connected to these rivers, leading to an indiscriminate discharge of various waste products. During the wet season, the magnitude of waste becomes huge due to run-off activities.

2.2. Sample collection and preparation

Sampling was carried out during the dry and wet seasons of 2020 to 2022, as recently reported.[37,42,43] Catfish (*C. gariepinus*) were sampled from all the investigated rivers. The ready availability at both seasons of the year was part of the reason for using this species in the current study. Fish were caught with the help of local fishermen. At each location, not less than fifteen (15) samples were obtained randomly, from which those considered of the same weight (115–122 g) and length (8.5–9.2 cm) were used for the analysis. Samples were washed with the river water, wrapped in hexane-treated aluminum foil, and placed inside an open-glass vessel within an ice box in the field. All samples were kept frozen at −20 °C on arrival in the laboratory before analysis. The head, backbone, fins, and tails were carefully removed from the partially frozen fish during preparation. Only the muscle of the fish, being the most desirable for consumption, was taken, chopped into small portions, homogenized, and separately pooled to form a composite for each river. The samples were later preserved under liquid nitrogen before analysis.

2.3. Chemicals and standards

All the chemicals used, including dichloromethane and *n*-hexane (BDH, Poole, UK), and a standard PCB mixture (Supelco, Bellefonte, PA, USA) were of pesticide residue grade. Sulfuric acid and silica gel were from Fisher Scientific. Other solvents and chemicals, including anhydrous sodium sulfate, were obtained from Darmstadt, Germany.

2.4. Extraction and analysis

The muscle tissues were sampled and processed for analysis, with three samples taken from each river, totaling eighteen samples per season. The extraction (in triplicate) follows EPA Method 8082A with a slight modification. Briefly, the fish samples (15 g each) were homogenized in excess anhydrous Na_2SO_4 (in a proportion of 3 : 1, Na_2SO_4: tissue) to remove excess moisture. The samples were Soxhlet extracted with 200 mL hexane: acetone (1 : 1 v/v) for 8 h (a mixture of hexane-acetone limits interference during extraction and improves the signal-to-noise ratio). Obtained fat extracts (after three cycles of

extraction) were cleaned on sulfuric acid (certified ACS, Fisher Scientific)–silica gel (40% [vol/wt], Fisher Scientific). The final volume of the purified extract was reduced to 2 mL and transferred into a vial ready for gas chromatography-electron capture detector (GC-ECD) analysis.

The extract was analyzed according to EPA Method 8082A with an Agilent 7820A gas chromatograph (Hewlett–Packard) coupled with an electron capture detector, as described previously.[37] Briefly, the carrier gas (He) flow rate is 6 mL min; the makeup gas (argon/methane or nitrogen) flow rate is 30 mL min; injector temperature is 205 °C; detector temperature is 290 °C; initial temperature is 140 °C which will be maintained for 2 min; the temperature program is from 140 °C to 240 °C with holding for 5 min; and the final temperature is 265 °C which will be maintained for 18 min. Data acquisition was performed using Varian Star software V5.52. PCB standards were used to prepare a calibration for the measurement.

Quantification was achieved using an external standard calibration method. The method requires that a five-point calibration is obtained for linearity and to establish the response factor for a minimum of 5 peaks from each standard. The five-point calibration generated (congener peak areas *versus* standard concentrations) produced a regression coefficient (r^2) of 0.9997.

2.5. Quality control and assurance

Apart from using pesticide residue grade reagents, this study employed matrix spike methods, procedural blanks ($n = 3$), and recoveries of standards ($^{13}C_{12}$-PCBs) for quality control and assurance. PCBs were not detected in the procedural blanks, indicating that there were no interference peaks present. Recoveries were determined by external standard calibration ($n = 3$) with five replicate samples and duplicate GC injections. The recovery from the matrix spiked sample ranged from 93.7–109%, while the surrogate PCB recoveries ranged from 88.9–96.7%. The limit of detection (LOD) and limit of quantification (LOQ, 0.2–0.6 mg kg^{-1}) were based on signal-to-noise ratios of 3 and 10, respectively.

2.6. Data analysis and risk assessment

In this study, we evaluated both carcinogenic and non-carcinogenic risks, following our recent publications.[9,37–43] The assessment of the non-carcinogenic risks involved the estimation of the target hazard quotient (THQ) and hazard index (HI). However, the assessment of carcinogenic risks was based on toxic equivalency factors (TEFs): measures of the toxicity of other congeners in relation to the most toxic dioxin compound (2,3,7,8-tetrachlorodibenzo-p-dioxin, 2,3,7,8-TCDD),[44] through which the toxic equivalent quantity (TEQ) was estimated following the referenced guidelines.[45] (See Text S1†).

The exposure assessment focused on the ingestion pathway, which is a primary route of exposure through consumption among children, adolescents, and adults. The exposure dose *via* ingestion was estimated based on established constants from the literature (See Table S1†).[46–48] In this study, the incremental lifetime cancer risk *via* ingestion (CR_{ig}) of *C. gariepinus* was estimated following the United States Environmental Protection Agency (USEPA) guidelines.[49] The qualitative description of the CR as prescribed by the Risk Assessment Guidance was strictly followed in assessing the model (See Text S1†).[49]

Descriptive statistics were estimated using the Origin 2018 software. The seasonal trend in PCBs was evaluated with a one-way analysis of variance (ANOVA). In this study, a p-value of < 0.05 was considered statistically significant.

3. Results and discussion

3.1. PCB distribution in fish

The seasonal mean contents of PCBs in edible tissues of *Clarias gariepinus* from Southwestern rivers in Nigeria are summarized in Table 1 and Fig. 2. Of the 209 PCB congeners, twenty-five congeners were detected across all locations. In general, all samples are mixtures of Aroclor. That is, no sample in the data set represents a 100% contribution from a single source. Varying contributions of multiple sources impact every sample. However, based on the Nigerian environment, and the percentage of chlorine content, Aroclor 1242, which contains 42% chlorine by weight, and Aroclor 1254, which contains 54% chlorine by weight in relation to their industrial applications in electrical components, including heat transfer fluids, transformers, and capacitors within Nigeria, are suspected Aroclor formulations for the congeners detected in this study. In approximately 85% of all the samples, the individual PCB concentrations were below 1.0 mg kg^{-1} across all locations and seasons. The muscle tissues' total PCB concentrations (the sum of the twenty-five PCB congeners) varied across the two seasons (Fig. 2). In the wet season of 2020 and 2021, the sum of the PCBs ranged from 5.43–22.14 mg kg^{-1} with a median value of 11.98 mg kg^{-1}, and from 5.26–23.52 mg kg^{-1} with a median value of 12.94 mg kg^{-1}, respectively. However, in the dry season of 2019, the PCB content ranged from 4.66–21.96 mg kg^{-1} with a median value of 11.79 mg kg^{-1}, while in 2020, it ranged from 4.63 mg kg^{-1}–21.50 mg kg^{-1} with a median value of 11.57 mg kg^{-1} across all locations (Table 1). The data showed no significant differences in the median values across the various sampling seasons, which suggests (i) a small increase in PCBs across months and (ii) stable sources of this pollutant regardless of the season involved. In addition, fish, being a migratory species, may not necessarily be stable at a location to bioaccumulate, considering the rivers in question. Thus, dilution as the fish migrates across different ionic strengths within the river may be responsible for the lack of major seasonal differences. A more critical assessment showed that the $\sum$PCB content recorded at each of the locations is slightly higher during the wet season (Fig. 2).

Fish sampled from the TMBO recorded the highest concentrations of PCBs. In general, based on the location, the PCB concentrations in the tissues decreased in the order TMBO > IBR > ONR > OGR > OGBR > ASR, with PCB-153 markedly dominant in all tissues. Thus, the study showed a statistically significant difference between the mean of all rivers as determined by one-way ANOVA [$F(2.516) = 0.0046$, $p < 0.05$]. Between ONR and OGR, there were no significant mean differences ($p > 0.05$). Similarly, between ASR and OGBR, no observable

Table 1 Mean concentration (mg kg^{-1}) of PCBs in fish across the two seasons (mean ± SD)[a]

PCB	K_{ow}	ASR	ONR	OGBR	OGR	IBR	TMB0
PCB 8	5.301	0.039 ± 0.009	0.023 ± 0.004	0.011 ± 0.004	0.033 ± 0.006	0.170 ± 0.020	0.142 ± 0.020
PCB 18	5.551	0.307 ± 0.020	0.277 ± 0.020	0.167 ± 0.020	0.325 ± 0.030	0.460 ± 0.040	0.659 ± 0.030
PCB 28	5.691	0.150 ± 0.020	0.194 ± 0.020	0.129 ± 0.020	0.229 ± 0.020	0.338 ± 0.020	0.487 ± 0.040
PCB 44	5.811	0.120 ± 0.010	0.228 ± 0.010	0.114 ± 0.010	0.191 ± 0.020	0.256 ± 0.020	0.331 ± 0.010
PCB 52	6.091	0.160 ± 0.020	0.260 ± 0.030	0.084 ± 0.009	0.280 ± 0.030	0.139 ± 0.020	0.630 ± 0.040
PCB 66	5.452	0.230 ± 0.040	0.270 ± 0.020	0.230 ± 0.020	0.310 ± 0.030	0.550 ± 0.030	0.693 ± 0.020
PCB 77	6.523	0.120 ± 0.020	0.240 ± 0.020	0.230 ± 0.310	0.340 ± 0.030	0.490 ± 0.030	0.830 ± 0.040
PCB 81	6.367	0.640 ± 0.030	0.870 ± 0.460	0.450 ± 0.020	0.640 ± 0.040	1.270 ± 0.050	2.080 ± 0.070
PCB 101	7.071	0.880 ± 0.030	2.090 ± 0.080	0.800 ± 0.030	1.680 ± 0.040	2.070 ± 0.040	2.720 ± 0.060
PCB 105	6.657	0.130 ± 0.020	0.540 ± 0.040	0.087 ± 0.020	0.540 ± 0.030	1.040 ± 0.050	1.440 ± 0.060
PCB 114	6.657	0.150 ± 0.020	0.340 ± 0.040	0.120 ± 0.020	0.290 ± 0.040	0.440 ± 0.030	0.630 ± 0.020
PCB 118	7.121	0.055 ± 0.006	0.010 ± 0.003	0.020 ± 0.006	0.009 ± 0.003	0.011 ± 0.003	0.022 ± 0.005
PCB 126	6.897	0.280 ± 0.020	1.570 ± 0.040	0.220 ± 0.020	1.660 ± 0.050	2.080 ± 0.070	2.870 ± 0.060
PCB 128	6.961	0.750 ± 0.030	1.760 ± 0.050	0.380 ± 0.020	1.450 ± 0.060	2.060 ± 0.080	2.450 ± 0.060
PCB 138	7.441	0.008 ± 0.004	0.007 ± 0.003	0.009 ± 0.003	0.008 ± 0.003	0.009 ± 0.003	0.008 ± 0.004
PCB 153	7.751	0.056 ± 0.009	0.010 ± 0.003	0.021 ± 0.003	0.008 ± 0.003	0.009 ± 0.004	0.030 ± 0.009
PCB 156	7.187	0.009 ± 0.003	1.050 ± 0.030	0.009 ± 0.003	0.007 ± 0.004	1.610 ± 0.060	0.060 ± 0.010
PCB 157	7.187	0.650 ± 0.020	0.912 ± 0.003	0.433 ± 0.010	1.700 ± 0.050	1.208 ± 0.004	2.070 ± 0.080
PCB 167	7.277	0.220 ± 0.010	0.009 ± 0.003	0.190 ± 0.005	0.009 ± 0.003	0.008 ± 0.003	0.010 ± 0.004
PCB 169	7.427	0.340 ± 0.030	0.490 ± 0.020	0.280 ± 0.010	0.540 ± 0.030	0.990 ± 0.020	0.018 ± 0.008
PCB 170	7.277	0.009 ± 0.003	0.008 ± 0.003	0.008 ± 0.002	0.011 ± 0.004	0.010 ± 0.005	0.010 ± 0.003
PCB 180	6.961	0.670 ± 0.030	0.902 ± 0.030	0.570 ± 0.020	0.960 ± 0.050	1.450 ± 0.040	2.060 ± 0.050
PCB 187	7.177	0.130 ± 0.010	0.009 ± 0.003	0.064 ± 0.005	0.330 ± 0.020	0.010 ± 0.005	0.008 ± 0.003
PCB 195	7.567	0.093 ± 0.020	0.530 ± 0.020	0.073 ± 0.005	0.130 ± 0.020	0.330 ± 0.030	0.440 ± 0.040
PCB 206	9.143	0.234 ± 0.008	0.350 ± 0.020	0.290 ± 0.008	0.390 ± 0.020	0.690 ± 0.020	1.660 ± 0.060
Mean∑PCB		6.797	13.306	5.246	13.584	17.735	22.618
∑Co-PCB		2.599 ± 0.226	6.031 ± 0.497	2.046 ± 0.154	5.742 ± 0.627	9.179 ± 0.677	10.042 ± 1.052
∑Marker-PCB		1.974 ± 0.345	3.477 ± 0.772	1.636 ± 0.319	3.178 ± 0.638	4.029 ± 0.838	5.952 ± 1.096
% Marker-PCB		30.73	26.83	32.74	26.30	22.78	26.61

[a] ∑Co-PCB: Coplanar PCBs – PCB-77, 81, 105, 114, 118, 126, 156, 157, 167, and 169. ∑Marker-PCB: ICES indicator PCBs – PCB-28, −52, −101, −118, −138, −153, and −180.

significant difference was recorded ($P > 0.05$). However, there are significant differences between IBR, TMBO, and other locations investigated in the study. The relatively lower concentrations of PCBs found at ASR and OGBR in fish indicate a relatively low rate of PCB pollution input.

3.2. Profiles of the PCB congeners in fish

Across all samples, seasons, and locations, the most dominant congener was PCB-101 (comprising 12.3–17.8% of the ∑PCB concentration), followed by PCB-128 (7.8–15.1%), PCB-126 (4.3–13.7%), PCB-180 (7.6–10.6%), PCB-157 (6.8–10.1%), and PCB-81

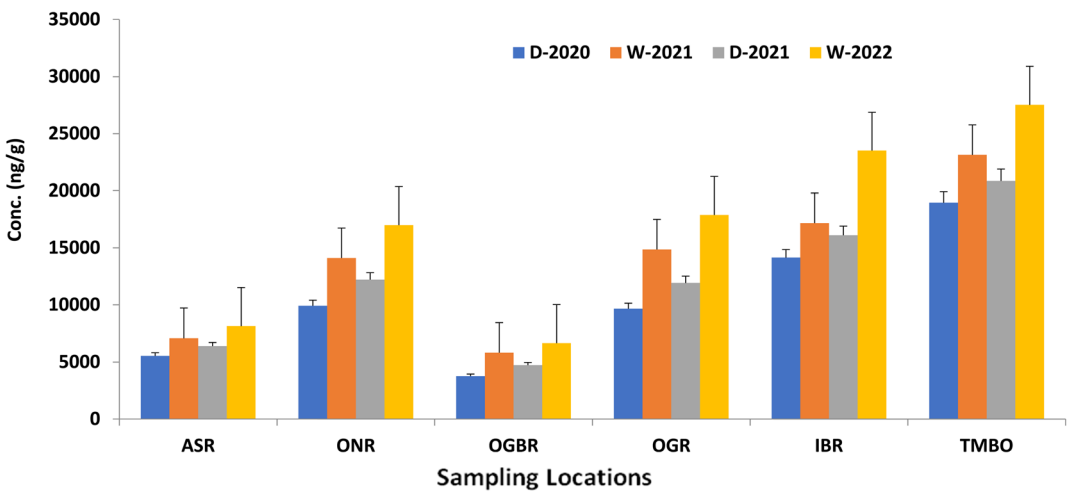

Fig. 2 Seasonal distribution of PCB across sampling locations. D-2019: Dry season of 2019; W-2020: Wet season of 2020; D2020: Dry season of 2020; and W2021: Wet season of 2021. ASR: Asejire River; ONR: Ona River; OGBR: Ogbese River; OGR: Ogun River; IBR: Ibe River; TMBO: Third Mainland Bridge Ocean.

(5.2–10.1%). The octanol–water partitioning coefficients (K_{ow}) of PCBs 81, 101, 126, 128, 157, and 180 are 6.367, 7.071, 6.897, 6.961, 7.187, and 6.961, respectively.[50] This parameter (K_{ow}) is very high among the respective isomeric group and PCB numbers detected in the study. These high K_{ow} values increase PCB bioaccumulation because they are more soluble in lipids, leading to accumulation in the fatty tissues of fish. Therefore, PCBs 81, 101, 126, 128, 157, and 180 can be considered to have high lipophilicity and persist in the tissues of the fish analyzed in the study. PCBs 101, 126, and 180 were particularly present in higher concentrations than the other congeners in the tissues. The two commonly detected PCB congeners in animal samples, PCBs 153 and 138, were detected in this study.[51] Interestingly, both PCBs were the least predominant in the study, even though they are highly lipophilic and have less tendency to be tightly bound to sediments, which should naturally enhance their availability for bioaccumulation.[52]

The coplanar (dioxin-like PCBs PCB-77, 81, 105, 114, 118, 126, 156, 157, 167, and 169) PCBs ranged from 2.05–10.04 mg kg^{-1} across all locations (Table 1), with OGR and TMBO recording the lowest and highest load, respectively. Similarly, the summarized statistical estimates of the Marker PCBs or Indicator PCBs PCB-28, 52, 101, 118, 138, 153, and 180 are included in Table 1. These indicator PCBs represent different PCB patterns to account for non-dioxin-like PCBs.[53] They are known to be persistent and highly bioaccumulative in the food chain.[53] Total indicator PCB concentrations (range) of 1.97 (0.008–0.879) mg kg^{-1}, 3.48 (0.008–2.093) mg kg^{-1}, 1.64 (0.009–0.800) mg kg^{-1}, 3.18 (0.008–1.682) mg kg^{-1}, 4.03 (0.009–2.071) mg kg^{-1}, and 5.95 (0.008–2.719) mg kg^{-1} were recorded at ASR, ONR, OGBR, OGR, IBR, and TMBO, respectively. Apart from PCBs-118, 138, and 153, all the other indicator PCBs exceeded the EU regulation of 0.075 mg kg^{-1} limit for ocean fish.[54] In general, the average percentage concentration of the sum of the indicator PCBs to ∑PCB across all locations (Table 1) was 27.7% (range 22.8–32.7%). This percentage is lower than the prescribed 50% attributable to the six indicator PCBs by the European Food Safety Authority.[53] The range falls within 24–42% in marine fish obtained from tsunami-stricken areas following the Great East Japan Earthquake in 2011.[55]

Bioaccumulation was reasonably high for PCBs 101 and 180 compared to PCBs 138 and 153, which have similar characteristics.[56] This suggests the contribution of other factors in controlling the rate of bioaccumulation, metabolism, and elimination from the organism.[57,58] The enhanced proportion of PCBs 101 (12.3–17.8%) and 180 (7.8–10.6%) of all PCB congeners across all locations possibly reflects a low rate of biotransformation, resistance to metabolism, and slow rate of elimination by cytochrome P-450 iso-enzymes, while reduced levels of PCBs 153 and 138 may reflect an increased metabolic and elimination rate.[59–61]

3.3. Influence of the physical and chemical properties of PCBs on bioaccumulation

The study shows that factors like equilibrium partition between the fish and ambient water possibly play a vital role in PCB uptake and elimination. *Clarias gariepinus* is a migratory species, capable of being exposed to PCBs either by dermal, inhalation, or ingestion routes. Consequently, defining levels of PCBs in fish revolves around several factors, including the physicochemical properties of the water. In this study, the order of decreasing homolog concentration follows TeCB > HeCB > PeCB > HpCB > TCB ≈ NoCB > OcCB > DCB (Fig. 3). It is not surprising that DCB is the lowest across all locations due to its ability to reach a quicker equilibrium between ambient water and *C. gariepinus* based on the relatively lower partition coefficient (K_{ow}). The trend equally reveals that highly chlorinated PCB congeners with high K_{ow} dominated the congener profiles. For instance, the high percentage levels (40–61%) of planar congeners (PCBs-77, 81, 105, 114, 118, 126, 156, 157, 167, and

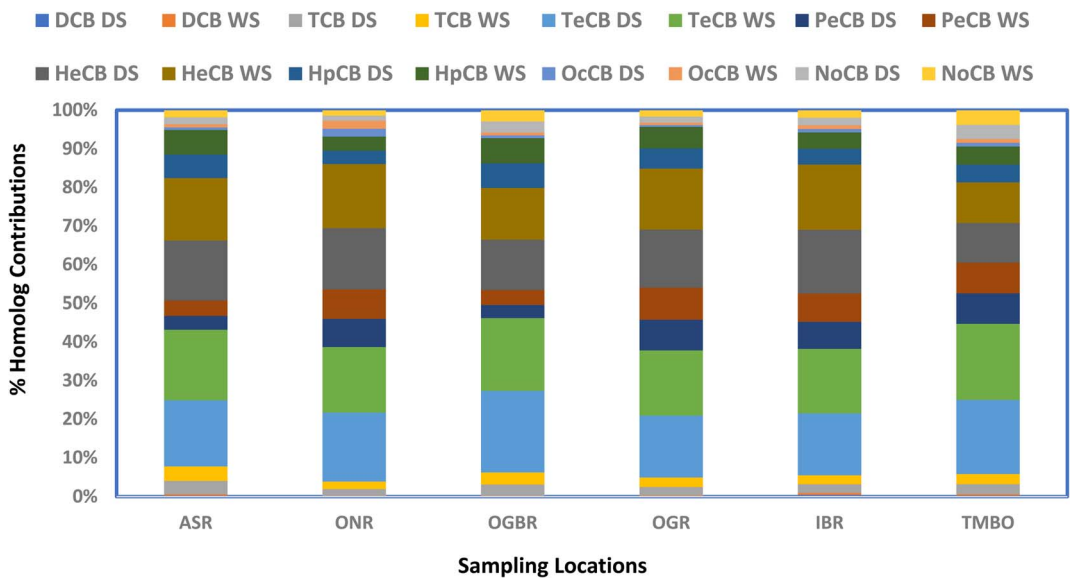

Fig. 3 Seasonal homolog contributions across sampling locations. Results are mean of each season.

169) can be attributed to their favorable accumulation *via* cell membranes.[62] In addition, varied PCB concentrations and congeners can also be related to the rate of metabolism.[63]

The trend as reported (proportion of the low-chlorinated PCBs) suggests the fish were not significantly contaminated with fresh PCBs because fish metabolize PCBs slowly compared to mammals.[64] By implication, the level of PCBs reported in this study may not be associated with fresh contamination of high levels of PCBs, which should have enhanced the proportions of the low-chlorinated PCBs in contrast to what was obtained in this study. Previous studies have shown that the detection of PCB-28 and PCB-52 suggests recent exposure.[65] However, the percentage contributions of these two PCBs (#28 : 1.76–2.36%; #52 : 0.93–2.80%) are lower compared to those of most other congeners detected in the study. In other words, the level of PCBs obtained in tissues of *C. gariepinus* in this study was most likely derived majorly from residual concentrations in the environment and less from recent introduction.

3.4. Comparison of PCB concentration with other studies

In comparison to previous studies (Table 2), the levels of PCBs in this study showed significant differences. The $\sum$25 PCB concentrations found in *Clarias gariepinus* from the rivers in this study were similar to the range of concentrations found in muscles of *Clarias gariepinus* from Ogun River, Nigeria (1.64–16.4 mg kg^{-1}),[66] and in fish from Galveston Bay, Texas (0.29–110 mg kg^{-1}).[67] However, the range of PCB concentrations in this study is much higher than those in several reports in Nigeria, including the levels found in *Tilapia zilli* from Lagos Lagoon (0.56–2.94 mg kg^{-1}),[68] fish from Eleyele Reservoir, Southwestern Nigeria (0.33–2.53 mg kg^{-1},[69] *Clarias gariepinus* from Ovia River, Southern Nigeria (0.001–0.003 mg kg^{-1}),[24] and *Clarias gariepinus* from Ogun River and Ona River (1.64–16.4 mg kg^{-1} and 0.56 mg kg^{-1}), respectively.[66]

In addition, various studies have reported lower concentrations of contaminants in different parts of the world. For instance, in Hartbeespoort Dam, South Africa, *Clarias gariepinus* had 0 mg kg^{-1} and *Cyprinus carpio* had 1.8 mg kg^{-1}.[33] Luxembourg River fish were reported to have concentrations ranging from 0.05 to 3.50 mg kg^{-1}.[70] Marine fish from tsunami-stricken areas of Japan showed concentrations ranging from 0.00044 to 0.086 mg kg^{-1},[55] while marine fish from the Persian Gulf exhibited concentrations from 0.0072 to 0.0902 mg kg^{-1}.[71] In the Red Sea coast (Saudi Arabia), PCB values were recorded in the range of 0.0002–0.0443 mg kg^{-1} for *C. Chanos* and 0.0339–0.0825 mg kg^{-1} for*M. cephalus*.[72] Additionally, muscles of *M. cephalus* from 15 sites on the coast of Florida, United States, were reported to have concentrations ranging from 0.0034 to 0.0593 mg kg^{-1},[73] while *Mugil* spp from Coastal lagoons, Central Mexican Pacific, showed concentrations from Bdl to 0.16 ng g^{-1} w/w,[34] which are far below the range recorded in this study. On the other hand, a much higher concentration of 128.0 mg kg^{-1}

Table 2 Comparison of PCB's concentration in muscles of *Clarias gariepinus* in southwestern rivers, Nigeria with national and global reports[a]

Locations	Name of species	Fish part	Concentrations	References
Southwest rivers, Nigeria	*Clarias gariepinus*	Muscle	DS: 4.63–21.96 mg kg^{-1} WS: 5.26–23.53 mg kg^{-1}	This study
Wupa river, Abuja, Nigeria	*Clarias anguillaris*		64–4254 ng g^{-1}	
Ovia river, Southern Nigeria	*Clarias gariepinus*	Muscle	0.001–0.003 mg kg^{-1}	Tongo and Ezemonye (2018)
Ogun river, Nigeria, Ona river, Nigeria	*Clarias gariepinus*	Muscle	1.64–16.4 mg kg^{-1} 0.56 mg kg^{-1}	Adeogun *et al.* (2016)
Lagos lagoon	*Tilapia zilli* (adult)	Muscle	00.56–2.94 mg kg^{-1}	Adeyemi *et al.* (2009)
Gulf of Mexico, Ionian sea	Bluefin tuna: *Thunnus thynnus* Yellowfin tuna: *Thunnus albacares*		8.08 ± 5.08 (w/w) 5.3–35.0 (15.9 ± 8.0) (w/w)	Nicklisch, *et al.* (2017), Storelli *et al.* (2008)
Hartbeespoort dam, South Africa	*Clarias gariepinus*	Muscle	0 mg kg^{-1} (5 year old catfish)	Rimayi and Chimuka (2019)
	Common carp (*Cyprinus carpio*)	Muscle	1.8 mg kg^{-1}	Rimayi and Chimuka (2019)
Laguna de Términos, Campeche	*Lepisosteus tropicus*	Muscle	0.782 ng g^{-1}	Carvalho *et al.* (2009)
	Crassostrea spp	Muscle	0.013–4.847 ng g^{-1}	Carvalho *et al.* (2009)
Coatzacoalcos river estuary, Veracruz, Mexico	*Centropomus parallelus*	Muscle	0.20 ng g^{-1}	Espinosa-Reyes *et al.* (2012)
Coast of Florida, United States	*M. cephalus*	Muscle	3.4–59.3 ng g^{-1}	Karouna-Renier *et al.* (2011)
Red sea coast, Saudi Arabia	*C. chanos*, *M. cephalus*	Muscle	0.22–44.3 ng g^{-1} (w/w) 33.9–82.5 ng g^{-1} (w/w)	Batang *et al.* (2016)
Zemplinska Sirara water reservoir	Common carp (*Cyprinus carpio*)	Muscle	10.2 mg kg^{-1}	Brazova *et al.* (2011)
Zemplinska Sirara water reservoir	Freshwater bream (*Abramis brama*)	Muscle	128.0 mg kg^{-1}	Brazova *et al.* (2011)
Coast of Malacca, Malaysia	*Lutjanus argentimaculatus*	Muscle	0.43–0.67 ng g^{-1} (w/w)	Mohamad *et al.* (2015)
Coastal lagoons, central Mexican Pacific	*Mugil* sp.	Muscle	Bdl–0.13 ng g^{-1} (w/w)	Ramírez-Ayala (2021)

[a] DS: Dry season; WS: Wet season.

Table 3 Summary of estimated daily intake (EDI, mg kg^{-1} per day) of PCBs ($\times 10^{-3}$) in children (CHD), adolescents (ADL), and adults (ADT)

		Minimum	Maximum	∑EDI
ASR	CHD	5.94×10^{-3}	0.671	4.894
	ADL	4.89×10^{-3}	0.551	4.027
	ADT	3.56×10^{-3}	0.402	2.935
ONR	CHD	5.72×10^{-3}	1.590	8.425
	ADL	4.70×10^{-3}	1.312	6.933
	ADT	3.43×10^{-3}	0.956	5.053
OGBR	CHD	5.91×10^{-3}	0.609	3.807
	ADL	4.85×10^{-3}	0.501	3.133
	ADT	3.54×10^{-3}	0.365	2.283
OGR	CHD	5.72×10^{-3}	1.295	9.208
	ADL	4.70×10^{-3}	1.066	7.577
	ADT	3.43×10^{-3}	0.777	5.522
IBR	CHD	6.27×10^{-3}	1.581	11.384
	ADL	5.16×10^{-3}	1.301	9.367
	ADT	3.76×10^{-3}	0.948	6.827
TMBO	CHD	5.85×10^{-3}	2.189	17.044
	ADL	4.81×10^{-3}	1.802	14.025
	ADT	3.51×10^{-3}	1.313	10.222

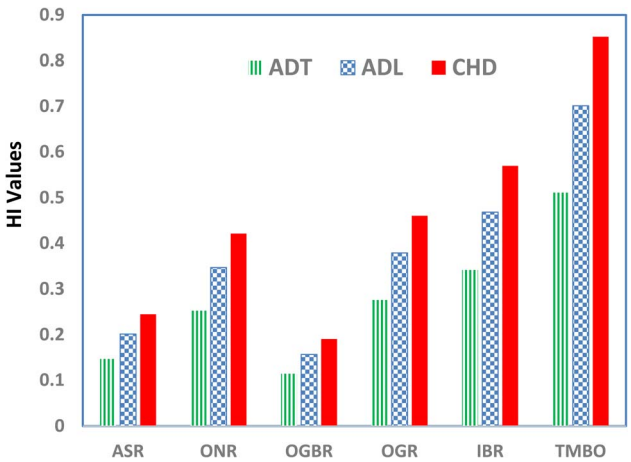

Fig. 4 Estimated hazard index of PCBs in Children (CHD), Adolescents (ADL), and Adults (ADT) through consumption of *Clarias gariepinus* from selected rivers from southwestern Nigeria. HI values all fell below the benchmark (1.0) of health concerns.

was found in *Abramis brama* from the Zemplinska Sirara Water reservoir, surpassing the mean concentration obtained in this study (Table 2).[74]

3.5. Human health risks

3.5.1. Regulatory risk assessment. Going by available regulatory criteria, often targeted at the protection of human health, a limit of 2.0 mg kg^{-1} PCBs was proposed by the United States Food and Drug Administration and the Swedish Food Regulation (SFR), while the Food and Agriculture Organization (FAO), World Health Organization (WHO), and Switzerland proposed a more stringent limit of 1.0 mg kg^{-1}.[75] The total concentrations obtained in the study at all locations are above these regulatory limits. Similarly, the PCB concentrations obtained were far above the EU's recommended safe limit of 0.000008 mg kg^{-1} for the sum of dioxins and dioxin-like PCBs in the muscle meat of fish.[76] Therefore, concerning human health risks, the level obtained at any location is of serious public concern. The enhanced PCB levels may be attributed to (i) an increase in the usage of PCB-containing materials in Nigeria, (ii) the effective uptake of PCBs by the species of fish used in the study (*Clarias gariepinus*),[33] and (iii) decreased PCB metabolism by *Clarias gariepinus*, which minimizes its elimination.[17]

3.5.2. Non-carcinogenic health risk assessment. Across various countries, varying estimated daily intake of PCBs from fish consumption have been reported, such as Spain (92 g per kg per day)[77] and China (1.62 ng per kg per day).[78] Using the mean PCB levels of both seasons, the summary statistics of the total estimated daily intake (∑EDI) showed ranges from 3.81×10^{-3} to 17.04×10^{-3} mg per kg per day, 2.35×10^{-3} to 10.53×10^{-3} mg per kg per day, and 2.28×10^{-3} to 10.22×10^{-3} mg kg^{-1} per day for children, adolescents, and adults, respectively (Table 3). The daily intake of PCBs across all locations was found to exceed the reference dose (2×10^{-5} mg per kg per day), indicating a high risk from fish consumption. The summary of the hazard quotient (HQ) is provided in Table S2.† The data estimated in this study showed that except for PCB 126 at TMBO, the HQs for PCBs were below the benchmark of 1.0 at all locations, for children, adolescents, and adults. Similarly, the estimated HI obtained across all locations was also generally low (<1.0), ranging from 0.19–0.85, 0.20–0.70, and 0.15–0.51, respectively (Fig. 4). This trend suggests no significant non-

Table 4 Estimated cancer risks of PCBs[a]

		ASR	ONR	OGBR	OGR	IBR	TMBO	SV
Children	Min	7.129×10^{-6}	6.855×10^{-6}	7.083×10^{-6}	6.855×10^{-6}	7.518×10^{-6}	7.015×10^{-6}	0.0260 (0.0066)
	Max	8.032×10^{-4}	1.913×10^{-3}	7.310×10^{-4}	1.554×10^{-3}	1.897×10^{-3}	2.626×10^{-3}	
	Total	5.871×10^{-3}	1.011×10^{-2}	4.567×10^{-3}	1.104×10^{2}	1.365×10^{-2}	2.044×10^{-2}	
Adolescents	Min	9.781×10^{-6}	9.405×10^{-6}	9.718×10^{-6}	9.405×10^{-6}	1.031×10^{-5}	9.624×10^{-6}	0.0425 (0.0106)
	Max	1.102×10^{-3}	2.625×10^{-3}	1.003×10^{-3}	2.131×10^{-3}	2.602×10^{-3}	3.603×10^{-3}	
	Total	8.054×10^{-3}	1.386×10^{-2}	6.266×10^{-3}	1.515×10^{-2}	1.873×10^{-2}	2.805×10^{-2}	
Adults	Min	1.189×10^{-5}	1.143×10^{-5}	1.181×10^{-5}	1.143×10^{-5}	1.253×10^{-5}	1.170×10^{-5}	0.0437 (0.0109)
	Max	1.339×10^{-3}	3.190×10^{-3}	1.219×10^{-3}	2.590×10^{-3}	3.163×10^{-3}	4.379×10^{-3}	
	Total	9.788×10^{-3}	1.685×10^{-2}	7.615×10^{-2}	1.842×10^{-2}	2.277×10^{-2}	3.409×10^{-2}	

[a] SV: Screening value: values in brackets are SV for carcinogenic risks while those outside brackets are for non-carcinogenic risks.

Fig. 5 (a) Estimates of carcinogenic potency of individual PCBs in Children. A target risk level of 10^{-6} as prescribed by USEPA denotes negligible cancer risks for individual chemicals including PCB (USEPA, 2010). (b) Estimates of carcinogenic potency of individual PCBs in Adolescents. All estimates were above the target risk level of 10^{-6} prescribed by USEPA (USEPA, 2010). (c) Estimates of carcinogenic potency of individual PCBs in Adults. CR: Cancer risk.

Table 5 Estimated toxic equivalent quantity (TEQ) of PCBs in fish[ab]

	PCBs	TEF	ASR	ONR	OGBR	OGR	IBR	TMBO
Non-ortho	77	0.005	0.058	0.118	0.116	0.169	0.246	0.415
	81	0.0003	0.019	0.026	0.014	0.019	0.038	0.062
	126	0.1	2.800	15.684	2.211	16.575	20.754	28.735
	169	0.01	1.026	1.481	0.849	1.629	2.963	0.054
∑(77–169)			3.904	17.308	3.189	18.393	24.001	29.266
Mono-ortho	105	0.0001	4.018×10^{-3}	0.016	2.624×10^{-3}	0.016	0.031	0.043
	114	0.0005	7.578×10^{-3}	0.017	5.763×10^{-3}	0.014	0.022	0.032
	118	0.0001	1.637×10^{-3}	2.947×10^{-4}	6.405×10^{-4}	2.873×10^{-4}	3.150×10^{-4}	0.001
	156	0.0005	4.425×10^{-4}	0.052	4.300×10^{-4}	3.750×10^{-4}	0.080	0.003
	157	0.0005	3.248×10^{-2}	5.800×10^{-4}	0.022	0.085	4.112×10^{-4}	0.104
	167	0.00001	6.622×10^{-4}	2.827×10^{-5}	5.801×10^{-4}	2.655×10^{-5}	2.527×10^{-5}	2.835×10^{-5}
∑(105–167)			0.047	0.087	0.032	0.117	0.134	0.182
Di-ortho	170	0.0001	2.580×10^{-4}	2.542×10^{-4}	2.325×10^{-4}	3.165×10^{-4}	3.360×10^{-4}	2.940×10^{-4}
	180	0.00001	6.672×10^{-4}	9.015×10^{-4}	5.728×10^{-4}	9.617×10^{-4}	1.453×10^{-3}	2.061×10^{-3}
∑(170–180)			9.252×10^{-4}	1.156×10^{-3}	8.053×10^{-4}	1.278×10^{-3}	1.789×10^{-3}	2.355×10^{-3}
TEQ = ∑ (77–180)			3.952	17.396	3.221	18.511	24.138	29.451
% Non-ortho PCBs			99.16	99.60	99.25	99.45	99.56	99.56
% PCB-126			54.18	82.60	55.35	85.78	80.12	80.81

[a] TEQ: toxic equivalent concentration = Σ [concentration of each dioxin-like congener X 2,3,7,8-TCDD TEF]. [b] TEF: toxic equivalent factor; $: Ding *et al.* (2012).

carcinogenic effect would arise from exposure to individual PCBs or their complex mixtures by consuming *Clarias gariepinus* from the investigated rivers. However, caution must be observed at TMBO based on the relatively high HI (0.51–0.85) across all age categories.

3.5.3. Assessment of carcinogenic health risks. PCBs have been classified as a group of carcinogens by USEPA. Therefore, indirect consumption of PCB-containing substances such as fish may promote both non-carcinogenic and carcinogenic effects. In this study, the average target cancer risks (CRs) estimated from the intake of PCBs (Table 4) ranged from 3.81×10^{-6} to 1.70×10^{-5}, 3.13×10^{-6} to 1.40×10^{-5}, and 2.28×10^{-6} to 1.02×10^{-5} for children, adolescents, and adults, respectively. The CRs were generally higher than the target risk level of 10^{-6} as prescribed by USEPA, which denotes cancer risks for individual chemicals including PCBs.[79] The highest CR (10^{-5}) was recorded at TMBO, while OGBR recorded the lowest. The results showed that PCBs 81, 101, 105, 26, 128, 157, 157, and 180 are major contributors to the CR, where each variably exceeded the safe limit (10^{-6}) across most locations. PCBs 105 and 206 equally exceeded the safe limit at TMBO across the age categories. In general, the cumulative cancer risks (∑CR) fall within the low CR classification (10^{-6}–10^{-4}) by USEPA for all age categories (Table 4). By implication, using the lowest values of CR for each age category across all locations only, the result implies that 1 person in every 262 634 children, 319 182 adolescents, and 437 915 adults may be at risk of cancer through consumption of fish from the investigated rivers (Fig. 5).

To further establish the potency of PCBs to cause carcinogenic health risks, toxic equivalency factors (TEFs) were used to estimate the toxic equivalent quantity (TEQ) (Table 5). Based on the mean PCB concentrations of both seasons, the total non-ortho PCBs (77, 81, 126, and 169) ranged from 3.19–29.27, the mono-ortho PCBs (105, 114, 118, 156, 157, and 167) ranged from 0.03–0.18, and the di-ortho PCBs (170–180) ranged from 8.05×10^{-4}–2.36×10^{-3}. The PCB-TEQ values ranged from 3.22 to 29.45 (Table 5). The TEQ range was comparatively and significantly higher than the estimated non-carcinogenic screening values (SV) of 0.026, 0.0425, and 0.0437, and carcinogenic SV of 0.0066, 0.0106, and 0.0109 for children, adolescents, and adults, respectively (included in Table 4). The SV represents the PCB threshold concentration of potential public health concern.[80] Based on the SV values, the PCB levels obtained in the current study are of serious potential health concern.

The non-ortho PCB congeners constituted over 99.2% of the ∑TEQ values across all the locations investigated in this study. One member, PCB-126, is the most toxic, representing between 55.4 and 85.8% of the total dioxin-like PCB contributions across all locations, in addition to the high contribution to the total PCBs in the study. In addition, the dioxin-like PCBs represented 40%, 47%, 41%, 48%, 52%, and 45% of the mean total PCBs recorded at ASR, ONR, OGBR, OGR, IBR, and TMBO, respectively.

It is evident from the study that PCBs continue to be present in fish, and the significance of intake *via* ingestion was also highlighted. High accumulation of PCBs in adults over several years is particularly of concern, especially in the mother's body where it is usually stored in the fat in breast milk through which breastfeeding infants are exposed.[81,82] Other studies have shown prenatal transfer of PCBs by crossing the placental barrier.[83,84] Therefore, relevant stakeholders at all levels of government are to take proactive measures that will prevent or at least minimize the release of these carcinogenic substances (PCBs) and other related carcinogens into the aquatic environment to safeguard human health.

4. Conclusions

This study found a seasonal increase in polychlorinated biphenyls (PCBs) in *Clarias gariepinus* fish from six rivers in southwestern Nigeria, with higher concentrations during the wet season. All recorded PCB levels exceeded regulatory limits, primarily featuring highly chlorinated PCBs, which made up over 86% of the total concentrations. The detection of PCB 28 and PCB 52 suggests ongoing contamination. Health risk assessments indicated no significant non-carcinogenic effects from PCB exposure through fish consumption, but low carcinogenic risks were noted, particularly for children, who are the most vulnerable. Recreational and subsistence fishers near the TMBO and IBR areas of Lagos State are especially at risk. To address these risks, public health policies should improve education on the importance of preserving aquatic ecosystems and preventing waste disposal into rivers. This study underscores the need for monitoring water and sediment quality for PCBs to reduce their environmental impact and supports a comprehensive assessment of aquatic species in southwestern Nigeria for PCBs and other persistent organic pollutants (POPs). These measures aim to enhance health risk mitigation and improve environmental safety.

Data availability

All data and related information not in the ESI† are available from the corresponding author (isaac.ololade@aaua.edu.ng).

Author contributions

Isaac Ayodele Ololade: conceptualization; data curation; formal analysis; funding acquisition; investigation; mthodology; resources; validation; project administration; visualization; writing – original draft; writing – review & editing; Abiodun Oyewumi Apata: investigation; methodology; writing – review & editing; Olubunmi Jerome Oloyede, Ifeoluwa Akindumila, and Olubunmi Jerome Oloyede: methodology, software, data curation, manuscript draft preparation. Omotayo Precious Asanga and Francis Femi Oloye: validation, software and data curation. Writing of the article and approval of the final manuscript were done by all authors.

Conflicts of interest

There are no conflicts to declare.

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