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Occurrence and risks of polychlorinated biphenyls in water, sediment, and fish of Wupa River, Nigeria

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Abstract

The occurrence and risks of polychlorinated biphenyls (PCBs) in water, sediment, and fish from the Wupa River, Nigeria, were assessed in this study. Water, sediment, and fish were collected from five locations in the Wupa River in November 2019. After extraction with dichloromethane, hexane, and acetone, the PCBs were determined using gas chromatography equipped with a quadrupole mass spectrometer. The hazard index and total cancer risk models were used for risk evaluation of the detected PCBs. The results of this study show that the ∑28 PCB concentrations in the water, sediment, and fish ranged from 0.04-11.42 ng/L, 5032-10132 ng/g, and 64-4254 ng/g, respectively. The hazard index values for children and adults were generally > 1, suggesting a potential non-carcinogenic risk for humans exposed to PCBs from the river. However, the total cancer risk values were above 1×10^{-6} and indicated that there is a carcinogenic risk for humans exposed to the PCBs from Wupa River.

Keywords: Polychlorinated biphenyls (PCBs), toxic equivalencies (TEQs), ecological risk, hazard index, total cancer risk

INTRODUCTION

Polychlorinated biphenyls (PCBs) are a group of chlorinated pollutants that have been produced and commercially accessible under different brand names since the 1920s[1]. PCBs include 209 congeners that result from the variation in the number and location of the chlorine atoms joined to the biphenyl rings[2]. The high stability, electric insulation tendencies, and low inflammation of PCBs are remarkable and have extended their applications to hydraulic fuels, plastics, refrigerants, printing ink, paints, wax extension systems, lubricants, adhesive products, *etc.*[1,3,4]. PCBs are non-ionizable, largely non-polar, and highly hydrophobic in nature. They are known suppressors of immune system function and cause neurobehavioral alterations, motor immaturity, *etc.*[2,5]. The use of PCBs is prohibited, but because they are recalcitrant, PCBs still exist in environmental matrices, biota and humans[6].

PCBs enter rivers through industrial discharge, surface runoff from non-point and point sources, atmospheric wet and dry deposition, sediment desorption, etc. In aquatic ecosystems, PCBs are adsorbed by particulate matter and precipitate in the sediment[7]. Nevertheless, PCBs can be resuspended by sediments with favorable environmental factors, and adsorbed PCBs are then released again into the water, starting another round of contamination and ultimately bioaccumulating in aquatic biota such as fish[8,9]. Since rivers are important for fishing and transportation, in addition to being a source of water for drinking, domestic use, agriculture irrigation, and recreational purposes, humans may become exposed to PCBs through consumption of agricultural foods, fish, and other seafood, drinking water, and dermal contact during transportation, recreational, and domestic water use[10]. Therefore, the monitoring and assessment of PCBs in water, sediment, and fish can indicate the status of aquatic contamination.

The Wupa River, which is about 16 km in length, is located around the Idu Industrial Layout in Abuja, Nigeria [Figure 1]. It is a branch of the Jabi River and lies within latitude 09° 01' 37.2054" N to 09° 04' 49.7215" N and longitude 07° 19' 22.7198" E to 07° 24' 45.0794" E. Next to the Wupa River is the Wupa Wastewater Treatment Plant (WWTP), Gosa dumpsite (GD), and the very busy Airport Road. The Wupa River receives effluents, waste discharges, and runoff from the WWTP, GD, and other industries sited in the Idu Industrial Layout. The Wupa River has been severely affected over the years by human activities along the river. Despite the length and intensity of these activities, there is limited information on the pollution status of the river. Thus, the objective of the present study was to assess the concentrations, sources, and risks of PCBs in the water, sediment, and fish from the Wupa River.

MATERIALS AND METHODS

Reagents

All solvents used for extraction (acetone, dichloromethane, and n-hexane) were of pesticide grade and products of Merck (Darmstadt, Germany). The PCB standard solution containing 28 PCBs (PCB8, PCB18, PCB28, PCB44, PCB52, PCB60, PCB77, PCB81, PCB101, PCB105, PCB114, PCB118, PCB123, PCB126, PCB128, PCB138, PCB153, PCB156, PCB157, PCB167, PCB169, PCB170, PCB180, PCB185, PCB189, PCB195, PCB206, and PCB209) was used for calibration (AccuStandard Inc., CT, USA). Only 28 PCBs, including the 7 indicator PCBs, 12 WHO dioxin-like PCBs, and some common PCBs, were chosen since the study focused on the occurrence and human health risks of PCBs. The PCB surrogate standard solution containing six isotopically labeled PCBs ($^{13}C_{12}$ PCB28, -52, -118, -153, -180, and -209) was a product of Cambridge Isotope Laboratories Inc. (MA, USA). Alumina, copper powder, anhydrous sodium sulfate, and silica gel were of analytical grade and obtained from BDH Chemicals (Poole, UK).

Collection of samples

Samples were collected in November 2019. Five water and sediment samples were collected from five different locations (SL1-SL5) along the Wupa River [Figure 1]. The grab sampling technique was used to collect water and sediment samples. At each location, three water and sediment samples were collected and combined to give a homogenous sample. Five different fish species were obtained from a local fisherman

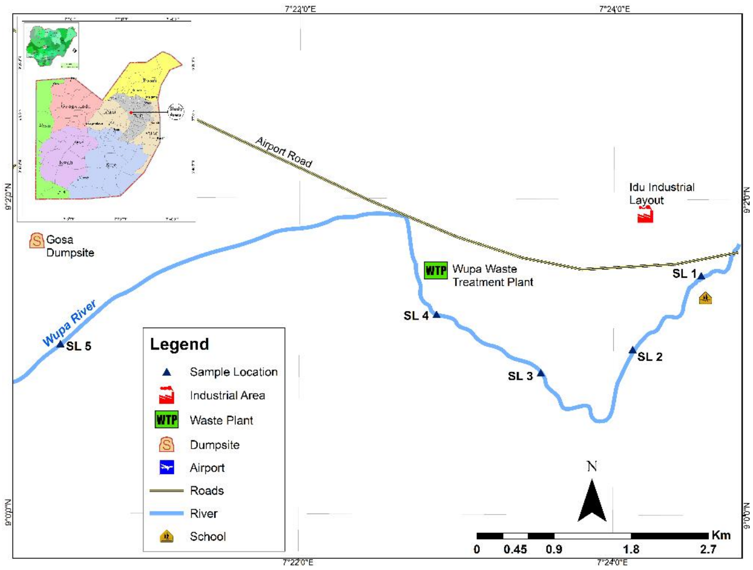

Figure 1. Map of study area.

during sampling. comprising *Cyprinus carpio* (CC), *Ethmalosa fimbriata* (EF), *Heterobranchus bidorsalis* (HB*)*, *Clarias anguillaris* (CA), and *Oreochromis niloticus* (ON). All samples were placed in amber containers, kept in an ice chest, and transported to the laboratory for analysis.

Extraction and cleanup of PCBs in water

The USEPA Method 3510 was used in the extraction of PCBs in water samples. About 100 mL of water sample was measured into a separating funnel containing 100 mL of dichloromethane (DCM). The mixture was extracted for 30 min and the extract was collected into a flask. The extraction process was repeated with another 100 mL of DCM and the extract was collected into the same flask. The combined extract was reduced to approximately 2 mL with a rotary evaporator (LabTech EV311H Rotary evaporator). The concentrated extract was cleaned in a column containing alumina-silica gel packed bottom to top with 4 g of neutral silica gel (5% deactivated), 2 g of neutral alumina (6% deactivated), and 5 g of anhydrous Na_2SO4. The PCBs were subsequently eluted with 50 mL of hexane from the column, concentrated to 2 mL, and stored in a vial prior to chemical analysis.

Extraction and cleanup of PCBs in sediment and fish

The USEPA Method 3540 C was used to extract the PCBs from the sediment and fish. A mass of 10.0 g of the sediment/fish was spiked with a mixed standard solution of isotopically labeled PCB congeners (200 ng g^{-1}) and Soxhlet extracted with 120 mL of an acetone/dichloromethane/n-hexane mixture (1:1:1 v/v) for 18 h in a water bath at 65 °C. Then, 3 g of anhydrous Na_2SO_4 and 1 g of activated copper were added to eliminate the possible water and sulfur, respectively. The extract was reduced to about 2 mL with a rotary evaporator and cleaned in a multilayer alumina-silica gel column packed bottom to top with 4 g of neutral silica gel (5% deactivated), 2 g of neutral alumina (6% deactivated), and 5 g of anhydrous Na_2SO_4. The PCBs were eluted with 40 mL mixture of hexane and dichloromethane (3:1 v/v). The eluate was evaporated to 2 mL and stored in a vial ready for chemical analysis.

Quantification of PCBs in samples

The concentrations of the 28 PCBs in the sample extracts were determined using an Agilent 7890A gas chromatograph interfaced with a 5876C mass selective detector (MSD) (Agilent Technologies Inc., Palo

Alto, CA, USA). The separation column was a HP5 (30 m × 320 µm × 0.25 µm). The mobile phase was high-purity helium gas at a constant flow velocity of 1.2 mL/min. The initial temperature of the column was fixed at 85 °C, held for 1 min, stepped up to 200 °C at 35 °C/min, and then increased from 200 to 300 °C at 10 °C/min. The injector temperature and that of the transfer line were maintained at 250 °C, while the injection volume was 1 µL. The mass spectrometer was operated at an electron impact energy of 70 eV and data acquisition was performed by selected ion monitoring (SIM). The PCB congeners were identified by matching the retention times of the PCBs in these samples with those obtained from authentic PCB standards.

Quality control and statistical analysis

Quality control and assurance were achieved with procedural blanks, recoveries of the $^{13}C_{12}$-PCBs, and matrix spike methods. Procedural blanks (n = 3) were analyzed following all the analysis steps but omitting the samples. PCBs were not detected in the procedural blanks. For the matrix spiked recovery method, a known standard of the PCBs was added to chosen fresh aliquots of samples (n = 3) that had already been analyzed and followed all the analytical steps. The percent recoveries of PCBs from the spiked matrices ranged from 95.3%-107%, 98.6%-106%, and 91.2%-106% for water, sediment, and fish, respectively. The surrogate PCB recoveries were 92.5%-99.5%, 90.9%-98.1%, and 89.7%-94.2% for water, sediment, and fish, respectively. The quantification of the PCBs was achieved by using an external calibration method consisting of five-point calibration lines obtained as a plot of the congener peak areas versus the standard concentrations. The regression coefficients (r^2) for the calibration lines ranged from 0.9992 to 0.9999. The limits of detection and quantification (3 and 10 times the noise levels of the baseline, respectively) for the PCBs were 0.01-0.4 and 0.03-1.2 ng L^{-1}, respectively. The precision of the method for replicate analyses was less than 8% relative standard deviation (RSD). The LODs, LOQs, RSD, r^2, and percentage recoveries of individual PCB congeners are shown in Supplementary Table 1.

One-way analysis of variance (ANOVA) was employed for the determination of significant variation (P < 0.05) in the PCB concentrations at the various sampling locations for each matrix. SPSS version 19.0 (SPSS Inc., Cary, NC) was used for statistical analysis.

Assessment of ecological risk from PCBs in Water, Sediment and Fish

The ecological risk of the dl- PCBs in the three matrices from the Wupa River was obtained using their toxic equivalencies (TEQs). The TEQs was computed with Equation (1)[11]:

$$TEQ = \sum TEF \times C \tag{1}$$

where C and TEF are the *dl*-PCB concentrations and toxic equivalence factors, respectively. The TEF values used are shown in Supplementary Table 2.

Assessment of human health risks

Assessment of the human health risk of PCBs in water, sediment, and fish from the Wupa River was done in terms of hazard index (HI) and total cancer risk (TCR), respectively, with the equations below[12,13].

For non-cancer risk,

$$\mathrm{HI} = \sum HQ = HQ_{ing} + HQ_{inh} + \mathrm{HQ}_{\mathrm{dermal}} \tag{2}$$

$$\mathrm{HQ} = \frac{CDInc}{RfD} \tag{3}$$

$$\mathrm{CDI}_{\text{ing-nc}} = \frac{C \times \mathrm{IngR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}_{nc}} \times 10^{-6} \tag{4}$$

$$\mathrm{CDI}_{\text{inh-nc}} = \frac{C \times \mathrm{InhR} \times \mathrm{EF} \times \mathrm{ET} \times \mathrm{ED}}{\mathrm{PEF} \times 24 \times \mathrm{AT}_{nc}} \tag{5}$$

$$\mathrm{CDI}_{\text{dermal-nc}} = \frac{C \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}_{nc}} \times 10^{-6} \tag{6}$$

For cancer risk,

$$\mathrm{Risk}_{\mathrm{ing}} = \frac{C \times IngR \times EF \times ED \times CF \times SFO}{BW \times ATca} \tag{7}$$

$$\mathrm{Risk}_{\mathrm{inh}} = \frac{C \times Inh \times EF \times ED \times IUR}{PEF \times ATca} \tag{8}$$

$$\mathrm{Risk}_{\mathrm{derm}} = \frac{C \times SA \times AF \times ABS \times EF \times ED \times CF \times SFO \times GIABS}{BW \times ATca} \tag{9}$$

The meaning and values of each term and variable are shown in Supplementary Tables 2 and 3 respectively. For water, the health risk was evaluated using only two routes of exposure (ingestion and dermal contact). For sediment, three routes of exposure (ingestion, inhalation, and dermal contact) were used. For fish, only the ingestion route was used. Generally, HI values greater than 1 indicate the presence of non-carcinogenic risk of PCB exposure and vice versa, while total cancer risk values greater than 1.0×10^{-6} indicate that there is a carcinogenic risk from PCB exposure and vice versa[14].

RESULTS AND DISCUSSION

PCB concentrations in the Wupa River

The results of the 28 PCB congeners determined in the water, sediment, and fish from the Wupa River are shown in Table 1 and Figures 2-4. Analysis of variance (ANOVA) showed that the concentrations of PCBs in each of the three matrices from the Wupa River varied significantly ($P < 0.05$) among the sampling locations. The PCB concentrations in the Wupa River may be a result of the human activities and industrial processes around the river, considering factors such as total organic matter, velocity of water flow, and transportation characteristics of PCBs[3,4,15].

Water

The total PCB (∑28 PCB) concentrations in the water samples of Wupa River varied between 3.13 and 11.42 ng/L. The maximum ∑28 PCBs was observed in sample W3, whereas sample W4 had the lowest concentration. The ∑28 PCB concentrations obtained in water samples of Wupa River may be a result of industrial releases from the industries in the Idu Industrial Layout of Abuja where the river is located. The water samples of the Wupa-Idu River have higher proportions of the more soluble, less chlorinated PCBs [Figure 2]. Similar observations have also been reported in the literature[16-18]. The occurrence pattern of the PCB homologs in the water samples of the Wupa River is shown in Figure 5. On average, the occurrence pattern was as follows: penta-PCBs > di-PCBs > hexa-PCBs > tetra-PCBs > tri-PCBs > hepta-PCBs > octa-PCBs. Nona- and deca-PCBs were not detected. The concentrations of total PCBs in water samples of Wupa

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Table 1. PCB concentrations in water (ng/L), sediment (ng/g), and fish (ng/g) from the Wupa River

	Water					Sediment					Fish				
	W1	**W2**	**W3**	**W4**	**W5**	**S1**	**S2**	**S3**	**S4**	**S5**	**CC**	**EF**	**HB**	**CA**	**ON**
PCB8	0.26	0.77	2.96	0.70	1.97	298	776	200	900	1200	550	152	328	92	20
PCB18	0.21	0.72	1.59	0.15	0.47	266	332	264	810	362	564	350	194	198	44
PCB28	ND	ND	ND	ND	ND	136	382	184	206	528	296	262	ND	ND	ND
PCB44	ND	ND	ND	ND	ND	274	744	116	246	704	ND	238	ND	ND	ND
PCB52	ND	ND	ND	ND	ND	268	246	144	258	526	180	54	ND	ND	ND
PCB66	0.85	0.71	0.6	0.40	ND	ND	478	116	208	288	712	138	ND	ND	ND
PCB77	0.56	0.90	0.43	0.35	ND	236	168	190	406	136	38	140	ND	ND	ND
PCB81	ND	0.92	ND	ND	ND	474	126	110	48	210	70	50	ND	ND	ND
PCB101	0.11	ND	ND	ND	2.62	244	276	324	844	452	62	426	188	ND	ND
PCB105	ND	0.18	0.35	ND	ND	1432	ND	90	ND	522	ND	104	ND	ND	ND
PCB114	ND	0.99	ND	0.67	ND	528	140	278	234	296	ND	ND	ND	ND	ND
PCB118	ND	0.56	ND	0.86	ND	188	188	220	306	272	ND	ND	ND	ND	ND
PCB123	0.16	ND	1.47	ND	0.75	398	266	232	220	302	452	346	244	ND	ND
PCB126	0.80	ND	0.13	ND	0.12	134	212	120	714	156	346	266	114	ND	ND
PCB128	0.06	ND	0.10	ND	0.40	218	308	230	468	132	394	364	134	ND	ND
PCB138	ND	ND	1.3	ND	ND	54	270	236	174	280	ND	ND	ND	ND	ND
PCB153	0.93	ND	0.14	ND	0.50	98	384	104	400	148	410	380	104	92	ND
PCB156	ND	ND	0.44	ND	ND	68	330	64	180	130	268	ND	ND	ND	ND
PCB157	ND	ND	ND	ND	ND	108	254	60	760	114	ND	ND	ND	ND	ND
PCB167	ND	ND	ND	ND	ND	102	188	28	310	66	16	ND	ND	ND	ND
PCB169	ND	ND	1.52	ND	ND	84	662	64	ND	188	42	ND	ND	ND	ND
PCB170	ND	ND	0.39	ND	ND	154	622	60	78	254	40	ND	ND	ND	ND
PCB180	ND	ND	ND	ND	0.96	110	246	442	874	526	78	500	ND	ND	ND
PCB187	ND	ND	ND	ND	ND	126	110	ND	258	154	48	ND	ND	ND	ND
PCB189	ND	ND	ND	ND	ND	212	134	74	ND	290	ND	ND	ND	ND	ND
PCB195	ND	0.91	ND	ND	ND	178	274	350	ND	228	ND	484	ND	20	ND
PCB206	ND	ND	ND	ND	ND	308	196	126	298	312	286	ND	286	ND	ND
PCB209	ND	ND	ND	ND	ND	422	276	606	932	ND	248	ND	248	ND	ND
TOTAL	**3.94**	**6.66**	**11.4**	**3.13**	**7.79**	**7118**	**8588**	**5032**	**10132**	**8776**	**5100**	**4254**	**1840**	**402**	**64**
Di-PCB	0.26	0.77	2.96	0.70	1.97	298	776	200	900	1200	550	152	328	92	20
Tri-PCBs	0.21	0.72	1.59	0.15	0.47	402	714	448	1016	890	860	612	194	198	44

Tetra-PCBs	1.41	2.53	1.03	0.75	0.00	1250	1762	676	1166	1864	1000	620	0.0	0.0	0.0
Penta-PCBs	1.07	1.73	1.95	1.53	3.49	2920	1082	1264	2318	2000	860	1142	546	0.0	0.0
Hexa-PCBs	0.99	0.00	3.50	0.00	0.90	732	2396	786	2292	1058	1130	744	238	92.0	0.0
Hepta-PCBs	0.00	0.00	0.39	0.00	0.96	602	1112	576	1210	1224	166	500	0	0	0
Octa-PCBs	0.00	0.91	0.00	0.00	0.00	178	274	350	0	228	0	484	0	20	0
Nona-PCBs	0.00	0.00	0.00	0.00	0.00	308	196	126	298	312	286	0	286	0	0
Deca-PCBs	0.00	0.00	0.00	0.00	0.00	422	276	606	932	0	248	0	248	0	0
LC-PCBs	2.95	5.75	7.53	3.13	5.93	4876	4334	2588	5400	5954	3270	2526	1068	290	64
HC-PCBs	0.99	0.91	3.89	0.00	1.86	2242	4254	2444	4732	2822	1830	1728	772	112	0

CC: Cyprinus carpio; CA: clarias anguillaris; EF: ethmalosa fimbriata; HB: heterobranchus bidorsalis; ON: oreochromis niloticus; PCB: polychlorinated biphenyl.

River were below the USEPA permissible limit of 500 ng/L PCBs in drinking water[19]. The National Oceanic and Atmospheric Administration (NOAA) set a maximum concentration (CMC) of 2000 ng/L and continuous concentration (CCC) of 14 ng/L for PCBs in water. The total PCB concentrations in water samples of Wupa River were below the CMC and CCC values. The PCB concentrations in water of the Wupa River were comparable to those reported for other rivers in the literature [Table 2].

Sediment

The ∑28 PCB concentrations in the sediment of the Wupa River ranged from 5032 ng/g for S3 to 10,132 ng/g for S4. All 28 PCB congeners except PCB66 were detected in sample S1. Only PCB105 and PCB187 were below their detection limits in samples S2 and S3, respectively. PCB105, PCB169, PCB189, and PCB195 were under their detection limits in sample S4, whereas all 28 PCBs were found in sample S5. The PCB congener distribution in the sediments of the Wupa River showed an even spread across chlorination levels, which indicated the presence of several different aroclors. The higher concentration of PCB8 in samples S2, S4, and S5 may be due to dechlorination or possibly an inadvertent PCB source. PCB209 was not only detected in all sediment samples but also in high proportions; in some samples (S3 and S4), it was the dominant congener. This is highly indicative of an incidental PCB source. On average, the occurrence pattern followed the order: penta-PCBs > hexa-PCBs > tetra-PCBs > hepta-PCBs > tri-PCBs > di-PCBs > deca-PCBs > nona-PCBs > octa-PCBs [Figure 5]. The lower chlorinated (LC) PCBs (di-PCBs to penta-PCBs) were the dominant PCBs in the sediment. The domination of the LC-PCBs in the sediments of the Wupa River may result from reductive dechlorination of higher chlorinated PCBs in the absence of free oxygen[3]. The ∑28 PCB concentrations in the sediment of the Wupa River were above the Dutch action value and Australia and New Zealand Ecological Investigation Level of 1000 ng/g[41,42], the Canadian Soil Quality Guideline value of 1300 ng/g[43], and the US EPA health-based screening level for total PCBs of 200 ng/g[44]. The ∑28 PCB concentrations obtained in the sediment of the Wupa River are compared with those reported elsewhere in Nigeria and other countries in Table 2. The PCB concentrations obtained in sediments of the Wupa River were comparable to the range of 226-31,900 ng/g reported for Escravos River[4] but higher than those reported for the Niger, Ase, Forcados, Nicholas, Ona, New Calabar, Ethiope, and Benin Rivers in Nigeria. They were also higher than the PCB concentrations reported for sediments from rivers in other countries. The PCB concentrations in the sediment of the Wupa River were in the same range of 3400-50200 ng/g

Table 2. Comparison of PCBs in water and sediment of the Wupa River with those in other rivers

Matrices	River System	Concentrations	Reference
Water (ng/L)	Wupa-Idu River, Nigeria	3.13-11.4	This Study
	River Niger, Nigeria	456-1139	[19]
	River Ethiope	0.0015-0.015	[20]
	Benin River	0.00003-0.00293	[20]
	River Nile, Egypt	0.014-0.02	[21]
	Pearl River Estuary, China	0.02-14.8	[22]
	Yangtze River, China	3.77-61.79	[22]
	Tonghui River of Beijing, China	31.58-344.9	[22]
	Minjiang River Estuary, China	204-2473	[22]
	Houston Ship Channel, USA	0.49-12.5	[23]
	Mississippi River, USA	22.2-163	[23]
	Delaware River, USA	0.42-1.65	[23]
	Hudson River, USA	<9.3-164.3	[23]
	Johannesburg River, South Africa	0.021-0.121	[23]
	Ebro River, Spain	43.2-108	[23]
	Bay of Bengal Coast, Bangladesh	32.17-199.4	[16]
Sediment (ng/g)	Wupa-Idu River, Nigeria	5032-10132	This Study
	Nigeria		
	Escravos River Basin, Nigeria	226-31900	[4]
	River Niger	13.5-277	[3]
	Ase River	ND-1633	[3]
	Forcados River	1.9-78.4	[3]
	Ogun River, Nigeria	323-2003	[24]
	Ona river, Nigeria	589-1354	[24]
	New Calabar River, Nigeria	210-2160	[25]
	Forcados River, Nigeria	2.7-202.3	[26]
	River Niger and Nicholas River	741-2964	[27]
	Ethiope River	0.73-6.7	[20]
	Benin River	0.35-15.15	[20]
	Africa		
	Umgeni River, South Africa	103-430	[28]
	Pangani River and its tributaries, Tanzania	0.36-11	[29]
	Lake Qarum, Egypt	1.48-137.2	[30]
	Lake Maryut, Egypt	3.06-388	[30]
	Lake Manzala, Egypt	2.53-76.37	[30]
	Monaslir Bay, Tunisia	1.1-9.3	[31]
	Congo River basin, Congo	Nd-1.4	[32]
	Other countries of the World		
	Ankara creek, Turkey	3.7-743.3	[33]
	Haihe River and Estuary, China	0.177-253	[34]
	Cienfuegos Bay, Cuba	2.50-15.49	[35]
	Chenab River, Pakistan	9.33-144.23	[36]
	Lianjiang River, China	4.70-743	[37]
	Lake Michigan, USA	53-35,000	[38]
	Northwest Persian Gulf, Iran	3400-50200	[39]
	Belford Harbor, Massachussets, USA	2800-109000	[40]

PCB: Polychlorinated biphenyl.

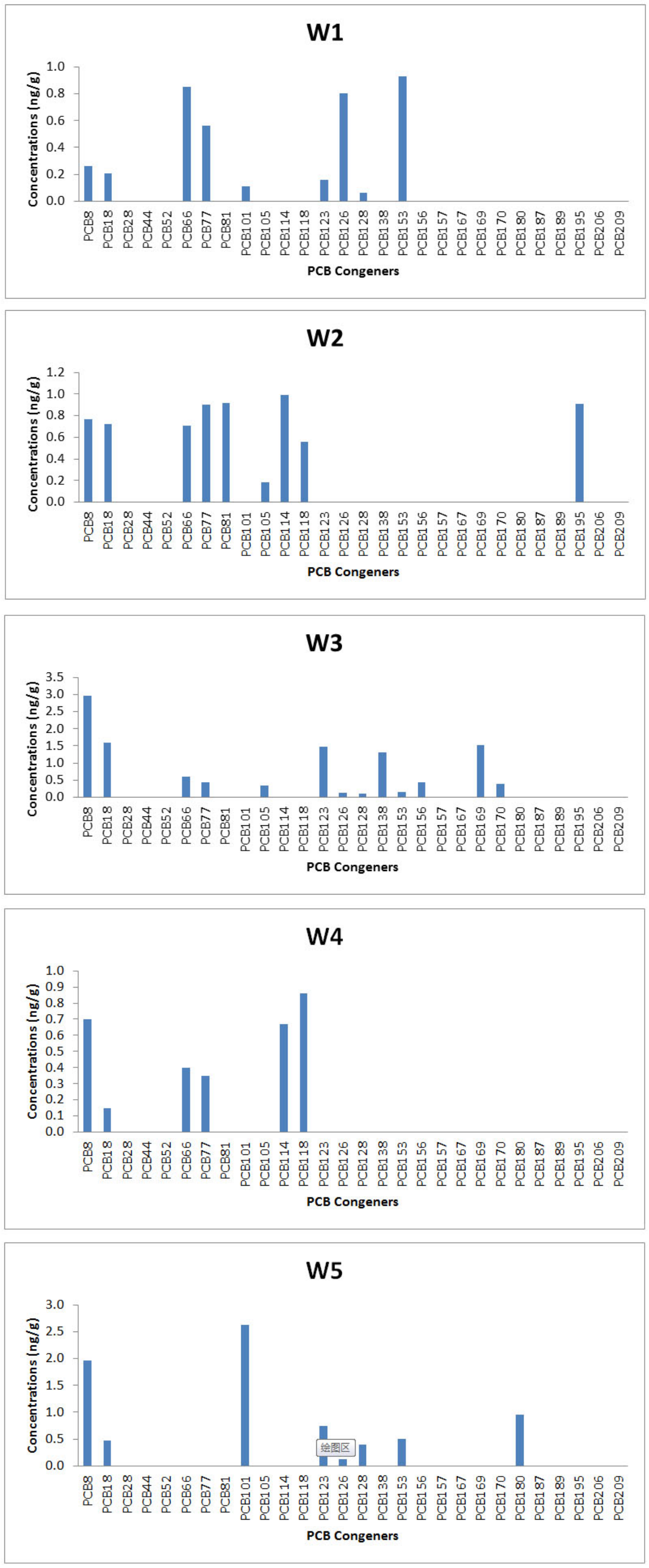

Figure 2. PCB congener profiles in water samples. PCB: Polychlorinated biphenyl.

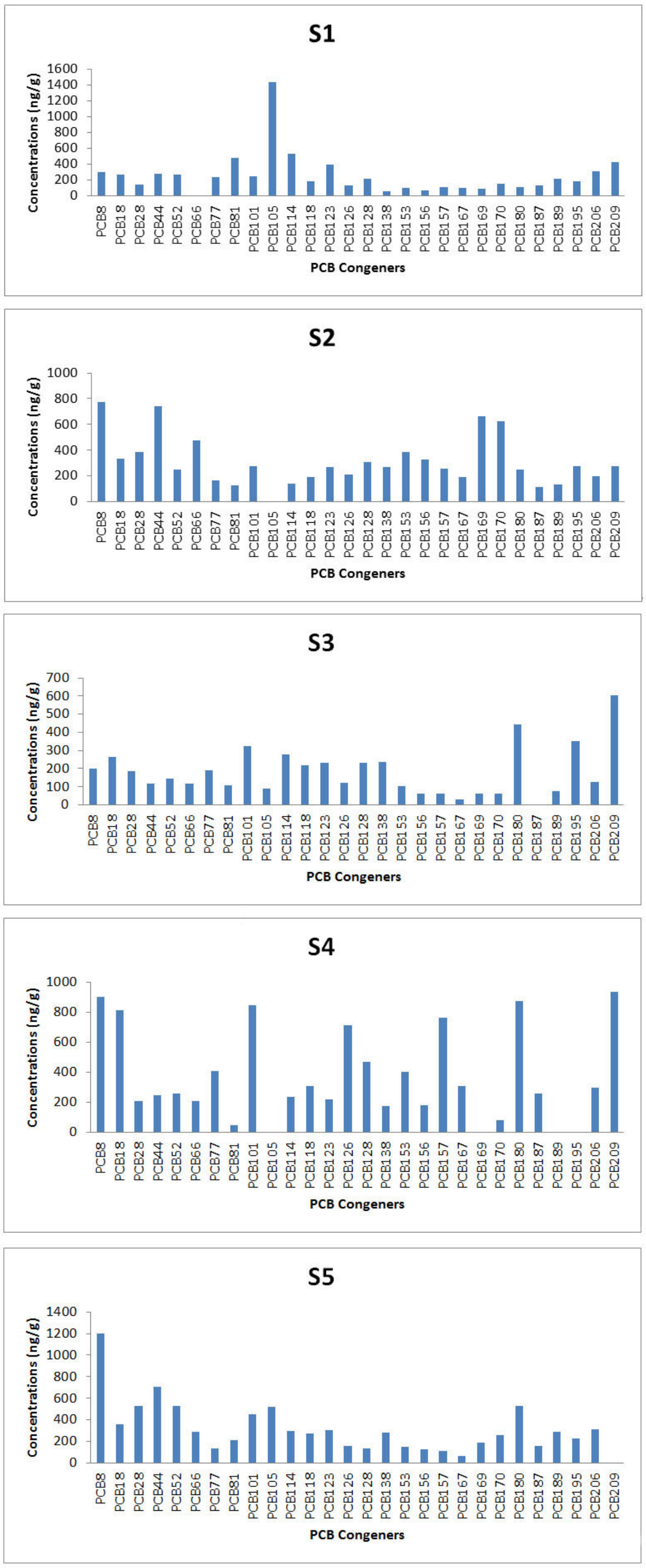

Figure 3. PCB congener profiles in sediment samples. PCB: Polychlorinated biphenyl.

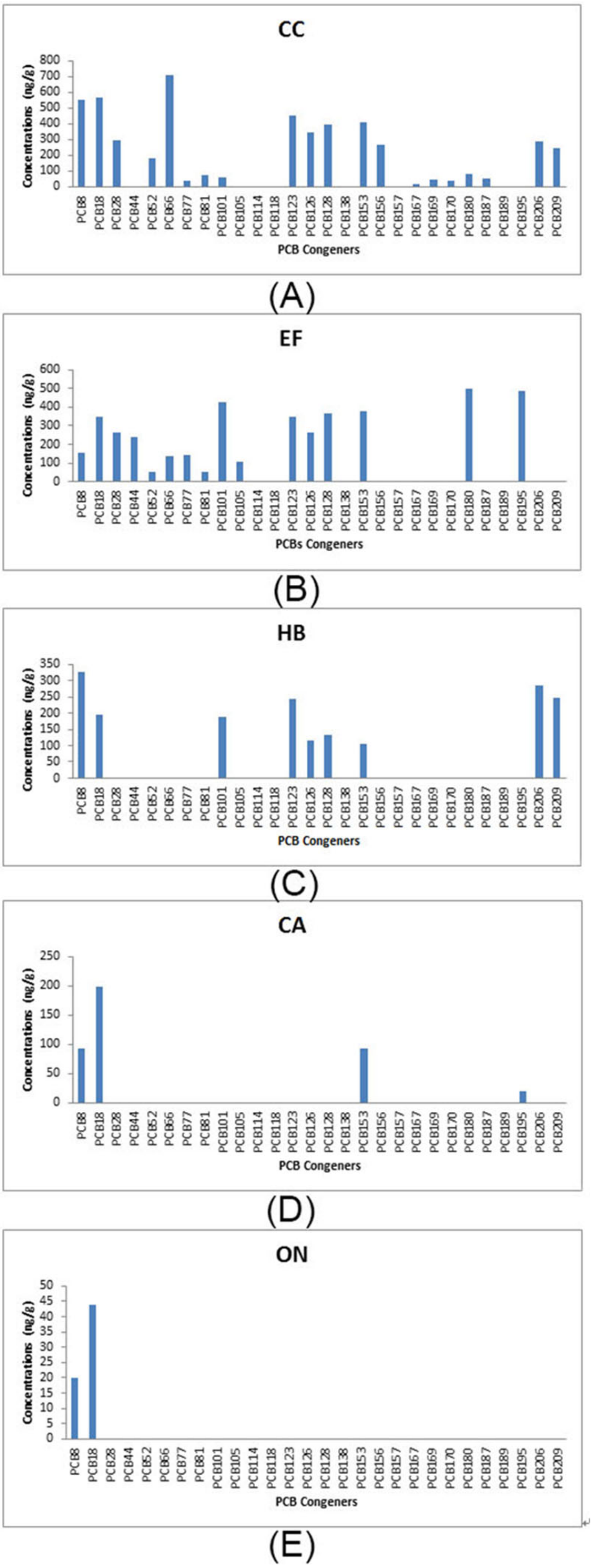

Figure 4. PCB congener profiles in fish samples: (A) cyprinus carpio; (B) ethmalosa fimbriata; (C) heterobranchus bidorsalis; (D) clarias anguillaris; and (E) oreochromis niloticus. CC: Cyprinus carpio; CA: clarias anguillaris; EF: ethmalosa fimbriata; ON: oreochromis niloticus; PCB: polychlorinated biphenyl.

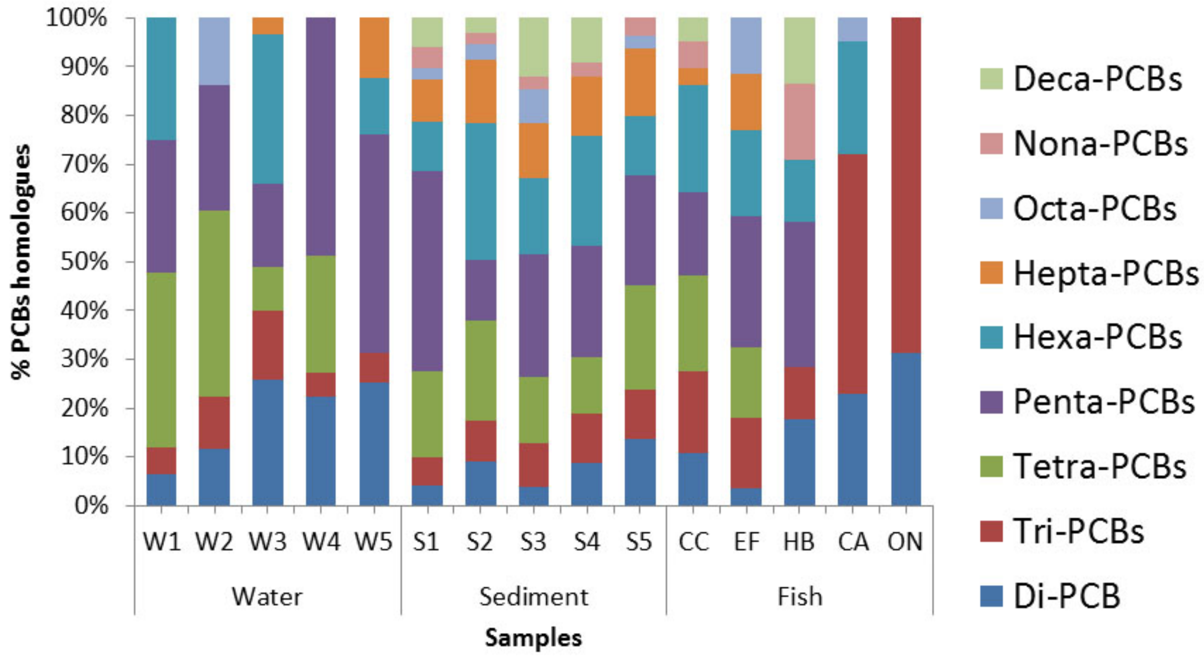

Figure 5. Occurrence profiles of PCBs in the water, sediment, and fish from the Wupa River. CC: Cyprinus carpio; CA: clarias anguillaris; EF: ethmalosa fimbriata; ON: oreochromis niloticus; PCB: polychlorinated biphenyl.

reported by Zahed *et al*. for the sediment of the Persian Gulf, Iran, 2800-109,000 ng/g reported for the sediment of Belford Harbor, MA, USA, and 53-35,000 ng/g reported for the sediment of Lake Michigan, USA[39,40].

Fish

The total PCB (∑28 PCBs) concentrations in the fish samples from the Wupa River varied between 64.0 and 5100 ng/g. The highest ∑28 PCB concentration was obtained in *Cyprinus carpio,* whereas the lowest concentration was obtained in *Oreochromis niloticus*. In *Cyprinus carpio*, PCB congeners 105, 114, 118, 157, 189, and 195 were below their detection limit. Similarly, in *Ethmalosa fimbriata*, PCB congeners 114, 118, 156, 167, 169, 170, 187, 189, 206, and 209 were under their detection limit. However, in *Heterobranchus bidorsalis* PCB congeners 8, 18, 101, 123, 126, 128, 153, 206, and 209 were detected. In *Clarias anguillaris*, only PCB congeners 8, 18, 153, and 195 were detected, while in *Oreochromis niloticus*, only PCB8 and PCB18 were detected.

The occurrence pattern of the PCBs was as follows: penta-PCBs > hexa-PCBs > tri-PCBs >tetra-PCBs > di-PCBs > hepta-PCBs > nona-PCBs > octa-PCBs >deca-PCBs [Figure 5]. The permissible limit of PCBs in fish set by the United States Food and Drug Administration (USFDA) and Swedish Food Regulation (SFR) is 2000 ng/g. The ∑28 PCB concentrations in *Cyprinus carpio* and *Ethmalosa fimbriata* were above the USFDA and SFR permissible limits. The ∑28 PCB concentrations obtained in fish from the Wupa River were comparable to the concentrations range of 20-6000 ng/g reported for fish from the Michigan River[45], 4300-10,000 ng/g reported for some fish species from the Great Lakes[46], 50-3500 ng/g reported for different Luxembourg River fish[47], 333-2531 ng/g reported for fish from Eleyele Reservoir, Southwestern Nigeria[22], 560-2940 ng/g in fish from Lagos Lagoon[48], and 290-110,000 ng/g in fish from Galveston Bay, TX[49]. The total PCB concentrations obtained in fish from the Wupa River in this study were higher than the undetected range of 94 ng/g reported for freshwater fish from Luxembourg[50], 1.50-280 ng/g for fish from the Belgian North and Western Scheldt Estuary[51], 0.24-1.4 ng/g in marine fish from Zhoushan City, China[52], 4.7-11.4 ng/g in different marine fish species in Nanjing city, China, 0.44-86 ng/g in marine fish from tsunami-stricken areas of Japan[53], and 7.20-90.19 ng/g in marine fish from the Persian Gulf[54].

Toxicity of dl-PCBs in Water, Sediment, and Fish

The computed dl-PCBs TEQs for the three matrices from the Wupa River are presented in Table 3. The TEQs ranged from 8.09.0 × 10^{-5} to 8.01 × 10^{-2} ng TEQ_{2005} g^{-1} in 80% of the water samples, from 16.2 to 71.5 ng TEQ g^{-1} in sediment, and from 11.4 to 35.9 ng $TEQ_{2005}g^{-1}$ in fish. The TEQ values obtained for the sediment of the Wupa River were more than the 21.5 pg TEQ g^{-1} limit stipulated by the Canadian Council of Ministers of the Environment (CCME) [43]. This implies that there are potential toxic effects as a result of PCB exposure in the Wupa River. With the exception of *Clarias anguillaris* and *Oreochromis niloticus* samples, the TEQs recorded in these fish samples from the Wupa River were greater than the upper limit of 6.5 pg TEQ_{2005} g^{-1} stipulated by the European Food Safety Authority (EFSA)[55] for dl-PCBs in fish, indicating that it is dangerous to consume these fish. PCB126 was the major donor to the TEQs obtained for these matrices from the Wupa River.

Human health risks

The computed non-cancer and cancer risks of PCBs in the three matrices from the Wupa River are displayed in Supplementary Tables 4-6. For water and sediment, the HQIng was greater than HQDerm, and HQinh was the lowest. The HI values of PCBs in the Wupa River for children and adults ranged from 14.8-14,512 and 4.43-4344, respectively for water, 3.07 × 10^{5}-1.82 × 10^{6} and 4.30 × 10^{4}-2.55 × 10^{5}, respectively for sediment and 2.17 × 10^{4}-6.58 × 10^{4} and 5.42 × 10^{3}-1.65 × 10^{4}, respectively for fish. The HI values for the three matrices were > 1, signifying the existence of non-cancer risk for individuals exposed to PCBs in the water, sediment, and fish from the Wupa River. However, the HI values for fish samples of the Wupa River were < 1, indicating that there is no adverse non-cancer risk for humans eating fish from the Wupa River.

The risk levels from PCB exposure in the water and sediments followed the same trend as the HQ. The TCR values of PCBs in the Wupa River for children and adults ranged from 1.33 × 10^{-3}-1.32 and 2.09 × 10^{-4}-2.07 × 10^{-1}, respectively, for water, 3.24 × 10^{1}-1.65 × 10^{2} and 2.50-1.27 × 10^{1}, respectively, for sediment and 1.97-6.21 and 2.71 × 10^{-1}-8.54 × 10^{-1}, respectively for fish. The TCR values for the three matrices from the Wupa River were above the 1 × 10^{-6} risk level, indicating the presence of potential cancer risk from PCB exposure in the Wupa River. However, the small sample size from which the risk data were derived is somewhat a limitation to this study and is well acknowledged.

CONCLUSION

The occurrence and risks of PCBs in water, sediment, and fish from the Wupa River, Nigeria, were assessed in this study. The concentrations of total PCBs in all the water samples from the Wupa River were below the USEPA permissible limit of PCBs in drinking water, while the concentrations of total PCBs in 60% of the fish samples were less than the USFDA and SFR permissible limits of PCBs in fish. However, the PCB concentrations in the sediments were above the Dutch action value, Australia and New Zealand Ecological Investigation Level, Canadian Soil Quality Guideline value, and the USEPA health-based screening level. PCB209 was the dominant PCB in some sediment profiles and is associated with the inadvertent PCB production. The risk assessment indicated that there are possible ecological and human health risks to biota and humans exposed to PCBs in the Wupa River.

Table 3. Toxic equivalence (ngTEQ$_{2005}$ g^{-1}) of PCBs in water, sediment, and fish from the Wupa River

Marices	Samples	PCB77	PCB81	PCB105	PCB114	PCB118	PCB123	PCB126	PCB156	PCB157	PCB167	PCB169	PCB189	TTEQ
Water	W1	5.60E-05	0	0	0	0	4.80E-06	8.00E-02	1.80E-06	0	0	0	0	**8.01E-02**
	W2	9.00E-05	2.76E-04	5.40E-06	2.97E-05	1.68E-05	0	0	0	0	0	0	0	**4.18E-04**
	W3	4.30E-05	0	1.05E-05	0	0	4.41E-05	1.30E-02	3.00E-06	1.32E-05	0	4.56E-02	0	**5.87E-02**
	W4	3.50E-05	0	0	2.01E-05	2.58E-05	0	0	0	0	0	0	0	**8.09E-05**
	W5	0	0	0	0	0	2.25E-05	1.20E-02	1.20E-05	0	0	0	0	**1.20E-02**
Sediment	S1	0.024	0.142	0.043	0.016	0.006	0.012	13.4	0.002	0.003	0.003	2.52	0.006	**16.2**
	S2	0.017	0.038	0	0.004	0.006	0.008	21.2	0.01	0.008	0.006	19.9	0.004	**41.2**
	S3	0.019	0.033	0.003	0.008	0.007	0.007	12	0.002	0.002	0.001	1.92	0.002	**14.0**
	S4	0.041	0.014	0	0.007	0.009	0.007	71.4	0.005	0.023	0.009	0	0	**71.5**
	S5	0.014	0.063	0.016	0.009	0.008	0.009	15.6	0.004	0.003	0.002	5.64	0.009	**21.4**
Fish	CC	3.80E-03	2.10E-02	0	0	0	1.36E-02	3.46E+01	8.04E-03	0	4.80E-04	1.26	0	**35.9**
	EF	1.40E-02	1.50E-02	3.12E-03	0	0	1.04E-02	2.66E+01	0	0	0	0	0	**26.6**
	HB	0	0	0	0	0	7.32E-03	1.14E+01	0	0	0	0	0	**11.4**
	CA	0	0	0	0	0	0	0	0	0	0	0	0	**0.0**
	ON	0	0	0	0	0	0	0	0	0	0	0	0	**0.0**

PCB: Polychlorinated biphenyl.

DECLARATIONS

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Authors' contributions

Performed data acquisition, as well as provided administrative, technical, and material support: Okoh A, Tesi GO

Made substantial contributions to conception and design of the study, editing and supervision: Dauda MS, Aliyu HD

Availability of data and materials

The data is available in the report. Additional data and information can be made available at request from individuals interested.

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Not applicable.

Consent for publication
Not applicable.

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