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Polychlorinated biphenyls (PCBs) in sediments and fish from dredged tributaries and creeks of river Ethiope, South-South, Nigeria: sources, risk assessment and bioaccumulation

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Abstract

Polychlorinated biphenyls (PCBs) are persistent organic pollutants that are ubiquitous in nature. In this study, the levels of PCBs were evaluated in sediments and fish samples obtained from dredged tributaries and creeks of River Ethiope. The work also assessed the possible relationship between the parameters and risks posed by polychlorinated biphenyls via several pollution indices. The mean concentrations of $\sum$ PCBs on the sediments spanned from 645 – 3,977 μg/kg (wet season) to 252 – 1,219 μg/kg (dry season) dry weight. The concentrations of PCBs in fishes were 1,688 μg/kg (wet season) and 557 μg/kg (dry season). Higher bioaccumulation factors were observed with lower molecular weight PCBs than the higher ones (9 – Hexa – PCB, 8- Tetra- PCB). The results of the ecological risk ($160 \leq$ Eri < 320), and human health risk ($\geq 10^{-4}$ to $10^{-3} - \leq 10^{-6}$), showed moderately to very high contamination and also moderately to very high cancer risk for children and adults. The strong positive correlation between PCB-114 and PCB-77, PCB-81, PCB-105 (r_2 = 1.00, 0.99 & 1.00, p < 0.01) and the risks assessment values which ranged from 6.10×10^{-3} to 1.47×10^{-2} for children and 6.30×10^{-4} to 1.11×10^{-3} for adults (wet season), 1.04×10^{-3} to 2.99×10^{-2} for children and 7.80×10^{-5} to 5.61×10^{-1} for adults (dry season), showed rarely to adversely high potential ecological risk, biological effect and human health risk across the environment. The data show that higher levels of PCBs were observed in sediments and fish samples when compared with available standards. Considering the ILCR, hazard index, toxic equivalent, the sediments and fish obtained from these sites would be considered risky for humans. Dredging activities are majorly responsible for the high levels of PCBs across the sites. These have contributed significantly to the environmental status of the studied area.

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1. Introduction

Persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) are very ubiquitous in nature as a result of their volatility, increase in sources of production and migra-

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tion mechanisms [1, 2]. About three decades ago, there was a drastic increase in the sources of PCBs [2, 3]. The contamination level of PCBs in the aquatic environment is alarming and it has become a worldwide problem [4]. Despite the disastrous effect of PCBs contamination in the ecosystem across the world, there has been none or little investigation of PCBs in dredged tributaries and creeks of River Ethiope. Across the world, the use of PCBs has been banned in many countries, but its presence is still on the increase in the environment due to indiscriminate dumping of e- wastes (electronics wastes), burning of materials containing PCBs and accidental spillage of oil, which migrate into the water bodies via runoffs from rains into the dredged river. In addition to their persistent nature, they are bio accumulative across the food chain which calls for the need for constant monitoring of the environment.

Dredging is a process of evacuating soil, sediments across water- ways, plant and bank of river and also the deposition of spoil at the river banks. It excavates huge quantities of sediments, denature features of habitation, alter the structure and movement at the bottom community and increase turbidity that serves as a barrier to primary productivity, thereby impeding the food chain in the aquatic environment [5]. River Ethiope is majorly a recreational river that has several beaches. Being a fresh water river, the water finds its uses in drinking, washing, bathing, swimming and other domestic activities [6]. It also serves the purposes of fishing and dredging of white sand for infrastructural purposes. The river has several tributaries and creeks across the towns and villages. It has an annual overflow at its banks and as a result, it carries along the wastes from indiscriminate dumping activities via runoffs from rains into the water body [7].

Sediments display the quality of the aquatic system as well as information of the impact of the pollution sources on the environment. Sediments are an important part of aquatic ecosystem which serve as habitat, feeding, spawning and rearing areas for various aquatic organisms [6]. Detailed study of sediments across the world show that sediments act as sinks and sources of pollutants in aquatic systems due to their variable physical and chemical properties [6]. Studies revealed linkages across several components like soil, sediments, water, fish and air as transportation media for PCBs in rivers. There is a developed model showing that sediments and fish serve as the highest reservoir for PCBs contaminations [8, 9]. Much quantity of PCBs (over 70 %) might be resident in the soil, but when there is a decline in emission, the soil may act as an important second source of PCBs with the surface sediments of the dredged river acting as long-term chamber of PCBs [8]. The change in climate (external conditions), internal conditions, biogeochemical conditions in sediments and spoil of dredging like pH, salinity and redox potential could lead to the transportation of particles and dissolved pollutants into the overlying water [8, 10, 11]. In an aquatic environment, pollution is majorly observed in sediments and fish which serve as the primary reservoir of chemical elements in marine and freshwater ecosystems.

In recent years, fishing activities across the dredging tributaries and creeks have increased as a result of different categories of fish that can be caught easily for consumption. These fishes have nutritional values and are sources of proteins, minerals and unsaturated fatty acids [2]. Consumption of fish once or twice a day has been recommended by the American Heart Association for a healthy daily consumption of Omega-3 fatty acids [12]. Sediment-bound pollutants move from one place to another in the aquatic environments by means of trophic movements, such as consumption of benthic organisms by fish. These contaminants stick to the sediments for a long time, which make the sediments serve as a sink for different types of organic and inorganic compounds [13]. Studies have confirmed that aquatic environments seem to be the major places for assessing accumulation of PCBs [14–16]. This study focuses on the sources, risk assessment and bioaccumulation of PCBs contaminants in sediments and fish of tributaries and creeks of River Ethiope, Delta State, South-South, Nigeria as well as to assess the possible sources and relationship via Pearson's correlation coefficient, hazard index (HI) and incremental life-time cancer risk (ILCR), ecological risk factor, potential ecological risk index and bioaccumulation across seasons.

2. Materials and methods

2.1. Study area

The geographical location of the study area across the tributaries and creeks of River Ethiope was Latitude 5^0 47′17″ N. Longitude 6^0 4′49″ E (Abraka), Latitude 5^0 47′11″ N, Longitude 6^0 4′32″ E (Eku) and Latitude 5^0 47′20″ N, Longitude 6^0 4′47″ E (Sapele). The location cuts across the stretch of the river at different towns and villages. The tributaries and creeks are rich in white sand for infrastructural development and aquatic organisms. Tributaries and creeks make up about 40 % of the mangrove swamp and the agricultural land. The water body covers about 10 % and about 50 % is slated for buildup land along the study area. Much rainfall is observed in the study area, which ranges from 1600 – 2300 mm and covers from April – September (wet season) with a temperature range of 30 – 35 °C (maximum) and 20 – 23 °C (minimum) in the dry season which covers October – March in most cases. The dredged tributaries and creeks cut across the stretch of the river, from Umuaja to Sapele where it links to Mosogar from where it enters the Benin River [6]. As a result of several economic activities like canoeing, fishing, factory products and agriculture in the study area, several untreated wastes, such as pesticides residues, agricultural and dredging activities that obstruct the pH, salinity and redox potential of the aquatic ecosystem can lead to a serious threat to the aquatic environment.

2.2. Collection of sediments and Fish samples

Eighteen sediments samples consisting 3 from each site and 2 Tilapia fish (*Oreochromis niloticus*) were collected from site 1 for each season from dredged tributaries and creeks of Ethiope River across three most developed communities in the area: Site 1 (Abraka), Site 2 (Eku) and Site 3 (Sapele) in June (wet season) and in December (dry season) 2022. Van Veen grab sample and fish net by fisher men were used to collect the samples.

Table 1. Estimation of human health risk assessment.

Parameter	Unit	Values		References
		Child	Adult	
EF	Day/Year	313	313	[17]
ED	Year	6	30	[17]
ET	h/d	8	8	[18]
IngR	Mg/d	200	100	[19]
SA	Cm^2/event	2800	5700	[19]
ABS	-	0.13	0.13	[20]
AF	mg/cm^2	0.2	0.07	[20]
BW	Kg	15	60	[21]
ATnc & Atca	D	ED x 365 & LT x 365		[19]
LT	Year	52 Years		[22]
PEF	m^3/Kg	1.36×10^9		[19]

Table 2. Statistical analysis of PCBs congeners in sediments and fish across sites in wet season.

Compounds	SEDIMENT-Wet Season												FISH-Wet Season			
	Mean ± SD	Mean ± SD	Mean ± SD	Median			Max.			Min.			Mean ± SD	Median	Max.	Min.
	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3				
PCB-8	ND	ND	60.8±83.7	N.D	N.D	7.53	N.D	N.D	120	N.D	N.D	1.53	ND	ND	ND	ND
PCB-18	4.34±5.18	294±393	48.6±67.0	0.67	32.4	6.52	8.00	572	96	0.67	16.4	1.22	24.7 ± 16.5	13.0989	48	1.47
PCB-28	ND	N.D	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-44	ND	N.D	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	172 ± 115	91.1465	334	10.2
PCB-52	ND	18.5±24.7	22.3±30.7	N.D	2.03	4.56	N.D	36	44	N.D	1.03	0.56	ND	ND	ND	ND
PCB-66	ND	ND	351±484.5	N.D	N.D	32.8	N.D	N.D	694	N.D	N.D	8.84	42.3 ± 28.1	22.3773	82	2.5
PCB-77	ND	ND	60.8±83.8	N.D	N.D	8.53	N.D	N.D	120	N.D	N.D	1.53	ND	ND	ND	ND
PCB-81	8.67±10.4	ND	116±160.56	234	N.D	22.9	16.0	N.D	230	1.34	N.D	2.93	ND	ND	ND	ND
PCB-101	18.4±22.0	ND	830±1144	2.86	N.D	40.9	34.0	N.D	1640	2.86	N.D	20.9	ND	ND	ND	ND
PCB-105	22.8±27.2	2.06±2.75	1,300±1792	3.53	0.11	55.7	42.0	4	2568	3.53	0.11	32.7	ND	ND	ND	ND
PCB-114	19.5±23.3	65.8±87.9	1023±1410	4.03	8.68	49.7	36.0	128	2020	3.03	3.68	25.7	93.8 ± 62.3	49.6667	182	5.56
PCB-118	15.2±18.1	ND	N.D	3.35	N.D	N.D	28.0	N.D	N.D	2.35	N.D	N.D	ND	ND	ND	ND
PCB-123	34.7±41.5	11.3±15.1	87.1±120	5.38	1.63	2.19	64.0	22	172	5.38	0.63	2.19	35.0 ± 23.3	18.5568	68	2.08
PCB-126	16.3±19.4	ND	760±105	1.52	N.D	1.91	30.0	N.D	150	2.52	N.D	1.91	32.9 ± 21.9	17.4652	64	1.95
PCB-128	10.8±13.0	ND	ND	1.51	N.D	N.D	20.0	N.D	N.D	1.68	N.D	N.D	ND	ND	ND	ND
PCB-138	2.17±2.59	ND	ND	0.34	N.D	N.D	4.00	N.D	N.D	0.34	N.D	N.D	ND	ND	ND	ND
PCB-153	19.5±23.3	36.0±48.1	ND	3.03	5.01	N.D	36.0	70	N.D	3.03	2.01	N.D	35.0 ± 23.3	18.5568	68	2.08
PCB-156	4.34±5.18	ND	ND	0.67	N.D	N.D	8.00	N.D	N.D	0.67	N.D	N.D	ND	ND	ND	ND
PCB-157	5.42±6.48	ND	ND	0.84	N.D	N.D	10.0	N.D	N.D	0.84	N.D	N.D	ND	ND	ND	ND
PCB-167	ND	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-169	33.6±40.2	54.5±72.8	ND	5.21	6.05	N.D	62.0	106	N.D	5.21	3.05	N.D	128 ± 85.7	68.2234	250	7.63
PCB-170	24.9±29.8	62.8±83.8	ND	3.87	8.51	N.D	46.0	122	N.D	3.87	3.51	N.D	72.1 ± 48.0	38.2051	140	4.27
PCB-180	20.6±24.6	77.2±103	ND	3.19	9.31	N.D	38.0	150	N.D	3.19	4.31	N.D	48.4 ± 32.2	25.652	94	2.87
PCB-187	41.2±49.2	114±152	ND	6.39	13.5	N.D	76.0	222	N.D	6.39	6.38	N.D	155 ± 103	81.8681	300	9.16
PCB-189	56.4±67.4	171±228	ND	6.74	15.5	N.D	104	332	N.D	8.74	9.54	N.D	362 ± 241	191.571	702	21.4
PCB-195	13.0±15.5	170±227	ND	3.02	9.48	N.D	24.0	330	N.D	2.02	9.48	N.D	66.0 ± 43.9	34.9304	128	3.91
PCB-206	136±162	326±435	ND	21	30.2	N.D	250	634	N.D	21	18.2	N.D	344 ± 229	182.293	668	20.4
PCB-209	138±165	387±516	ND	21.3	31.6	N.D	254	752	N.D	21.3	21.6	N.D	76.3 ± 50.7	40.3883	148	4.52
Total	645±771	1790±2390	3977±5483	332	174	233.2	1190	3480	7854	100	100	100	1688 ± 1122	894	3276	100

Both sediments and fish samples were conveyed to the laboratory and stored in a deep freezer at -20 °C prior to further analysis. The muscle tissue of the fish was used for the analysis. Standard analytical procedures were used for the analysis of the PCBs contaminant in both samples [23].

Table 3. Statistical analysis of PCBs congeners in sediments and fish across sites in the dry season.

Compounds	SEDIMENT-Dry Season												FISH-Dry Season			
	Mean ± SD	Mean ± SD	Mean ± SD	Median			Max.			Min.			Mean ± SD	Median	Max.	Min.
	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3				
PCB-8	5.21 ± 3.38	26.4 ± 15.2	N.D	2.82	15.67	ND	10.0	48.0	ND	0.43	4.89	ND	ND	ND	ND	ND
PCB-18	6.26 ± 4.06	6.61 ± 3.81	1.25 ± 0.53	3.38	3.92	0.87	12.0	12.0	2.00	0.51	1.22	0.49	4.39 ± 2.55	2.59	8.00	0.79
PCB-28	N.D	N.D	N.D	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-44	N.D	N.D	N.D	- ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-52	20.9 ± 13.5	11.0 ± 6.35	1.25 ± 0.53	11.28	6.53	0.87	40.0	20.0	2.00	1.71	2.04	0.49	27.5 ± 15.9	16.2	50.0	4.93
PCB-66	22.9 ± 14.9	74.9 ± 43.2	4.99 ± 2.13	12.41	44.39	3.49	44.0	136	8.00	1.88	13.9	1.98	ND	ND	ND	ND
PCB-77	3.13 ± 2.03	N.D	1.25 ± 0.53	1.69	ND	0.87	6.00	ND	2.00	0.26	ND	0.49	7.69 ± 4.46	4.54	14.0	1.38
PCB-81	196 ± 127	9.92 ± 5.72	2.50 ± 1.06	106.06	5.87	1.74	376	18.0	4.00	16.1	1.83	0.99	36.3 ± 21.0	21.4	66.0	6.51
PCB-101	69.9 ± 45.4	20.9 ± 12.1	2.50 ± 1.06	37.80	12.40	1.74	134	38.0	4.00	5.73	3.87	0.99	4.39 ± 2.55	2.59	8.00	0.79
PCB-105	82.4 ± 53.5	9.92 ± 5.72	125 ± 0.53	44.57	5.87	0.87	158	18.0	2.00	6.76	1.83	0.49	4.39 ± 2.56	2.59	8.00	0.79
PCB-114	126.2 ± 81.9	76.0 ± 43.8	9.98 ± 4.26	68.26	45.04	6.97	242	138	16.0	10.4	14.1	3.96	69.2 ± 40.2	40.8	126	12.4
PCB-118	19.8 ± 12.9	16.6 ± 9.53	1.25 ± 0.53	10.72	9.79	0.87	38.0	30.0	2.00	1.63	3.06	0.49	15.4 ± 8.92	9.07	28.0	2.76
PCB-123	124.1 ± 80.6	9.92 ± 5.72	1.25 ± 0.53	67.13	5.87	0.87	238	18.0	2.00	10.2	1.83	0.49	6.59 ± 3.82	3.89	12.0	1.18
PCB-126	76.1 ± 49.4	8.81 ± 5.08	3.74 ± 1.60	41.18	5.22	2.61	146	16.0	6.00	6.24	1.63	1.49	2.20 ± 1.27	1.29	4.00	0.39
PCB-128	1.04 ± 0.68	N.D	N.D	0.56	ND	ND	ND	ND	ND	0.09	ND	ND	ND	ND	ND	ND
PCB-138	N.D	N.D	N.D	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PCB-153	7.30 ± 4.74	12.1 ± 6.99	2.50 ± 1.06	3.95	7.18	1.74	14.0	22.0	4.00	0.60	2.24	0.99	7.69 ± 4.46	4.54	14.0	1.38
PCB-156	N.D	N.D	N.D	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-157	N.D	N.D	N.D	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-167	N.D	N.D	N.D	ND	ND	N.D	N.D	N.D	N.D	N.D	N.D	N.D	ND	ND	ND	ND
PCB-169	9.38 ± 6.09	36.4 ± 20.9	2.50 ± 1.06	5.08	21.54	1.74	18.0	66.0	4.00	0.77	6.72	0.99	10.9 ± 6.37	6.48	20.0	1.97
PCB-170	16.7 ± 10.8	15.4 ± 8.89	1.25 ± 0.53	9.03	9.14	0.87	32.0	28.0	2.00	1.37	2.85	0.49	38.5 ± 22.3	22.7	70.0	6.90
PCB-180	37.5 ± 24.4	26.4 ± 15.2	11.23 ± 4.79	20.31	15.67	7.84	72.0	48.0	18.0	3.08	4.89	4.46	27.5 ± 15.9	16.2	50.0	4.93
PCB-187	235 ± 152	36.4 ± 20.9	2.50 ± 1.06	126.94	21.54	1.74	450	66.0	4.00	19.2	6.72	0.99	8.79 ± 5.10	5.18	16.0	1.58
PCB-189	23.9 ± 15.6	40.8 ± 23.5	7.49 ± 3.19	12.98	24.15	5.23	46.0	74.0	12.0	1.97	7.54	2.97	1.10 ± 0.64	0.65	2.00	0.20
PCB-195	31.3 ± 20.3	50.7 ± 29.2	2.50 ± 1.06	16.92	30.03	1.74	60.0	92.0	4.00	2.57	9.37	0.99	47.2 ± 27.4	27.7	86.0	8.48
PCB-206	93.9 ± 60.9	28.7 ± 16.5	178 ± 76.1	50.77	16.97	125	180	52.0	286	7.70	5.29	70.8	24.2 ± 14.0	14.3	44.0	4.34
PCB-209	10.4 ± 6.77	23.1 ± 13.3	12.5 ± 5.32	5.64	13.71	8.71	20.0	42.0	20.0	0.86	4.28	4.95	213 ± 124	126	388	38.3
Total	1219 ± 791	541 ± 312	252 ± 215	659.50	320.50	176	2338	982	404	100	100	100	557 ± 323	329	1014	100

2.3. Analytical procedures

Extraction, purification/fractionation of the chromatographic separation and quantification were performed for the determination of PCBs in samples collected from the aquatic environment.

Table 4. Statistical analysis of PCBs congeners in sediments and fish across sites in the dry season.

	PCB-8	PCB-18	PCB-28	PCB-44	PCB-52	PCB-66	PCB-77	PCB-81	PCB-101	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-128	PCB-138	PCB-153	PCB-156	PCB-157	PCB-167	PCB-169	PCB-170	PCB-180	PCB-187	PCB-189	PCB-195	PCB-206	PCB-209
PCB-8	1.00																											
PCB-18	-0.37	1.00																										
PCB-28	ND	ND	1.00																									
PCB-44	ND	ND	ND	1.00																								
PCB-52	0.63	0.48	ND	ND	1.00																							
PCB-66	1.00*	-0.37	ND	ND	0.63*	1.00																						
PCB-77	1.00*	-0.37	ND	ND	0.63*	1.00	1.00																					
PCB-81	1.00*	-0.43	ND	ND	0.58	1.00*	1.00*	1.00																				
PCB-101	1.00*	-0.39	ND	ND	0.62*	1.00*	1.00*	1.00*	1.00																			
PCB-105	1.00*	-0.39	ND	ND	0.62*	1.00*	1.00*	1.00*	1.00*	1.00																		
PCB-114	1.00*	-0.33	ND	ND	0.66*	1.00*	1.00*	0.99*	1.00*	1.00*	1.00																	
PCB-118	-0.50	-0.62	ND	ND	-0.99	-0.50	-0.50	-0.44	-0.48	-0.49	-0.53	1.00																
PCB-123	0.95*	-0.63	ND	ND	0.37	0.95*	0.95*	0.97*	0.96*	0.96*	0.94*	-0.22	1.00															
PCB-126	1.00*	-0.39	ND	ND	0.62	1.00*	1.00*	1.00*	1.00	1.00	1.00*	-0.48	0.96*	1.00														
PCB-128	-0.50	-0.62	ND	ND	-0.99	-0.50	-0.50	-0.44	-0.48	-0.49	-0.53	1.00*	-0.22	-0.48	1.00													
PCB-138	-0.50	-0.62	ND	ND	-0.99	-0.50	-0.50	-0.44	-0.48	-0.49	-0.53	1.00*	-0.22	-0.48	1.00*	1.00												
PCB-153	-0.89	0.76*	ND	ND	-0.21	-0.89	-0.89	-0.92	-0.90	-0.90	-0.87	0.05	-0.99	-0.90	0.05	0.05	1.00											
PCB-156	-0.50	-0.62	ND	ND	-0.99	-0.50	-0.50	-0.44	-0.48	-0.49	-0.53	1.00*	-0.22	-0.48	1.00*	1.00*	0.05	1.00										
PCB-157	-0.50	-0.62	ND	ND	-0.99	-0.50	-0.50	-0.44	-0.48	-0.49	-0.53	1.00*	-0.22	-0.48	1.00*	1.00	0.05	1.00*	1.00									
PCB-167	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00								
PCB-169	-0.92	0.70*	ND	ND	-0.29	-0.92	-0.92	-0.95	-0.93	-0.93	-0.91	0.13	-1.00	-0.93	0.13	0.13	1.00*	0.13	0.13	ND	1.00							
PCB-170	-0.80	0.85*	ND	ND	-0.04	-0.80	-0.80	-0.84	-0.81	-0.81	-0.78	-0.12	-0.94	-0.81	-0.12	-0.12	0.99*	-0.12	-0.12	ND	0.97*	1.00						
PCB-180	-0.71	0.92*	ND	ND	0.10	-0.71	-0.71	-0.75	-0.72	-0.72	-0.68	-0.26	-0.89	-0.72	-0.26	-0.26	0.95*	-0.26	-0.26	ND	0.92*	0.99*	1.00					
PCB-187	-0.78	0.87*	ND	ND	0.00	-0.78	-0.78	-0.82	-0.79	-0.78	-0.75	-0.16	-0.93	-0.79	-0.16	-0.16	0.98*	-0.16	-0.16	ND	0.96*	1.00*	0.99*	1.00				
PCB-189	-0.75	0.89*	ND	ND	0.03	-0.75	-0.75	-0.80	-0.77	-0.76	-0.73	-0.19	-0.92	-0.77	-0.19	-0.19	0.97*	-0.19	-0.19	ND	0.95*	1.00*	1.00*	1.00*	1.00			
PCB-195	-0.56	0.98*	ND	ND	0.29	-0.56	-0.56	-0.61	-0.57	-0.57	-0.52	-0.44	-0.78	-0.57	-0.44	-0.44	0.88*	-0.44	-0.44	ND	0.83*	0.94*	0.98*	0.96*	0.97*	1.00		
PCB-206	-0.81	0.84*	ND	ND	-0.06	-0.81	-0.81	-0.85	-0.83	-0.82	-0.79	-0.10	-0.95	-0.83	-0.10	-0.10	0.99*	-0.10	-0.10	ND	0.97*	1.00*	0.99*	1.00*	1.00*	0.94*	1.00	
PCB-209	-0.77	0.88*	ND	ND	0.00	-0.77	-0.77	-0.81	-0.78*	-0.78	-0.75	-0.16	-0.93	-0.78	-0.16	-0.16	0.98*	-0.16	-0.16	ND	0.96*	1.00*	1.00*	1.00*	1.00*	0.96*	1.00*	1.00

* Significant correlation at the p < 0.01 level.

2.4. *Sample preparations*

The sediment samples were dried in the oven at 60 °C, ground and filtered with 2 mm mesh sieve according to a previous method [16]. The fish was prepared in accordance with guidelines [23]. The aluminum foil covering the fish was removed including the fillet of both sides of the fish, rinsed with tap water and distilled water before dissection. The muscle tissue was obtained and ground with mortar and pestle, mixed

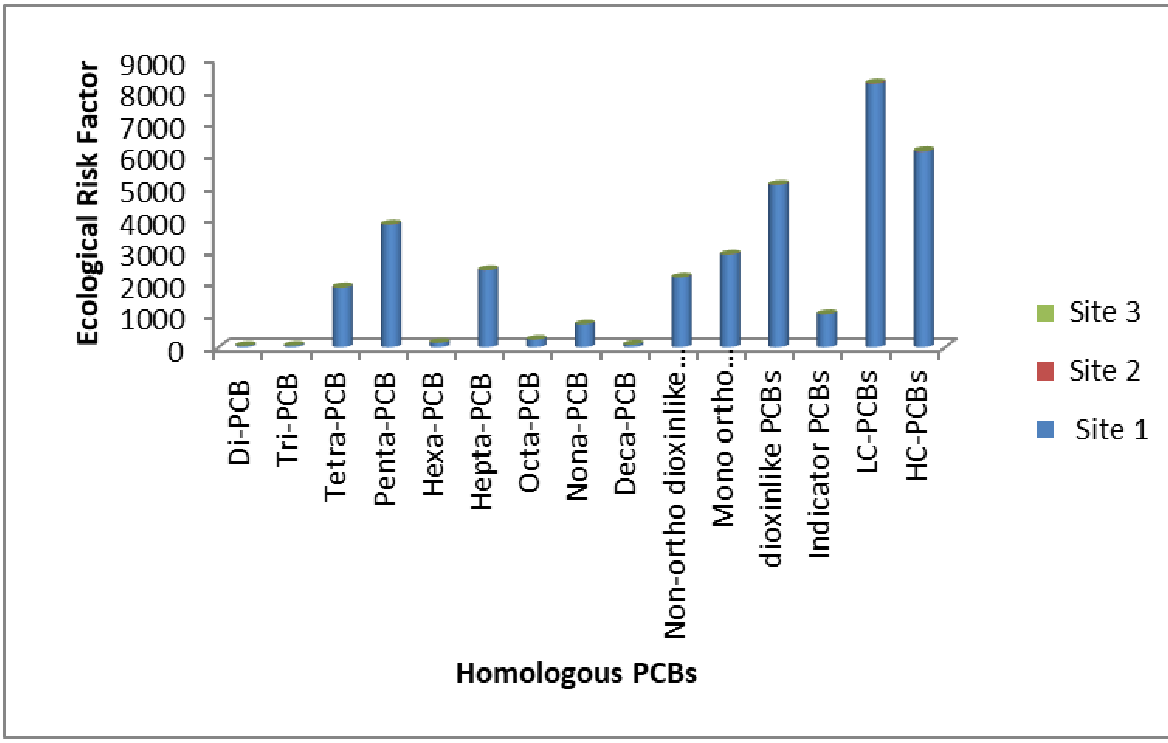

Figure 1. Ecological risk factor of the homologous PCBs across the sites in wet season.

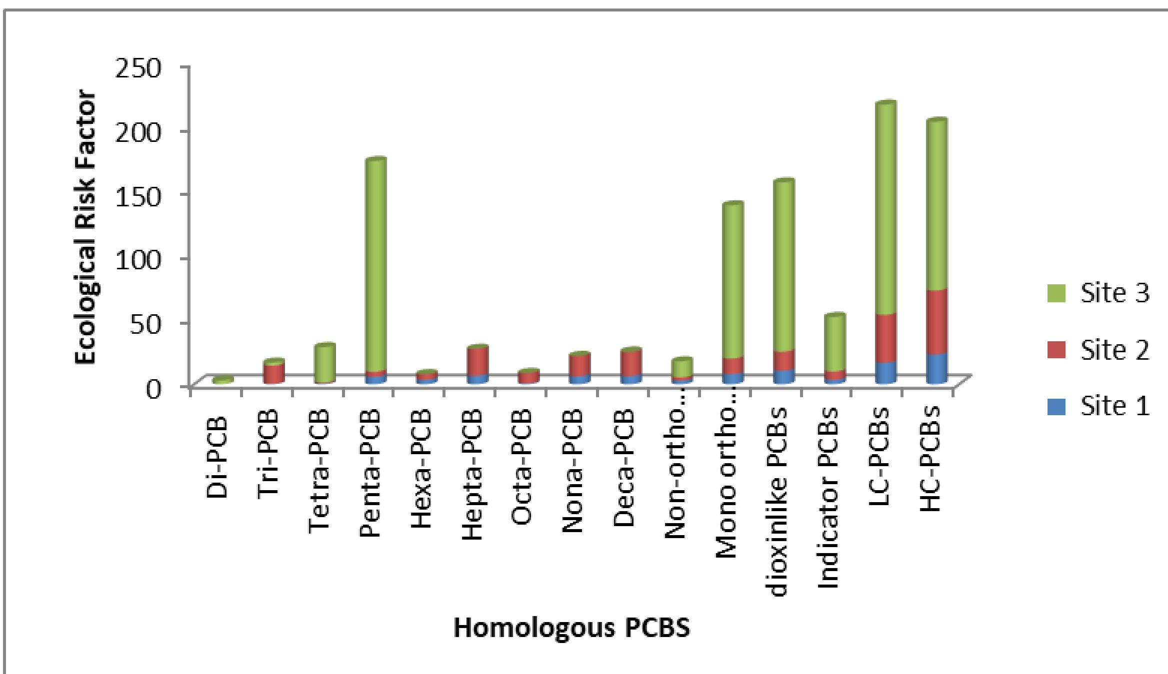

Figure 2. Ecological risk factor of the homologous PCBs across the sites in dry season.

with anhydrous sodium sulphate and homogenized. Thereafter, 10 g of the sample was used for extraction.

2.5. *Extraction of samples*

After drying, the sediments were spiked with surrogate mixture of 2 ng/kg of PCB IUPAC # 65 and PCB IUPAC # 166, 5 g aliquot was extracted three times by sonication with 15 mLl dichloromethane (1:1) for 20 minutes. There was concentration and fractionation of the organic extract which was carried out with 3 g of neutral alumina Carlo Erba, followed by deactivation with 3% (w/w) Milli-Q water and the PCBs were eluted with 5.5 and 6 mL of hexane and hexane/ethyl acetate (9:1)

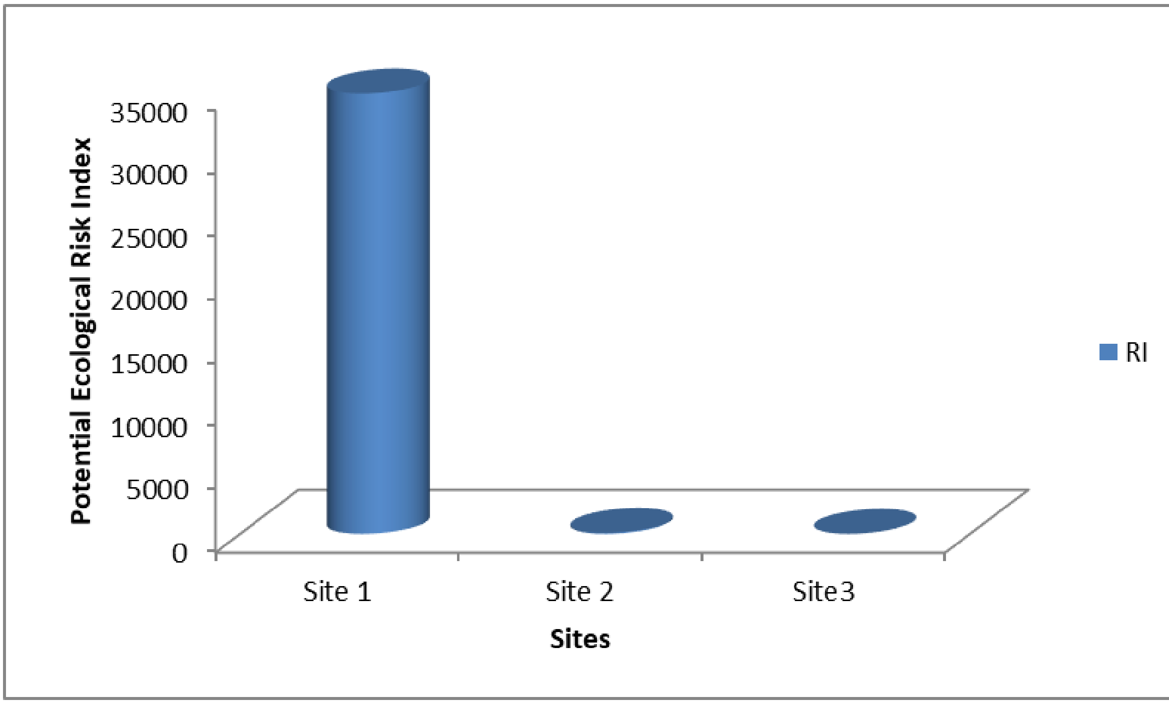

Figure 3. Potential ecological risk index of the homologous PCBs across the sites in wet season.

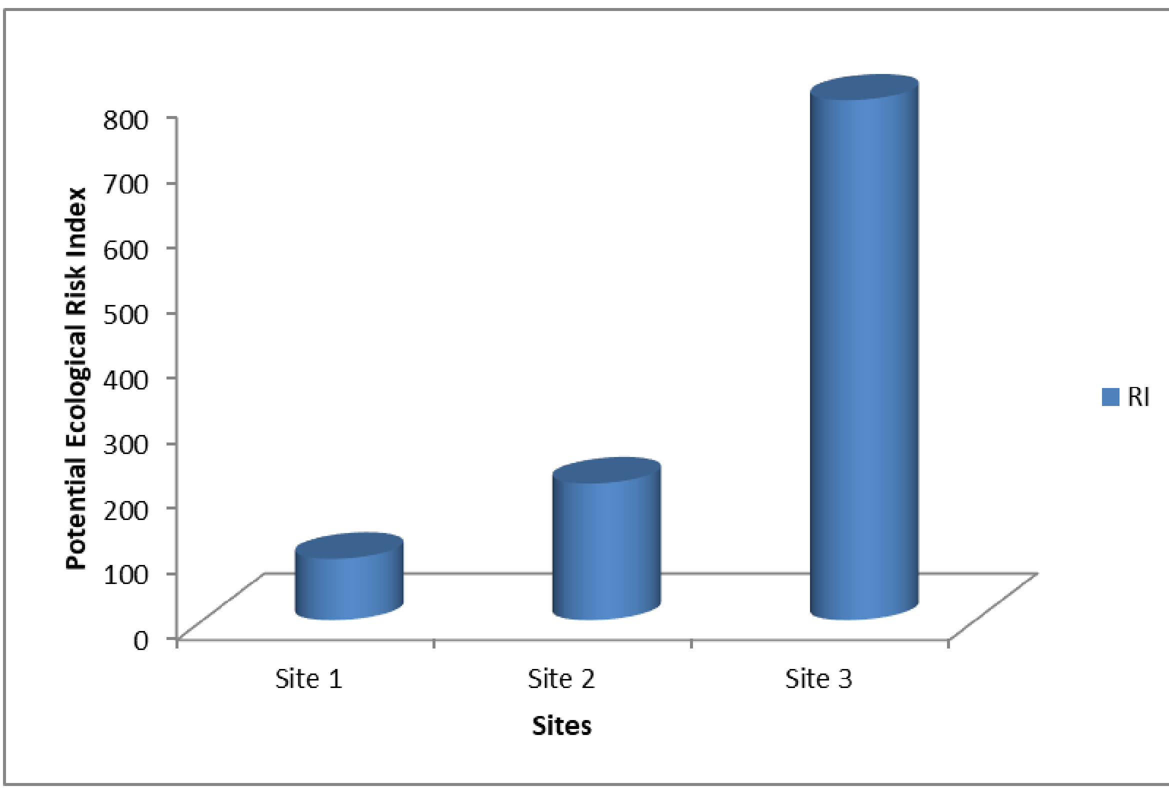

Figure 4. Potential ecological risk index of the homologous PCBs across the sites in dry season.

(Merck) in separate fractions. Then, the final elution of the column was done with 12 mL of ethyl acetate in another fraction having more polar compounds [16]. The fish sample extraction was done according to a modified standard procedure [24] (extract concentration was made to 2 mlL and not 1 mL). Tetrachloro-m-xylene used as surrogate standard was added to the sample before extraction. The extraction was carried out using hexane and acetone mixed in the ratio 1:1 for 20 – 22 hours with Soxhlet apparatus turning 5 – 6 times/hour. The concentration of the extract was made to 2 mL with the aid of a rotary evaporator at 40 °C. 1 mL of the concentrated extract was used for cleanup, while the leftover of 1 mL was used to conduct gravimetric determination.

2.6. Extract cleanup

The cleanup was done with strong acid, after which partitioning was carried out with florisil as follows: 2 mL of concentrated sulphuric acid was mixed with 1 mL of the extract with thorough shaking to avoid co-extract interference. The cleanup was done severally until acid phase was colourless. The extract was dried with anhydrous sodium sulphate before fractionation with florisil. The packing of the glass column was done with 10 g of florisil (60 – 100 mesh) with addition of 2 g anhydrous sodium sulphate at the upper part of the column to remove water vapor. About 1 mL of the extract was inserted into the column followed by elute of PCBs with 80 mL hexane. The concentration of elute was carried out with a rotary evaporator and a gradual nitrogen stream. About 1 mLl hexane was used to dissolve the PCB fraction and conveyed to vials of Gas chromatograph awaiting Gas chromatography analysis. Gas Chromatography-Mass Spectrometry (GC – MS) (Clarus 680, Perkin-Elmer Inc., Waltham MA, USA) analysis was done in single ion monitoring (SIM) mode for assessment and quantification of PCB congeners after a known amount of IUPAC no. 209 internal standard was added to the cleaned extract.

2.7. Determination of PCBs congeners

PCBs determination was done on sediments and fish samples with the help of Hewlett-Packard 7890 Gas chromatograph coupled with a Hewlett Packard Model 5975 mass analyzer: quadrupole and automatic sampler. The column made up of fused silica capillary was a 30 m DB – 1 ms (100% dimethylsiloxane) (Cj & W Scientific, CA, USA) (0.25 mm i.d. x 0.25 µm of the thickness of film). The temperature of the oven was initially programmed at 100 °C (standing for 1 min) – 325°C at the ratio of 15 °C/min for 5 min. The conditions under which the gas chromatograph was carried out were 250 °C injection temperature and 280 °C transfer line temperature. Helium was used as a carrier gas through a steady flow count of 1.2 mL/min, with an elevated oven temperature of 120 °C for 0.5 min to 210 °C at 30 °C/min to 300 °C at 6 °C for 5 min. The running conditions of the mass spectrometer were 250 °C used for transfer line and ionization source temperature, 70eV was used as ionization voltage, the time used for scanning was 0.3s with 0.5s scanning delay and 5 minutes delay solvent. Statistical analysis for all data obtained was done using Microsoft Excel (Microsoft Inc., USA) and SPSS 22.0 (SPSS Inc., Chicago II., USA).

2.8. Identification and quantification of PCBs

The analyzed compounds identification was based upon the ion ratio, the spectra and the retention time. These were done by internal standard procedures using peak areas. The MS analysis was done using single ion monitoring mode where 2 masses were monitored in the mass spectrum. The chromatogram retention time shifts were corrected with the use of internal standard retention time and agreement of about ± 0.1 minute of the retention time expected for the positive confirmation of the analyte. The linear range of the detector was evaluated by preparing several standard solutions which were injected at various concentrations. The calibration curves were within the range of 10 – 200 ng/ml, the correlation coefficient (r_2) was above 0.999 for all compounds analyzed.

2.9. Bioaccumulation factor

The determination of the bioaccumulation factor of the analytes in fish was done relative to the dissolved concentration of the analytes in the sediments (sediment-based bioaccumulation factor, SBF). That is, by dividing the concentration of the chemical compound in the fish tissue by the concentration of the same compound in the sediment according to a previous method [2].

2.10. Human health risk and toxic equivalency (TEQ) assessment

Hazardous index (HI) and incremental life cancer risk (ILCR) were used for quantitative measurement of the non-cancer and cancer effects of PCBs in sediments and *Oreochromis niloticus* for humans. HI and ILCR showed the potential non cancer and cancer risk of individual. The calculated non-cancer risk for children and adults was done using Equations (1) – (13).

$$CDI_{Ing-nc} = \frac{C \times IngR \times EF\,XED}{BW \times ATnc} x10^{-6} \quad (1)$$

$$CDI_{Inh-nc} = \frac{C \times EF \times ET \times ED}{PEF \times 24\,ATnc} \times 10^{6} \quad (2)$$

$$CDI_{dermal-nc} = \frac{C \times SA \times AF \times ABSd \times EF \times ED}{BW \times ATnc} \quad (3)$$

$$\text{Hazard } Quotient\ (HQ) = \frac{CDInc}{RfD} \quad (4)$$

$$\text{Hazard } Index\ ((HI)) = \sum (HQ) = \text{HQing} + \text{HQinh} + \text{HQderm}.$$

$$\text{Hazard } Index(HI) = \frac{CDIing - nc}{RfDing} + \frac{CDIinh - nc}{RfCinh} + \frac{CDIdermal - nc}{RdDermal}. \quad (5)$$

The calculated incremental life cancer risk was done using Equation (6) – (13).

$$CDI_{\ Ing-ca} = \frac{C \times Ing\,Radj \times EF}{ATca} \times 10^{-6}, \quad (6)$$

$$IngRadj = \frac{EDchild \times IngRchild}{BWchild} + \frac{(EDadult - EDchild)}{BWadult} \times IngRadj, \quad (7)$$

Table 5. Pearson correlation coefficient for sediments in dry season.

	PCB-8	PCB-18	PCB-28	PCB-44	PCB-52	PCB-66	PCB-77	PCB-81	PCB-101	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-128	PCB-138	PCB-153	PCB-156	PCB-157	PCB-167	PCB-169	PCB-170	PCB-180	PCB-187	PCB-189	PCB-195	PCB-206	PCB-209
PCB-8	1.00																											
PCB-18	0.66	1.00																										
PCB-28	ND	ND	1.00																									
PCB-44	ND	ND	ND	1.00																								
PCB-52	0.17	0.85	ND	ND	1.00																							
PCB-66	1.00	0.72	ND	ND	0.24	1.00																						
PCB-77	-0.61	0.19	ND	ND	0.68	-0.55	1.00																					
PCB-81	-0.29	0.53	ND	ND	0.90*	-0.21	0.93	1.00																				
PCB-101	-0.07	0.70	ND	ND	0.97*	0.01	0.83	0.98	1.00																			
PCB-105	-0.23	0.58	ND	ND	0.92	-0.15	0.91	1.00*	0.99	1.00																		
PCB-114	0.24	0.89	ND	ND	1.00	0.32	0.62	0.86	0.95	0.89	1.00																	
PCB-118	0.49	0.98	ND	ND	0.94	0.55	0.39	0.70	0.84	0.74	0.97	1.00																
PCB-123	-0.26	0.55	ND	ND	0.91	-0.19	0.92	1.00	0.98	1.00	0.87	0.72*	1.00															
PCB-126	-0.26	0.55	ND	ND	0.91	-0.18	0.92	1.00	0.98	1.00	0.87	0.72*	1.00	1.00														
PCB-128	-0.32	0.50	ND	ND	0.88	-0.24	0.94	1.00	0.97	1.00	0.84	0.67	1.00	1.00*	1.00													
PCB-138	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00												
PCB-153	0.93	0.90	ND	ND	0.53	0.95	-0.27	0.10	0.31	0.16	0.59	0.78*	0.12	0.13	0.06	ND	1.00											
PCB-156	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00										
PCB-157	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00									
PCB-167	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00								
PCB-169	1.00	0.67*	ND	ND	0.19	1.00	-0.60	-0.27	-0.05	-0.21	0.26	0.50	-0.24	-0.24	-0.30	ND	0.93	ND	ND	ND	1.00							
PCB-170	0.56	0.99	ND	ND	0.91	0.63	0.31	0.63*	0.78	0.67	0.94*	1.00*	0.65	0.65*	0.60	ND	0.84	ND	ND	ND	0.58	1.00						
PCB-180	0.26	0.90	ND	ND	1.00	0.33	0.60	0.85*	0.95*	0.88	1.00*	0.97*	0.86*	0.87	0.83	ND	0.61	ND	ND	ND	0.28	0.94*	1.00					
PCB-187	-0.19	0.61	ND	ND	0.93	-0.12	0.90	1.00*	0.99*	1.00	0.90	0.76*	1.00*	1.00	0.99	ND	0.19	ND	ND	ND	-0.18	0.70*	0.90	1.00				
PCB-189	0.93	0.89	ND	ND	0.52	0.95	-0.27	0.09	0.31	0.15	0.59	0.78*	0.12	0.12	0.06	ND	1.00	ND	ND	ND	0.94	0.83*	0.60	0.18	1.00			
PCB-195	0.89	0.93	ND	ND	0.60	0.92	-0.18	0.19	0.40	0.25	0.66	0.84	0.22	0.22	0.16	ND	1.00	ND	ND	ND	0.89	0.88*	0.68	0.28	0.99*	1.00		
PCB-206	-0.96	-0.84	ND	ND	-0.43	-0.98	0.38	0.02	-0.20	-0.04	-0.49	-0.70	-0.01	-0.01	0.05	ND	-0.99	ND	ND	ND	-0.97	-0.76*	-0.51	-0.07	-0.99	-0.98	1.00	
PCB-209	0.98	0.50	ND	ND	-0.03	0.96	-0.76	-0.47	-0.27	-0.42	0.05	0.31	-0.45	-0.44	-0.50	ND	0.83	ND	ND	ND	0.98	0.39	0.06	-0.38	0.84	0.78	-0.89	1.00

$$CDI_{Inh-ca} = \frac{C \times EF \times ET \times ED}{PEF \times 24\,ATca} \times 10^3, \quad (8)$$

$$CDI_{dermal-ca} = \frac{C \times ABSd \times EF \times DFSadj}{ATca} \times 10^{-6}, \quad (9)$$

$$DFS_{adj} = \frac{EDchild \times SAchild \times AFchild}{BWchild} + \frac{(EDadult - EDadult) \times SAadult \times AFadult}{BWadult}, \quad (10)$$

Table 6. Pearson correlation coefficient for Oreochromis *niloticus* in wet seasons.

	PCB-8	PCB-18	PCB-28	PCB-44	PCB-52	PCB-66	PCB-77	PCB-81	PCB-101	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-128	PCB-138	PCB-153	PCB-156	PCB-157	PCB-167	PCB-169	PCB-170	PCB-180	PCB-187	PCB-189	PCB-195	PCB-206	PCB-209
PCB-8	1.00																											
PCB-18	ND	1.00																										
PCB-28	ND	ND	1.00																									
PCB-44	ND	ND	ND	1.00																								
PCB-52	ND	1.00*	ND	ND	1.00																							
PCB-66	ND	ND	ND	ND	ND	1.00																						
PCB-77	ND	1.00*	ND	ND	1.00*	ND	1.00																					
PCB-81	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00																				
PCB-101	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00																			
PCB-105	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00																		
PCB-114	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00																	
PCB0-118	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00																
PCB-123	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00															
PCB-126	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00														
PCB-128	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00													
PCB-138	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00												
PCB-153	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00	1.00*	1.00*	1.00	1.00*	1.00	ND	ND	1.00											
PCB-156	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00										
PCB-157	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00									
PCB-167	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00								
PCB-169	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00*	ND	ND	ND	1.00							
PCB-170	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00*	ND	ND	ND	1.00*	1.00						
PCB-180	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00					
PCB-187	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00	ND	ND	ND	1.00*	1.00*	1.00*	1.00				
PCB-189	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00			
PCB-195	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00		
PCB-206	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00	
PCB-209	ND	1.00*	ND	ND	1.00*	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	ND	ND	1.00	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00

*Significant correlation at the $p < 0.01$ level.

$$Risk = CDI_{ca} \times CSF, \quad (11)$$

$$\text{Total } Risk = Risk_{ing} + Risk_{inh} + Risk_{dermal}, \quad (12)$$

$$\text{Total } Risk = CDI_{Ing-ca} \times CSF_{Ing} + CD_{Inh-ca} \times IUR + \frac{CDIdermal \times CSFing}{ABS_{GI}}, \quad (13)$$

Table 7. Pearson correlation coefficient for *Oreochromis niloticus* in dry seasons.

	PCB-8	PCB-18	PCB-28	PCB-44	PCB-52	PCB-66	PCB-77	PCB-81	PCB-101	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-128	PCB-138	PCB-153	PCB-156	PCB-157	PCB-167	PCB-169	PCB-170	PCB-180	PCB-187	PCB-189	PCB-195	PCB-206	PCB-209
PCB-8	1.00																											
PCB-18	ND	1.00																										
PCB-28	ND	NS	1.00																									
PCB-44	ND	1.00*	ND	1.00																								
PCB-52	ND	ND	ND	ND	1.00																							
PCB-66	ND	1.00*	ND	1.00*	ND	1.00																						
PCB-77	ND	ND	ND	ND	ND	ND	1.00																					
PCB-81	ND	ND	ND	ND	ND	ND	ND	1.00																				
PCB-101	ND	ND	ND	ND	ND	ND	ND	ND	1.00																			
PCB-105	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00																		
PCB-114	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00																	
PCB0-118	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00																
PCB-123	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00															
PCB-126	ND	-1.00	ND	-1.00	ND	-1.00	ND	ND	ND	ND	-1.00	ND	-1.00	1.00														
PCB-128	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00													
PCB-138	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00												
PCB-153	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00	-1.00	ND	ND	1.00											
PCB-156	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00										
PCB-157	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00									
PCB-167	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.00								
PCB-169	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00							
PCB-170	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00						
PCB-180	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00					
PCB-187	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00*	1.00				
PCB-189	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00			
PCB-195	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00		
PCB-206	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00*	1.00*	1.00*	1.00*	1.00	
PCB-209	ND	1.00*	ND	1.00*	ND	1.00*	ND	ND	ND	ND	1.00*	ND	1.00*	-1.00	ND	ND	1.00*	ND	ND	ND	1.00*	1.00*	1.00*	1.00	1.00*	1.00*	1.00*	1.00

* Represent a significant correlation at the $p < 0.01$ level.

where EF = Exposure frequency, ED = Exposure duration, ET = Exposure time, IngR = Ingestion rate for receptor, InhR = Inhalation rate, SA = Skin surface area, ATnc = Average time for non cancer, ATca = Average time for cancer, PEF = Sediment to air particulate emission factor, ABS = Dermal absorption factor for PCBs, AF = Sediment to skin adherences factor, BW = average body weight, LT = Life time, CDIing, CDIinh, CDIdermal = Chronic daily intake or dose contacted

Table 8. Incremental life-time cancer risk for children and adults across sites and seasons.

	Sites	Cancer risk for children				Cancer risk for adults			
		ILCRIng	ILCRInh	ILCRDerm	TCR	ILCRIng	ILCRInh	ILCRDerm	TCR
Wet	Site 1	6.10x10^{-10}	2.20x10^{-10}	2.0x10^{-10}	6. 1x10^{-3}	4.2x10^{-4}	1.0x10^{-12}	2.2x10^{4}	6.3x10^{-4}
	Site 2	1.10x10^{-2}	3.59x10^{-10}	3.93x10^{-3}	1.47x10^{-2}	7.36x10^{-4}	2.33x10^{-12}	3.82x10^{-4}	1.11x10^{-3}
	Site 3	6.44x10^{-2}	2.14x10^{-9}	2.34x10^{-2}	8.78x10^{-2}	4.39x10^{-3}	1.20x10^{-11}	2.28x10^{-3}	6.67x10^{-3}
Dry	Site 1	1.84x10^{-2}	6.13x10^{-10}	1.15x10^{-2}	2.99x10^{-2}	1.25x10^{-3}	3.97x10^{-12}	6.51x10^{-4}	1.91x10^{-3}
	Site 2	5.42x10^{-3}	1.80x10^{-10}	1.97x10^{-4}	7.38x10^{-3}	3.69x10^{-4}	1.17x10^{-12}	1.91x10^{4}	5.61x10^{4}
	Site 3	7.65 x10^{-4}	2.54 x10^{-11}	2.79 x10^{-4}	1.04x10^{-3}	5.23 x10^{-5}	1.65 x10^{-13}	2.57 x10^{-5}	7.80 x10^{-5}

Note: ILCR = Incremental life-time cancer risk, Ing = ingestion, In = Inhalation, Derm = Dermal.

Table 9. Incremental life-time cancer risk for children and adults across sites and seasons.

	Sites	Non-cancer risk for children				Non-cancer risk for adults			
		HQIng	HQInh	HQDerm	HI	HQIng	HQInh	HQDerm	HI
Wet	Site 1	4.22x10^{-1}	2.22x10^{-3}	1.5x10^{-1}	5.72x10^{-1}	5.2x10^{-2}	9.0 x10	4.6x10^{-4}	6.2x10^{-2}
	Site 2	7.39x10^{-1}	3.80x10	2.69x10^{-1}	1.01	9.23x10^{-2}	1.59x10^{-2}	8.23x10^{-4}	1.09x10^{-1}
	Site 3	4.41	2.27x10^{-2}	1.61	6.04	5.51x10^{-1}	9.46x10^{-2}	4.90x10^{-2}	6.51x10^{-1}
Dry	Site 1	1.26	6.49x10^{-3}	4.72x10^{-1}	1.74	1.58x10^{-1}	2.70x10^{-2}	1.40x10^{-3}	1.86x10^{-1}
	Site 2	3.71x10^{-1}	1.91x10^{-3}	1.31x10^{-1}	5.04x10^{-1}	4.63x10^{-2}	7.95x10^{-3}	4.12x10^{-4}	5.47x10^{-2}
	Site 3	5.24 x10^{-2}	2.70 x10^{-4}	1.91 x10^{-2}	7.18 x10^{-2}	6.56 x10^{-3}	1.12 x10^{-3}	5.83 x10^{-5}	7.74 x10^{-3}

Note: ILCR = Incremental life-time cancer risk, Ing = ingestion, In = Inhalation, Derm = Dermal.

Table 10. Incremental life-time cancer risk for children and adults across sites and seasons.

	Sites	PCB-77	PCB-81	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-156	PCB-157	PCB-167	PCB-169	PCB-189	TTEQ
Wet	Site 1	ND	ND	ND	3.80X10^{-3}	ND	7.00X10^{-4}	2.20	ND	ND	ND	31.8	0.01	34.0
	Site 2	ND	5.00X10^{-4}	ND	1.30X10^{-3}	ND	8.00X10^{-4}	6.40	2.00X10^{-4}	ND	ND	18.6	3.10X10^{-3}	25.0
	Site 3	ND	6.90X10^{-3}	ND	6.10X10^{-2}	ND	5.20X10^{-3}	15.0	ND	ND	ND	ND	ND	15.0
Dry	Site 1	ND	11.3X10^{-2}	ND	7.30X10^{-3}	ND	7.10X10^{-3}	14.6	ND	ND	ND	5.40	7.00X10^{-4}	20.0
	Site 2	ND	5.00X10^{-4}	ND	4.10X10^{-3}	ND	5.00X10^{-4}	1.60	ND	ND	ND	19.8	2.20X10^{-3}	21.4
	Site 3	ND	1.00X10^{-4}	ND	5.00X10^{-4}	ND	6.00X10^{-5}	0.60	ND	ND	ND	1.20	4.00X10^{-4}	1.80

Table 11. Toxic equivalence (ngTEQ2005 g-1) of PCBs in *Oreochromisniloticus* across season.

Compounds	PCB-77	PCB-81	PCB-105	PCB-114	PCB-118	PCB-123	PCB-126	PCB-156	PCB-157	PCB-167	PCB-169	PCB-189	TTEQ
Wet	ND	ND	ND	5.46X10^{-3}	ND	2.04×10^{-3}	1.92×10^{1}	ND	ND	ND	7.50	2.11×10^{-2}	26.7
Dry	1.40×10^{-3}	1.98×10^{-2}	2.40×10^{-4}	3.78×10^{-3}	8.4×10^{-4}	3.60×10^{-4}	4.00×10^{-1}	ND	ND	ND	6.00×10^{-1}	6.00×10^{-5}	1.03

through ingestion, inhalation and dermal contact with sediment, RFD = Reference dose, RfDing = Chronic oral reference dose, RfCinh = Chronic inhalation reference concentrations, RfD-dermal = Chronic dermal reference dose = RfDing XABSGI through three exposure rout, CSFing = Chronic oral slope factor through ingestion, IUR = Chronic inhalation unit risk, CSF-dermal = Chronic dermal slope factor = CSFing/ABSGI and C = Concentration of PCBs in sediment respectively.

The Toxic Equivalency (TEQ) values of PCBs were determined by multiplying the detected concentration with the corresponding Toxic Equivalent Factor (TEFs) taken from WHO [25].

2.11. Potential ecological risk evaluation

The potential ecological risk (RI) showed the impact of all the PCBs pollutants present in the environment, which was measured quantitatively to classify the extent of the risk. The ecological risk factors (*Eri*) were quantitatively determined to express the potential ecological risk index with Equation (15) and (16).

$$RI = \Sigma \tag{14}$$

The RI can be classified as follows; RI < 150 indicates a low risk, 150 ≤ RI < 300 a moderate risk, 300 ≤ RI < 600 a considerable risk, where RI ≥ 600 shows a very high risk.

$$E_r^i = T^i x\, C_f^i \tag{15}$$

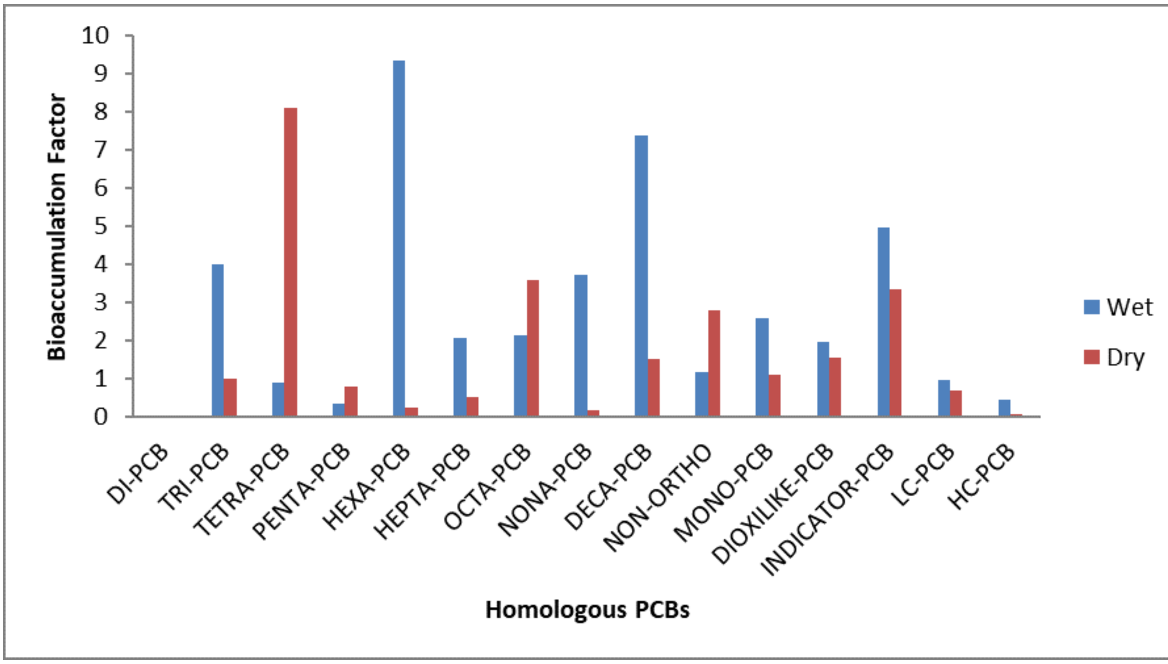

Figure 5. Sediment-based Bioaccumulation Factor (SBF) for *Oreochromis niloticus* across the seasons.

$$C_f^i = \frac{C^i}{C_n^i}, \tag{16}$$

Where C_f^i = the pollution index of a pollutant, C^i = the determined concentration of contaminants in sediments, *ni* = the global contaminant level in the pre-industrial sediments for each pollutant (10 μg/kg) and *Ti* = toxic response parameters for each pollutant in the study according to the standardization developed by Montuori *et al.* [16] and Han *et al.* [26]. The classification of the risk levels follows $Eri < 40$ is a minor ecological hazard, $40 < Eri$ is a medium ecological hazard, $80 \leq Eri \leq 40$ is a strong ecological hazard, $160 \leq Eri \leq 320$ is a very strong ecological hazard, $Eri > 320$ is pole strength ecological hazard [26].

2.12. Quality assurance and data analysis

Blank sample analysis with recoveries of surrogate standard for each sample was done as a monitoring measure for all the procedures. Each batch (4 per batch) was analyzed with a blank across all analytical phases. The mean recoveries for all the PCBs congeners were calculated. For extraction purposes, 10 g of muscle tissue and 2 g of sediments were used to extract the PCBs. Data analysis was done with Microsoft Excel Inc., USA by evaluating the Pearson's correlation analysis-assessing the relationship between the contaminant, the mean, median and standard deviation. The detection and quantification limits (LODs & LOQs) were determined with the ratio of 3 and 10 for signal-to-noise for all the concentrations, which ranged from 0.003 – 0.013 μg/kg (LODs) and 0.01 – 0.04 μg/kg (LOQs). From all the PCBs investigated, the blanks were totally cleared. Calibration was done via injection of standard solutions of mixed PCBs at 6 concentrations level.

3. Results and discussion

Several substances including heavy metals can contaminate the soil, but this work was on PCBs. Table 1 gives the estimation of human health risk assessment. Tables 2 and 3 are the statistical analysis of the concentrations of PCBs obtained from sediments and fish samples. The mean of Σ28-PCBs levels in the sediments was above 500 µg/kg guideline of Environmental Protection Agency [27] in some sites across seasons. These are 645, 1,790 and 3,977 µg/kg at Sites 1, 2 and 3 for the wet season and 1,219 µg/kg at Site 1 for the dry season. To compare the evaluated PCBs concentrations with reports from other studies is complex due to differences in PCBs congeners selected and the statistical methods used. However, comparing the total PCBs concentrations observed in this study (252 – 3,977 µg/kg) with other related studies showed that the result obtained in this study was relatively higher for sediments in Nigeria 0.026 – 0.198 µg/kg [28], China 1.75 – 92.27 µg/kg [29], China 16.15 – 477.85 µg/kg [30], and China 0.12 – 1.25 µg/kg [26]. There is a significant statistical difference of $p < 0.01$ and $p > 0.05$ between Sites across seasons. The concentrations of the 28- PCBs congeners detected across sites and seasons differ remarkably, suggesting that their occurrence is not from a common source of the dredging activities, but from other anthropogenic activities like domestic refuse disposal and chemical residues from nearby farmlands that enter the creek and tributaries through runoffs. This infers that there is possibility of degradation of some PCBs contaminants from Falcorp mangrove, agricultural activities [13] and from the polyvinyl chloride (PVC) pipes used for the dredging. The mean and the median values also showed significant differences ($p > 0.05$), which suggest that there is abnormal distribution of the data sets, indicating that they were log transfer [31]. The concentrations of PCBs in fish also exceeded the European guideline value of 220 µg/kg fresh weight across seasons and 1000 µg/kg guideline of the World Health Organization (WHO) and Food and Agricultural Organization (FAO) in the wet season [32]. The highest mean concentration in Oreochromis niloticus was observed in PCB-206 (344 ± 229 µg/kg) in wet the season and PCB-209 (213 ± 124 µg/kg) in the dry season. The total 28-PCB concentrations in fish were 1,688 ± 1,122 µg/kg for the wet season and 557 ± 323 µg/kg for the dry season. Out of the 28-PCBs congeners considered in *Oreochromis niloticus*, 16-PCB congeners were detected in the wet season while 19-PCB were detected in the dry season.

Comparing the total PCBs levels in *Oreochromis niloticus* with the regulatory criteria of PCBs in fish for human health protection in North America, including Canada with the value of 2000 µg/kg [32], which is higher than the values of this study, it can be recommended that fish in the studied sites is safe for consumption. However, the limits set by WHO, FAO and Switzerland (1000 µg/kg) suggest that the fish in the study areas are not safe for consumption. The higher mean in 28-PCBs observed in the wet season than in dry season suggests that PCBs contaminants enter the dredged river more than during the wet season. This could be ascribed to agricultural runoffs from nearby farmlands and dredging activities. The total PCBs concentrations in *Oreochromis niloticus* tissue are also higher than the maximum tissue concentrations of 100, 110 and 500 µg/kg of British Columbia, New York and Australia respectively [32].

3.1. Ecological risk assessment

The figures presenting the assessment of PCB levels in sediments across various sites and seasons, utilizing ecological risk factors and indices, are shown in Figures 1 to 4. Specifically, Figures 1 and 2 illustrate the ecological risk factors of sediments, highlighting variations across different sites and throughout the seasons.

The Di-PCB, Tri- PCB, Hexa-PCB and Deca-PCB showed medium and strong ecological risk factors based on the classification of $40 \leq Eri < 80$ and $80 \leq Eri < 160$. Octa-PCB display very strong ecological risk factor ($160 \leq Eri < 320$), the other homologous PCBs are in the category of ($Eri \geq 320$) in the wet season. In the dry season across sites, the homologous PCBs are in the categories of slight to very strong ecological risk factors. Figures 3 and 4 reveal the ecological risk index across the sites and seasons. In the wet season, RI shows a very high-risk index based on the classification of RI ≥ 600 and in the dry season, Sites 1 and 2 show low and moderate risk index, while Site 3 was in the category of very high ecological risk index. The ecological risk calculations of PCBs concentrations from sediment samples obtained from the dredged river across the three Sites, with the method proposed by Hakanson [33] show that there was a medium to very high-risk factor and low to very high-risk index. These will rarely lead to positive biological effects and generate a high potential risk to the environment [26]. This may also be associated with local hydrogeological and geomorphological conditions [26, 30]. The PCBs ecological risk factors obtained from sediment samples across sites are high majorly in wet season and low in dry season, hence attention is needed to avoid long term accumulation by taking appropriate measures, so as to reasonably evacuate the old dredging pipes undergoing deterioration and to monitor the indiscriminate dumping of refuge and the use of pesticides along the nearby farmlands.

Tables 4 – 7 are the results of the correlation analysis of Sites 1, 2 and 3 across seasons for sediments and *Oreochromis niloticus*. As can be seen from the Tables, there is a strong positive correlation between PCB-114 and PCB-77, PCB81, PCB-105 (r_2= 1.00, 0.99 & 1.00, $p < 0.01$) and also between PCB-126 and PCB-77, PCB-81, PCB-105, PCB-114, PCB-123 (r_2 = 0.95, 0.97, 0.95, 0.93 & 0.96, $p < 0.01$). Similarly, a strong positive correlation occurred between PCB- 157 and PCB-118, PCB-156 (r_2 = 1.00, 1.00, $p < 0.01$) in the wet season for the sediments, while in dry the season, there was a strong positive correlation between PCB-81 and PCB-77 (r_2 = 0.93), between PCB-105 and PCB-77, PCB-81 (r_2 = 0.91, 1.00, $p < 0.01$), between PCB-114 and PCB81, PCB-105 (r_2 = 0.86, 0.89, $p < 0.01$), between PCB-126 and PCB-77, PCB-81, PCB-105, PCB114,PCB—118 (r_2 = 0.92, 1.00, 1.00, 0.87, 0.72, $p < 0.01$), between PCB-123 and PCB-77, PCB81, PCB-105, PCB-114, PCB-118 (r_2 = 0.92, 1.00, 1.00, 0.87, 0.72). In fish, there was significant positive and negative correlation of (r_2 = 100 & -100, $p < 0.01$) across the detected PCB congeners. The positive correlation observed indicates that there is positive relationship between the contaminants of PCBs congeners detected and their persistence has a significant effect on the environment. The negative correlation showed that their persistence in the sediment and fish does not have any significant effect in them [34].

3.2. Human health risk

Table 8 is the incremental life-time cancer (ILCR) risk values for sediments obtained from the dredged river across sites and seasons. The total cancer risk (TCR) values ranged from 6.10 x 10-3 to 1.47 x 10-2 for children and 6.30 x 10-4 to 1.11 x 10-3 for adults across sites in the wet season, while in the dry season, the values ranged from 1.04 x 10-3 to 2.99 x 10-2 for children and 7.80 x 10-5 to 5.61 x 10^{-1} for adults across sites. The TCR across sites and seasons for adults were lower than those for children. This might be as a result of high tendency of dermal contact of children with sediments. Considering the categories of life time cancer risk by New York State Department of Health [35] (≥ 10-4 = very high, > 10-3 to 10-1 = high, ≥ 10-4 to 10-3 = moderate, ≥ 10-6 to 10-4 =low and ≤ 10-6 = very low), the PCBs cancer risks observed in this study showed moderate to very high risk for both children and adults across the sites and the seasons. This implies that the sediment samples from the dredged river were subject to generation of cancer in humans via dermal contact. The calculated PCBs hazard index (HI) values in this work are in Table 9. The HI values range from 5.75 x 10-1 to 6.04, 7.18 x 10-2 to 1.74 for children and 6.20 x 10-2 to 1.09 x 10-1, 7.74 x 10-3 to 1.86 x 10-1 for adults across the sites and seasons. The HI values for children were >1 in some sites across seasons, suggesting that there was probability of health effect with PCBs exposure across the sites. For adults, the HI values were < 1 indicating that there was no carcinogenic risk i.e., there was no health effect associated with

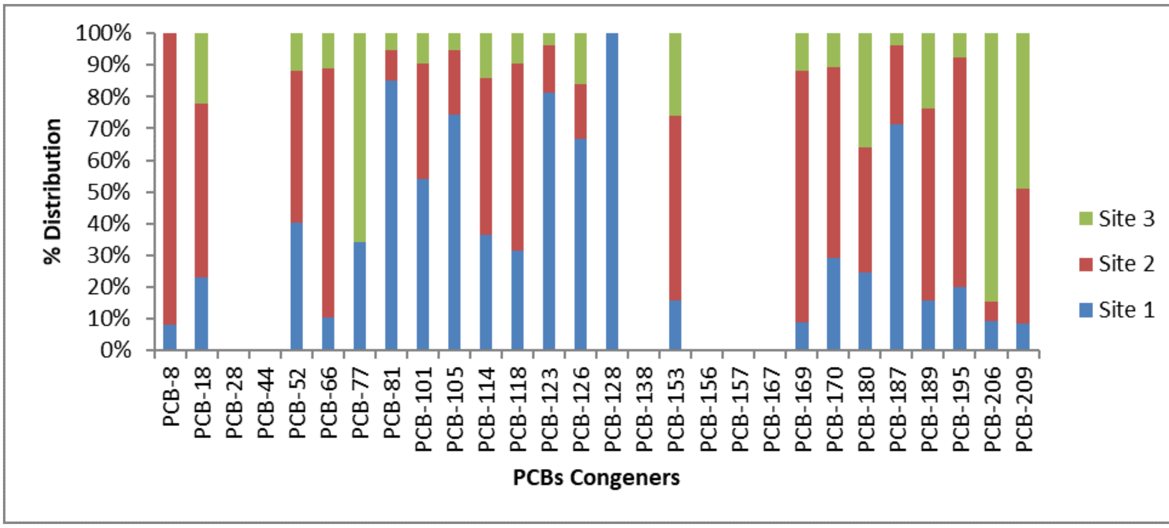

Figure 6. Percentage distribution of PCBs congeners in sediments across Sites in the dry seasons.

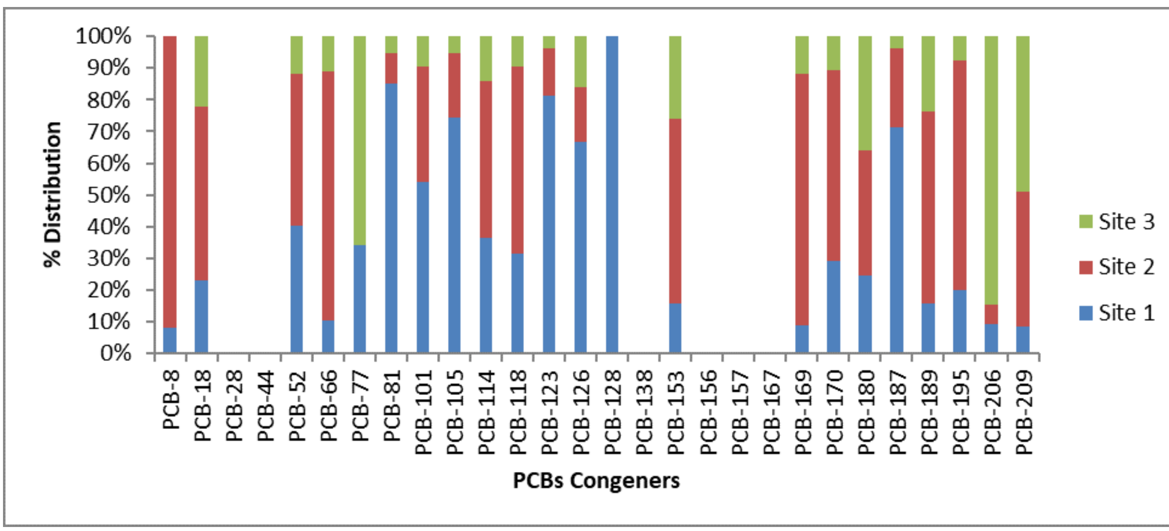

Figure 7. Percentage distribution of PCBs congeners in sediments across Sites in the wet seasons.

PCBs exposure across sites and seasons. Table 10 is the toxic equivalent quotients (TEQs) of PCBs obtained from sediments in the dredged river, which is used to evaluate the human exposure and health through food intakes contaminated with the dioxin-like PCBs congeners. The values in sites 1 and 2 (34.0 and 25.0 µg/kg) across the wet season exceed 25.0 pg/g of high risk (HR) to sensitive species [26], indicating high ecological risk of dioxin-like (DL)-PCBs in the sediments. At site 3 (wet season) and across all sites in the dry season, the values were below 25.0 pg.g of HR limit which signifies low ecological risk at these sites and seasons. The values in this study are higher in contamination level than those of Montuori *et al.* [16] (0.002 – 0.33 ng/g) and Han *et al.* [26] (0.0014 –9.663 ng/g). This might be ascribed to the season of sample collection. Since the TEQs values in this study for sediments were very high, the PCBs toxicities of the dredged river will adversely affect the ecological environment and human health via biomagnifications [16].

Table 11 shows the TEQs values of PCBs obtained from *Oreochromis niloticus* samples across the seasons. The values were 26.7 µg/kg (wet season) and 1.03 µg/kg (dry season). The values obtained in wet season exceeded the guideline recommended for fish (not consumable) with concentration greater than 25.0 nglTEQ/kg on wet weight basis [32]. Due to the bio accumulative nature of dioxin-like PCBs, there is complication in their regulation for the protection of wildlife. As a result, the Canadian Council of Ministers of Environment proposed tissue residues guideline of 50 nglTEQ/kg for aquatic life protection [32]. Comparing this to the values obtained, it implies that the fish in the studied environment are safe for consumption.

Figure 5 showcases the Sediment-based Bioaccumulation Factor (SBF) for *Oreochromis niloticus*, detailing seasonal variations. Furthermore, the distribution percentages of PCB congeners in sediments, differentiated by site and season, are depicted in Figures 6 and 7.

The bioaccumulation factors for fish are displayed in Figure 5. From the profile, it is observed that the optimal level of

SBF is in the lower molecular PCBs congeners (Tetra and Hexa) across seasons. This might be ascribed to the lower degradation of these congeners, thereby accumulating to a high level in aquatic ecosystem leading to bioaccumulation. This might also be anchored on easy transfer of least chlorinated compounds to aquatic organisms, due to their easy penetration in cell membrane [2, 36] as a result of their reduced number of chlorine atoms. This result corroborates with the study of Ernesto *et al.* [2]. Next to hexa on the level of bioaccumulation in the study are the Deca and Indicator-PCBs, which are known to have a high molecular weight and bioaccumulate because they have higher partition coefficient, indicating high lipophilicity. They tend to accumulate in lipid and across the trophic chain.

The percentage distributions of the 28-PCBs congeners are shown in Figures 6 and 7. Across sites, the percentage values range from below detectable limit to 19.3 % (site 1), from below detectable limit to 13.9 % (Site 2) and from below detectable limit to 70.8 % (site 3) in dry season. While in the wet season, the values range from below detectable limit to 21.4 % (Site 1), from below detectable limit to 21.6 % (Site 2) and from below detectable limit to 32.7 % (Site 3). The highest percentage was observed in high molecular weight PCBs (PCB-206) and low molecular weight (PCB-105) in dry and wet season. The dominance high PCBs in dry season at higher molecular weight could be due to weak volatility and solubility as a result of increase in the number of chlorine atoms and strong binding capacity of the PCBs contaminants. The highest observed in low molecular weight PCBs at wet season might be due to high volatility and solubility as a result of decrease in the number of chlorine atoms [16].

4. Conclusion

The ecological risk and human health risk from the sediments and fish from dredged tributaries and creeks of River Ethiope from PCBs concentrations across sites and seasons are of moderately to very high contamination and also moderately to very high cancer risk for adults. No carcinogenic risk for children was observed. This is because children are not fully involved in the dredging activities. The Pearson correlation coefficient analysis showed evidence of a strong positive correlation in both sediments and fish samples and the bioaccumulation levels in fish are higher in the lower molecular weight PCBs across seasons. The sources of PCBs in the tributaries of River Ethiope from the correlation point of view were majorly from the dredging activities. These results generate ideas for management and control of pollution at the tributaries of River Ethiope.

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