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Polychlorinated Biphenyls Contamination of Drinking Water Source of the Pra River: Congener Distribution and Associated Risk

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Abstract

PCB pollution of the environment and the associated ecological and health impacts have been of great concern since the 1970s, yet PCBs persist in the environment. In this study, the spatial distribution and impacts of PCBs at twenty different locations in the drinking water source of the Pra River in Ghana were determined in the wet and dry seasons. Extraction of PCBs was performed using solid-phase extraction and a solvent extraction system, respectively, and the analytes were identified and quantified by gas chromatography. PCBs 18, 153, and 180 were the dominant congeners in the river water. The mean Σ_{12}PCBs concentration ranges in the wet season were 0.04 - 11 ng/L, 0.33 - 69 ng/g, and 0.72 - 153 ng/L. The dry season PCB concentrations were higher at all locations. The Pra River has low levels of PCB pollution at all the sites compared with other regions. In general, the hexa-chlorinated PCBs biphenyls were the most dominant homologue. The estimated probabilistic PCB cancer risk is much lower than the acceptable risk of one chance in a million (1.0E−06) of exposed persons to suffer the risk of cancer. The mean TEQs for fish, birds and humans/mammals were very low, and none of the samples exceeded the environmental quality standard of 1.0 pg-TEQ/L. The PCB concentration suggests that the river water does not pose a danger to aquatic organisms and human health.

Subject Areas

Environmental Sciences

Keywords

Pra River Water, PCBs, Pollution, Toxicity, Human and Wildlife

1. Introduction

Polychlorinated Biphenyls (PCBs) are one of the Persistent Organic Pollutants (POPs) that has been used as dielectric fluids in transformers and capacitors, as plasticizers and in carbonless copy paper, hydraulic fluids and as additives in paint [1]. Technological polychlorinated biphenyls consist of a mixture of homologous and/or isomeric chlorinated biphenyls. The compounds/congeners have various degrees of chlorination between 21% and 68%. PCBs with an average content of 42 to 54% chlorine during the production and use [2]. These toxic compounds have been banned in transformers since July 1979. However, in England, Scotland, Wales and EU transformers whose oil contains more than 0.005% (500 pm) PCBs but less than 0.05% (500 ppm) and a total PCB volume of 50 ml are currently being used, and will be held until the end of their useful life. These facilities containing PCBs are expected to be decommissioned or disposed of by 31st December 2025 [3].

Globally, PCBs exist in various environmental and human matrices because of their widespread production, use and their propensity for long-range atmospheric transport [4]-[8]. Also, PCB compounds are generated from thermal processes such as waste incineration, plasticizer burning, and vehicle exhaust emissions [9] [10]. The continued dispersive released from primary and diffuse secondary sources has led to serious contamination [11] [12]. In recent years, this class of pollutants have become an increasing concern in aquatic environments [13]-[15], especially in sediments of rivers, lakes, and seas. PCBs enter aquatic systems through various paths, via air deposition, wastewater discharge and runoff and are transported from one media to another through the atmosphere, rainfall, or land runoff [16] [17]. PCBs have been determined in several environmental media worldwide [18]-[22], these toxic compounds have been identified amongst pollutants that pose ecological risk [23] [24], and have contributed significantly to disease as a result of widespread human exposure to these pollutants [25]-[31].

Though PCBs were never produced in Ghana, yet the usage of PCB oils in electric transformers and capacitors, storage and leaks have led to the contamination of the Ghanaian environment with PCBs. Surrounding areas of pre-1972 transformers country wide have been identified as PCB contaminated sites in Ghana [32] [33]. Another probable source of PCBs is one of the world's largest ships' graveyards, just south of Nouadhibou Mauritania, where hundreds of wrecks have been grounded and left to decay [34]-[36].

Reports indicate the occurrence of PCBs in environmental samples in Ghana, such as sediments, soils, oysters, and lagoon water and sediments, electronic waste and air samples [37]-[40]; and in breast milk at levels that might pose health risk to children [41] [42]. The presence of these toxic compounds in the Pra River, a major drinking water source in the central region of Ghana, may pose adverse health and ecological effects. The sediment PCB profile and potential hazards due to exposure to Pra River sediments have been studied [43]. However, there is no report on the incidence of PCBs in the drinking water source of the Pra Rives. The potential health effects and ecological impacts due to the use of the river water have not been adequately investigated. This paper presents the PCB distribution of drinking water sources of the Pra River, the potential health risks, and impacts on humans and wildlife.

2. Study Area and Methodology

2.1. Sampling

The Pra River is a river in Ghana, the easternmost and the largest of the three principal rivers that drain the area south of the Volta divide. Rising in the Kwahu Plateau near Mpraeso in the eastern region then flowing southward for 240 km (149.129 miles) into the Gulf of Guinea east of Takoradi in the western region [44]. River water samples were collected along the western and central region sections of the river (**Figure 1**). The locations were chosen based on their proximity to settlements, unregulated dumping sites, local palm oil processing factories and urbanized water channels into the river. This choice guarantees that the analysis includes both high-risk regions and more general regional patterns in the distribution of PCBs. All samples were put in an icebox, transported to the laboratory, and stored in a freezer at −20˚C. The sampling locations are designated as P1 - 20 (**Table 1**).

2.2. Chemicals

Standard PCB mixture of "CEN PCB Congener Mix 1" (IUPAC No.'s 18, 28, 31, 44, 52, 101, 118, 138, 149, 153, 180, and 194) (Supelco, USA), which comprises 10 μg/L of each component in heptane was employed for the six-point calibration technique. For the analysis, an internal standard solution was added to samples before extraction and it consisted of 50 ng/L 4,4'-dibromobiphenyl, Supelco (USA) in n-heptane. First-grade quality solvents, namely n-heptane, methanol and acetone were also used for the analytical procedure, respectively. These solvents were procured from the Wako Pure Chemicals. The laboratory blanks used were 18Ω water purchased from Sigma-Aldrich.

2.3. Extraction and Cleanup of PCBs

Water samples from the Pra River basin were spiked with 4,4'-dibromobiphenyl as internal standard and extracted with n-hexane/acetone (1:1 v/v). All eluates from the Silicagel and Florisil cartridges (Sep-Pac1 Vac 6 cc, Waters) were

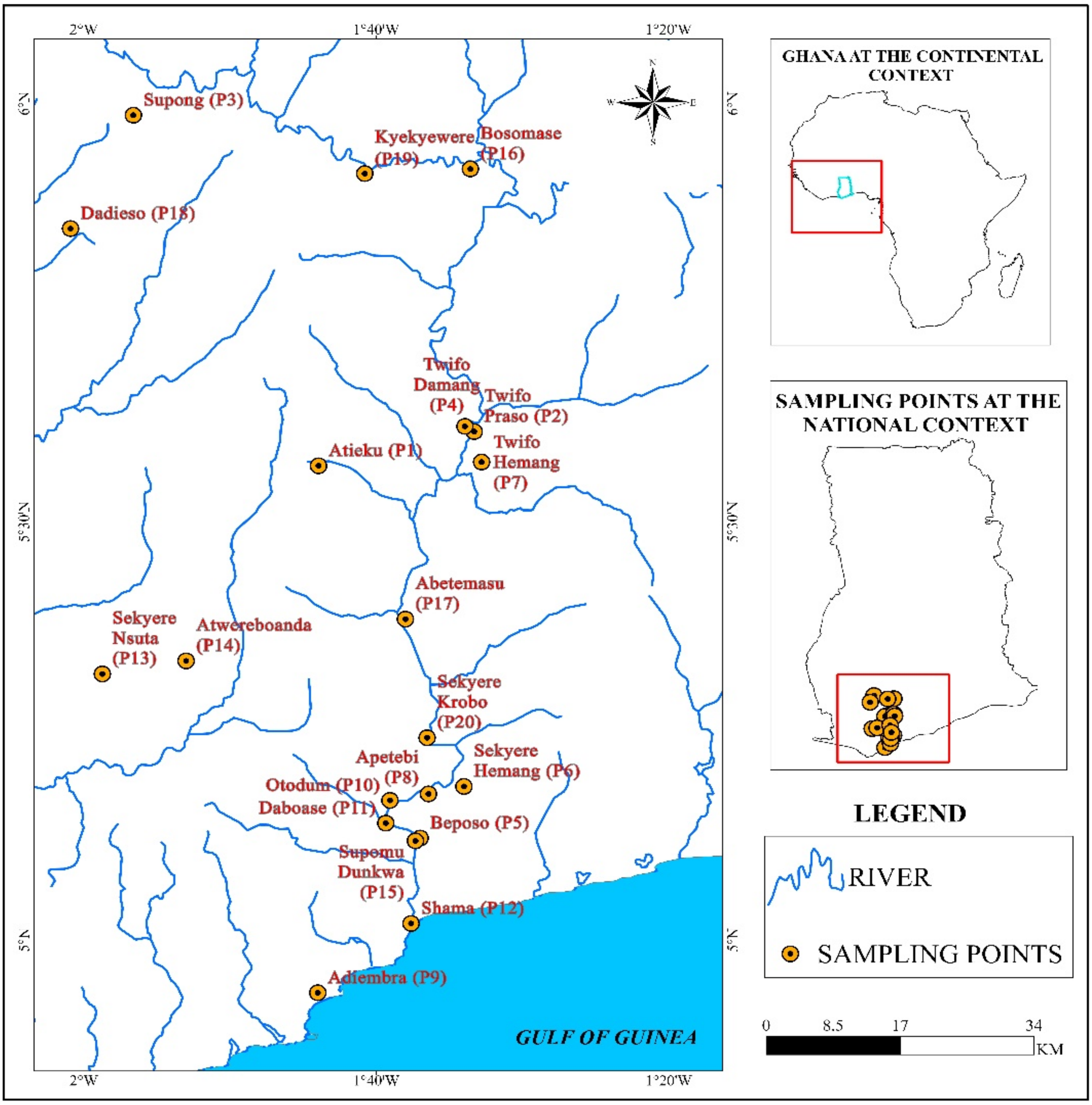

Figure 1. Study area showing sampling sites [43].

Table 1. Sampling locations with their coordinates.

Sampling Site Name	Sampling Site Number	Coordinates (N, W)
Atieku	P1	5.56667, −1.73240
Twifo Praso	P2	5.36599, −1.32599
Supong	P3	5.98580, −1.94344
Twifo Damang	P4	5.61379, −1.56545
Beposo	P5	5.12163, −1.61658

Continued

Sekyere Hemang	P6	5.183333, −1.56667
Twifo Hemang	P7	5.57118, −1.54650
Apetebi	P8	5.17440, −1.60710
Adiembra	P9	4.93673, −1.73319
Otodum	P10	5.16667, −165100
Daboase	P11	5.13850, −1.65910
Shama	P12	5.01948, −1.62690
Sekyere Nsuta	P13	5.31778, −1.97879
Atwereboanda	P14	5.33333, −1.88333
Supomu Dunkwa	P15	5.118269, −1.62188
Bosomase	P16	5.92160, −1.55930
Abetemasu	P17	5.38333, −1.63333
Dadieso	P18	5.85025, −2.01507
Kyekyewere	P19	5.91599, −1.67971
Sekyere Krobo	P20	5.24156, −1.60860

concentrated in volume to 0.5 ml under nitrogen gas flow. Sample extraction and chemical analyses were performed following previous methods [45]-[47].

2.4. Analysis of PCBs

The gas chromatography-mass spectrometry (GC-MS) (Shimadzu QP-2010 Ultra Japan equipped with flame ionization detector), analysis of the samples was conducted using a DB-5MS capillary column of length 30 m (0.25-μm internal diameter and 0.25-μm film thickness). Helium was used as a carrier gas with a flow rate of 0.72 mL/min and a total flow of 31.8 mL/s and a linear velocity of 32.2 cm/s at purge flow of 3.0 mL/min using splitless injection mode. The injection and detector temperatures were set at 150˚C and 320˚C, respectively. The oven temperature was programmed to increase from 80˚C to 310˚C at a rate of 40˚C/min and then from 310˚C to 320˚C at a rate of 2˚C/min. A 1-μL injection volume was used. PCB homologues were determined by single ion monitoring (SIM).

The following quantifying and qualifying ions were monitored simultaneously: m/z values were 256 and 258 for trichlorobiphenyls; 290 and 292 for tetrachlorobiphenyls; 326 and 328 for pentachlorobiphenyls; 360 and 362 for hexachlorobiphenyls; 394 and 396 for heptachlorobiphenyls; and 312 for 4,4’-dibromo biphenyl. All the quality control steps were followed. Laboratory blanks were subjected to the same analytical procedures as applied to the original samples. However, no significant peaks were observed for the analytes of interest.

The identification of PCB congener peaks was based on their relative retention times (RRTs) and on intensity ratios of the monitored ions for quantification using gas chromatography- mass spectrometry (GC-MS). The target individual PCBs were quantified using the internal standards calibration method

2.5. Health and Ecological Risk

2.5.1. Health Risk Estimation

The carcinogenic risk associated with a potential receptor exposure to PCBs in water was estimated using the linear low-dose model [48]. This model assumes that there are multiple stages for cancer, and it is only applicable at low risk levels (*i.e.*, estimated risks < 0.01). The assessment risk was based on ingestion and dermal contact.

2.5.2. Ecological Risk Analysis

TEQ method quantitatively was used to assess the ecological-toxicity. The TEQ values of the dioxin-like PCB118 were obtained by multiplying the WHO-TEF of the PCB 118 congener for humans and mammals, fish, and birds by the PCB concentrations [49] [50].

2.6. Quality Control

Procedural blanks, matrix-spiked blanks, duplicate samples, internal standards and surrogate standard were analyzed with the field-sampless. The recoveries of spiked 12 indicator polychlorinated biphenyl (PCB) congeners, nos.18, 28, 31, 44, 52, 101, 118, 138, 149, 153.194 and 180; PCBs from water were 71.20% - 105.7%, with precision as relative percent difference (RPD) range of 2.41% - 10.79%. The limit of detection (LODs) determined for all congeners were in the range of 0.0047 - 0.057 ng/L, and the LOQ from 0.017 - 0.098 ng/L. The calibration linearity was good and ranged from $r^2 = 0.992 - 0.998$, in line with the acceptance criteria of $r \leq 0.995$ (or $r^2 \leq 0.990$) for GC analysis of environmental samples.

The ∑PCB concentration was defined as the sum of 12 PCB congeners. Data analyses were performed using Excel, SPSS 14.0 (SPSS Inc., Chicago, 1L, USA). For similarity comparisons cluster analysis (CA) and principal component analysis (PCA) were conducted on the mean concentrations of PCB congeners in water, in addition to the use of coefficient of determination, R^2. Details on the interpretation of R^2 and the multivariate analysis are found in Bentum *et al.* [43].

3. Results and Discussion

3.1. PCBs Recovery Rates

The recovery rates of polychlorinated biphenyls (PCBs) during solid-phase extraction (SPE) and solvent extraction processes ranged from 75.20% to 105.70%. These values fall within the acceptable limits based on standard analytical guidelines, where recovery rates between 70% and 120% are generally considered satisfactory for environmental sample analyses [51].

The recovery range observed in this study closely aligns with previously reported values of 72% - 108% for PCBs extracted from water samples using SPE

[52]. Similarly, the recovery range of 70% - 110% reported by Guo in 2017 [53], using a combination of SPE and GC-MS for PCB analysis in soil and sediment samples, is comparable to the findings of the current study. Another study investigating water samples reported a slightly broader recovery range of 68% - 112% [54], with some values falling just below the 70% threshold, indicating minor deviations when compared to the present study.

3.2. PCBs Distribution in the Pra River

The PCB congener detection frequency of various congeners is shown (**Figure 2**). The detection frequency ranged from 50% - 100%. The predominant congeners in both seasons were PCB 18, 153 and 180 (**Table 4**). In all seasons these congeners recorded detection. PCB 31 recorded 100% detection in the dry season only. These congeners were thus detected in samples from all the sampling sites. PCB 194 recorded the least percentage detections of 50%. The PCB congener distribution for the wet and dry seasons are shone (**Table 2** and **Table 3**).

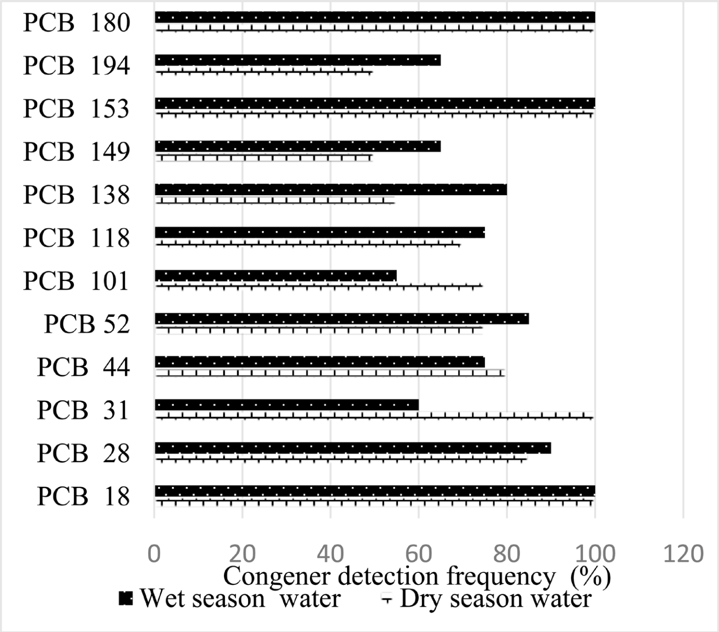

Figure 2. Proportions of PCB congeners detect in the river Pra Basin water.

Table 2. PCBs distribution in river water at sampling sites in the minor wet season.

	PCB congener concentration (ng/L)												
Site	18	28	31	44	52	101	118	138	149	153	194	180	ΣPCBs
P1	0.039	0.033	0.05	nd	0.036	0.16	0.67	0.038	nd	0.049	nd	0.16	1.235
P2	0.61	0.017	n.d	0.31	0.044	nd	nd	0.083	0.084	0.081	nd	0.56	1.789
P3	0.55	0.018	0.035	0.35	0.089	0.21	0.064	0.02	0.056	0.27	0.12	0.89	2.672
P4	0.58	0.021	0.078	nd	0.075	nd	0.087	0.025	0.045	0.74	0.034	0.32	2.005
P5	0.088	0.065	0.061	0.24	0.079	nd	0.083	0.029	0.065	0.18	0.056	0.11	1.056

Continued

P6	0.14	0.065	0.077	0.26	0.025	0.26	0.08	0.047	0.073	0.04	0.034	0.2	1.301
P7	0.16	0.02	0.045	nd	0.047	nd	0.085	0.055	0.047	0.18	0.098	0.03	0.767
P8	0.054	0.028	n.d	nd	0.010	0.15	0.023	0.023	nd	0.13	0.035	0.13	0.583
P9	0.094	0.025	n.d	nd	0.008	nd	0.095	0.011	nd	0.14	0.046	0.11	0.529
P10	0.064	0.055	n.d	0.38	0.092	0.32	0.097	0.032	nd	0.37	nd	0.2	1.61
P11	0.23	0.078	0.034	0.67	nd	0.54	nd	0.12	0.075	0.073	0.008	0.12	1.948
P12	0.13	n.d	n.d	0.56	0.038	nd	0.023	nd	0.084	0.091	0.061	0.65	1.637
P13	0.046	n.d	n.d	0.85	nd	nd	0.087	0.023	0.045	0.082	nd	0.54	1.673
P14	0.085	0.027	0.034	nd	0.17	0.86	nd	nd	nd	0.39	nd	0.34	1.906
P15	0.14	0.14	0.078	0.93	nd	0.05	nd	nd	nd	0.78	0.097	0.78	2.995
P16	0.041	0.067	0.089	0.4	0.064	nd	0.041	0.21	0.053	0.45	0.032	0.087	1.534
P17	0.25	0.02	n.d	nd	0.083	0.45	0.067	0.081	0.043	0.54	nd	0.012	1.546
P18	0.13	0.023	n.d	0.36	0.032	0.12	nd	0.12	0.045	0.097	0.065	0.87	1.862
P19	0.058	0.045	0.035	0.45	0.067	nd	0.043	nd	0.057	0.64	nd	0.42	1.815
P20	0.09	0.078	0.054	0.93	0.041	0.075	0.045	0.047	nd	0.76	0.008	0.13	2.258
Σ	3.579	0.825	0.67	6.69	0.999	3.195	1.59	0.964	0.772	6.083	0.694	6.659	32.720

Table 3. PCBs distribution in river water at sampling sites in the dry season.

	PCB congener concentration (ng/L)												
Site	18	28	31	44	52	101	118	138	149	153	194	180	ΣPCBs
P1	0.081	0.056	0.061	0.086	0.63	0.059	nd	0.13	nd	0.67	0.34	0.17	2.283
P2	0.098	0.073	0.56	0.056	0.58	0.065	0.087	0.083	0.078	0.08	0.067	0.45	2.277
P3	0.07	0.05	0.056	0.057	0.81	nd	nd	nd	0.057	0.089	nd	0.84	2.029
P4	0.098	0.081	0.19	0.34	0.68	nd	0.095	0.059	0.086	0.14	0.084	0.25	2.103
P5	0.81	nd	0.23	nd	0.56	nd	0.091	nd	0.76	0.48	0.096	0.11	3.137
P6	0.037	nd	0.094	0.048	0.93	nd	0.25	nd	0.095	0.09	nd	0.87	2.414
P7	0.026	0.042	0.057	0.095	0.2	nd	0.088	0.870	0.087	0.76	1.04	0.15	3.415
P8	0.083	nd	0.083	nd	0.98	0.089	0.29	0.230	nd	0.13	0.082	0.56	2.527
P9	0.51	nd	0.052	0.059	nd	nd	nd	nd	nd	0.76	0.94	0.11	2.431
P10	0.45	0.23	0.062	1.39	0.76	0.075	0.55	0.064	nd	0.097	nd	0.29	3.968
P11	0.37	0.52	0.34	nd	nd	0.083	0.16	nd	nd	0.84	0.54	0.65	3.503
P12	0.28	0.44	0.095	0.097	nd	0.062	0.78	nd	nd	1.67	0.069	0.95	4.443
P13	0.52	0.23	1.02	0.083	nd	0.079	0.86	nd	nd	0.86	nd	0.78	4.432
P14	0.089	0.28	0.081	0.2	0.27	0.12	0.085	nd	nd	0.072	nd	0.94	2.137
P15	0.78	0.056	0.46	0.13	nd	0.076	0.079	nd	nd	0.08	nd	0.56	2.221

Continued

P16	0.069	nd	0.47	nd	0.64	0.18	nd	0.210	0.087	1.24	nd	0.083	2.979
P17	0.076	0.051	0.093	0.68	0.83	0.064	nd	0.290	nd	0.84	nd	0.29	3.214
P18	0.73	0.32	0.059	0.89	0.82	0.98	nd	0.460	0.66	0.47	nd	0.48	5.869
P19	0.97	0.054	0.098	0.067	0.69	0.054	0.62	0.93	0.67	0.84	nd	0.42	5.413
P20	0.89	0.064	0.14	0.075	0.71	0.085	0.89	0.0471	0.84	0.18	0.008	1.32	5.249
Σ	7.03	2.547	4.301	4.353	10.09	2.071	4.925	3.3731	3.42	10.388	3.266	10.273	66.044

Table 4. Distribution Statistics of PCB Congener Profile of Water Samples from the Pra River Basin.

		Concentration of PCB Congeners (ng/L) in Water Samples												
		0.18	28	PCB 31	44	52	101	118	138	149	153	194	180	Σ_{12}PCB
August-December	Mean	0.29	0.11	0.15	0.291	0.28	0.13	0.16	0.11	0.12	0.43	0.11	0.42	2.60
	Std	0.29	0.15	0.23	0.35	0.34	0.22	0.25	0.21	0.25	0.39	0.23	0.33	1.33
	CV	102.04	130.0	161.16	118.7	121.7	167.24	155.52	96.03	198.68	89.72	214.42	77.25	51.09
	Median	0.13	0.06	0.06	0.11	0.08	0.06	0.08	0.03	0.05	0.32	0.03	0.33	2.30
	Min	0.03	nd	nd	nd	nd	nd	nd	nd	nd	0.04	nd	0.01	0.77
	Max	0.97	0.52	1.02	1.39	0.98	0.98	0.89	0.93	0.86	1.67	1.04	1.32	6.02
	PCB%	10.94	4.34	5.58	11.19	10.65	5.06	6.26	4.07	4.77	16.66	4.17	16.27	100.00
August-September	Mean	0.22	0.04	0.03	0.34	0.05	0.160	0.08	0.04	0.04	0.35	0.05	0.33	1.73
	Std	0.23	0.03	0.03	0.32	0.04	0.23	0.14	0.05	0.03	0.30	0.08	0.29	0.71
	CV	107.94	81.24	95.6	95.95	83.49	144.18	180.62	115.74	81.53	84.81	156.19	85.76	41.27
	Median	0.130	0.028	0.035	0.330	0.043	0.063	0.06	0.03	0.045	0.225	0.03	0.20	1.66
	Min	0.01	nd	nd	nd	nd	nd	nd	nd	nd	0.01	nd	0.01	0.03
	Max	0.85	0.14	0.09	0.93	0.17	0.86	0.67	0.21	0.08	0.97	0.34	0.89	6.20
	PCB%	12.57	2.39	1.938	19.35	2.89	9.243	4.60	2.51	2.233	20.123	2.892	19.26	100.00
November-December	Mean	0.35	0.19	0.26	0.25	0.51	0.10	0.25	0.17	0.21	0.52	0.17	0.51	3.47
	Std	0.33	0.18	0.29	0.37	0.35	0.21	0.31	0.28	0.33	0.46	0.31	0.35	1.23
	CV	93.61	97.39	113.55	150.15	69.59	204.38	126.12	165.44	157.07	87.84	187.29	67.66	35.40
	Median	0.19	0.08	0.10	0.09	0.64	0.07	0.09	0.05	0.03	0.48	0.03	0.47	3.09
	Min	0.03	nd	0.05	nd	nd	nd	nd	nd	nd	0.07	nd	0.08	2.03
	Max	0.97	o.97	1.02	1.39	0.98	0.98	0.89	0.93	0.86	1.67	1.04	1.32	6.02
	PCB%	10.13	5.32	7.40	7.13	14.52	2.98	7.09	4.86	6.04	14.95	4.80	14.79	100.00

The mean PCB congener concentration in the surface water were generally lower in the wet season, than the drying season (**Figure 3**) Apart from PCB 44 (0.34 ng/L) and PCB 101 (0.16 ng/L) that recorded relatively higher concentrations in the wet seasons, all other congeners recorded lower concentrations in the wet season. The mean congener concentrations in the wet season ranged from 0.03 ± 0.03 ng/L (PCB 31) - 0.35 ± 0.30 ng/L for PCBs 153, and decrease in the order congener 153 > 44 > 180 > 18 < 101 > 118 > 194 = 52 > 149, 138, 28 > 31.

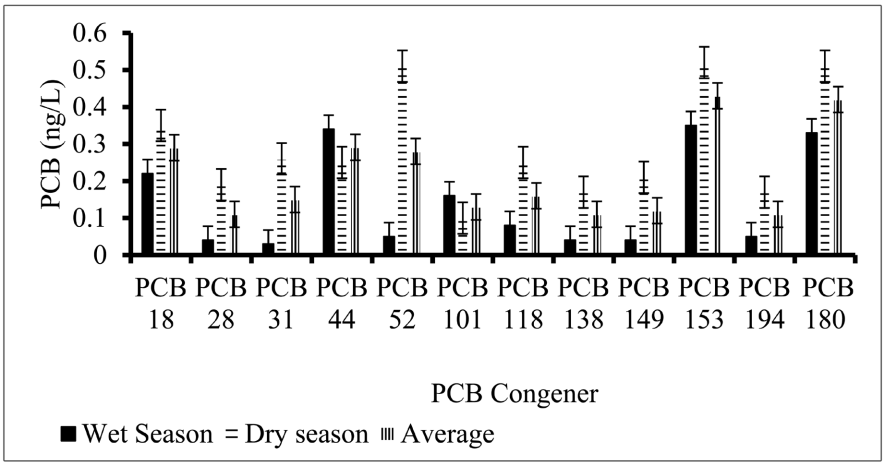

Figure 3. Average PCB congener profile of the Pra river water.

The concentrations in the dry season ranged from 0.10 0 ± 0.21 ng/L (PCB 101) - 0.52 ± 0.46 ng/L (PCB 153), and followed the order by PCB congeners 44 >153 >105 >180 >70 >101 >28 > 52. The total PCB concentration for the dry and wet seasons were respectively 3.47 ± 1.23 ng/L and 1.7 ± 0.71.ng/L. The overall mean was ΣPCB was 2.60 ± 1.33 ng/L. Significant variation in the PCB distribution, 51.09% as revealed by the coefficient of variation. The variation recorded for the congener distribution in the dry and wet seasons were respectively 67.66% - 204.38% and 81.24% - 180.62%; with respective means 35.40% and 41.27%. However, along the River Nile, Egypt, PCB 138 (10.119 μg/L) has been reported as the highest congener concentration and PCB118 (1.009 μg/L) the lowest [55]. Also, the levels of PCB 28 (0.018 - 0.042 μg/L), PCB52 (0.006 - 0.015 μg/L) and PCB 101 (0.001 - 0.039 μg/L) reported in Mexico City [56] were found to be lower than those recorded in the Pra River basin in this study.

The total PCB concentrations (Σ_{12}PCB) for the water from the sampling points in the dry season and wet seasons, is shown (**Figure 4**). The box plot (**Figure 5**) shows the ranges of Σ_{12}PCB for the dry season was higher than the wet season mean. Except for sites P3 and P15, which recorded higher PCB concentrations during the wet season, all other sites showed elevated concentrations in the dry season. Surface runoff during the wet season can transport PCBs into the river system from contaminated sources, and desorption from bottom sediments may also contribute to increased levels. The rise in PCB concentrations at sites P3 and P15 is likely due to this influx. In contrast, evaporation and reduced dilution during the dry season tend to concentrate PCBs in the water.

Additionally, anthropogenic activities further amplify these seasonal patterns. Contributing factors such as illegal mining (galamsey), discharges of engine and lubricating oils, improper waste disposal, and agricultural runoff create a complex, seasonally-driven variation in PCB distribution.

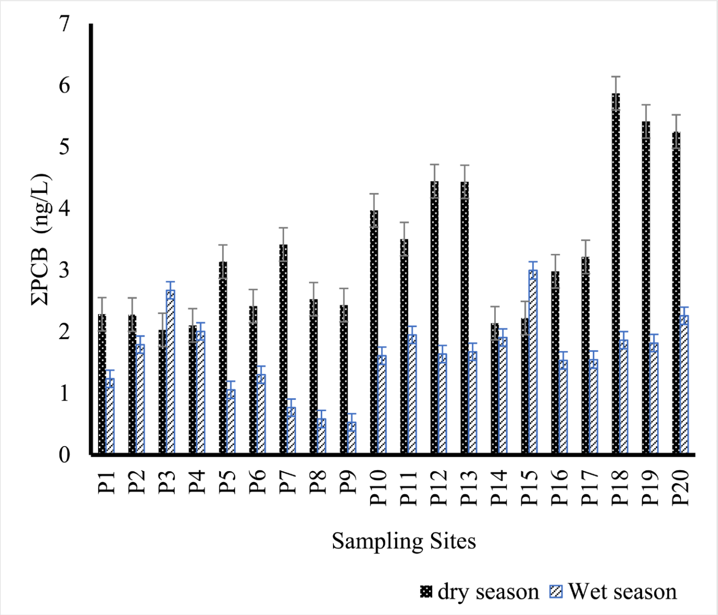

Figure 4. Total PCB levels in river water at the sampling sites.

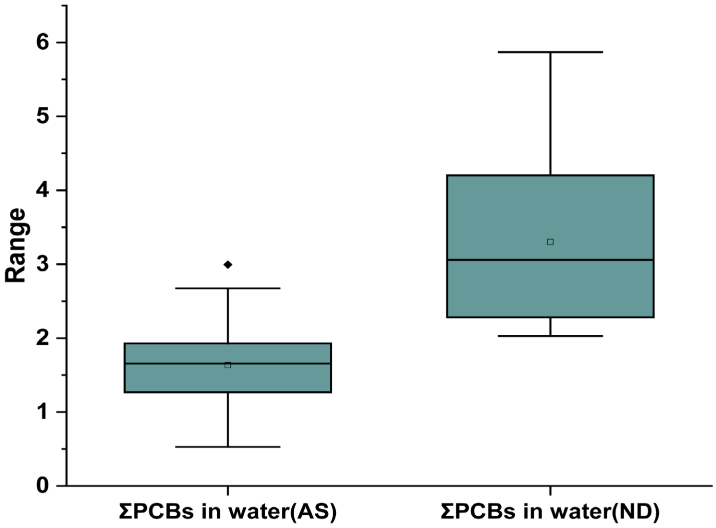

Figure 5. Box-plot of PCB distribution in water from the Pra River.

The highest concentration, ΣPCB, in the dry and wet season respectively were recorded at sites P18 (5.869 ng/L) and P15 (2.995 ng/L). The least concentrations were recorded at sites P3 (2.029 ng/L) and P9 (0.5285 ng/L) respectively in the dry and wet season. The means Σ_{12}PCB for the dry, wet seasons as well as the overall mean ΣPCB for the two seasons, 2.60 ± 1.33 ng/L, were all below the USEPA's recommended standard for freshwater (<14 ng/L) [57], suggesting exposure at these sites does not pose potential risks from PCBs. The ΣPCBs observed for this study are comparable to those reported in literature for river waters elsewhere, as shown in Table 5.

Table 5. Comparison of the Pra river PCB concentrations with other studies.

Location	Concentration ΣPCBs (ng/L)	Reference
Pra river basin	0.5285 - 5.869	This study
Hudson river estuary, USA	0.86 - 6.0	[58]
Houston waterway	0.49 to 12.49 ng/L	[59]
Yangtze River	0.04 - 11 ng/L	[60]
Rivers in Shanghai, China	0.04 - 11 ng/L	[61]
Estuary of Dagu River, China	ND-34.8	[62]
Southern Moravia region, Czech Republic	5.2 - 190.8	[63]
Udu River, Nigeria	20 - 1860	[64]
Ganga and Brahmaputra. Rivers, India	ND - 142	[65]
Ebro River	(up to 108 ng/L	[66]

The most dominant homologs detected in water samples were the hexachlorobiphenyl homologs (6-CBs), which is made up of PCBs 138, 149 and 153 constituted 25.51% of the total PCBs in water. PCB153 was the most dominant congener constituting 16.66%. The homologs order 6-CBs > 7-CBs (16.27%) > 4-CBs > 5-CBs > 3-CBs > 8-CB (4.17%). Whiles previous reports have suggested a slight increase in PCB concentration with higher chlorine numbers [57], this study found that the concentration of PCBs was not dependent on the number of chlorine atoms present.

3.3. PCB Comparison Using R^2

The degree of similarity between any two sites congener profiles is identified through the use of coefficient of determination R^2 *[67]* [68]. An R^2 value of 0.9 to 1.0 shows a perfect match between the profiles; R^2 value of 0.8 to 0.89 means very similar fingerprints; 0.7 to 0.78 suggests similar fingerprints, while a value of 0.0 implies no relationship between profiles. Very few sampling sites, P6 and P1, P5 and P4, P5 and P3, P10 and P9, P11 and P8, P14 and P9, P14 and P10, and P20 and P7 showed fingerprints match in the dry season; with $R^2 = 0.945$ ($r = 0.972$). More sites showed very similar degree of fingerprint. Sites that showed ambiguous fingerprints had R^2 values ranging from 0.6 to 0.69 ($r = 0.787 - 0.814$), while sites with distinctly different fingerprints recorded $R^2 < 0.6$ ($r < 0.775$). In the wet season, many sites showed perfect march, and very similar and similar fingerprints in their congener profiles than in the dry season. An R^2 of 0.2 ($p = 0.05$) was obtained for PCB congener profile comparison of samples for dry and wet seasons. Thus only 20% of the variation in the PCB profile in one season is dependent on the profiles of the other season. The water samples for the two seasons had distinctly different PCB congener fingerprint.

3.4. PCB Comparisons Based on HCH and PCA

3.4.1. Hierarchical Cluster Analysis

The dendrograms show the hierarchical clustering of the sampling sites based on their PCB congener profiles. Clusters with the least Euclidean distance (shortest branch) are most similar. The clusters are dissimilar if the distance is about half the whole Euclidian distance [43]. The clustering of the sampling sites for water (**Figure 6**), revealed that sites P3 and P6 had the highest level of similarity in terms of their PCB profiles. The degree of similarity followed the order: P3and P6 > P16 and P17 > P12, P13 and P15 > P2, P3 and P6 > P4 and P19.

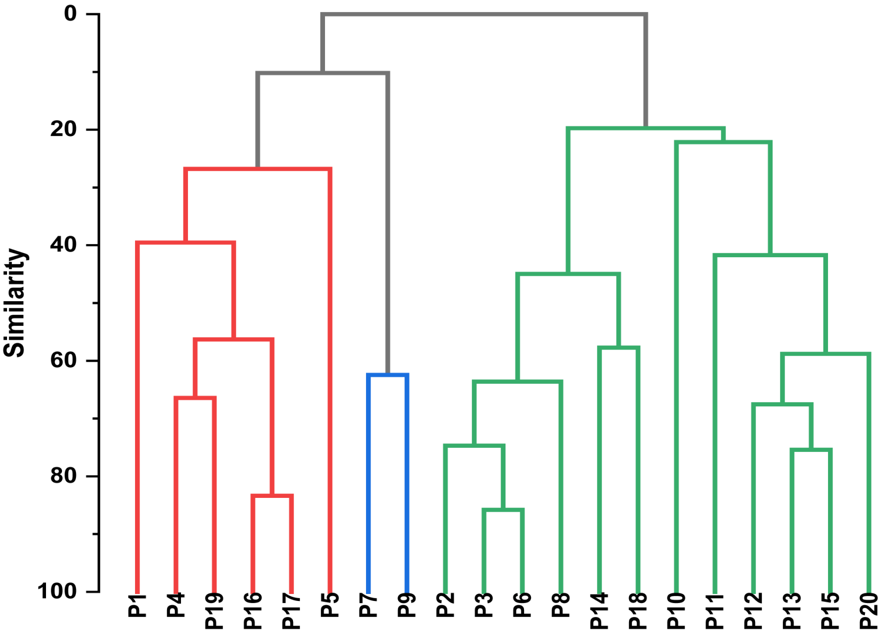

Figure 6. Dendrogram of water sampling sites for PCB profiles.

3.4.2. PCB Fingerprint Based on PCA

To examine the relationships, similarities among the PCB congeners and sampling points; the PCB profiles (mean of PCB data) obtained were subjected to Principal Component Analysis (PCA). The results are shown (**Figures 7-9**), were interpreted based on the guide reported [43] [67]-[70]. The Principal Components of PCBs observed in 3D (**Figure 7**) and 2D (**Figure 8** and **Figure 9**) for water samples are shown. The results show that 56.68% of the total variation in congener data is explained by the first three components. Principal Component 1 (PC1), plotted on the horizontal axis, explains most (23.53%) of the variability. The second component (PC2) captured 17.45% and the third (PC3), 15.70% (**Figure 9**). Principal Component 1 is loaded with PCBs 18, 28, 44, 101, 138, 180 and 194 as these are closer (more parallel) to PC1 axis (**Figure 6**).

However, PCB 44 with the largest score (longest vector) contributes greatest to the variability followed by PCB 194, 180 and 28. PCBs vectors of 138 and 194 pointing in opposite directions to the others have a *negative* relationship. Vectors pointing in the same direction (PCB 18, 44 and 180; PCB 28 and 31) correlate positively. The correlations suggest that these congeners have common sources or mutual dependence and/or similar behavior during transport, or are subject to common factors of control [71].

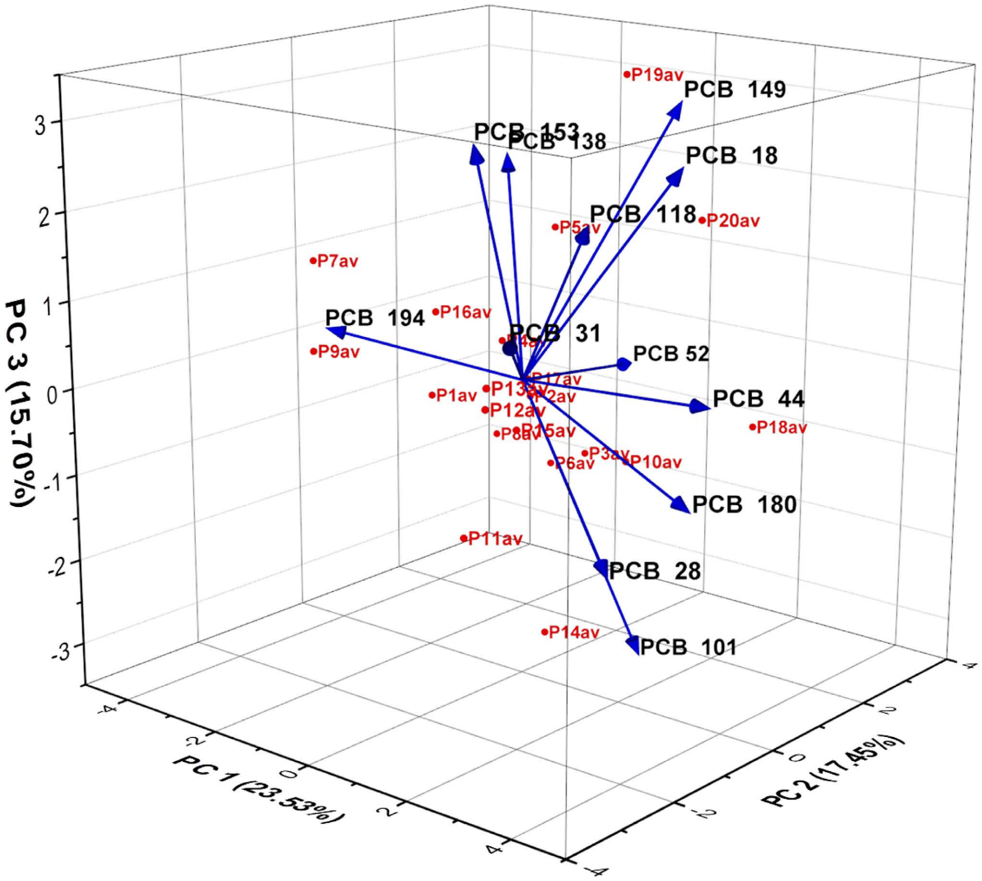

Figure 7. A 3D Principal Components of PCBs observed in the Pra river water samples.

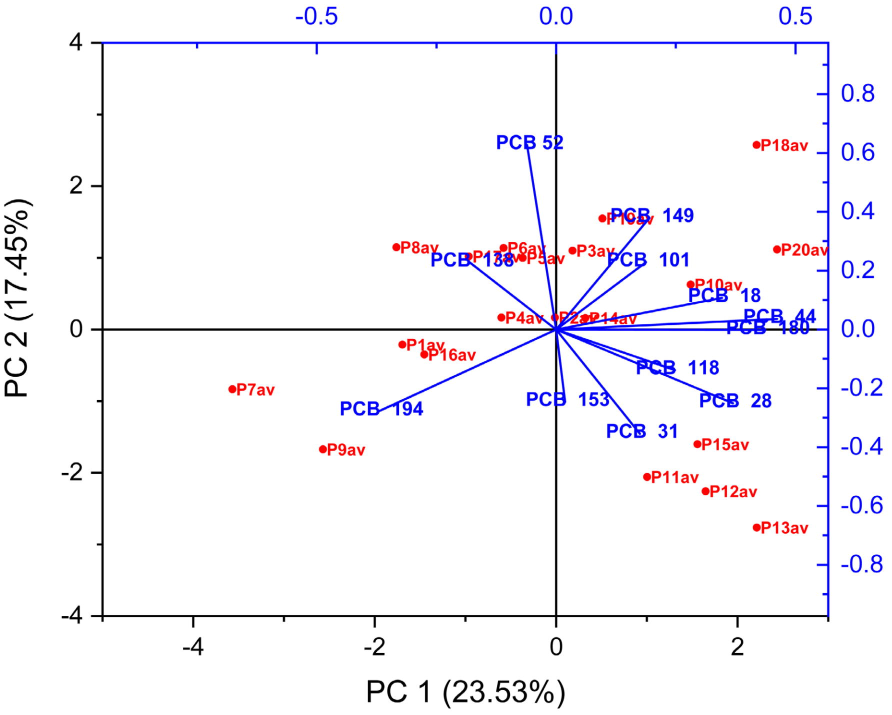

Figure 8. Principal Components Analysis of PCBs projected onto PC 1 and PC 2.

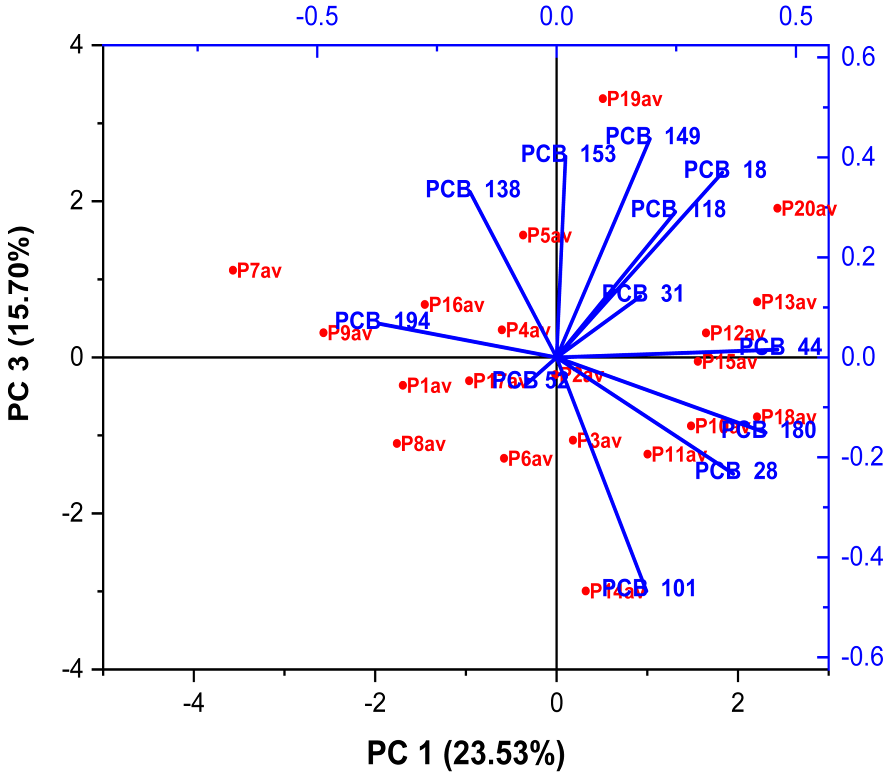

Figure 9. Principal Components Analysis of PCBs projected onto PC 1 and PC 3.

The second component (PC2) is loaded with PCBs 31, 52, 101, 149 and 153 as these are closer (more parallel) to PC2 axis. However, PCB 52 with the largest score (longest vector) contributes greatest to the variability followed by PCB 31. PCBs of 52 and 149 correlate negatively with PCBs 31 and 153. PCBs 52, and 149; whiles PCBs 28 and 31 correlated positively.

Principal Component 3 is loaded explains 15.70% of the variations, and loaded with PCBs 44, 180 and 194. The sampling sites (points P) that show similarity have similar or close scores, included P3 and P6; P2, P16 and P17; P12, P13 and P15; P4 and P19. These are in agreement as those observed with the cluster analysis in Figure 6.

3.5. Human Cancer Health Risk Assessment

The PCB cancer risk from use of water from Pra River Basin is mainly due to PCB118, the only dioxin like PCB detected in this study. The mean and ranges of cancer risk values for human exposure to PCBs via ingestion (CR_{ing}) and dermal contact (CR_{der}) are respectively 4.8E−09 (0 to 2.7E−08) and 4.5E−10 (0 to 2.0E−09). The total cancer risk value is 5.3E−09 (Table 6). Thus, a maximum of three chance in a hundred million and averagely five out of a billion of the population are at risk of exposure to cancer or related illness as a result of PCB118 exposure. The observed estimated probabilistic PCB cancer risk is much lower than the acceptable risk of one chance in a million (1.0E−06) of exposed persons to suffer the risk of cancer. This implies a very low PCB cancer risk from the use of water from the Pra river.

Table 6. PCB Cancer Risk Values Associated with the use of Water from Pra River.

	CR_{ing}			CR_{der}			CRI_{total}		
PCB	min	max	mean	min	max	mean	min	max	mean
PCB118	0	2.7E−08	4.8E−09	0	2.0E−09	4.5E−10	0	2.9E−08	5.3E−09

3.6. Non-Cancer Health Risk Assessment

The mean HQs due to the individual PCBs is shown (Rable 4). ranged from 3.9×10^{-04} - 1.9×10^{-03}. The hazard index (HI) which is the sum of the HQs [72] had a mean of 1.1×10^{-02}, and ranged from 3.2×10^{-04} to 5.8×10^{-02}. The HQs and the HI were all lower than 1. Thus, this indicates the potential for non-cancer hazarded negligible.

However, in children, chronic exposure to lower chlorinated PCBs is associated with lower IQ, attention deficits and behavioral problems [73]. Also, the lower chlorinated PCBs can accumulate in aquatic organisms especially fish leading to reproductive toxicity, growth inhibition, and immune suppression [74]. Communities relying on the Pra River are at risk. Ingesting contaminated fish leads PCB accumulation in human beings particularly affecting liver function, endocrine disruption, and neurological development [75].

3.7. Potential Ecotoxicological Risks

The TEQs for PCBs in samples were calculated by using toxicity equivalence factors (WHO-TEFs). The total TEQ was calculated by multiplying the actual concentration of PCBs in the environmental sample by the corresponding toxic equivalency factor (TEF). The TEQ values are shown (Table 7). The TEQs calculated for the water in the wet and dry seasons respectively range from 0.005 - 0.030 pg/L and 0.020 - 0.059 pg/L for birds; 0.003 - 0.0145 pg/L and 0.011 - 0.029 pg/L for fish, and 0.053 - 0.267 pg/L and 0.203 - 0.587 pg/L for human and mammals. All the TEQs are lower than the recommend WHO PCB-TEQ value 0.003 ng/L (3.0 pg/L) [76].

Table 7. PCB-TEQ values for wildlife and humans/ mammals.

	PCB-TEQ (pg/L)					
	Dry season			Wet season		
	Birds	Fish	Humans	Birds	Fish	Humans
P1	0.023	0.012	0.228	0.012	0.006.	0.123
P2	0.023	0.011	0.280	0.018	0.009	0.179
P3	0.020	0.010	0.203	0.027	0.013	0.267
P4	0.021	0.011	0.210	0.020.	0.010	0.201

Continued

P5	0.032	0.016	0.314	0.011	0.005	0.106
P6	0.024	0.012	0.241	0.013	0.007	0.130.
P7	0.034	0.017	0.342	0.008	0.004	0.077
P8	0.025	0.013	0.253	0.006	0.003	0.058
P9	0.024	0.012	0.243	0.005	0.003	0.053
P10	0.040	0.020	0.397	0.016	0.008	0.161
P11	0.035	0.018	0.350	0.020	0.010	0.195
P12	0.034	0.017	0.344	0.016	0.008	0.164
P13	0.044	0.022	0.443	0.017	0.008	0.167
P14	0.021	0.011	0.214	0.019	0.010	0.191
P15	0.022	0.011	0.222	0.030	0.015	0.300
P16	0.040	0.020	0.398	0.015	0.008	0.154
P17	0.032	0.016	0.032	0.016	0.007	0.155
P18	0.059	0.029	0.587	0.019	0.009	0.186
P19	0.054	0.027	0.541	0.018	0.009	0.182
P20	0.053	0.026	0.525	0.023	0.011	0.226
Mean	0.033	0.017	0.330	0.016	0.008	0.164
std	0.012	0.006	0.119	0.006	0.003	0.063
CV	36.05.	36.06	3606	38.25	38.25	38.25

The coefficient of variation ranged from 36.06% - 38.25%, suggesting significant variation in the TEQs at the sampling sites. However, the variation in TEQs for the wet and dry sessions was very minimal. The levels recorded in the Pra River is within levels reported worldwide. In rivers in Shanghai, China it ragged from nd–1135.63 pg TEQ/L [61]; in Lake Baikal, Russia 10^{-4} to 3.3×10^{-3} pg/L [77], iin surface water east Slovakia, 2.86 ng/L [78]; Korean river, <0.01 - 238 pg/L [79]; Yamuna River (<1 - 1600 pg/L) and the Delhi River (<0.01 - 2314 pg/L) [80]. High TEQs beyond the WHO acceptable limit has been reported in in River Otamiri, Imo State, Nigeria [81].

The World Health Organization has recommended a limit of 0.003 $ng \cdot L^{-1}$ TEQ value for PCBs in surface water [76], and the European Union, 1 ng/L [82]. It can be seen that none of toxicity levels at the study sites exceeded the WHO acceptable limit for PCBs in surface. This suggests the river water is safe for humans and wildlife. However, the toxicity of an environmentally derived mixture of chemicals, including dioxinlike PCBs, have been observed to act in an additive or even synergistic manner [83]; the toxicity of PCB presents in the Pra river to human and wildlife may therefore be higher than what has been observed in this study as the toxicity was based on PCB 118 only.

4. Conclusions

The PCBs at twenty different locations in the drinking water source of the Pra River were determined. Among all congeners, PCB18, 153, and 180 have the highest detection frequency in water. The Σ_{12}PCBs concentration for the wet and dry seasons respectively ranges from 0.539 - 2.995 ng/L and 2.103 - 5.869 ng/L. The Pra River has low levels of PCB pollution at all the sites compared with other regions. In general, the hexa-chlorinated PCBs biphenyls were the dominant congeners. The mean and ranges of cancer risk values for human exposure to PCBs via ingestion and dermal contact were respectively 4.8E−09 (0 to 2.7E−08) and 4.5E−10 (0 to 2.0E−09). The total cancer risk value is 5.3E−09. Thus, a maximum of three chances in a hundred million, and averagely, five out of a billion of the population are at risk of exposure to cancer or related illness. The observed estimated probabilistic PCB cancer risk is much lower than the acceptable risk of one chance in a million (1.0E−06) of exposed persons to suffer the risk of cancer. All of the TEQ were lower than the environmental quality standard in Japan (1.0 pg-TEQL (−1)). This implies negligible PCB risk from the use of water from the Pra River. The potential ecological risk from PCBs in the water is very low. The PCB concentration in water suggests that the river water does not pose a danger to aquatic organisms and human health.

Sites identified as contaminated with PCBs should be thoroughly surveyed and mapped to determine the various sources contributing to PCB leakage into the Pra River. Targeted interventions such as the removal of PCB-containing equipment, sediment dredging, capping, phytoremediation, public education, and stricter regulatory enforcement can significantly reduce PCB concentrations and help prevent further contamination within the Pra River Basin.

Authors' Contributions

J.K.B., D.K.E., and A.E. designed and supervised the study; participated in laboratory work J.K.B., D.K.E., and oversaw the laboratory work. J.K.B., D.K.E., A.E, J.K.E, P. B, O.O.O., J.W.A.J. All authors performed data interpretation, cleaning and analysis; and participated in drafting and editing the manuscript. J.K.B., D.K.E., A.E, J.K.E, P. B, O.O.O., J.W.A.J. All authors reviewed the final version of the manuscript to be published.

Conflicts of Interest

The authors declare no conflicts of interest.

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