



Assessment of Polychlorinated Biphenyls (PCBs) in Water, Sediments and Biota from E-waste Dumpsites in Lagos and Osun States, South-West, Nigeria

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ABSTRACT: The levels of PCBs in sediments, water, leachate and aquatic fauna (*Tilapia guineensis*, *Callinectes amnicola* and *Cardiosoma armatum*) found in and around e-waste dumpsites in Lagos and Osun States, South-West, Nigeria were analyzed using Gas Chromatography Electron Capture Detector (GC ECD) Agilent 7820A. All the 28 PCBs congeners studied were detected with the Σ hexa-PCBs dominating in Lagos (21%) while the Σ tetra-PCBs (24%) enriched the samples from Osun State. The concentrations of Σ -PCBs in decreasing order were Lagos: sediment > fish gill > fish muscles > water > crab > leachate and Osun: fish gill > fish muscle > sediment > crab > water > leachate. The concentrations of total indicator PCBs (Σ_7 PCBs) in the sediment from Lagos (4.19 $\mu\text{g}/\text{kg}$) and Osun (8.58 $\mu\text{g}/\text{kg}$) exceeded the Canadian Sediment Quality standard threshold effect level (CSQ TEL) (0.03 $\mu\text{g}/\text{kg}$) and the National Oceanic Atmospheric Administration threshold effect level (NOAA TEL) for fresh and marine sediments. The calculated toxic equivalent quotient (TEQ) for fish from Lagos and Osun (3.7 and 4.4) respectively further suggests a likely occurrence of adverse effects to humans who consume the fish. This study reveals the high health and ecological risks associated with e-waste pollution in the aquatic environment.

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PCBs are a class of non-polar toxic chemical compounds consisting of 209 congeners (Adeyemi *et al.*, 2009), out of which only about 130 (di- to deca-PCBs) are found in commercial mixtures. They are water soluble organic compounds with 1 to 10 chlorine atoms attached to biphenyl, which is a molecule composed of two benzene rings with a chemical formula of $\text{C}_{12}\text{H}_{10-x}\text{Cl}_x$, where $x = 1-10$. They are also persistent organic pollutants whose effects remain long after their usage. They are used and applied widely in dielectric fluids such as in transformers, capacitors, coolants and other agricultural purposes. They are detected in a wide range of environmental compartments including biota (fish and other benthic organisms), soil, water, air, sediment, plants and animal tissue in all regions of the earth. Their reported half-lives in soil and sediment range from months to years. Due to their very low solubility in water and low volatility, most of them are contained in sediments that serve as environmental reservoirs from which they may continue to be released over a long period of time for example, the life expectancy of electrical transformers that contain PCBs is 30 years or more. (Wang and Zhong, 2011). They were synthesized for the first time in the last century and produced at an industrial level in the 1930's (Jang and Townsend, 2011).

The core indicator PCBs are the 7 ICES (International Council for the Exploration of the Sea) monitored congeners namely CB-28, CB-52, CB-101, CB-118, CB-138, CB-153 and CB-180 (Boalt *et al.*, 2014). They are listed as mandatory contaminants to be analyzed and reported within both the OSPARCOM (Oslo/Paris Convention for Protection of the Marine Environment of the North-East Atlantic (1992) and the HELCOM (Helsinki Commission) Conventions. They are also classed as priority POPs under the Stockholm Convention and the Marine Strategy Framework Directive (MSFD).

Twenty members of the 209 PCB congeners attain coplanarity (Wong and Wong, 2007). This is due to non-ortho chlorine substitution in the biphenyl ring effects that is (four non-ortho substituted PCBs: CB-77, CB-81, CB-126, CB-169, IUPAC and eight mono-ortho substituted: CB-105, CB-118, CB-156, CB-157, CB-167, CB-114, CB-123, CB-189, IUPAC) (Burreau *et al.*, 2006). These are the dioxin-like PCBs (dl-PCBs) because they have a coplanar structure very similar to that of dioxins. They are known to elicit common toxic and biological responses including body weight loss, thymicatrophy, dermal disorder, hepatic damage,

teratogenicity, reproductive toxicity, immunotoxicity, and high induction potency of 3-methyl cholanthrene type hepatic microsomal enzymes (Wang and Zhong, 2011).

Exposure to PCBs may enhance the genotoxic activity of similar compounds to cause more severe toxic effects which are explained by the binding of planar (non-ortho) PCBs and PCDD/Fs to the aryl hydrocarbon (Ah) receptor (Boalt *et al.*, 2014). The author further mentioned that their lipophilic characteristics are responsible for their ability to bioaccumulate particularly in tissues and organs rich in lipids which leads to their consequent possible connection with carcinogenesis in living organisms. They are rapidly accumulated by aquatic organisms and bioaccumulated through the aquatic food chain. Greater relative amounts of PCBs are usually found in the liver, adipose tissue, skin, and breast milk. It has been shown that absorption by nursing infants of tetra and higher chlorinated congeners from breast milk ranges from 90% to 100% of the dose (Wang and Zhong, 2011).

Therefore, this study investigated the levels of PCBs in the leachate, water, sediment and some aquatic fauna found in and around e-waste dumpsites as well as assessed the ecological risk posed to the organisms by PCBs contamination.

MATERIALS AND METHODS

Study Areas and Collection of Surface Water, Leachate and Animal Specimen: Sampling was performed in Lagos (Alaba International Market) and Osun (Ajegunle Market) States of South-West Nigeria (Figure 1). To measure the levels of the PCBs four samplings were performed at each of the two dumpsites (point of entry into the water body, 500 m away from the dumpsite, 1000 m away from the dumpsite and 2000 m upstream which was the control). Surface water samples were collected from the study stations by dipping water sampler 6-7 cm below the surface film. Leachates samples were also collected from 10 different points around the dumpsites with a 1dm³ water sampler, mixed together and filtered using 15 cm whatman filter paper (Okunola *et al.*, 2012). Both samples were stored in 1litre water bottle each with a screw cap, transported to the laboratory in a cooler of dry ice for further analysis.

Sediment samples (0-10 cm depth) were collected from the study sites using a stainless Van-Veen grab as previously described (Okunola *et al.*, 2012). The sediment samples (100-200 g) were wrapped in aluminium foil and placed into cleaned polythene bags (Ziploc) and transported to the laboratory where they were pooled to form a composite sample for each study site. Sediments were air dried for one month and ground

to powder using a laboratory mortar and pestle, passed through a 2 mm sieve and stored at -2⁰C until analysed (Okunola *et al.*, 2012). Professional anglers used gill nets of 1-3cm diameter and cylindrical crab traps (Ayoola and Kuton, 2009). The biological samples used for the test were *Tillapia guinensis*, *Cardiosoma armatum* and *Callinectes amnicola*. The choice of the study animals was based on their availability in the environment.



Fig 1: Map of Study Area

Determination of Polychlorinated Biphenyls (PCBs): PCBs were analyzed following a previously developed method of Chu *et al.*, (1996). Sample aliquots of 50mL /5 g of sample (water, leachate/sediment and biota) were ground with 5 g anhydrous sodium sulfate into a dry homogenate. The biota samples were saponified with an ethanolic solution of potassium hydroxide (Baumard *et al.*, 1998). An internal standard was added and ultrasonic extraction was used to extract PCBs from the samples with 50 mL hexane/acetone (1:1 v/v). The extract was concentrated to approximately 3 mL by rotary evaporator. For the clean-up, the sample solution was shaken with concentrated H₂SO₄ in a test tube, after centrifugation, the acid layer was discarded. This treatment was repeated several times until the hexane layer was dried with anhydrous sodium sulfate, and then concentrated to approximately 1 mL for column chromatographic clean-up. The concentrations of 28 PCB congeners were measured by GC-ECD Agilent 7820A. The recovery rate of individual PCB congeners ranged from 87% to 100%.

Ecological Risk Assessment (ERA): To assess toxicity posed to the benthic community, three different categories of consensus based sediment quality guidelines (SQG) namely threshold effect concentration

(TEC), probable effect concentration (PEC) and extreme effect concentration (EEC) according to Long *et al.*, (1995) were used.

Total Equivalent Quotient (TEQ): To express the overall toxicity of the non-ortho substituted coplanar PCBs (CB-77, CB-81, CB-126, and CB-169) in the fish from each State, a TEQ was calculated according to SQUIRT by NOAA (1999)

Statistics : Concentrations of PAHs in the samples from each study site were reported as mean values. Analysis of variance was performed on all experimental data and means were compared using SPSS version 20.0 software.

RESULTS AND DISCUSSION

Distribution of individual PCB Congeners in the Studied Samples from the two Dumpsites: The total PCBs ranged from 0.22 µg/L in Osun leachate to 21.04 µg/kg in Lagos sediment (Figure 2). The concentrations of Σ -PCBs were in the following decreasing order in Lagos: sediment > fish gill > fish muscles > water > crab > leachate and Osun: fish gill > fish muscle > sediment > crab > water > leachate. Generally relatively higher PCBs burden was recorded in Lagos (56.21%) than in Osun (43.7%). This was attributed to a higher density market in Lagos than in Osun. The Σ -hexa-PCBs dominated (21%) in Lagos samples while Osun samples were dominated by Σ -tetra-PCBs accounting for about 22.84%. The sum of the coplanar PCBs accounted for about (20.2%) in Lagos and 19.4% in Osun samples, an indication of highly toxic samples (Wang and Zhong, 2011).

Distribution Patterns of Individual PCBs Congeners in Leachate from the two Dumpsites: Leachate samples from both States had the lowest recorded concentrations of PCBs congeners (Table 1). Indicator PCBs in the leachate samples were dominated by CB-52 ranging from 0.07 µg/L in Lagos samples to 0.03 µg/L in Osun samples. Congeners CB-118, CB-153 and CB-180 were not detected in Osun while all congeners screened were present in Lagos leachate. The low levels of PCBs concentration in the leachate from both states were attributed to dilution from water. A similar observation was reported by Dannon-schaffer (2010).

Distribution Patterns of Individual PCBs Congeners in Water from the two Dumpsites: In Lagos water samples, indicator CB-28 had the highest level of 0.57 µg/L while the lowest recorded level occurred in CB-180 (0.11

µg/L) as shown in Table 1. All the coplanar PCBs screened were detected in Osun water with CB-156 having the highest occurrence (0.79 µg/L) while the lowest level was CB-157 (0.23 µg/L). When the concentrations of these contaminants in the water bodies are high, the sediments may accumulate excessive quantities that will directly or indirectly disrupt the ecosystem causing significant contamination and loss of desirable species (Burton, 2002).

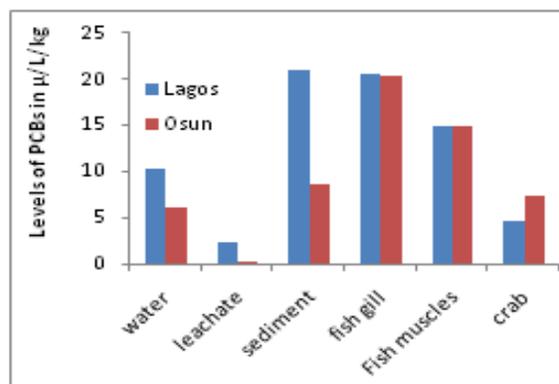


Fig 2: Total concentrations of PCBs congeners in Osun and Lagos e-waste (n=8)

Distribution Patterns of Individual PCBs Congeners in Sediment from the two Dumpsites: In Table 1 the highest recorded level of PCBs congeners occurred in Lagos sediment (16.02%) while the most dominant indicator PCBs in Lagos was CB-118 (1.73 µg/kg) and the lowest in concentration was CB-180 (0.11 µg/kg). In Osun CB-28 was observed to have the highest level of 0.56 µg/kg with the lowest level of 0.15 µg/kg recorded for CB-180. The high level of PCBs found in the sediments have been widely reported (Wong and Wong, 2007; Wang and Zhong, 2011) and it is suggestive of the fact that persistent organic pollutants are strongly particle associated due to their hydrophobic properties and tend to be accumulated in sediments ((Apitz *et al.*, 2006; Gomez-Gutierrez *et al.*, 2007).

Distribution Patterns of Individual PCBs Congeners in Biota from and around the two Dumpsites: All the 7 indicator PCBs investigated were detected in Lagos and Osun crabs (Table 1). Congener CB-101 had the highest level of indicator PCBs (0.22 µg/kg) in Lagos crab while congener CB-28 dominated in Osun crab (0.92 µg/kg). Out of the 12 coplanar PCBs (non and mono-ortho) investigated, it was observed that CB-156 had the highest concentration of 0.71 µg/kg in Lagos crab while Osun crab was dominated by CB-126 (0.72 µg/kg) in Osun crab.

Table 1: Concentration of PCBs in water, leachate ($\mu\text{g/L}$), sediment, *Tilapia guineensis* (muscles and gill) and crab (*Callinectes amnicola* and *Cardiosoma armatum*) $\mu\text{g/kg}$ from Lagos and Osun States

Congeners	Lagos						Osun					
	water	leach	Sed	Fish gill	Fish muscles	Crab	Water	leach	sed	Fish gill	Fish muscles	crab
CB8	0.62	0.27	5.28	1.26	2.83	0.14	0.15	ND	0.77	0.72	0.70	0.81
Σ di-PCB	0.62	0.27	5.28	1.26	2.83	0.14	0.15	ND	0.77	0.72	0.70	0.81
CB18	1.08	0.07	1.07	1.27	1.82	0.20	0.66	0.02	0.50	0.99	3.86	0.56
CB28	0.57	0.06	0.78	0.48	0.40	0.13	0.23	0.02	0.56	0.50	0.17	0.92
Σ tri-PCB	1.65	0.13	1.85	1.75	2.22	0.33	0.89	0.04	1.06	1.49	4.03	1.48
CB44	0.44	0.05	0.59	0.44	0.78	0.07	0.23	ND	0.34	0.36	0.31	0.15
CB52	0.39	0.07	0.46	0.50	1.35	0.10	0.44	0.03	0.20	0.50	0.32	0.22
CB60	0.40	0.04	0.93	0.63	0.35	0.12	0.23	0.02	0.34	0.67	0.31	0.16
CB77	0.53	0.01	1.31	0.33	0.54	0.12	0.27	ND	0.45	0.68	0.33	0.15
CB101	0.39	0.05	0.87	1.05	0.60	0.22	0.15	0.02	0.42	0.93	0.79	0.17
CB81	0.47	0.05	0.48	0.92	0.57	0.23	0.05	ND	0.43	1.50	0.62	0.28
CB105	0.31	0.03	0.28	0.69	0.25	0.06	0.04	0.02	0.07	0.56	0.54	0.08
Σ tetra-PCB	2.93	0.30	4.92	4.56	4.44	0.92	1.41	0.09	2.25	5.20	3.22	1.21
CB114	0.37	0.05	0.57	1.08	0.39	0.10	0.34	0.02	0.58	0.77	0.98	0.22
CB118	0.43	0.06	1.73	0.19	1.95	0.14	1.38	ND	0.18	0.45	0.22	0.13
CB123	0.31	0.06	0.28	0.98	0.43	0.11	0.11	ND	0.11	0.82	0.67	0.11
CB126	0.25	0.35	0.25	0.55	0.17	0.15	0.06	0.02	0.22	0.46	0.27	0.72
Σ penta-PCB	1.36	0.52	2.83	2.80	2.94	0.50	1.89	0.04	1.09	2.50	2.14	1.18
CB128	0.11	0.03	0.19	0.89	0.09	0.14	0.05	ND	ND	0.81	0.23	0.18
CB138	0.23	0.03	0.11	1.16	0.15	0.12	0.11	0.02	0.22	0.70	0.30	0.18
CB153	0.24	0.06	0.09	1.61	0.39	0.18	0.05	ND	0.16	1.43	0.23	0.26
CB156	0.79	0.09	0.70	1.16	0.69	0.71	0.51	ND	0.81	1.12	0.30	0.37
CB157	0.23	0.03	0.12	1.39	0.28	0.31	0.14	ND	0.30	0.76	0.36	0.24
CB167	0.17	0.03	0.04	0.65	0.11	0.07	0.09	0.02	0.04	0.63	0.09	0.22
CB169	0.25	0.08	0.11	0.51	0.10	0.21	0.06	ND	0.11	0.58	0.15	0.20
Σ hexa-PCB	2.02	0.35	1.36	7.37	1.81	1.74	1.01	0.04	1.64	6.03	1.66	1.65
CB170	0.11	0.03	0.18	0.50	0.09	0.10	0.10	ND	0.17	0.47	0.17	0.17
CB180	0.13	0.03	0.15	0.90	0.10	0.11	ND	ND	0.15	0.77	0.31	0.13
CB185	0.19	0.05	0.24	0.44	0.16	0.51	0.16	ND	0.29	0.56	0.10	0.17
CB189	0.10	0.05	0.14	0.39	0.08	0.09	0.09	ND	0.42	0.52	0.08	0.14
Σ hepta-PCB	0.53	0.16	0.77	2.23	0.43	0.81	0.39	ND	1.03	2.34	0.66	0.61
CB195	0.10	0.03	0.36	0.29	0.04	0.11	ND	ND	0.20	0.28	ND	0.13
Σ octa-PCB	0.10	0.03	0.36	0.29	0.04	0.11	ND	ND	0.20	0.28	ND	0.13
CB206	0.08	0.04	0.15	0.11	0.09	0.06	0.11	ND	0.32	0.63	ND	0.06
Σ nano-PCB	0.08	0.04	0.15	0.11	0.09	0.06	0.11	ND	0.32	0.63	ND	0.06
CB209	1.02	0.50	3.57	0.29	0.09	0.03	0.23	ND	0.22	1.20	2.54	0.18
Σ deca-PCB	1.02	0.50	3.57	0.29	0.09	0.03	0.23	ND	0.22	1.20	2.5	0.18
Σ total PCBs	10.31	2.24	21.04	20.65	14.89	4.65	6.04	0.22	8.58	20.38	14.96	7.32
Σ ICES indicator PCBs	2.38	0.36	4.19	5.89	4.94	1.00	2.36	0.09	1.89	5.28	2.34	2.01
*coplanar PCBs	4.21	0.92	5.90	7.84	5.56	2.30	3.14	0.08	3.72	8.85	4.61	2.86

ND-not detected (or value 0.00). Total International Council for the exploration of the sea (ICES) indicator PCBs=sum of concentrations of PCB-28, -52, -101, -118, -138, -153, -180. *Total dioxin-like (coplanar) PCBs = Sum of concentration of PCB -77, -81, -105 -114, -123 -118 -126 -156, -157 -167 -169 and -189

Congener CB-118 a coplanar PCB had the highest recorded concentration of $1.95\mu\text{g/kg}$ in fish muscles from Lagos while fish gill from Osun accumulated the highest recorded level of the pollutant ($21.5\mu\text{g/Kg}$) than all other samples from the State. This result further confirms that these pollutants are absorbed by organisms directly from the water through their gills, hard exoskeleton, head, telson and other tissues (Onwumere, *et al.*, 1990). The levels of the non-ortho- substituted PCBs in fish from both Lagos and Osun States respectively were higher than the NOAA (1998) fish TEF. This has a serious health implication as coplanar PCBs are known to elicit common toxic and biological responses (Wang and Zhong, 2011). Relatively higher PCBs levels occurred in the dry than in the wet seasons (Figures 3 and 4). Water and sediment from Station 2 in Lagos State were more enriched with PCBs than other stations, while in Osun State station 1 was the most polluted with the Σ penta PCBs having the highest concentration in water (Figure 5). This was attributed to the nature of the water bodies in the two States. Water in Lagos is lotic while that of Osun is lentic.

Evaluation of Ecological Risk Assessment

The concentration of total indicator PCBs (Σ_7 PCBs) in the sediment from Lagos ($4.19\mu\text{g/kg}$) and Osun ($8.58\mu\text{g/kg}$) exceeded the CSQ TEL ($0.03\mu\text{g/kg}$) and the NOAA TEL levels for fresh and marine sediments. The levels of the non-ortho- substituted PCBs (CB-77, CB-81, CB-126, and CB-169) in fish from both States were found to be higher than the NOAA (1998) fish TEF. The calculated TEQ for fish from Lagos and Osun were found to be 3.7 and 4.4 respectively.

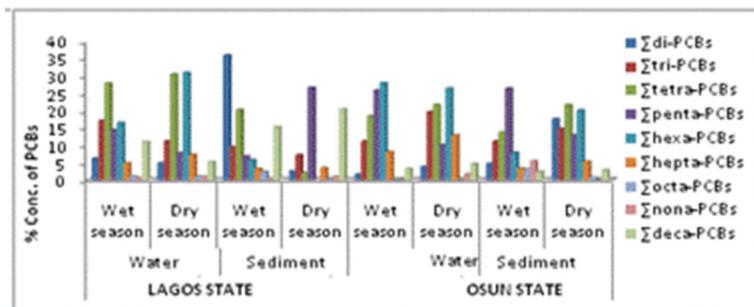


Figure 3: Percentage Seasonal Variation of PCBs found in the Water and Sediment Samples from Lagos and Osun e-waste dumpsites.

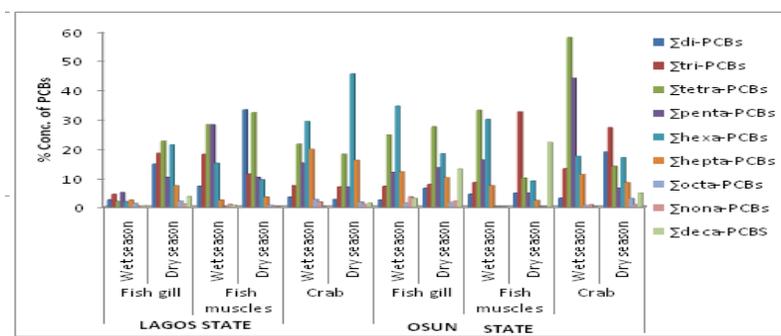


Figure 4: Percentage Seasonal Variation of PCBs in *Tilapia guineensis* (gill and muscles) and crab (*Callinectes amnicola* and *Cardiosoma armatum*) from Lagos and Osun States

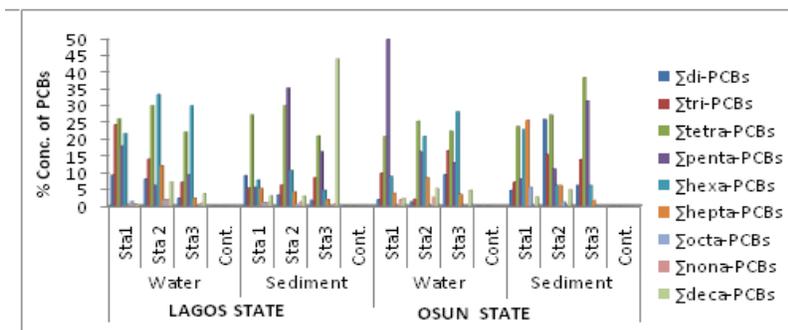


Figure 5: Percentage Spatial variations of the Percentage Concentrations of PCBs in Lagos and Osun States.

This is suggestive of a likely frequent toxic occurrence to the benthic community which is of ecological risk. Also the result further show that consumption of this fish from these contaminated sites by humans is likely to cause serious adverse health effects. A study by Qing *et al.*, (2006) reported that the United States Environmental Protection Agency (USEPA) has classified PCBs as probable human carcinogens (Group B2). The study reported that as at 1998, 37 states in the United States of America have issued 679 fish advisories for PCBs. These advisories educate the public on the high concentrations of PCBs found in local fish at levels of public health concern and recommend either limiting or avoiding consumption of certain fish from specific water bodies (USEPA, 1999).

Acute and chronic high-level exposures of laboratory animals to PCBs have resulted in liver and kidney damage, neurological effects, developmental

effects, endocrine effects, hematological effects, and death. This has made PCBs to be included in the list of priority pollutants that have attracted global attention (Osibanjo, 2012).

Conclusion:

Finally, this study established the environmental risk and contamination of water, sediment and biota by PCBs in South-West Nigeria. It also revealed that the levels of non-ortho and indicator PCBs in sediments and fish in both States exceeded the CSQ TEL and the NOAA TEL (1998) acceptable limit standards for fresh and marine sediments as well as the fish TEF. Consequently from the above findings, it is obvious that there is urgent need for a concerted effort to be made by the relevant authorities to address the environmental and health hazards posed by the exposure to e-waste.

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