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The link to this publication is https://ajoeer.org.ng/otn/ajoeer/2022/qtr-2/05.pdf

POLYCHLORINATED BIPHENYLS (PCBS) AND HEAVY METALS CONCENTRATIONS IN PERIWINKLE IN SOME RIVERS IN RIVERS STATE, NIGERIA.

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ABSTRACT

Pollution of aquatic environments in the Niger Delta remains a major source of food chain contamination. In this study, the concentrations of some trace metals and Polychlorinated biphenyls (PCBs) in biota were determined. Standard laboratory and environmental techniques were employed in sampling and analysis of samples. Periwinkle samples were collected from Borokiri, Kaa, Okrika and Eagle Island rivers. The concentrations of PCBs were determined using gas chromatography equipped with flame ionization detector (GC-FID) while trace metals were determined using atomic absorption spectrophotometer (AAS). The results were analyzed using descriptive and inferential techniques. The results of the study showed that concentrations of PCBs in the periwinkle ranged from 0.95 – 0.98 mg/kg. Trace metals concentrations (mg/kg) in periwinkle ranged as 0.848- 9.719, 4.325-6.115, 5.917-9.013, 4.369-7.395, 0.619-1.462, 3.712-5.653 and 2.911-5.614 for Cd, Cu, Zn, Cr, Ba, Pb and Ni respectively. While the concentrations of the PCB were within regulatory limits; those of trace metals were all more than the permissible limits by World Health Organization (WHO). It is necessary to consistently monitor the levels of these pollutants in the rivers as they can contaminate the food chain thereby inducing toxicities in humans.

Keywords: Polychlorinated biphenyls, trace metals concentrations, food chain, contamination.

1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are group of non-polar synthetic toxic chemical compounds consisting of 209 congeners (Adeyemi et al., 2009; Kampire et al., 2015). They are classified as persistent organic pollutants (POPs) (Igbo et al., 2018; Yawei et al., 2008), and are bio-accumulative causing serious damage to the environment and have been identified worldwide (Ahmed, 2003; Hu et al., 2010; Cimenci et al., 2013; Gdaniec-Pietryka et al., 2013). They are of global concern due to their persistency, bio-accumulative properties, and their toxicity to humans and wildlife (United Nations Environmental Programme UNEP, 2016; Yawei et al., 2008). Polychlorinated biphenyls (PCBs) are either oily liquids or solids and are colorless to light-yellow having no smell or taste. PCBs do not occur naturally in the environment. They are water soluble organic compounds consisting of the biphenyl structure with two linked benzene rings in which some or all the hydrogen atoms have been substituted by chlorine atoms with a chemical formula $C_{12} H_{10-x} Cl_x$, where x=1-10 (World Health Organization WHO, 2000; Igbo et al., 2018). PCBs are introduced into the environment primarily as a result of anthropogenic activities (Iwata et al., 1993).

Due to their resistance to electrical, thermal and chemical processes, it is used in a wide variety of application since their commercial production in 1929 (Anyasi & Atagana, 2011). PCBs are used and applied widely in dielectric fluids such as in transformers, capacitor, coolants, lubricant and other agricultural purposes. Their resistivity to degradation and low flammability also enabled it use as additives in sealants and as heat transfer agents (Erickson & Kaley 2011; Hagmar, 2003). PCBs, apart from making capacitors flame-resistant they also allowed capacitors to be made smaller, hence lowering cost of equipment (Agency for Toxic Substances and Diseases Registry ATSDR, 2000). Notably, PCBs are taken up by small benthic organisms like periwinkle (*L. littorea*) and fish in sediments and water. They are also taken up by other animals that eat these aquatic animals as food. PCBs accumulate in fish and marine mammals, reaching levels that may be many thousands of times higher than in sediments and water. PCBs influence patterns of survival, reproduction, growth, enzyme activities and accumulation in representative aquatic organisms. PCB can also cause mortality or lethality in organisms when exceeded the normal body dosage.

Food chain contamination by heavy metals is also a growing global issue and has currently reached an alarming rate. Heavy metals are metals having density greater than $5g/cm^3$ (Ademoriti, 1999). Heavy metals occur in the environment with large variation in concentrations. The increase in the level of the heavy metals in the environment from various anthropogenic sources has become a source of major concern for environmentalist (Opeolu et al., 2008) and a worldwide scientific problem because they are not degradable and most of them have toxic effects on organisms (Abiaobo & Asuquo, 2020). Heavy metals enter the environment from various natural and anthropogenic sources such as rocks, soil, fertilizers, oil spills and discharge from industrial wastes. Heavy metals are toxic or poisonous at low concentration. They are not degraded, when released into the environment, they tend to persist indefinitely accumulating in living tissues through food chain. In aquatic environment, heavy metals in dissolved form are easily taken up by aquatic organisms and accumulate in their tissues and may affect organisms directly by accumulation in their body or indirectly by transferring to the next tropic level of the food chain. Simultaneously, PCBs and heavy metals can be released from the sediment-water interface under certain conditions and then ingested by benthic organisms and further accumulate in the food chain (Zhao et al., 2009). PCBs and heavy metals concentrations are detected in lipid-rich tissues such as liver and muscle of fatty fish. Periwinkle (*L. littorea*) consumption is an important constituent of diets among the people of Rivers State and Niger-Delta at large. It contains massive amount of omega -3 fatty acids which in turn lowers cholesterol, cancer risk and blood pressure levels. It is also rich in proteins and vitamins for growth and development and good healthy body (www. greenfact.org). However, despite the beneficial aspects of consumption, periwinkle (*L. littorea*) may contain contaminants such as PCBs and trace metals. This study therefore is undertaken to determine the concentrations of PCBs and trace metals in tissues of periwinkle (*L. littorea*) in some rivers in Rivers State.

2. Materials and Methods

2.1 Sample Collection

Eighty samples of periwinkle (*L. littorea*) used for the study were collected from each of these rivers, Kaa, Okrika, Borokiri and Eagle Island rivers of River State using the composite technique.

2.2. Sample Preparation

2.2.1 Sample preparation, extraction and cleanup of Periwinkle (*L. littorea*) for PCB and Trace metals analysis

The tissues (edible parts) of the periwinkle were extracted from the shell with an acetone-rinsed knife. The periwinkle tissues were rinsed thoroughly with tap water and then with distilled water. For PCB analysis, samples from each location were crushed, homogenized and mixed with anhydrous sodium sulphate (1:4). The extractions of PCBs from the tissues were performed using 1,1,2-trichlorotrifluoroethane. The extract was later purified to reduce interference. For trace metals analysis, 2g of the crushed periwinkle samples were digested using 10ml of nitric acid and 2ml of perchloric acid.

2.3 Instrumental Analysis

2.3.1 Determination of PCBs

PCBs were analyzed using a gas chromatography (Agilent 7890A) equipped with flame ionization detector (GC-FID) system on Chemstation software with injection of 1µg on a programmable temperature operating in splitless mode of 250^0C. Helium was the carrier gas.

2.3.2 Determination of Trace Metals

The digested periwinkle samples were subjected to analysis for trace metals (Cd, Cu, Zn, Cr, Ba, Pb and Ni) by the use of Solar Thermo Elemental Atomic Absorption spectrophotometer (AAS) model SN. SG-710960.

2. 4 Statistical Analysis

The concentrations obtained from the analyses were recorded as mean ± SD. Descriptive and inferential statistics were employed in the analysis of the obtained results. Particularly the mean, standard deviation and the t-test were used. Possibilities less than 0.05 ($P < 0.05$) were considered as statistically significant.

3. Results and Discussion

3.1 Concentrations of PCBs in periwinkle *(L. littorea)*

As depicted in Table 1, the concentrations of PCBs in periwinkle from Okrika river was 0.98 mg/kg. PCBs in the periwinkle (*L. littorea*) from Borokiri and Eagle Island were 0.95 and 0.96 mg/kg respectively. Hence, PCBs in the periwinkle (*L. littorea*) sample ranged from 0.95-0.98 mg/kg with 100% of the samples having PCB concentrations below the permissible limit set by United State Food Drugs and Administration (USFDA). The low concentrations of PCBs in the periwinkle obtained from Okrika, Borokiri and Eagle Island may be attributed to less industrial activities that use/produce PCB products/wastes. The low concentrations of PCB may also be connected to dilution from the river water. PCB sources may also be from domestic wastes. Notably, the PCBs in the periwinkle samples from Kaa river was below the detection limit of the GC. This may be attributed to the fact that the domestic waste generated in the area contains very low PCBs.

To show if there is significant difference between the PCB concentrations from the various sampling locations, a one-sample t-test was employed. The results indicate that no statistically significant difference was found in the concentrations of PCBs in periwinkle collected from the different sampling locations ($p>0.05$). Elsewhere in the State and overseas, the levels of PCBs have been determined with values either above or below the concentrations determined in the present study. In Southwest Nigeria, the distributions of polychlorinated biphenyl (PCB) congeners were determined in sediments and Biota from the Ogun and Ona rivers, southwest Nigeria (Adeogun et al., 2016). The results revealed significant site-related PCB congener distribution patterns for higher molecular weight (HMW) PCB in samples from the Ogun river (71.3%), while the Ona river (42.6%) showed significant PCB congener patterns for low-molecular-weight (LMW) congeners. Igbo et al., (2018) reported the levels of PCBs in water, sediments and Biota *(Tilapia guneensis, Callinectes amnicola and Cardiosoma annatum)* from E-waste Dumpsites in Lagos and Osun States, South-West Nigeria. In their studies, 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.

Zhao et al., (2009) studied the spatial distribution and bioaccumulation of organochlorine pesticides in surface sediment and benthic organisms from Taihu Lake, China. The results of the study revealed that OCPs were detected in all sediment samples with concentrations ranging from 4.22-461 ng g(-1) dw.

In another study Kampire et al., (2015) in the North End Lake in Port \Elizabeth, South African, the concentrations and distributions of 6 PCB indicator congeners (IUPAC nos. 28, 52, 101, 138, 153, and 180) were measured in 236 organ samples of fish *(Cyprinus carpio and Oreochromis mossambicus)*. The findings of the study revealed that the concentrations of total PCBs in the liver, gonads, gills and muscle were 95.69, 57.49, 44.63, 34.14 ng·g-1 lipid weight (lw) in *C. Carpio* and 119.73, 59.21, 49.78, 34.63 ng·g-1 (lw) in *O. mossambicus*, respectively.

Notably, PCBs are taken up by small organisms and fish in sediments and water. They are also taken up by other animals that eat these aquatic animals as food. PCBs accumulate in fish and marine mammals, reaching levels that may be many thousands of times higher than in sediments and water. PCBs influence patterns of survival, reproduction, growth, enzyme activities and accumulation in representative aquatic organisms. PCB can also cause mortality or lethality in aquatic organisms when exceeded the normal body dosage. PCBs especially those with 2, 3, 7, 8-tetrachlorodibenzeno dioxin (TCDD) type activity adversely affect reproductive sources success of spawning female Chinook salmon. PCBs can also impair the reproductive capacities of marine mammals. Mixtures of planner PCBs and Dioxins, however produces synergism of Aquatic Animal Health (AAH) activity in fish liver at low rate and antagonism at high dose. These substances have the tendency to bio-accumulate in organisms.

3.2 Concentration of Trace Metals in periwinkle (*L. littorea*)

The concentrations of trace metals in periwinkle (*L. littorea*) samples are presented in Table 2**.** These were able to provide information on the extent of contamination of the periwinkle from Okrika, Borokiri, Eagle Island and Kaa rivers.

Cadmium (Cd)
The concentrations of Cd observed in periwinkle (*L. littorea*) samples from the various sampling stations ranged from 0.848-1.719mg/kg (mean ± SD=1.289±0.483) with highest concentration recorded in Eagle Island while the least concentration was recorded in Kaa. This obtained concentration is higher than the permissible limit of 0.050 mg/kg in sea food (shell fish) by WHO (2000).
The concentration obtained was higher than that recorded from Uta Ewa creek, Imo river estuary, South Eastern Nigeria (Abiaobo & Asuquo, 2020) and upper reaches of the Bonny estuary, Nigeria (Moslen et al., 2017). The high concentrations of Cd recorded in the tissues of periwinkle (*L. littorea*) in the present study areas may be attributed to bioaccumulation of the metals in the organism. High concentrations of Cd have been reported in fish which may pose some health challenges (Ekpete & Obunwo, 2016). Cd in some enzymes alters the stereo-structure of the enzymes and impairing its catalytic activity (Bhatia, 2006).

Copper (Cu)
The obtained concentrations of Cu in the tissues of periwinkle (*L. littorea*) samples ranged from 4.325 -6.115 mg/kg (mean ± SD= 5.535±0.823) which is higher than the WHO permissible limit of 0.50 PPM in fish (including shell fish). Highest concentration of Cu was observed in sample from Okrika while the least in sample from Borokiri. The concentration obtained in the present study is not at variance with the concentration reported in some marine species particularly periwinkle (*L. littorea*) collected during different seasons from Ondo coastal region, Nigeria (Ololade et al., 2008).

The high concentrations of Cu observed in periwinkle samples collected from Okrika, Borokiri, Eagle Island and Kaa rivers may be attributed to anthropogenic activities such as steel fabrication, electrical production, mining and farming (Ekpete et al., 2019). Cu is essential for the growth of plants and animals. In humans, it helps in the production of blood hemoglobin. In high doses Cu cause anaemia, liver and kidney damage, stomach and intestinal irritation.

Zinc (Zn)

The concentrations of Zn in the tissues of periwinkle (*L. littorea*) samples from the various sampling locations in the rivers ranged from 5.917-9.013mg/kg (mean ±SD=7.669± 1.336).

These are higher than the value of 5.0 PPM specified for human consumption by WHO. The highest concentrations of Zn were observed in samples from Kaa river while the least concentrations were observed in samples from Okrika river. The concentrations of Zn observed in periwinkle samples in the present study is higher than those observed in periwinkle (*L. littorea*) collected during different seasons from Ondo coastal region, Nigeria (Ololade et al., 2008), were the concentration was as low as 0.10±0.11 and 0.11±0.09mg/kg for the dry and wet seasons respectively and those observed in periwinkle *(Tympanotonus fuscatus)* samples from Uta Ewa creek, Imo river estuary, South East Nigeria whose concentrations ranged as 1.08 – 2.29 mg/kg (1.70±0.50 mg/kg) (Abiaobo & Asuquo, 2020). However, the Zn mean ±SD values of 18.34±2.091 mg/kg reported by Nwineewii et al., (2019) in crab *(Potamon fluviatile)* from the New Calabar river of Rivers State is higher than concentrations observed in present study.

Zn occurs naturally in air, water and soil. Zn is essential for a healthy body, but excess Zn could be harmful and cause health effect. Zn have been reported to be toxic to bacteria, plants, invertebrates and vertebrate fish (Rout & Das, 2003; Muyssen et al., 2006).

Table 1: Concentrations (mg/kg) of PCB in periwinkle *(L. littorea)* from various stations.

S/N	Sampling Stations	Concentration (mg/kg)	USFDA (ppm)
1	Okrika river	0.98	2.00
2	Borokiri river	0.95	2.00
3	Eagle Island river	0.96	2.00
4	Kaa river	BDL	2.00

BDL = Below Detection limit

Table 2: The concentrations (mg/kg) of trace metals in periwinkle from various sampling stations.

Sampling Stations	**Cd**	**Cu**	**Zn**	**Cr**	**Ba**	**Pb**	**Ni**
Okrika river	0.894	6.115	5.917	6.192	1.462	3.712	3.917
Borokiri river	1.693	4.325	7.423	7.395	0.619	5.653	5.614
Eagle Island river	1.719	5.729	8.315	5.713	0.830	4.061	4.604
Kaa river	0.848	5.972	9.013	4.369	1.327	4.326	2.911
Mean ± SD	1.289±0.483	5.535±0.823	7.667±1.336	5.917±1.251	1.060±0.400	4.438±0.848	4.261±1.139
WHO Permissible Limits	0.050	0.50	5.000	0.050	1.000	0.050	0.50

Table 3: Descriptive statistics of trace metals in periwinkle

	Minimum	Maximum	Mean	Std. Error	Std. Deviation	Variance
Cd	0.848	1.719	1.289	0.241	0.483	0.233
Cu	4.325	6.115	5.535	0.411	0.823	0.677
Zn	5.917	9.013	7.667	0.668	1.336	1.785
Cr	4.369	7.395	5.917	0.626	1.251	1.566
Ba	0.619	1.462	1.060	0.200	0.400	0.160
Pb	3.712	5.653	4.438	0.424	0.848	0.719
Ni	2.911	5.614	4.261	0.569	1.139	1.297

Table 4: One sample T-test of trace metal concentrations in Periwinkle

	Test Value = 0					
					95% Confidence Interval of the Difference	
	T	Df	Sig. (2-tailed)	Mean Difference	Lower	Upper
Cd	5.340	3.000	0.013	1.289	0.521	2.056
Cu	13.459	3.000	0.001	5.535	4.226	6.844
Zn	11.479	3.000	0.001	7.667	5.541	9.792
Cr	9.457	3.000	0.003	5.917	3.926	7.909
Ba	5.298	3.000	0.013	1.060	0.423	1.696
Pb	10.465	3.000	0.002	4.438	3.089	5.788
Ni	7.484	3.000	0.005	4.261	2.449	6.073

Chromium (Cr)
The observed concentrations of Cr in tissues of periwinkle (*L. littorea*) samples from the various sampling stations ranged from 4.369-7.395 mg/kg (mean ±SD=5.917±1.251mg/kg) with highest concentrations observed in samples from Borokiri while the least concentrations were observed in sample from Kaa river. These values are higher than the recommended limit of 0.050 PPM set by WHO. The mean ± SD of Cr reported in crab *(Potamon fluviatile)* from the New Calabar river by Nwineewii et al., (2019) is higher than the mean ±SD observed in the present study. However, the concentrations observed in the present study is higher than concentrations observed in periwinkle *(Tympanotonus fuscatus)* samples from Uta Ewa creek, Imo river estuary south eastern Nigeria (Abiaobo & Asuquo, 2020). Cr occurs in several oxidation states in the environment ranging from Cr^{2+} to Cr^{6+} (Rodriguez et al., 2007) but preferably, Cr^{3+} and Cr^{6+} are known stable states (Ekpete et al., 2019) and are toxic to animals, humans and plants (Mohanty & Kumar-Patra, 2013). Cr upset stomach and ulcer as well alteration of genetic material. High concentrations of Cr in tissues of periwinkle observed from various sampling locations may be attributed to industrial activities, sewage and fertilizers (Ghani, 2011).

Barium (Ba)
The concentrations of Ba in periwinkle (*L. littorea*) ranged from 0.619 –1.462 (mean± SD=1.060±0.400mg/kg). The concentrations of the metal however varied from location to location. The concentrations observed in samples from Okrika and Kaa rivers were slightly higher than the recommended limit of 1.000 PPM set by WHO as their concentration were 1.462mg/kg and 1.317mg/kg respectively. The concentrations 0.619mg/kg and 0.830mg/kg were observed in samples from Borokiri and Eagle Island which was below the standard limit set by the WHO. Ba is most commonly found as the mineral barite ($BaSO_4$) and witherite ($BaCO_3$) and is primarily produced through the electrolysis of barium chloride ($BaCl_2$). $BaCl_2$ is used as water softener. A small amount of water soluble barium may cause breathing difficulties, increase blood pressures, stomach irritation, muscle weakness, swelling of brain and liver, damage kidney and heart.

Lead (Pb)
The observed concentrations of Pb in the periwinkle (*L. littorea*) samples from the various sampling stations ranged from 3.712–5.653mg/kg (mean±SD = 4.438±0.848mg/kg). These are higher than the 0.050 PPM value stipulated by WHO. The highest concentrations of Pb were recorded in samples from Borokiri river while the least was recorded in samples from Okrika river. The concentrations of Pb observed in the tissues of periwinkle in the present study is higher than value observed by Onwuli et al., (2014) in periwinkle *(Tympanotonos fuscatus)* from Eagle Island river, Port Harcourt, Rivers State and Moslen et al., (2017) from Azuabie creek, Okujagu axis, Bonny estuary. Pb concentrations may be connected with illegal oil bunkering activities, local dredging and discharged effluents and agricultural farmlands (Ekpete et al., 2019).
The consumption of foods contaminated with lead is worrisome and may pose some heath challenges (Ekpete & Obunwo, 2016) such as loss of appetite, headache, hypertension, abdominal pain, sleeplessness, arthritis, mental retardation, birth defects, brain and kidney damage and even death (Martine & Griswold, 2009).

Nickel (Ni)
The concentrations of Ni in the tissues of periwinkle (*L. littorea*) samples from the various stations varied from 2.911– 5.614 mg/kg (mean±SD = 4.261±1.139mg/kg). These are higher

than 0.50PPM permissible limit set by WHO. The highest concentration of Nickel was observed in sample from Borokiri river and the least observed in samples from Kaa river. The concentration of the metal in tissues of periwinkle in the present study is lower than concentrations observed in crab *(Potamon fluviatile)* from the New Calabar river (Nwineewii et al., 2019).

The source of Ni in the tissues of periwinkle from the various sampling locations might be from accumulation of domestic effluents and agricultural discharges as well as illegal oil bunkering and burning of fossil fuels. Ni is essential in small doses, but can be dangerous when the maximum tolerable amounts are exceeded. Concentration of Ni greater than normal concentration in food causes lung disease in dogs and rats and affects the stomach, blood, liver, kidneys and immune system in rats and mice (Nwineewii & Edem, 2014) and possibly humans. To show if there is significant difference between the trace metals concentrations from various sampling location, a one-sample t-test was employed. The results shown in Table 4 indicate that statistically significant differences were found in the concentrations of all the metals in periwinkle collected from the different sampling locations ($p < 0.05$).

4. Conclusion

The findings of the study clearly indicate levels of these contaminants (PCBs and trace metals) in periwinkles. PCB concentrations in periwinkle from Okrika, Borokiri and Eagle Island rivers were below the regulatory limit set by USFDA while periwinkle sample from the Kaa river contains no PCB. The concentrations (mg/kg) of trace metals Cd, Cu, Zn, Cr, Pb, Ba and Ni in periwinkle (*L. littorea*) from the various sampling locations were at elevated levels. These high concentrations can be injurious to human health as they were above regulatory limit set by the WHO except for Ba in Borokiri and Eagle Island rivers which were below WHO regulatory limit.

The levels of these metals reported in this study can pose great threat to the aquatic organisms and humans. Humans can suffer carcinogenic and non-carcinogenic effects such as hepatotoxicity, nephrotoxicity, immunotoxicty and even brain tissue damage following exposures. Monitoring the levels of the contaminants in these rivers will play a critical role in identifying levels beyond regulatory limits which may induce some forms of toxicity following entry into food chain and food web.

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