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Ecological and Human Health of Polychlorinated Biphenyls (PCBs) Across Tide and Saline Influenced Hydrocarbon Mining Zones of Tropical Deltaic Wetlands, Southern Nigeria

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Abstract: Polychlorinated Biphenyls (PCBs) concentrations across the Lower Orashi and Sombriero which are tide and saline influenced were measured across eight (8) locations. These unique environments are affected by hydrocarbon mining especially in the Tropical Deltaic Wetland zones. Sediment and water samples were analyzed to ascertain potential human and ecological implications. Standard laboratory methods were adopted for physico-chemical parameters of water while PCB sequencing/speciation was done by Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The PCB speciation chemistry reveals non-similarity in their characteristic bioabundance across all stations. However, similarity exists in PCBs concentrations in the marine media. PCB_{water} locational dominance is in the order: SR3 (104.02) > TZ2 (5.16) > SR2 (3.60) > TZ1 (3.35) > OR3 (3.04) > OR2 (2.91) > SRI (2.39) > OR1 (1.38). $\sum PCB_{water}$ (125.85) > $\sum PCB_{sediment}$ (47.02), implying the surface waters bioaccumulates more PCBs than sediments comparatively. PCB-66 2,3',4,4'-Tetrachlorobiphenyl (5.13) ranks the highest ecotoxin in the study area. SR3 and OR1 ranks highest and least in PCBs locational concentrations respectively. Ecologically, Sombriero River (SR = 110.01, %) > Transition Zone (TZ = 8.51, %) > Orashi River (OR = 7.33, %) in PCB bioabundance, indicating locational and media variations at P < 0.05. The results show riparian populations' exposure to PCBs posing likely health impacts. Review, monitoring and stringent enforcement of importation, application, disposal regulations of PCB laden equipment and appliances is recommended amongst others.

Keywords: *Environmental Pollution, Human Health Assessment, Organochlorine, Niger Delta, Lower Sombriero, Orashi Zon.*

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I. INTRODUCTION

Polychlorinated Biphenyls (PCBs) are artificial/ synthetic (man-made) ecologically bioavailable, bioaccumulative and biomagnifying persistent organochlorine range of industrial chemicals. These chemicals possess multiple chlorine compounds (Gerald and David, 2018). The chemical formula for PCBs is ($C_{12}H_{10-x}Cl_x$). PCBs were first manufactured in the USA by Messrs. MOSANTO (Gerald and David, 2018; OSPAR Commission, 2020). PCBs are highly toxic to marine and freshwater ecosystems, aquatic species; and are human carcinogens (Zhang et al., 2019). The Hazardous Substances in the Baltic Sea Commission, HELCOM (2010), documented that PCBs are water insoluble but soluble in fats and acetone/hexane solutions, and also share great affinity with suspended particulate matter (SPM) in surface water, lithogeneous, hydrogenous, biogeneous and cosmogeneous sediments (Jing, Fusi and Kjellerup, 2018; Obanya, Ntor, Okoroafor and Nwanze, 2019).

PCBs have wide industrial and commercial applications in manufacturing and urbanization equipment such as electrical coolants in capacitors and transformers; petrochemical lubricants, hydraulic fluids, paints and dyes, pesticides and wide range of household appliances

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(Filipkowski, 2013). Hence, they are widely distributed in space especially in developing countries.

Globally, ecological systems, particularly the marine ecosystems provide for and sustain man and aquatic species, with food, water, fishes (pelagic, mesopelagic and benthic), spirituality, wildlife, coral products, sea mammals, crustaceans, relaxation, medicines, energy, transportation, economic and recreation, essential faunal communities, embedded natural resources and climate change mitigation characteristics (Ute et al., 2020; Otiasah, et al., 2022).

Studies have implicated pollution of global marine ecologies to introduction of novel anthropogenic chemicals and related substances of organic and inorganic origins inimical to and beyond the carrying capacities of these ecosystems. Loss of marine productivity, biological diversities and ecosystem services are direct consequences of declining ecosystems' health occasioned by polluting chemicals (Otiasah, Ezekwe and Lawal, 2020).

Polychlorinated biphenyls (PBCs) are a range of highly persistent ecotoxic chemicals inimical to marine ecosystems' health. PCBs environmental persistence enables long distance travel including spatially remote environments allochthonous to its point of entry, aided by meteorological and fluvial conditions (Breivik, 2007 in Filipkowski, 2013). Six basic structural PCBs exist including the Single-Sided, Double-Sided, Multilayered, Rigid, Flex and Rigid-Flex PCBs; and their 209 different yields/mixtures popularly referred to as "Congeners" in global environment (Ibrahim et al., 2018). They have further characteristics such as chemical stability, non-inflammability, and high boiling point and usefulness in a wide range of electrical applications (EFSA, 2010; Zhang et al., 2019; Tang et al., 2020).

PCBs are further classified into Indicator (Ind-PCBs) and Co-Planer PCBs (Co-PCBs). Indicator PCBs (ind-PCBs) generally viewed as a measure of the bioavailability and quantitative abundance whereas Co-Planer PCBs (Co-PCBs) gives a measure of their hyper-reactivity and ecotoxicity (Igbo et al., 2018). Seven basic ind-PCBs have their IUPAC numbers 28, 52, 101, 118, 138, 153 and 180. Ind-PCBs have their chlorine atoms from three to seven. On the hand, two groups of 68 coplanar congeners exist. The first set consists of 20 referred to as CP0 with their chlorine substitution at none of the ortho positions on the biphenyl. The second set of 48 congeners referred to as CP1 or mono-ortho congeners have their chlorine substitution at only one of the ortho positions. 4CL 169 congeners have four or more chlorine substituent notwithstanding the ortho position. PP are 54 congeners having all para position chlorinated. 2M congeners are 140 in number having two or more of the meta positions chlorinated (Xiaoai et al., 2020).

Ute et al., (2020) observed that PCBs were classified by various international frameworks in 2001 such as the Stockholm Convention on Persistent Organic Pollutants (POPs), the Baltic Marine Environment Protection Commission - an integrated thematic assessment of hazardous substances in the Baltic Sea - HELCOM, the Commission for the Protection of the Marine Environment of Northeast Atlantic - OSPAR and Programme for the Assessment and Control of Pollution of the Mediterranean River - MEDPOL; as 'persistent organic pollutants' (POPs).

PCBs have equally been classified as "probable human carcinogens" causing a variety of cancer in the human body (Ibrahim et al., 2018; Zhang et al., 2019; IARC 2019). Other probable human carcinogens are Formaldehyde, Benzene, Benzidine, Asbestos, Butadiene, Ethylene oxide, Aminobiphenyl, Vinyl chloride, Erionite, Benzo[a]pyrene, Acetaldehyde, Cadmium, Busulfan, Acrylamide, DDT, Aflatoxins, Aristolochic acids, Hexavalent chromium, Trichloroethylene, Dimethyl sulfate, Benz[a]anthracene and N-Nitrosodimethylamine (IARC, 2019; IUPAC Glosary of Toxicology and US National Library of Medicine Technical Bulletine, 2019; WHO, 2020).

Studies have reported safe levels of ecological and human PCBs bioabundance in Nigeria and global marine media while others documented serious adverse public health implications arising from exposure to PCBs. Human PCBs exposure risks conditions include developmental and nervous system impairment in fetuses, low birth weights and body sizes in infants; circulatory and hastened senility, dysfunctional immune system, endocrine and digestive system hormones disruptions (USEPA, 2000, 2004; Filipkowski, 2013). In adults, visual recognition, protracted muscles development, degrees of memory loss, cancers of the brain, biliary, gastrointestinal tracts, gall bladder, liver, lungs, breast; and increased melanomas have been documented (FAO/WHO, 1999; JECFA, 2000; USEPA, 2014a; 2014b and 2015). EFSA (2010) identified neurological and psychomotor control ailments such as low intelligence quotient (IQ) and dysfunctional memory, obscured head size of infants and abnormal behaviors amongst children whose mothers were exposed to PCBs prior to or during pregnancy.

Carcinogenicity, chronic and acute neurological ailments, hypothyroidism, reproduction difficulties, high serum lipids, infertility, cardiovascular dysfunction, liver and lung cancers, asthma and arthritis are some other public health impacts of PCBs exposure (Breivik, 2007 in Filipkowski, 2013). Similarly, growth retardation, low sperm count, stunted puberty development, alteration and malfunctioning of human hormones and reproductive organs, estrogen mimics, incomplete menstrual circle, decreased metabolism of sex steroids, irregularity in estrogen and testosterone levels are other public health impacts of PCBs (ICES, 2008; Sonal, Ajay-Moham and Vikas, 2022). PCBs transforms from higher to lower chlorinated forms with greater human estrogen toxicological hyper reactivity (Molenie, 2018). PCBs are equally responsible for disequilibrium of the immune and thyroid hormones causing taunted growth. Specifically, dioxin enjoys great affinity with body receptors where they retard production of lymphocytes in proper abundance (USEPA, 2000; Weisglas-Kuperus et al, 2000 and EFSA, 2010).

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Xiaoai et al., (2020) documented hydrodynamics; median particulate size/crystal size, temperature, covalent bonds, mineral composition and the effect of van der Waals forces as major drivers of PCBs bioabundance in marine ecosystems. Similarly, PCBs transport/diffusion from sediment to water is enabled when the fugacity fraction $ff_{sw} \geq 0.50$, while at $ff_{sw} \leq 0.50$ that of water to sediment is enabled. PCB transportation/redistribution/diffusion is from higher to lower fugacity. Net redistribution of PCBs/xenobiotics is zero for chemicals of equal fugacity (ie, Equilibrium State).

Ingestion of contaminated fish from bioaccumulated benthic and pelagic planktons, algae, invertebrates and during swimming; transported along the delicate food chain to higher trophic levels and humans in addition to atmospheric inhalation has been identified as major pathways for human PCB exposure (Nizzetto et al, 2012 reported in Ute et al, 2020). Evaporation of contaminated surface and settled water are means of atmospheric PCBs diffusion inhaled by humans. PCBs uptake from contaminated soils by plants transported along the food web to humans through consumption of meat and dairy products from animals grazed with such affected plants are other sources of human exposure (Mai et al, 2016; Sonal et al, 2020).

Despite global PCBs ban in the 1980s due to their high ecotoxicity and public health implications from consumption of contaminated water, fish and other seafood, studies have observed that fish PCB contamination increased by 177% afterwards forcing issuance of food consumption advisories by various nations. The report also indicated that over two million (Something is missing here) of total sediments and surface water bodies were affected by one or multiple PCB contaminations globally (HELCOM, 2010; OSPAR Commission; 2020).

Tissues of marine whales, dolphins, birds, seals and reptiles are destroyed by PCBs bioaccumulation for their lipophilic characteristics (Gideon et al., 2020; Sonal, Ajay-Moham and Vikas, 2022). Bioaccumulated PCBs undergo upward transportation along the fragile food chain increasing biomagnification which adversely affect the health and reproductive abilities of top predators; and are eventually taken up by man (WHO, 2020). PCBs have been implicated in the reduction in the quantity and mortality of eggs, leading to reproduction disruption and declining vulnerable marine animals, mammals and seabird populations (Molenie, 2018; Zhang et al., 2019; Tang et al., 2020). The immune systems of marine organisms are systematically damaged and made dysfunctional by PCB exposure making them vulnerable to diseases. Disruptions of the immune systems of marine animals cause birth difficulties, growth and developmental abnormalities; and deformities in skeletal structures. Finally, altered behavioral characteristics (schooling, shoaling, foraging, courtship and mating etc) are documented to be consequences of high levels of PCBs exposure (USEPA, 2014a; 2014b and 2015; Wang et al., 2019).

Obanya et al (2019) carried out a Gas Chromatography-Electron Capture Detector (GC-ECD) analytical study of the seasonal stoichiometric abundance of PCBs in three marine matrices of sediment, surface water and blackchin tilapia fish (sarotherodon melannotheron) of the Ologe Lagoon, a major effluent receiver. The study found concentration levels of PCBs in all stations and media to be within safe ecological and public health limits except in the surface water of a particular zone during the rainy season. Only 2,2,4' – Trichlorobiphenyl or PCB28 (2,2,4' TCB/PCB28) congener was reported in the media. Concentrations in sediment ranges from 0.0033 ± 0.00333 to 0.430 ± 0.00351ng/g. 2,2,4' TCB/PCB28 abundance was reported to be lower in the sediment compared to the surface water and fish samples from all stations. Variations were observed within and across matrices of the 2,2,4' TCB/PCB28 levels in sediment and surface water, but not in fish. However, concentration of 2,2,4' TCB/PCB28 in the fish was below WHO allowable limits for food, indicating that the fish of Ologe River were safe and healthy for consumption; wherein, continuous monitoring was recommended notwithstanding.

In a comparison study of concentrations of PCBs in two locations - Mediterranean Sea and Southwestern Scotland, Filipkowski, (2013) found quantitative abundance of most congener PCBs within permissible thresholds except one. Of the two marine environmental matrices (sediment and fish), elevation of PCBs were reported to be lower in the sediment compared to fish irrespective of levels of contamination and location. Finally, the study acknowledges greater difficulty in the determination of PCBs of low molecular weight (Tri & Tetrachlorinated PCBs) compared to those of higher weights (Penta & Hexachlorinated PCBs).

Studies have traced African and Nigerian environmental entry and fate of PCBs to high patronage of imported electrical and electronics equipment and their arbitrary disposal, including drainage of terrestrial wastes of electrical, medical, automobile, civil and mechanical origins; and direct atmospheric deposition into the surface waters (Amiard-Triquet, 2015. These sources contribute to the marine ecosystem as the greatest global pollutant sink. PCBs are transboundary, non-aboriginal and allochthonous in presence from their entry points and sources (Unyimandu et al., 2017; Unyimandu et al., 2018a; Unyimandu et al., 2018b).

The Lower Orashi and Sombriero contiguous River systems are vital ecosystems to Nigeria that supports various ecological, economic and transportation activities; including fishing, agriculture, urbanization, mining, manufacture and related industrial processes; thus, leading to pollution of the marine environment and PCB contamination. According to Otiasah, Otiasah & Ohanuna (2024), the Lower Orashi and Sombriero River systems is host to ten (10) crude oil and gas fields, a gas plant, four crude oil transportation trunk lines and other hydrocarbon exploration, production, liquefaction and transportation assets. Otiasah, Ezekwe, B abatunde & Otiasah (2023) reported that population in the study area are rural fishermen, farmers, hunters, petty

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traders on marine resources and artisans who depend entirely on the resources of the surrounding Rivers and wetlands for their daily sustenance. Contamination of their River waters and sediments pose grievous existential consequences to them. Furthermore, population growth, rapid urbanization, increased hydrocarbon (oil and gas) mining activities, infrastructural development, industrialization, massive importation of foreign chemicals, materials and equipment containing PCBs in the study area and their drainages has made scientific knowledge of PCBs in the coastal marine ecology inevitable. Specific scientific study or data on PCB contamination in the surface waters and sediments of these contiguous River systems is non-existent in present knowledge. This study therefore investigated the sources, levels of bioabundance and spatial characteristics of Polychlorinated Biphenyl (PCBs) in the surface waters and sediments of the Lower Orashi and Sombriero Rivers Systems, Rivers State, Nigeria; to ascertain their potential ecological and human health implications with a view to emplacing effective remediation and monitoring protocol in the wider Niger Delta marine ecological environment.

II. MATERIALS AND METHODS

The Lower Orashi and Sombriero River estuaries covering territories of Ahoada West, Abua/Odual, Degema, Akuku-Toru and Asari-Toru Local Government Areas of Rivers State, Nigeria (Lawal, 2015, Otiasah et al., 2024) (Figure 1). This constituted the study area. The area is described as a freshwater and saline mangrove wetland of coastal lowland rain forest vegetation dominated by economic trees (*pentaclethramacrophylla, chrysophyllumalbidum and irvingiagabonenesis*), of tidal and semi tidal flat mud characteristic of the Niger Delta (Abam & Okagbue, 1997; Niger Delta Development Commission NDDC, 2006). The Orashi, Sombriero, Santh Batholomew and Santa Barbara are the major Rivers shaping the entire flow pattern of the area. It is predominantly humid tropical climate with long annual rainy season spanning March - October, a shorter dry season between November -February, and two characteristic South-West Monsoon wind and North East trade wind (Oyegun, 1997). The monsoon is dominant during the rainy season while the trade wind dictates activities during the dry season. It is of a low-lying plain of fresh unconsolidated fluvial sediments of Quaternary Age, characterized by layered heterogeneous sediment structure of alternating sands, silts and clay, of the Benin, Agbada and Akata Formations (Abam & Okagbue, 1997). The soil is majorly Entisols and Inceptisols occasionally mixed with Alfisols and elevated ≤ 7meters above mean sea level (Awosika, 1995; Aweto, 2002). Most of its freshwater between 100 – 200m in the Benin Formation is 2000m thick; consolidated at near bottom depth, having sprouting shallow water aquifer (Ekundayo & Obuekwe, 2001). The area is characterized by subsistence farming on the parchment of lands, fishing, weaving, hunting, palm oil production, timber logging, canoe-carving, red palm oil making and wine tapping, artisans, petty trades, hydrocarbon mining and gas reserves of the tertiary Niger Delta (Otiasah, et al, 2023). The Orashi River, from Mbiama to Hulk is fresh water while the Sombriero River from Degema behe ading the Orashi River (Transition Zone) to Ebemaboko is saline water. Both Rivers are intertidal and waterlogged mud flatlands with complex strata of diverse flora/trees and rich fauna.

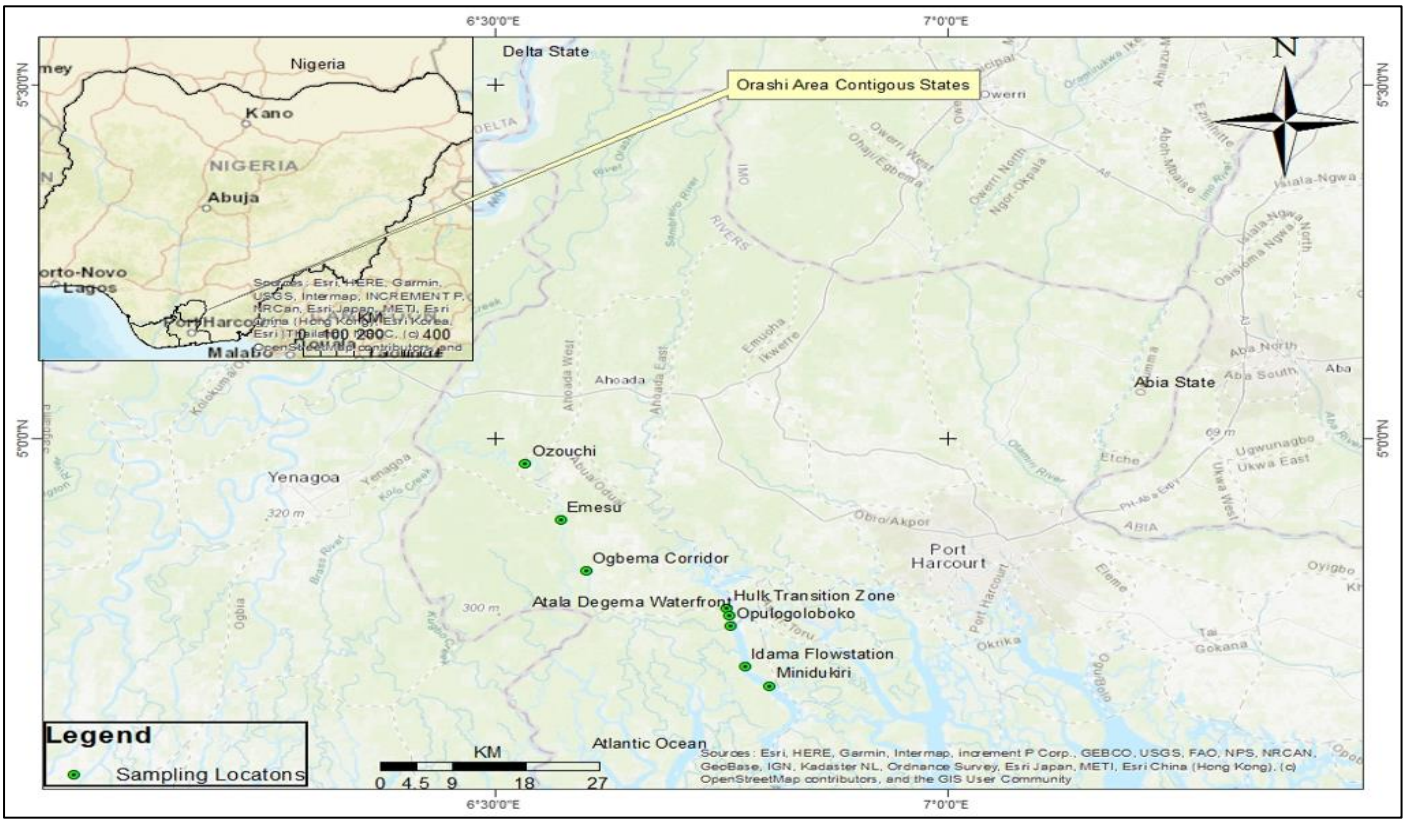

Fig 1 The Study Area Showing Sample Locations.

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➢ *Sample and Sampling Technique*

A total of eight (8) sampling sites were identified for this study, based on the suggestions of Otiasah et al, (2024). Water and marine sediment samples were collected in all eight sites. The sampling sites stretch from the Lower section of Orashi and Sombriero Rivers (Figure 1). Mbiama water front through the Sombriero River down to the feeding/entry point of the Atlantic Ocean specifies the sampling areas. Hulk (Agada)/Degema opposite points of the Transitional Zone (TZ) where the Sombriero beheaded the Orashi River, intersection point between the two River systems. Direct field measurements and results of laboratory analysis of surface waters and sediments for PCBs constituted the data for this study. Standard laboratory methods following the methods of Sharma et al., (2015) were adopted for physico-chemical parameters of water while Inductively Coupled Plasma Mass Spectrometer (ICP-MS) was used for PCBs sequencing/speciation. The data obtained in this study were analyzed with descriptive and comparative statistics.

Table 1 Sampling Sites and their GPS Coordinates

S/No	Sampling Sites	Designation	GPS
1	Ozuochi	OR1	E006º 32 02.0” N04º57 45.5”
2	Emesu	OR2	E006º 34 29.9” N04º53 03.1”
3	Ogbema Corridor	OR3	E006º 36 07.0” N04º48 44.3”
4	Hulk-Transition Zone	TZ1	E006º 45 31.4” N04º45 46.7”
5	Atala-Degema Waterfront	TZ2	E006º 45 51.4” N04º45 40.4”
6	Opulogoloboko	SR1	E006º 45 34.1” N04º44 08.3”
7	Idama Flow Station	SR2	E006º 46 34.7” N04º44 38 3”
8	Minjidukiri	SR3	E006º 46 52.5” N04º40 36.9”

III. RESULTS AND DISCUSSIONS

➢ *Polychlorinated Biphenyls (PCBs) in Water and Bottom Sediment*

Tables 2 and 3 present the results of PCBs speciation chemistry for surface water and bottom sediment from sections of the Orashi and Sombriero Rivers for the study respectively.

Table 2 Result of Speciation Chemistry for PCBs in Water Samples of the Study.

SN	PCBs COMPD, ppb	OR1	OR2	OR3	SR1	SR2	SR3	TZ1	TZ2	Mean
1	PCB-8 2,3’ Dichlorobiphenyl	0.01	0.88	0.43	N/D	0.91	N/D	0.09	0.06	2.38
2	PCB-18 2,2’,5-Trichlorobiphenyl	0.04	0.97	0.11	0.37	0.45	N/D	0.62	0.85	1.47
3	PCB-28 2,4,4’-Trichlorobiphenyl	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	
4	PCB-44 2,2’3,5’- Tetrachlorobiphenyl	N/D	N/D	N/D	N/D	0.67	N/D	N/D	N/D	0.67
5	PCB-52 2,2’5,5’-Tetrachlorobiphenyl	0.02	0.21	0.14	0.34	0.76	0.27	0.19	0.23	2.16
6	PCB-66 2,3’,4,4’-Tetrachlorobiphenyl	0.32	5.38	0.40	0.87	0.53	12.83	0.25	0.19	20.77
7	PCB-77 3,3’,4,4’-Tetrachlorobiphenyl	0.06	1.16	0.02	0.14	0.70	2.21	0.99	0.72	6
8	PCB-81 3,4,4’,5-Tetrachlorobiphenyl	0.13	N/D	0.12	0.85	0.92	3.23	0.40	1.56	7.21
9	PCB-101 2,2’4,5,5’ –Pentachlorobiphenyl	0.02	0.74	0.06	N/D	0.26	N/D	0.31	0.36	1.75
10	PCB-105 2,3,3’4,4’ –Pentachlorobiphenyl	0.05	1.59	0.11	0.40	0.22	N/D	0.39	0.44	3.2
11	PCB-114 2,3,4,4’,5–Pentachlorobiphenyl	0.02	N/D	0.04	N/D	0.56	0.20	N/D	0.26	1.08
12	PCB-118 2,3’,4,4’,5–Pentachlorobiphenyl	0.01	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.01
13	PCB-123 2,3’4,4’5’–Pentachlorobiphenyl	0.03	N/D	0.28	N/D	0.52	N/D	0.14	0.72	1.69
14	PCB-126 3,3’4,4’5’–Pentachlorobiphenyl	0.11	0.18	0.03	0.40	0.58	1.02	0.41	0.81	3.54
15	PCB-128 2,2’3,3’4,4’–Hexachlorobiphenyl	0.00	N/D	N/D	N/D	0.31	1.10	N/D	N/D	1.41
16	PCB-138 2,2’3,4,4’5’–Hexachlorobiphenyl	N/D	N/D	0.04	0.52	0.21	0.91	N/D	0.27	1.95
17	PCB-153 2,2’4,4’5,5’–Hexachlorobiphenyl	0.03	0.32	0.20	0.97	0	37.64	0.02	0.93	40.11
18	PCB-156 2,3,3’4,4’5–Hexachlorobiphenyl	0.01	N/D	0.05	N/D	0.28	N/D	0.32	N/D	0.66
19	PCB-157 2,3,3’4,4’5’–Hexachlorobiphenyl	0.00	0.48	0.03	0.09	0	1.45	0.34	0.81	3.2
20	PCB-167 2,3’4,4’5,5’–Hexachlorobiphenyl	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	
21	PCB-169 3,3’4,4’5,5’–Hexachlorobiphenyl	0.07	0.73	0.29	0.34	0.12	31.54	0.50	0.73	34.32
22	PCB-170 2,2’3,3’4,4’5–Hexachlorobiphenyl	0.41	0.45	0.14	0.32	0.58	0.60	0.33	0.56	3.39
2 3	PCB-180 2,2’3,4,4’5,5’Heptachlorobiphenyl	0.32	0.22	0.34	0.32	1.64	2.76	0.26	0.63	6.49
24	PCB-187 2,2’3,4’5,5’6’–Heptachlorobiphenyl	0.15	0.86	0.18	0.10	0.27	29.79	0.68	0.56	32.59
25	PCB-189 2,3,3’,4,4’5,5’–Heptachlorobiphenyl	0.02	0.14	0.15	0.13	0.33	23.76	0.49	0.77	25.79
26	PCB-195 2,2’3,3’4,4’5,6’–	0.19	0.26	0.68	0.40	0.03	6.56	0.22	0.97	9.31

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	Octachlorobiphenyl									
27	PCB-2062,2'3,3'4,4'5,5'6' Nonachlorobiphenyl	0.07	0.11	1.08	0.42	0.24	4.59	0.54	0.89	7.94
28	PCB-209 Decachlorobiphenyl	0.15	0.14	0.18	0.36	0.39	4.42	0.33	0.05	6.02
	TOTAL	1.38	2.91	3.04	2.39	3.60	104.02	3.35	5.16	125.85

Source: Researcher's Fieldwork

Table 3 Results of Speciation Chemistry for PCBs in Bottom Sediment Samples of the Study.

SN	PCBs COMPD, ppb	OR1	OR2	OR3	SR1	SR2	SR3	TZ1	TZ2	MEAN
1	PCB-8 2,3' Dichlorobiphenyl	3.94	1.99	1.62	N/D	0.91	0.98	0.18	N/D	1.20
2	PCB-18 2,2',5-Trichlorobiphenyl	4.81	3.51	1.18	N/D	1.74	0.02	1.71	0.15	1.64
3	PCB-28 2,4,4'-Trichlorobiphenyl	N/D	N/D	N/D	N/D	N/D	N/D	0.50	N/D	0.06
4	PCB-44 2,2'3,5'- Tetrachlorobiphenyl	4.41	4.26	0.82	N/D	0.35	0.53	0.59	1.23	1.52
5	PCB-52 2,2'5,5'-Tetrachlorobiphenyl	7.65	6.39	2.52	1.81	0.61	1.78	0.06	2.94	2.97
6	PCB-66 2,3',4,4'-Tetrachlorobiphenyl	1.10	7.91	1.78	4.27	16.73	5.82	1.55	1.84	5.13
7	PCB-77 3,3',4,4'-Tetrachlorobiphenyl	1.55	3.15	1.74	8.75	2.87	6.30	0.28	0.74	3.17
8	PCB-81 3,4,4',5-Tetrachlorobiphenyl	6.89	9.38	2.75	2.09	6.01	1.77	2.94	0.58	4.05
9	PCB-101 2,2'4,5,5' –Pentachlorobiphenyl	1.75	11.62	3.01	1.74	2.48	3.23	1.05	0.25	3.14
10	PCB-105 2,3,3'4,4' –Pentachlorobiphenyl	7.39	2.73	3.64	1.06	2.99	5.46	1.22	0.99	3.19
11	PCB-114 2,3,4,4',5–Pentachlorobiphenyl	9.43	3.61	0.57	4.95	0.75	7.22	0.07	1.81	3.55
12	PCB-118 2,3',4,4',5–Pentachlorobiphenyl	3.60	2.18	1.34	1.88	0.72	4.36	0.68	2.77	2.19
13	PCB-123 2,3'4,4'5'–Pentachlorobiphenyl	0.33	5.13	2.29	2.65	0.94	0.25	0.07	0.65	1.54
14	PCB-126 3,3'4,4'5'–Pentachlorobiphenyl	2.61	1.50	0.73	3.05	3.35	2.99	0.03	0.19	1.80
15	PCB-128 2,2'3,3'4,4'–Hexachlorobiphenyl	N/D	0.31	0.23	N/D	0.16	0.62	0.01	0.57	0.24
16	PCB-138 2,2'3,4,4'5'–Hexachlorobiphenyl	N/D	0.26	N/D	N/D	0.15	0.53	0.01	0.54	0.18
17	PCB-153 2,2'4,4'5,5'–Hexachlorobiphenyl	3.19	1.47	1.07	1.16	0.58	2.93	0.05	1.99	1.56
18	PCB-156 2,3,3'4,4'5–Hexachlorobiphenyl	N/D	0.19	N/D	N/D	0.14	0.38	0.02	N/D	0.09
19	PCB-157 2,3,3'4,4'5'–Hexachlorobiphenyl	N/D	0.19	N/D	N/D	0.18	0.38	0.01	N/D	0.05
20	PCB-167 2,3'4,4'5,5'–Hexachlorobiphenyl	8.00	1.43	4.72	1.22	1.66	2.86	0.18	0.91	2.62
21	PCB-169 3,3'4,4'5,5'–Hexachlorobiphenyl	3.87	0.89	2.21	8.42	0.57	1.78	0.07	0.38	2.27
22	PCB-170 2,2'3,3'4,4'5–Hexachlorobiphenyl	1.13	0.96	0.63	0.59	0.31	1.93	0.04	1.47	0.88
23	PCB-180 2,2'3,4,4'5,5'–Heptachlorobiphenyl	N/D	0.97	N/D	1.24	0.22	1.94	0.07	1.39	0.73
24	PCB-187 2,2'3,4'5,5'6'–Heptachlorobiphenyl	N/D	0.86	0.56	N/D	0.41	1.71	0.54	0.82	0.61
25	PCB-189 2,3,3',4,4'5,5'–Heptachlorobiphenyl	N/D	0.68	N/D	1.00	1.75	1.36	0.13	0.59	0.69
26	PCB-195 2,2'3,3'4,4'5,6'–Octachlorobiphenyl	N/D	0.41	N/D	N/D	1.12	0.82	0.25	N/D	0.33
27	PCB-206 2,2'3,3'4,4'5,5'6'–Nonachlorobiphenyl	N/D	2.87	N/D	N/D	1.25	0.74	0.41	N/D	0.66
28	PCB-209 Decachlorobiphenyl	N/D	3.18	N/D	N/D	3.04	0.37	1.07	N/D	0.96
29	Total	71.65	72.53	33.41	45.88	49.34	58.06	13.79	5.56	47.02

Source: Researcher's Fieldwork.

The concentration of polychlorinated biphenyls for the different sampling locations in water and sediment ranged from CB8- 0.01±0.91 to to 0.37±3.18 (Table 1 and Table 2)

The results of speciation chemistry of polychlorinated biphenyls for water and sediment respectively reveal non-similarity/uniformity in their characteristic concentration or bioabundance across all stations of the study. However, similarity exists in terms of PCBs presence in the marine ecological media. Only one dichlorobiphenyl (2, 3' – CB8) was detected in all the water and sediment stations of the study. Two Trichlorobiphenyls (2, 2', 5 – CB18 and 2, 4, 4' - CB28) were detected across all stations of water and sediment. Five Tetrachlorobiphenyls (2,2',3,5' - CB44, 2,2,5'5' - 52, 2,3',4,4' - 66, 3,3',4,4' - CB77 and 3,4,4'5 - CB81) were equally observed in all the water and sediment sources of the study. Six Pentachlorpbiphenyls (2,2',4,5,5' - CB101, 2,3,3',4,4' - CB105; 2,3,4,4'5 - CB114; 2,3',4,4'5 - CB118; 2,3',4,4',5' - CB123 and 3,3',4,4'5 - CB126) were detected in all water and sediment stations of the study. Eight Hexachlorinatedbiphenyls (2,2',3,3',4,4' - CB128; 2,2'3,4,4'5' - CB138; 2,2',4,4'5',5' - CB153; 2,3,3',4,4'5 - CB156; 2,3,3'4,4'5' - CB157; 2,3',4,4'5,5' - CB167; 3,3',4,4',5,5' - CB169 and 2,2',3,3',4,4'5 - CB170) were detected from all water and sediment locations. Three Heptachlorobiphenyl (2,2',3,4,4'5,5' – CB180; 2,2',3,4',5,5',6' - CB187 and 2,3,3',4,4',5,5' – CB189) were detected in all stations. Also, one each of Octochlorobiphenyl (2,2',3,3',4,4',5,6' CB195) and Nonachlorobiphenyl (2,2',3,3',4,4',5,5'6' - CB206) were

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observed across all water and sediment stations. Finally, a single Decachloro biphenyl was observed across both environmental media of the study.

From table 1, the dominant ecological PCBs in the study surface water from their horizontal mean concentrations is in the order: PCB-66 2,3',4,4'-Tetrachlorobiphenyl (5.13) > PCB81 (4.05) > PCB114 (3.55) > PCB105 (3.19) > PCB77 (3.17) > PCB 101 (3.14) > PCB 52 (2.97) > PCB 167 (2.62) > PCB 169 (2.27) > PCB 118 (2.19) > PCB 126 (1.80) > PCB-18 (1.64) > PCB-153 (1.56) > PCB-123 (1.54) > PCB-44 (1.52) > PCB-8 (1.20) > PCB- 209 (0.96) > PCB- 170 (0.88) > PCB- 180 (0.73) > PCB- 189 (0.69) > PCB-206 (0.66) > PCB-187 (0.61) > PCB- 195 (0.33) > PCB- 128 (0.24) > PCB- 138 (0.18) > PCB- 156 (0.09) > PCB-28 (0.06) > PCB- 157 (0.05), indicating that PCB-66 2,3',4,4'-Tetrachlorobiphenyl (5.13) is the highest ecotoxin in the marine waters of Orashi and Sombriero River systems.

Similarly, PCBs locational dominance in the surface waters by their vertical total contribution is in the order: SR3 (104.02) > TZ2 (5.16) > SR2 (3.60) > TZ1 (3.35) > OR3 (3.04) > OR2 (2.91) > SRI (2.39) > OR1 (1.38). This shows that Sombriero River location 3 has the highest concentration of PCBs whereas Orashi River Location 1 is the least. Ecological zone-wise, the Sombriero River (SR) at a total of 110.01 representing % ranks highest in PCB bioabundance followed by the Transition Zone at 8.51(%) and Orashi River the least at 7.33 (%), ie SR > TZ > OR. This indicates both locational and media variations.

Results from this study shows that $\sum PCB_{water}$ (125.85) > $\sum PCB_{sediment}$ (47.02), implying the PCB bioabundance/ bioaccumulation is more in the surface waters compared to the sediments in the study area. It explains that PCBs in the study area are relatively fresh and less dense terrestrial drains and direct atmospheric depositions exhibiting their characteristic water insolubility and yet to fully sediment through vertical downward travel due to less affinity with surface water particulate matters. By implication of this finding, it is clear that consumption of water, pelagic fishes, species and their derived products pose greater health risks than crustaceans and other benthic mammals, species and their products. This study therefore disagrees with the findings of Gideon et al., (2020) that found greater sediment PCB bioaccumulation than the surface waters in their study of the spatial characteristics, sources, and human health risks of polychlorinated biphenyls in sediments from some river systems in the Niger Delta, Nigeria. However, the two studies are in concordance that lower chlorinated PCBs dominated the marine environmental media with potential adverse health risks associated with exposure of species and humans in the Niger Delta. Again, the study agrees with Ute, et al., 2020), that there exists spatial variations in downward vertical strata zonation in accretion, sedimentation in PCBs abundance, sinking, disintegration, transport negligible background rocks geochemical inputs and stratigraphic abundance accounted for higher water bioabundance relative to sediment. This study found no recent secondary contamination of PCBs arising from either of scavenging or exhumation of legacies as against the findings of higher marine sediment legacy reported in (Fu and Wu, 2006 in Obanya et. al, 2019; Xiaoai et al., 2020). Similarly, the study finds negligible organic content but high synthetic content in the sources and environmental characteristics of PCBs, hence its fate and diffusion were not influenced by the hydrophobic and lipophilic nature of high organic matter content in both marine ecological media (sediment & surface water), hence divergent with the findings in (Obanya et. al, 2019).

- *Ecological Impact Health Implication of Exposure to PCBs and Human Health Implications of PCBs in Tropical Deltaic Wetlands*

The ecological impacts of PCBs have been widely studied. Xiaoai et al., (2020) investigated polychlorinated biphenyl in the drinking water source of the Yangtze River as characteristics for risk assessment and found that PCBs in water, sediment and suspended particulate matter (SPM) ranged between 0.04 – 11ng/L, 0.33 – 69 ng/L and 0-72 – 153 ng/L respectively. Thus, $PCB_{spm} > PCB_{sediment} > PCB_{water}$. This is indicative that PCBs biaccumulate more in marine particulate matters and sediments than other matrices. The median values of the Co-PCBs for the three phases were 0.03 ng/L, 0.03 ng/L and 0.23 ng/L for water, sediment and SPM respectively. Comparison of the total PCBs (∑PCBs) of individual matrices shows that ∑PCB in water elevated more than ∑PCBspm and ∑PCBsediment. The study equally reported PCBs of lower molecular weight (ie. di, tri & tetra-CBs) dominated the midstream ecological matrices. Poor water solubility, diminuated polarity and poor content of the Co-PCBs in water were adduced as factors responsible for the lower Co-PCBs in the waters of the Yangtze River. Similarly, TEQ of PCBs in the waters of the Yangtze River ranged between ND-5.55 pg – TEQ/L which is lower than the permissible value of 1.00 pg – TEQ/L, according to Zhou et al., (2014) and poses no ecotoxic risk. Finally, carcinogenic and noncarcinogenic risk value of PCBs in the ecological matrices of the Yangtze River ranges between 3.87×10^{-11} to 9.67×10^{-9} and 6.78×10^{-8} to 1.69×10^{-5} respectively. This is lower than the USEPA permissible limit of 1×10-6; hence poses no ecotoxicologic and human cancer risk at less than 1.

However, this study found the stoichiometric bioabundance of all PCBs above the WHO/USEPA permissible limit of 1×10-6 matrices and locations except in locations not detected; hence poses ecotoxicologic and human cancer risks even at less than 1 in all detected locations of the study environmental matrices. The study therefore disagrees with (Obanya et. al, 2019) in the spatial quantitative bioabundance of PCBs but agrees on lower presence of PCBs in sediments compared to surface water; and the occurrence of variations within and across locations and matrices.

This study found the dominance of lower congeners in both study matrices, indicating higher reactivity in ecological and human toxicity, hence is at variance with Filipkowski (2013) regarding the presence PCBs within permissible limits in all stations and matrices. It also

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disagrees on the presence of low molecular weight (tri & tetrachlorinated PCBs) compared to those of higher weights (penta & hexachlorinated PCBs); but agrees on lower presence of PCBs in sediments compared to other marine media.

IV. CONCLUSION

Environmental interactions are intrinsically interlinked in nature, especially in their relations with and between organisms and matrices. Natural phenomena are causal in nature with causes and effects as it affects human and the environment they are a part of. This study investigated the sources, levels of bioabundance and spatial characteristics of Polychlorinated Biphenyl (PCBs) in the surface waters and sediments of the Lower Orashi and Sombriero Rivers Systems, Rivers State, Nigeria. This was done identify their potential ecological and human health implications with a view to emplacing effective remediation and monitoring protocol in the wider marine ecological environment of Niger Delta. Findings show that all Ind-PCBs evaluated were higher than institutional permissible limits, whereas the sequencing and speciation chemistry analysis found the CO-PCBs to be hyperactive, thus implicated in the surface water, bottom sediments and ancillary species' pollution. The speciation analyses and sequencing characteristics of polychlorinated biphenyls (PCBs) for water and sediment were dissimilar in their abundance across all stations of the study. Though, PCBs quantitative abundance is higher in the surface waters than sediments in all sampling locations and ecologies, concentrations of PCBs in the surface water and bottom sediments of the two River systems differ significantly across different sections of the river courses. Measured PCBs, concentration levels were all above the WHO/USEPA/EFSA/JECFA/FAO respective permissible limits. This then suggests that the water from the sampled river systems is not suitable for aquatic water organisms' water utilization and associated sediment quality. The larger implication of this finding is that the riparian population and others within their trade chain who consume water and ancillary species (benthic & pelagic) and their derived products are exposed to PCB toxicity with likely risks of any or multiple associated health impacts. Furthermore, consumption of water, pelagic fishes, species and their derived products pose greater health risks than crustaceans and other benthic mammals, species and their products. Ecologically, while all the ecological zones pose health threats to the riparian populations, those in the Sombriero River (SR) are exposed to relatively higher degrees of PCB associated health risks followed by those of the Transition Zone (TZ) and Orashi River (OR) respectively; since their ecological PCB means are above the threshold values of 4.0 pg/g offered by Canadian guidelines.

It is arising from these findings, that this study suggests an inclusive multi-stakeholders articulation, review and stringent enforcement of regulations on importation, application, disposal and monitoring of PCB laden equipment and appliances is recommended for the Niger Delta region, with legally empowered independent environmental regulatory agency away from government and devolution from the Concurrent to the Residual Legislative Lists is recommended.

- *Data Availability Statement*

The data sources that support the findings of this study have been provided in the body of the study and available from the corresponding author upon reasonable request.

- *Funding Statement*

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- *Conflict of Interest Statement*

The authors declare that they do not have any conflict of interest.

- *Ethics Statement*

There are no ethical considerations for this research.

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