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The Determination and Impact Analysis of Petroleum Hydrocarbons in Surface waters, Sediments, and some Aquatic Species in Ogbia LGA, Bayelsa, Nigeria

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Abstract: The study determined and analyzed the impacts of petroleum hydrocarbons in surface waters, sediments, and some aquatic species in Ogbia LGA. Composite samples from these matrices were taken at four locations from each of the four different Clans that make up the LGA, for the months of November, February, June, and September in the year 2024-2025. After extractions and clean-up using standard methods, aliphatic fractions (aliphatic components and total aliphatic hydrocarbons – TAH) were analyzed with gas chromatography-flame ionization detector (GC-FID); while the polycyclic aromatic hydrocarbons (PAHs) were determined with gas chromatography-mass spectrometry (GC-MS). The summations of TAHs and PAHs gave the total petroleum hydrocarbons (TPHs) which ranged from 308.87 – 16,582.25 µg/L in waters; 188.93 – 3,308.22 mg/kg in sediments, and 55.37 – 203.61 mg/kg in aquatic species. The range of values were much higher than the 50mg/kg DPR target value for soil and sediment; and the 300 µg/L and 2 mg/kg EU limits for water and foods respectively. Source diagnosis reveals that the source of hydrocarbons in the waters were largely petrogenic, that of sediments were both a petrogenic and biogenic; while aquatic species showed much evidence of biogenic inputs with obvious anthropogenic petroleum impacts. The calculated hazard quotients (HQ) and Hazard indices (HI) were much larger than one (1) for the different waters and sediments in the different Clans indicating the ability of petroleum hydrocarbons in waters and sediments to cause great harms to the environment. The hazard indices for the aquatic species however, ranged from 0.488 – 2.01 implicating medium to slightly high risks to human health. There is the need to mitigate anthropogenic petroleum inputs in order to sustain aquatic lives and preserve public health.

1. Introduction

Water in adequate quantity and quality is essential for the sustenance of life and regular functioning of the ecosystems. Surface waters such as rivers, streams, creeks, lakes, and ponds serve as valued fresh water source for mankind. The contamination of this source therefore, may represent a major threat to human health and ecosystem functions. Surface water has many functions which include: recreational and domestic uses, transportation of goods, generation of electric power,

industrial uses such as production of goods and cooling of power plants. Surface water also finds use in the irrigation of lands and serves as support for all forms of aquatic lives. The pollution of water bodies may hinder its capacity to support and sustain life. The productivity, stability, species abundance, composition and diversity as well as physiological conditions of indigenous organisms may also be largely affected by its pollution (Chokor, 2021a; Alaqarbeh *et al.,* 2022). Surface water is indeed considered as impaired, when it fails to performed at least one its functions, which include: protection and propagation of aquatic lives (such as fishes), recreation, and public water supply, etc (EPA, 2015; Nasri *et al.,* 2024). Several factors have been implicated for the pollution of surface water – both natural and anthropogenic. However, most surface water pollution is almost entirely the results of human activities (i.e., anthropogenic) (Abouri *et al.,* 2024).

In Bayelsa, a major anthropogenic source of pollution of water bodies has been as results of oil related activities which have often resulted to petroleum hydrocarbons pollution. The discovering and exploration of oil in the Niger-Delta, Nigeria to which Bayelsa belong is not without its attendant woos as oil spillage arising from improper handling of sites facilities as well as intentional bunkering activities have often presented great damage to the environment. Petroleum hydrocarbons refer to hydrocarbons originating from crude oil and its refined products.

However, since there are several hundreds of chemical compounds in crude oil and its refined products, it is too costly and cumbersome to measure each compound separately in the environment. Therefore, Total Petroleum Hydrocarbons (TPHs) measure is often used as definitive picture of the whole. TPH is frequently used gross parameter for the quantification of environmental contaminants originating from crude oil and its products such as fuels, oils, lubricants, waxes etc, and is majorly composed of hydrogen and carbon, thus, the name petroleum hydrocarbon (Schwartz, 2012). TPHs are available in the range of C_8 – C_{40} as mixtures composing hundreds to thousands of hydrocarbons including aliphatic (straight carbon chain, branched chains and cycloalkanes), and aromatics (carbon ring) compounds.

Though hydrocarbons are found naturally in the environment, large amount of it in a contaminated environment comes from the activities of man. Petroleum hydrocarbons gets into water bodies through the activities of man such as: oil exploration and exploitation, pipelines leakages and vandalisms, runoff from petroleum contaminated soils, leakages from farm tanks, tankers, trucks, vessels and ships, oil spills, and indiscriminate dumping of petroleum products on water ways (Chokor, 2021b). Sewage, municipal and industrial discharges, automobile wastes and vehicular emission arising from incomplete combustion brought down by the rains, carried by runoff and emptied into water bodies constitute other sources of anthropogenic petroleum hydrocarbons (Chokor, 2021b).

Hydrocarbons being lipophilic, are adsorbed unto suspended particulates in the water column and are eventually settled down in sediments where they may accumulate to high concentrations. Factors that may impact petroleum hydrocarbons concentrations in sediments include: sediment type and size, geographical location, water migration, etc (Zhao *et al*., 2017). However, change in environmental conditions may cause adsorbed hydrocarbons to be release in to water column by physicochemical and biological processes, leading to secondary pollution of the aquatic environment. The contamination of these water bodies by petroleum hydrocarbons may spell danger to aquatic organisms and human health. Aquatic lives may be affected through direct smothering, toxicity, and bioaccumulation through the food chain (Al-Jasimee and Hussein, 2021). Human lives are threatened when they ingest the water (domestic use) and edible aquatic species that have accumulated these hydrocarbons. Dermal contact with contaminated water either trough bathing, recreation activities or

sourcing for foods and resources in it also constitute source of threat to man. The impairment of growth and developmental rates, feeding mechanisms, and increased susceptibility to diseases and other histopathological disorders are some subtle acute effects that have been reported for exposure to petroleum hydrocarbons (Al-Shwafi 2008; Enuneku *et al*., 2015). Chronic low-level exposure to hydrocarbons could results to physiological impairment, shorten survival rate and reduced reproductive success (Enuneku *et al*., 2015; Lee *et al*., 2015). Several diseases conditions have also been linked to petroleum hydrocarbons contaminations; such as: the disruption in the activities of various body organs (such as: the pancreas, kidney, liver, blood circulatory system) and ultimately death; humans health complications (like skin irritation and rashes, genotoxicity, respiratory system disorders, cancers of different parts of the body, deoxyribonucleic acid (DNA) damage, birth defects, childhood leukaemia, infertility and miscarriages in women) (Asghar *et al*., 2016; Oyinbo *et al*., 2018; Briggs and Briggs, 2018; Ite *et al*., 2018; Chokor, 2022). The impact of petroleum hydrocarbons on the aquatic system and human health in general is a function of: persistence and bioavailability of specific hydrocarbon, ability of organisms to accumulate and metabolize various hydrocarbons, fate of the metabolized products, and interference of specific hydrocarbons with normal metabolic processes (Lamardo *et al*., 2013; Lee *et al*., 2015; Zhao *et al*., 2020; Chokor, 2022).

Many authors have reported TPHs values for several surface waters and sediments. Akporido and Onianwa (2015) reported a range of 1,658.47 – 5,435.75µg/L for surface water at the Esi River, Western Niger Delta, Wokoma (2014), and Daniel and Nna (2016) however reported ranges of 15,600 – 23,400µg/L and 13,161.81 – 24,854.62µg/L respectively for TPHs in subsurface seawater from the Kua/Kinabere creek, in Ogoni Land – estuary of Bonny River and surface water in Cross River Estuary, Niger Delta. Also, Adewuyi and Olowu (2012) reported a range of 20,340 – 27,400µg/L for surface water within the vicinity of Nigeria National Petroleum Corporation (NNPC) depot in Apata, Ibadan. Mean values of 73,500µg/L and 23,600µg/L were respectively recorded by Adewuyi *et al* (2011) and Clinton *et al* (2009), for surface water at Ubeji River, Warri, and oil polluted mangrove wetland of Niger Delta, Nigeria. A much lower value range of 4 – 8µg/L were reported by Ikpe *et al* (2016) for the Ethiope River, Delta, Nigeria. Adeniji et al (2017a,b) reported for the surface waters in Algoa Bay and Buffalo River Estuary, both in Eastern Cape, South Africa values range of 45.07 - 307µg/L and 7.65 – 477.07µg/L respectively. Elsewhere, around the world, values of 2.0 – 40.8µg/L have been reported for the Dungun Rver Basin, Malaysia (Suratman, 2013), while range of 60,000 – 260,000µg/L were recorded for the Deepwater Horizon, Gulf of Mexico (Sammarco *et al*., 2013). Sogril (2014) also reported a range 19 - 88µg/L for the the Levantine Basin water of Israeli Coastline.

TPHs level in sediments of the Niger River at Okpu and Iyiowa-Odekpe axes was reported to be of the range of 108.0437 – 1,091.4635 mg/Kg (Chokor, 2022). Values of: 270 – 830 mg/Kg was reported for the Qua-Iboe River, Akwa-Ibom State, Nigeria (Iyang *et al*., 2018), Ranges of 112.30 – 657.30mg/Kg was observed for the New Calabar River, South-Southern Nigeria (Ibigoni *et al*., 2009), 54.72 – 2,002 mg/Kg for River Oluwa, western Nigeria (Fagbote and Olanipekun, 2013), 400 – 6,205 mg/Kg for sediments from the Upper Reaches of the Sombreiro River, Niger Delta, Nigeria (Howard *et al*., 2012), 1,403 – 3,755 mg/Kg for sediments of polluted tidal creek in Bonny River, Nigeria (Wokoma, 2014), and 1,242 – 5,200 mg/Kg for soils in communities of Niger Delta region, Nigeria (Alinnor *et al*., 2014), 25.46 – 69.35 mg/Kg for Onyima Creek, Niger Delta, Nigeria (Edori and Edori, 2021), 3.162 – 8.758 mg/Kg for Woji Creek, Niger Delta Estuary of Rivers State, Nigeria (Ihunwo *et al*., 2021),. Elsewhere, around the world, ranges of values of: 12.59 – 1,100 mg/Kg was

reported for the sediment of the Buffalo River Estuary, Eastern Cape Province, South Africa (Adeniji *et al*., 2017b), 50 – 1,122 mg/Kg for Arabian Gulf sediments in Kuwait (Massoud et al., 1996), 35.6 – 1,466.1 mg/Kg for surface sediments from South China Sea of Kuching Division (Yusoff *et al.,* 2012), 496 – 8,972 mg/Kg for sediment of Ceuta Harbor, North Africa, Spain (Guerra-Garcia and Garcia-Gomez, 2005), 1,116.3 – 2,137.4 mg/Kg for sediments along discharged Basin of Suez oil Refinery Company, South West of the Suez Gulf (Farid *et al* ., 2014), 0.72 – 27.03 mg/Kg for Algoa Bay, Eastern Cape, South Africa (Adeniji et al., 2017a), 0.05 – 20 mg/Kg for Bizerte Lagoon, Tunisia (Mzoughi *et al*., 2005), 3.88 – 24.7 mg/Kg for Candarli Gulf, Turkey (Filiz *et al*., 2012), 3.88 – 24.7 mg/Kg for the Mediterranean Sea, Dameltal harbour (Shereet, 2009), 2.1 – 310 mg/Kg) for the Black Sea, Monaco (Readman *et al*., 2002), and the 0.52 – 4.59 mg/Kg and 0.26 – 1.64 mg/Kg respectively reported for the coaster areas of Papar and Putatan, both in Sabah, Malaysia (Siti Aishah *et al*., 2013). Mean TPHs (mg/kg) of 58334±32 and 58314±35 have been reported for tissues of fishes from the Odidi River and Eqwa River, both in Delta State, Nigeria by Ogeleka *et al*. (2016). The author also reported mean values of 103380±98 and 103180±99 respectively for snail's tissues from the same source. Akinola *et al*. (2020) recorded value of 1995.99 – 3401.55mg/kg for the N. hastatus in Coastal area of Ondo State, Nigeria. While value ranges (mg/kg) of 11.44 – 48.16 and 3.87 – 57.5 were reported for the Iraqi marine waters and Shatt Al-Basrah Canal, Iraq respectively (Nasir, 2005; Galo *et al*., 2022). Mean values of 4.2±2.3 and 6.8±3.6 mg/kg were observed by Jamoussi *et al* (2022) for Epinephelus tauvina and Cephalopholis argus from Jeddah, Saudi Arabia.

While works on petroleum hydrocarbons have been done in other areas, works in Ogbia LGA are still scanty, yet Ogbia LGA, Bayelsa occupies ecological and economic significant place in the Niger Delta. Also, the few works that have been carried out focus only on either surface water or sediments or aquatic species, without examining these entire media constituting the aquatic ecosystem at the same time in one area. In this work, we concurrently sampled and determine the levels of TPHs in surface water, sediments, and aquatic species in Ogbia LGA with view to:

1. Quantifying the levels of TPHs in water, sediments, and aquatic species
2. Predicting the potential health risk of TPHs to the environment and human health
3. Comparing the levels in the different media, and determine the extent of accumulation in sediments and bioaccumulation in species if any and
4. Evaluating the source of hydrocarbons using some diagnostic indices

2. Methodology

2.1 Study area

The study area is Ogbia LGA, Balyesa State, Nigeria. It is located in the heart of the Niger Delta The primary occupation of the Ogbia people include: farming, fishing, and trade, with agriculture occupying a principal role in their local economy. The mangroves, rivers, and creeks predominance in the region are very vital for the support of diverse ecosystem and sustenance of the local economy. Being an oil producing region, it is also characterized by numerous oil and gas operations which on one hand have contributed to the growth of the economy, and on the other hand to environmental and social challenges.

2.2 Sampling method

2.2.1 Collections of water and sediment samples

Composite samples of surface waters and sediments were taken from each of the four (4) clans (CN) that make up Ogbia LGA. Viz: Oloibiri (CN1), Kolo creek (CN2), Anyama (CN3), and Abureni

(CN4) (Table 1). A total of 128 composite samples from the two matrices were investigated. Samples were taken in batches (November, February, June and September). Five composite samples of water and sediments were collected at four different locations in each of the four clans. At each location, five samples were taken at random and composited to form a representative sample for each location. Each 1-liter water sample was collected using clean glass amber bottles, pre-rinsed with surrounding water, and taken just below the surface. The bottles were tightly covered with Teflon-lined caps to prevent any sort of contamination. Sediment samples were taken from the bottom surface (5– 10cm) with a stainless steel Van Veen grab sampler and kept in pre cleaned wide-mouth amber bottles. Both water and sediment samples were carefully labeled then place in ice chests at temperature below 4°C for onward transfer to the laboratory for analysis (Ikpe *et al*., 2016; Chokor, 2024).

Table 1: Sampled communities locations

Ogbia Clans	Communities	Latitude(N)	Logitude(E)
Oloibiri (CN1)	Opume	4°37'41.6''	6°21'37.5''
	Akipelai	4°37'55.9''	6°20'37.8''
	Otuabagi	4°42'45.5''	6°22'00.3''
	Otuabula	4°42'41.0''	6°17'17.8''
Kolo	Imiringi	4°51'06.7''	6°22'28.1''
Creek(CN2)	Kolo	4°53'15.9''	6°22'25.6''
	Otuasega	4°54'22.6''	6°24'06.0''
	Oruma	4°54'58.4''	6°25'23.2''
Anyama(CN3)	Okodi	4°39'50.0''	6°13'31.2''
	Ologi	4°47'27.3''	6°15'00.2''
	Epebu	4°39'09.7''	6°13'00.5''
	Ewoama	4°35'20.6''	6°17'03.4''
Abureni (CN4)	Idema	4°37'48.3''	6°20'55.2''
	Amorikeni	4°45'27.2''	6°20'42.7''
	Ebor	4°36'27.2''	6°21'54.2''
	Odeduma	4°37'49.8''	6°22'42.5''

2.2.2 Collection of aquatic species samples

Samples of aquatic species viz; *Trichiurus lepturus* (Hairtail), *Chrysichthys nigrodigitatus* (Silver catfish), *Ethmasola fimbriata* (Bonga), and *Galatea paradoxa* (Clam) were collected from the different deports/markets in the four clans viz: CN1 (Oloibiri), CN2 (Kolo creek), CN3 (Anyama), and CN4 (Abureni) in Ogbia LGA. Samples were taken for both wet and dry seasons (February and, September). A total of 96 specimens of the 4 species were investigated. Samples of fish species were wrapped in aluminum foil, placed in polyethylene bags and housed in a cooler at 4°C for onward transportation to the laboratory. In the laboratory, the scales and viscera were removed leaving only the edible portions; which were cleaned in tap water to remove any dirt. They were then placed in a well labeled sample bottles and refrigerated at < 4°C awaiting extractions (Chokor and Ogonegbu, 2023).

2.3 Extraction methods

2.3.1 Water

Water samples were Liquid-liquid extracted with dichloromethane (DCM), after spiking with mixed surrogate standard (1mL of 10 μg/mL 1-chlorooctadecane and ortho-terphenyl) (Iyang et *al*., 2018; Udosen *et al*., 2020; Chokor, 2022). The extract was then concentrated under reduced pressure using a rotary evaporator; before column cleans-up.

2.3.2 Sediment

Sediment samples were air dried at room temperature for a period of five (5) days; sieved and homogenized. The aliquot of the air-dried samples (10g) was mixed with sufficient quantity of anhydrous sodium sulphate (Na_2SO_4) (about 5g) to remove moisture, spiked with surrogate standards (10 µg/mL of 1-chlorooctadecane and ortho-terphenyl), wrapped in a filter paper, placed in a thimble and then loaded into the main chamber of the soxhlet extractor. Extraction was done with dichloromethane for 17 hr. Extracts were dried by passing through column of anhydrous sodium sulphate; and reduced to about 2mL with a rotator evaporator prior to column clean-up (Adeniji *et al*, 2017a; Iyang *et al*., 2018; Chokor, 2024).

2.3.3 Aquatic species

The dried fish samples were ground into a fine powder using a mortar and pestle; spiked with 10 *µ*g/mL 1-chlorooctadecane and ortho-terphenyl used as surrogate standards, and solvent extracted with hexane in a Soxhlet extractor for 17 h. The solvent was then evaporated under reduced pressure using a rotary evaporator to obtain concentrated extracts, ready for column cleanup (Owoh-Etete *et al*., 2023; Chokor and Ogonegbu, 2023).

2.4 Sample clean-up and separation

The concentrated extracts of surface waters, sediments, and edible tissues of aquatic species were transferred into a chromatographic column (10 mm i.d. X 30 cm) parked with 10g activated silica gel slurry about 2cm anhydrous Na_2SO_4 layer on top. The samples were first eluted with 30 mL of n-hexane to obtain the hydrocarbon fraction followed by 30 mL of DCM to obtain the aromatic fraction. The eluates were concentrated to approximately 2 mL with rotator evaporator at 30 °C; 1.5mL of each were transferred into chromatographic vials and stored at 4 °C awaiting gas chromatographic analyses. Blank samples were processed the same way for the purpose of quality assurance (Maioli *et al*., 2011; Adeniji *et al*., 2017a; Iyang *et al*., 2018).

2.5 Analyses of samples

The determination of total aliphatic hydrocarbons (TAHs) were done with gas chromatography joined to a flame ionization detector (GC-FID); Agilent 6890N system equipped with DB-5 capillary column with dimension of 30 m X 0.32 mm X 0.25 µm. The column was conditioned to: initial temperature of 50 °C for 5 minutes then rose to 150 °C at rate of 10 °C /min. for 15 minutes; after which it was increased at 16 °C/min to 280 °C and held for 5 minutes. The temperatures of the injector and detector were set at 200 and 300 °C respectively. Sample volume injected was 1 µL in a split less mode; the carrier gas was helium at a flow rate of 1mL/min. The sum of the sixteen priority polycyclic aromatic hydrocarbons (Σ16PAHs) were determined in the aromatic fractions by the same version of gas chromatography (Agilent 6890N) above but couple to a mass spectrometer as detector. The gas chromatographic column had an initial temperature of 70 °C, which was held for 20 min. This was ramped to 150°C at rate of 25°C min^{-1}, and further increased to 200 °C at 3 °C min^{-1}, and finally raised to 300 °C at 2 °C min^{-1}. Sample injection into the GC was via a pulsed spit-less mode with an injection volume of 1µL. The sum of all aliphatic and aromatic hydrocarbons measured by the GC provides a measure of total petroleum hydrocarbons (TPH) concentration.

2.6 Identification and quantification

Aliphatic hydrocarbons and polycyclic aromatic hydrocarbons (PAHs) were identified by comparison of their retentions time with those of respective standards. The response factors associated with the respective internal standards based on five-point calibration curve for the fractionated n-alkanes (nC_8- nC_{40}), and individual PAH were used for quantifications. The nC_8- nC_{40} standard mixtures (internal standard), and standard solution of 1-chlorooctadecane (surrogate standard) were used for the quantifications of the aliphatic hydrocarbons. Deuterated PAH internal standard solutions (naphthalene-d8, acenaphthene-10, phenanthrene-d10, chrysene-d12, and perylene-d12) and surrogate standard solution (σ-terphenyl-d14) were however, used in the quantifications of PAHs and its procedural recovery.

2.7 Risks Assessments

2.7.1 Assessment of Ecological Risks

Ecological risks for water and sediment were determined using fractional approach; the equations 1 and 2 were applied:

$$HQ = \frac{Concentration\ of\ group\ of\ hydrocarbons\ in\ the\ medium}{Toxicity\ reference\ value} \quad (1)$$

$$HI = \sum HQ \quad (2)$$

Where, hazard index (HI) is the sum of the hazard quotient (HQ) of each group hydrocarbons. HI (HQ) > 1 connotes high risk whereas 0.1 ≥ HI (HQ) ≤ 1, and HI (HQ) < 0.1 represent medium and low risks respectively (Bandowe *et al*., 2014; Chokor, 2023). Toxicity reference values (TVR) were obtained from Hawail Department of Health (HiDOH, 2018). For the surface water TVRs for C_5-C_8, C_9-C_{18}, and C_{19}-C_{36} were 24, 43, and 6800µg/L respectively, while TVRs values of 1.591 (C_5-C_8), 5.543 (C_9-C_{18}), and 9.883 (C_{19}-C_{36}) mg/kg were applied for the sediments.

2.7.2 Assessment of health risks

Health risks due to consumption of aquatic species caught from these water bodies were assessed using fractional approach; were TPHs were grouped into three defined carbon ranges viz: low (C_5-C_8), medium (C_9- C_{18}), and high (C_{19}-C_{36}) with their characteristic reference doses of 0.04, 0.01, and 3.0mg$Kg^{-1}Day^{-1}$ (HIDOH, 2018). The indices of: Daily Dietary intake (DDI), Hazard quotient (HQ), and Hazard index (HI) were used to evaluate exposure to TPHs (Tongo *et al*., 2017; Chokor and Ogonegbu, 2023).

$$DDI = [C \times CR]/Bw \quad (3)$$

$$HQ = DDI/ RfD \quad (4)$$

$$HI = \sum_{i=1}^{n} HQ \quad (5)$$

where, DDI refers to the daily dietary intake (mgKg^{-1}bwDay^{-1}), C is the concentrations (mg/Kg) of individual or fractions TPHs; CR is the consumption rate (g/Day), Bw is the average body weight (60Kg) of an adult, and RfD represent the reference dose for the fractions of TPH.

The DDIs were calculated using consumption rate of 7.6 Kg per capita per annum, equivalent to 20.8 g/day. HQ (Hazard Quotient) and HI (Hazard Index) values above one represents possible adverse

health effects whereas those between 0.1 to 1.0 and less than 0.1 represent medium and negligible health effects (Bandowe *et al*., 2014; Chokor, 2024).

3. Results and Discussion

3.1 The levels of TPHs in the surface water

Fig. 1 and **Table 2** show the mean concentrations of TPHs in the sampled waters in the four clans (CN1, CN2, CN3, & CN4) for the months of Nov, Feb, Jun, and Sep. The calculated mean for each month sampled for all clans, annual mean for each clan, as well as the overall annual mean for all the months/clans are presented. The large variations between sites, months, and clans evidenced anthropogenic contaminations. The average values of TPHs for the months of November, February, June, and September in the four Clans were: 5773.78±5909.13; 5182.12±6208.32; 6981.56±6333.61; and 7258.98±6673.29 (µg/L) respectively. While, the annual mean concentrations (µg/L) for each Clan were: CN1 (5916.91±2168.05 µg/L), CN2 (15042.05±1128.66 µg/L), CN3 (3697.54±751.50 µg/L), and CN4 (539.95±298.96 µg/L). The overall annual mean for all months sampled at all Clans was 6699.11±1068.61 µg/L. The highest monthly mean level of TPHs (16582.25 µg/L) was obtained in September at Kolo Creek (CN2), while the lowest (308.87 µg/L) was observed for the month of February at Aburinni (CN4). The general orders for TPHs pollutions were: Sep. > Jun. > Nov. > Feb., for the months, and CN2 > CN1 > CN3 > CN4, for the different Clans.

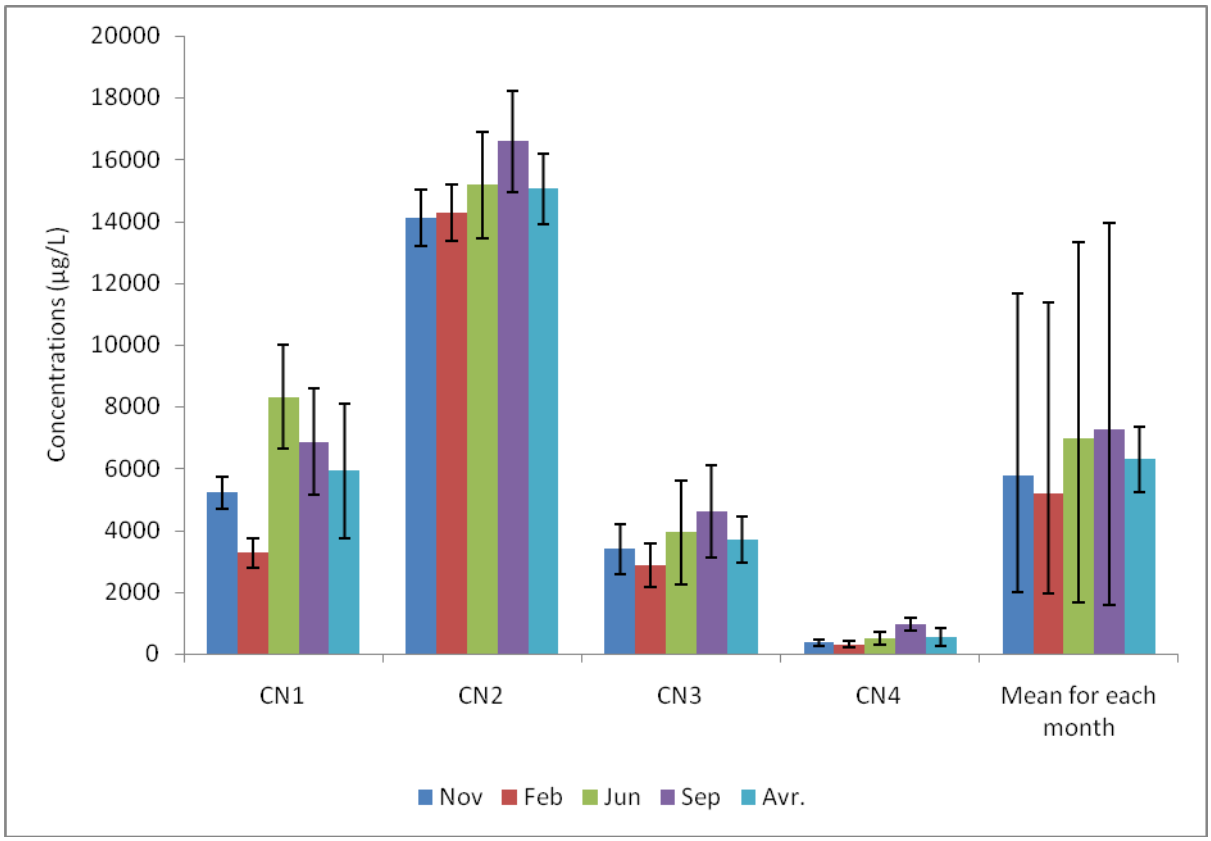

Fig.1: Concentrations of TPHs in surface waters (µg/L) at different clans in the different months

* CN1 = Oloibiri, CN2 = Kolo Creek, CN3 = Anyama, CN4 = Abureni., Avr = Averages

The range of values for the wet season in all Clans was 505.63 – 16582.25 µg/L with a mean of 7120.27µg/L, while that for the dry season was: 308.87 – 14284.31 µg/L with an average value of 5477.94µg/L. The mean for the wet season was significantly higher than that of the dry season; probably as a result of washing of TPHs into the water bodies from land and road surface during the

rains, and re-suspension of TPHs from sediments due to increased water current arising from increase discharged during the raining season (Chokor, 2021b). Also, increased volatilization and degradation (due to photo-oxidation and biodegradations of microorganisms) of TPHs accompanying the increased temperature during the dry season can encourage significant difference in TPHs values between the seasons (Chokor, 2021b; Galo *et al*., 2022). The range of values 308.87 – 16582.25 µg/L with a mean of 6299.11±1068.61 µg/L obtained in this report, are higher than the European Union (EC, 2005) Standard limit of 300µg/L. They were also higher than those reported by Ikpe *et al* (2016), Adeniji *et al* (2017ab), and Suratman (2013) (Table 3), but lower than those recorded by Wokoma (2014), Daniel and Nna (2016), and Ahmed *et al* (2015) (**Table 3**).

Table 2: Average concentrations of TPHs in surface waters (µg/L) and sediments (mg/kg) at different clans in the different months

Clan	Surface water (µg/kg)					
	Nov	Feb	Jun	Sep	range	Mean
CN1	5209.56	3278.95	8312.85	6866.46	3278.95 – 9312.85	5916.91 ±2168.05
CN2	14117.38	14284.31	15184.42	16582.25	14117.37 – 16582.25	15042.05 ±1128.66
CN3	3394.13	2856.36	3923.72	4615.94	2856.36 – 4615.94	3697.54 ±751.50
CN4	374.06	308.87	505.63	971.27	308.87 – 971.27	539.95 ±298.96
mean	5773.78 ±5909.13	5182.12 ±6208.32	6981.56 ±6333.61	7258.98 ±6673.29	308.87 – 16582.82	6299.11 ±1068.61
	Sediments (mg/kg)					
CN1	722.46	701.2	954.32	821.34	701.20 – 954.32	799.83 ±115.53
CN2	1697.45	1473.39	2777.9	3308.22	1473.39 - 3308.22	2314.24 ±873.77
CN3	438.57	306.96	611.98	761.45	306.96 – 761.45	529.74 ±198.65
CN4	211.7	188.93	270.56	331.81	188.93 – 331.81	250.75 ±64.05
mean	767.54 ±654.20	667.62 ±580.12	1153.69 ±1118.21	1305.705 ±1352.70	188.93 – 3308.22	973.64 ±304.85

* CN1 = Oloibiri, CN2 = Kolo Creek, CN3 = Anyama, CN4 = Abureni.

3.2 The concentrations of TPHs in sediments

The concentrations of TPHs in sediments **(Table 2)** tended to followed similar pattern with those of surface waters with the highest concentration (3308.22mg/kg) also recorded in Kolo Creek (CN2) and in the month of September. While the lowest value (188.93mg/kg) was observed in February at Aburinni (CN4). The mean values (mg/kg) for the clans for the months sampled in decreasing order were: CN2: (2314.24), CN1: (799.83), CN3 (529.74), and CN4: (250.75). The overall mean for all clans and all months was 973.64± 304.85mg/kg. Hydrocarbons in polluted waters tend sorbs to organic matters in water column and with time they settled down as sediment. To some extent, the sediments can be regarded as TPHs remover of the water column. This account for the several order of accumulations of TPHs in sediments compared to the water column. However, TPHs in sediments will re-enter the water column with re-suspension of sediment during storms or any forms of

perturbations, resulting in secondary pollutions of the surface water. This perhaps explains the large correlations of TPHs in both water and sediments in this study area. Four levels of hydrocarbon pollution have been proposed by Massoud *et al* (1996) for the assessment of marine sediments viz: unpolluted (10 -15mg/kg), slightly polluted (15 – 50mg/kg), moderately polluted (50 – 200mg/kg), and heavily polluted (>200mg/kg). However, the Nigerian Department of Petroleum Resources (DPR, 2002) prescribed target and intervention values of 50 and 5000 mg/kg respectively for sediments/soil TPHs. The values obtained in this study were all greater than the target values; and tend to suggest heavily polluted status resulting from anthropogenic addition. However all the values were still less than the DPR intervention value for sediment guidelines.

Table 3: Comparison of TPHs levels in surface waters in this study with those of others

Region	Range and/ or mean* of TPHs (µg/L)	References
Ethiope River, Delta, Nigeria	4 - 8	Ikpe *et al*, 2016
Algoa Bay & Buffalo River Estuary, South Africa	45.02 – 307 7.65 – 477.07	Adeniji *et al*., 2017ab
Dungun River Basin, Malaysia	2.0 – 40,8	Suratman, 2013
Kua/Kinebere Creek, Ogoni Land Estuary of Bonny River, Nigeria	15,600 – 23,400	Wokoma,2014
Cross River Estuary, Niger-Delta, Nigeria	13,161.81 – 24,854.62	Daniel and Nna, 2016
Alexandria Coasts, Egypt	2,421 – 183,817 (25,613.88)	Ahmed *et al*., 2015b
Creeks and Streams, Ogbia LGA, Nigeria	308.87 – 16,582.25 (6299.11± 1068)	This study

* mean in Brackets

The range of values (188.93 – 3,308.22mg/kg; mean: 973.64mg/kg), obtained in this study, were comparable to the 270 – 830 mg/Kg; mean: 606.83mg/kg reported for the Qua-Iboe River, Akwa-Ibom State, Nigeria (Iyang *et al*., 2018), and the 400 – 6,205 mg/Kg recorded for the sediment from the Upper Reaches of Sombreiro River, Niger Delta, Nigeria (Howard *et al*., 2012). They are however, much higher than those reported in literatures by Ihunwo *et al* (2021), Edori and Edori (2021), Adeniji *et al* (2017a), Mzoughi *et al* (2005), Shereet (2009) and Kucuksezgin *et al* (2012) for the Woji Creek, Niger Delta, Nigeria; Onyima Creek, Niger Delta, Nigeria; Algoa Bay, Eastern Cape, South Africa; Bizerte lagoon, Tunisia; Mediterranean Sea, Damelta harbor; and Candarli Gulf, Turkey; respectively. The mean values reported for sediments of some other Nigerian Rivers such as: the Ubeji River (1,602.4±8.9 mg/Kg) (Adewuyi *et al*., 2011), the Benin River, Adjacent a lubricating oil production factory (41,900 mg/Kg) (Akporido and Ipeaiyeda, 2014) and the Odidi and Eqwa Rivers (215,730±81 & 215,700±77 mg/Kg) (Ogeleka *et al*., 2016) were much higher than that reported in this study. The 496 – 8,972 mg/Kg recorded for the sediment of Ceuta harbor, North of Africa, Spain (Guerra-Garcia and Garcia-Gomez, 2005) was also slightly higher than that reported in this study.

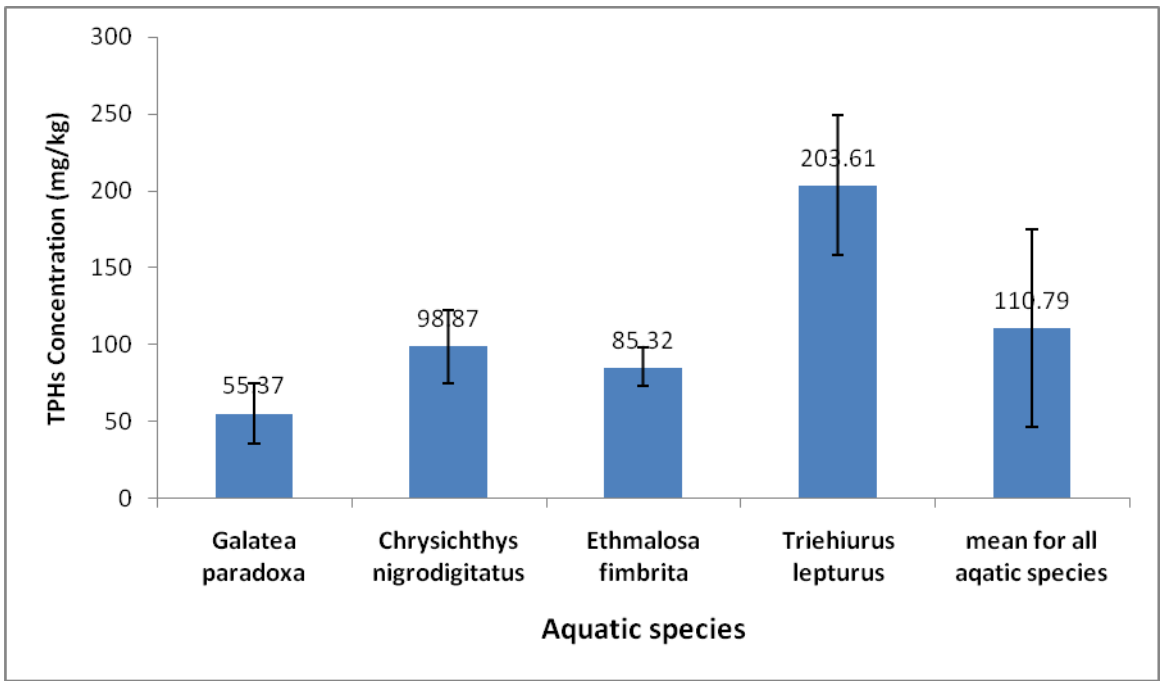

Fig. 2: Annual mean concentrations of TPHs in the aquatic species

3.3 TPHs levels in tissues of aquatic species

The annual mean TPHs levels (mg/kg) in tissues of aquatic species (Fig 2) range from 55.37 in Galatea paradoxa (Clam) to 203.61 in Trichiurus lepturus (Hairtail). The average concentration for all the aquatic species sampled was 110.79 ± 64.49mg/kg. The order of concentrations which was: Trichiurus lepturus > Chrysichthys nigrodigitatus (Silver catfish) > Ethmasola fimbriata (Bonga) > Galatea paradoxa tended to show some correlations with habitat niche and positions in trophic levels. Trichiurus lepturus which accumulated the greatest amount of TPH, is high up the food chain, is demersal and strictly carnivorous. Chrysichthys nigrodigitatus and Ethmasola fimbriata are both omnivores. Chrysichthys nigrodigitatus is however, benthopelagic whereas Ethmasola fimbriata is purely pelagic. Since the concentrations of TPHs are much higher in sediments than in the water column, it is expected that demersal benthivorous organisms will concentrate more TPHs in their tissues compared to the pelagic ones. This perhaps explains why Chrysichthys nigrodigitatus have more concentration of TPH (98.87mg/kg) in comparison with Ethmasola fimbriata (85.32mg/kg). The aquatic organism with the least TPHs concentration was clams (Galatea paradoxa) which are primarily detritus feeders and occupies the low end of the food chain. Various factors such as the exposure time, the lipid content of tissues, species of organism, age, sex, and environmental conditions (including marine habitat, dietary habits, and different depths which the specie lives in the marine environment) are known to affects the accumulations of petroleum hydrocarbons in aquatic species (Ashraf and Mian, 2010; Galo *et al*., 2022). There was large variability within specie and among species sampled. Values of TPHs in tissues were significantly higher ($p < 0.05$) in the rainy season as compared to the dry season. This variation tended to reflect the seasonal fluctuations in TPHs levels of water and sediment. The values for all investigated species tissues were higher than the European Union (EC, 2005) recommended value of 2mg/Kg for foods. The values were much higher than those reported aquatic species by Al-Ali *et al* (2016), Al-Imarah et al (2020), and Jamoussi *et al* (2022); but were comparable to that observed by Aderinola *et al* (2018) (Table 4).

They were however much lower than those reported by Ogeleka *et al* (2016), Mousa et al (2011), Ahmed *et al* (2014), and Akinola *et al* (2020) (Table 4).

Table 4: Comparison of TPHs levels in aquatic species tissues in this study with those of others

Region	Range and/ or mean* of TPHs (mg/Kg)	References
Odidi and Eqwa Rivers, Delta State, Nigeria(fish tissues)	(58334±32), (58314±35)	Ogeleka *et al*., 2016
Egyptian Mediterranean Sea water (tissues of 8 fish species)	2100 – 4249 Winter 790 – 9186 Summer	Mousa *et al*.,2011
Suez Gulf Coast (aquatic species)	987.43 – 2754.2	Ahmed *et al*., 2014
Coastal area of Ondo State, Nigeria (N. hastatus)	1995.99 – 3401.55	Akinola *et al*., 2020
NW Arabian Gulf	2.45 – 7.65	Al-Ali *et al*., 2016
Shatt Al-Arab River & Southern Iraqi Marshes (tissues of 5 fish species)	6.98 – 60.60	Al- Imarah *et al*., 2020
Jeddah (Soudi Arabia) (Epinephelus tauvina, Cephalopholis argus, Plectroponus pessuliferus	(4.2 ±2.3), (6.8±3.6), (7.4±3.2)	Jamoussi *et al*., 2022
Badagry Creek, Lagos, Nigeria (Chrysichthys nigrodigitatus, Macrobrachium macrobrachion	(27.2±15.4), (219.56±171.8)	Aderinola *et al*., 2018
Ologe Lagoon, Lagos, Nigeria (Chrysichthys nigrodigitatus, Macrobrachium macrobrachion	(104.5±23.3) (211.56±127.9)	Aderinola *et al*., 2018
Ogbia LGA, Nigeria (aquatic species: edible portion)	55.37 – 203.61 (110.79 ± 64.4)	This study

* mean in Brackets

3.4 Ecological risks assessment of TPHs in water and sediment

Table 5 gives the mean hazard quotients (HQ) and hazard indices (HI) of waters from the various Clans in the different months. The HQs values for the C_5-C_8; and C_9-C_{18} fractions were all greater than one signifying their capabilities in causing harm to the aquatic environment. Note that the C_{18}-C_{36} fraction hazard quotients were not included because the calculated values were infinitesimally small and were thus insignificant. The hazard index range was 6.16 – 13.47 (mean: 9.16) in Aburinni Clan (CN4); 51.11 – 83.59 with a mean of 61.81 in Anyama Clan (CN3); 60.82 – 112.63 (mean: 84.08) in Oloibiri Clan (CN1); and 42.82 – 254.88 (mean: 110.61) in Kolo Clan (CN2). These high indices connote the abilities the TPHs to cause great harms to the environment. The order of ability being: CN2 > CN1 > CN3 > CN4.

Table 6 represents the mean hazard quotients and indices calculated from the hydrocarbons profiles of sediments at the different clans. The HQs and HIs values for the different clans were all larger than one (1) indicating that there are ecological damages resulting from the presence of the individual hydrocarbons groups as well as the overall total hydrocarbons (TPHs). Based on the TPHs concentrations alone, the order of contaminations was: CN2 > CN1 > CN3 > CN4; however based on the hazard indices, the order was: CN2 > CN1 > CN4 >CN3. This implies that the proportion of each

fraction in the overall TPHs actually determined the extent of pollution emphasizing the need for TPHs profiling.

Table 5: Calculated mean values for hazard quotients and indices for the surface waters in the different months for the different Clans

Clan	month	HQ C8	HQ C9-C18	HI (∑HQ)	Clan	month	HQ C8	HQ C9-C18	HI (∑HQ)
CN1	Nov	1.27	61.26	62.53	CN2	Nov	22.29	61.30	83.59
	Feb	33.84	66.50	100.34		Feb	16.14	41.76	57.90
	Jun	56.43	56.20	112.63		Jun	27.86	26.75	54.61
	Sep	24.42	32.40	60.82		Sep	13.98	37.13	51.11
	Ave	29.99	54.09	84.08		Ave	20.06	41.74	61.81
CN2	Nov	2.39	40.44	42.82	CN4	Nov	3.43	4.06	7.49
	Feb	26.83	59.23	86.06		Feb	4.81	4.71	9.53
	Jun	12.64	46.02	58.66		Jun	2.57	3.58	6.16
	Sep	144.12	110.77	254.88		Sep	4.85	8.61	13.47
	Ave	46.95	64.11	110.61		Ave	3.92	5.24	9.16

* CN1 = Oloibiri, CN2 = Kolo Creek, CN3 = Anyama, CN4 = Abureni.

Table 6: Mean HQs and HIs in sediment of the different clans

Clan	HQ (C8)	HQ(C9-C18)	HQ(C19-C36)	HI (∑HQ)
CN1	1.58	99.86	21.31	122.74
CN2	80.88	205.04	102.09	388.02
CN3	4.07	41.20	26.19	71.46
CN4	54.20	13.98	7.18	75.35

* CN1 = Oloibiri, CN2 = Kolo Creek, CN3 = Anyama, CN4 = Abureni.

Table 7: Calculated mean DDI, HQ, and HI for fish samples from the different Clans

Indices	CN1	CN2	CN3	CN4	overall mean
DDI_{C5-C8}	0.0000336	0.0000667	0.0000192	0.0000215	0.0000353
HQ_{C5-C8}	0.000841	0.00167	0.000480	0.000538	0.000882
DDI_{C9-C18}	0.0201	0.0147	0.00578	0.00478	0.0113
HQ_{C9-C18}	2.01	1.47	0.578	0.478	1.13
$DDI_{C19-C36}$	0.0112	0.0514	0.0120	0.0240	0.0247
$HQ_{C19-C36}$	0.00373	0.0171	0.00401	0.00802	0.00823
ΣDDI	0.0313	0.0662	0.0178	0.0288	0.0361
ΣHQ	2.01	1.490	0.583	0.486	1.140

3.5 Health risks assessment of TPHs due to consumption of aquatic species

The values for calculated daily dietary intake (DDI), hazard quotient (HQ), and hazard Index (HI) (ΣHQ) for the aquatic species samples from the different Clans are as shown in Table 7. The health risks were assessed using fractional approach which grouped the TPHs into three different fractions viz: C_5- C_8, C_9-C_{18}, and C_{19}-C_{36}. The determination method used in this study did not allow for complete determination of the first group (C_5-C_8). However, the C_8 values determined were used. The estimated DDI for the petroleum hydrocarbons fractions ranges from 0.0000215 – 0.0662 $mgKg^{-1}bwDay^{-1}$ in the aquatic species samples from the different Clans. The sums of the DDIs for all

fractions were: 0.0178, 0.0288, 0.0313, and 0.0662mgKg^{-1}bwday^{-1} for Anyama (CN3) Abureni (CN4), Oloibiri (CN1) and Kolo Creek (CN2) respectively. The hazard quotients for the fractions were of the range: 0.000480 – 2.01, while the sum of the hazard quotients (hazard indices) for the various Clans were in order decreasing values were: Oloibiri (CN1) (2.01), Kolo Creek (CN2) (1.490), Anyama (CN3) (0.583), and Abureni (CN4) (0.486). The overall mean for the Hazard indices for all clans was 1.140. Hazard quotients and indices values greater than one (>1) suggest that these fractions individually and collectively are capable of causing harm to the human health. However, values between 0.1 to 1.0 and less than 0.1 represent medium and negligible health effects (Bandowe *et al*., 2014; Chokor, 2024). The range of values obtained for HI in this study, implicated medium to slightly high health risks to human.

3.6 Mean distributions of aliphatic hydrocarbons in the different matrices and the bioaccumulation factors for fish/water and fish/sediment

Table 8, shows the mean hydrocarbons profiles for the surface waters, sediments, and fish species along with the bioaccumulation factors for fish/water and fish/sediment in the sampled area. The most dominant hydrocarbons in water column were those of C_8 – C_{23}. There was also large presence of C_{26} and C_{28}. The other hydrocarbons appear in relatively smaller amount in the water column. The wide ranges of abundance of the low molecular weight hydrocarbons (less than C_{23}) tend to suggest recent anthropogenic contaminations. Various activities such occasional spills, illegal oil bunkering and refining, transportations, and run-off from domestic waste and sewage discharge could have been responsible for these contaminations.

In the sediments, the mean distribution profile reveals also the prevalence of low molecular weight hydrocarbons (C_8 – C_{20}) in comparison to higher molecular weight ones; thus, evidencing anthropogenic source. The large presence of C_{16}, C_{18}, and C_{20} also gave support to petroleum anthropogenic source. However, the very large value of C_{15} in the sediment gives strong credence to the contribution of biogenic source.

The most abundant hydrocarbons in the fish species were in the range of C_{13} – C_{25}. The C_{25} particularly, showed very large dominance in comparison with others. The dominance of this odd numbered alkane is an indication of biogenic inputs. This was supported by the Pr/Ph ratio that was larger than unity (1). However, the large presence of some even numbered aliphatic hydrocarbons particularly C_{16} tend to portray anthropogenic sources. In all sample types (water, sediment, and fish species) the total contributions of polycyclic aromatic hydrocarbons (PAHs) to the total petroleum hydrocarbons (TPHs) were relatively small with mean values of: 3.1% in surface waters, 1.64% in sediments, and 2.30% in fish species.

The fish/water accumulation factors (**Table 8**) for individual hydrocarbons range from 0.17 in C_8 to 952.89 in C_{25}. The C_{25} showed the highest bioaccumulation in fish tissues, followed by C_{36} (107.29), C_{15} (105.52), C_{38} (59.76), C_{17} (29.39), and C_{16} (22.06). Overall, the total petroleum hydrocarbons accumulation factor has a mean value of 17.59. In contrast, only few hydrocarbons showed fish/ sediment accumulation factors that were higher than one (1). These included: C_{25} (120.70), C_{27} (1.45), C_{23} (1.44), and C_{29} (1.03). The overall total petroleum hydrocarbons (TPH) accumulation factor for the fish/ sediment was 0.11 (**Table 8**), which was less than one (1). This shows that it is much easier for the contaminant hydrocarbons to be transfer (either by adsorption or ingestion) from water to fish than it does with sediment, most probably due to differential partitioning behaviour related to their physicochemical properties (Karim *et al*., 2016; Udosen *et al*., 2020; Opia and Chizoruo, 2024).

Table 8: Mean hydrocarbons profiles in water, sediment, and fish species and the bioaccumulations factors for fish/water and fish/sediment in the sampled area (n = 64 for water, n = 64 for sediment, n = 96 for fish species

Components	Concentrations			Bioaccumulation factors	
	Water (mg/L)	Sediments(mg/kg)	Fish species (mg/kg)	fish/water	fish/sediment
C8	0.603±0.428^{a}	55.97±61.94^{b}	0.102±0.063^{c}	0.17	0.00
C9	0.270±0.195^{a}	71.59±97.82^{b}	0.067±0.050^{c}	0.24	0.00
C10	0.107±0.115^{a}	26.44±24.69^{b}	0.082±0.031^{a}	0.77	0.00
C11	0.120±0.089^{a}	2.04±1.54^{b}	0.022±0.016^{c}	0.18	0.01
C12	0.252±0.217^{a}	39.94±31.80^{b}	0.116±0.108^{c}	0.46	0.00
C13	0.119±0.135^{a}	6.95±3.08^{b}	0.360±0.587^{c}	3.03	0.05
C14	0.095±0.112^{a}	46.95±40.43^{b}	0.802±1.213^{c}	8.44	0.02
C15	0.067±0.062^{a}	152.57±213.97^{b}	7.070±6.858^{c}	105.52	0.05
C16	0.359±0.211^{a}	59.22±57.98^{b}	7.918±5.890^{c}	22.06	0.13
C17	0.245±0.168^{a}	14.87±27.82^{b}	7.200±5.727^{c}	29.39	0.48
Pristane	0.176±0.152^{a}	20.54±21.79^{b}	3.476±2.595^{c}	19.75	0.17
C18	0.141±0.118^{a}	57.86±57.35^{b}	4.309±3.206^{c}	30.56	0.07
Phytane	0.225±0.191^{a}	19.23±23.64^{b}	1.294±1.368^{c}	5.75	0.07
C19	0.119±0.115^{a}	10.65±5.29^{b}	2.327±3.293^{a}	19.55	0.22
C20	0.111±0.120^{a}	47.47±43.73^{b}	0.201±0.245^{a}	1.81	0.00
C21	0.112±0.116^{a}	1.89±2.78^{b}	1.026±1.049^{b}	9.16	0.54
C22	0.828±1.599^{a}	37.17±29.83^{b}	2.249±1.626^{c}	2.72	0.06
C23	0.431±0.815^{a}	2.12±2.20^{b}	3.045±3.168^{b}	7.06	1.44
C24	0.066±0.078^{a}	95.59±134.88^{b}	0.516±0.501^{c}	7.82	0.01
C25	0.057±0.093^{a}	0.45±0.19^{b}	54.315±40.78^{c}	952.89	120.70
C26	0.483±0.917^{a}	70.34±93.84^{b}	1.436±0.994^{c}	2.97	0.02
C27	0.077±0.096^{a}	0.40±0.15^{b}	0.578±0.624^{b}	7.51	1.45
C28	0.353±0.606^{a}	50.78±60.11^{b}	0.597±0.963^{a}	1.69	0.01
C29	0.091±0.083^{a}	0.51±0.17^{b}	0.523±0.554^{b}	5.75	1.03
C30	0.102±0.073^{a}	24.72±37.67^{b}	0.606±0.522^{c}	5.94	0.02
C31	0.081±0.086^{a}	0.46±0.09^{b}	0.203±0.185^{c}	2.51	0.44
C32	0.086±0.064^{a}	0.45±0.07^{b}	0.293±0.201^{c}	3.41	0.65
C33	0.072±0.058^{a}	0.48±0.09^{b}	0.140±0.242^{c}	1.94	0.29
C34	0.078±0.071^{a}	13.96±16.87^{b}	0.361±0.653^{c}	4.63	0.03
C35	0.074±0.077^{a}	1.95±2.79^{b}	0.948±1.234^{b}	12.81	0.49
C36	0.017±0.016^{a}	8.74±8.96^{b}	1.824±1.640c	107.29	0.21
C37	0.023±0.015^{a}	1.95±2.35^{b}	0.740±1.449^{c}	32.17	0.38
C38	0.054±0.032^{a}	12.64±8.39^{b}	3.227±2.625^{c}	59.76	0.26
C39	0.007±0.006^{a}	0.41±0.12^{b}	0.026±0.035^{c}	3.71	0.06
C40	0.002±0.002^{a}	0.33±0.18b	0.250±0.480^{b}	125.00	0.76
TAHs	6.103±0.209^{a}	957.63±31.85^{b}	108.252±63.27^{c}	17.74	0.11
$\sum PAHs$	0.196±0.020^{a}	16.01±2.31^{b}	2.543±1.218^{c}	12.97	0.16
TPHs	6.299±1.069^{a}	973.64±304.85^{b}	110.794±64.49^{c}	17.59	0.11

*TAH: Total Aliphatic Hydrocarbons, ∑PAH: Total Poly Aromatic Hydrocarbons, TPH: Total Petroleum Hydrocarbons; Same superscripts (a, b, c) denote no significant differences between samples within a row ($P < 0.05$) based on Duncan's Multiple Range Test.

In sediments, hydrocarbons are adsorbed strongly to soil particles, and must be desorbed into water column before dermal adsorption by organisms. Also, ingested sediment containing hydrocarbons may pass though the aquatic body system unabsorbed and un-retained. These factors culminate to the lower accumulation factors observed in fish/sediment in comparison with fish/water.

Table 9: Source diagnostic indices of water samples for the different Clans in the different Months

Clan	Index	Nov	Feb	Jun	Sep	Ave
CN1 (Oloibiri)	E/O	1.07	1.64	1.86	1.78	1.59
	∑LMW/∑HMW	1.46	1.97	2.91	2.47	2.21
	LHC/SHC	0.21	0.27	0.41	0.32	0.30
	nC31/nC19	0.34	0.58	1.00	0.77	0.67
	Pr/Ph	1.06	1.00	0.72	0.73	0.88
	nC16 index	11.76	7.56	10.44	7.88	9.41
	CPI	0.89	0.87	1.06	1.03	0.96
CN2 (Kolo Creek)	E/O	1.12	1.02	1.44	1.65	1.30
	∑LMW/∑HMW	0.05	0.21	0.17	1.79	0.55
	LHC/SHC	2.00	1.26	2.12	0.49	1.47
	nC31/nC19	1.39	1.09	1.22	0.73	1.11
	Pr/Ph	1.17	1.29	1.24	1.46	1.29
	nC16 index	47.16	45.27	45.96	17.18	38.89
	CPI	0.19	0.21	0.20	0.93	0.38
CN3 (Anyama)	E/O	1.06	3.11	2.99	1.25	2.10
	∑LMW/∑HMW	4.44	3.28	0.17	0.77	2.16
	LHC/SHC	0.17	0.35	3.83	0.49	1.21
	nC31/nC19	0.32	0.16	1.11	0.35	0.48
	Pr/Ph	1.22	0.68	0.52	0.70	0.78
	nC16 index	4.67	6.55	28.05	8.42	11.92
	CPI	0.68	0.49	0.66	0.64	0.62
CN4 (Abureni)	E/O	1.66	1.55	3.10	1.22	1.88
	∑LMW/∑HMW	2.39	1.37	2.43	1.40	1.89
	LHC/SHC	27.39	42.67	23.02	122.85	53.97
	nC31/nC19	0.72	0.73	0.19	0.37	0.50
	Pr/Ph	0.91	0.94	1.04	0.91	0.95
	nC16 index	7.18	11.39	4.60	12.77	8.98
	CPI	0.91	0.96	0.64	0.86	0.84

3.7 Hydrocarbons source identification

Table 9 reveals the source diagnostic indices for the different months in the different clans in the surface waters. The even-to-odd numbered alkanes (E/O) ratios reflected petrogenic inputs in all clans with an average of 1.59, 1.30, 2.10, and 1.88 for clan CN1, CN2, CN3, and CN4 respectively. According to Adeniji *et al* (2017b), and Chokor (2022, 2024), Even (nC_{16}, nC_{18}, $nC_{20,}$) to odd (nC_{15}, nC_{17}, nC_{19}) numbered alkane ratio (E/O) less than one (1) indicate biogenic sources whereas values greater than one are implicative of petroleum inputs. The petrogenic source of hydrocarbons inputs were buttressed by the sum of low molecular weight to high molecular weight hydrocarbons (∑LMW/∑HMW) ratios that were larger than one; except at Kolo (CN2) in the month of September and at Oloibiri clan (CN1) in the months of June and September which highlight contributions from

biogenic sources. According to Farid *et al* (2014), Kanzari *et al* (2014), and Chokor (2022), ∑LMW/∑HMW ratio close to or greater than one implicate petroleum and plankton origin whereas values less than one (1) represent hydrocarbons sourced from higher plants, marine animals, and sedimentary bacterial.

The carbon preference index (CPI) values at Oloibiri and Aburinni Clans were close to one indicating crude oil sources, whereas those at Anyama and Kolo clans were much below unity (1); an affirmation of degraded petroleum. CPI values higher than one especially in the range of 3 – 10; indicate biogenic sources; while values in the neighborhood of one, and less than one represent petroleum sources and degraded petroleum respectively (Onyema *et al*., 2013; Abdallah *et al*., 2015; Chokor, 2023). The CPI also measures the odd to even numbered hydrocarbon but it is calculated in a different form (Eq. 6).

$$CPI_{25\text{-}33} = 0.5 \times [(C_{25} - C_{33})/ (C_{24} - C_{32})] + [(C_{25} - C_{33})/ (C_{26} - C_{34})] \quad (6)$$

Biogenic hydrocarbons are characterized by the dominance of pristine (Pr) over phytane (Ph); giving a high ratio of Pr/Ph which reliably implicates the absence of petroleum (Abdullah *et al*., 2015; Chokor, 2022). In this study, the ratios ranged from 0.52 – 1.46 in all samples with mean of: CN1 (0.88), CN2 (1.29), CN3 (0.78), and CN4 (0.95). This evidenced that the sources of hydrocarbons in Anyama, Oloibiri, and Aburinni Clans were largely petrogenic, while that at Kolo Creek showed strong evidence of biogenic contributions. The n16 –index also suggested that source of hydrocarbons were petrogenic; however evidences of mixed origin also abound. The n16 –index is computed by dividing the total n-alkanes concentrations by the concentration of nC_{16}. The value is low (<16) with oil contaminated samples; it is larger (>50) when the contamination is biological (Tolosa *et al*., 2004; Chokor, 2023). Biogenic sources could come from marine algae or terrestrial plant waxes. The formal is indicated by large presence of odd numbered hydrocarbons in the range of nC_{15} – nC_{21}; whereas the latter is implicated by the dominance of nC23 – nC_{31} odd numbered alkanes. The long chain to short chain hydrocarbons (LHC/SHC) ratio and the nC_{31}/nC_{19} ratio are useful in this regard. The LHC/SHC ratio is computed from Eq. 7

$$LHC/SHC = [\sum (nC_{27} + nC_{29} + nC_{31})/\sum (nC_{15} + nC_{17} + nC_{19})] \quad (7)$$

LHC/SHC ratio greater than 4 is implicative of terrestrial plant waxes, ratios within 0.21 – 0.80 suggests phytoplankton (marine) origins. A mixture of both source is implied when the ratio falls between 2.38 and 4.33 (Fagbote and Olanipekun, 2013; Adeniji *et al*., 2017a). The distributions ratios of LHC/SHC in this study, gave much credence to marine inputs in CN3 (Anyama) and CN1 (Oloibiri). However, CN4 (Aburinni) was particularly noted for terrestrial inputs, while Kolo Clan showed evidence of mixture of both sources. The nC_{31}/nC_{19} ratio however showed evidence of terrestrial input in CN1 and CN2, while the other two clans radiated mixture of both sources. The conclusion drawn from the above diagnoses is that the presences of TPHs in the water columns were largely due to petroleum anthropogenic sources with very little contributions from natural biogenic source.

Similar diagnostic indices applied to sediments and the aquatic species reveal that sediment's TPHs were due to both anthropogenic and biogenic inputs; while that of the aquatic species indicated large biogenic inputs but the impacts of anthropogenic sources were obvious.

4. Conclusion

The study evaluated the status, source, and impacts of hydrocarbons in water, sediment, and aquatic species of the Creeks in Ogbia LGA. The mean values of 6299.11±1068.61 µg/L, 973.64 ± 304.85 mg/kg, and 110.79 ± 64.4mg/kg obtained for water, sediment, and aquatic species respectively, were much higher than the established statutory guidelines. The large variations in temporal and spatial distributions are indicative of anthropogenic contaminations. Aliphatic and source fingerprinting implicated anthropogenic petroleum inputs as major in the pollution of the aquatic environment. Levels of petroleum hydrocarbons were significantly higher in the wet seasons than in dry ones. Strong correlations were found for the levels of TPHs in water and sediment. The TPHs concentrations in aquatic species tended to reflect same pattern; however bioaccumulations in the species were a function of their trophic levels and their habitat niche. The calculated hazard indices implicated abilities to cause great ecological harms, and medium to slightly high health risks to human.

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