

The Distribution and Ecological Risk Assessment of Polyaromatic Hydrocarbons in the Surface Water of Okpare River, Olomu, Delta State, Nigeria

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Abstract: The Okpare River, a tributary of the Niger Delta, plays a crucial role in sustaining numerous communities in the Olomu kingdom, Delta state, Nigeria. Unfortunately, the river has been significantly affected by oil spills resulting from illegal bunkering activities in the Okpare community. Given that crude oil is a source of polycyclic aromatic hydrocarbons (PAHs), understanding their source, distribution and ecological risk is imperative for developing effective mitigation strategies and safeguarding the ecosystem health. This study utilized the risk quotient method and diagnostic ratio to determine the ecological risk and the sources of the 16 priority PAHs listed by USEPA in the surface water of Okpare River. Samples were collected for 12 months, encompassing both wet (May 2022 to October 2022) and dry seasons (November 2022 to April 2023). A total of eighty-four Samples were obtained from seven sampling stations positioned 100 meters apart along the Okpare river. All samples were collected and analyzed adhering to the standard methods and procedures. Among the 16 priority PAHs determined Benzo(b)fluoranthene concentrations with average values of 46.50 ng/L and 13.17 ng/L had the highest concentration in the wet season while dibenz (a, h) anthracene levels with average values of 24.00 ng/L and 53.00 ng/L was higher in the dry season. The distribution of PAHs in the Okpare River in the wet season was in the order 5 rings > 6 rings > 4 rings > 3 rings > 2 rings; the order of magnitude in the dry season was 6 rings > 5 rings > 4 rings > 3 rings > 2 rings. BaA, BbF, BkF, BaP, InP, DBA, and B(ghi)p posed a high ecological risk in the wet season, while BbF, DBA, and B(ghi)p demonstrated high ecological risk during the dry season. The source of PAH in this study may be due to pyrogenic and petrogenic sources. Effective mitigation strategies should target these pollution sources, underscoring the importance of adopting sustainable practices to preserve the ecological balance of the Okpare River and enhance the well-being of adjacent communities.

Keywords: Ecological Risk, Petrogenic, Pollutant, Okpare, Risk Quotient, Season.

1. INTRODUCTION

The advent of cutting-edge technology has initiated an era of industrialization, fostering an improved utilization of natural resources. This has resulted in the facilitation of daily life and the enhancement of the human quality of living. Nevertheless, this advancement has imposed a significant toll due to the negative impacts of pollutants discharged into the environment during the extensive exploration, extraction, and processing of natural resources on an industrial scale. Environmental pollution emerges as the predominant challenge facing humanity today, given its role as a major contributor to mortality and morbidity in numerous societies (Ukaogo *et al.* 2020). Environmental pollution is the contamination of air, land, and water's physical, chemical, and biological components to an extent that adversely affects normal environmental processes

(Pankaj *et al.* 2020). Contamination can be characterized as the presence of impurities or substances that are not necessarily harmful, finding themselves in inappropriate locations, or the existence of substances at concentrations surpassing background levels in a physical body, material, or the natural environment (Chapman, 2007; Frank, 2017). The difficulties presented by environmental pollution encompass climate change, deforestation, loss of biodiversity, water scarcity, and disease outbreaks (Baguma *et al.* 2022). Environmental pollution originates from both natural processes, such as volcanic eruptions, rock weathering, dust storms, and wildfires, and human activities like urbanization and mining (Pankaj *et al.* 2020). However, it is human activities that evoke heightened concern, as they contribute a larger quantity of pollutants at levels considered harmful to plants, animals, and humans (Ming *et al.* 2021).

Pesticides, polycyclic aromatic hydrocarbons (PAHs), and trace metals are a few contaminants that can affect water quality. PAHs represent chemical compounds comprising two or more fused aromatic rings (Liu *et al.* 2020). These compounds arise from incomplete combustion of organic materials like fossil fuels, wood, and tobacco, with additional natural occurrences during events such as forest fires and volcanic eruptions. Anthropogenic sources, including vehicle emissions, power plants, chemical and oil-shale industries, coke production, and urban sewage, contribute significantly to environmental PAHs levels (Liu *et al.* 2020; Ruqayah *et al.* 2023). PAHs can be classified into pyrogenic, petrogenic, and biogenic/diagenetic types based on their thermodynamic properties, originating from different sources (Grmasha *et al.* 2023). Parent PAHs are made up of fused rings, ranging from benzene to nine rings, and do not contain heteroatoms or alkyl groups. Alkyl PAHs are defined as PAHs having an alkyl group (Stogiannidis, 2015). PAHs are divided into high molecular weight and low molecular weight categories according to their properties and molecular weights (Liu *et al.* 2020). Low molecular weight (LMW) compounds have two to three rings, whereas high molecular weight (HMW) compounds have four to six rings (Eno *et al.* 2015). LMW PAHs are not known for carcinogenic effects, in contrast, HMW PAHs, with higher molecular weights, are significantly carcinogenic and mutagenic (Karlsson and Viklander 2008; Stogiannidis, 2015). The United State Environmental Protection Agency, (2014) has identified 16 priority polycyclic aromatic hydrocarbons (PAHs), which are classified by the International Agency for Research on Cancer (IARC) as likely or possibly carcinogenic. Specifically, attention has been focused on several PAHs including benz(a)anthracene, chrysene, dibenz(a,h)anthracene, and benzo(a)pyrene. Additionally, benzo(ghi)perylene, indeno(1,2,3-c,d)pyrene, benzo(b)fluoranthene, and benzo(k)-fluoranthene are highlighted among PAHs with carcinogenic properties. Of these, benzo(a)pyrene stands out as the most extensively studied PAH concerning its carcinogenic characteristics (IARC, 2010; Samburova *et al.* 2017). The Sixteen PAHs classified as priority pollutants are of particular concern due to their mutagenic properties, with some recognized as probable human carcinogens (Halfadji *et al.* 2021).

The Okpare River, a tributary of the Niger Delta, plays a crucial role in sustaining numerous communities in the Olomu kingdom, Delta state, Nigeria. It serves as a primary source for fish, domestic water supply, irrigation, and recreational activities. With historical significance as a trading port during the colonial era, Okpare River continues to function as a local trading hub. Unfortunately, the river has been significantly affected by oil spills resulting from illegal bunkering activities in the Okpare community (Edjere and Asibor, 2020; Onajite and Ovie, 2022). Given that crude oil is a source of polycyclic aromatic hydrocarbons (PAHs), understanding their source, distribution and ecological risk is imperative for developing effective mitigation strategies and safeguarding the ecosystem health and human well-being associated with the Okpare River and its surrounding communities.

2. MATERIALS AND METHOD

The study area

The study area is Okpare River in Olomu, Ughelli South Local Government Area, Delta State, Nigeria. Olomu is one of the oldest kingdoms in the Urhobo ethnic nationality of the Niger Delta region (Ekeh *et al.* 2012) and has crude oil wells. The Okpare River took its source from Umuaja in Umutu and empties into the Atlantic Ocean at Forcados Estuary (Onajite and Ovie, 2022). The study area is located between latitude 05°27'N 47.0 and N05°27'28.0 and longitude E005°54'19 and E005°54'3. The river runs North-West to South-East. The region experiences a dry season from November to April, characterized by a dusty haze from the northeast winds, the wet season, runs from May to October with a brief dry spell in August (Ekanem and Nwagbara, 2005). The area is mostly rainforest with occasional swamplands and is rich in fruit trees, palm trees, and lumber trees. The major occupations of the people in the region include fishing, transportation, farming, and trading. Fig. 1 shows the map of the study area showing the sampling points.

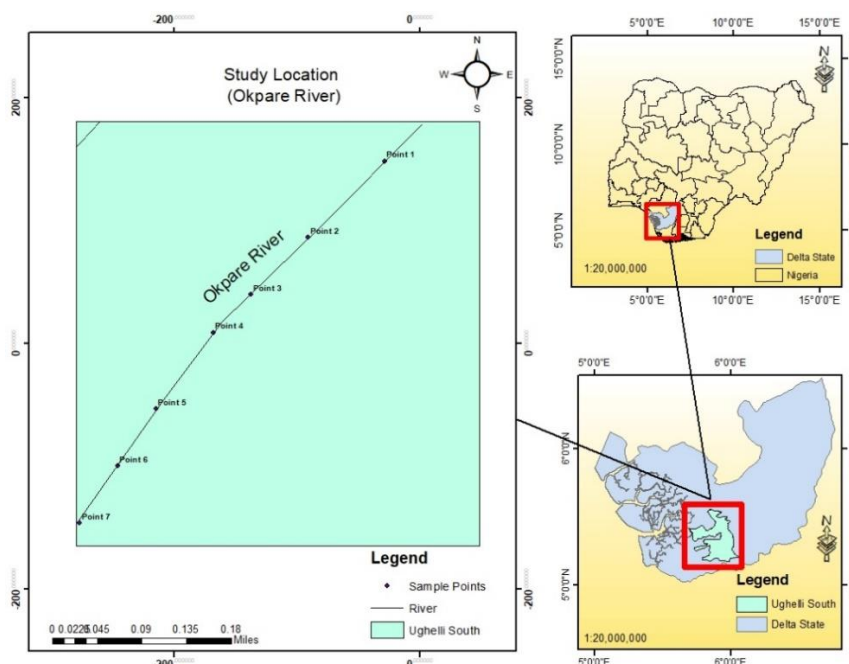


Figure 1: Map of the Study Area

Sampling of Surface Water

Surface water samples were collected monthly for 12 months during both wet and dry seasons. In the wet season (May 2022 to October 2022) and dry season (November 2022 to April 2023) (Ekanem and Nwagbara, 2005) using water sampler. Samples were collected from 7 sampling stations situated 100 meters apart along the river, according to APHA, (2017) standard procedures. Amber glass bottles were employed to collect water samples for PAHs determination. All water samples were preserved by cooling at approximately 4°C in an ice box. All ex-situ analysis was performed at Dukoria Laboratory Ltd in Effurun, Delta State.

Analytical Methods

Extraction of Water Samples for PAHs was done Using Liquid-Liquid Extraction Chromatographic Method (APHA 6440 B). The water samples were combined with dichloromethane in a separating funnel to isolate the organic layer from the aqueous layer. After concentration of the organic layer in a round-bottom flask to approximately 2–3 mL using a rotary evaporator, it was then transferred to a clean beaker and covered with foil. To begin column chromatography, a silica gel-packed column was primed with a 15 mL n-Hexane slurry, followed by the gradual introduction of the concentrated organic extract into the column. Subsequently, 25 mL of n-Hexane was introduced to the column to fractionate the aromatics into a separate clean beaker, which was also covered with foil. Additionally, 2–3 spatulas of anhydrous sodium sulfate were added to each beaker to eliminate any remaining water traces. Utilizing a pipette, the clear portion of the extract was drawn into a clean Gas Chromatography vial, corked, covered, and appropriately labeled for subsequent analysis of PAHs via Gas Chromatography (APHA, 2017).

Statistical Analysis

The data generated underwent descriptive statistical analysis. The risk quotient method and the diagnostic ratio calculations was carried out with Microsoft Excel Office 365.

Ecological Risk Assessment

The impact of individual PAHs on aquatic organisms in water can be effectively assessed using the risk quotient (RQ) method, as outlined by Cao et al. (2010), Wang et al. (2021). The RQ is calculated through the formulas:

$$RQNCs = CPAHs / (CQ(NCs)) \dots\dots\dots \text{(Equation 1)}$$

$$RQMPCs = CPAHs / (CQ(MPCs)) \dots\dots\dots \text{(Equation 2)}$$

The risk quotient method was first proposed by Kalf et al. (1997). The risk quotient method proposed by Kalf et al. (1997) could only account for the ecological risk assessment of only 10 PAHs of the United State Environmental Protection Agency listed priority PAHs and could not account for 6 PAHs (acenaphthylene, Acenaphthene, fluoranthene, pyrene, benzo(a)anthracene, benzo(b)fluoranthene). This method was however modified by Cao et al. (2010) to account for the ecological risk of the 16 PAHs marked by USEPA (Lan et al. 2020). Table 1 presents the risk categories in the risk quotient method used to ascertain the ecological risk of PAHs in water (Lan et al. 2020).

Table 1: Risk Quotient Classification for Ecological Risk

RQNCs	RQMPCs	Risk Rank
0		Risk Free
≥ 1	< 1	Moderate Risk
	≥ 1	High Risk

3. RESULTS AND DISCUSSION

In the study area, the concentrations of Naphthalene had mean values of 1.50 ng/L and 0.17 ng/L in the wet and dry seasons respectively. Acenaphthylene levels had mean concentrations of 0.5 ng/L and 3.00 ng/L in the wet and dry seasons respectively. Meanwhile, Acenaphthene concentrations had average values of 0.17 ng/L and 3.67 ng/L in the wet and dry seasons respectively. Fluorene concentrations had mean levels of 0.33 ng/L and 1.33 ng/L in the wet and dry seasons respectively.

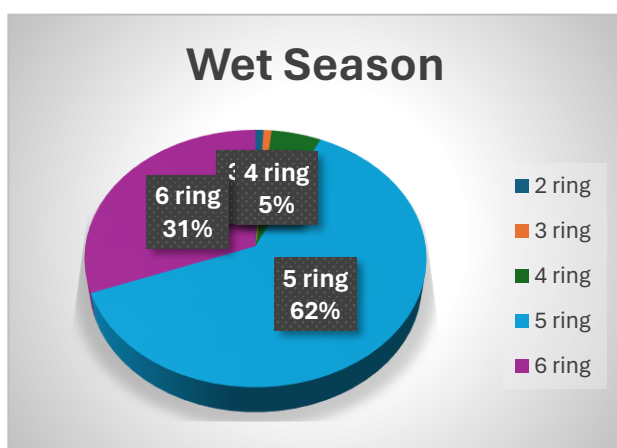


Figure 2: PAHs Distribution During the Wet Season

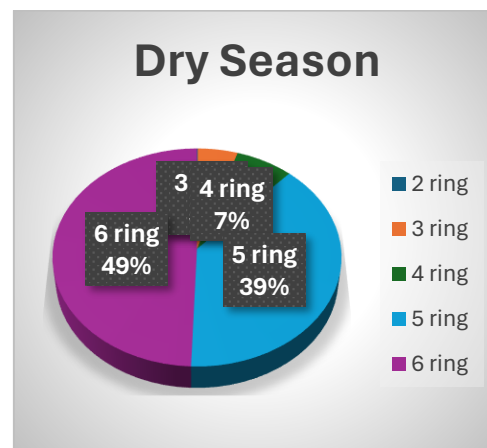


Figure 3: PAHs Distribution During the Dry Season

Phenanthrene concentrations had mean values of 0.33 ng/L and 0.33 ng/L during the wet and dry season. Anthracene concentrations had average values of 0.17 ng/L and 0.33 ng/L during the wet and dry seasons respectively. Fluoranthene concentrations had mean values of 0.17 ng/L and 0.50 ng/L during the rainy and dry season. Pyrene levels had average values of 0.50 ng/L and 0.33 ng/L during the wet and dry seasons respectively. Chrysene levels had average values of 3.17 ng/L and 6.50 ng/L. Benz(a)anthracene levels had average values of 5.50 ng/L and 5.17 ng/L. Benzo(b)fluoranthene concentrations had average values of 46.50 ng/L and 13.17 ng/L during the wet and dry seasons respectively. Benzo(b)fluoranthene had the in the highest mean concentration in the surface water during the wet season (Fig. 4). Benzo(k)fluoranthene concentrations had mean values of 3.50 ng/L and 34.67 ng/L during the rainy and dry season. Benzo(a)pyrene levels had average values of 31.33 ng/L and 18.17 ng/L during the wet and dry seasons respectively. Benzo(a)pyrene concentrations are within the permissible limits set by World Health Organization, (2017) (Table 2). Indeno (1,2,3-cd) pyrene levels had average values of 31.83 ng/L and 3.67 ng/L. Dibenz (a, h) anthracene levels had average values of 24.00 ng/L and 53.00 ng/L. Dibenz(a,h)anthracene had the highest mean concentrations in the surface water during the dry season (Fig. 4). Benzo (g,h,i) perylene concentrations had average values of 17.83 ng/L and 35.83 ng/L during the wet and dry seasons respectively.

Table 2: Polyaromatic Hydrocarbons Concentrations in the Study Area

Polyaromatic Hydrocarbons	Number of Rings	Wet Season		Dry Season		WHO, 2017
		Range (ng/L)	Mean (ng/L)	Range (ng/L)	Mean (ng/L)	
Naphthalene	2	1.00 – 3.00	1.50	0.00 – 4.00	0.17	
Acenaphthylene	3	0.00 – 2.00	0.50	0.00 – 11.00	3.00	
Acenaphthene	3	0.00 – 1.00	0.17	0.00 – 8.00	3.67	
Fluorene	3	0.00 – 1.00	0.33	0.00 – 4.00	1.33	
Phenanthrene	3	0.00 – 1.00	0.33	0.00 – 1.00	0.33	
Anthracene	3	0.00 – 1.00	0.17	0.00 – 1.00	0.33	
Fluoranthene	4	0.00 – 1.00	0.17	0.00 – 2.00	0.50	
pyrene	4	0.00 – 1.00	0.50	0.00 – 1.00	0.33	
Chrysene	4	1.00 – 7.00	3.17	0.00 -24.00	6.50	
Benz(a)anthracene	4	1.00 – 13.00	5.50	1.00 – 12.00	5.17	
Benzo(b)fluoranthene	5	10.00 – 71.00	46.5	0.00 – 22.00	13.17	
Benzo(k)fluoranthene	5	10.00 – 82.00	3.50	0.00 – 98.00	34.67	
Benzo(a)pyrene	5	25.00 – 54.00	31.33	4.00 – 28.00	18.17	700
Indeno (1,2,3-cd) pyrene	5	8.00 – 49.00	31.83	18.00 – 48.00	3.67	
Dibenz(a,h)anthracene	6	10.00 – 42.00	24.00	31.00 – 103.00	53.00	
Benzo(g,h,i)perylene	6	6.00 – 42.00	17.83	24.00 – 46.00	35.83	
Total		-	167.33	-	179.84	

Note: WHO, 2017 – World Health Organization permissible limits for drinking water quality.

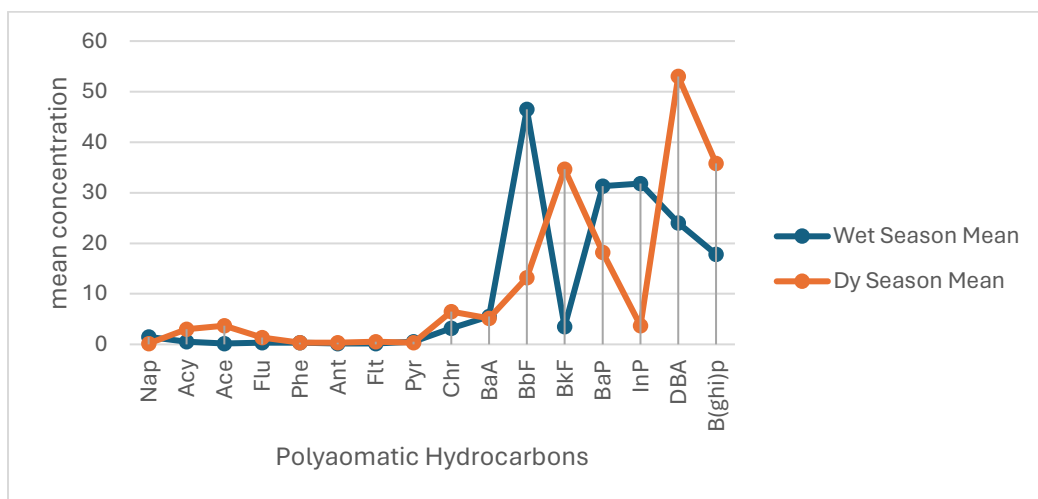


Figure 4: PAHs obtained in the Surface Water during the wet Season Compared to the Dry Season.

4. DISCUSSION

Distribution of PAHs in the Surface Water

Two categories of PAHs were identified from the measurements in this study: 2 to 3 rings (low molecular weight, LMW), four rings, and 5 to 6 rings (high molecular weight, HMW). The distribution of PAHs in the Okpare River in the wet season was in the order 5 rings > 6 rings > 4 rings > 3 rings > 2 rings (Figure 2) while the order of magnitude in the dry season was

6 rings > 5 rings > 4 rings > 3 rings > 2 rings (Fig. 3). The percentage of high molecular weight PAHs (PAHs with 4 – 6 rings) are higher than the low molecular weight PAHs (PAHs with 2 – 3 rings). LMW PAHs are not known for carcinogenic effects and in contrast, HMW PAHs, with higher molecular weights, are significantly carcinogenic and mutagenic (Karlsson and Viklander 2008; Stogiannidis, 2015). The PAHs distribution obtained in this study is not in agreement with higher concentration of low molecular weight PAHs in the order 3 rings > 4 rings > 5 rings > 2 rings > 6 rings reported by Lui et al. (2020) in a similar study conducted in the Wuhai and Lingwu section of the Yellow River, China. Eno et al. (2015) reported higher concentration of high molecular weight PAHs in the surface water of the lower reach of Qua Iboe River Estuary, Akwa Ibom State. The potential volatilization of LMW PAHs to the environment due to their relatively high vapour pressure could account for the low percentage of these PAHs in the study area (Eno et al. 2015). Furthermore, two- to three-ring PAHs biodegrade more quickly and as opposed to 4-6 ring PAHs, volatilize. Additionally, the reason for the greater concentrations of HMW-PAHs in this study might be that they are attached to soot from the burning of bush and fossil fuel which finally ends up in the water (Eno et al. 2015). The 16 USEPA listed priority PAHs in the range 0.17 – 46.50 ng/L (wet season) and 0.17 – 53.00 ng/L (dry season) obtained in this study are lower than the range of 464.00 – 992.00 ng/L obtained from Euphrates River in Iraq (Grmasha et al. 2023), 46.53 – 222.00 ng/L in Shanghai river (Lui et al. 2016) but comparable to the range 0.24 – 58.72 ng/L reported by Hassan et al. 2019 from a study conducted in Euphrates estuary.

Ecological Risk Assessment

The ecological risk assessment results of the 16 listed priority polycyclic aromatic hydrocarbons (PAHs) in the surface water of the Okpare River during the wet and dry seasons are shown in Table 4.8 and Table 4.9. The RQNCs values of the individual PAHs did not exceed 1, except for Benz(a)anthracene (BaA), Benzo(b)fluoranthene (BbF), Benzo(a)pyrene (BaP), Indeno(1,2,3-cd) pyrene (InP), Dibenz(a,h)anthracene (DBA) and Benzo(g,h,i)perylene (B(ghi)p) indicating that PAHs with RQNCs surpassing 1 pose a moderate ecological risk (Table 3). While the RQMPCs values for Benz(a)anthracene (BaA), Benzo(b)fluoranthene (BbF), Benzo(a)pyrene (BaP), Indeno(1,2,3-cd) pyrene (InP), Dibenz(a,h)anthracene (DBA), and Benzo(g,h,i) perylene (B(ghi)p) exceeded 1, signifying a high ecological risk associated with these specific PAHs. During the dry season, the RQNCs values for all PAHs were above 1, except for Naphthalene (Nap), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt) and Pyrene (Pyr) suggesting that PAHs with values surpassing 1 represented a moderate ecological risk (Table 3). Additionally, the RQMPCs values for BbF, DBA, and B(ghi)p were higher than 1, indicating a high ecological risk associated with these PAHs.

Table 3: Ecological Risk Assessment of Polyaromatic Hydrocarbons in the Study Area

Polyaromatic Hydrocarbons	Abbreviation	Mean of the study (ng/L)	NCs/ (ng/L)	MPCs/ (ng/L)	RQNCs	RQMPCs	Mean of the study (ng/L)	NCs/ (ng/L)	MPCs/ (ng/L)	RQNCs	RQMPCs
Naphthalene	Nap	1.50	12	1200	0.13	0.0013	0.17	12	1200	0.014	0.00014
Acenaphthylene	Acy	0.50	0.7	70	0.71	0.007	3.00	0.7	70	4.29	0.043
Acenaphthene	Ace	0.17	0.7	70	0.24	0.002	3.67	0.7	70	5.24	0.052
Fluorene	Flu	0.33	0.7	70	0.47	0.0047	1.33	0.7	70	1.90	0.019
Phenanthrene	Phe	0.33	3	300	0.11	0.0011	0.33	3	300	0.11	0.0011
Anthracene	Ant	0.17	0.7	70	0.24	0.0024	0.33	0.7	70	0.47	0.0047
Fluoranthene	Flt	0.17	3	300	0.057	0.00057	0.50	3	300	0.167	0.00167
Pyrene	Pyr	0.50	0.7	70	0.71	0.0071	0.33	0.7	70	0.47	0.0047
Chrysene	Chr	3.17	3.4	340	0.93	0.009	6.50	3.4	340	1.91	0.019
Benz(a)anthracene	BaA	5.50	0.1	10	55.00	0.55	5.17	0.1	10	51.70	0.52
Benzo(b)fluoranthene	BbF	46.5	0.1	10	465.00	4.65	13.17	0.1	10	131.70	1.32
Benzo(k)fluoranthene	BkF	3.50	0.4	40	8.75	0.0875	34.67	0.4	40	86.68	0.87
Benzo(a)pyrene	BaP	31.33	0.5	50	62.66	0.63	18.17	0.5	50	36.34	0.3634
Indeno(1,2,3-cd) pyrene	InP	31.83	0.4	40	79.58	0.79	3.67	0.4	40	9.18	0.09
Dibenz(a,h)anthracene	DBA	24.00	0.5	50	48.00	0.48	53.00	0.5	50	106.00	1.06
Benzo(g,h,i)perylene	B(ghi)p	17.83	0.3	30	59.43	0.59	35.83	0.3	30	119.43	1.194

Note: NCs – Lowest Risk Standard Value; MPCs – Highest Risk Standard Value; RQNCs – Lowest Risk Quotient Value; RQMPCs – Highest Risk Quotient Value.

Table 4: The Results of Diagnostic Ratio of PAHs in the Study Area

Diagnostic Ratio	Ant/Ant+Phe		InP/InP + B(ghi)p		Flt/Flt+Pyr	
	Wet	Dry	Wet	Dry	Wet	Dry
Diagnostic Ratio Value	0.16	0.5	0.64	0.09	0.25	0.6
Source	Combustion Biomass	Combustion of Biomass	Combustion of Biomass	Petroleum Contamination	Petroleum Contamination	Combustion of Biomass

Source Apportionment

Diagnostic ratios or indicator ratios play a crucial role in distinguishing between various PAHs sources. These ratios serve as valuable tools for discerning petrogenic and pyrogenic sources. Analyzing these ratios facilitates the identification of the specific origins of PAHs contamination (Szopińska *et al.* 2019). A ratio of Ant / (Ant + Phe) less than 0.10 suggests petroleum input, while a value exceeding 0.1 is indicative of the combustion of coal or biomass (Hussain *et al.* 2016; Dong *et al.* 2021). Similarly, Inp /Inp + B(ghi)p ratio, less than 0.2 signifies a petroleum origin, between 0.2 and 0.5 indicates petroleum combustion, and values exceeding 0.5 are typical of the combustion of coal, wood, and biomass. In terms of Flt/ (Flt + Pyr) ratio, values below 0.4 imply petroleum origin, 0.4– 0.5 suggest petroleum combustion, and values exceeding 0.5 imply the combustion of coal and biomass. The diagnostic ratios for the 16 priority Polycyclic Aromatic Hydrocarbons (PAHs) examined in the study area, during the wet and dry seasons, are detailed in Table 4. The Ant/Ant+Phe ratios during both seasons were greater than 0.1, suggesting a likely source of PAHs stemming from biomass combustion. Furthermore, the InP/InP + B(ghi)p ratios surpassed 0.5 during the wet season, indicative of biomass combustion, whereas during the dry season, these ratios were below 0.2, implying a petroleum origin. Moreover, the Flt/Flt+Pyr ratio values during the wet season were below 0.4, further supporting the inference of a petroleum origin. Conversely, during the dry season, Flt/Flt+Pyr ratio values exceeded 0.5, suggesting a shift towards biomass combustion as the predominant source of PAHs during this period.

5. CONCLUSION

This study examines the concentration, distribution, sources, and ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in the surface water of the Okpare River. High molecular weight PAHs were found at elevated concentrations compared to low molecular weight PAHs, with 5-ring compounds dominating in the surface water during the wet season and 6-ring compounds in the dry season. Ecological assessment revealed a high level of ecological risk associated with some of the PAH's concentrations in the surface water. The source of PAHs in the study area is likely linked to both pyrogenic and petrogenic origins. These findings underscore the urgent need for measures to mitigate the ecological risk posed by PAHs in the aquatic environment of our study area.

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