



Source Apportionment and Health Risk of Some Organic Contaminants in Water and Suspended Particulate Matter from Imo River, Nigeria

NNA, PJ; *ORIE, KJ; KALU, NAS

Department of Chemistry, Ignatius Ajuru University of Education Port-Harcourt, Rivers State, Nigeria.

*Corresponding Author Email: oriekingsley81@gmail.com

Co-Authors Email: agaraprince@yahoo.com; okwutem2002@gmail.com

ABSTRACT: The objective of this study was to provide the source identification and assessment of potential health associated with some organic contaminant detected in water and suspended particle matter (SPM) from the Imo River, Nigeria using gas chromatography with a flame ionization detector (GC/FID) after 1:1 ratio solvent mixture (n-hexane and dichloromethane) extraction. The findings on SPM revealed the values; pH (5.42 ± 0.097 - 5.567 ± 0.057), conductivity (74.58 ± 5.390 - 87.6 ± 0.9 uS/cm), and total organic carbon (TOC) (3.626 ± 0.366 - 6.143 ± 0.176), and surface water; pH (6.077 ± 0.049 - 6.46 ± 0.52), conductivity (49.03 ± 1.430 - 58.12 ± 0.553 uS/cm). The total concentrations of both low and high molecular PAHs in SPM recorded at stations 1-3 were 4.384 mg/kg, 16.87 mg/kg, and (22.69 mg/kg); surface water 2.329 mg/L, 7.428 mg/L and 6.657 mg/L. The source apportionment of PAHs in SPM and surface water via molecular diagnostic ratios identified both petrogenic source (crude oil, petroleum spillages, and seepages) and pyrogenic sources (combustion of grass, wood, coal, and gas flaring). A further analysis with Principal Component Analysis (PCA) revealed two principal components, with 65.568% for component 1 and 34.432% for component 2 for SPM, and 62.80% for principal components and 37.2% for principal component 2 for surface water. The correlation analysis of PAHs affirmed common sources such as petrogenic and pyrogenic origins. The toxic equivalent factor of total benzo(a)pyrene for 6 Σ PAHs in SPM was recorded as 3.693 mg/kg, 1.605 mg/kg, and 0.453 mg/kg; surface water was recorded as 0.823 mg/L, 1.858 mg/L, and 0.353 mg/L, while the mutagenic equivalency quantities of SPM were recorded as 3.693 mg/kg, 1.146 mg/kg, and 0.316 mg/kg; surface water 0.739 mg/L, 0.938, and 0.577 for stations 1-3. The effect of carcinogenic PAHs in SPM was higher in adults than in children, with regards to ILCR analysis. Users of Imo Rivers should be guided on the exposure of SPM and surface water since the toxic, mutagenic, and cancer risk levels of the river are above the permissible limit in some stations.

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The issue of water quality has emerged as a significant concern for both human and animal populations in recent times, mostly as a result of the rapid expansion of human populations, intensified industrial activities, and the accelerated pace of urban development. Industrial effluents and a majority of commercial wastes are discharged onto the land, subsequently

leading to their transport into nearby rivers. The presence of persistent organic pollutants such as PAHs and PCBs is indicated by the diverse range of activities observed within this environment (Yahaya *et al.*, 2019; Adedosu *et al.*, 2013). River systems are subject to the inflow of wastewater originating from residential areas, agricultural practices, and industrial

*Corresponding Author Email: oriekingsley81@gmail.com

activities due to the swift progress of urbanization, agriculture, and industry (Wang *et al.*, 2018; Yahaya *et al.*, 2019). Suspended particulate matter (SPM) has a significant role in polluting aquatic habitats, particularly in rivers, where it serves as the primary driver of pollutant movement and transformation. Previous studies have demonstrated the significant influence of SPM on ecosystem development and the formation of stagnant water bodies (Liu *et al.*, 2016; Vercruyssen *et al.*, 2017). The primary origin of SPM can be attributed to agricultural activities, together with the erosion of weathered soil and rock caused by surface and riverbed erosion (Abali *et al.*, 2023). A small fraction of SPM originates from the synthesis of organic matter (OM) and minerals within river systems. The phenomenon of sedimentation of SPM in rivers entails the amalgamation of SPM with contaminants, which is then followed by its subsequent deposition on the riverbed (Lamba *et al.*, 2015). It has been observed that SPM typically exhibits an average particle size ranging from 0.45 to 63 μm (Zheng *et al.*, 2008; Lamba *et al.*, 2015). The uptake of contaminants onto the surface of SPM involves various mechanisms, including velocity, sedimentation, suspension, and resuspension (Abali *et al.*, 2023). The downstream transportation of these pollutants leads to a substantial degradation in water quality. According to the findings, the primary factors influencing the mobility of chemicals in water are the concentration, surface activity, and composition of SPM (Horowitz and Stephens, 2008; Jiann *et al.*, 2013). The size distribution of SPM plays a crucial role in the absorption of contaminants from the surrounding environment and the behaviour of pollutants (Liu *et al.*, 2016). The reaction encompasses various components, including alterations in water turbidity and particle composition, the photosynthetic activity of plankton, and variations in sediment flux downstream (Liu *et al.*, 2016; Vercruyssen *et al.*, 2017). In addition, it is noteworthy to acknowledge that SPM plays a vital function in governing the rate of reaction between water and sediment, the cycling of the food chain, and the metabolic rate of organisms (Binding *et al.*, 2010). Organic contaminants are carbon-based chemicals that possess the potential to cause adverse impacts on both the environment and human well-being. The presence of these contaminants can be attributed to a range of origins, encompassing industrial activity, agricultural practices, and domestic waste (Haritash and Kaushik, 2009; Ahrens *et al.*, 2010; Edori and Nna, 2018).

The entry of these substances into the environment can occur via air, water, and soil pathways, and their presence can endure for extended durations, resulting in the accumulation of these substances in organisms

(Brennan *et al.*, 2021; Owwoeke *et al.*, 2023). The characteristics and consequences of organic pollutants is of utmost importance in order to devise efficient approaches for reducing their occurrence and mitigating their detrimental impacts. A crucial element in comprehending organic pollutants lies in the recognition of their multifarious origins (Ahrens *et al.*, 2010). Industrial operations, encompassing activities such as manufacturing and mining, have the potential to discharge a diverse array of chemical substances into the surrounding environment. The substances present in the environment and pose a risk of air, water, and soil contamination, encompass heavy metals, solvents, and pesticides (Rouf *et al.*, 2022). Moreover, the implementation of agricultural techniques, such as the application of fertilizers and pesticides, has the potential to introduce organic pollutants into the surrounding ecosystem (Owwoeke *et al.*, 2023). The presence of organic pollutants can be influenced by home waste, which encompasses sewage and household chemicals (Brennan *et al.*, 2021).

Polycyclic aromatic hydrocarbons (PAHs) are a significant category of organic contaminants distinguished by the inclusion of several fused aromatic rings. These molecules are produced by the process of incomplete combustion of organic material, such as fossil fuels, wood, and tobacco (Lapworth *et al.*, 2012; Donlawson *et al.*, 2020). PAHs, owing to their chemical characteristics, exhibit persistence and have the capacity to accumulate within the environment, hence presenting potential hazards to both human health and the ecosystem (Ambade *et al.*, 2021; Orie *et al.*, 2022). Polycyclic aromatic hydrocarbons (PAHs) are recognized for their limited solubility in water and strong attraction to organic substances. This characteristic facilitates their effective attachment to particles, enabling their dispersion in both air and water medium (Barkhordarian *et al.*, 2012; McCarthy *et al.*, 2017). Consequently, PAHs can be transported over considerable distances, resulting in their occurrence in geographically isolated regions. Furthermore, PAHs have the potential to undergo several transformation processes within the natural environment, including degradation and bioaccumulation, which can significantly influence their fate and behaviour (Mathew & Orie, 2015). The occurrence of harmful health impacts, such as cancer, respiratory issues, and developmental difficulties, has been linked to the exposure of individuals to polycyclic aromatic hydrocarbons (PAHs). Moreover, the existence of these species within the environment has the potential to inflict harm onto aquatic organisms, disturb the delicate balance of food chains, and exert an influence

on the total biodiversity (Barkhordarian *et al.*, 2012; Ambade *et al.*, 2021).

Source apportionment plays a pivotal role in comprehending the relative contributions of various pollution sources to the overall air quality. Its aids in the identification of primary sources of pollutants, including industrial emissions, automotive exhaust, and natural sources such as dust and pollen (Rouf *et al.*, 2022). Source apportionment studies offer useful insights into the health hazards associated with certain pollutants by assessing the impact of each source. The concept of health risk pertains to the likelihood of a person or a group encountering adverse health consequences due to their exposure to specific variables or circumstances (Uma *et al.*, 1989; Rouf *et al.*, 2022). Various factors can contribute to this phenomenon, encompassing environmental dangers, lifestyle decisions, genetic predispositions, and the existence of underlying medical disorders (Haritash and Kaushik, 2009). Thus, objective of this study was to provide the source identification and assessment of potential health risk associated with some organic contaminants detected in water and suspended particle matter (SPM) from the Imo River, Nigeria

MATERIALS AND METHODS

Study area: The Imo River flows from Ndiianiche in Imo State's Onuimo Local Government Area to

Arondizuogu Village. The location's geographical coordinates are within the latitudinal range of 06° 49'N - 06° 47'N and the longitudinal range of 08° 17'E - 08° 16'E. Figure 1 depicts the geographical representation of the research area, which shows that the river covers an estimated area of 9100 km². The Oramirukwa—Otamiri sub-basin and the Aba River sub-basin as the primary sub-basins contained within the basin was identified by researchers (Uma *et al.*, 1989). The occurrence of orographic rainfall is widely recorded in the region, including an area that exceeds 25 Km². The average annual precipitation varies between 1800mm and 2500mm, but the average annual temperature reaches 20 °C. The region consists of a sparsely vegetated shrubby rainforest, which is distinguished by a higher prevalence and diversity of plant species observed at particular elevations on the windward sides of hills.

Table 1: Coordinates of the Area Sampled in Imo River

S/No	Stations	Name	Latitude	Longitude
1	Station 1	WS-1	6.724848	8.349022
2		WS-2	6.718754	8.355788
3		WS-3	6.711308	8.359382
4		WS-4	6.703998	8.362911
5	Station 2	WS-5	6.696190	8.367720
6		WS-6	6.689409	8.372068
7		WS-7	6.682995	8.378971
8	Station 3	WS-8	9 6.777613	83284690
9		WS-9	6.671852	8.390907
10		WS-10	6.666399	8.395739

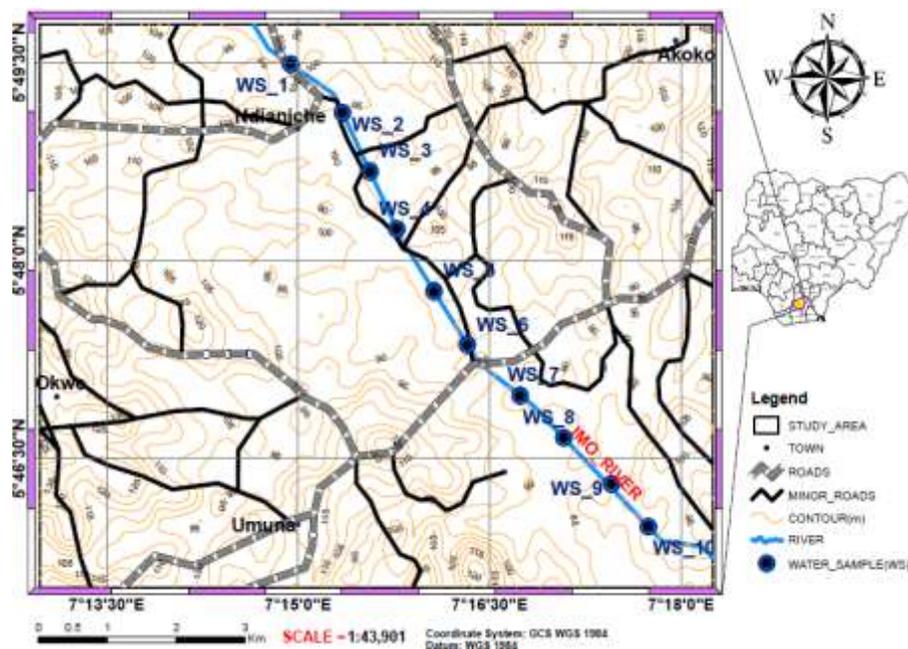


Fig 1: Geographical representation of the research area

Sample collection: Water samples were collected from ten (10) gauge stations along a 100-metre section of the river, with the labels WS1 through WS10. Using 25-litre containers that had been previously cleaned,

water samples were taken from the surface. When we got back to the lab, we used a filtration method to get samples of the suspended particles. Filters constructed of 0.7-µm GF/F glass fibres were used for this purpose.

The GF/F filters were thereafter wrapped in aluminium foil and stored in double-layered plastic bags at -18 C until testing could begin.

Preparation of crude extracts

SPM sample extraction: An approximately 10-gram well-mixed sample of SPM was transferred to a solvent-free beaker. The next step was to dilute the sample using a 50 ml mixture of n-hexane and dichloromethane, with a volumetric ratio of 1:1. After that, the sample was subjected to sonication at 70 °C for 10 to 15 minutes. A clear extract was obtained by adding 10 g of anhydrous sodium sulphate to the sample. After solvent extraction, the mixture was concentrated to a volume of about 2 ml in a flask with a round bottom using a vacuum rotary evaporator. Solid-phase extraction with alumina/silica gel was used to successfully recover PAH extracts from SPM. The concentrated solvent (2 ml) was carefully added to the filled column. After that, the sample was fractionated using 10 ml of dichloromethane. A 20-ml volume of n-hexane was used to elute saturated aliphatic hydrocarbons, while an aromatic hydrocarbon mixture of 30-ml volume was prepared by mixing hexane with dichloromethane in a 90:10 (v/v) ratio. The aromatic hydrocarbon solvent that was extracted from the combination was re-concentrated. After that, the material was moved into a vial for gas chromatography with a flame ionisation detector (GC/FID) analysis.

Water sample extraction: Precisely 10 g of uniformly mixed SPM was transferred into a beaker that had been washed with solvent. We next treated the sample with 50 mL of a solvent mixture consisting of n-hexane and dichloromethane in a volumetric ratio of 1:1. After that, the sample was subjected to a sonicator set to 70 °C for a duration of 10 to 15 minutes. A clear extract was obtained by treating the sample with approximately 10 g of anhydrous sodium sulphate. After solvent extraction, the mixture was concentrated to a volume of about 2 ml in a flask with a round bottom using a vacuum rotary evaporator. The PAH extracts from SPM were obtained by means of solid-phase extraction employing alumina/silica gel. It was with great care that the filled column was given the concentrated solvent (2 ml). Then, 10 millilitres of dichloromethane were added for fractionation. To separate saturated aliphatic hydrocarbons from aromatic hydrocarbons, 20 mL of n-hexane was used, and 30 mL of a 90:10 (v/v) combination of hexane and dichloromethane was used. An aromatic hydrocarbon, which served as the solvent in the combination, was re-concentrated. The next step was to transfer the substance to a vial for analysis using gas

chromatography in conjunction with a flame ionisation detector (GC/FID).

Health risk assessment: Potential toxicity based on carcinogenic PAHs (CPAHs): The hydrodynamics of river systems can result in the re-suspension of pollutants that have accumulated in the sediments of river beds (Tavakoly-Sany *et al.*, 2014; Spahr *et al.*, 2020). In this investigation, the researchers utilized the benzo(a)pyrene toxic equivalency quotient (TEQ) and incremental lifetime cancer risk (ILCR) factors to assess the potential risks associated with exposure to PAHs present in the SPM of the river within the designated study area. The existing body of research has thoroughly investigated the potential risks linked to PAHs present in SPM and several other environmental mediums. In order to assess the relative risks, this research has employed carcinogenic equivalency factors (BaPTEQ) (equation 1) and mutagenic equivalence factors (BaPMEQ) (equation 2) to evaluate the hazardous nature of benzo[a]pyrene (BaP). The evaluation of the BaP carcinogenic equivalency factor (BaPTEQ) for polycyclic aromatic hydrocarbons (PAHs) was conducted utilizing the subsequent equation 1:

$$BaPTEQ = \sum Ci \times BaPTEF \quad 1$$

Where BaP_{TEF} is the cancer potency relative to BaP, and Ci is the individual PAH concentration.

The BaP mutagenic equivalency factor (BaP_{MEQ}) for the PAHs was evaluated by using Equation 2:

$$BaPMEQ = \sum Ci \times BaPMEF \quad 2$$

Where BaP_{MEF} is the mutagenic potency relative to BaP, and Ci is the individual PAH concentration. The BaP carcinogenic equivalency factors (BaP_{TEFs}) of the seven carcinogenic PAHs used were BaP (1), BaA (0.1), BbF (0.1), BkF (0.01), Chry (0.001), DahA (1) and IndP (0.1) (Spahr *et al.*, 2020). The BaP mutagenic potency factors (BaP_{MEFs}) were BaP (1), BaA (0.082), BbF (0.25), BkF (0.11), Chry (0.017), DahA (0.29) and IndP (0.31) (Cao *et al.*, 2010; Howard *et al.* 2021) (Tavakoly-Sany *et al.*, 2014).

Incremental Lifetime Cancer Risk (ILCR): In order to calculate the ILCR, the chronic daily intake (CDI) is computed with Equation 3, thereafter, the ILCR was computed with Equation 4;

$$CDI = \frac{C \times Ingr \times EF \times ED \times CF \times SFO}{BW \times AT} \quad 3$$

$$ILCR = CDI \times CSF \quad 4$$

The oral slope factor (SFO) is $(\text{mg/kg/day})^{-1}$. Bamuwamye *et al.* (2015) reported the following SFO concentrations (mg/ kg/ day) : BaA = 7.3×10^{-1} ; Chry = 7.3×10^{-3} ; BbF = 7.3×10^{-1} ; BkF = 7.3×10^{-2} ; BaP = 7.3; IndP = 7.3; and DahA = 7.3. Table 2 shows the ILCR parameters.

Table 2: Parameters for estimating human cancer risk.

Exposure variable	Child	Adult
Body weight, BW (kg)	15	60
Exposure duration, ED (years)	6	24
Exposure frequency, EF (days/year)	313	313
Averaging time, AT (days)	$52 \times 365 = 18,980$	$52 \times 365 = 18,980$
Ingestion rate, IngR (mg/kg)	200	100
Adherence factor, AF (mg/cm ²)	0.2	0.07
Adsorption fraction, ABS (unitless)	0.13	0.13
Averaging time, AT _h (h)	$52 \text{ yrs} \times 365 \text{ days/yr} \times 24 \text{ h/day} = 455,520$	$52 \text{ yrs} \times 365 \text{ days/yr} \times 24 \text{ h/day} = 455,520$
Conversion factor, CF	1×10^{-6}	1×10^{-6}
Exposure time, ET (hr/day)	8	8

Cao et al., 2017; Liu et al., 2019; Howard et al., 2021)

The total incremental cancer risk (Total ILCR) was estimated with equation 5

$$\text{Total ILCR} = \text{ILCR1} + \text{ILCR2} + \text{ILCR3}, \dots, \text{ILCRn} \quad 5$$

In the given equation, the variable "n" represents the individual carcinogenicity of each PAH present in water or SPM. The acceptable level of cancer risk, also known as the Incremental Lifetime Cancer Risk

(ILCR), is typically deemed to be within the range of 10^{-4} to 10^{-6} for regulatory purposes (Liu *et al.*, 2019).

RESULTS AND DISCUSSION

Physical properties of suspended particulate matter and water: The physical properties of water and SPM are presented in Table 3, and the parameters presented are pH and conductivity for water samples and pH, conductivity and total organic carbon (TOC).

Table 3: Physical properties of water and suspended particulate matter

Sample Description	Water sample			Suspended particulate matter sample		
	pH	Conductivity ($\mu\text{S/cm}$)		pH	Conductivity ($\mu\text{S/cm}$)	TOC
Station 1	1	6.96	52.14	5.4	73.5	4.05
	2	6.40	53.12	5.3	75.8	4.75
	3	6.72	52.32	5.5	68.0	5.92
	4	5.76	41.12	5.5	81.0	4.30
Mean/SD	6.46±0.52	49.68±5.719		5.42±0.097	74.58±5.390	4.76±0.829
Station 2	5	6.63	58.19	5.0	87.6	6.09
	6	6.61	58.64	5.1	88.5	6.00
	7	6.65	57.54	5.3	86.70	6.34
Mean/SD	6.63±0.02	58.12±0.553		5.133±0.153	87.6±0.9	6.143±0.176
Station 3	8	6.02	50.12	5.5	82.45	3.97
	9	6.11	49.56	5.6	81.3	3.67
	10	6.10	47.41	5.6	79.4	3.24
	Mean/SD	6.077±0.049	49.03±1.430		5.567±0.057	81.05±1.540

The pH and conductivity values for SPM in the Imo River varied from 5.00 to 5.70 and 68.0 to 87.89 $\mu\text{S/cm}$, respectively. Water conductivity ranged from 41.12 to 60.15 $\mu\text{S/cm}$, with PH values ranging from 5.76 to 6.96. Oyem *et al.* (2014) discovered that the pH and conductivity values for the Niger Delta area were 5.43 and 5087.89 $\mu\text{S/cm}$, respectively.

Giann *et al.* (2013) found elevated pH and conductivity values in SPM in a river in Texas, United States. Zheng *et al.* (2013) reported on the pH and conductivity values of SPM observed in Taihu Lake sediment. The pH was 6.17, and the conductivity was measured at 98.29 $\mu\text{S/cm}$. The findings are consistent

with the work of Edori *et al.* (2019), who investigated the physicochemical parameters of surface water and sediment in the Silver River in Southern Ijaw, Bayelsa State, Nigeria's Niger Delta.

The TOC concentrations in sediment samples from several Imo River locations range from 3.35 to 6.34. Station 2 had the highest TOC concentration, whereas station 3 had the lowest. Previous studies found comparable levels of TOC in Niger Delta sediments (Sojину *et al.*, 2010). Adedosu *et al.* (2013) discovered 2.56 to 4.65 TOC levels in soil samples from affected Niger Delta sites. Ahrens *et al.* (2010) detected 4.16–

6.10 TOC in SPM and sediment samples from Tokyo Bay, Japan.

PAHs in SPM and Surface water from Imo River: The SPM of PAHs surface water from Imo River is presented in Table 4. Stations 1-3 had concentrations of 5.05 mg/L, 1.05 mg/L, and 2.05 mg/L of low-molecular-weight PAHs in the SPM, as shown in Table 4.

Station 1's SPM contains the greatest concentration of fluorene (2.29 mg/L) among the low-molecular-weight PAHs. Station 3's SPM content of 0.45 mg/L and Station 2's concentration of 1.86 mg/L indicate that naphthene contains the largest quantities of low-

molecular-weight PAHs, respectively (Bamuwamy *et al.*, 2015). At stations 1-3, the amounts of high-molecular-weight PAHs were found to be 17.64 mg/L, 11.82 mg/L, and 3.148 mg/L, consequently. Stations 1-3's surface water samples showed concentrations of 0.689 mg/L, 1.987 mg/L, and 0.796 mg/L of PAHs with low molecular weights, respectively.

A comparable analysis was performed on the surface water samples obtained from the stated sites to determine the concentrations of PAHs, or high-molecular-weight polycyclic aromatic hydrocarbons (Zheng *et al.*, 2013; Edori *et al.*, 2019).

Table 4: Concentrations of PAHs (mg/L) in surface water

Sample Description	SPM			Surface Water		
	Station 1	Station 2	Station 3	Station 1	Station 2	Station 3
Naphthalene	0.98	1.86	0.45	0.031	0.067	0.46
Acenaphthene	0.26	0.23	0.023	0.022	0.25	0.029
Acenaphthylene	0.62	0.26	0.13	0.031	0.082	0.043
Anthracene	0.46	0.84	0.023	0.302	1.12	0.078
Fluorene	2.29	1.12	0.24	0.012	0.45	0.17
Phenanthrene	0.44	0.74	0.37	0.291	0.018	0.016
Total LPAHs	5.05	5.05	1.236	0.689	1.987	0.796
Mean/SD	0.841667	0.841667	0.206	0.1148	0.331	0.132
Pyrene	2.34	2.04	1.02	0.063	0.15	0.97
Fluoranthene	0.48	0.66	0.10	0.027	0.47	0.04
Chrysene	1.80	5.37	0.70	0.622	0.46	1.54
Benzo[a]anthracene	0.37	0.13	0.013	0.01	1.27	0.20
Benzo[a]pyrene	1.76	0.38	0.16	0.680	0.56	0.23
Benzo[k]fluoranthrene	6.48	0.89	0.35	0.03	2.006	0.26
Dibenz[a,h]anthracene	1.77	1.12	0.28	0.14	0.150	0.011
Benzo [ghi] perylene	2.05	0.45	0.45	0.056	0.37	1.73
Indeno(1,2,3-cd)pyrene	0.59	0.78	0.075	0.012	0.005	0.88
Total HPAHs	17.64	11.82	3.148	1.64	5.441	5.861
Mean/SD	1.96	1.313	0.349	0.182	0.6045	0.6512
Total PAHs	22.69	16.87	4.384	2.329	7.428	6.657

The subsequent analyses revealed values of 1.64 mg/L, 5.441 mg/L, and 5.861 mg/L, correspondingly. Surface water PAH concentrations were found to be 2.329 mg/L, 7.428 mg/L, and 6.657 mg/L, according to the quantification results. The results shown in Table 3 clearly show that the levels of PAHs organic pollutants found in SPM were significantly higher than those found in the surface water at the specified sampling sites along the Imo River.

Guo *et al.* (2007) examined PAH concentrations in surface water and discovered a wide range, from 946.1 to 13448.5 ng/L. Similarly, values ranging from 317.5 to 238518.7 ng/g were reported in SPM with a dry weight.

The Chinese Daliao River basin was the site of these measurements. In the upper part of the Huaihe River in China, researchers Liu *et al.* (2016) found lower quantities of PAHs in surface water and SPM. Nevertheless, PAH concentrations were estimated by

the researchers and ranged from 79.94 to 421.07 ng/L for SPM and 10.87 to 201.42 ng/L for surface water. The present results are further supported by the research of Zhao *et al.* (2015). The Yellow River is located in Northwestern China, and throughout their analysis, the researchers found PAH concentrations in water ranging from 548 to 2598 ng/L and in SPMs from 1502 to 11,562 ng/g. The findings are in agreement with those of Chen *et al.* (2015), who studied the Weihe River in Northwest China and found SPM concentrations ranging from 3557 ng/L to 147,907 ng/L and water dissolved phase (WDP) concentrations from 351 to 4427 ng/L.

Source Apportionment of PAHs in Imo Rivers: Molecular diagnostic ratios of PAHs SPM and Surface water: The two main anthropogenic causes of PAH pollution are petrogenic and pyrogenic. The molecular diagnostic ratios of PAHs in SPM and surface water in Imo River are depicted in Table 5

Table 5: Molecular diagnostic ratios of PAHs in SPM and surface water

Molecular diagnostic ratios of PAHs SPM					
Stations	Fl/Py	Ph/An	Fl/(Fl+Py)	Ind/Ind+B[ghi]P	LPAHs/HPAHs
Station 1	0.205	0.957	0.17	0.22	0.286
Station 2	0.323	0.881	0.24	0.63	0.427
Station 3	0.098	16.087	0.10	0.14	0.393
Molecular diagnostic ratios PAHs surface water					
Station 1	0.429	0.725	0.3	0.174	0.420
Station 2	3.133	0.016	0.758	0.013	0.365
Station 3	0.041	0.205	0.04	0.337	0.136

The fluoranthene /anthracene ratio of this study was reported as 0.098 to 0.205 for SPM, whereas that of surface water was 0.429 and 0.041 (station 1 & 3) and 3.133 (station 2). A Fl/Py ratio more than one (Fl/Py>1) is indicative of a pyrolytic origin, whereas a ratio less than one (Fl/Py<1) is associated with a petrogenic source (Qiu *et al.*, 2011). The PAHs in both SPM and surface water were of petrogenic source for all the stations except for station 2 of surface water that was pyrogenic source. This is consistent with the research conducted by Qiu *et al.* (2011) and Anyakora *et al.* (2013), who identified PAHs in SPM water with petrogenic sources. The phe/ant ratios of PAHs in SPM were estimated as 0.957, 0.881 and 16.087, and that of surface water were 0.725, 0.016, and 0.205 for stations 1-3. Based on classification PAHs that phe /An ratio of less than 10 is pyrogenic sources, and Ph/An greater 10 is petrogenic source (Qiu *et al.*, 2011), all the identified PAHs in SPM and surface water in stations 1-3 are pyrogenic source, except station 3 for SPM that was identified with petrogenic origin. The finding of this research is supported by Zhao *et al.* (2015), who classified the isomeric ratio of phe/ant of PAHs in SPMs in the Yellow River, Northwestern China, to be of petrogenic origin, and Liu *et al.* (2016), with the report of pyrogen origin for a phe/ant molecular diagnostic ratio of 14 and above for suspended particular matter from the upper reach of the Huaihe River, China. The Fl/(Fl+Py) ratio of PAHs in SPM were reported as 0.17, 0.24, and 0.10 and that of the surface water were 0.30, 0.758 and 0.04 for stations 1-3 respectively. The Fl/(Fl+Py) ratio less than 0.4 has been associated with petrogenic sources (Anyakora *et al.*, 2013). Thus, based on the ratio, the PAHs in both SPM and surface water were associated with petrogenic origin. The current finding fits with what Leizou studied in 2021, which showed that polycyclic aromatic hydrocarbons (PAHs) found in urban soils in Yenagoa City, Bayelsa State, Nigeria, came from fossils. The diagnostic ratios of fluorine (Fl) to the sum of fluorine and pyrite (Fl+Py) were determined to be 0.2. Yogaswara *et al.* (2020) did a study where they looked at the Fl/(Fl+Py) ratio of polycyclic aromatic hydrocarbons (PAHs) in the brackish water and sediments of the Citarum Irrigation System in Pais Jaya, Karawang, Indonesia. The observed Fl/(Fl+Py) ratio was found to be 0.21. The

Ind/Ind+B[ghi]P ratio of PAHs in SPM were recorded as 0.22, 0.63, and 0.14 while that of surface water were 0.174, 0.013 and 0.337 for stations 1-3 respectively. According to the findings of Kim *et al.* (2013), an Ind/Ind+B[ghi]P ratio below 0.2 signifies the presence of petroleum sources. The ratios falling within the range of 0.2 to 0.5 indicate the contribution of petroleum combustion sources, while ratios over 0.5 suggest the involvement of combustion sources related to grass, wood, and coal. The PAHs in SPM at station 1 are associated with petroleum combustion sources; the PAHs in station 2 are associated with grass, wood, and coal combustion sources; and the PAHs in station 3 are attributed to petroleum sources (Sojinu *et al.*, 2010; Leizou, 2021). According to Yogaswara *et al.* (2020), a ratio more than 1 of LPAHs/HPAHs indicates a potential petroleum origin, but a ratio less than 1 suggests the involvement of pyrogenic processes. The investigation of PAHs in SPM and surface water in the Imo River, as illustrated in Table 3, is postulated to have arisen from pyrogenic pathways (Kafilzadeh *et al.*, 2011; Emoyan *et al.*, 2015).

Principal Component Analysis (PCA) of PAHs in SPM and Surface Water: Principal component analysis identified SPM PAH variable causes and sources. PCA source allocation of PAHs in SPM is shown in Table 6.

Table 6: Principal Component Analysis (PCA) of PAHs in SPM and Surface Water

PAHs	SPM		Surface Water	
	Component		Component	
	1	2	1	2
Naphthalene	.408	.794	-.821	.570
Acenaphthene	.991	.134	.856	.317
Acenaphthylene	.859	-.511	.736	.676
Anthracene	.742	.670	.952	.305
Fluorene	.938	-.346	.437	.771
Phenanthrene	.436	.900	-.001	-1.00
Pyrene	.999	.033	-.815	.580
Fluoranthene	.841	.541	.857	.516
Chrysene	.479	.878	-.924	.382
Benzo[a]anthracene	.892	-.452	.792	.610
Benzo[a]pyrene	.785	-.420	.703	-.711
Benzo[k]fluoranthrene	.754	-.357	.812	.583
Dibenz[a,h]anthracene	.980	-.197	.893	-.450
Benzo [ghi] perylene	.700	-.514	-.759	.651
Indeno(1,2,3-cd)pyrene	.870	.492	-.866	.401
% of Variance	65.568	34.432	62.800	37.2

Two primary components were retrieved, where the first component accounted for 65.568% and the second principle component accounted for 34.432%. In the first part, high levels of polycyclic aromatic hydrocarbons (PAHs) like Ace, Acn, Ant, Flu, Pyr, Flo, BaA, BaP, BkF, DahA, BghiP, and IdP indicate the presence of petrogenic sources like crude oil, oil spills, and seepages. Conversely, in the second component, Naph, Phe, and Chry exhibited the highest loading, indicating the influence of pyrogenic sources such as the combustion of grass, wood, coal, and gas flaring. Petrol emissions are shown by Ace, Flu, Pyr, and DbA compounds. On the other hand, diesel emissions are marked by BbF, BkF, and Phe compounds, which are the main polycyclic aromatic hydrocarbon (PAH) species (Marr *et al.*, 1999; Mabilia *et al.*, 2004). Two main constituents were obtained for the surface water, wherein constituent 1 constituted 62.800% and constituent 2 constituted 37.200%. The studies conducted by Leizou (2021) and Emoyan *et al.* (2015) reported the highest values for Ace, Ant, Pyr, Flu, Chr, BaA, BaP, BkF, Da, Ha, BghiP, and IcdP. These findings suggest a common petrogenic source for these compounds. Additionally, Nap, Flu, and Phe were found to indicate pyrogenic sources.

Hu *et al.* (2019) detected 16 PAHs in water and SPM samples from the main rivers in northeastern China's Liao River basin. Diagnostic ratios and PCA showed that petroleum and fossil fuel burning may be the source of these polycyclic aromatic hydrocarbons

(PAHs). Liu *et al.* (2016) examined Huaihe River surface water and SPM. The investigation found pyrogenic and petroleum-derived PAH pollution in the river. These findings match Imo River research. Wang *et al.* (2018) found that vehicle emissions generated 46.3% of polycyclic aromatic hydrocarbons (PAHs) in Yangtze River suspended particulate matter (SPM) samples from China. Researchers found that 40.4% of the water samples' polycyclic aromatic hydrocarbons (PAHs) came from wood and coal burning. However, 13.3% of PAHs came from petrogenic sources. Leizou (2021)'s study demonstrated that PAHs in urban soils in Yenagoa City, Bayelsa State, Nigeria, come from rock sources. The diagnostic ratios of fluorine (Fl) to fluorine and pyrite (Fl+Py) were 0.2. Yogaswara *et al.* (2020) examined the Fl/(Fl+Py) ratio of PAHs in brackish water and sediments of the Citarum Irrigation System in Pais Jaya, Karawang, Indonesia. Both water and sediment samples had Fl/(Fl+Py) ratios of 0.21.

Human Health Risk Assessment

Potential toxicity based on carcinogenic PAHs: Benzo (a)pyrene (BaP) is a PAH compound that is often used as a reference for assessing the toxicity and mutagenicity of other PAHs. BaP is a known carcinogen and is frequently used to estimate the potential carcinogenic and mutagenic effects of mixtures of PAHs (Türk-Çulha *et al.*, 2020; Aigberua and Seiyaboh, 2021). The toxicity of benzo (a)pyrene related to PAHs in SPM and water are presented in Table 7.

Table 7: Benzo (a)pyrene toxic equivalency quantities (BaPTEQ and mutagenic equivalency quantities (BaPmeq) levels

PAHs	Benzo(a)pyrene toxic equivalency quantities (BaPTEQ)			Water		
	Station 1	Station 2	Station 3	Station 1	Station 2	Station 3
Bap	1.76	0.38	0.16	0.680	1.56	0.23
BaA	0.037	0.013	0.0013	0.001	0.127	0.02
Bkf	0.0648	0.0089	0.0035	0.0003	0.02006	0.0026
Chry	0.0018	0.00537	0.0007	0.000622	0.00046	0.00154
DahA	1.77	1.12	0.28	0.14	0.150	0.011
Indp	0.059	0.078	0.0075	0.0012	0.0005	0.088
∑PAHs	3.6926	1.605	0.453	0.823	1.858	0.353
Mean	0.615	0.268	0.0755	0.137	0.309	0.059
St. dev.	0.891	0.443	0.118	0.272	0.616	0.089
Benzo(a)pyrene Mutagenic Equivalency Quantities (BaPmeq) Levels						
Bap	1.76	0.38	0.16	0.680	0.56	0.23
BaA	0.03034	0.01066	0.001066	0.00082	0.10414	0.0164
Bkf	0.7128	0.0979	0.0385	0.0033	0.22066	0.0286
Chry	0.0306	0.09129	0.0119	0.010574	0.00782	0.02618
DahA	0.5133	0.3248	0.0812	0.0406	0.0435	0.00319
Indp	0.1829	0.2418	0.02325	0.00372	0.00155	0.2728
∑PAHs	3.229	1.146	0.316	0.739	0.938	0.577
Mean	0.538	0.191	0.053	0.123	0.156	0.096
St. dev.	0.658	0.146	0.059	0.273	0.214	0.121

The toxic equivalent factor (TEF) of all six PAHs and benzo(a)pyrene (BaP) found in the Imo River's SPM was found to be 3.693. Additionally, the average TEF

for stations 1-3 were found to be 0.615, 1.605, and 0.453. The toxicity associated with the concentration of benzo(a)pyrene in station 1 was found to be greater

compared to stations 2 and 3. This observation suggests that station 1 exhibits a higher level of toxicity due to the presence of BaP-related PAHs in SPM (Jung *et al.*, 2010). The concentration of the hazardous equivalent factor of total BaP for the 6PAHs in water was found to be significantly high at station 2 (1.858), while stations 1 and 3 have 0.823 and 0.35. The present discovery is compatible with Dong *et al.* (2021), who found a high toxicity equivalent factor of total benzo(a)pyrene in Taihu Lake's bay and river in China. Studies by Iwegbue and Obi (2016) found elevated toxicity in urban dust samples from the Nigerian Niger Delta. Reizer *et al.* (2019) report that the WHO recommends a drinking water guideline of 0.7 µg/L. This shows that the toxicity equivalent factor of total benzo(a)pyrene recorded in Imo River stations was below the WHO standard. At station 1, the total and mean mutagenic equivalency quantities of benzo(a)pyrene levels for 6 Σ PAHs in SPM were recorded as 3.229 and 0.538, respectively. The expected values for station 2 and station 3 were 1.146 (0.191) and 0.316 (0.053), respectively. The concentrations of benzo(a)pyrene mutagenic

equivalency quantities were observed to be elevated for PAHs present in SPM at the three designated sites; however, they were comparatively lower for PAHs in water. In a previous investigation conducted by Olatunji *et al.* (2014), the recorded mutagenic equivalency levels of benzo(a)pyrene were found to exceed the estimated value for the present study. In their study, Wang *et al.* (2018) examined the mutagenic equivalency quantities of benzo[a]pyrene (BaP) in various layers of soil throughout petroleum-contaminated regions of the Loess Plateau, China. The mean values obtained for BaP concentrations were 8.18 ± 4.05 mg/kg and 6.94 ± 4.36 mg/kg. In a comparable investigation, Ofori *et al.* (2021) documented the BaPMEQ values of 3×10^{-5} mg/kg and 5.2×10^{-4} mg/kg for roadside dust in Nigeria.

Incremental lifetime cancer risk: Carcinogenic PAHs ingested are assessed using incremental lifetime cancer risk (Howard *et al.*, 2021; Ambade, 2021). Table 8 shows estimations for children and adults' PAH exposure means.

Table 8: Incremental lifetime cancer risk of children and adult for PAHs in SPM Imo Rivers

	Children			Adult		
	SPM			SPM		
PAHs	Station 1	Station 2	Station 3	Station 1	Station 2	Station 3
Bkf	1.56E-7	2.14E-8	8.43E-9	6.4E-7	8.79E-8	3.46E-8
Bap	4.24E-6	9.15E-7	3.85E-7	1.74E-4	3.75E-6	1.58E-6
BaA	8.91E-8	3.13E-8	3.13E-9	3.65E-7	1.28E-7	1.28E-8
Chry	4.33E-9	1.29E-8	1.69E-9	1.78E-8	5.3E-8	6.91E-9
DahA	4.26E-6	2.70E-6	6.74E-7	1.75E-5	1.11E-5	2.77E-6
Indp	1.4E-7	1.88E-7	1.81E-8	5.83E-7	7.7E-7	7.41E-8
Σ PAHs	1.03E-05	4.06E-06	7.18E-06	1.93E-04	1.59E-05	4.48E-06
Mean	1.72E-06	6.77E-07	1.20E-06	3.22E-05	2.65E-06	7.46E-07
St. dev.	2.045E-06	1.05E-06	2.72E-06	6.98E-05	4.38E-06	1.17E-06
	Surface water			Surface water		
Bkf	7.22E-10	4.83E-8	6.02E-9	2.96E-9	1.98E-7	2.57E-8
Bap	1.64E-6	1.35E-6	5.54E-7	6.72E-6	5.53E-6	2.27E-6
BaA	2.41E-9	3.06E-7	4.82E-8	9.88E-9	1.25E-9	1.98E-7
Chry	1.50E-9	1.11E-9	3.71E-9	6.14E-9	4.54E-9	1.52E-8
DahA	3.37E-7	3.61E-7	2.65E-8	1.38E-6	1.48E-6	1.09E-7
Indp	2.89E-9	1.20E-9	2.12E-7	1.19E-8	4.94E-9	8.69E-7
Σ PAHs	1.98E-06	2.07E-06	8.50E-07	8.13E-06	7.22E-06	3.49E-06
Mean	3.31E-07	3.45E-07	1.42E-07	1.36E-06	1.20E-06	5.81E-07
St. dev.	6.55E-07	5.17E-07	2.17E-07	2.68E-06	2.20E-06	8.87E-07

The mean cancer risks for PAHs in SPM ingested by children were estimated at 1.72E-06, 6.77E-07, and 1.20E-06, while those of adults were recorded at 3.22E-05, 2.65E-06, and 7.46E-07, respectively, for stations 1-3. The cancer risk of ingested surface water with PAHs for children were 3.31E-07, 3.45E-07 and 1.42E-07, and that of the adult was 1.36E-06, 1.20E-06, and 5.81E-07 respectively for stations 1-3. The stations 1 and 2, the effect of carcinogenic PAHs in SPM was higher in adults than the children, whereas, for station 3, the effect of carcinogenic PAHs was high for children than the adults. This is on the basis that

the summation of all the PAHs in SPM from different stations (1-3) for adults was recorded as 1.93E-04, 1.59E-05, and 4.48E-06, while that of the children was estimated as 1.03E-05, 4.06E-06 and 7.18E-06 for the respective stations. The incidence of cancer in adults who consumed surface water was found to be greater than that observed in children at stations 1-3. The above discovery fits with the results of a study by Howard *et al.* (2021), which found that polycyclic aromatic hydrocarbons (PAHs) found in suspended particulate matter (SPM) were linked to a significant increase in the lifetime risk of cancer in adults living

in the Niger Delta region of Nigeria. Hu et al. (2016) did a study that found the cumulative lifetime cancer risk (ILCR) was significantly higher for people who were exposed to suspended particulate matter (SPM) and surface water in the major rivers of the Liao River drainage basin in northeast China. Emmanuel *et al.* (2022) provided more support for the research by doing an investigation on the evaluation of human health risks related to polycyclic aromatic hydrocarbons (PAHs) in three senatorial districts of Anambra State, Nigeria. Asagbra *et al.* (2015) provided regulatory guidelines indicating that values of ILCR $< 10^{-6}$ indicate the absence of risk or low risk, but values $\geq 10^{-4}$ indicate a significant level of risk. Adverse health impacts are observed when the cumulative ILCR value for each category exceeds a threshold of 10^{-4} . Nevertheless, it is worth noting that all of the averages lie within the range of $\leq 10^{-6}$ and ILCR $< 10^{-4}$. Consequently, it can be inferred that the risk value remains below the established baseline thresholds. Hence, it is plausible that the residents of the region may face a prospective hazard of developing cancer in the forthcoming period (Duan *et al.*, 2016; Howard *et al.*, 2021). The incremental lifetime cancer risk (ILCR) for stations 1–3 pertaining to children was found to be below the regulatory threshold of 10^{-4} , indicating that it falls within the acceptable range of the ILCR for polycyclic aromatic hydrocarbons (PAHs). The cumulative incremental lifetime cancer risk (ILCR) for station 1 among adults was found to be in line with the regulatory guidelines. Consequently, the consumption of water from the Imo River at this station would present a heightened health hazard to people. Stations 1 and 2 remained positioned at the safety line due to their individual lifetime cancer risk (ILCR) exceeding the threshold of 10^{-4} .

Conclusions: The source of PAHs in SPM and surface water in Imo River were associated with petroleum, grass, wood, coal and combustion. The PCA of SPM was recorded at 65.568% for component 1 and 34.432% for principal component 2. The PCA of surface water recorded 62.80% for principal components and 37.2% for principal component 2. There is a possibility that people within the sampled stations will have the risk of cancer, based on toxic equivalent factor and the mutagenic equivalency quantities of total benzo(a)pyrene. Also, the effect of carcinogenic PAHs in SPM was higher in adults than in children, with regards to ILCR analysis.

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