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Risk Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Sediments and Freshwater Samples of Cross River Channel (Etono Central Beach), Biase, Nigeria

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The purpose of this study was to assess the levels of polycyclic aromatic hydrocarbons (PAHs) in sediments and freshwater samples of Cross River Central Channel (Etono Central Beach) in Biase, Nigeria and the associated risk. Methodologically, five samples each of sediment and water were collected at five different locations of the Etono Central Beach in dry and wet season and analyzed for PAHs and physico-chemical parameters as applicable using laboratory apparatus and GC-FID. The results of the findings revealed that the mean values of PAHs in water ranged from (0.2017-2.8409) mg/L and (0.0172-5.8899) mg/L for dry and wet season, respectively. In sediments, they ranged from (0.1019-4.6825) mg/L and (0.0018-0.1435) mg/L for dry and wet season, respectively. For cancer risk assessment (CRA) associated with the PAHs, the values ranged from (9.3210×10^{-3} – 2.4154×10^{-1}), (7.7601×10^{-3} – 4.5458×10^{-1}) and (8.8801×10^{-3} – 2.3027×10^{-1}) for 10, 25 and 70 years, respectively in dry season. In wet season, (1.1011×10^{-3} – 1.4365), (2.5280×10^{-3} – 1.1971) and (1.1011×10^{-3} – 1.3694) for 10, 25 and 70 years, respectively. The implication is that the sediments contained more PAHs than water and were greater in the dry season than the wet season. The cancer risk assessment (CRA) revealed that exposure to certain polycyclic aromatic hydrocarbons (PAHs) posed a cancer risk to individuals within the 70-year age bracket that exceeded acceptable limits. This risk was higher during the dry season compared to the wet season, with benzo[*k*]fluoranthene and chrysene identified as the most concerning compounds.

Keywords: Cancer risk assessment, Sediment contamination, Cross River, Etono Central Beach, Environmental toxicology.

INTRODUCTION

The presence of pollutants (toxic substances) like organochlorine pesticides, polychlorinated biphenyl (PCBs) and polycyclic aromatic hydrocarbons in the water environments has been of great interest due to their negative impact on the health of aquatic life and man who ends up utilizing these aquatic resources for food and other purposes. Prolonged human exposure to these toxic substances has been linked to several adverse health effects, including low birth weight, immune system disorders, neurological problems, developmental delays in children and an increased risk of cancer [1].

The PAHs containing five- or six-membered rings are the most common and those composed exclusively of six-membered rings are referred to as alternate PAHs, consisting of benzenoid structures [2]. While smaller PAHs are more water soluble and represent prominent aquatic contaminants, larger PAHs are generally insoluble in water and only weakly soluble in organic solvents and lipids. They are persistent environmental pollutants associated with serious health effects, including carcinogenicity and developmental toxicity [3]. Thousands of PAHs are present in the environment but practically, PAHs assessment is limited to a few, most of which are the 16 priority PAHs (acenaphthene, acenaphthylene, phenanthrene, anthracene, naphthalene, fluoranthene, benzo[*a*]anthracene, fluorene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, indeno[1,2,3-*c*,*d*]-pyrene, chrysene, dibenzo[*a*,*h*]anthracene, benzo[*a*]pyrene, benzo[*g*,*h*,*i*]perylene and pyrene) labelled by US EPA as naturally toxic [4]. Humans, wildlife and fish will not come into contact with a single PAH, but a combination of PAHs. Studies in animals have also revealed that PAHs can inflict harmful on the body fluids, skin and the body's mechanism for fighting

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diseases after long- and short-term exposure. Molecular and epidemiological studies have shown that fetuses and babies are more prone to the harmful effects of a variety of environmental pollutants than adults, including PAHs [5].

PAHs originate from natural, petrogenic, pyrolytic and anthropogenic sources. Pyrolytic PAHs formed during incomplete combustion of organic matter [6], while natural sources include oil seeps and plant pigments [7]. However, these natural inputs contribute minimally to environmental PAHs levels [8]. Major anthropogenic sources include vehicle emissions, industrial processes, coal tar and asphalt production, smoking, cooking and waste incineration [9]. Petrogenic as well as pyrogenic PAHs are highly abundant and are normally generated by the reduction of biogenic precursors [10,11]. PAHs enters into the environment through the deliberate or accidental liberation of petroleum or its refined products from places that utilizes the products, underground storage pipes leakage, destruction of pipelines and oil spillages [12]. PAHs from pyrolytic sources are generated through chemical transformations induced by heat. This process involves the thermal decomposition of organic substances, leading to the formation of new compounds, including PAHs [13].

Exposure to PAHs has been linked to various adverse health effects in humans, including cancer, skin diseases, emphysema, and disorders of the nervous, reproductive and endocrine systems [14,15]. Laboratory studies in animals have confirmed PAHs' toxic and carcinogenic potential, with benzo[*a*]pyrene notably identified as a potent carcinogen [3], [16]. Cancer risk assessments (CRA) of PAHs in water samples have shown varying risk levels across different age groups, with some compounds like benzo[*b*]fluoranthene reaching levels of public health concern [17]. Non-carcinogenic effects are evaluated using hazard quotients (HQ), while carcinogenic risks (CR) estimate lifetime cancer probability from PAHs exposure [18,19].

Given the study area's dependence on the Cross-River channel for water and livelihood, long-term exposure to PAHs is a significant public health concern. This study aims to quantify PAHs concentrations in sediments and water, assess the associated carcinogenic risks, and provide recommendations to mitigate potential health hazards. The findings will serve as baseline data for environmental monitoring and public health interventions in the region.

EXPERIMENTAL

The scientific instruments like conductivity meter (4510 Conductivity meter JENWAY, Bibby Sci. Ltd., Dunmow, England.), pH meter (pH meter SD20, Mettler-Toledo, India), oven (model D-91126, Memmert GmbH+ Co., Germany), Rotatory evaporator (model Stuart RE300B, Barloworld Sci. Ltd., UK), balance (model PL303, Mettler Toledo, India) and Agilent 7890B gas chromatograph equipped with a flame ionization detector (FID) were used in this study. Reagents used include distilled water, *n*-hexane and acetone, all of which were of analytical grade and made by the British Drug House, Poole, England.

Description of study/sampling area: Etono Central is located in Biase Local Government area of Cross River State, Nigeria. The Cross River is bounded by two tropical moist forest Eco-regions: the Cross-Niger transition forests, lying west of the river between the Cross and Niger Rivers and the Cross-Sanga-Bioko Coastal Forests, lying to the east between the Cross River and the Cameroonian Sanga River. A greater percentage of the land in the community is submerged in water, with fishing and farming the primary source of livelihood of the inhabitants. Its geographical coordinates are 5º38′0″ North, 8º1′0″ East (Fig. 1). The Etono River/beach is the major water source for the community.

The sample locations are the main beach where the villagers fetched their domestic and drinking water from, it is also used as berthing port canoes and boats especially during the raining seasons. Other activities carried out here include swimming, laundering, fishing and other activities.

Sample collection/procedure: Twenty samples (ten water samples and ten of sediment) were collected randomly from the study area within the two seasons of the year; dry season (December) and rainy season (July). Five water samples and five samples of sediment were collected at random locations about 60 m apart for each season, making a total of 20 samples that were taken from the study area. The sampling points were carefully chosen from the points where the villagers carry out main activities in the river. Some of these activities include boating/transportation, fishing, swimming/bathing, sand mining and other domestic activities. A dirt-free and repeatedly rinsed transparent 1 L capacity bottles were used for sampling at the surface level of the water [20]. The freshwater samples were stored in an ice-packed cooler and were transferred to the lab for analysis. Intertidal sediment samples were collected with a hand trowel into polyethylene bags.

Physico-chemical properties

Electrical conductivity: The electrical conductivity (EC) of the water samples was measured using an electrical conductivity meter. The five samples at room temperature were collected into clean and sterilized beakers. Each beaker was placed on a flat, stable surface. The measuring lead EC meter was inserted into each sample after it was turned on and the displayed measurements in µS (micro-Siemens) by the meter were recorded. The measuring lead was rinsed with distilled water after each measurement to ensure the accuracy of results.

Determination of pH: The water sample's pH was measured with a pH meter. The pH probe was calibrated with distilled water before/after each analysis. The pH probe was inserted into each water sample and allowed for the reading to stabilize and the value recorded.

Total suspended solids (TSS): Each shaken water sample was filtered through a pre-weighed filter paper and 50 mL of the filtrate was collected into a clean beaker. After filtering, the paper was dried in an oven at 60 ºC, then, it was put in a desiccator for 2 h. The TSS was calculated using eqn. 1 thus:

$$\text{TSS (ppm)} = \frac{W_2 - W_1 \times 10^3}{V} \tag{1}$$

where W_1 = initial weight of filter paper; W_2 = weight of the filter paper and residue; V = filtered sample's volume.

Dissolved oxygen (DO): The dissolved oxygen for each water samples were measured with a dissolved oxygen meter.

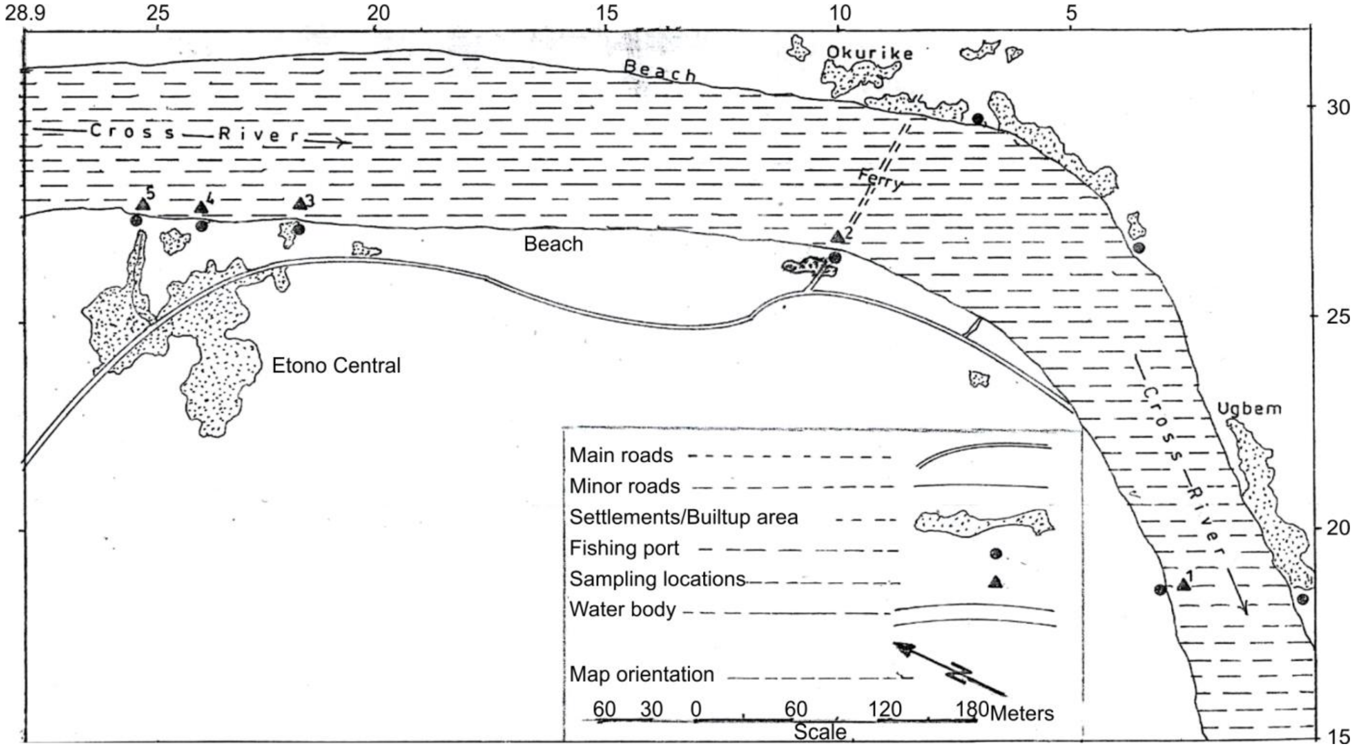

Fig. 1. Map of Etono Central Beach and environs, showing sampling locations

The five samples at room temperature were collected into clean and sterilized beakers. Each beaker was placed on a flat, stable surface. After transferring on the dissolved oxygen (DO) meter, the probe was inserted into each sample, and the displayed readings (in mg/L) were recorded. To ensure the accuracy, the probe was rinsed with distilled water between measurements.

Determination of organic matter in sediments: A clean, dry and empty porcelain crucible was weighed. Then, portion of the dried test specimen previously used for the moisture content determination was placed into the crucible, and the combined mass of the crucible and specimen was recorded. The crucible was placed in a muffle furnace, where the temperature was gradually increased to 700 ºC and maintained for 40 min. After ashing, the crucible was allowed to cool to room temperature (approximately 25 ºC) before final weighing. The dish with the ash (burnt soil) was weighed and recorded thus:

$$\text{Organic matter (\%)} = \frac{\begin{array}{c}\text{Ignited weight (weight of sample +}\\ \text{crucible)} - \text{Weight of empty crucible}\end{array}}{\text{Weight of sample taken}} \times 100 \quad (2)$$

Determination of polycyclic aromatic hydrocarbons (PAHs): The reagents and equipment used in this study included *n*-hexane and acetone as solvents, a rotary evaporator for solvent removal and a gas chromatography system equipped with a flame ionization detector (GC-FID) for compound analysis.

Extraction of samples using ultrasonic bath: Each soil sample was finely pulverized using a mortar and pestle and precisely 5 g was weighed for analysis. The samples were put in a beaker and 10 mL acetone:hexane (1:1) was added. The samples were sonicated using an ultrasonic bath for 20 min. The mixture was allowed to settle, after which the solvent layer was decanted and concentrated to 2 mL using a rotary evaporator.

Gas chromatography analysis: Sample analysis was conducted with an Agilent 7890B Gas Chromatograph equipped with a flame ionization detector (FID) and a DB-1 capillary column (30 m × 0.32 mm i.d., 0.25 μm film thickness) coated with 5% phenyl methyl siloxane. Each sample (1 mL) was injected in splitless mode at 220 ºC and 14.861 psi, with a total flow rate of 21.364 mL/min and a purge flow to the split vent of 15 mL/min after 0.75 min. The oven temperature was initially set at 100 ºC for 2 min, operated at 10 ºC min^{-1} to 280 ºC for 4 min, and then further operated at 10 ºC min^{-1} to 300 ºC. The FID was maintained at 300 ºC, with hydrogen and air flow rates of 30 mL min^{-1} and 300 mL min^{-1}, respectively, and nitrogen as make-up gas at 18 mL min^{-1}. Concentrations were calculated after calibration and chromatograms were analyzed.

Data analysis: The data were analyzed and presented using descriptive statistics (mean and standard deviation) in MS-Excel application to ensure accuracy and precision). SPSS version 20 software was used to carry out the significant test on the mean of PAHs in the samples for both seasons using paired t-test and one-way ANOVA at $p < 0.05$.

Human health risk assessment: In this study, the human health risk assessment focused on the exposure of inhabitants and users of the Etono River beach to carcinogenic PAHs. The assessment employed the following indices:

Chronic daily intake (CDI) index: The CDI of pollutants in river water is assessed from variuos exposure scenarios like ingestion, dermal contact and inhalation, but in this study, the CDI was estimated using the ingestion pathway.

Oral exposure (ingestion): The following equation was used in estimating the CDI through Oral (Ingestion) exposure as proposed by USEPA [21]:

$$\text{CDIing} = \frac{\text{CW} \times \text{IR} \times \text{EF} \times \text{ED} \times \text{CF1}}{\text{BW} \times \text{AT}} \qquad (3)$$

where CDIing = chronic daily intake through ingestion (mg/kg day), CW = pollutant concentration in drinking water (μg/L), IR = rate of drinking water ingestion (L/day), EF = frequency of exposure (days/year), ED = duration of exposure (year), BW = body weight (kg), AT = average time (days) and CF1 = conversion factor of mass from μg to mg (0.001) [21].

Parameters for cancer risk assessment of PAHs: The following parameters were calculated using the data derived from local residence with reference values of USEPA and as described by Emrah [19].

IR = rate of drinking water ingestion (L/day), EF = frequency of exposure (days/year), ED = duration of exposure (year), BW = body weight (kg), AT = average time (days).

Carcinogenic risk (CR) index: The CR index was estimated using the equation below:

$$\text{CR} = \text{CDIing} \times \text{SF} \qquad (4)$$

where: CDIing = chronic daily intake by ingestion (mg/kg day), SF = Slope factor of the pollutant $(\text{mg/kg/day})^{-1}$. SF is the oral slope factor in mg of toxin/kg of body weight daily for a lifetime [21].

Cancer slope factor: Cancer slope factor (CSF) was used to assess the risk of cancer related to a potentially carcinogenic substance or carcinogen exposure. A slope factor is an upper limit 95% confidence limit approximation, of the increased risk of cancer from a lifetime exposure to an agent *via* inhalation or ingestion. This estimate is normally expressed in units of affected population per mg of the substance per kg body weight, usually reserved for the low-dose region of the dose-response relationship or exposure-response relationship, which corresponds to risks less than 1 in 100 exposures [21].

Toxicologically available data are enough to serve the purpose of deriving a guideline value for benz[*a*]pyrene only, one of the highly potent carcinogenic PAHs tested to date. IARC have categorized benz[*a*]pyrene as a probable carcinogen to humans. The derivation of the guideline value is based therefore on the cancer risk for a lifetime assessed *via* extrapolation of the tumor incidence data studied in the most appropriate carcinogen bioassay in animals. According to the Integrated Risk Information System (IRIS) of US EPA, the ingestion slope factor ranges from 4.5 to 11.7 per (mg/kg/day), with a geometric average of 7.3 per (mg/kg/day) [1].

Relative potency factor: Guideline values of slope factor for all PAHs have not been given except for benzo[*a*]pyrene due to insufficient data to set guidelines. But, the use of relative potency can be used to offer guidance to the relative contributions of PAHs to the risk associated their presence [17].

The World Health Organization (WHO) has determined the relative carcinogenic potencies of several PAHs, as shown in Table-1. PAHs with a relative carcinogenic potency below 0.01 (*i.e.* 100 times less carcinogenic than benzo[*a*]pyrene) are generally excluded from risk assessments due to their minimal risk in water. By multiplying the relative potency factor of each PAH by the cancer slope factor of benzo[*a*]pyrene, an estimated cancer slope factor for each compound can be derived [17,21].

TABLE-1
HUMAN HEALTH TOXICOLOGICAL
DATA FOR CARCINOGENIC PAHs

PAHs	Relative potency factor	Cancer slope factor (mg/kg/day)
Benz[*a*]anthracene	0.1	7.3×10^{-1}
Benzo[*a*]pyrene	1	7.3
Benzo[*k*]fluoranthene	0.1	7.3×10^{-1}
Chrysene	0.01	7.3×10^{-2}
Dibenz[*a*,*h*]anthracene	1	7.3
Fluoranthene	0.1	7.3×10^{-1}
Indeno[1,2,3-*c*,*d*]pyrene	0.1	7.3×10^{-1}

Source: [4,17,21]

RESULTS AND DISCUSSION

The samples were obtained from 5 different points of the Cross river channel (Etono Central Beach) where the inhabitants carry out most of their activities. The sampling points were carefully chosen to be the main activities points of the villagers with the river body. Some of the activities include boating, fishing, swimming/bathing, farming, sand mining and domestic activities.

Physico-chemical parameters in water and sediment: The physico-chemical parameters measured include pH, DO, conductivity, TSS and organic matter (Table-2). The physico-chemical parameters, except for organic matter, were analyzed in the water samples. This exclusion was due to the fact that the water samples were collected directly above the exact location of the sediment sampling points.

The pH for wet season ranges from 6.00-6.20 with a mean of 6.05. The lowest pH value 6.00 occurred at location 4 while the highest pH value 6.20 occurred at location 3. In dry season, the pH ranged from 6.04-6.37 with a mean of 6.23, therefore showing a slight difference between the two seasons. In wet season, conductivity (μS/m) ranged from 0.08 to 0.09, with a mean value of 0.0086 (mg/L). The 0.09 value occurred at locations 1, 4 and 5 while the 0.08 value occurred at locations 2 and 3. The conductivity in the dry season, varied more from location to location, having its maximum value of 0.11 at location 4 and the lowest value of 0.02 at location 1. The lowest concentration of TSS (g) in the wet season was 1.05 and the highest concentration was 1.42 occurring at location 3 and 1, respectively, with a mean value of 1.194. Generally, the TSS was higher in the wet season than dry and this can be attributed to the influx of materials *via* run-off from land sources. The concentration of dissolved oxygen during the wet season ranged from 2.88 to 3.44, with a mean value of 3.01. The highest concentration was 3.01 occurred at location 5 while the lowest concentration 2.88 occurred at location 1, while during dry season it ranged from 2.59-2.90, with the highest occuring at location 4 and the lowest at location 3.

Organic matter, the only parameter analyzed using sediment samples, recorded its highest concentration at location 2 (9.078) and the lowest at location 4 (4.311). All the measured physico-chemical parameters in the study remained within the permissible limits established by the World Health Organization (WHO).

TABLE-2
PHYSICO-CHEMICAL PARAMETERS FOR WATER SAMPLES IN WET AND DRY SEASON

Locations	Major activities	pH		Conductivity (µS/m)		TSS (g)		DO (mg/L)		OM (g)	
		Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
1	Boating/transportation	6.04	6.01	0.02	0.09	0.61	1.42	2.79	2.88	4.291	4.667
2	Boating	6.37	6.03	0.10	0.08	0.56	1.12	2.88	2.93	0.708	9.078
3	Settlement area washing, fishing and other domestic	6.34	6.20	0.08	0.08	1.01	1.05	2.59	2.77	8.022	4.666
4	Farming swimming, washing, *etc.*	6.30	6.00	0.11	0.09	1.16	1.13	2.97	3.44	4.232	4.311
5	Farming, sand mining	6.10	6.05	0.09	0.09	1.01	1.25	2.89	3.01	3.959	4.653
	Total	31.15	30.29	0.40	0.43	4.35	5.97	14.12	15.03	28.21	27.38
	Mean	6.23	6.06	0.080	0.07	0.87	1.19	2.82	3.01	5.64	5.48

Levels of PAHs in sediment and water samples

Water samples: The mean values of PAHs in water samples during the dry season for the study locations (Table-3) ranged from 0.2017 mg/L to 2.8409 mg/L. The highest mean value occurred at location 1 and the lowest at location 4. The range for individual PAHs for the different locations are ND-11.0294; ND-6.3880, ND-2.8796, ND-6.3880 for locations 2, 3, 4 and 5, respectively. PAHs concentrations for location 1 were below the detection limit. Location 2 has the highest concentration value of PAHs for water samples in the dry season with a sum of 43.6525 mg/L and the lowest concentration was found in location 4 with a value of 0.3477 mg/L. Naphthalene, indeno[1,2,3-*c*,*d*]pyrene, dibenzo[*a*,*h*]anthracene, benzo[*g*,*h*,*i*]-perylene, benzo[*g*]fluoranthene, benzo[*a*]pyrene, were below the detection limits in water samples across all locations. The mean level for individual PAHs ranged from 0.6718 mg/L for fluorene-2.85196 mg/L for benzo[*k*]fluoranthene. The low value of fluorene amongst other PAHs was also reported in water samples from Egi, Omoku and Egbema communities in River States as reported by Inam *et al*. [17]. Benzo[*k*]fluoranthene was also the highest occurring individual PAHs across all the locations with a 7.8718 mg/L value in at location 3. Acenaphthene had the lowest value amongst all 16 PAHs, with a value of 0.34772 at location 4. It was observed that individual PAH levels within the study locations were relatively low for three-ringed *viz.* acenaphthylene, acenaphthene, fluorene and higher than four and five-ringed fluoranthene, chrysene and benzo[*k*]fluoranthene, respectively. No six-ringed PAHs were detected for the water sample in the dry season.

During the wet season, individual PAHs concentration drastically reduced as compared to the dry season. The influx of freshwater from, rain, runoff and surrounded streams are suspected for this sharp decrease in the value of PAHs concentration in water samples of the wet season. This is similar to the work reported in a study within Warri and its surrounding areas [22]. The highest mean of PAHs concentration occurred in location 4 with a value of 5.8899 and the lowest occurring at location 1 with a mean value of 0.01721 (Table-4). None of the sixteen PAHs under study were detected at location 3. The highest occurring individual PAHs were pyrene with an individual mean value of 7.0063. Its highest value occurred at location 4 with a relatively high value of 83.5703 mg/L.

Sediment samples: In the dry season, the mean value for sediment samples across locations (Table-3) ranged from 0.1109 mg/kg in location 5 to 4.6825 mg/kg in location 1. The individual mean value ranged from 0.1294 mg/kg for acenapthylene to 13.6858 mg/kg for chrysene. Individually, the highest occurring PAH was chrysene at location 1 with a value of 60.848 mg/kg. During the wet season, the mean value for sediment samples across locations (Table-4) ranged from 0.0037 mg/kg in location 1 to 0.0291 mg/kg at location 4. Meanwhile, the mean value for location 2 was not detected. The individual PAHs mean value ranged from 0.0271 mg/kg for phenanthrene to 0.1881 mg/kg for pyrene, with few others like dibenzo-[*a*,*h*]anthracene, naphthalene, indeno[1,2,3-*c*,*d*]pyrene, fluorene, *etc.* were below the detection limits. The concentrations of PAHs in sediment for the dry season were generally higher than that of the wet season.

Ring-based analysis of PAHs in water and sediment

Two to three-ringed PAHs: The results for the analysis revealed that two to three-ringed PAHs concentrations were higher during the dry season for both water and sediment samples. Anthracene and Phenanthrene were the predominant PAHs for both samples in both seasons. The concentrations of two to three-ringed PAHs were generally higher in sediments than water samples for both seasons, with dry season recording the greater concentration.

Four-ringed PAHs: Fluoranthene and pyrene (which is the overall predominant PAHs) were the most predominant four-ringed PAHs with a mean value in water samples of 0.4743 mg/L and 7.0063 mg/L, respectively in the wet season (Table-4) and 2.0234 mg/L and 0.9501 mg/L, respectively for the dry season (Table-3). Their concentration in sediment samples was 0.819 mg/kg and 2.3899 mg/kg, respectively in the dry season and 0.174 and 0.1881 mg/kg for the wet season. Pyrene had the highest concentration among the four-ringed PAHs occurring at location 1 with a value of 83.5702 mg/L, recorded in the water samples.

Five-ringed PAHs: Dibenzo[*a*,*h*]anthracene recorded the highest occurring concentration (3.9592 mg/kg, Table-3) among the five ringed PAHs. During the dry season, five-ring PAHs were moderately present in sediment samples, although benzo-[*a*]pyrene was absent across all locations.

Six-ringed PAHs: Benzo[*g*,*h*,*i*]perylene and indeno[1,2,3-*c*,*d*]pyrene were the least occurring PAHs, a similar result were also recorded in sediments and soil samples in Warri and its surrounding areas [22]. The two PAHs were both below detection limits in water and sediment samples for both seasons and so no concentration was recorded.

Location analysis: In the dry season (Table-3), location 2 had the highest overall concentration of PAHs in the water

TABLE-3
DRY SEASON CONCENTRATION OF PAHs IN SEDIMENT AND WATER SAMPLES (mg/kg)

Locations/PAHs concentration	Sample	Locat 1	Locat 2	Locat 3	Locat 4	Locat 5	Total	Mean
Acenaphthene	Sd	1.7501×10^{-1}	5.960×10^{-1}	8.2610×10^{-1}	5.6410×10^{-1}	4.7510×10^{-1}	2.6401	0.5274
	W	ND	1.8908	8.2955×10^{-1}	3.4776×10^{-1}	8.2955×10^{-1}	3.8977	0.7858
Acenaphthylene	Sd	6.470×10^{-1}	ND	ND	ND	ND	6.4701×10^{-1}	0.1294
	W	ND	3.9289	ND	ND	ND	3.9289	0.7858
Anthracene	Sd	ND	ND	1.6078	1.0995	ND	2.7073	0.5415
	W	ND	2.9562	3.2909	ND	3.2909	9.5385	1.9076
Benz[*a*]anthracene	Sd	ND	ND	2.0231	ND	ND	2.0231	0.4046
	W	ND	11.0294	ND	ND	ND	11.0294	2.2059
Benzo[*a*]pyrene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*b*]fluoranthene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*g*,*h*,*i*]perylene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*k*]fluoranthene	Sd	ND	ND	3.6209	2.9302	ND	6.5512	1.3102
	W	ND	7.8718	6.3880	ND	6.3880	20.6478	4.1296
Chrysene	Sd	60.8481	1.1333	3.3379	3.1100	ND	68.4292	13.6858
	W	ND	6.2272	4.2503	ND	4.2503	14.7278	2.9456
Dibenzo[*a*,*h*]anthracene	Sd	2.6329	2.9214	5.6956	4.5869	ND	15.8369	3.9592
	W	ND	ND	ND	ND	ND	ND	ND
Fluoranthene	Sd	6.9310×10^{-1}	5.1801×10^{-1}	8.2820×10^{-1}	1.4186	6.3700×10^{-1}	4.0900	8.1903×10^{-1}
	W	ND	3.8086	3.1543	ND	3.1540	10.1172	2.0234
Fluorene	Sd	3.7211×10^{-1}	3.6401×10^{-1}	2.4800×10^{-1}	3.3500×10^{-1}	ND	ND	2.6410×10^{-1}
	W	ND	1.7237	8.1767×10^{-1}	ND	8.1768×10^{-1}	3.3590	0.6718
Indeno[1,2,3-*c*,*d*]pyrene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Naphthalene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Phenanthrene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	1.7910	2.9388	ND	2.9387	7.6685	1.5337
Pyrene	Sd	6.4119	3.1859	4.9301×10^{-1}	1.3385	5.2010×10^{-1}	11.9495	2.3899
	W	ND	2.4249	1.1628	ND	1.1629	4.7506	0.9501
ΣPAHs	Sd	71.8168	8.7168	18.7161	15.3814	1.6310	116.5378	24.0305
	W	ND	43.6525	22.8323	0.3477	22.8320	89.6645	17.9329
Range	Sd	0.1750-60.8481	0.3640-3.1859	0.2480-5.6956	0.3350-4.5869	0.4751-0.6370	0.6470-68.4292	0.1294-13.6858
	W	ND	1.7237-11.0294	1.1629-6.3880	0.3477-2.8796	0.8177-6.3880	3.3590-14.2598	0.6718-2.8519
Mean	Sd	4.6825±1.08	5.8663×10^{-1} ±1.02	1.2563±0.67	1.0438±0.39	0.1109±0.2	7.6881±1.88	1.6031±0.39
	W	NIL	2.8409±0.85	1.5626±0.81	0.2017±0.08	1.5600±0.81	5.4977±0.56	1.0995±0.07

N/B: ND = Not detected, Sd = Sediment, W = water and Loct. = Location

samples and location 3 was next to it, while surprisingly all 16 USEPA priority PAHs under investigation were below detection in location 1. The degrees of concentration of PAHs in the locations were as follows; location 2 > location 3 > location 5 > location 4 > location 1 (no PAH detected in location 1).

In sediment samples, location one had the highest overall concentration, next to location 3. The order is as follows; location 1 > location 3 > location 4 > location 2 > location 5. Chrysene and benzo[*k*]fluoranthene were the highest occurring PAHs in sediment and water samples, respectively in the dry season. In wet season (Table-4), the concentration of PAHs was lower for both samples across the locations than in dry season. For the sediment sample, location 4 had the highest overall PAH concentration. Few PAHs were actually above the detection limit in sediment samples in the wet season. The order of concentration was as follows: location 5 > location 4 > location 3 > location 1> location 2 (no PAH detected in location 2). For water samples, location 4 had the highest overall concentration (pyrene alone made up 88.67%). The degree of concentration is as follows: location 4 > location 2 > location 5 > location 1> location 3 (no PAH detected in location 3).

Significant difference in mean PAHs of samples using One-way ANOVA: The significant difference in mean PAHs of samples using the 1-way ANOVA are shown in Table-5. The paired sample correlation showed insignificant inverse relationship between dry and wet seasons at *p*-value > 0.05. Table-5 has shown that the difference in the average of water in both seasons is not significant at F = 1.183, *p*-value > 0.05. Likewise, the difference in the mean of sediment in both seasons was also not significant F = 3.426 $p > 0.05$.

Human health risk assessment: Some PAHs have been proven to be carcinogenic to both humans and animals, and are classified as such by the U.S. Agency for Toxic Substances and Disease Registry (ATSDR) and the U.S. Environmental Protection Agency (EPA). These PAHs include benzo[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo(*b*)fluoranthene, benzo-

TABLE-4
WET SEASON CONCENTRATION OF PAHs IN SEDIMENT AND WATER SAMPLES (mg/kg)

R	Sample	Locat 1	Locat 2	Locat 3	Locat 4	Locat 5	Total	Mean
Acenaphthene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Acenaphthylene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Anthracene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benz[*a*]anthracene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*a*]pyrene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*b*]fluoranthene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*g*,*h*,*i*]perylene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Benzo[*k*]fluoranthene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Chrysene	Sd	ND	ND	ND	2.5011×10^{-1}	ND	2.5010×10^{-1}	0.0624
	W	ND	ND	ND	ND	ND	ND	ND
Dibenzo[*a*,*h*]anthracene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	6.5594	ND	6.5594	1.3119
Fluoranthene	Sd	0.0603	ND	1.5101×10^{-1}	0.1142	5.4401×10^{-1}	0.8690	1.7410×10^{-1}
	W	2.7534×10^{-1}	1.3853×10^{-1}	ND	1.7541	2.0346×10^{-1}	2.3715	0,4743
Fluorene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Indeno[1,2,3-*c*,*d*]pyrene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Naphthalene	Sd	ND	ND	ND	ND	ND	ND	ND
	W	ND	ND	ND	ND	ND	ND	ND
Phenanthrene	Sd	ND	ND	ND	ND	1.3500×10^{-1}	0.1354	0.0271
	W	ND	ND	ND	ND	4.8313×10^{-1}	4.8313×10^{-1}	0.0966
Pyrene	Sd	ND	ND	ND	ND	9.410×10^{-1}	0.9405	0.1881
	W	ND	1.4610	ND	83.5703	ND	85.0313	7.0063
ΣPAHs	Sd	0.0603	NIL	0.1514	0.3641	1.6194	2.1949	0.0098
	W	2.7534×10^{-1}	1.5996	ND	94.2386	6.8665×10^{-1}	94.4453	8.8891
Range	Sd	ND-0.0603	NIL	ND-0.1514	0.1010-0.2501	0.1350-0.9410	0.1013-0.9405	0.0203-0.1881
	W	ND – 0.2753	0.1385-1.4610	ND	1.7541-83.5703	0.2035-0.4832	0.4832-85.0313	0.0966-7.0063
Mean	Sd	0.0037±0.02	NIL	0.0095±0.04	0.0291±0.07	0.1011±0.26	0.1435±0.31	0.0018±0.06
	W	0.0172	0.0999±0.81	ND	5.8899±3.91	0.0429±0.19	5.9028±2.17	0.5556±1.75

N/B: ND = Not detected, Sd = Sediment, W = water and Loct. = location.

TABLE-5
SIGNIFICANT DIFFERENCE IN MEAN PAHs OF WATER AND SEDIMENT
IN DRY AND WET SEASON USING THE ONE-WAY ANOVA

	Sum of squares		df		Mean square		F		Sig.	
	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment
Between groups	2.367	19.809	1	1	2.367	19.809	1.183	3.426	0.285	0.074
Within groups	60.011	173.456	30	30	2.000	5.782				
Total	62.378	193.265	31	31						

[*b*]fluoranthene, benzo[*k*]fluoranthene, indeno[1,2,3-*c*,*d*)pyrene and dibenzo[*a*,*h*]anthracene.

Benzo[*a*]pyrene as a group 1 agent (carcinogenic to humans); dibenz[*a*,*h*]anthracene as group 2A (probable carcinogen to humans); benzo[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, chrysene and indeno[1,2,3-*c*,*d*]pyrene are classified into group 2B (possibly carcinogenic to humans), [22,23]. However, guideline values have only been established for benzo[*a*]pyrene [22,23], as data for other PAHs remain insufficient. Nonetheless, the use of relative potency factors (RPFs) can help estimate the relative contribution of various PAHs to overall risk [17].

The human health risk assessment was determined by estimating the chronic daily intake (CDI) using the levels of PAHs detected in water samples and the parameters data collected from the residents of the study location with reference values of USEPA. The Cancer Risk was calculated by multiplying the chronic daily intake by the slope factor of the PAHs.

Chronic daily intake index (oral exposure ingestion): The data presented in Table-6 summarize the mean values obtained from residents across the five sampling locations (as shown on the study area map), with reference to USEPA guidelines. Health risk estimations were conducted for three

TABLE-6
PARAMETERS USED IN THE CANCER
RISK ASSESSMENT OF PAHs

Definition	Mean			Units
Age	10	25	70	years
Ingestion rate (IR)	0.60	1.30	2.00	Litre/Day
Exposure frequency (EF)	365	365	365	years
Body weight (BW)	35	60	70	Kg
Average time (AT)	3650	9125	25550	Days
Exposure duration (ED)	10	25	70	years

age groups with upper age limits of 10, 25 and 70 years, to assess age-dependent exposure and vulnerability.

Carcinogenic risk (CR) index: In estimating cancer risk, CDI is multiplied by the cancer oral slope factors (OSF) of the substance under investigation [17] (eqn. 3):

$$\text{CR} = \text{CDIing} \times \text{SF} \tag{3}$$

where CDIing = chronic daily intake by ingestion (mg/kg day) and SF stands for slope factor of the pollutant $(\text{mg/kg/day})^{-1}$. CR value over 1.00×10^{-5} indicates the potential carcinogenic risk under most regulatory programs.

Cancer risk assessment

The results for the risk assessment in Tables 7 and 8 showed the cancer risk associated with polycyclic aromatic hydrocarbon ingestion at the various locations for both wet and dry seasons in the study area. The results for the selected PAHs to be used for the CR assessment were not detected for location 2 in the dry season and location 3 in the wet season, so no CR assessment was carried out for them.

Dry season: In dry season (Table-7), location 2 showed a range of 1.9089×10^{-2} to 3.946×10^{-1} for 10 years old, 3.1457×10^{-2} to 4.5458×10^{-1} for 25 years old and 1.08625×10^{-2} to 8.8801×10^{-1} for 70 years old. The overall PAHs cancer risk values were 8.2911×10^{-1}, 1.0310, 1.5401 for 10 years, 25 years and 70 years, respectively. The PAH with the highest risk value was benz[*a*]anthracene, being the highest for all the age brackets. Next to it were benzo[*k*]fluoranthene and chrysene with a slight difference in their values for the three age brackets. The overall CR values of the PAHs examined were above the tolerable limit of 1.00×10^{-5} with the highest cancer risk occurring for the 70 years' bracket. The results obtained for this location in the dry season is generally higher than that reported by Inam *et al*. [17], for fresh water samples from Egbema/Ogba/Ndoni communities of Rivers state, Nigeria.

Location 3 had an overall CR value 3.2390×10^{-1}, 3.9601×10^{-1} and 4.5500×10^{-1} for age 10, 25 and 70, respectively. The highest value also occurred for age 70 like in location 1. The PAH with the highest CR value was benzo[*k*]fluoranthene with CR value of 1.3989×10^{-1}, 1.1658×10^{-1} and 1.3336×10^{-1} for age 10, 25 and 70, respectively. This aligns with the findings reported by Ekere *et al*. [24], for fish and water samples. The results again were higher than the acceptable limit of 1.00×10^{-5} for these PAHs in the water samples.

The overall CR values in location 5 for 10, 25 and 70 years were 6.33×10^{-1}, 6.64×10^{-1} and 7.50×10^{-1}, respectively. This was higher than the permissible limit of 1.00×10^{-5}. The PAHs with the greatest concern were benzo[*k*]fluoranthene and chrysene. The 70 years' age bracket still had the highest

TABLE-7
CANCER RISK ASSESSMENT OF PAHS IN DIFFERENT LOCATIONS IN DRY SEASON

PAHs	Age (Years)	Location 2	Location3	Location 5
Benz[*a*]anthracene	10	2.4154×10^{-1}	Not detected	Not detected
	25	2.0129×10^{-1}	Not detected	Not detected
	70	2.3027×10^{-1}	Not detected	Not detected
Benzo[*a*]pyrene	10	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected
Benzo[*b*]fluoranthene	10	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected
Benzo[*k*]fluoranthene	10	1.9089×10^{-2}	1.3989×10^{-1}	1.3918×10^{-1}
	25	1.4366×10^{-1}	1.1658×10^{-1}	1.1653×10^{-1}
	70	1.0863×10^{-1}	1.3336×10^{-1}	1.3339×10^{-1}
Chrysene	10	1.3637×10^{-1}	9.3210×10^{-3}	9.3299×10^{-3}
	25	4.5458×10^{-1}	7.7601×10^{-3}	7.7760×10^{-3}
	70	8.8801×10^{-3}	8.8801×10^{-3}	8.8901×10^{-3}
Dibenz[*a*,*h*]anthracene	10	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected
Fluoranthene	10	3.7748×10^{-2}	1.7910×10^{-2}	1.8125×10^{-2}
	25	3.1457×10^{-2}	1.4920×10^{-2}	1.5105×10^{-1}
	70	3.5986×10^{-1}	1.7070×10^{-2}	1.7279×10^{-1}
Indeno[1,2,3-*c*,*d*]pyrene	10	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected
Total	10	4.3501×10^{-1}	1.6701×10^{-1}	6.3310×10^{-1}
	25	7.0210×10^{-1}	1.3910×10^{-1}	6.6400×10^{-1}
	70	1.1700E+00	1.5900×10^{-1}	7.5000×10^{-1}

TABLE-8
CANCER RISK ASSESSMENT OF PAHS IN DIFFERENT LOCATIONS IN WET SEASON

PAHs	Age (Years)	Location 2	Location3	Location 4	Location 5
Benz[*a*]anthracene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Benzo[*a*]pyrene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Benzo[*b*]fluoranthene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Benzo[*k*]fluoranthene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Chrysene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Dibenz[*a*,*h*]anthracene	10	Not detected	Not detected	1.4365	Not detected
	25	Not detected	Not detected	1.1971	Not detected
	70	Not detected	Not detected	1.3694	Not detected
Fluoranthene	10	1.1011×10^{-3}	3.3069×10^{-3}	3.8389×10^{-3}	4.4557×10^{-3}
	25	2.5280×10^{-3}	2.7557×10^{-3}	3.1645×10^{-3}	3.6970×10^{-3}
	70	1.1011×10^{-3}	4.3168×10^{-3}	3.6620×10^{-3}	6.0420×10^{-1}
Indeno[1,2,3-*c*,*d*]pyrene	10	Not detected	Not detected	Not detected	Not detected
	25	Not detected	Not detected	Not detected	Not detected
	70	Not detected	Not detected	Not detected	Not detected
Total	10	1.1011×10^{-3}	3.3069×10^{-3}	1.4403E+00	4.4557×10^{-3}
	25	2.5280×10^{-3}	2.7557×10^{-3}	12287E+00	3.6970×10^{-3}
	70	1.1011×10^{-3}	4.3186×10^{-3}	1.4060E+00	6.0420×10^{-1}

overall cancer risk, with a value of 4.55×10^{-1}. It was higher than that reported by other researchers [17,19], but similar to that recorded long West Coast Peninsular Malaysia (CR value 10^{-2}) reported by Mirsadeghi *et al*. [25].

Wet season: The CR values were generally higher in the dry season, likely due to reduced dilution of PAHs from rainfall and runoff. At location 1 (Table-8), only fluoranthene was detected, with CR values of 6.03×10^{-3}, 5.03×10^{-3} and 5.75×10^{-3} for ages 10, 25 and 70 years, respectively, indicating that all values exceeding the USEPA limit of 1.00×10^{-5} for PAHs in freshwater.

During the wet season at location 2, only fluoranthene was detected for CR assessment, with values of 3.77×10^{-2}, 3.15×10^{-2} and 3.60×10^{-2} for ages 10, 25 and 70 years, respectively exceeding values reported in Abu Qir Bay, Egypt (below USEPA limits) [26] and Çankırı, Turkey (1.53×10^{-6}) [19]. At location 4, both fluoranthene and dibenz[*a*,*h*]anthracene were present, with CR values of 3.84×10^{-2}, 3.16×10^{-2} and 3.66×10^{-2} for fluoranthene, and 1.44, 1.20 and 1.37 for dibenz[*a*,*h*] anthracene across the same age groups. At location 5, fluoranthene was again the only PAH detected, with CR values of 4.46×10^{-3}, 3.70×10^{-3} and 6.04×10^{-3}. Across all locations, the highest CR values were consistently recorded for the 70-year age group, followed by 25 and 10 years.

Generally, it was observed that the PAHs concentration level for the wet season showed a marked decline from that of the dry season. This may be due to dilution of the river by rainfall, erosion and flood. Most PAHs were below the detection limits and so had no CR value estimated for them. In dry season, benzo[*k*]fluoranthene and chrysene were the most dominant PAHs, having values above the USEPA acceptable limit of 1.00×10^{-5} and were found at all locations but location 4 in the dry season. The least CR value recorded for benzo[*k*]fluoranthene and chrysene were 1.1344×10^{-3} and 2.068×10^{-3}, respectively, although their concentrations were below the detection limit at location 4 in some locations. Whereas, for the wet season, the dominant PAHs were fluoranthene and dibenz[*a*,*h*]anthracene, the former being the most dominant.

Conclusion

The findings of this study indicate that the measured physico-chemical parameters *viz*., pH, TSS, DO, conductivity and organic matter, were within the recommended limits. However, the presence of polycyclic aromatic hydrocarbons (PAHs) in the study area points to a notable degree of pollution, with higher concentrations observed during the dry season compared to the wet season. Overall, the sediment samples contained both a greater number and higher concentrations of PAHs than water samples in both seasons. The cancer risk assessment (CR) revealed that some PAHs present in the environment pose risks above acceptable limits, particularly during the dry season. Among all age groups assessed, individuals in the 70-year age bracket faced the highest overall cancer risk. The PAHs of greatest concern based on their concentrations and toxicity were benzo[*k*]fluoranthene and chrysene. These findings highlight the need for continuous monitoring and mitigation strategies to reduce PAH contamination and associated health risks in the region.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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