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Assessment of Polycyclic Aromatic Hydrocarbons Concentration in Surface Water, Sediment and Blue Crab, *Callinectes Amnicola* (De Rochebrune, 1883) from Buguma Creek, Rivers State, Nigeria

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Abstract: *Polycyclic aromatic hydrocarbons (PAHs) are persistent toxic contaminants that pose ecological and human health risks. This study assessed PAH levels in surface water, sediment, and Callinectes amnicola from Buguma Creek, Rivers State, Nigeria. Surface water, sediment and C. Amnicola samples were collected bimonthly from three stations between May and September 2022 for PAHs analysis by gas chromatography. Physico-chemical parameters such as temperature (°C), salinity (‰), pH, dissolved oxygen (DO, mg/L), and biological oxygen demand (BOD, mg/L) were measured in surface water. Data were statistically analysed by Analysis of Variance (ANOVA). Results showed significant no spatial but temporal variations in DO, BOD, and salinity, with the highest values recorded for DO (5.3±0.4 mg/L) and BOD (5.9±0.3 mg/L) in July, while salinity peaked in May (19.0±0.0 ‰). Of the 17 PAHs listed by the agency of toxic substances and disease registry, 12 (including Naphthalene which was not listed in ATSDR) were detected, excluding benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene, and benzo(g,h,i)perylene. PAHs concentration followed the trend: sediment > water > C. amnicola, with the highest levels recorded in July. PAH sources were both petrogenic and pyrogenic, with petrogenic inputs dominating surface water, pyrogenic sources in C. amnicola, and both contributing equally to sediment contamination. Biota-water accumulation factor (BWAF) and biota-sediment accumulation factor (BSAF) indicated C. amnicola primarily accumulated PAHs from water rather than sediment. PAHs concentrations in all matrices exceeded by several magnitude the US Environmental Protection Agency's limits (<1000 ng/g for sediment, <1000 ng/L for water). The presence of carcinogenic PAHs, including benzo(a)anthracene, benzo[b]fluoranthene, and benzo(a)pyrene in water and sediment, and chrysene and benzo[b]fluoranthene in C. amnicola, raises concerns over bioaccumulation and trophic transfer. In conclusion, Buguma Creek is polluted with PAHs from both pyrogenic and petrogenic sources. Given the potential ecological and human health risks associated with PAHs, regular monitoring and pollution prevention measures are essential for mitigating anthropogenic impacts and ensuring sustainable ecosystem.*

Keywords: contaminants, PAH, shellfish, brackish water creek, water quality.

INTRODUCTION

The Niger Delta, Nigeria, is rich in oil and gas resources, with a long history of exploration and development (Ite *et al*., 2013). However, sabotage, crude oil theft, artisanal refining, and operational spills have significantly impacted terrestrial and aquatic ecosystems (Lindén and Pålsson, 2013; United Nations Environment Programme, 2011). Of particular concern are crude oil spills in intertidal mangrove swamps, including Buguma Creek (4° 44'N, 6° 52'E), which has not been exempt from such contamination. Buguma, characterised by its extensive mangrove swamps, interconnecting creeks, and proximity to the ocean, relies heavily on fishing for both subsistence and commercial purposes. Seafood is a dietary staple, forming an essential component of their local cuisines. A major class of environmental pollutants resulting from crude oil contamination in coastal waters is polycyclic aromatic hydrocarbons (PAHs) (Pegg and Zabbey, 2013). PAHs are organic compounds produced during the incomplete combustion of organic materials (Ofori *et al*., 2020; Abdel-Shafy and Mansour, 2015; Russell, 2013) and are ubiquitous in soil, sediment, air, water, and biota (Lawal, 2017) and food (Ab-Latif *et al.*, 2024; Abdel-Shafy and Mansour, 2015). In coastal waters, PAHs originate from of accidental oil spillage or through human discharge of petroleum byproducts (petrogenic sources), combustion processes of fuel and other organic substances (pyrogenic sources), or natural and biological activities (Okedere and Elehinafe, 2022; Abdel-Shafy and Mansour, 2015). Based on their benzene rings, PAHs are classified as low molecular weight, LMW PAHs (PAHs with 2-3 benzene rings) and high molecular weight (HMW) PAHs have 4 or more benzene rings (Ofori *et al*., 2020; Olayinka *et al*., 2019; Stogiannidis and Laane, 2015; Abou-Arab *et al*., 2014). For instance, anthracene and phenanthrene with 2 and 3 benzene rings are categorised as LMW PAHs while, pyrene and benzo(a)pyrene with 4 and 5 benzene rings respectively are classed as HMW PAHs.

Agency for Toxic Substances and Disease Registry (ATSDR,1995), listed 17 priority PAHs include acenaphthene, acenaphthylene, anthracene, benzo[a]anthracene, benzo[a]pyrene, benzo[e]pyrene, fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[j]fluoranthene benzo[g,h,i]perylene, dibenz[a,h]anthracene, chrysene, phenanthrene, indeno[1,2,3-c,d]pyrene, fluorene, and pyrene. According to Menzie and Potocki, (1992), eight PAHs namely: chrysene, benzo(a)anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo(a)pyrene, dibenz[a,h]anthracene, indeno[1,2,3-c,d]pyrene, and benzo[g,h,i]perylene has been identified and profiled as potent carcinogens, mutagens and teratogens.

All components of the aquatic environments (water, sediment and aquatic organisms) are mostly described as the ultimate sinks of PAHs (Ayandiran *et al*., 2022). For instance, research conducted around the Atlas Cove Jetty in Lagos, Nigeria, detected 17 PAH congeners in sediment samples and 10 in biota samples, with total PAHs concentrations in sediments ranging from 2.15 to 36.46 mg/kg and in fish samples from 11.89 to 71.06 mg/kg. Similarly, Aigerua and Seiyaboh (2021) reported PAHs concentrations in sediment ranging from <0.01 to 3,965.4 µg/kg, with most detected compounds exceeding regulatory limits. High molecular weight (HMW) PAHs (4-6 rings) accounted for 94.29% of the total PAHs distribution, while low molecular weight (LMW) PAHs (2–3 ring) were detected in trace amounts, constituting approximately 5.71% of total PAHs. Furthermore, Ihunwo and Ibezim-Ezeani (2021) recorded PAHs concentrations in surface water (ΣPAH_{16} = 0.125±0.011 mg/L) and in *C. amnicola* tissues (ΣPAH_{16} = 10.659±2.399 mg/kg). PAHs, being lipophilic and chemically stable, readily bioaccumulate in aquatic organisms (Nkpaa *et al*., 2013), with uptake by aquatic organisms occurring rapidly through contaminated water, sediments, and food chains and may accumulate in tissue in high concentrations (Abdel-Shafy and Mansour, 2015). Studies have demonstrated PAHs presence in various aquatic species, including crabs, which serve as bioindicators of PAHs pollution (Nyarko *et al*., 2011; Ofori *et al*., 2020).

Crabs, are invertebrates belonging to the order Decapoda, are widely distributed and serve as an important protein source for coastal communities (Olalekan and Lawal-Are, 2013; Oduro *et al*., 2001). *Callinectes amnicola* (Rochebrune, 1883), from the family Portunidae, is a common estuarine species in tropical and subtropical regions, including Nigeria (Ezekiel and Bernard, 2014). In Buguma, *C. amnicola* (locally called *Ikoli*) is the most frequently fished, highly prized and consumed crab species, particularly in traditional dishes such as *onunu* and seafood okro soup (Amachree, 2018, *unpublished*). While crabs may constitute a relatively small portion of the overall diet, their contribution to PAH intake can be significant. Given the mutagenic, carcinogenic, and teratogenic properties of PAHs, this poses a potential health risk (Okedere and Elehinafe, 2022). Studies have detected PAHs in *C. amnicola* from Woji Creek (Ihunwo et al., 2019, 2021) and in other Nigerian waters including: *Callinectes species* from the Ondo coastal area (Oladele *et al*., 2008), Azuabie Creek (Daka and Ugbomeh, 2013), polluted creeks in Rivers State (Anyanwu and Chris, 2023), and Atlas Cove Jetty, Lagos (Olayinka *et al*., 2019). A recent study by Ihunwo and Ibezim-Ezeani (2021) reported that excess cancer risk from PAH exposure exceeded safety limits across all age groups consuming *C. amnicola* from Woji Creek. However, there is a lack of data on PAH levels in *C. amnicola* from Buguma Creek, despite its significance as a dietary staple. This study aimed to assess PAH concentrations in water, sediment, and *C. amnicola* from Buguma Creek.

MATERIALS AND METHODS

Study Area and sampling Stations

The study was carried out in Buguma Creek in Asari-Toru Local Government of Rivers State, Nigeria. The creek system within the study area consists of Amanayabo Okolo with associated interconnecting creeks such as Ido Canal and Jordan Creek which interconnect and surround Buguma and Ido communities. The study area is dominated by mangroves and few Nypa palm (*Nypa fructicans*). The indigenes depend on the creek for food, recreation, open defaecation, transportation, spiritual purposes and general livelihood (Amachree and Soberekon, 2022).

Three sampling stations were established for the study (Figure 3.1). Station 1: Amanyanabo Okolo (Maryhood Bridge; Latitude 4°44'19.0932"N; Longitude 6°52'3.8568"E) is downstream and 1.3km apart from station 2, activities include landing sites for artisanal bunkering products,

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mangrove firewood, domestic waste disposal, open defecation and fishing. Station 2: Amanyanabo Okolo (trans-Kalabari highway/Buguma North; Latitude 4°44'40.9488"N; Longitude 6°51'56.7036"E) is downstream and ~1 km apart from station 3, activities include fishing and mangrove firewood cutting. Station 3: Ido Canal (Latitude 4°44'40.722"N; Longitude 6°51'25.7544"E) is upstream and the activities include sand mining, fishing and picking of periwinkle.

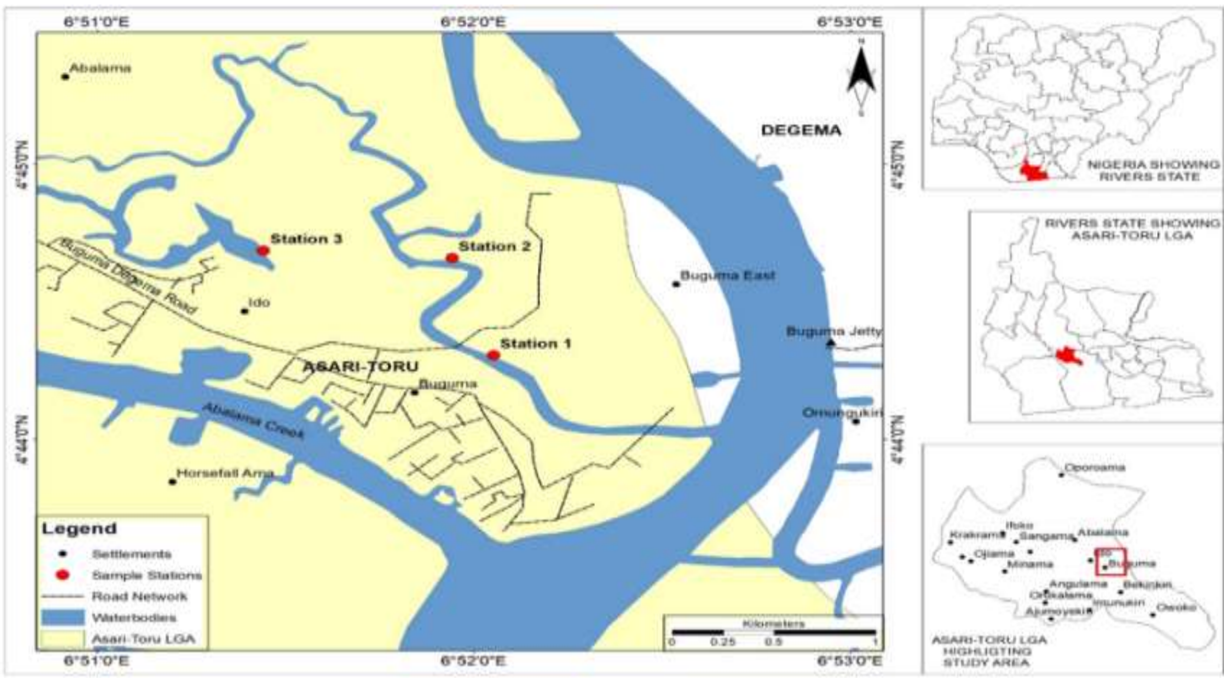

Figure 1: Map of the study area showing the sampling stations

Determination of Physico-Chemical Parameters

The water quality was monitored for temperature, hydrogen-ion ncentration (pH), dissolved oxygen (DO), Biological oxygen demand (BOD) and salinity. Surface water temperature (°C), pH and salinity (‰) were measured *in-situ* with mercury-in-glass thermometer, pH meter (Model HI 9812, Hannah Products, Portugal) and hand-held Refractometer (Model RE 6783, Atago Products, Portugal) respectively. Water samples for dissolved oxygen, DO (mg/l) and Biochemical oxygen demand, BOD_5 (mg/l) were collected in amber bottle and taken to laboratory for analysis of the individual parameters according to standard procedures set by the American Public Health Association (APHA, 2017).

Collection of surface water, sediment and *C. amnicola* for PAHs Analysis

Samples were collected during ebb tide from the three stations bimonthly between May and September 2022 (Edori and Kpee, 2019). Surface water samples were collected in pre-acid-washed glass bottles while sediment samples were collected using a stainless-steel spoon and stored in polyethylene bags (Rizk *et al*., 2022). *Callinectes amnicola* were collected with drag nets and identified using identification keys (Schneider, 1992; Manning and Holthuis, 1981). Thereafter, all sample were taken to a reputable laboratory in Port Harcourt to analysis for PAHs concentration according to American Standard Test Method for Oil and Grease and Petroleum Hydrocarbons in Water, ASTM D7679-16 (ASTM 2016).

Preparation and Preservation of Water, Sediment and *C. amnicola*
Samples were prepared and preserved as described (ASTM D7679-16). Water Samples Sediment and *C. amnicola* samples were prepared in clean glass bottles packed and stored below 4 ℃, for a maximum of 3 days prior to extraction and 14 days after extraction prior to analysis. However, before packing and storage, surface water was acidified to pH below 2 with concentrated hydrochloric acid.

Water Sample Extraction
Extraction was done by the method as described (ASTM D7679-16). Preserved surface water samples were thoroughly mixed by shaking. Thereafter, 10 ml of dicloromethane (DCM) solvent was added to the water sample and recapped immediately. The water sample was mixed vigorously for 2 mins by shaking while releasing pressure. The sample was transferred into a separatory funnel and allowed to stand for 5 minutes in the separatory funnel until the content settles. Thereafter, the precipitate was transferred into a clean volumetric flask through a filter paper containing 5g of solvent pre-wetted sodium sulphate for total oil and grease (TOG) analysis and addition of silica gel of 4g for total hydrocarbon content (THC) and Total petroleum hydrocarbon (TPH) analysis. The extract volume was recorded and the remaining contents of the separatory funnel were drained into a 1000ml graduated cylinder and the volume was recorded for later calculations. An aliquot of the extract was read using gas chromatography (Agilent 7890A) and the result calculated and the result expressed in mg/l.

Sediment and *C. amnicola* Extraction
The sediment and *C. amnicola* were extracted as described (ASTM D7679-16). Sediment or *C. amnicola* sample was homogenized and 15.0 ± 0.1g weighed out into a clean 250ml extraction bottle. Thereafter, 30ml of extraction solvent (dichloromethane) was added to the sample in the 250ml bottle and covered properly with aluminium foil paper. Sediment or *C. amnicola* samples were swirled for 2mins while releasing the cover to control pressure build up and allow the extract to settle for 15mins. The extracted mixture was passed through a filter paper containing 6g of solvent pre-wetted sodium sulphate only for removal of moisture content for TOG and activated silica gel of 4g for THC and TPH analysis. The volume of the extract obtained was recorded for further calculations. Thereafter, an aliquot of the extract was transferred into a vial for reading gas chromatography (Agilent 7890A) and the result expressed in mg/kg weight of sediment or crab.

Statistical Analysis and Calculations of Data
Statistical analysis was carried out on all data using the Minitab version 16 for Microsoft Windows. Data were presented as mean ± standard deviation (STD). Data were analyzed by one-way analysis of variance (ANOVA) and descriptive statistics (sum, mean and STD). Tukey’s post-hoc at 95%

confidence limit was used to provide which means are significantly different from each other. Calculation of bioaccumulation factor, BAF (i.e., the transfer factor in *C. amnicola* from the aquatic ecosystem, which includes water and sediments) was calculated as described (Rashed, 2001; Kalfakakour and Akrida-Demertzi, 2000) as follows:

$$BAF = \frac{PAH\ Concentration\ in\ biota}{PAH\ concentration\ in\ sediment\ or\ water}$$

RESULTS

Physico - Chemical Characteristics of Surface Water

The results of the spatial and temporal variations in the physico-chemical characteristics of surface water are presented on Tables 1. There were no spatial but temporal statistically significant differences ($p<0.05$) for most of the parameters measured (Table 1). No differences were seen in temperature and pH but for DO, BOD and salinity with July showing highest for DO and BOD, while May peaked for salinity (Table 2).

Table 1: Temporal and spatial variations in physico-chemical characteristics of surface water.

PARAMETER	MONTH			
	MAY	**JULY**	**SEPT**	**P-value**
Temperature (°C)	26.00 ± 0.00^a	26.00 ±0.00^a	26.33 ± 0.58^a	0.422
DO (mg/l)	2.40 ± 0.55^b	5.27 ± 0.45^a	3.02 ± 0.02^b	0.000
pH	7.00 ± 0.00	7.00 ± 0.00	7.00 ± 0.00	-
BOD (mg/l)	3.57 ± 0.21^b	5.87 ± 0.32^a	3.85 ± 0.13^b	0.000
Salinity (‰)	19.00± 0.00^a	15.00 ± 0.00^b	10.93 ± 0.06^c	0.000
		STATION		
	MHB	**TKH**	**IC**	**P-Value**
Temperature °C	26.00 ± 0.00^a	26.00 ± 0.00^a	26.33 ± 0.58^a	0.422
DO (mg/l)	3.79 ± 1.30^a	3.30 ± 1.39^a	3.59 ±1.88^a	0.927
pH	7.00± 0.00	7.00 ± 0.00	7.00 ± 0.00	-
BOD (mg/l)	4.60± 1.30^a	4.23 ± 1.10^a	4.45± 1.37^a	0.939
Salinity (‰)	15.00 ± 4.00^a	14.97 ± 4.05^a	14.97 ± 4.05^a	1.000

Data are mean±SD for *n*=3. Different letter within a row indicates statistically significant difference ($p<0.05$). Note: Maryhood bridge (MHB), Trans-Kalabari highway (TKH) and Ido canal (IC).

Distribution of PAHs in water (mg/l), sediment (mg/kg) and *Callinestes amnicola* (mg/kg)

The distribution of PAHs in surface water, sediment, and *C. amnicola* is presented in Table 2 and 3. Spatial and temporal variations were observed in the number of individual PAHs detected in all matrices. Of the 17 PAHs listed by the agency of toxic substances and disease registry (ATSDR, 1995), 12 (including Naphthalene which was not listed in ATSDR) excluding benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene, and benzo(g,h,i)perylene were identified in Buguma Creek.

In surface water, nine PAHs were detected, comprising five low molecular weight (LMW) PAHs (naphthalene, acenaphthylene, acenaphthene, fluorene, and phenanthrene) and four high molecular weight (HMW) PAHs (fluoranthene, benzo(a)anthracene, benzo(b)fluoranthene, and benzo(a)pyrene). In May, only acenaphthene was found at Maryhood Bridge (MHB) and Trans-Kalabari Highway (TKH), while fluorene was detected at Ido Canal (IC). In July, acenaphthylene, fluorene, and benzo(b)fluoranthene were recorded at MHB, whereas TKH contained naphthalene, acenaphthylene, fluorene, and benzo(b)fluoranthene. Ido Canal had acenaphthylene and benzo(b)fluoranthene. By September, all stations exhibited the same six PAHs: naphthalene, acenaphthylene, phenanthrene, fluoranthene, benzo(a)anthracene, and benzo(a)pyrene. Across all stations and months, the total concentration of LMW PAHs (2–3 rings; petrogenic) exceeded that of HMW PAHs (4–6 rings; pyrogenic) (Tables 2 and 3).

In sediment, eight PAHs were identified: four LMW PAHs (naphthalene, acenaphthylene, phenanthrene, and anthracene) and four HMW PAHs (pyrene, benzo(a)anthracene, benzo(b)fluoranthene, and benzo(a)pyrene). In May, only pyrene was detected across all stations. In July, MHB contained acenaphthylene, phenanthrene, anthracene, and benzo(b)fluoranthene, while TKH had naphthalene, acenaphthylene, phenanthrene, anthracene, and benzo(b)fluoranthene. At IC, acenaphthylene, anthracene, and benzo(b)fluoranthene were recorded. In September, all stations had the same five PAHs: naphthalene, acenaphthylene, anthracene, benzo(a)anthracene, and benzo(a)pyrene. The total HMW PAHs concentrations were higher in May (11.20 mg/kg) and July (89.50 mg/kg) compared to LMW PAHs counterparts, but in September, HMW PAHs (1.06 mg/kg) were lower than LMW PAHs (9.30 mg/kg). Spatially, the sum of HMW PAHs was higher than LMW PAHs at MHB (34.11 vs 16.28 mg/kg), TKH (29.82 vs 19.15 mg/kg), and IC (37.83 vs 13.66 mg/kg).

In *C. amnicola*, three PAHs were identified: one LMW PAH (acenaphthylene) and two HMW PAHs (chrysene and benzo[b]fluoranthene). In May, pyrene was detected at MHB, while TKH contained acenaphthene and pyrene, and IC had acenaphthene alone. In July, anthracene, chrysene, and benzo(b)fluoranthene were found at MHB and IC, whereas TKH had anthracene and benzo(b)fluoranthene. In September, all stations recorded the same PAHs: anthracene, chrysene, and benzo(b)fluoranthene. The total concentration of LMW PAHs was higher than HMW PAHs in May (5.28 mg/kg) and September (3.11 mg/kg), whereas in July, HMW PAHs (21.35 mg/kg) exceeded LMW PAHs (15.21 mg/kg). Spatially, LMW PAH concentrations at MHB and TKH were lower than their HMW counterparts, while at IC, LMW PAHs (10.62 mg/kg) were higher than HMW PAHs (5.60 mg/kg) (Tables 2 and 3).

Overall, total PAH concentrations followed the trend: sediment > water > *C. amnicola* across all stations and months, except in September, where surface water (4.93 mg/L) and *C. amnicola* (4.93 mg/kg) had equal PAHs concentration (Tables 2 and 3).

Bioaccumulation Factor for *C. amnicola*

The biota-water accumulation factor (BWAF) and biota-sediment accumulation factor (BSAF) across different stations and months are presented in Tables 2 and 3. Generally, both BWAF and BSAF values were less than 1, except for BSAF in May and BWAF in September, which exceeded this threshold. For instance, at Maryhood Bridge, the BWAF was 0.78, while the BSAF was 0.38 (Tables 2 and 3).

PAHs concentration in surface water, sediment and *C. amnicola*

The results of PAHs concentration in surface water, sediment and *C. amnicola* are presented in Figure 2. Monthly variations were observed in PAHs concentration in surface water, with the highest value recorded in July (19.40 ± 1.63 mg/L), followed by May (3.22 ± 0.24 mg/L) and September (1.64 ± 0.03 mg/L). However, no significant spatial differences were observed in PAH concentrations in surface water (Figure 2b; $p > 0.05$). The mean concentrations (± SD) were 8.19 ± 10.21 mg/L at MHB, 7.48 ± 8.78 mg/L at TKH, and 8.60 ± 10.50 mg/L at IC.

Similarly, monthly variations were observed in PAHs concentration in sediment, with the highest values recorded in July (42.93 ± 1.69 mg/kg), followed by May (3.75 ± 0.17 mg/kg) and September (3.58 ± 0.12 mg/kg). In contrast to the temporal variations, no significant spatial differences were found in sediment PAH concentrations ($p > 0.05$). The mean concentrations (± SD) were 16.80 ± 23.07 mg/kg at MHB, 16.34 ± 21.99 mg/kg at TKH, and 17.16 ± 23.35 mg/kg at IC.

Likewise, PAHs concentration in *C. amnicola* varied significantly across months but not across stations. The highest concentration (8.04 ± 1.57 mg/kg) was recorded in July, followed by May (3.19 ± 0.26 mg/kg) and September (1.17 ± 0.02 mg/kg).

TYPE OF PAH	NAME OF PAH	MONTH								
		MAY			JULY			SEPTEMBER		
	MATRIX	Water	Sediment	*C. amnicola*	Water	Sediment	*C. amnicola*	Water	Sediment	*C. amnicola*
2-3 rings	Naphthalene	-	-	-	2.90	3.00	-	0.67	5.55	-
	Acenaphtylene	-	-	-	18.19	16.90	-	1.49	2.10	-
	Acenapthene	6.23	-	5.28	-	-	-	-	-	-
	Fluorene	3.45	-	-	15.01	-	-	-	-	-
	Phenanthrene	-	-	-	-	6.39	-	-	-	-
	Anthracene	-	-	-	-	13.50	15.21	1.14	1.66	3.11
	SUM	**9.68**	**-**	**5.28**	**36.10**	**39.79**	**15.21**	**3.31**	**9.30**	**3.11**
4-6 rings	Fluoranthene	-	-	-	-	-	-	1.54	-	-
	Pyrene	-	11.20	3.95	-	-	-	-	-	-
	B (a) ANT	-	-	-	-	-	-	0.03	0.52	-
	Chrysene	-	-	-	-	-	4.38	-	-	1.22
	B (b) FLU	-	-	-	22.09	89.50	16.97	-	-	0.60
	B (k) FLU	-	-	-	-	-	-	-	-	-
	B (a) pyrene	-	-	-	-	-	-	0.05	0.54	-
	Di (a,h)ANT	-	-	-	-	-	-	-	-	-
	IN(cd) PYR	-	-	-	-	-	-	-	-	-
	B(ghi) PYL	-	-	-	-	-	-	-	-	-
	SUM	**-**	**11.20**	**3.95**	**22.09**	**89.50**	**21.35**	**1.62**	**1.06**	**1.82**
	GRAND TOTAL	**9.68**	**11.20**	**9.23**	**58.19**	**129.29**	**36.55**	**4.93**	**10.36**	**4.93**
	BWAF			**0.95**			**0.67**			**1.00**
	BSAF			**1.23**			**0.45**			**0.69**

Table 2: Temporal Partition of PAH in Water (mg/l), Sediment (mg/kg) and *Callinestes amnicola* (mg/kg)
Data are Sum (*n*=3). Note: Benzo(a)anthracene, [B(a)ANT]; Benzo(b)fluoranthene, [B(a)FLU]; Benzo(k)fluoaranthene, [B(k)FLU]; Benzo(a)pyrene, [B(a)PYR]; Indeno(1,2,3-cd)pyrene, [IN(cd)PYR]; Dibenzo(a,h)anthracene, [Di(ah)ANT]; Benzo(g,h,i)perylene, [B(ghi)PYL]; Biota-Water Bioaccumulation factor [BWAF]; Biota-Sediment Bioabbumulation factor [BSAF].

Table 3: Spatial Partition of PAH in Water (mg/l), Sediment (mg/kg) and *Callinestes amnicola* (mg/kg)

TYPE OF PAH	NAME OF PAH	STATION								
		Maryhood bridge			Trans-Kalabari highway			Ido canal		
	MATRIX	Water	Sediment	*C. amnicola*	Water	Sediment	*C. amnicola*	Water	Sediment	*C. amnicola*
2-3 rings	Naphthalene	0.20	1.74	-	3.08	4.96	-	0.29	1.85	-
	Acenaphtylene	6.19	7.62	-	4.57	6.27	-	8.92	5.10	-
	Acenapthene	2.97	-	-	3.26	-	1.90	-	-	**3.38**
	Fluorene	9.25	-	-	5.76	-	-	3.45	-	-
	Phenanthrene	-	2.47	-	-	3.92	-	-	-	-
	Anthracene	0.41	4.45	6.02	0.40	4.00	5.06	0.34	6.72	**7.24**
	SUM	**19.02**	**16.28**	**6.02**	**17.06**	**19.15**	**6.96**	**13.00**	**13.66**	**10.62**
4-6 rings	Fluoranthene	0.52	-	-	0.50	-	-	0.52	-	-
	Pyrene	-	3.70	2.47	-	3.63	1.48	-	3.87	-
	B (a) ANT	0.01	0.03	-	0.01	0.21	-	0.01	0.28	-
	Chrysene	-	-	4.48	-	-	0.39	-	-	**0.72**
	B (b) FLU	5.00	30.10	6.20	4.84	25.80	6.50	12.25	33.60	**4.88**
	B (k) FLU	-	-	-	-	-	-	-	-	-
	B (a) pyrene	0.01	0.27	-	0.02	0.19	-	0.02	0.08	-
	Di (a,h)ANT	-	-	-	-	-	-	-	-	-
	IN(cd) PYR	-	-	-	-	-	-	-	-	-
	B(ghi) PYL	-	-	-	-	-	-	-	-	-
	SUM	**5.55**	**34.11**	**13.15**	**5.37**	**29.82**	**8.37**	**12.80**	**37.83**	**5.60**
	GRAND TOTAL	**24.57**	**50.39**	**19.17**	**22.43**	**48.97**	**15.33**	**25.80**	**51.49**	**16.22**
	BWAF			**0.78**			**0.68**			**0.72**
	BSAF			**0.38**			**0.31**			**0.36**

Data are Sum (*n*=3). Note: Benzo(a)anthracene, [B(a)ANT]; Benzo(b)fluoranthene, [B(a)FLU]; Benzo(k)fluoaranthene, [B(k)FLU]; Benzo(a)pyrene, [B(a)PYR]; Indeno(1,2,3-cd)pyrene, [IN(cd)PYR]; Dibenzo(a,h)anthracene, [Di(ah)ANT]; Benzo(g,h,i)perylene, [B(ghi)PYL]; Biota-Water Bioaccumulation factor [BWAF]; Biota-Sediment Bioabbumulation factor [BSAF].

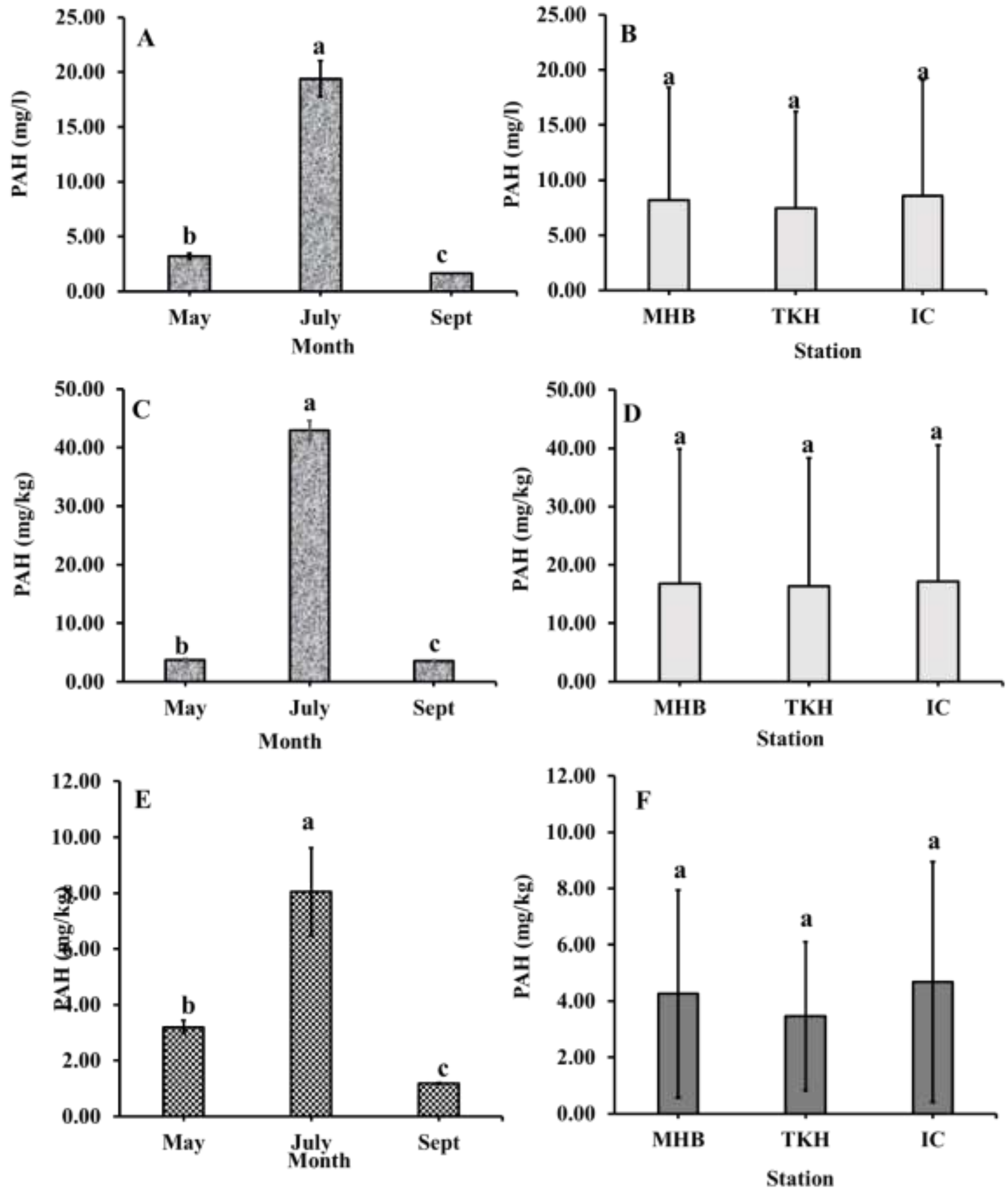

Figure 2. Temporal and spatial PAH concentration in surface water (A and B), sediment (C and D) and C. Amnicola (E and F) from Buguma Creek. Data are mean ±SD, (n=3). Different letters represent significant differences, $p<0.05$). MHB=Maryhood bridge; TKH=Trans-Kalabari highway; IC=Ido canal.

DISCUSSION

Physico-Chemical Characteristics of Surface Water

The spatial and temporal physico-chemical characteristics were monitored in the study area. The values of the physico-chemical parameters measured between the stations were similar but varied significantly between the sampling months. However, values were within the range previously reported in Buguma creek (Amachree and Soberekon, 2022; Oribhabor, 2010). The lack of significant differences between stations could be attributed to the homogeneous nature of the study area. The uniformity in the stations suggested that creek during the sampling period experienced well-mixed water conditions, possibly due to tidal influences, thereby resulting in no considerable variation in the physicochemical characteristics (Wang *et al*., 2018). A similar lack of spatial variation was reported in the Bodo Creek, where physico-chemical parameters remained consistent across different stations (Vincent-Akpu *et al*., 2015).

Temporally, no significant variations were observed in temperature and pH levels. This stability suggests a consistent thermal and acidic/alkaline environment throughout the study period. Similar findings were reported in the Ikoli Creek, where temperature and pH remained relatively constant across seasons (Seiyaboh *et al*., 2016). July recorded the highest levels of DO and BOD, this elevation can be due to the rainy season. Rainfall can increase water flow and turbulence, promoting the mixing of oxygen-rich surface water (USEPA, 2024). The increased BOD might be due to the influx or resuspension of organic matter during the rainy seasons (Anhwange *et al*., 2012; Suratman *et al*., 2016). The results are similar with earlier observations in the Forcados River, where higher DO and BOD levels were noted during specific periods (Abija *et al*., 2018). In increased salinity in could result from reduced freshwater input during the dry season, leading to higher salt concentration (Ayers *et al*., 2017; Krishan *et al*., 2010). A similar trend hand been earlier observed in the Bonny/New Calabar River Estuary, where salinity levels were higher during the dry season due to decreased freshwater influx (Chindah *et al*., 2004).

Distribution of PAH in surface water, sediment and *C. amnicola*

The distribution of PAHs in surface water, sediment and *C. amnicola* was evaluated. The results showed that out of the 17 priority PAHs listed by agency of toxic substances and disease registry (ATSDR. 1995), 12 (including naphthalene which was not listed in ATSDR) excluding benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, dibenzo(a,h) anthracene, benzo(e)pyrene, and benzo(g,h,i) perylene were detected in Buguma Creek. Spatial and temporal variations were observed in the number of individual PAHs accumulated in each matrix (surface water, sediment, and *C. amnicola*), indicating contamination likely from both petrogenic (oil-related) and pyrogenic (combustion-related) sources (Oluwafunmilayo *et al*., 2019). PAHs in the aquatic environment can originate from anthropogenic activities such as industrial discharge, oil spills, and incomplete combustion of fossil fuels (Okedere and Elehinafe, 2022; Abdel-Shafy and Mansour, 2015), all of which are prominent in Buguma Creek. The findings of this study contrast with those reported by Anyakora *et al*. (2005) and Akinsanya *et al*. (2018), who detected 16 PAHs in Siokolo Fishing

Settlement and two sites (Ibasa and Ilase creeks) in Snake Island, Lagos, respectively. Similarly, Okpashi *et al.* (2017) identified 17 PAHs in the Qua Iboe River, while Ihunwo *et al*. (2019) and Ihunwo and Ibezim-Ezeani (2021) recorded 10 and 16 PAHs respectively in Woji Creek. Eight PAHs viz: benzo(a)anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo(a)pyrene, dibenzo[a,h]anthracene, indeno[1,2,3-cd]pyrene, and benzo[g,h,i]perylene, are considered possible carcinogens and mutagens (Menzie and Potocki, 1992). In particular, benzo(a)pyrene has been identified as highly carcinogenic (Moorthy *et al*., 2015; David and Gooderham, 2018). Of these eight PAHs, benzo(a)anthracene, benzo[b]fluoranthene, and benzo(a)pyrene were found in both surface water and sediment, while chrysene and benzo[b]fluoranthene were detected in *C. amnicola*, suggesting that Buguma Creek has carcinogenic and mutagenic potential. The findings are in line with Ihunwo and Ibezim-Ezeani (2021) and Akinsanya *et al*. (2018) who reported carcinogenic PAHs in Woji creek and snake Island, Lagos respectively.

Also, the findings of the present revealed distinct temporal and spatial variations in the concentrations of low molecular weight (LMW) and high molecular weight (HMW) PAHs within the matrices. Across all stations and months, the total concentration of LMW PAHs (2-3 rings; petrogenic origin) in surface water was consistently higher than that of HMW PAHs (4-6 rings; pyrogenic origin). This pattern suggested significant inputs from petrogenic sources, such as oil spills and urban runoff, introducing lighter PAH compounds into the aquatic environment. This might be the case of Buguma creek as there is continuous landing of products from nearby artisanal refineries (bunkering activities) as well as runoff from contaminated mangrove swamps or infiltrations from nearby oil installations. The LMW PAHs are more water soluble hence remain dissolved in the water column. Similar observations have been reported in other studies, indicating that LMW PAHs are more prevalent in surface waters due to their solubility and petrogenic sources (Ihunwo and Ibezim-Ezeani 2021; Lv *et al*., 2014).

Sediments exhibited higher concentrations of HMW PAHs compared to LMW PAHs in May and July, indicating a dominance of pyrogenic sources during these months. This could be attributed to atmospheric deposition from combustion processes such as emissions from artisanal refineries and waste dumps along the riverbank, which are common in the region. However, in September, LMW PAHs (9.30 mg/kg) exceeded HMW PAHs (1.06 mg/kg), possibly due to increased petrogenic inputs from runoff during the rainy season. These temporal shifts highlight the dynamic nature of PAH sources and deposition in the creek's sediments. Spatial analysis revealed that HMW PAHs consistently exceeded LMW PAHs at all sampled locations: Maryhood Bridge (HMW: 34.11 mg/kg; LMW: 16.28 mg/kg), Trans-Kalabari Highway (HMW: 29.82 mg/kg; LMW: 19.15 mg/kg), and Ido Canal (HMW: 37.83 mg/kg; LMW: 13.66 mg/kg). The predominance of HMW PAHs in sediments across these stations suggests ongoing contributions from pyrogenic sources, possibly linked to local artisanal refineries and waste dumps, as previously mentioned. The affinity of HMW PAHs to adsorb onto particulate matter could lead to their accumulation in sediments. Studies have shown that sediments act as sinks for HMW PAHs due to their hydrophobic nature and strong association with organic matter (Aigberua and Seiyaboh, 2021; Sun *et al.*, 2017; Zhang *et al.*, 2017).

In *C. amnicola*, PAH accumulation patterns varied over time. In July, HMW PAHs (21.35 mg/kg) were more concentrated in crab tissues than LMW PAHs (15.21 mg/kg), possibly due to increased environmental levels of HMW PAHs leading to higher uptake. Conversely, LMW PAHs were more abundant in *C. amnicola* in May (5.28 mg/kg) and September (3.11 mg/kg), reflecting their greater bioavailability in the water column during these months. The metabolic capacity of *C. amnicola* to process different PAH compounds may also influence these patterns. Research indicates that aquatic organisms (e.g., fish and invertebrates) bioaccumulate PAHs, with variations depending on the molecular weight of the compounds and environmental concentrations (Ihunwo and Ibezim-Ezeani, 2021; Ikenaka *et al*., 2008). Spatially, HMW PAHs were more prevalent in crabs from Maryhood Bridge and Trans-Kalabari Highway, whereas crabs from Ido Canal contained higher levels of LMW PAHs (10.62 mg/kg) than HMW PAHs (5.60 mg/kg). This distribution may reflect local differences in PAH sources and environmental conditions influencing PAH bioavailability and uptake. Factors such as proximity to pollution sources, water flow patterns, and sediment characteristics can affect the types and concentrations of PAHs to which aquatic organisms are exposed.

In this study, the order of PAH accumulation across matrices was sediment > water > *C. amnicola*. The highest accumulation was recorded in sediments, possibly due to increased mobility and bioavailability of PAHs through sorption to colloids. According to Ghandourah (2022), PAHs, particularly HMW PAHs, are relatively immobile in sediments because their non-polar structures inhibit dissolution in water. However, LMW PAHs are more soluble and become bioavailable in the water column. Additionally, the presence of organic colloids in water can increase PAH concentrations beyond their aqueous solubility, as PAHs adsorb onto these colloids, which are then transported through sediment pore spaces. Although previous studies have reported rapid PAH accumulation in aquatic organisms, often at higher concentrations than in their surrounding environment, this was not the case in the present study. *C. amnicola* exhibited the lowest PAHs accumulation (Tables 2 and 3), which is in line with the findings of Akinsanya *et al*. (2018), who reported low bioaccumulation and biomagnification potential in biota (benthos, fish and parasite).

Bioaccumulation Factor for *C. amnicola*

The transfer factor (BWAF and BSAF) in *C. amnicola* from the aquatic ecosystem was calculated (Kalfakakour and Akrida-Demertzi, 2000). BSAF and BWAF for most PAHs measured were <1, with BWAF showing higher values than BSAF indicating that accumulation of PAH by *C. amnicola* is mainly from water than from sediment. The implication of BSAF and BWAF values <1 for most of the measured PAHs in *C. amnicola* is that the bioaccumulation of these PAHs in the species from the aquatic ecosystem is relatively low (Olayinka *et al.,* 2019; Chris and Oghenetekevwe, 2022; Ukaogo *et al*., 2022; Saunders *et al*., 2022). This could be attributed to the crabs' metabolic capacity to biotransform and excrete PAHs thereby reducing their bioaccumulation potential. However, certain months showed BSAF (May) and BWAF

(September) values exceeding 1, indicating periods of increased bioavailability and uptake of PAHs. Factors such as seasonal variations, changes in environmental conditions, and differences in PAH sources could influence these fluctuations. Studies have shown that bioaccumulation factors can vary with the organism's exposure duration, metabolic rate, and the physicochemical properties of the specific PAHs (Ihunwo and Ibezim-Ezeani, 2021). For example, Although, *C. amnicola* exhibited some capacity of regulating PAH accumulation, their presence in the crabs' edible tissues poses potential health risks to consumers. Regular monitoring and assessment of PAH levels in aquatic ecosystems and biota are essential to mitigate adverse ecological and human health effects. Additionally, the higher values of BWAF compared to BSAF suggest that water is a more important source of PAHs for *C. amnicola* than sediment. This information could be used to develop strategies for reducing the input of PAHs into aquatic environments, particularly water sources, to further reduce the accumulation of these contaminants in aquatic organisms.

PAH Concentration in Surface Water, Sediment and *C. amnicola*

Research on PAHs concentration in the environment has received much attention due to their persistent and toxic properties (IARC, 1983; see reviews of Abdel- Shafy and Mansour, 2015 and Okedere and Elehinafe, 2022). PAH accumulate in sediments, aquatic pelagic and benthic food webs and pose a risk to human health (Malins *et al*., 1988; Baumard *et al*., 1998; Law and Klungsoyr, 2000). The uniform distribution of PAHs (for example in surface water: mean values were 8.19 ± 10.21 mg/L, 7.48 ± 8.78 mg/L, and 8.60 ± 10.50 mg/L for Maryhood bridge, Trans-Kalabari highway and Ido canal respectively) across the stations suggested widespread contamination, possibly due to the creek's hydrodynamics facilitating the even dispersal of pollutants. Similar findings have been reported in other Niger Delta creeks, where PAH contamination remains consistent across different sites due to diffuse pollution sources (Olawoyin et al., 2012; Agbozu and Opuene, 2009).

Temporally, PAHs concentration in all matrices (surface water, sediment and *C. amnicola*) followed the same trend, July > May > September. This trend indicated enhanced input from petroleum spills, industrial discharges, and atmospheric deposition, consistent with previous studies on PAH distribution in the Niger Delta (Anyakora *et al*., 2005). Increased runoff and anthropogenic activities during the rainy season increased PAHs concentration in the surface water, contributed to higher PAH deposition in sediments and increased PAH bioavailability to the crabs. Crabs, being benthic organisms, fed predominantly on detritus, crustaceans, pisces, algae filaments, diatoms and sand grains can accumulate PAHs primarily through direct sediment contact and contaminated food sources (Olayinka *et al*., 2019; Tongo *et al*., 2017; Arimoro and Idoro, 2007). This trend supported the previous observations where higher concentrations of pollutants (e.g., Total petroleum hydrocarbon and heavy metals) were recorded during the rainy season due to increased influx of runoff and anthropogenic activities (Anyanwu *et al.*, 2023).

CONCLUSION

In conclusions, Buguma Creek is polluted with PAHs from both pyrogenic and petrogenic sources, with petrogenic inputs dominating in surface water and pyrogenic sources in *C. amnicola*, while both contributed equally to sediment pollution. PAH partitioning followed the trend: sediment > water > *C. amnicola*, with concentrations in all matrices exceeding US EPA limits (<1000 ng/g for sediment, <1000 ng/L for water) by several magnitudes. The high PAH levels raise concerns about bioaccumulation and trophic transfer, as crabs are key food web components, posing risks to higher predators. The detection of benzo(a)anthracene, benzo[b]fluoranthene, and benzo(a)pyrene in surface water and sediment, along with chrysene and benzo[b]fluoranthene in *C. amnicola*, indicated potential carcinogenic and mutagenic risk. To mitigate anthropogenic impacts and ensure ecological sustainability of the creek, regular monitoring and pollution control measures are essential.

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