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Genotoxicity of Polycyclic Aromatic Hydrocarbons in Processed Seafood from Coastal Communities in the Niger Delta, Nigeria: Implication of Processing on Public Health

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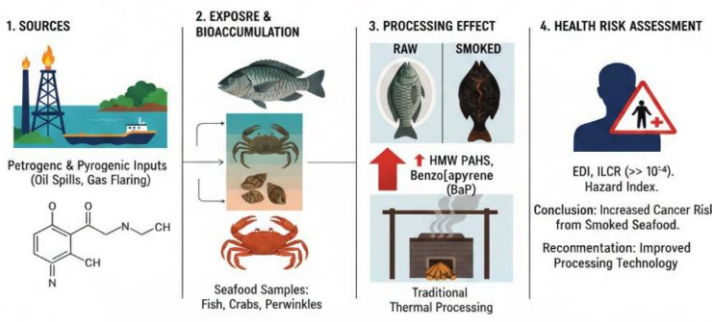
ABSTRACT

The persistence, potential for bioaccumulation, mutagenicity and carcinogenicity of polycyclic aromatic hydrocarbons (PAHs) make them environmental hazards of concern. This study evaluated the human health risks of PAHs in processed seafood obtained from the waters of coastal communities of the Niger Delta, Nigeria. Twenty-four (24) Oreochromis niloticus and twenty-four (24) mugil cephalus samples were collected from four locations and analysed using gas chromatography with flame ionisation detection (GC-FID). Human health risk assessment was conducted using two approaches: margin of exposure (MOE) for BaP, PAH4, and PAH8, and cancer risk (CR) based on the total of 16 priority USEPA PAHs. The concentrations of 16PAHs varied across species and sites. Higher values were observed in smoked samples, indicating that thermal processing significantly enhanced PAH formation. Diagnostic ratios of Ant/(Ant+Phe), Flt/(Flt+Pyr), BaA/(BaA+Chr), and ΣLMW/ΣHMW revealed mixed pyrogenic and petrogenic sources, reflecting contributions from both oil-related discharges and combustion during traditional fish smoking. Mean MOE values were markedly below the European Food Safety Authority (EFSA) benchmark of 10,000. The children were more vulnerable than adults due to higher exposure per their body weight. Estimated CR values, particularly at Akiama and Orosikiri, exceeded the USEPA acceptable range (10^{-6}–10^{-4}), indicating potential carcinogenic concern from chronic seafood consumption. The study concludes that PAH contamination in seafood from the Niger Delta arises from both environmental pollution and processing activities, posing a tangible public health risk. Adoption of cleaner seafood-smoking technologies and stricter environmental regulations is recommended to safeguard consumer health and ensure seafood safety in the region.

Keywords: *Polycyclic Aromatic Hydrocarbons, Processed Seafood, Margin of Exposure, Cancer Risk, Niger Delta*

Graphical Abstract



Introduction

The oil and gas industry, which is significant in Nigeria's coastal areas, is the country's largest industry. This dominance has its resultant impact on the aquatic environment, especially in relation to the potential enhancement of environmental pollutants across the coastal region. Because of their proximity to the source, they receive the greatest chemical contamination discharges. Thus, it could be said that the industries potentially increase organic and inorganic contaminant concentration across the Niger Delta region [1, 2, 3].

Seafood is a key part of the diet of coastal residents. Seafood, especially fish, a low-fat, high-quality protein, is a vital part of a healthy diet and an important source of omega-3 fatty acids, especially eicosapentaenoic acid (EPA) and DHA (docosahexaenoic acid). In addition to macro and micronutrients such as potassium, phosphorus, calcium, iron, zinc, iodine, and magnesium, they also contain high amounts of cholecalciferol (vitamin D) and riboflavin (vitamin B2). Seafood has many health benefits, including reducing the risk of cardiovascular diseases, triacylglycerol levels, and blood pressure, slowing plaque growth in arteries, improving cardiovascular risk factors and protecting against inflammation [4, 5]. It aids brain development in infants, vision and nerves during pregnancy, and decreases the risk of depression, arthritis, Alzheimer's disease and dementia. While seafood has nutritional benefits, it also has potential risks. Seafood can take in harmful chemicals like polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), heavy metals (HMs) from the water and the food they eat. Seafood and foods can be readily contaminated by organic compounds like polycyclic aromatic hydrocarbons (PAHs) [6, 7, 8].

The presence of PAHs in seafood is a major health concern due to their well-known carcinogenic, teratogenic, mutagenic and persistence qualities [6]. In addition to harming human health, toxic substances can have an impact on marine organisms' survival, reproduction, and productivity. Coastal monitoring programs are useful for evaluating the existing condition of coastal habitats, detecting possible sources of contamination to stop future issues, and figuring out trends in contaminants over time and location [9]. Polycyclic aromatic hydrocarbons (PAHs), belong to a class of semi-volatile compounds containing two or more condensed aromatic rings arranged linearly, angularly, or in clusters. They are significant organic contaminants found in food, the environment, and coastal seas. Fossil fuel combustion, sewage and sewage sludge discharge, industrial and mining effluents, fertiliser and pesticide residues, and other sources can all release PAH pollutants into the environment [10].

Usually, food represents the chief source of exposure. The Scientific Committee on Food (SCF) advised the monitoring of 15 PAHs in 2002 after concluding that 15 of the 33 PAHs under investigation exhibit mutagenic/genotoxic effects and, except for benzo[ghi]perylene, have also demonstrated definite carcinogenic effects. SCF has emphasised the necessity of ongoing data collection on various PAH levels in foods for risk assessments [11]. Three major sources can produce PAHs: petroleum (petrogenic), combustion (pyrogenic), and natural (biogenic) [12]. Pyrogenic PAHs are produced when organic matter is processed at high temperatures, whereas biogenic PAHs are produced by living things.

Natural processes like petrol leaks and seepage from fossil fuels allow petrogenic PAHs to enter the environment [13]. Numerous PAHs are genotoxic and mutagenic, and they can create DNA adducts in both living things and in laboratory settings. [14]. PAHs can cause cancer as well as other negative effects, such as changed thymus weight and serum immunoglobulin in rats, decreased ovarian follicles and ovary weight, and neurobehavioral abnormalities in young animals [15, 16].

According to the European Food Safety Authority, it is more appropriate to use PAH4 (the sum of benzo[a]anthracene (BaA), chrysene (Chr), benzo[a]pyrene (BaP), and benzo[b]fluoranthene (BbF)) or PAH8 which is the sum of PAH4 plus benzo[k]fluoranthene (BkF), indeno[1,2,3-cd]pyrene (InP), dibenz[a,h]anthracene (DBA) and benzo[g,h,i]perylene (BgP) than using exclusively BaP level as indicator for risk assessment in seafood [17]. The principal route of human exposure to PAHs is consumption, and the risk of exposure to PAHs in aquatic products has increased due to increasing seafood consumption and human degradation of the marine environment [16, 18, 19]. Seafood is now the main dietary source of PAH exposure, according to additional research [20]. The levels of the 16 priority USEPA PAHs in processed seafood from coastal waters in Nigeria's Niger Delta were examined in this study, along with the potential health risks associated with consuming seafood by the local populace.

Materials and Methods

Table 1: Study location and Seafood samples collected

Sampling site	Longitude	Latitude	Activities
Orosikiri Rivers State	N 7.1708	E 4.437	Waterfronts serve as places for fishing activities, receive raw or untreated industrial effluents from oil and gas companies around that vicinity, and discharges from oil and gas tankers, barges, and passenger boats.
Akiama Rivers State	N 7.1984	E 4.4356	
Orerokpe Delta State	N 5.63473	E 5.89968	These waterfronts along Erwin Creek in Warri South serve as places for fishing activities, as well as receiving raw or untreated industrial effluents from oil and gas companies and discharges from small oil and gas tankers, barges, and passenger boats
Osubi Delta State	N 5.59601	E 5.85001	
S/N	Local name	Scientific Name	English Name of seafood
1	Atagbala	*Oreochromis niloticus*	Tilapia
2	Ndege	*Mugil cephalu*	Mullet

Sample Collection

Seafood (fish and benthic organisms) samples were obtained straight from four different sampling locations in the river and the creeks using a fishing net and hand-selected, respectively, by the fishermen. A total of one hundred and twenty-four (124) samples of different species of seafood were collected from the four sampling locations of Orosikiri and Akiama creeks in Bonny Local Government Area, Rivers State, and Orerokpe and Osubi creeks in Okpe Local Government Area, Delta State, Nigeria for this study. Six tilapia, six mullet, six giant land crab and six Africana rainbow crab samples were harvested from Akiama and Orosikiri creeks. Nine tilapia, nine mullet, six giant land crab, six Africana rainbow crab and forty periwinkle samples were collected from Orerokpe and Osubi creeks. All the samples collected were properly labelled and kept cold in an icebox after being wrapped in sterile plastic bags. The labelled samples were transferred to the laboratory and stored in a refrigerator at 4 °C to avoid microbial action until further use.

Four sets of three (3) tilapia and three mullets were taken from samples of the four sampling locations and smoked. Another two sets of three tilapia and three mullets from Orerokpe and Osubi samples were sun-dried. The gas chromatography-flame ionisation detector (GC-FID) device was used to measure PAH levels, and PAH standards were used for calibration. For every seafood sample, analyses were performed in triplicate.

Digestion of Samples

PAHs Extraction, Clean-up, and Analysis

PAHs were determined in seafood samples using conventional USEPA organic pollutant analysis techniques. The analysis involved three major stages: extraction, clean-up and gas chromatographic determination [22, 23].

Samples Pre-Treatment & Extraction

Approximately 10 g of homogenised air-dried sample was weighed into a pre-cleaned amber glass bottle to prevent photodegradation of PAHs.

Anhydrous sodium sulfate (Na_2SO_4) was added to remove residual moisture, and the sample was thoroughly mixed. A surrogate standard solution (1-chlorooctadecane, 300 µg/mL) was spiked into each sample to monitor extraction efficiency and recovery. Thereafter, 30 mL of dichloromethane (DCM) was added as the extraction solvent. The bottle was tightly capped with a Teflon-lined lid and agitated on a mechanical shaker for 5 to 6 hours at room temperature to facilitate efficient solvent-sample interaction. To ease phase separation, the mixture was shaken and then left undisturbed for approximately an hour. Whatman No. 1 filter paper (110 mm) was used to carefully filter the supernatant (organic layer) into a beaker that had been cleaned and rinsed with solvent. After that, the filtration was gently evaporated in a fume cupboard to concentrate it to about 1 mL under a stream of nitrogen gas to minimise loss of volatile analytes.

Sample Purification

The concentrated sample was subjected to a clean-up to remove co-extracted matrix interferences. A glass column with a tiny plug of glass wool at the base and 10 g of activated silica gel (previously heated at 130 °C for 16 hours and cooled in a desiccator) was used for the cleanup process. A 2 cm coating of anhydrous sodium sulphate was encrusted on top of the silica gel to prevent moisture from entering it. The column was preconditioned with 10 mL of n-pentane before sample application. The concentrated extract was mixed with a small volume of cyclohexane and quantitatively transferred onto the column. Sample elution was carried out using 20 mL of n-pentane to remove aliphatic hydrocarbons, followed by 10 mL of dichloromethane (DCM) to elute the target PAHs. The eluate was collected in a clean beaker and concentrated to a final volume of 1 mL by ambient vaporization in a fume cupboard.

GC-FID Analysis

PAHs were separated and quantified using an Agilent 6890N Gas Chromatograph equipped with a Flame Ionization Detector (GC–FID) in compliance with USEPA [24]. The Flame Ionization Detector (FID) measured the PAH compounds based on their ionization response, and quantification was achieved through external calibration using certified PAH standards. The concentrations of each PAH in the samples were reported in milligrams per kilogram (mg/kg) on a dry-weight basis.

Quality Control

Method blanks, duplicate samples, and matrix spikes were used to guarantee analytical quality assurance and control. The extraction efficiency was evaluated using surrogate recoveries [25], and acceptable recoveries varied from 70% to 120%. Calibration curves were prepared using certified PAH standards, and correlation coefficients ($R^2 \geq 0.995$) were maintained for all target analytes. The described procedure ensured accurate, reproducible, and reliable quantification of PAHs in sediment and seafood matrices.

Dietary exposure and health risk estimations

Exposure Assessment

Depending on the levels of contamination found in this investigation, daily seafood consumption was used to measure consumer exposure to PAH.

Estimation of toxic potency

The overall PAH concentration is expressed as BaP equivalent concentration (BEC) to show the potency since BaP is assumed to be the most potent carcinogenic PAH [24]. The BEC was calculated as the sum of the BaP_{eqi} values for PAH congeners. Four PAHs (PAH4) are defined as the sum of BaP, Chr, BaA, and BbF, while eight PAHs (PAH8) are defined as the sum of BaP, Chr, BaA, BbF, BkF, DBA, BgP, and InP.

Based on each PAH congener concentration (C_i) in the sample, the BEC value was determined. This was multiplied by its corresponding toxic equivalent factor (TEF_i).

$BEC_i = C_i \times TEF_i$ (1)

$BEC = \sum_{i=1}^{n} BEC_I$ (2)

The toxic equivalent factors (TEFs) of PAHs used in the calculation, except for DBA, were as reported in [26].

Exposure assessment

The estimated daily intake, EDI, is calculated by the equation (3)

$$EDI = \frac{BEC \times IR \times EF \times ED}{BW \times At_{nc}} \quad (3)$$

Where EDI is the estimated daily intake (mg/kg-day); BEC is the BaP equivalent concentration of PAH4, PAH8 and 16PAHs (mg BEC/kg) for each seafood; EF is the frequency of exposure (350 days/year) [27, 28]. Two age groups—children (0–17 years old) and adults (>18 years old)—were used to estimate exposure. The ED is exposure time expressed in years (Children: ED = 6, adults: ED = 30). IR is the average amount (kg/day) of seafood consumed by an individual for each age group (fish - children: 0.0273 kg/day and adults: 0.0364 kg/day [29]; shellfish - children: 0.012 kg/day and adults: 0.016 kg/day [30], and BW is the consumer's average body weight (kg) (children: 21.1 years and adults: 63 kg [31].

Risk Characterization

Health risk characterization was performed using two complementary approaches: the margin of exposure (MOE) for BaP, PAH4, and PAH8 [32], and the cancer risk (CR) for the total 16 PAHs (TPAHs) [24, 31].

The cancer risk is estimated using equation (4):

$$CR = \frac{BEC \times CSF \times 3\sqrt{(BW/70)} \times IR \times EF \times ED}{BW \times At_c} \quad (4)$$

where CR stands for cancer risk (unitless); CSF is known as the cancer slope factor (kg-day/mg) of BaP (7.3 kg-day/mg) and AT_c is the averaging time for carcinogenic (365 x LT) (LT: life span; 54.7 years [33].

Margin of exposur

$$MOE = \frac{BMDL_{10}}{EDI} \quad (5)$$

where MOE is the margin of exposure (dimensionless); EDI is the estimated daily intake (mg/kg-bw/day); $BMDL_{10}$ is the benchmark dose lower limit, $BMDL_{10}$ of PAH4 = 0.34 mg/kg bw/day and $BMDL_{10}$ of PAH8 = 0.49 mg/kg-bw/day [32]. An MOE value lower than 10,000 indicates a potential health concern (i.e. serious adverse health effects), requiring risk management, while an MOE of 10,000 or higher suggests low concern for human health, that is, no significant toxic effects to humans [31].

Statistical Analysis

All data were statistically analysed using IBM SPSS software (version 23). The statistically significant difference was ascertained using a one-way ANOVA. At p-levels of less than 0.05, or a 95% confidence level, the findings were deemed meaningful. For additional statistical analysis and computations, Microsoft Excel was utilised.

Results and Discussion

Concentration of PAHs in the raw and processed seafood

The compositions of the PAHs in Seafood from the various sample locations are represented in Figure 1. No single seafood has a complete 16 USEPA PAHs. Low-molecular-weight PAHs make up the majority of them. The smoked *Oreochromis niloticus* (SON) from Akiama creek has the highest total PAHs concentration of 194.49 mg/kg, followed by SON from Orosikiri creek (123.29 mg/kg), while sun-dried *Mugil cephalus* (DMC) has the lowest TPAHs concentration of 6.93 mg/kg.

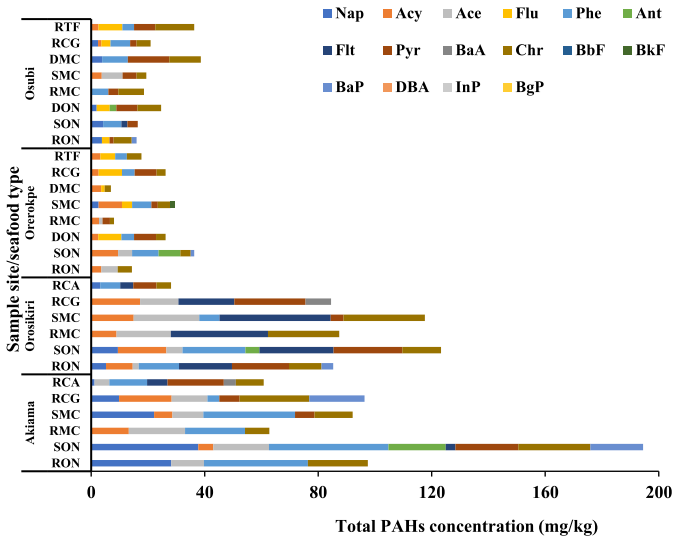


Figure 1: The total PAHs and distribution pattern in the raw, smoked and dried seafood samples from different sites, Niger Delta (seafood starting with R, S and D denote raw, smoked and sun-dried, respectively)

RON: Raw *Oreochromis niloticus*, SON: Smoked *Oreochromis niloticus*, RMC: Raw *Mugil cephalus*, SMC: Smoked *Mugil cephalus*, RCG: Raw *Cardisoma guanhumi*, RCA: Raw *Cardisoma armatum*, DON: Dried *Oreochromis niloticus*, DOM: Dried *Oreochromis niloticus*, RTF: Raw *Tympanotonus fuscatus*

Estimated Daily Intake

Equations (1) and (2) were used to determine the estimated daily intakes (EDIs) of PAHs resulting from seafood consumption for each population group based on the BEC values, as indicated in Table 2. The results of BaP equivalent concentrations (BECs) and estimated daily intakes (EDIs) of PAHs (mg BEC/kg-bw/day) are presented in Table 1.

Table 1: BaP equivalent concentration (BEC) and estimated daily intake (EDI) of BaP, PAH4, PAH8 and TPAHs

Sample site	Seafood type	BEC (mg kg−1)				EDI (mg-BEC kg^{-1} day^{-1})							
						BaP		PAH4		PAH8		TPAHs	
		BaP	PAH4	PAH8	TPAHs	Child	Adult	Child	Adult	Child	Adult	Child	Adult
Akiama													
	RON	4.63	4.68	4.68	4.69	5.75E-03	2.57E-03	5.81E-03	2.59E-03	5.81E-03	2.59E-03	5.83E-03	1.43E-03
	SON	6.52	6.60	6.60	6.69	8.10E-03	3.62E-03	8.23E-03	3.66E-03	2.74E-03	3.66E-03	8.31E-03	2.03E-03
	RMC	0.00	0.07	0.07	0.08	0.00E+00	0.00E+00	8.16E-05	3.63E-05	2.72E-05	3.63E-05	9.89E-05	2.42E-05
	SMC	0.00	0.08	0.08	0.12	0.00E+00	0.00E+00	1.05E-04	4.68E-05	3.51E-05	4.68E-05	1.49E-04	3.65E-05
Orosikiri													
	RON	4.14	4.19	4.19	4.24	5.14E-03	2.29E-03	5.23E-03	2.33E-03	1.74E-03	2.33E-03	5.26E-03	1.29E-03
	SON	4.94	5.01	5.01	5.12	6.14E-03	2.74E-03	6.25E-03	2.78E-03	2.08E-03	2.78E-03	6.35E-03	1.56E-03
	RMC	8.15	0.08	0.08	0.11	1.01E-02	4.52E-03			3.39E-05	4.52E-05	1.34E-04	3.28E-05
	SMC	0.00	0.12	0.12	0.16	0.00E+00	0.00E+00	1.53E-04	6.80E-05	5.10E-05	6.80E-05	2.00E-04	4.89E-05
Orerokpe													
	RON	0.00	0.05	0.05	0.06	0.00E+00	0.00E+00	6.19E-05	2.75E-05	2.06E-05	2.75E-05	7.72E-05	1.89E-05
	SON	1.19	1.23	1.23	1.34	1.48E-03	6.62E-04	1.53E-03	6.82E-04	5.11E-04	6.82E-04	1.66E-03	4.07E-04
	DON	0.00	0.03	0.03	0.08	0.00E+00	0.00E+00	4.23E-05	1.88E-05	1.41E-05	1.88E-05	9.77E-05	2.39E-05
	RMC	0.00	0.01	0.01	0.02	0.00E+00	0.00E+00	1.79E-05	7.95E-06	5.96E-06	7.95E-06	2.60E-05	6.37E-06
	SMC	0.34	0.38	0.53	0.55	4.22E-04	1.89E-04	4.80E-04	2.13E-04	2.20E-04	2.93E-04	6.88E-04	1.69E-04
	DMC	0.00	0.02	0.02	0.03	0.00E+00	0.00E+00	2.70E-05	1.20E-05	8.98E-06	1.20E-05	3.58E-05	8.77E-06
Osubi													
	RON	1.68	1.74	1.74	1.75	2.08E-03	9.31E-04	2.17E-03	9.66E-04	7.25E-04	9.66E-04	2.17E-03	5.32E-04
	SON	2.11	2.19	2.19	2.21	2.62E-03	1.17E-03	2.74E-03	1.22E-03	9.13E-04	1.22E-03	2.75E-03	6.72E-04
	DON	1.68	1.75	1.75	1.78	2.08E-03	9.31E-04	2.18E-03	9.71E-04	7.28E-04	9.71E-04	2.22E-03	5.43E-04
	RMC	0.34	0.43	3.01	3.02	4.22E-04	1.89E-04	5.37E-04	2.39E-04	1.25E-03	1.67E-03	3.76E-03	9.20E-04
	SMC	1.79	1.90	5.11	5.14	2.22E-03	9.93E-04	2.37E-03	1.05E-03	2.13E-03	2.83E-03	6.38E-03	1.56E-03
	DMC	0.34	0.42	3.00	3.03	4.22E-04	1.89E-04	5.29E-04	2.35E-04	1.25E-03	1.67E-03	3.76E-03	9.22E-04

RON: Raw Oreochromis niloticus, SON: Smoked Oreochromis niloticus, RMC: Raw Mugil cephalus, SMC: Smoked Mugil cephalus, DON: Dried Oreochromis niloticus, DOM: Dried Oreochromis niloticus

Risk Assessment

The health risk due to PAHs exposure in seafood via consumption for the study region was assessed using the margin of exposure (MOE) for BaP, PAH4 and PAH8, and cancer risk (CR) for 16 priority USEPA PAHs (TPAHs). The descriptive statistics of MOE and CR for the two age groups are shown in Table 2.

Table 2: Descriptive statistical analysis of margin of exposure (MOE) (BaP, PAH4 and PAH8) and cancer risk (TPAH) in seafood from the coastal communities in the Niger Delta, Nigeria

Sample site	Statistical parameter	Margin of exposure, MOE						Cancer risk, CR	
		BaP		PAH4		PAH8		TPAHs	
		Child	Adult	Child	Adult	Child	Adult	Child	Adult
Akiama	Min	3.61E+00	8.08E+00	1.72E+01	3.86E+01	3.07E+01	5.56E+01	9.31E-05	2.99E-04
	Max	6.59E+00	1.47E+01	3.19E+03	7.18E+03	1.38E+04	1.03E+04	1.08E-02	3.48E-02
	Mean	4.89E+00	1.09E+01	1.08E+03	2.43E+03	4.66E+03	3.50E+03	4.27E-03	1.37E-02
Orosikiri	Min	2.24E+00	5.02E+00	5.37E+01	1.21E+02	2.32E+02	1.74E+02	2.18E-05	7.00E-05
	Max	1.36E+01	3.05E+01	1.21E+04	2.72E+04	5.22E+04	3.92E+04	3.49E-03	1.12E-02
	Mean	9.09E+00	2.04E+01	2.77E+03	6.23E+03	1.08E+04	8.07E+03	1.19E-03	3.82E-03
Orerokpe	Min	4.72E+01	1.06E+02	2.22E+02	4.99E+02	9.58E+02	7.19E+02	1.40E-05	4.49E-05
	Max	1.66E+02	3.71E+02	1.93E+04	4.35E+04	8.36E+04	6.27E+04	8.92E-04	2.87E-03
	Mean	1.07E+02	2.39E+02	9.69E+03	2.18E+04	4.18E+04	3.13E+04	1.78E-04	5.72E-04
Osubi	Min	2.67E+01	5.98E+01	1.24E+02	2.79E+02	2.30E+02	1.73E+02	1.92E-05	1.40E-04
	Max	1.66E+02	3.71E+02	1.26E+04	2.82E+04	5.43E+04	4.07E+04	3.43E-03	1.10E-02
	Mean	7.62E+01	1.71E+02	2.37E+03	5.33E+03	9.60E+03	7.20E+03	1.42E-03	4.60E-03

Table 3: Molecular Diagnostic ratio for PAHS in seafood from Akiama and Orosikiri

Diagnostic Ratio	Petrogenic	Pyrogenic	This study											
			Akiama						Orosikiri					
			RON	SON	RMC	SMC	RCG	RCA	RON	SON	RMC	SMC	RCG	RCA
Ant/(Ant+Phe)	< 0.1	> 0.1	0.000	0.339	0.000	0.000	0.000	0.000	0.000	0.329	0.000	0.000	0.000	0.000
Flt/(Flt+Pyr)	< 0.4	> 0.4	0.000	0.296	0.000	0.000	0.000	0.421	0.466	0.518	1.000	0.769	0.601	0.360
BaA/(BaA+Chr)	< 0.200	> 0.350	0.000	0.000	0.000	0.000	0.191	0.300	0.000	0.000	0.000	0.000	1.000	0.000
ΣLMW/ΣHMW	> 1	< 1	1.673	1.334	2.171	1.889	0.916	0.506	0.802	1.007	0.876	0.583	0.650	0.574
	Fuel combustion	Grass/wood combustion	Petrogenic	pyrogenic/petrogenic	Petrogenic	Petrogenic	Pyrogenic	mixture of petrogenic and pyrogenic	Pyrogenic	Pyrogenic	Pyrogenic	Pyrogenic	Pyrogenic	pyrogenic/petrogenic

Ant (anthracene), Phe (phenanthrene), Flt (fluoranthene), Pyr (pyrene), BaA (Benzo [a] anthracene), Chr (chrysene), ∑LMW (sum of low molecular PAHs), and ∑HMW (sum of high molecular PAHs).

Table 4: Molecular Diagnostic ratio for PAHS in seafood from Orerokpe and Osubi

Diagnostic Ratio	Petrogenic	Pyrogenic	This study															
			Orerokpe								Osubi							
			RON	SON	DON	RMC	SMC	DOM	RCG	RTF	RON	SON	DON	RMC	SMC	DOM	RCG	RTF
Ant/(Ant+Phe)	< 0.1	> 0.1	0.000	0.453	0.425	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.000	0.000	0.000	0.000	0.000	0.000
Flt/(Flt+Pyr)	< 0.4	> 0.4	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.370	0.000	0.000	0.000	0.000	0.000	0.000
BaA/(BaA+Chr)	< 0.200	> 0.350	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
ΣLMW/ΣHMW	> 1	< 1	2.527	4.699	2.704	1.000	2.525	1.034	1.397	2.456	0.684	0.654	0.939	0.641	1.085	0.484	1.897	0.713
	Fuel combustion tion	Grass/wood combustion	Petrogenic	pyrogenic/petrogenic	pyrogenic/petrogenic	Petrogenic	Petrogenic	Petrogenic	Petrogenic	Petrogenic	Pyrogenic	pyrogenic/petrogenic	Pyrogenic	Pyrogenic	Petrogenic	Pyrogenic	Petrogenic	Pyrogenic

Ant (anthracene), Phe (phenanthrene), Flt (fluoranthene), Pyr (pyrene), BaA (Benzo [a] anthracene), Chr (chrysene), ∑LMW (sum of low molecular PAHs), and ∑HMW (sum of high molecular PAHs).

Molecular Diagnostic Ratios of PAHs in Seafood

The molecular diagnostic ratios (MDRs) presented in Tables 3 and 4 provide valuable insight into the possible sources and transformation pathways of PAHs detected in seafood from the studied coastal communities. Ratios such as Ant/(Ant + Phe), Flt/(Flt + Pyr), BaA/(BaA + Chr), and ΣLMW/ΣHMW were employed in this study to determine the sources and to differentiate between petrogenic (originating from petroleum inputs) and pyrogenic (derived from incomplete combustion) sources [34, 35]. The results revealed a mixture of petrogenic and pyrogenic signatures across the seafood species and sampling sites. Assessments of Ant/(Ant + Phe) and ΣLMW/ΣHMW > 1 in some seafood samples indicated petrogenic influence, likely from crude oil and industrial discharges characteristic of Niger Delta waters. Conversely, Flt/(Flt + Pyr) and BaA/(BaA + Chr) > 0.4 in other seafood samples suggested pyrogenic inputs associated with biomass and fossil fuel combustion, including traditional fish-smoking processes.

The coexistence of both source signatures implies that PAH contamination in the studied seafood is multifactorial, resulting from a combination of oil-related pollution and thermal degradation during processing. The dominance of HMW PAHs in many samples further supports the contribution of pyrogenic processes, as HMW PAHs are typically formed at higher combustion temperatures. These findings are consistent with those of Ofosu et al. [13] and Habibullah-Al-Mamun et al. [20], who identified mixed PAH origins in aquatic biota from oil-impacted environments. Overall, the diagnostic ratio results highlight the dual impact of industrial emissions and traditional smoking methods on PAH accumulation in seafood, reinforcing the need for integrated source control and improved processing hygiene practices in the Niger Delta coastal communities.

Human Health Risk Assessment of PAHs in Raw and Processed Seafood

Estimated Daily Intake of PAHs in Seafood

The estimated daily intakes of PAHs were determined using the measured concentrations of PAHs in seafood and the exposure parameters defined above. Toxic potency was expressed in terms of Benzo[a]pyrene equivalent concentration (BEC) using established toxic equivalency factors [30, 31]. The EDI values were computed for two age groups (children and adults) according to Equation (3) in the methodology, incorporating exposure duration, frequency, and body weight.

The EDIs for PAH4, PAH8 and TPAHs are presented in Table 1. The EDI values varied markedly across sampling sites and seafood species. The EDI values of PAH4 for both children and adults followed the descending order: SON > RON > SMC > RMC, SON > RON > SMC, SON > SMC > RON > DON > DMC > RMC and SON > SMC > RON = DON > RMC > DMC for Akiama, Orosikiri, Orerokpe and Osubi, respectively. The EDI values of TPAHs followed the same trend as that of EDI values of PAH4/PAH8 for Akiama and Orosikiri, SON > SMC > RON > DON > DMC > RMC, and SON > SMC > DON > RON > RMC > DMC for Orerokpe and Osubi, respectively.

The higher values occurred in seafood collected from Akiama, followed by Orosikiri, while relatively lower levels were recorded in samples from Orerokpe and Osubi. For instance, the EDI in raw *O. niloticus* (RON) from Akiama was 5.75×10^{-3} mg-BaP/kg-bw/day for children and 2.57×10^{-3} mg-BaP/kg-bw/day for adults. In smoked *O. niloticus* (SON) from the same sampling location, the EDI increased to 8.10×10^{-3} mg/kg-bw/day (children) and 3.62×10^{-3} mg/kg-bw/day (adults), confirming the effect of processing (smoking) on PAH accumulation. A similar pattern was observed for *Mugil cephalus*, where processed samples exhibited higher PAH burdens than their raw counterparts. The general trend observed across all locations followed the order: Smoked > Raw > Dried seafood, indicating that thermal processing significantly enhanced the formation of genotoxic PAHs. This agrees with the findings from EFSA [32] and Alomirah et al. [36], who reported elevated PAH concentrations in smoked fish products. The higher exposure levels observed in children are attributable to their lower body weight and developing physiological systems, which increase their vulnerability to PAH-related genotoxicity.

Margin of Exposure (MOE)

The Benchmark dose lower confidence limits ($BMDL_10$) for BaP, PAH4 and PAH8 used for the computation of the MOE in this study are 0.07, 0.34, and 0.49 mg/kg-bw/day, respectively [31]. Margin of exposure (MOE) serves as an inverse indicator of risk - lower values signify greater potential concern. The results (Table 3) revealed that children's MOE values were consistently lower than adults', reflecting higher exposure per unit body weight. The lowest MOE values were observed at Akiama and Orosikiri, with mean MOE (BaP) values of 4.89×10^0 and 9.09×10^0, respectively. These values are far below the conservative reference level of 10,000 suggested by EFSA [32], indicating a potential genotoxic risk. In contrast, samples from Orerokpe and Osubi recorded higher MOE values (up to 10^2–10^3), suggesting comparatively lower but still notable concern.

Cancer Risk (CR)

The cancer risk (CR) associated with exposure to total PAHs was estimated using the cancer slope factor (SF = 7.3 kg-day/mg) for BaP equivalents [37, 38]. Acceptable risk limits generally range between 1×10^{-6} and 1×10^{-4} [24]. The calculated CR values in this study exceeded these limits in most cases. Specifically, mean CR values ranged from 4.27×10^{-3} (children) to 1.37×10^{-2} (adults) at Akiama, and from 1.19×10^{-3} (children) to 3.82×10^{-3} (adults) at Orosikiri. Even at Orerokpe and Osubi, where lower concentrations were found, mean CR values of 1.78×10^{-4} and 1.42×10^{-3} for children and adults, respectively were still above the negligible risk threshold. The higher CR values, reaching 3.48×10^{-2} (adults) and 1.08×10^{-2} (children) at Akiama, suggest a potentially significant carcinogenic risk correlated to chronic consumption of contaminated seafood.

Overall, the findings demonstrate that seafood from Akiama and Orosikiri presents the greatest potential health risk, reflecting the influence of intense oil and gas activities and pyrogenic contamination in these areas. The elevated PAH concentrations, low MOE values, and high cancer risk estimates suggest that long-term consumption of such seafood could contribute to increased carcinogenic and genotoxic exposure to the local population. The pattern of higher PAH levels in smoked products further implicates local processing methods as an additional exposure pathway. Given the dietary dependence on seafood in these communities, these results underscore the public health relevance of continuous environmental monitoring, cleaner processing practices, and regulatory intervention to mitigate PAH exposure risks.

Conclusion

This study demonstrated that seafood from coastal waters in the Niger Delta is contaminated with varying concentrations of PAHs, originating from both petrogenic and pyrogenic sources.

Elevated PAH contents in smoked samples compared with raw and dried ones indicate that thermal processing significantly enhances PAH formation, while proximity to oil and gas operations contributes to environmental accumulation. The results highlight that seafood from Akiama and Orosikiri contained the highest PAH levels, reflecting the influence of industrial discharges and traditional smoking practices in these areas.

The human health risk assessment revealed that both adults and children are chronically exposed to genotoxic and carcinogenic PAHs through seafood consumption, with children being more vulnerable due to lower body weight. The calculated margin of exposure (MOE) values were well below the EFSA safety threshold, and cancer risk (CR) estimates in several cases exceeded the USEPA acceptable range (10^{-6}–10^{-4}), indicating potential public health concern. These results emphasise the critical need for ongoing seafood PAH monitoring, promotion of cleaner processing methods, and stricter environmental control of effluent discharges to protect the health and livelihoods of Niger Delta coastal populations.

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Conflict of Interest

Regarding this article, the authors state that they have no conflicts of interest.

CRediT author statement

BMOO: Conceptualisation, Resource, Writing – review & editing of manuscript, validation, Supervision. IJG: Data Curation, Investigation, data analysis and drafting of manuscript; TGA: Data Curation, data analysis; DCB: Supervision.

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