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Levels and Associated Human Health Risks of Polycyclic Aromatic Hydrocarbons (PAHs) in Fish and Shellfish from Kuruama Community, Bonny River, Nigeria

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Abstract

Residual levels and human health risk assessment of polycyclic aromatic hydrocarbons (PAHs) in Fish (Mullet fish-*Mugil cephalus)* and Shellfish (Tiger prawn-*Penaeus Monodon* and crab-*Uca tangeri)* samples from fishing areas affected by oil and gas exploration activities in Kuruma Community, along Bonny River, Southern Nigeria, were studied. Determination and quantification of PAHs in the biota was carried out following standard procedures using Gas chromatography while human health risk were assessed using human health risk assessment models. Mean concentration of total PAHs ranged from 0.015 to 0.139mg/kg (fish), 0.008 to 0.194 mg/kg (prawn) and 0.015 to 0.140 mg/kg (crab) with considerable predominance of the 3 and 4-rings PAHs. Mean concentrations for total carcinogenic PAHs accounted for 46%, 37% and 36% in prawn, fish and, crab respectively. Total mean PAH concentrations were higher in prawn (0.064 mg/kg) than fish and crab, however, concentrations were not significantly different between the species ($p>0.05$). Low molecular weight (LMW) PAHs were the predominant compounds in fish and crab accounting for 53% of the total PAH residues, suggesting that gill-water transfer might be the dominant exposure route of PAHs into these organisms. Evaluation of human health risk using individual carcinogenic potencies as well as excess cancer risk (ECR) indicated that PAHs in fish and shellfish would induce potential carcinogenic effects.

Keywords: PAHs, Fish, Shellfish, Human health risk

Introduction

Nigeria has no doubt experienced rapid development in the petroleum sector over the last five years, however, along with this development comes the deterioration of environmental quality as a result of incessant spills. Pollution caused by spills from crude oil is one of the most predominant problems in the environment and is, therefore, receiving global attention (Marinescu *et al.,* 2010).Concern about crude oil pollution has been its contamination of various environmental components (Onwurah *et al.,* 2007). Contamination and toxicity of crude oil pollution arise from the fact that Crude oil are complex mixtures of chemicals one of which are polycyclic aromatic compounds (Domask, 1984).

Polycyclic aromatic hydrocarbons (PAHs) form an important class of environmental contaminants. They are ubiquitous and some exhibit carcinogenic or mutagenic potentials (Ravindra *et al.,* 2008). Sixteen of them are listed as priority pollutants by the US Environmental Protection Agency (EPA) as a result of their potential adverse effects on organisms, including human health (USEPA 1987). Due to their hydrophobic nature, they tend to be absorbed rapidly on suspended materials and sediment, becoming bioavailable to fish, shellfish and other marine organisms through the food chain (Latimer and Zheng 2003; Perugini *et al.,* 2007). Ensuing oil spills, contamination of fisheries by PAHs and possible increased health risks associated with consumption of these fisheries in spill areas are of major concern (Wickliffe *et al.,* 2018*).* PAHs are therefore some of the most important chemicals of concern regarding oil-spill-contaminated fishery resources associated with oil spills (Wickliffe *et al.,* 2018). Fish and shellfish are among the various fishery resources vulnerably exposed to PAHs as a result of exposure to oil spills (Copat *et al.,* 2013), which most times accumulate in fish and shellfish, binding to fatty tissues or muscle tissues (Copat *et al.,* 2013).

The Bonny River is one of the rivers in Nigeria affected by oil spills (Awajiusuk, 2015). It is a terminal for crude oil export through a network of pipelines covering 7,000 kilometers (Awajiusuk, 2015). Along the Bonny river are fishing settlements and landing site for fish catch. Kuruama is one of such fishing settlement along the Bonny River affected by oil and gas exploration activities. It is a landing site for fisheries especially, for prawn. The main objective of this study is to determine the levels of PAHs in fish (Mullet fish-*Mugil cephalus)* and Shellfish (Tiger prawn-*Penaeus Monodon* and crab-*Uca tangeri)* from Kuruama in order to assess the potential human health risk associated with consumption.

Materials and Methods

The Bonny River (4° 26′ 0″ N and 7° 10′ 0″ E) is an arm of the Niger River Delta in Rivers Sate, Southern Nigeria. The River is a terminal for crude oil export and along its coast are three oil and gas exploration companies (Shell Nigeria, Mobil producing and Nigeria Liquefied Natural Gas (NLNG)). There is also an awareness of illegal bunkering activities by militants. Kuruama (4^0 28' 42.488"N and 7^0 6' 50.416"E) is a small Island and a fishing settlement in the Bonny Kingdom and a landing site for fisheries especially, for prawn (Figure 1).

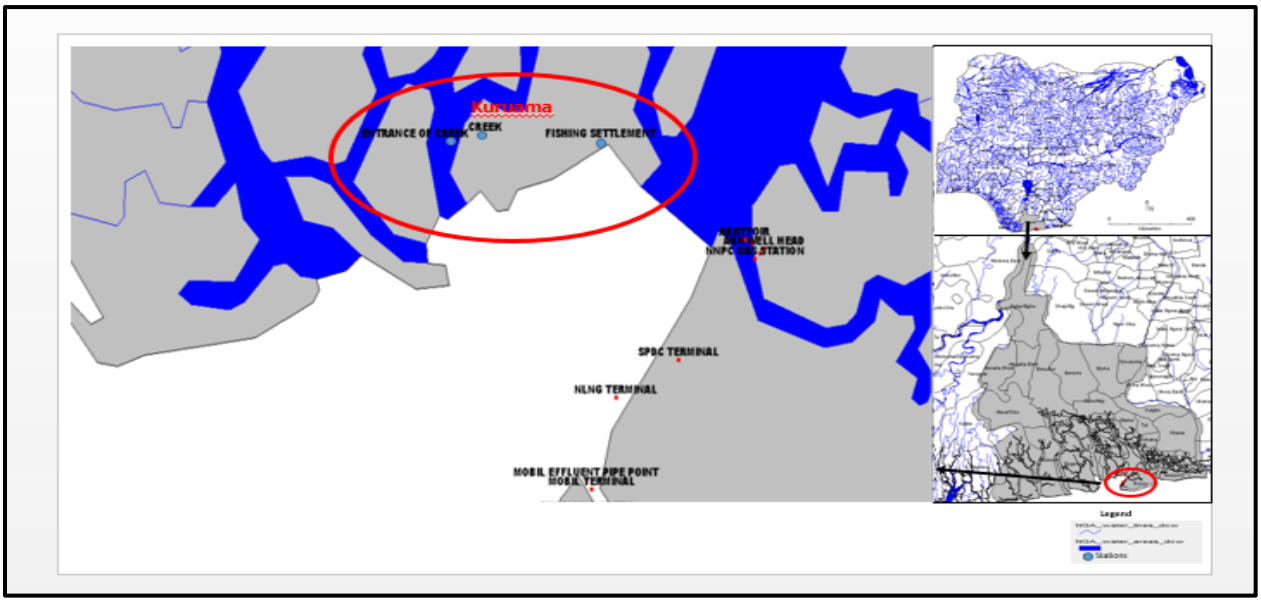

Figure 1: Map of Bonny River, showing Kuruama Community with Sampling Stations

Sample Collection

Mullet fish *(Mugil cephalus*), Tiger prawn (*Penaeus monodon*) and crab (*Uca tangeri*) samples were purchased from local fishermen at sampling locations. All samples were weighed (g), washed then wrapped in aluminum foil and transported immediately to the laboratory in polythene bags. They were refrigerated at 4 °C until extraction (Ezemonye *et al.* 2008).

Analytical procedures

The whole samples of biota were analyzed for PAHs. Analytical procedures for PAHs used in this study are described in detail previously (US EPA, 1986). Frozen composite whole-body tissue was inserted into a homogenizer cup and 100 ml of acetone was added. Samples were homogenized for 20 minutes at 100 rpm and mixed further with 5g of anhydrous sodium sulphate. Extraction was done using soxhlet extraction for approximately 5 hours using dichloromethane and n-hexane mixture. The resulting solvent was eluted with 50 ml n-hexane solvent, evaporated again until 1 - 3 ml. Determination of PAHs in the biota was carried out following standard procedures using Gas chromatography (GC, Hewlett-Packard HP-5890 Series II *with* flame ionization detection (*GC-FID*)).

Human Health Risk Assessment

Human health risk assessment was carried out to estimate the probability of adverse health effects in humans as a result of exposure to PAHs through consumption of contaminated fish. All calculations were done based on USEPA standards (USEPA, 1996). The assessment was carried out for adults (70kg) for both non-carcinogenic and carcinogenic health risk. The description and values of the parameters used for the various calculations are presented in Table 1.

Table 1: Parameters used for estimating exposure assessment through Fish Consumption

Parameters	Unit	Value	Reference
Mean concentration of PAHs (Cf)	mg/kg-fish/Prawn/Crab	Table 2	Table 2
Reference Dose (RfD)	mg/kg/day	USEPA, 1993	USEPA, 1993
Fish/Crustacean ingestion rate *(IFR)*	Kg/capita/day	0.85 (Marine Fish) 0.33 (Crustacean)	FAO, 2014
Adult body weight (BW)	kg	70	Tongo *et al., 2015*
Oral Slope Factor *(SF)*	mg/kg/day	US EPA 2005	US EPA 2005
Toxicity equivalence factor ($TEFi$)	No Unit	Nisbet and LaGoy, 1992; EU, 2006	Nisbet and LaGoy, 1992

Estimated daily intake (EDI)

The estimated daily intake (EDI) (mg/kg/day) of PAHs in fish, prawn and crab samples were estimated using Equation 1

$$\text{Estimated Daily Intake (EDI)} = \frac{Cf\ X\ IFR}{BW} \quad \ldots\ldots\ldots\ldots\ldots \textit{Equation 1} \text{ (USEPA, 1996)}$$

Assessment of non-carcinogenic and carcinogenic health risks

Assessment of non-carcinogenic and carcinogenic health risks was achieved by estimating the hazard quotient (HQ) and hazard index (HI), while the carcinogenic potency of individual PAHs and Excess Cancer Risk (ECR) were used specifically to further estimate carcinogenic health risk. The HQ for non-carcinogenic risks from exposure to PAHs was calculated by dividing the EDI by reference dose (RfD) (Equation 2), while the HQ for carcinogenic risks was estimated using Equation 3.

$$\text{Hazard Quotient (HQ}_{\text{Non-carcinogenic}}) = \frac{EDI}{RfD} \quad \ldots\ldots\ldots\ldots \textit{Equation 2} \text{ (USEPA, 1996)}$$

$$\text{Hazard Quotient (HQ}_{\text{Carcinogenic}}) = EDI\ X\ SF \quad \ldots\ldots\ldots \textit{Equation 3} \text{ (USEPA, 1996)}$$

The hazard index, which estimates the total risk from multiple contaminant pathways, was obtained by summing the HQ of the contaminant pathway (Equation 4). Risk was evaluated for both non-carcinogenic and carcinogenic risks. Values of HQ and HI of contaminants under one (1) are considered as safe (USEPA, 1986).

$$\text{HI} = \sum_{i=1}^{n} HQi \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \textit{Equation 4} \text{ (USEPA, 1986)}$$

The carcinogenic potency of individual PAHs was determined as the product of the concentration of individual PAH congeners and their toxicity equivalency factor (TEF) (Equation 5), while ECR was estimated using Equation 6.

Carcinogenic potencies for PAHs (B(A)Pteq) = $PAHi\ X\ TEFi$ ……. *Equation 5* (USEPA, 1996)

Results and Discussion

PAHs levels in Fish, Prawn, and Crab

Levels of PAHs in fish and shellfish samples from Bonny River, Southern Nigeria are presented in Table 2. Mean PAH concentrations in fish and crab ranged from 0 to 0.059 mg/kg while PAH levels in prawn ranged from 0 to 0.064 mg/kg. Total mean PAH concentrations were higher in prawn (0.064 mg/kg) than fish and crab (0.059 mg/kg), however, concentrations were not significantly different between the species ($p>0.05$, $F= 0.02$).

Table 2: Mean concentration of PAHs in Fish and Shellfish from Kuruama Community, Bonny River, Nigeria

		Fish	Prawn	Crab
PAHs (mg/kg)		Mean±SD	Mean±SD	Mean±SD
Naphthalene	NaP	0.000±0.001	0±0	0±0.001
Acenaphthylene	AcPY	0.005±0.011	0.002±0.004	0.005±0.011
Acenaphthene	AcP	0.010±0.013	0.006±0.012	0.010±0.013
Fluorene	Flu	0.002±0.003	0.001±0.001	0.002±0.003
Phenanthrene	Phe	0.009±0.011	0.015±0.024	0.009±0.011
Anthracene	Ant	0.006±0.012	0.005±0.011	0.006±0.013
Fluoranthene	FL	0.005±0.011	0.005±0.011	0.005±0.011
Pyrene	Pyr	0.001±0.002	0.001±0.003	0.001±0.002
Benzo(a)anthracene	BaA	0.017±0.021	0.015±0.014	0.017±0.021
Chrysene	Chr	0.002±0.004	0±0	0.002±0.004
Benzo(k)fluoranthrene	BkFL	0±0	0±0	0±0
Benzo(a)pyrene	BaP	0.001±0.003	0.011±0.021	0.001±0.003
Benzo(b)fluoranthrene	BbFL	0.002±0.004	0.004±0.007	0.002±0.004
Indeno(1,2,3)pyrene	Ind	0±0	0±0	0±0
Dibenzo(a,h)anthracene	DBA	0±0	0±0	0±0
Benzo(g,h,i,)perylene	BP	0±0	0±0	0±0
TOTAL PAH	∑PAH	0.059±0.056	0.064±0.088	0.059±0.057
Total Carcinogenic PAHs	∑CPAH	0.022±0.031	0.029±0.042	0.022±0.031

Benzo(a)anthracene concentrations in fish and crab samples were significantly higher ($p<0.05$) than the other congeners with mean concentrations of 0.017±0.021 mg/kg, accounting for 28.2% and 27.9% of the total PAHs in fish and crab respectively (Figure 2). In addition, benzo(a)anthracene and Phenanthrene in prawn samples were significantly higher than the other congeners ($p<0.05$) with mean concentrations of 0.015±0.014 mg/kg and 0.015±0.024 mg/kg (Table 2)., accounting for 22.8% and 23.6% respectively (Figure 2). Among the sixteen target PAHs assessed, indeno(1,2,3)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i,)perylene were not detected in any of the biota samples. Mean concentrations (mg/kg) for total carcinogenic PAHs (sum of BaA, Chr, BkFL, BaP, BbFL, Ind, DBA, BP) were 0.022, 0.029 and 0.022 for fish, prawn and, crab, accounting for 37%. 46% and 36% respectively. Total mean carcinogenic PAH concentrations were higher in prawn, but differences in concentrations were not statistically significant between the species ($p>0.05$, $F= 0.07$).

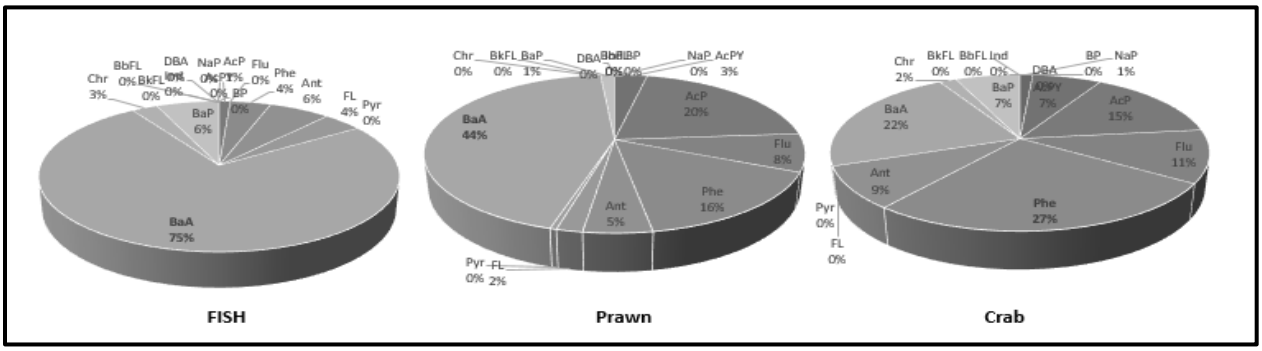

Figure 2: Mean percentage composition of PAHs in biota from Kuruama Community, Bonny River, Nigeria.

The high concentrations of PAHs in prawn than fish and crab could be attributed to the fact that crustaceans usually have reduced rates of biological elimination and thus are more likely to be contaminated (Law *et al.,* 2002). The occurrence of pollutants in fish and shellfish, however, depends largely on environmental concentrations of these compounds and on the physiology and ecological characteristics of the species *(*Meador *et al.,* 1995). A comparison of the concentrations of PAHs in prawns reported in this study with those in previous survey (Nkpaa *et al.,* 2013; Llobet *et al.,* 2006) revealed higher concentrations indicating higher level of contamination. In addition, concentration of benzo(a)pyrene (BaP) in prawn exceeded the safe level of 0.0005 mg/kg for consumption of crustaceans (shellfish) ((Table 2). The high benzo(a)pyrene (BaP) concentrations in prawn exceeding the EU recommended safe limit thus calls for serious health concerns.

PAH composition pattern by ring type showed the predominance of the three-ring and four-ring type PAHs (Fig. 3).

The mean concentrations of the lower molecular weight PAHs (LWPAHs) (two to three rings) was higher than the higher molecular weight PAHs (HWPAHs) (four to six rings) in fish (0.031 mg/kg) and crab (0.032 mg/kg) (Figure 3), accounting for 52.6% and 53.2% respectively of the total PAH, while for prawn the mean percentage concentration of the HWPAHs was 0.036 mg/kg and concentrations were higher than the LWPAH accounting for 55.9% of the total PAHs in prawn (Figure 3). Differences in concentrations between the HWPAH and LWPAH PAHs among the species were however not statistically significant ($p>0.05$).

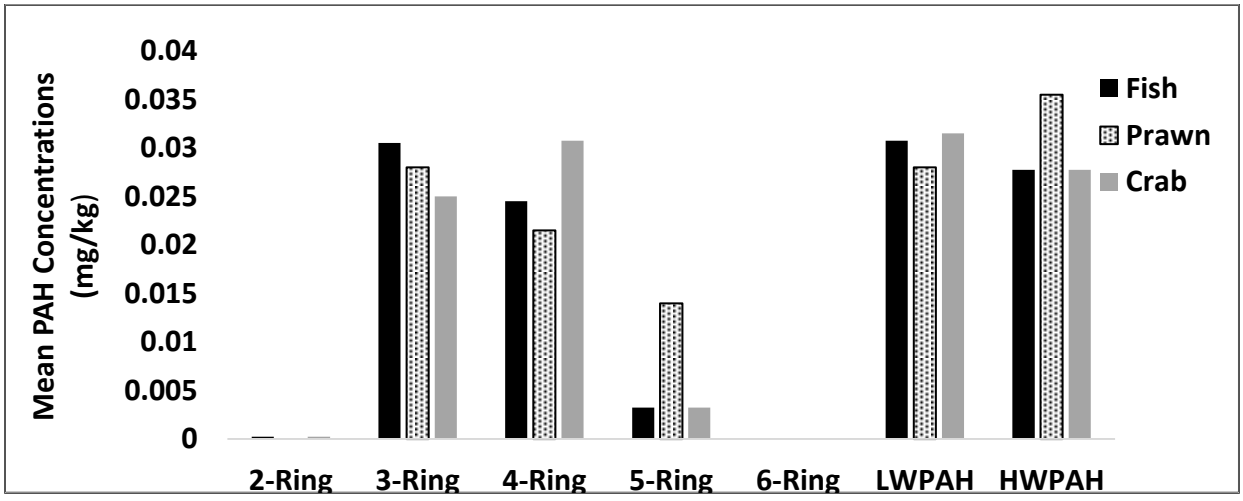

Figure 3: Mean composition of PAHs by ring-type in biota from Kuruama Community, Bonny River, Nigeria

Human Health Risk Assessment of PAHs levels in Fish, Prawn, and Crab

Intake through fish and shellfish consumption is a major route of human exposure to organic contaminants such as PAHs (Dougherty *et al.*, 2000). Risk evaluation for human health was therefore carried out by assessing the average dietary exposure to PAHs, non-carcinogenic and carcinogenic risks. The estimated daily exposure (mg/kg body weight/day) to PAHs from consumption of fish and shellfish from Kuruama Community, Bonny River, Nigeria is presented in Table 3. Consumption of fish contributed to the highest daily intake of PAHs, with EDI values ranging from 0 to 0.0002 mg/kg body weight/day. EDI values for Carcinogenic PAHs were, however, highest in prawn, accounting for 46% of the total estimated daily intake of PAHs in prawn. The estimated daily intake of PAHs in all the species analysed was however observed to be lower than the reference dose (RfD) indicating low risk through consumption. The average HQs and HIs for PAHs in fish and shellfish samples for non-carcinogenic and carcinogenic health risk also showed no potential negative health effect on consumers as values were below 1.

Cancer risk due to dietary exposure to PAHs was further evaluated using individual carcinogenic potencies for PAHs. Benzo(a)pyrene had the highest carcinogenic potency in prawn (0.01 mg/kg) (Table 3) while benzo(a)anthracene and had the highest carcinogenic potency in fish and crab (0.002). Results for individual carcinogenic potencies for benzo(a)anthracene and Benzo(a)pyrene in fish and shellfish showed values exceeding the guideline screening value of 0.00067 mg/kg (wet wt) (USEPA 2000), for human consumption indicating a high potential for carcinogenic risk through consumption.

Table 3: Estimated daily intake, Non-Carcinogenic and Carcinogenic Risk of PAHs for adult (70-kg body weight) from consumption of fish and shellfish

	Fish				Prawn				Crab			
PAHs	EDI	HQ (Non-carcinogenic)	HQ Carcinogenic	B(A)Pteq	EDI	HQ (Non-carcinogenic)	HQ Carcinogenic	B(A)Pteq	EDI	HQ (Non-carcinogenic)	HQ Carcinogenic	B(A)Pteq
NaP	3.04E-06	0.000	NA	2.50E-07	0.00E+00	0.00E+00	NA	0.00E+00	1.18E-06	5.89E-05	NA	2.50E-07
AcPY	6.38E-05	0.016	NA	5.25E-06	9.43E-06	2.36E-03	NA	2.00E-06	2.48E-05	6.19E-03	NA	5.25E-06
AcP	1.15E-04	NA	NA	9.50E-06	2.71E-05	NA	NA	5.75E-06	4.60E-05	NA	NA	9.75E-06
Flu	1.82E-05	0.000	NA	1.50E-06	2.36E-06	3.93E-05	NA	5.00E-07	7.07E-06	1.18E-04	NA	1.50E-06
Phe	1.03E-04	0.003	NA	8.50E-06	6.84E-05	1.71E-03	NA	1.45E-05	4.01E-05	1.00E-03	NA	8.50E-06
Ant	6.98E-05	NA	NA	5.75E-05	2.48E-05	NA	NA	5.25E-05	2.95E-05	NA	NA	6.25E-05
FL	6.38E-05	0.000	NA	5.25E-06	2.48E-05	8.25E-05	NA	5.25E-06	2.48E-05	8.25E-05	NA	5.25E-06
Pyr	1.21E-05	0.000	NA	1.00E-06	5.89E-06	1.47E-04	NA	1.25E-06	4.71E-06	1.18E-04	NA	1.00E-06
BaA	2.00E-04	0.007	NA	1.65E-03	7.07E-05	2.36E-03	NA	1.50E-03	7.78E-05	2.59E-03	NA	1.65E-03
Chr	2.13E-05	NA	1.55E-05	1.75E-05	0.00E+00	NA	0.00E+00	0.00E+00	8.25E-06	NA	6.02E-06	1.75E-05
BkFL	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00
BaP	1.52E-05	NA	1.11E-07	1.25E-03	4.95E-05	NA	3.61E-07	1.05E-02	5.89E-06	NA	4.30E-08	1.25E-03
BbFL	2.43E-05	NA	1.77E-05	2.00E-04	1.65E-05	NA	1.20E-05	3.50E-04	9.43E-06	NA	6.88E-06	2.00E-04
Ind	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00
DBA	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00
BP	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00	0.00E+00	NA	0.00E+00	0.00E+00
		HI= 0.026	HI= 3.34E-05			HI= 0.007	HI= 1.24E-05			HI= 0.010	HI= 1.30E-05	

***NA-Not Available**

Conclusion

The study revealed varying levels of PAHs in Fish and Shellfish from in Kuruama Community, Bonny River, Nigeria. The concentration of PAHs in these organisms should be considered alarming in view of the carcinogenic potency of many individual PAHs especially for benzo(a)anthracene and benzo(a)pyrene in which results from the study showed high potential for carcinogenic risk through consumption.

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