

ESTIMATION OF HUMAN HEALTH RISK FROM EXPOSURE TO POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) THROUGH DIETARY INTAKE OF FISH AND SHELLFISH FROM COAL BEACH, BONNY RIVER, SOUTHERN NIGERIA

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ABSTRACT

*Dietary intake is one of the major human exposure pathways to PAHs. Consequently, the objective of the study was to estimate possible human health risk from consumption associated with PAHs concentrations in fish (Mullet fish-*Mugil cephalus*) and shellfish (Tiger prawn-*Penaeus monodon* and crab-*Uca tangeri*) samples, from Coal Beach, a landing site for fish catch along Bonny River, Southern Nigeria. Fish samples were observed to record the highest total mean PAH concentration of 0.126 mg/kg with total carcinogenic PAHs accounting for 47.3% of the total PAHs. The highest average concentration of 0.060, 0.033 and 0.021 mg/kg was observed for benzo(a)anthracene in fish, prawn and crab respectively. Consumption of fish contributed to the highest intake of PAHs with carcinogenic PAHs accounting for 47% of the total estimated dietary intake. The estimated daily intake of PAHs in all the species analysed were however observed to be lower than the reference dose (RfD) indicating low risk through consumption. However, further risk assessment using individual carcinogenic potencies, revealed that individual carcinogenic potencies for benzo(a)anthracene and benzo(a)pyrene in fish and shellfish exceeded the guideline screening value for human consumption indicating a high potential for carcinogenic risk.*

Keywords: PAHs, Fish, Shellfish, Health risk

INTRODUCTION

Fish, seafood products, meat and dairy products make up more than 90% of the daily intake of food for the general population (Lee and Shim, 2007). Fish and shellfish are traditionally important in diet as they constitute an important source of proteins, minerals, and vitamins (Lee and Shim, 2007; Kris-Etherton *et al.*, 2002). These aquatic organisms are however vulnerably exposed to toxic chemicals released from industrial, agricultural and municipal sources (Copat *et al.*, 2013).

Many of these chemicals, which most times are carcinogenic accumulate by binding to fatty tissues or muscle tissues of these organisms (Copat *et al.*, 2013). One of such contaminants fish and shellfish are exposed to, are Polycyclic Aromatic Hydrocarbons (PAHs). It is a large heterogeneous group of organic contaminants that are persistent with a wide range of distribution in various environmental media (Wu *et al.*, 2012). They are important components of crude oil and have been reported in areas of crude oil spills (Awajiusuk, 2015). The Bonny River

is one of such rivers 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, is the presence of oil and gas industries with their coherent gas flaring sites. The Coal Beach community is a landing site for all boats coming into Bonny from Port Harcourt and also a landing sites for boats from the surrounding villages of Bonny Local Government Area, consequently a major landing site for fish catch along Bonny River. This area is affected by most of these spills. A mobile Nigeria National Petroleum Corporation (NNPC) filling station, the only filling station which supplies maritime transport fuel is located along the coast of Coal beach and forms a potential source of PAHs to the Bonny River.

PAHs found in crude oil have the potential to accumulate in fish and crustaceans, such as shrimps, prawns, and crabs (Law *et al.*, 2002) and can subsequently result in potential health risk through ingestion of contaminated seafood (Yender *et al.*, 2002). PAHs have been reported in different environmental media including fish and shellfish in this region (Nkpaa, *et al.*, 2013; Nwaichia and Ntorgbob, 2016). However, only a few reports exist on the human health risk associated with dietary intake of PAH-contaminated fish and shellfish from Bonny

River. The study was therefore carried out to estimate possible human health risk from consumption associated with PAHs concentrations in fish (Mullet fish, *Mugil cephalus*) and shellfish (Tiger prawn, *Penaeus monodon* and crab, *Uca tangeri*) samples, from Coal Beach, a fishing area along Bonny River, Southern Nigeria and a fish landing site affected by oil and gas exploration activities.

MATERIALS AND METHODS

Study area

The Bonny River (4° 26' 0" N and 7° 10' 0" E) is an estuary on the coast of Rivers State, Southern Nigeria that drains the River Niger. 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) (Figure 1). There is also an awareness of illegal bunkering activities by militants. Coal Beach (4⁰ 27' 24.104"N and 7⁰ 10' 35.935"E) is the commercial nerve center of Bonny town hosting major economic activities. It is a landing site for fishermen who have ready buyers for their fish catch. It is located North East of NLNG export site. In addition, there is an abattoir situated about 500m from the Coal beach jetty where waste is emptied directly into the Bonny River (Figure 1).

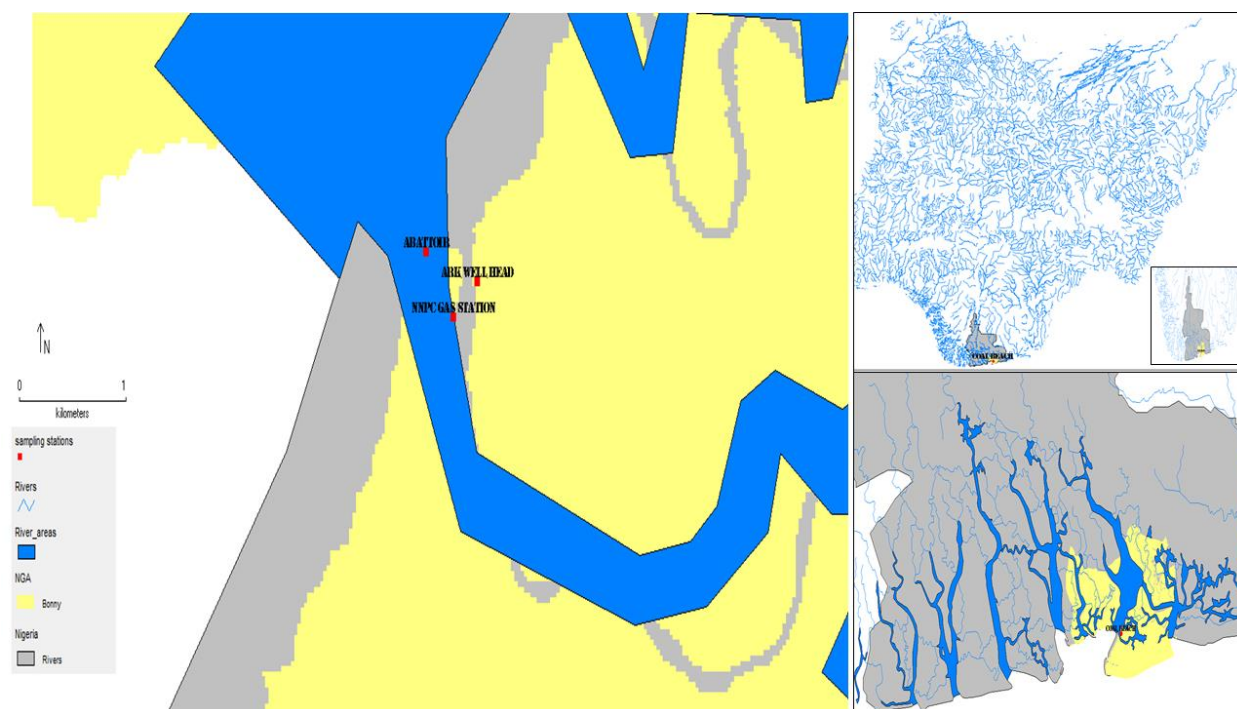


Figure 1: Map of Bonny River, showing Coal Beach with Sampling Stations

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, wrapped in aluminum foil, labeled and transported on ice to the laboratory. 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 details previously (USEPA, 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 I.

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

Parameters	Unit	Value	Reference
Mean concentration of PAHs	mg/kg-fish, Prawn, and Crab	Table 2	Table 2
Reference Dose (<i>RfD</i>)	mg/kg/day	Table 2	USEPA (1993).
Fish/Crustacean ingestion rate (<i>IFR</i>)	Kg/capita/day	0.85(Marine Fish) 0.33 (Crustaceans)	FAO (2014).
Exposure Duration (<i>ED</i>)	years	70	Qu <i>et al.</i> , (2015).
Exposure Frequency (<i>EF</i>)	Days/year	365	Qu <i>et al.</i> , (2015).
Adult body weight (<i>BW</i>)	kg	70	Tongo <i>et al.</i> , (2017)
Average life span (<i>ATn</i>)	days	25550	Papadakis <i>et al.</i> , (2015)
Carcinogenic potency of benzo[a]pyrene (<i>Q</i>)	mg/kg/day	7.3	Qu <i>et al.</i> , (2015).
Oral Slope Factor (<i>SF</i>)	mg/kg/day	USEPA (2005)	USEPA (2005)
Toxicity equivalence factor (<i>TEFi</i>)	No Unit	Nisbet and LaGoy, (1992).	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.

$$\text{Estimated Daily Intake (EDI)} = \frac{Cf \times IFR}{BW} \quad \text{Equation 1}$$

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.

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

$$\text{Hazard Quotient (HQ}_{\text{Carcinogenic}}) = EDI \times SF \quad \text{Equation 3}$$

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).

$$HI = \sum_{i=1}^n HQ_i \quad \text{Equation 4}$$

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.

$$\text{Carcinogenic potencies for PAHs (B(A)Pteq)} = PAH_i \times TEF_i \quad \text{Equation 5}$$

$$\text{Excess Cancer Risk (ECR)} = \frac{\sum Q \times B(A)P_{teq} \times IFR \times ED}{BW \times ATn} \quad \text{Equation 6}$$

Where

Q = Carcinogenic potency of benzo[a]pyrene (mg/kg/day) (Table 1)

$B(A)P_{teq}$ = Carcinogenic potencies for PAHs

IFR = Fish/Crustacean ingestion rate (Kg/capita/day) (Table 1)

ED = Exposure Duration (Years) (Table 1)

BW = Adult body weight (Kg) (Table 1)

ATn = Average life span (Days) (Table 1)

RESULTS

PAHs levels in Fish, Prawn, and Crab

Results of PAH congeners in fish and shellfish samples from Coal beach, Bonny River, Southern Nigeria is presented in Table 2.

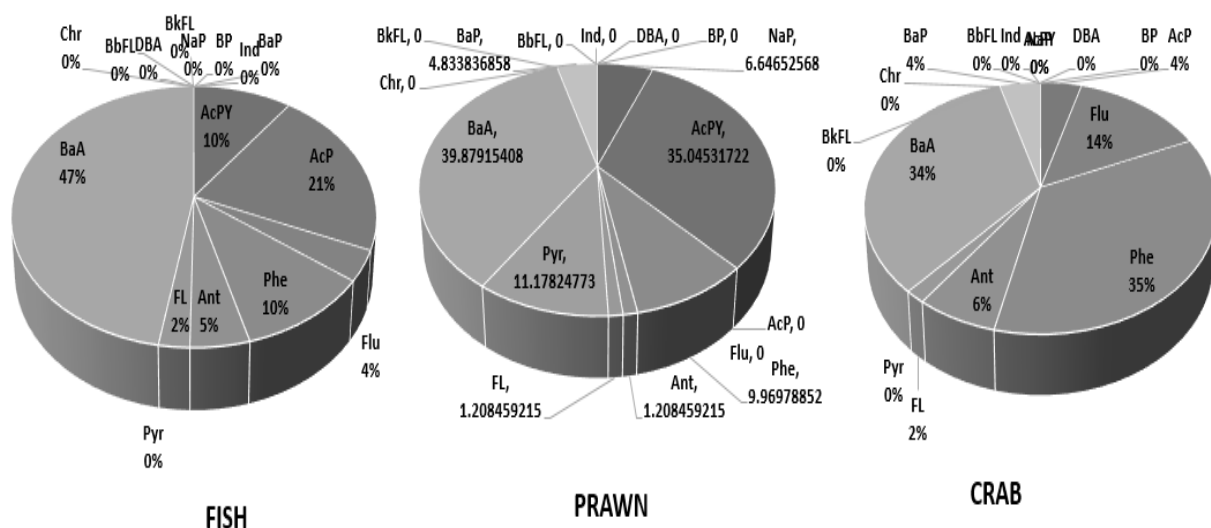
Mean concentration of total PAHs was higher in fish (0.126 mg/kg) than prawn and crab (Table 2), however, concentrations were not significantly different between the species ($p > 0.05$, $F = 0.47$). For individual concentrations of PAHs, benzo(a)anthracene was the most dominant congener in fish and prawn samples (Table 2) with mean concentrations of 0.060 ± 0.06 and 0.033 ± 0.03 mg/kg (Table 2), accounting for 47% and 40% of the total PAHs in fish and prawn respectively. However, concentrations were not

statistically significant between the congeners. Phenanthrene was the most dominant congener in crab with a mean concentration of 0.022 ± 0.025 mg/kg and a percentage contribution of 35% (Figure 2). However, Phenanthrene concentrations in crab were not significantly higher than the other congeners ($p > 0.05$).

Mean concentrations for total carcinogenic PAHs (sum of BaA, Chr, BkFL, BaP, BbFL, Ind, DBA, BP) accounted for 47%, 35% and 38% of the total PAHs in fish, prawn and crab respectively (Table 2). Total mean carcinogenic PAH concentrations were higher in fish (0.060 mg/kg) than prawn and crab, but differences in concentrations were not statistically significant between the species ($p > 0.05$, $F = 0.19$).

Table 2: Mean concentration of PAHs in Fish and Shellfish from Coal Beach, Bonny, Nigeria

PAHs (mg/kg)		Fish	Prawn	Crab
		Mean±SD	Mean±SD	Mean±SD
Naphthalene	NaP	0±0	0.006±0.011	0±0
Acenaphthylene	AcPY	0.013±0.016	0.029±0.057	0±0
Acenaphthene	AcP	0.027±0.035	0±0	0.003±0.003
Fluorene	Flu	0.005±0.006	0±0	0.009±0.014
Phenanthrene	Phe	0.013±0.015	0.008±0.017	0.022±0.041
Anthracene	Ant	0.006±0.010	0.001±0.002	0.004±0.005
Fluoranthene	FL	0.003±0.006	0.001±0.002	0.001±0.002
Pyrene	Pyr	0±0	0.009±0.019	0±0
Benzo(a)anthracene	BaA	0.060±0.060	0.033±0.030	0.021±0.025
Chrysene	Chr	0±0	0±0	0±0
Benzo(k)fluoranthrene	BkFL	0±0	0±0	0±0
Benzo(a)pyrene	BaP	0±0	0.004±0.008	0.003±0.006
Benzo(b)fluoranthrene	BbFL	0±0	0±0	0±0
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	ΣPAH	0.126±0.104	0.083±0.110	0.063±0.081
Total Carcinogenic PAHs	ΣCPAH	0.060±0.060	0.029±0.036	0.024±0.028

**Figure 2: Mean percentage composition of PAHs in biota from Coal Beach, Bonny, Nigeria**

The PAH composition pattern by ring type showed a considerable predominance of the three-ring and four-ring type PAHs (Figure 3).

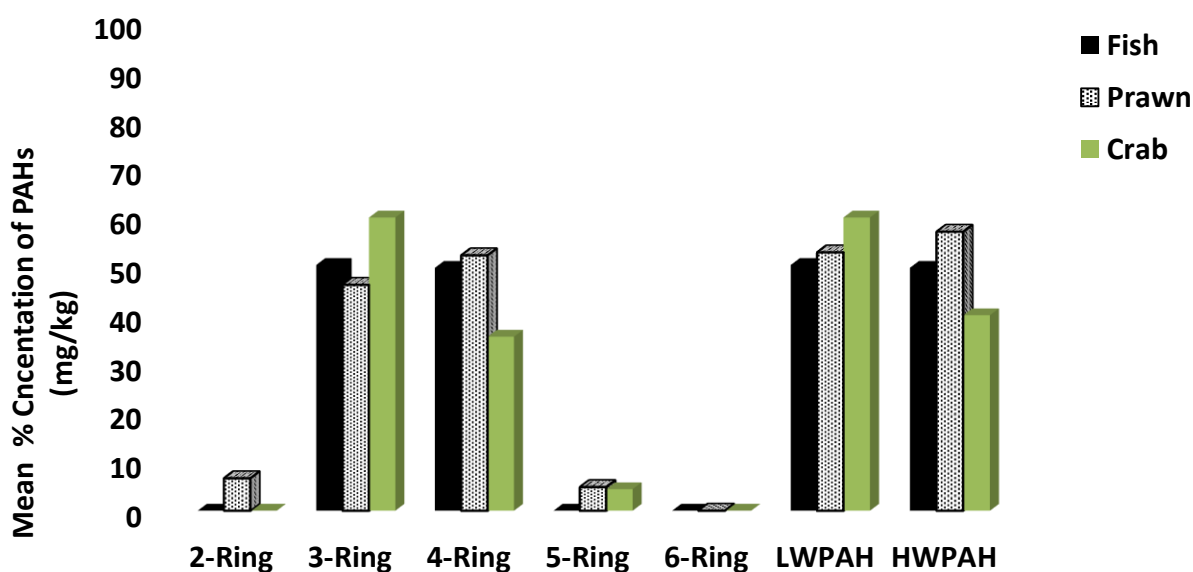


Figure 3: Mean percentage composition of PAHs by ring-type in biota from Coal Beach, Bonny, Nigeria

The mean percentage concentration of the lower molecular weight PAHs (LWPAHs) (two to three rings) was higher than the higher molecular weight PAHs (HWPAAHs) (four to six rings) in fish and crab accounting for 50% and 60% respectively of the total PAH, while for prawn the mean percentage concentration of the HWPAAHs was higher than the LWPAH accounting for 57% of the total PAHs in fish (Figure 3). Differences in concentrations between the HWPAAH and LWPAH PAHs among the species were however not statistically significant ($p > 0.05$).

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

For risk assessment, dietary exposure to PAHs, the non-carcinogenic and carcinogenic risks were estimated. Daily dietary intake of PAHs (mg/kg body weight/day) through fish and shellfish consumption for an adult (70kg) is shown in Table 3.

EDI values (mg/kg body weight/day) ranged from 0 to 0.0002 (prawn), 0 to 0.0007 (fish) and 0 to 0.0001 (crab) with consumption of fish contributing to the highest intake of PAHs. EDI values for Carcinogenic PAHs accounted for 47%, 35% and 38% in fish, prawns, and crabs respectively.

DISCUSSION

Pollutant levels in fish and shellfish depend principally on environmental concentrations and on the physiology and ecological characteristics of the species (Meador *et al.*, 1995). PAH concentrations in fish samples were higher than concentrations in prawn and crab. The observed high concentrations are not consistent with the fact that fishes are known to metabolise PAH (Law *et al.*, 2002; Di Giulio and Hinton, 2016), and crustaceans are especially likely to be contaminated because of reduced rates of biological clearance of PAHs in these species. Nonetheless, studies have reported

the accumulation of PAHs in fish (Nkpaa, *et al.*, 2013; Zhang *et al.*, 2004) and this could explain the reason for the higher concentrations of PAHs in fish compared to prawn and crab.

For risk assessment, 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 to consumers as values were below 1.

The potency of PAHs in fish and shellfish to cause carcinogenic health risk was evaluated using individual carcinogenic potencies for PAHs. Benzo(a)anthracene had the highest carcinogenic potency (mg/kg) in prawn (0.004) and crab (0.003)

while Benzo(a)pyrene had the highest carcinogenic potency (mg/kg) in fish (0.006)(Table 3). 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 0.67 ng/g wet wt (USEPA, 2000), for human consumption indicating high potential carcinogenic risk.

In addition, results of the estimated excess cancer risk (ECR) from lifetime exposure to PAHs through fish and shellfish consumption showed ECR value for Benzo(a)anthracene in fish (Table 3) exceeding the threshold value of 1×10^{-6} set by USEPA (Ding *et al.*, 2012) indicating that lifetime exposure to benzo(a)anthracene through fish consumption would result in cancer risk.

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

Prawn						Fish					Crab				
PAHs	EDI	HQ(Non-carcinogenic)	HQ Carcinogenic	B(A)P _{10q}	ECR	EDI	HQ(Non-carcinogenic)	HQ Carcinogenic	B(A)P _{10q}	ECR	EDI	HQ(Non-carcinogenic)	HQ Carcinogenic	B(A)P _{10q}	ECR
NaP	2.59286E-05	0.001296429	NA	0.0000055	5.18571E-10	0	0	NA	0	0	0	0	NA	0	0
AcPY	0.000136714	0.034178571	NA	0.000029	2.73429E-09	0.000154821	0.038705	NA	0.00001275	3.09643E-09	0	0	NA	0	0
AcP	0	NA	NA	0	0	0.000327857	NA	NA	0.000027	6.35714E-09	1.29643E-05	NA	NA	0.00000275	2.59286E-10
Flu	0	0	NA	0	0	5.76786E-05	0.000961	NA	0.00000475	1.15357E-09	0.00004125	0.000688	NA	0.00000875	8.25E-10
Pha	3.88929E-05	0.000972321	NA	0.00000825	7.77857E-10	0.000160893	0.004022	NA	0.00001325	3.21786E-09	0.000103714	0.002593	NA	0.000022	2.07429E-09
Ant	4.71429E-06	NA	NA	0.00001	9.42857E-10	6.98214E-05	NA	NA	0.0000575	1.39643E-08	1.88571E-05	NA	NA	0.00004	3.77143E-09
FL	4.71429E-06	1.57143E-05	NA	0.000001	9.42857E-11	3.64286E-05	0.000121	NA	0.000003	7.28571E-10	4.71429E-06	1.57E-05	NA	0.000001	9.42857E-11
Pyr	4.36071E-05	0.001090179	NA	0.00000925	8.72143E-10	0	0	NA	0	0	0	0	NA	0	0
BaA	0.000155571	0.005185714	NA	0.0033	3.11143E-07	0.000725536	0.024185	NA	0.005975	1.45107E-06	0.000100179	0.003339	NA	0.002125	2.00357E-07
Chr	0	NA	0	0	0	0	NA	0	0	0	0	NA	0	0	0
BkFL	0	NA	0	0	0	0	NA	0	0	0	0	NA	0	0	0
BaP	1.88571E-05	NA	1.37637E-07	0.004	3.77143E-07	0	NA	0	0	0	1.29643E-05	NA	9.46393E-08	0.00275	2.59286E-07
BbFL	0	NA	0	0	0	0	NA	0	0	0	0	NA	0	0	0
Ind	0	NA	0	0	0	0	NA	0	0	0	0	NA	0	0	0
DBA	0	NA	0	0	0	0	NA	0	0	0	0	NA	0	0	0
BP	0	NA	0	0	NA	0	NA	0	0	0	0	NA	0	0	0
		HI=0.043	HI=1.377E-07				HI=0.068	HI=0				HI=0.007	HI=9E-08		

*NA-Not Available

CONCLUSION

The present study showed varying levels of PAHs in fish and shellfish from Coal Beach in Bonny along the Bonny Estuary with high potential for carcinogenic risk in humans from fish consumption. The study, therefore, provides reasonable evidence of the need to fully evaluate the risks of PAHs in fish and shellfish from fishing sites affected by oil and gas exploration activities to safeguard the health of consumers.

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