



## Bioaccumulation of Polycyclic Aromatic Hydrocarbon (PAH) in a bivalve (*Arca senilis*- blood cockles) and health risk assessment

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### ARTICLE INFO

#### Keywords:

Bioaccumulation  
PAH  
Bivalves  
Human health risks  
Southern Nigeria

### ABSTRACT

Concentration of PAH in bivalves (*Arca senilis*) and human health risks due to consumption was examined in samples collected from southern Nigeria and analysed using gas chromatography. Mean PAH concentration ( $\text{ngkg}^{-1}$ ) ranged from  $12.0 \pm 5.0$ – $5500.0 \pm 1000$  with a significant difference ( $p < 0.001$ ) while total PAH ranged from 3000.0–16,000.0. Concentrations ( $\text{ngkg}^{-1}$ ) of PAH4 varied from 250 to 15268.0 while concentrations of PAH8 ranged from 542.0 to 15620.7 with significant difference ( $p < 0.001$ ). Diagnostic ratios for PAH source distinction suggested mixture of petrogenic and pyrogenic sources. Dietary daily intake-DDI ( $\text{ng/kg/day}$ ) of individual PAHs ranged from 1.04 to 9.86 while DDI for PAH4 and PAH8 were 340.8 and 379.8 respectively. Carcinogenic potencies ( $\text{ngkg}^{-1}$ ) varied from 0.012 to 900.0 for individual PAH while carcinogenic toxic equivalent (TEQs) values were 1916.2, 572.49 and 1914.4 for total PAH, PAH4 and PAH8 respectively. The Excess cancer risk (ECR) for individual PAHs, PAH4 and PAH8 were all  $< 10^{-6}$ . DDI and ECR values obtained were below USEPA threshold concentration/limits indicating minimal health risk concerns while PAH4 and PAH8 concentrations were also below the EU regulatory limits ( $30 \mu\text{g kg}^{-1}$ ) for PAH4. The margin of exposures were above the 10,000 critical limit proposed by EFSA while incremental life cancer risk (ILCR) value ( $10^{-5}$ – $10^{-9}$ ) also suggests low potential health risk for consumers of the sea food. The screening value (SV) was 0.095 but lower than observed TEQs values indicating potential health concerns. The study concluded that consumers of bivalves (*Arca senilis*) in southern Nigeria generally have minimal health risk concern via consumption but regular monitoring is required to detect changes.

### 1. Introduction

The Niger Delta region of Nigeria is impacted due to contamination traceable to oil and gas activities. Polycyclic Aromatic Hydrocarbon (PAH) is a component (0.2–7%) of hydrocarbon [1] and is a major group of chemicals of concern in the environment due to its carcinogenic and mutagenic potencies. Researchers have reported the presence of PAH in the region [2–4]. PAHs sources can be petrogenic emanating from petroleum-related activities or pyrogenic (pyrolytic), from the incomplete combustion of diesel fuel and engine oil [5], wood, coal, biomass of forest, grass fires, waste incinerators, and fossil fuels that are used in industrial operations and power plants [6–9]. Due to their low vapour pressure and solubility in sea water PAH attach to suspended organic matter and eventually sink to the bottom of sediments [10] and remain persistent for years. The sediment is home for most bivalves such as *Arca senilis*, a filter feeding mollusc of the order Arcoida in the

family Arcidae. *Arca senilis* satisfies basic biomonitoring conditions due to wide distribution along coastal areas, sessile lifestyle, easy to handle and a filter feeder with the ability to accumulate heavy metals and contaminants without appreciable metabolism [11–13]. In view of the above, the organism gives a time-integrated indication of environmental contamination because it accumulates considerable level of contaminants during indiscriminate filter feeding. It is a major sea food (rich in protein and vitamins) in the south coast of Nigeria and consumed in a variety of delicacies. Average consumption of seafood in some coastal communities of the Niger Delta region of Nigeria is about 370 g per week in terms of dry weight of sea food per meal consumed [14]. Bioaccumulation and toxic effects associated with such marine organisms and onward transmission to humans via the food chain is a major concern due to negative effects. The alternation in gametogenesis, gender determination, and growth [15] and increased risk of cancer and mutation are major adverse effects on living organisms [16].

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<https://doi.org/10.1016/j.toxrep.2019.09.006>

Received 15 July 2019; Received in revised form 4 September 2019; Accepted 20 September 2019

Available online 25 September 2019

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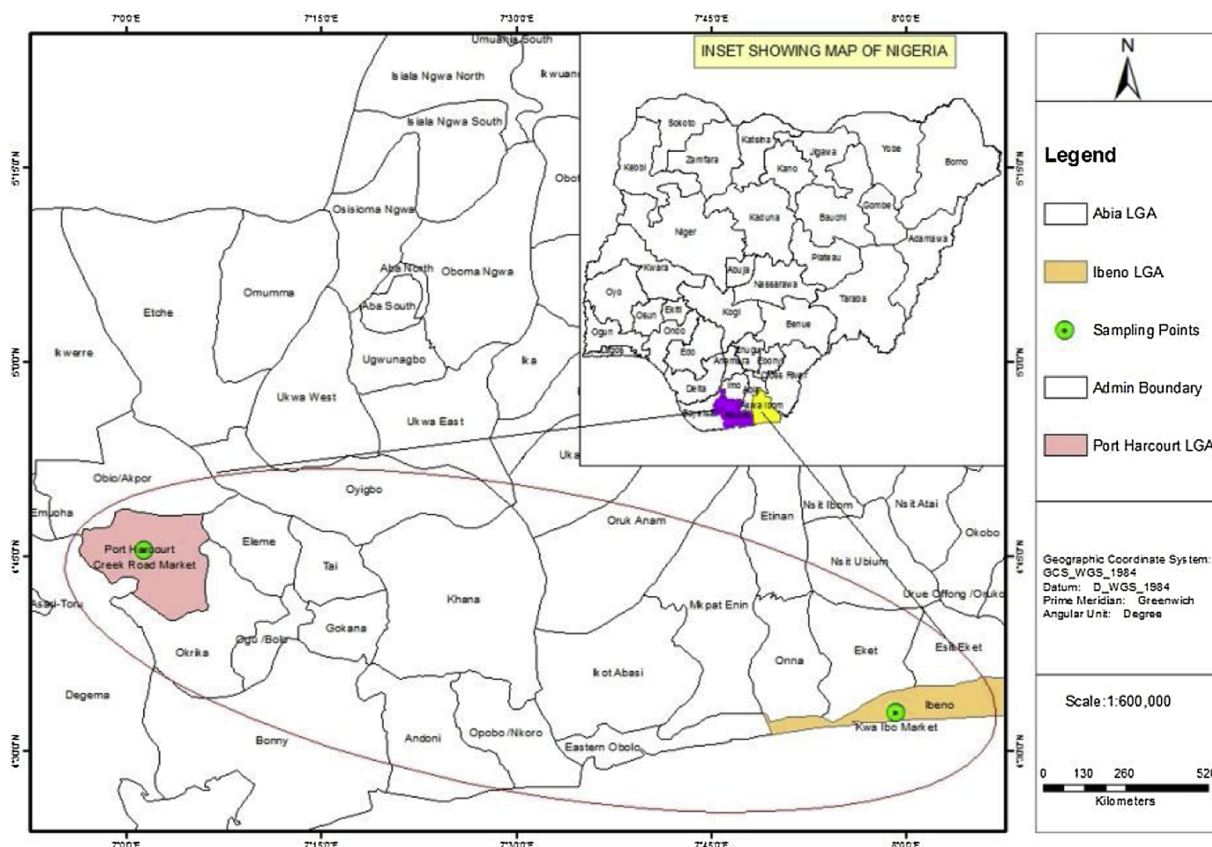


Fig. 1. Map showing study area.

European Union has stressed and recommended that PAHs be measured as wide as possible in food products in order to obtain data on the occurrence and specific concentrations in a variety of matrices [17]. There is dearth of information on bioaccumulation of PAH in bivalves such as *Arca senilis* in Nigeria, particularly in the southern region. The aim of this study was therefore, to determine concentrations of PAH in *Arca senilis* obtained from markets in southern Nigeria with a view to assessing potential health risk associated with consumption.

## 2. Materials and methods

### 2.1. Sample collection

Samples of bivalve (*Arca senilis*) were obtained from two regional markets in Port Harcourt (4°45′29.64″N 7° 1′25.30″E) and Eket - Kwa Ibo (4°32′47.93″N 7°59′25.85″E) in southern Nigeria (Fig. 1). These markets are major landing points for fresh sea foods such as fish, oysters, periwinkles and bivalves. The samples were collected fresh, wrapped in aluminum foil, labeled and immediately transported to the laboratory in ice-pack coolers for analysis. Fifty samples were collected on a monthly basis for twelve months (October 2017 to September 2018). This was done to satisfy incremental sampling in order to give aggregate samples representative of lots or sub-lots for laboratory analysis [18].

### 2.2. Extraction and analysis

Extraction of PAH was done using the methods described by Pena et al. [19]. Bivalve samples were extracted from the shell, oven dried and properly homogenized. 10 g of the bivalve sample was carefully mixed with anhydrous  $\text{Na}_2\text{SO}_4$ . For extraction, 20 ml of dichloromethane was added to the sample and then covered with aluminum foil to avoid evaporation. This was then sonicated to separate

supernatants of extracts and evaporator was used to concentrate the extracts. A chromatographic column packed with 1 cm glass wool at the base was used to clean up the extracts. The column was added with 2 g of silica gel and 1 cm of anhydrous  $\text{Na}_2\text{SO}_4$  and pre-eluted with 20 ml dichloromethane. Analytes (extracts) were concentrated and put in vials of 3 ml size.

### 2.3. Gas chromatographic (GC) analysis

Gas Chromatography (GC model: HP5890 Series II GC-FID (made in the USA) was used for the analysis. The extract was analysed for the PAH congeners (Table 1). The GC was set for an initial temperature of 60 °C (2 min) and inclined at 25 °C/min and raised to 300 °C for 5 min, allowed to stay for 15 min, making a total of 22 min. Using 2  $\mu\text{l}$  level splitless injection mode, the injection port temperature was put at 250 °C while the injection port of the flame ionization detector (FID) was maintained at 300 °C. A standard mixture of 16 priority PAHs (NaP, AcPY, AcP, Flu, Phe, Ant, FL, Pyr, BaA and Chr, BbFL, BkFL, BaP, Ind, DBA and BP) was recorded and used for the analysis. Comparison of the retention time of standards to that from the extracts and individual analysis of PAHs were used for identification and quantization of different components observed. The carcinogenic PAHs evaluated included BaA, Chr, BkFL, BaP, BbFL, Ind, DBA and BP while the non carcinogenic PAHs evaluated included Nap, AcPY, AcP, Flu, Phe, Ant, FL, Pyr [20]. The limit of detection (LOD) was  $0.0001\mu\text{gkg}^{-1}$ . The recovery method was by use of surrogate standard. Recovery rate was by spiking the sample with known concentration of the surrogate and normal extraction was done. Injection into the GC gives the concentration of the surrogate alongside the other samples [21].

### 2.4. Analytical standards and reference

The internal standard solutions consisted of naphthalened<sub>8</sub>,

**Table 1**  
PAH Components Analysed.

Full Name of PAH	Abbreviation	Markings
Naphthalene	NaP	*
Acenaphthylene	AcPY	*
Acenaphthene	AcP	*
Fluorine	Flu	*
Phenanthrene	Phe	*
Anthracene	Ant	*
Fluoranthene	FL	*
Pyrene	Pyr	*
Benzo [a] anthracene	BaA	***
Chrysene	Chr	***
Benzo [b] fluoranthene	BbFL	***
Benzo [k] fluoranthene	BkFL	**
Benzo [a] pyrene	BaP	***
Indeno [1, 2, 3-cd] pyrene	Ind	**
Dibenzo [a, h] anthracene	DBA	**
Benzo [g, h, i] perylene	BP	**

\*Non-Carcinogenic PAHs. \*\* Carcinogenic PAHs. \*\*\* Carcinogenic PAH and PAH used to derive the PAH4 Index. [3].

acenaphthene-d10, chrysene-d12, and perylene-d12 in DCM at 1000 µg/mL (Accu Standard Inc., New Haven, CT, USA). The Analytical Standards and Reference used during the analysis of samples are as used by [21,22].

2.5. Comparison of PAH concentrations with legal limits

The health risk associated with consumption of bivalves (*Arca senilis*) contaminated with PAHs concentrations was evaluated by comparing measured PAHs in biota with available regulatory limits. Individual PAH concentrations, total PAHs concentrations (sum of all evaluated PAHs) and total carcinogenic PAHs (sum of measured carcinogenic PAHs [20] including BaA, Chr, BkFL, BaP, BbFL, Ind, DBA and BP were evaluated and compared with available limits and literature.

2.6. Exposure assessment

The Dietary Daily Intake (DDI) via consumption of PAHs contaminated seafood (bivalves - *Arca senilis*) was evaluated. The DDI of PAH's in the bivalve species was assessed for adult population using Eq. 1 [23]. Average weight of adult in Nigeria was considered to be 70 kg [24] obtained from data of the Food and Agriculture Organization [25] on Fishery and aquaculture statistics [3,23].

$$\text{The Dietary Daily Intake (DDI)- ng/day} = C_i \times \text{IFR} \quad (1)$$

$C_i$  = concentration of PAH in bivalve samples  
IFR = fish ingestion rate (IFR) [25]

2.7. Risk characterisation

The cancer risk due to exposure via consumption of PAH contaminated bivalves (*Arca senilis*) was evaluated by using the indices of PAH4 (sum of BaA, Chr, BbFL, and BaP and PAH8 (the sum of BaP, BaA, BbFL, BkFL, BP, Chr, DBA and Ind) and individual PAH carcinogenic potencies, carcinogenic toxic equivalents (TEQs) and excess cancer risk (ECR). Margin of exposure (MOE), incremental life cancer risk (ILCR) and screening value were also evaluated. The calculated values in bivalve samples were then compared with regulatory limits to assess possible risk of exposure and effects.

To calculate PAH4, it is the summation of BaA, Chr, BbFL and BaP [3,18,].

$$\text{PAH4 Index (PAH4)} = \Sigma \text{BaA} + \text{Chr} + \text{BbFL} + \text{BaP} \quad (2)$$

Where, BaA is benz[a]anthracene, Chr is Chrysene, BbFL is benzo[b] fluoranthene, BaP is benzo[a]pyrene [3]

$$\text{Carcinogenic potencies of individual PAHs (B(A)Pteq)} = C_i \times \text{TEFi} \quad (3)$$

TEFi = toxicity equivalency factor as used by [26]

$$\text{Carcinogenic toxic equivalents (TEQs)} = \Sigma \text{B(A)Pteq} \quad (5)$$

as used by [27]

The excess cancer risk due to consumption of PAH contaminated bivalve was evaluated using Eq. 6 [28].

$$\text{Excess Cancer Risk (ECR)} = \Sigma Q \times B(A) \text{ Pteq} \times \text{IFR} \times \text{ED} / (\text{BW} \times \text{ATn}) \quad (6)$$

Where

Carcinogenic potency of BaP (Q) mg kg<sup>-1</sup> d – 7.3 mg/kg/d [27]

Exposure Duration (ED) - 30 years [29], Adult body weight (BW) - 70 kg [30], Average life span (ATn) - 8760 days [31]

The screening value was calculated using the Eq. 7

$$\text{Screening Value (SV)} = (\text{RL/SF}) \times \text{BW} / \text{IFR} \quad (7)$$

as used by [32,33].

Where

Maximum acceptable risk level (RL) dimensionless = 10<sup>-5</sup> [34],

Oral Slope Factor (SF) mg/kg/day [20].

i Evaluation of Margin of Exposure (MOE)

MOE was also evaluated as an acceptable method of risk assessment approved by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) and the European Food Safety Authority (EFSA) [35,36,37]

$$\text{MOE} = \text{BMD}_{10} \times \text{BW/E} \quad (8)$$

Where

BMD<sub>10</sub> = 70 µg/kg bw/day (for BaP) or 340 µg/kg bw/day (for PAH4) [38].

E = DDI = Dietary Daily Intake (DDI) [3,23]

BW = Adult body weight (BW) - 70 kg [30]

i Evaluation of Incremental Life cancer Risk (ILCR):

The ILCR was evaluated using the equation

$$\text{ILCR}_{\text{ingestion}} = \text{CS} \times \text{CSF}_{\text{ingestion}} \times 3\sqrt{(\text{BW}/70)} \times \text{IR} \times \text{EF} \times \text{ED}/\text{BW} \times \text{AT} \times 10^6 \quad (9)$$

as used by [39]

where

CS = Sum of converted PAHs concentrations based on TEQ values as proposed by Nisbet and Lagoy [26]

CSF<sub>ingestion</sub>: Carcinogenic slope factor for ingestion of B[a]P (7.3 mg kg<sup>-1</sup> day<sup>-1</sup>) [40]

BW = Body weight of adult (70 kg) [30],

IR = fish ingestion rate (IFR) as used in eqn 1 [3,23]

EF = Exposure Frequency 365 days/year as used by [41,42]

ED = Exposure Duration (ED) - 30 years [29]

AT = Average Time (AT) [31,43].

2.8. Evaluation of PAH sources

The Ant/Ant + Phen, Flu/Flu + Py [44] BaA/(BaA + Chry) [45] and LMW/HMW [46] ratios were used to estimated possible sources of PAH in bivalve samples examined.

**Table 2**  
Mean concentrations (ngkg<sup>-1</sup>) ± standard error (SE) of PAH congeners in bivalve samples examined during the study.

PAH components	MEAN ± SE (n = 12)
NaP	130.0 ± 40.0
AcPY	88.0 ± 30.0
AcP	24.0 ± 10.0
Flu	12.0 ± 5.0
Phe	68.0 ± 20.0
Ant	45.0 ± 20.0
FL	100.0 ± 70.0
Pyr	190.0 ± 60.0
BaA	5500.0 ± 1000.0
Chr	230.0 ± 10.0
BbFL	410.0 ± 200.0
BkFL	200.0 ± 90.0
BaP	79.0 ± 30.0
Ind	240.0 ± 100.0
DBA	180.0 ± 10.0
BP	92.0 ± 40.0

Maximum levels: BaP = 6.0 µgkg<sup>-1</sup>; PAH4 = 35.0 µgkg<sup>-1</sup> [18,47].

2.9. Statistics

Microsoft Excel was used for basic statistics and graphs. Analysis of variance (general linear model) was used to test for significant difference while Tukey test was used for post hoc analysis at 95% CIs for mean based on pooled standard deviation was done using Minitab 16 software.

3. Results and discussion

3.1. Individual PAH concentrations in bivalves (*Arca senilis*)

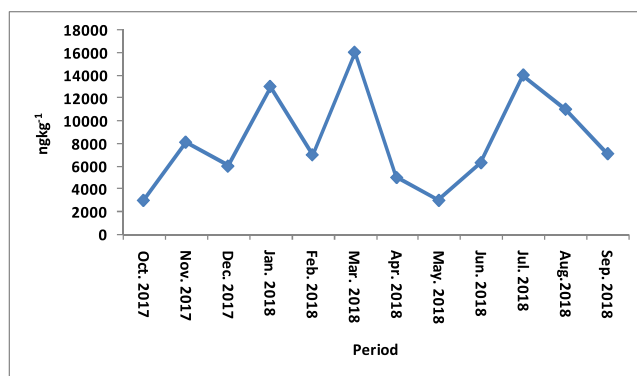
The mean concentrations of the component PAHs in bivalve samples (*Arca senilis*) during the entire study period is presented in Table 2 while Table 3 shows summary of ANOVA for PAH concentrations in bivalve samples. ANOVA shows significant difference in terms of concentrations between PAH congeners while alphabets depicts actual point of difference using post hoc analysis (Tukey test).

Results show that Fluorene had the least mean concentration (ngkg<sup>-1</sup>) of 12 ± 5 while BaA had the highest mean concentration (5500 ± 1000), followed by BbFL with 410.0 ± 200 in bivalve samples examined. ANOVA showed significant difference (p < 0.001) in the concentration of PAH congeners and mean comparison further showed that the significant difference was between the levels of BaA and all other PAH components in the sample (Table 3). This was due to the consistently elevated levels of BaA in the samples observed. Tongo [3] had reported higher levels (0.049 ± 0.048 0.047 ± 0.042

**Table 3**  
Summary of ANOVA for PAH variables.

Variables	MS value	F-value
All PAH components	0.0000221	13.9***
PAH4	0.0000848	17.16***
PAH8	0.0000428	16.78***
BaA	A	
Chr	B	
BbFL	B	
BkFL	B	
BaP	B	
Ind	B	
DBA	B	
BP	B	

PAH components with different alphabets are significantly different.  
\*\*\* = significant (p < 0.001).



**Fig. 2.** Total concentrations of PAHs (sum of PAHs/month) showing temporal trend.

0.013 ± 0.018 mg/kg) of BaA in fish and shellfish from the same region. Concentrations of PAH components observed in this study were within levels recorded in a similar study [14] however, high values reported by [14] was attributed to major hydrocarbon pollutions within the Niger Delta region. Concentrations of BbFL and Ind in this study were below the European Union (EU) limit of 12.000 µg kg<sup>-1</sup> and 30 µg kg<sup>-1</sup> for the respective compounds. Lee and Shim [48] had reported use of BaP as a marker for the occurrence and effect of carcinogenic PAHs in food. The mean concentration (79.0 ± 30 ng kg<sup>-1</sup>) of BaP in bivalves of this study was lower than those reported in fish and shell fish from the Niger Delta region [3]. The BaP in bivalves of this study was also lower than the European Union regulatory limits of 0.002 mg/kg and 0.0005 mg/kg in sea foods of fish and shellfish respectively. This indicates minimal health risk concern for consumers of this shell fish (bivalves) in the southern part of Nigeria.

3.2. Total PAH concentrations in bivalves (*Arca senilis*)

The sum of all PAH (total PAHs) on monthly trend is presented in Fig. 2. There were noticeable variations in the total PAHs (sum of all PAH) across study periods. The sum of all PAHs observed showed that the least concentration (3000 ng kg<sup>-1</sup>) in bivalve samples was observed in October 2017 and May 2018 while the highest concentration (16,000 ng kg<sup>-1</sup>) was recorded in March 2018. The observed temporal variation in total PAH was however, not statistically significant (p > 0.05) between the study periods. Total PAH concentrations of this study were at variance with levels (7.150 ± 0.040, 35.800 ± 0.100 and 30.100 ± 0.090 µg kg<sup>-1</sup>) reported by [14] in similar organisms from the Niger Delta region. Further breakdown of total PAH indicated that 47% of PAH concentration came from PAH8 while 42% was contributed by PAH4 and 11% came from noncarcinogenic PAH. Total PAHs in bivalves reported by other studies is given in Table 4. Differences in observed PAH could be due to varying contaminant levels in the environment as well as intake and excretion rates (metabolic pathways) of different organisms. Meador [49] had reported occurrence of PAH in fish and shell fish to depend largely on environmental

**Table 4**  
Total PAHs in bivalves reported by other studies around the world.

Bivalve	Total PAH	Reference
<i>Crassostrea.sp</i>	524(µgkg <sup>-1</sup> )	Bay of Biscay [51] 2008
<i>Ostera edulis</i> *	125 (ngg <sup>-1</sup> )	Lebanon [52] 2008
<i>Crassostrea Virginia</i> -	312	Mobile Bay [53] 2003
<i>Mytilus galloprovincialis</i>	98.80 (ngg <sup>-1</sup> )	Mediterranean [54] 1998
<i>Saccostrea cucullata</i> -	66 (ngg <sup>-1</sup> )	Oman [55] 2005
<i>Circentia callipyga</i> -	105(ngg <sup>-1</sup> )	Qatar [55] 2005
<i>Barbatia helblingii</i>	421.86(ngg <sup>-1</sup> )	Bushehr [56] 2011
<i>Arca senilis</i>	3000 - 16,000 ng kg <sup>-1</sup>	This study

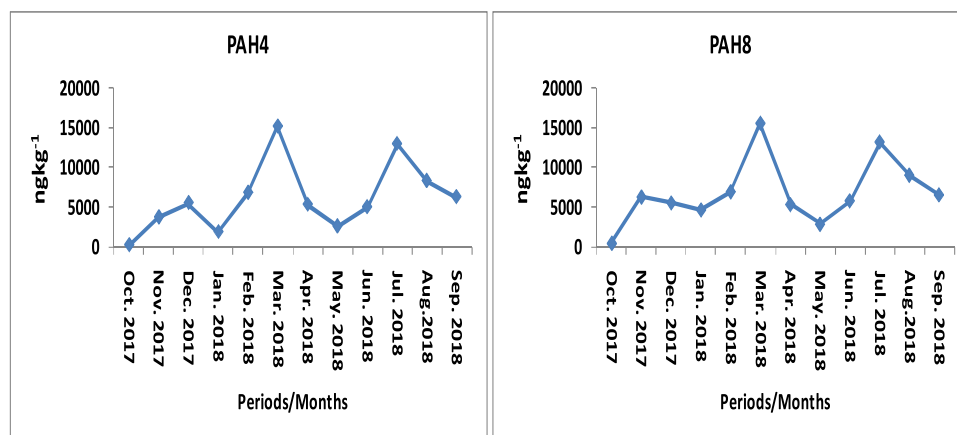


Fig. 3. PAH4 and PAH8 showing temporal variations and trend.

concentrations, physiology and ecological characteristics of the species. Total PAH values of this study was lower compared to concentrations ( $\mu\text{g kg}^{-1}$ ) reported in related bivalves (oyster: 289–370, mussel: 268–351, Clam: 134–342) [50].

### 3.3. PAH4 and PAH8 concentrations in bivalves (*Arca senilis*)

The sum of PAH4 and PAH8 on monthly variations was expressed graphically in Fig. 3. Minimal differences existed between values of PAH4 and PAH8 in bivalve samples observed but both varied noticeably across study periods. PAH4 varied from  $250.0 \text{ ng kg}^{-1}$  in October 2017 to  $15,268.0 \text{ ng kg}^{-1}$  in March 2018 while PAH8 ranged from  $542.0 \text{ ng kg}^{-1}$  in October 2017 to  $15,620.7 \text{ ng kg}^{-1}$  in March 2018 with both showing similar trend in their temporal variation. Values obtained in this study were well below those reported by [14] for carcinogenic PAHs. There was significant difference ( $p < 0.001$ ) between concentrations of PAH4 and PH8 components due to the consistently elevated concentrations of BaA relative to other components (Table 2). Variations of PAH4 components was however, not significantly different ( $p > 0.05$ ) between study period but PAH8 components showed varied significant difference ( $p < 0.05$ ) between study periods. BaP was also considered a biomarker for the occurrence and effect of carcinogenic PAHs in food by the European Union Commission [57]. This guideline gives maximum concentration of PAH congeners in bivalves as  $10 \text{ ng kg}^{-1}$  (fresh weight) which is above what was observed in this study. This also indicates minimal health risk exposure to consumers of bivalves (*Arca senilis*) in the southern region of Nigeria. Studies on individual PAHs, such as BaP have shown toxicological effects, such as haematological effects, reproductive, developmental toxicity and immunotoxicity but the carcinogenic and genotoxic (DNA-damaging) potential of these compounds have caused most concern [18]. The International Agency for Research into Cancer (IARC) in 2012 concluded that benzo[a]pyrene is a human carcinogen [18]. Based on data collected from member states, the European Food Safety Authority (EFSA) Panel on Contaminants in the Food Chain (CONTAM) in 2008 concluded that BaP alone was not a suitable indicator for the occurrence of

PAHs in food. The Panel found that PAH4 (the sum of BaP, BaA, BbFL and Chr and PAH8 (the sum of BaP, BaA, BbFL, BkFL, BP, Chr, DBA and Ind were the most suitable indicators for PAHs in food, with PAH8 not providing much added value compared to PAH4 [18]. The fact that PAH8 does not provide much added value is in consonance with the findings of this study where the difference between PAH4 and PAH8 was quite minimal (Fig. 3). The concentrations of PAH4 and PAH8 in samples of bivalve (*Arca senilis*) in this study were below the EU regulatory limits of maximum levels of  $30 \mu\text{g kg}^{-1}$  for PAH4 in the Commission Regulation (EU) No 835/2011. This implies reduced risk of carcinogenic potentials for consumers of this bivalve (*Arca senilis*) from the southern region of Nigeria.

### 3.4. Sources of PAHs

The sources of PAHs can either be petrogenic i.e., released from petroleum products or pyrogenic due to the combustion of biomass. Diagnostic ratios have been designed and used to distinguish the sources of PAHs due to their stability, physical and chemical attributes [46,58]. Table 5 presents commonly used ratios for PAHs source determination. This diagnostic ratios used for water and sediments was also applied to the biota (bivalve) examined in this study.

The Ant/Ant + Phen, Flu/Flu + Py, BaA/(BaA + Chry) and LMW/HMW ratios obtained for this study were 0.398, 0.345, 0.071 and 0.66 respectively suggesting PAHs sources of pyrogenic origin in the samples examined. However, Flu/Flu + Py (0.345) suggest PAH component of petrogenic source indicating a possible mixture of PAH source for this study. The BaA/(BaA + Chry) mean value of 0.071 found in this study agrees with the result of [14] who reported BaA/(BaA + Chry) ratio of 1.00 and 0.890 in molluscs from the Niger Delta and considered the PAHs more of pyrogenic in origin. Nyarko [45] had earlier reported BaA/(BaA + Chry) ratio  $> 0.350$  to indicate pyrogenic sources but values  $< 0.200$  was attributed to petrogenic origin. These sources were not distinguishable for ratios in the range 0.200–0.350 [45]. Major anthropogenic activities linked to petrogenic and pyrogenic source include illegal petroleum refining/bunkering activities, burning of

Table 5  
Diagnostic ratios values for particular PAHs source determination.

Diagnostic Ratio	Petrogenic	Pyrogenic	References	This study
Ant/Ant + Phen	$< 0.1$	$> 0.1$	Brandli et al. 2007 [44]	0.398
Flu/Flu + Py	$< 0.4$	$> 0.4$	Brandli et al. 2007 [4]	0.345
BaA/(BaA + Chry)	$< 0.2000$	$> 0.350$	Nyarko et al. 2011 [45]	0.071
LMW/HMW-PAHs	$> 1$	$< 1$	Nasher et al. 2013 [46]	0.66
	Fuel combustion	Grass/coal/wood combustion		

Key: Ant is anthracene, Phen is phenanthrene, Flu is fluoranthene and Py is pyrene.

**Table 6**

Calculated dietary daily intake (DDI), Carcinogenic potencies (B(A)Pteq) and Excess cancer risk (ECR), MOE and ILCR of PAHs in bivalves (*Arca senilis*) from markets in the Southern region Nigeria.

PAH component	MEAN ± SE (ngkg <sup>-1</sup> )	RFD	TEFs	DDI (CixIFR) ngkg <sup>-1</sup>	B(A)Pteq (CixTEFi) ngkg <sup>-1</sup>	ECR ngkg <sup>-1</sup>	MOE
NaP	130.0 ± 40.0	0.02	0.001	7.124	0.13	2.5 × 10 <sup>-12</sup>	6.8 × 10 <sup>5</sup>
AcPY	88.0 ± 30.0	NA	0.001	4.822	0.088	1.7 × 10 <sup>-11</sup>	1.0 × 10 <sup>6</sup>
AcP	24.0 ± 10.0	0.06	0.001	1.315	0.024	4.7 × 10 <sup>-12</sup>	2.7 × 10 <sup>6</sup>
Flu	12.0 ± 5.0	0.04	0.001	6.576	0.012	2.3 × 10 <sup>-12</sup>	7.4 × 10 <sup>5</sup>
Phe	68.0 ± 20.0	NA	0.001	3.726	0.068	1.3 × 10 <sup>-12</sup>	1.3 × 10 <sup>6</sup>
Ant	45.0 ± 20.0	0.3	0.01	2.466	0.45	8.8 × 10 <sup>-11</sup>	1.9 × 10 <sup>6</sup>
FL	100.0.0 ± 70.0	0.04	0.001	5.48	0.1	1.3 × 10 <sup>-12</sup>	8.9 × 10 <sup>5</sup>
Pyr	190.0 ± 60.0	0.03	0.001	1.041	0.19	3.7 × 10 <sup>-12</sup>	4.7 × 10 <sup>6</sup>
BaA	5500.0 ± 1000.0	NA	0.1	3.014	550	1.0 × 10 <sup>-8</sup>	1.6 × 10 <sup>6</sup>
Chr	230.0 ± 10.0	NA	0.01	1.260	2.3	4.5 × 10 <sup>-11</sup>	3.8 × 10 <sup>6</sup>
BbFL	410.0 ± 200.0	NA	1	2.246	410	8.0 × 10 <sup>-9</sup>	2.1 × 10 <sup>6</sup>
BkFL	200.0 ± 90.0	NA	0.1	1.096	20	3.9 × 10 <sup>-10</sup>	4.4 × 10 <sup>6</sup>
BaP	79.0 ± 30.0	NA	0.1	4.329	7.9	1.5 × 10 <sup>-10</sup>	1.1 × 10 <sup>6</sup>
Ind	240.0 ± 100.0	NA	0.1	1.315	24	4.6 × 10 <sup>-10</sup>	3.7 × 10 <sup>6</sup>
DBA	180.0 ± 10.0	NA	5	9.864	900	1.7 × 10 <sup>-8</sup>	4.9 × 10 <sup>5</sup>
BP	92.0 ± 40.0	NA	0.01	5.041	0.92	1.7 × 10 <sup>-11</sup>	9.7 × 10 <sup>5</sup>
TEQ for Total PAHs	1916.182						
TEQ = PAH4	572.49						1.4 × 10 <sup>4</sup>
TEQ = PAH8	1914.39						1.2 × 10 <sup>4</sup>
ILCR (All PAHs)	1.36 × 10 <sup>-5</sup>						
ILCR (PAH4)	4.1 × 10 <sup>-9</sup>						
ILCR (PAH8)	1.3 × 10 <sup>-5</sup>						
SV	0.095						

confiscated petroleum products on water ways and mangrove environments by security agencies, combustion of tyres/plastics and other organic substances of non-point sources within the Niger Delta region. The findings of this study compares favourably with that of [56] who reported a mixture of petrogenic and pyrogenic origin of PAH in clams using Ant/Ant + Phen, Flu/Flu + Py ratios.

### 3.5. Potential human health risk from consumption of contaminated seafood (bivalves)

Health risk is mainly via exposure by consumption of contaminated seafood. The dietary daily intake (DDI) was assessed to determine human health risk via exposure through consumption of contaminated bivalves. Carcinogenic risks was determined via the carcinogenic potencies of individual PAHs (B(A)Pteq and those of PAH4 and PAH8, carcinogenic toxic equivalents (TEQs) and the Excess Cancer Risk (ECR) Index. Margin of exposure (MOE) and incremental life cancer risk (ILCR) were also assessed. Values obtained for the evaluated health indices are given in Table 6.

The concept of DDI in evaluating health risks from toxicants is usually applied due to differences in fish consumption rates [59]. Exposure route in this case is mainly via consumption of contaminated seafood (bivalves). DDI of individual PAHs ranged from 1.04 to 9.86 ng kg<sup>-1</sup> to while the DDI for PAH4 and PAH8 were 340.8 and 379.8 ng kg<sup>-1</sup> respectively (Table 6). The DDI of individual PAHs were generally lower than the available reference oral dose but those of PAH4 and PAH8 had elevated DDI values above those of individual PAHs which could be a source of concern due to the fact that PAHs may not act in isolation when consumed. The DDI for individual PAH concentrations were also compared to the available reference dose [20] to determine long-term risk from exposure to PAHs residues through the consumption of contaminated bivalves. The recommended allowable daily intake for BaP in Italy is 0.04 to 0.42 g day<sup>-1</sup> [60] which is above the DDI concentration observed in this study for both carcinogenic and noncarcinogenic PAHs. The (B(A)Pteq of individual PAHs varied from 0.012 to 900.0 ng kg<sup>-1</sup>

The Excess cancer risk (ECR) could be induced by dietary exposure via consumption of bivalves contaminated with PAH by humans. ECR (ngkg<sup>-1</sup>) values of individual PAHs ranged from 1.0 × 10<sup>-8</sup> to 4.7 × 10<sup>-12</sup> while those of PAH4 and PAH8 were 5.9 × 10<sup>-8</sup> and

1.6 × 10<sup>-7</sup>. The calculated ECR for lifetime exposure to PAHs via consumption of bivalves were compared to the acceptable guideline value of 10<sup>-6</sup> of USEPA [61]. This guideline value suggests that a level of cancer risk of one in one million (ECR = 10<sup>-6</sup>) in a 70 year period of lifetime, is considered acceptable but a case of an additional lifetime cancer risk of one in ten thousand or greater (ECR = 10<sup>-4</sup>) is considered serious [62]. Results of this study for individual PAHs, PAH4 and PAH8 were all below the USEPA threshold concentration indicating minimal health risk concerns. Studies in fish [63] and other foods [64,65], have reported ECR values above USEPA regulatory value.

Margin of Exposure: The Joint FAO/WHO Expert Committee on Food Additives (JECFA) and the European Food Safety Authority (EFSA) in 2005 developed guidance on risk assessment of genotoxic and carcinogenic compounds [35]. The JECFA noted that genotoxic and carcinogenic compounds could give non-linear dose–response relationships, but the no-observed-effect-level in a study of carcinogenicity depicts the limit of detection in that bioassay, rather than an estimate of a possible threshold. MOE thus, is the ratio of the no-observed-adverse-effect level (NOAEL) or benchmark dose lower confidence limit (BMDL) for the critical effect to the theoretical, predicted, or estimated exposure dose or concentration [66]. MOEs for individual PAH congeners in this study ranged from 4.9 × 10<sup>5</sup> - 4.4 × 10<sup>6</sup> while those of PAH4 and PAH8 were 1.4 × 10<sup>4</sup> and 1.2 × 10<sup>4</sup> respectively (Table 6). MOEs for individual PAH congeners, PAH4 and PAH8 were above the 10,000 critical limit proposed by EFSA indicating an exposure margin of low health risk concern for consumers of *Arca senilis* in southern Nigeria. MOEs of this study were lower than those (150,000 - children and 230,000 - adults) calculated by Veyrand for mean exposure to PAH4 in French total diet study [67]. MOEs (BaP: 14960, PAH4: 7723) calculated by [37] were also less than values obtained in this study, in which Wu attributed it to high pollution rate in their study area.

Incremental Life Cancer Risk (ILCR): The exposure risk for PAHs could quantitatively be assessed by using the approach of incremental lifetime cancer risk (ILCR) [68–70]. Excess lifetime cancer risk of ≥ 10<sup>-6</sup> has been consistently considered insignificant and ≥ 10<sup>-4</sup> as significant, where actions are usually taken in order to reduce risk when cancer risk falls within the latter group [71]. Values of ILCR obtained in this study were in the range of 10<sup>-5</sup> - 10<sup>-9</sup> suggesting low health risk concern for consumers of this bivalve (*Arca senilis*) in southern Nigeria

with regards to incremental life time exposure via consumption. The ILCR values obtained in this study compares with that ( $2.62 \times 10^{-5}$ -adults;  $7.08 \times 10^{-6}$ -children) reported [72] in a multi-pathway assessment of human health risk posed by polycyclic aromatic hydrocarbons. This guide line may vary slightly depending on the region and policy meaning no absolute criteria for acceptable number of additional cancers over a lifetime. In most jurisdictions, the one-in-one-million ( $10^{-6}$ ) chance of additional cancers proposed by the USEPA regulatory is used as a management tool for risks posed by environmental contamination. However, risks ranging from 1-in-10,000 ( $10^{-4}$ ) to 1-in-1,000,000 ( $10^{-6}$ ) are generally considered acceptable, depending on the situation and circumstances of exposure [73].

Screening value (SV) indicates the threshold level of chemicals in edible tissue which is of potential public health concern [32,33]. SV was assessed in this study to determine health risks of PAHs to humans that consume such bivalves. The SV value of this study was compared to the TEQ value to determine health risk to humans via consumption of PAH contaminated bivalves. The SV of this study was 0.095 was observed to be lower than the carcinogenic toxic equivalents (TEQs) values of this study. This may suggest potential health risks for consumers of bivalves in southern Nigeria. The result of this current study is consistent with those of [74], in a study of PAH concentrations in *Chrysidichthys nigrodidatatu* in southern Nigeria who also reported TEQ values above screening values suggesting potential health effects. The result of this study also agrees with that of [75] who reported higher TEQ values in relation to observed screening values for PAH in sea foods (fish, crab, and bivalve) in Iran.

#### 4. Conclusion

Health risks due to consumption of PAH contaminated sea foods is a major concern requiring regular monitoring to detect changes as recommended by regulatory agencies. This study examined levels of PAH in bivalves and possible health risks due to consumption. Concentrations of individual PAHs congeners and total PAH in samples examined were minimal compared to regulatory limits. Health indices such as carcinogenic potencies, carcinogenic toxic equivalents, excess cancer risks, margin of exposure, incremental life cancer risk and screening value were assessed and found to be lower than regulatory limits of USEPA and European Union. Diagnostic ratio for PAH source evaluation suggested mixture of petrogenic and pyrogenic origin. The study therefore, concluded that consumers of bivalves (*Arca senilis*) in southern Nigeria have minimal health risk concerns via consumption of such sea foods but regular monitoring is needed to detect perturbations.

#### Declaration of Competing Interest

The authors hereby declare that there is no conflict of interest with regards to this paper.

#### Acknowledgement

The authors acknowledge the efforts of Grant Ogbe in the production of study map.

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