



Assessment of PAHs levels in some fish and seafood from different coastal waters in the Niger Delta



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ABSTRACT

Levels of sixteen polycyclic aromatic hydrocarbons (PAHs) in 30 edible tissues of selected frequently-consumed fish and seafood collected from three coastal waters of Niger Delta, namely, Sime, Kporghor and Iko were investigated in 2014. Gas chromatographic analysis were employed for PAHs determination. Observed mean PAHs levels in the samples ranged from below detection limit (BD) of analytical instrument to $22.400 \pm 0.050 \mu\text{g kg}^{-1}$ wet wt. in *Littorina littorea*, BD to $87.400 \pm 0.030 \mu\text{g kg}^{-1}$ wet wt. in *Crassostrea virginica* and from BD to $171.000 \pm 0.430 \mu\text{g kg}^{-1}$ wet wt. in *Periophthalmus koeleuteri*. The highest average concentration of $171.000 \pm 0.430 \mu\text{g kg}^{-1}$ wet wt. was recorded for Indeno [1,2,3-cd]pyrene from Sime water. High molecular weight PAHs (HMW-PAHs) were generally predominant compared to low molecular weight PAHs (LMW-PAHs). The LMW- PAH/HMW-PAH ratio was <1 for all species, indicating anthropogenic origin of PAHs in the coastal waters of Niger Delta environment. Moreover, the study of the PAHs fingerprints, using specific ratios, suggests the predominance of a pyrolytic origin for observed PAHs.

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1. Introduction

Marine and terrestrial environments, in the recent times, have been tagged with abundance of persistent organic pollutants. This sordid situation is worsened by the emergence of densely populated industrial districts as both environmental components are important in most activities leading to economy development, and at the same time cast an ominous pall on the environment directly or indirectly. Oshineye [23] reported Nigeria's oil reserves of about 31.5 billion barrels and crude oil production of about 2.118 million barrels per day in the post- 2000 era, underscoring the enormity of oil and related activities in Nigeria. There are 606 oil fields in the Niger Delta of which 360 are onshore and 246 are offshore [18]. Associated gas flaring, above ground pipeline leakage, oil waste dumping, sabotage and oil spills lead to environmental pollution. Pollution caused by petroleum and its derivatives is the most prevalent problem in the environment. The release of crude oil into the environment by oil spills is receiving worldwide attention [17]. Crude oil spills of January 12, 1998 and July,

1979 at Mobil Unlimited Idoho platform (45,000 barrels) and West of Shell operated forcados terminal storage facility (560,000 barrels) respectively spilled into the Atlantic coastal line in Nigeria, its surrounding land, mangrove swamps and territorial waters. Similarly, other anthropogenic activities in coastal areas contributing to PAHs contamination of coastal environments could include use of creosote-treated wood in aquaculture, bush burning, industrial effluent discharge, dense vehicular emissions, artisanal refining of petroleum products as seen in the Niger Delta among others. These activities Such impact however, is often assessed from changes in the physical, chemical [19] and biological components of the ecosystem.

Human health is largely determined by the diet and recommendable diet should be able to provide sufficient nutrients and low levels of pathogenic microorganisms, as well as chemical contaminants. Fish constitutes an important source of proteins, vitamins and unsaturated essential fatty acids (PUFA), especially omega-3 PUFA's [3]. In contrast to the potential health benefits of dietary fish intake, say in the prevention of coronary heart disease, an issue of concern related to frequent fish consumption is the risk derived from exposure to chemical pollutants. The pollution of the environment by PAHs is a major concern [20]. PAHs are large class of persistent organic pollutants containing two or more fused benzene

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rings. They are known to be ubiquitous in both marine and terrestrial environments [5] and are included in the EU and USEPA priority pollutant list due to their mutagenic and carcinogenic properties [28]. Predominant exposure route is dietary, excluding smokers and occupationally exposed populations. As chemically stable and lipophilic compounds [5], they can easily cross lipid membrane and have the potential to bioaccumulate in aquatic organisms. Based on physical and biological properties, PAHs are classified into high molecular weight (HMW) and low molecular weight (LMW) types. Those consisting of 4–6 aromatic rings are termed HMW and have been shown to be less readily bio-degraded by native microorganisms and can bioaccumulate in the aquatic organisms like fish and mussels. On the other hand, LMW PAHs consists of 2–3 aromatic rings and although less carcinogenic than HMW type [6] a pose toxic to many aquatic organisms. [32] reported the possibility of using Periwinkles and Oysters as pollution biomonitors due to the fact that they are sedentary or bottom feeders they are good accumulators of heavy. Although for most people, fish and seafood represents only a small part of the total diet, this trend may differ in the Niger Delta communities. European Union established a maximum level of $1 \mu\text{g g}^{-1}$ wet weight for benzo (a) pyrene in foodstuff and is used for the carcinogenic risk of PAHs in muscle meat of fish [11] but more recently, it was attributed to dibenzo(a,1) pyrene a carcinogenic potency that is about 100 times that of benzo(a) pyrene [22].

PAHs cause tainting on seafoods, have relatively low solubility and high affinity for particles, and most PAHs can therefore be found attached to particles that have settled or are suspended in the water column [25]. European Union has stressed and recommended that PAHs be measured in as wide as possible in food products in order to obtain data on the occurrence and specific concentrations in a variety of matrices [31].

Kporghor and Iko rivers are located in Eastern Obolo Local Government Area (LGA) of Akwalbom state (lying by Rivers state), and have been exposed to oil spill from the Shell Petroleum Development Company (Nigeria) Ltd pipeline, artisanal refining and pipeline vandalization. Sime (Tai) water is located in Tai LGA Rivers State (a known oil polluted zone), all in the Niger Delta region of Nigeria. Map of study area is as shown in Fig. 1 as reported in a study report by UNEP (2011). Average consumption of seafood in these communities is about 370 g per week going by measured dry weight of possible number at random per meal consumed. This study seeks to assess the levels of PAHs in commonly consumed and commercially viable fish and seafood species, *Periophthalmus koeleuteri* (Mudskipper), *Littorina littorea* (Periwinkle) and *Crasostrea virginica* (Oyster) from Nigerian coastal waters and identify the probable sources.

2. Material and methods

2.1. Sample collection and preparation

Fresh samples of *L. littorea*, *P. koeleuteri* and *C. virginica* were collected from landing beaches of Kporghor, Iko and Sime towns using harvesting buckets provided by local fishermen. At each site, ten individual Mudskippers, Periwinkles and Oysters of similar size and species were collected, cleaned and wrapped in aluminum foils, and kept frozen in an ice chest cooler for onward transportation to the laboratory for analysis.

2.2. Determination of polycyclic aromatic hydrocarbons levels

Fresh samples were well cleaned in distilled water to remove any external dirt. Dissection was performed on fresh samples, using instruments and glass dishes rinsed with solvent. Tissues

Table 1

PAHs levels ($\mu\text{g kg}^{-1}$ wet wt.) for *Littorina littorea* from the study areas (Sime, Kporghor and Iko coastal waters).

PAHs	Sime Tai	Kporghor	Iko
Naphthalene	BD	BD	BD
Acenaphthylene	BD	BD	BD
Acenaphthene	BD	BD	BD
Fluorene	BD	0.040 ± 0.004	BD
Phenanthrene	0.030 ± 0.004	$0.010^* \pm 0.002$	0.020 ± 0.003
Anthracene	0.020 ± 0.002	$0.010^* \pm 0.000$	0.020 ± 0.003
Fluoranthene	0.010 ± 0.000	0.030 ± 0.004	0.030 ± 0.004
Pyrene	0.030 ± 0.004	0.040 ± 0.004	0.040 ± 0.003
Benzo[a]anthracene	0.050 ± 0.004	$0.002^* \pm 0.004$	0.080 ± 0.004
Chrysene	$0.080^* \pm 0.004$	BD	$0.010^* \pm 0.002$
Benzo[b]fluoranthene	$4.240^* \pm 0.010$	13.300 ± 0.030	16.800 ± 0.040
Benzo[k]fluoranthene	0.004,001	BD	$0.040^* \pm 0.004$
Benzo[a]pyrene	$0.010^* \pm 0.000$	$0.010^* \pm 0.000$	$0.020^* \pm 0.003$
Indeno[1,2,3-cd]pyrene	$2.650^* \pm 0.010$	$22.400^* \pm 0.050$	$13.100^* \pm 0.020$
Dibenzo[a,h]anthracene	$0.010^* \pm 0.002$	0.002 ± 0.000	0.002 ± 0.000
Benzo[g,h,i]perylene	$0.020^* \pm 0.002$	0.010 ± 0.002	0.010 ± 0.002
Total	$7.150^* \pm 0.040$	$35.800^* \pm 0.100$	$30.100^* \text{h} \pm 0.090$
LMW-PAH/HMW-PAH	$0.010^* \pm 0.001$	$0.002^* \pm 0.000$	$0.001^* \pm 0.000$
BaA/(BaA + Chry)	0.380 ± 0.001	$1.000^* \pm 0.003$	0.890 ± 0.001

Means with superindices (*) across rows are significantly different ($P < 0.05$), values are mean \pm S.E.M ($n = 10$). BD = below detection limit of 0.0001.

were dissected and minced into smaller pieces, and a subsample was taken from the homogenate. The samples were then blended and kept in air tight containers prior to extraction process. Two grams of samples were weighed into a clean extraction container (50 ml beaker). A 10 ml analar grade extraction solvent (dichloromethane) was added into the sample and mixed thoroughly and allowed to settle. The mixtures were carefully filtered into clean solvent rinsed extraction bottle, using filter paper fitted into Buchner funnels. Transferred extracts were concentrated to $2 \mu\text{l}$ for cleanup/separation in gas chromatographic analysis (HP 5890 series II, GC apparatus, coupled with flame ionization detector (FID) HP Wilmington, DE, USA equipped with HP chemstation Rev. A 09:01 (10206) software). Elution protocol as given in instruction manual was strictly followed as in high pressure solvent extraction. Separation occurred as the vapor constituent partition between the gas and liquid phase and the sample was automatically detected as it eluted from the column (at constant flow rate) by the FID detector which response is dependent upon the composition of the vapor. To determine whether analyte detection was affected by the difference between diluent used for PAHs extraction and the experimental sample matrix, prepared standard curves were used to extrapolate the amount of added analyte in each case which denoted the spike recovered. There were no discrepancies observed. This was done by an addition of evenly spaced four (4) aliquots of spike to samples and these spiked aliquots were used to generate a calibration line and amount in the sample was calculated.

2.3. Statistical analysis

Means of ten replicates were subjected to ANOVA using Excel windows 10 and Duncan Multiple Range Test was employed for comparisons.

3. Results

From the results obtained, mean levels ($\mu\text{g kg}^{-1}$ wet wt.) of the sixteen PAHs distribution in *L. littorea*, *C. virginica* and *P. koeleuteri* collected from Sime, Kporghor and Iko coastal waters are as shown in Tables 1, 2 and 3 respectively. Also, low molecular weight: high molecular weight PAHs, LMW-PAH/HMW-PAH and Benzo(a) anthracene divided by the sum of Benzo(a) anthracene and Chrysene, BaA/(BaA + Chry) ratios



Fig. 1. Map of Ogoniland showing sampling area (UNEP, 2011).

[24] for all samples were shown. Average levels of individual PAHs ranged from below detection limit (BD) of analytical instrument to $22.400 \pm 0.050 \mu\text{g kg}^{-1}$ wet wt. with the highest ($22.400 \pm 0.050 \mu\text{g kg}^{-1}$ wet wt.) recorded for carcinogenic

Indeno[1,2,3-cd]pyrene for *L. littorea* from Kporghor coastal water. Similarly, total PAHs levels were 7.150 ± 0.040 , 35.800 ± 0.100 and $30.100 \pm 0.090 \mu\text{g kg}^{-1}$ wet wt. for Sime, Kporghor and Iko samples respectively (Fig. 2) and were statistically significant for *L. littorea*.

Table 2
PAHs levels ($\mu\text{g kg}^{-1}$ wet wt.) for *Crossostrea virginica* from the study areas (Sime, Kporghor and Iko coastal waters).

PAHs	Sime Tai	Kporghor	Iko
Naphthalene	BD	BD	BD
Acenaphthylene	BD	0.002 ± 0.00	0.010 ± 0.002
Acenaphthene	BD	0.003 ± 0.00	0.060 ± 0.004
Fluorene	BD	0.04 ± 0.004	0.120 ± 0.010
Phenanthrene	0.060 [*] ± 0.004	0.01 [*] ± 0.00	0.130 [*] ± 0.010
Anthracene	0.050 ± 0.010	0.05 ± 0.004	0.020 ± 0.002
Fluoranthene	0.090 [*] ± 0.004	0.490 ± 0.010	0.230 ± 0.010
Pyrene	0.050 ± 0.010	0.030 ± 0.002	0.050 ± 0.004
Benzo[a]anthracene	0.030 [*] ± 0.003	0.003 [*] ± 0.000	0.010 [*] ± 0.002
Chrysene	0.030 ± 0.003	BD	0.090 ± 0.004
Benzo[b]fluoranthene	2.720 [*] ± 0.010	87.400 [*] ± 0.030	63.90 [*] ± 0.004
Benzo[k]fluoranthene	0.020 ± 0.002	0.010 ± 0.000	0.060 [*] ± 0.010
Benzo[a]pyrene	0.070 ± 0.004	0.010 [*] ± 0.002	0.020 ± 0.002
Indeno[1,2,3-cd]pyrene	0.010 [*] ± 0.002	16.900 [*] ± 0.030	32.100 [*] ± 0.020
Dibenzo[a,h]anthracene	0.490 [*] ± 0.004	0.050 [*] ± 0.004	0.170 [*] ± 0.010
Benzo[g,h,i]perylene	0.050 ± 0.01	0.020 ± 0.002	0.180 [*] ± 0.004
TOTAL	3.670 [*] ± 0.050	105.000 ± 0.090	97.200 ± 0.090
LMW-PAH/HMW-PAH	0.500 ± 0.002	0.010 ± 0.000	0.004 [*] ± 0.000
BaA/(BaA + Chry)	0.500 [*] ± 0.010	0.030 [*] ± 0.001	0.100 [*] ± 0.002

Means with superindices (*) across rows are significantly different ($P < 0.05$), values are mean ± S.E.M ($n = 10$). BD = below detection limit of 0.0001.

Table 3
PAHs levels ($\mu\text{g kg}^{-1}$ wet wt.) in *Periophthalmus koeleuteri* from the study areas (Sime, Kporghor and Iko coastal waters).

PAHs	Sime Tai	Kporghor	Iko
Naphthalene	BD	BD	BD
Acenaphthylene	BD	0.002 ± 0.000	BD
Acenaphthene	BD	0.001 ± 0.000	BD
Fluorene	0.010 ± 0.000	0.003 ± 0.000	BD
Phenanthrene	0.030 ± 0.004	0.240 [*] ± 0.040	BD
Anthracene	BD	0.040 ± 0.003	BD
Fluoranthene	0.060 [*] ± 0.004	2.460 [*] ± 0.010	0.010 [*] ± 0.002
Pyrene	0.050 ± 0.010	0.090 ± 0.004	0.010 ± 0.000
Benzo[a]anthracene	0.003 [*] ± 0.000	0.002 [*] ± 0.000	0.030 [*] ± 0.010
Chrysene	BD	BD	0.090 ± 0.004
Benzo[b]fluoranthene	0.860 [*] ± 0.004	10.400 [*] ± 0.040	49.300 [*] ± 0.050
Benzo[k]fluoranthene	0.001 ± 0.000	0.001 ± 0.000	0.001 ± 0.002
Benzo[a]pyrene	BD	BD	0.002 ± 0.000
Indeno[1,2,3-cd]pyrene	171.000 [*] ± 0.430	32.700 [*] ± 0.030	3.670 [*] ± 0.010
Dibenzo[a,h]anthracene	0.002 ± 0.000	BD	0.001 ± 0.002
Benzo[g,h,i]perylene	0.001 ± 0.000	BD	0.004 ± 0.000
TOTAL	171.900 [*] ± 0.450	45.900 ± 0.120	53.100 ± 0.070
LMW-PAH/HMW-PAH	0.000 [*] ± 0.000	0.010 [*] ± 0.000	5.310 [*] ± 0.090
BaA/(BaA + Chry)	0.100 [*] ± 0.004	1.000 [*] ± 0.020	0.430 [*] ± 0.050

Means with superindices (*) across rows are significantly different ($P < 0.05$), values are mean ± S.E.M ($n = 10$). BD = below detection limit of 0.0001.

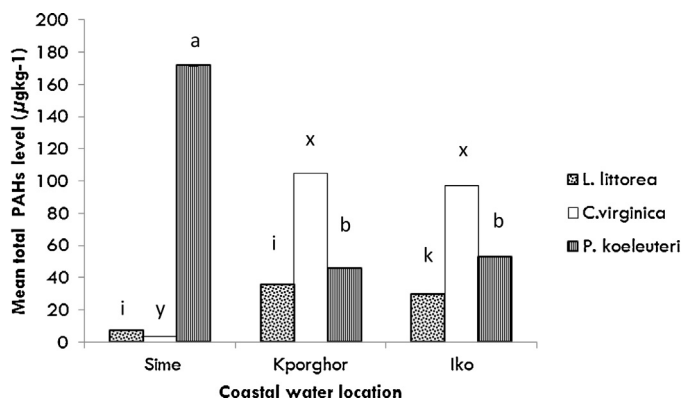


Fig. 2. Mean total PAHs level (mean ± SEM) in study samples from Sime, Kporghor and Iko coastal waters. Means with same alphabets are not significantly different ($P < 0.05$). Sime, Kporghor and Iko are various coastal water locations to show sample source.

Presence of Seriously carcinogenic B[a]P was observed (Fig. 3) for all samples. LMW-PAH/HMW-PAH ratios for samples from Iko waters (0.001 and 0.004) were markedly lower relative to the ratios found in Sime (0.002 and 0.010) and Kporghor (0.010 and 0.500) for *L. littorea* and *C. virginica* respectively. LMW-PAH/HMW-PAH ratios for *L. littorea* species from all the sites were < 1 indicating anthropogenic contributions. Attempted classifications of constituent PAHs based on carcinogenicity are also discussed.

4. Discussion

Useful ratio, BaA/(BaA + Chry) (Benzo(a) anthracene divided by the sum of Benzo(a) anthracene and Chrysene), was adopted to analyse possible sources of polycyclic aromatic hydrocarbons in study samples. *L. littorea* from Kporghor and Iko waters gave mean value > 0.890 while that from Sime water was markedly below. BaA/(BaA + Chry) ratios for *L. littorea* samples were 0.380, 1.000 and 0.890 from Sime, Kporghor, and Iko waters respectively. The BaA/(BaA + Chry) ratio for *L. littorea* from Kporghor is, however, markedly higher relative to the ratios found for Sime and Iko samples. This implies more of pyrogenic sources of PAHs as [21] reported that a BaA/(BaA + Chry) ratio > 0.350 indicates pyrogenic or combustion sources while those < 0.200 has been attributed to petrogenic sources. These sources are however indistinguishable for ratios in the range 0.200–0.350 [21]. Car-

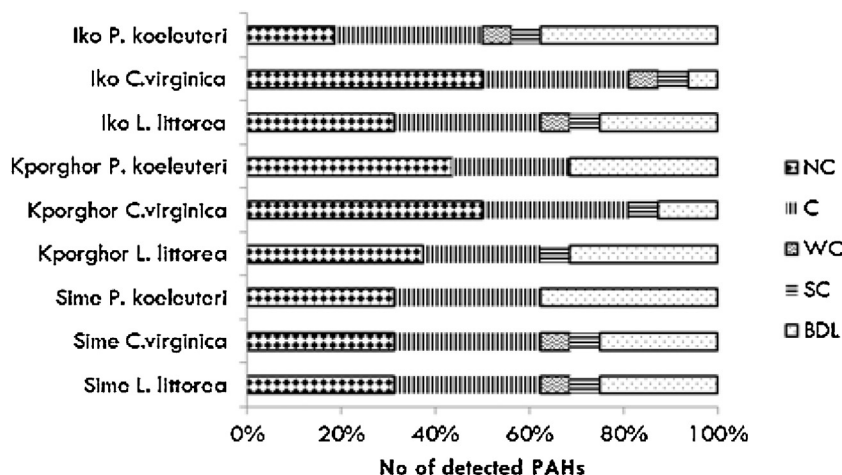


Fig. 3. Distribution of PAHs by type in study samples: BDL, SC, WC, C and NC represent below detection limit of the analytical instrument, Strongly carcinogenic, Weakly carcinogenic, Carcinogenic and Not carcinogenic PAHs, respectively. Iko, Kporghor and Sime appended to sample names (scientific) are various coastal water locations to show source.

cinogenic Benzo[b]Fluoranthene levels in *L. littorea* samples from Kporghor and Iko waters exceeded the European Union (EU) limit of $12.000 \mu\text{g kg}^{-1}$ wet wt. while those from Sime were below. Collectively, bioaccumulation of strongly and weakly carcinogenic (Fig. 3) PAHs was not expressed in *P. koeleuteri* samples harvested from Kporghor and Sime water bodies. Non-carcinogenic types however, were more in abundance of the total 16 PAHs observed.

Up to $171.900 \mu\text{g g}^{-1}$ wet wt. of PAHs was obtained for *P. koeleuteri* and far exceeded highest amount of 70.440 ng g^{-1} wet wt. obtained among five species of fish samples collected along the harbor line, Mumbai [9] and 3.365 ppm in edible fishes of the Gomti river, Lucknow, India [15].

After reviewing the available data on occurrence and toxicity of the 15 PAHs identified by SCF in 2002 and additionally BF which had been suggested by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 2005, the CONTAM Panel [10] recommended that groups of PAHs termed PAH4 (Benzo (a) pyrene, Chrysene, Benzo (a) anthracene and Benzo (b) fluoranthene) were better indicators of PAHs occurrence than Benzo (a) pyrene on its own in food. From this study *P. koeleuteri* samples ($49.420 \mu\text{g kg}^{-1}$) from Iko coastal waters far exceeded the EU regulatory limits of Maximum levels of $30 \mu\text{g kg}^{-1}$ wet wt. for PAH4 in Commission Regulation (EU) No 835/2011. In the same vein, samples of *C. virginica* from Kporghor and Iko coastal waters gave PAH4 values of $87.410 \mu\text{g kg}^{-1}$ and $63.930 \mu\text{g kg}^{-1}$ respectively and could be highly implicated in their potentials as carcinogens.

The sources of PAHs in coastal environment are described as either petrogenic (if the source is derived from petroleum, e.g. natural oil seepage and oil spills) or pyrogenic (if the source is derived from the incomplete combustion of organic matter and fossil fuel [1] and the ratio of low molecular weight PAHs (LMW-PAHs) to high molecular weight PAHs (HMW-PAHs) has been used to characterize the origin of PAHs in the environment [29,26]. Results gave global distribution pattern characterized by an abundance of heavy compounds illustrated by a median LMW-PAH/HMW-PAH ratio value of 0.002 (Table 1). This high concentration of heavy molecular weight PAHs indicates a predominant pyrolytic origin for the PAH pollution. This approach has been used to characterize the sources of PAHs in sediments and was adopted to characterize the sources of PAHs in the fish and seafoods analysed. Combustion processes, therefore appear as the main formation mechanism for the PAHs bioaccumulated in study species. Predominance of the pyrolytic origin may be linked to intense illegal refining in the region and other linear and diffused sources may be linked to the great number of linear and diffuse combustion sources.

Samples of *C. virginica* gave mean PAHs distribution in Sime, Kporghor and Iko coastal waters as shown in Table 2. A total of sixteen PAHs were detected of which 10%, 15% and 15% distribution pattern were seen in Sime, Kporghor and Iko water samples respectively. Average levels of these PAHs ranged from BD to $87.400 \pm 0.030 \mu\text{g kg}^{-1}$ wet wt. The highest average individual concentration of $87.400 \pm 0.030 \mu\text{g kg}^{-1}$ wet wt. was recorded for carcinogenic Benzo[b]Fluoranthene from Kporghor. Fig. 3 illustrates the uniform clear distribution of strongly carcinogenic PAHs in *C. virginica* samples across locations. The LMW-PAH/HMW-PAH ratios in Iko samples were markedly lower relative to those of Sime and Kporghor and were < 1 for *C. virginica* species from all the sites (Tables 1–3). The BaA/(BaA + Chry) ratios for *C. virginica* from Sime was, however, markedly higher relative to the ratios found in *C. virginica* from Kporghor and Iko waters. The BaA/(BaA + Chry) ratios for *C. virginica* from Sime and Iko waters were > 0.03 as against that of Kporghor. Indeno[1,2,3-cd]pyrene levels in *C. virginica* from Iko and Kporghor locations exceeded the European Union (EU) limit of $5 \mu\text{g kg}^{-1}$ wet wt. B[a]P recommended allowable daily intakes range from 0.04 to 0.42 g day^{-1} in Italy [14] for different foodstuffs and had its lower limit exceeded for *C. virginica* in this study and

could imply danger. Several carcinogenic PAHs have been reported in water and food samples [33]. As a result, the accumulation in the environment has become an issue of global health concern, the principal route of exposure to man, being the consumption of contaminated water, foods and marine products most especially fish [13,2].

Mean PAHs distribution in *P. koeleuteri* collected from study locations are as shown in Table 3, alongside their LMW-PAH/HMW-PAH and BaA/(BaA + Chry) ratios. A total of 16 PAHs were detected and average individual levels of these PAHs ranged from BD to $171 \pm 0.430 \mu\text{g kg}^{-1}$ wet wt. The highest level of $171 \pm 0.430 \mu\text{g kg}^{-1}$ wet wt. was recorded for Indeno[1,2,3-cd]pyrene from Sime. Total PAHs levels in this sample were significantly higher ($171 \pm 0.450 \mu\text{g kg}^{-1}$ wet wt.) in Sime samples (Fig. 2). The LMW-PAH/HMW-PAH ratios for Sime samples were markedly lower ($p \leq 0.05$) relative to the ratios found in Kporghor and Iko waters. BaA/(BaA + Chry) ratios for *P. koeleuteri* were 0.100, 1.000 and 0.430 for Sime, Kporghor, and Iko waters respectively. The BaA/(BaA + Chry) ratios for *P. koeleuteri* from Kporghor, is markedly higher relative to the ratios found for Sime and Iko samples. Carcinogenic Benzo[b]fluoranthene levels in *P. koeleuteri* from Iko location exceeded the European Union (EU) limit of $12 \mu\text{g kg}^{-1}$ wet wt. It is important to note that observed PAHs levels exceeded reported measurable but low levels in fresh fish and seafoods studied in similar communities [27]. For Sime, *C. virginica* accumulated significantly lower ($P < 0.05$) concentrations of total PAHs than *L. littorea* and *P. koeleuteri*. This is possibly due to local physical mixing, as a result of fishing activities, exposing the fishes to PAHs irrespective of where these fishes may be found. The observed differences in PAH bioaccumulation in study species may also be attributed to differences in feeding preferences and general behavior [12]. The LMW-PAH/HMW-PAH ratios indicate that the HMW-PAHs were generally predominant compared to the LMW-PAHs. The predominance of HMW-PAHs may be due to preferential degradation during PAHs transport and burial into sediments [4].

Levels of PAHs in *L. littorea*, *C. virginica* and *P. koeleuteri* reflect the state of contamination of the environment, although highest PAHs level of $171.900 \mu\text{g kg}^{-1}$ wet wt. for Sime samples (Table 3), with less distribution of carcinogenic PAHs (Fig. 3), illustrates the less carcinogenicity of its heterogenous components. The LMW-PAH/HMW-PAH ratios observed in the four species from all three locations were < 1 , indicating that the sources of these PAHs in study species are mainly pyrogenic. This is thus a clear indication of anthropogenic pollution of PAHs in the coastal marine environment. The observed BaA/(BaA + Chry) ratios for the three species from all the locations were > 0.350 except *C. virginica* (Kporghor and Iko locations) and *P. koeleuteri* (Sime location). This also indicated abundant pyrogenic sources of PAHs contamination. Possible anthropogenic sources include combustion of petroleum, automobile tyre, wood and vehicle emissions. Bioaccumulation of strongly carcinogenic PAHs was least with *P. koeleuteri* and hence it may not constitute a good biomonitor. The coastal people who tend to consume larger quantities of fish [30] could be at a greater risk to health issues like growth reduction, endocrine alteration [16], cancer, mutations and birth defects. Also, malformations of embryo and larvae [8] and DNA damage [7] in fish could give residual effects in man. Suitable clean-up technologies, awareness campaign and mitigation measures, are therefore recommended.

5. Conclusion

The present study demonstrated the suitability of *C. virginica* $> L. littorea$ in environmental monitoring over *P. koeleuteri* and also underscored the need for a holistic characterization in monitoring and response as bulk concentrations could be misleading. Gener-

ally, observed elevated PAHs in fish and seafoods harvested from 3 coastal waters in the Niger Delta region were implicated to be of anthropogenic origin. The investigation of the aromatic compound distributions in all of the 30 fish and seafood samples have underlined that there is a heterogeneous PAHs background pollution. All of degradative activities and their effects left an environmental footprint and needs attention.

Transparency document

The [Transparency document](#) associated with this article can be found in the online version.

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