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# Distribution, source identification and eco-toxicological risks of PAHs in sediments of Aba River at Ogbor-Hill region, Nigeria

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## ABSTRACT

The distribution, identification of potential sources, and eco-toxicological risks assessment of polycyclic aromatic hydrocarbons (PAHs) in the sediments of Aba River at Ogbor Hill segment in Aba, South-Eastern Nigeria was investigated. Composites samples were collected from sediments at five (5) different locations (LT-A, LT-B, LT-C, LT-D & LT-E), and a Soxhlet extraction with dichloromethane (DCM) and GC-MS was utilized to simultaneously determine the sixteen (16) US EPA listed priority PAHs. The total PAHs (∑16PAHs) concentrations was in 481.95–2,562.55 µg/Kg range with average 1,122.64±839.70 µg/Kg. The total PAHs range was below the 4,022 µg/Kg Effect Range Low (ERL) target value for sediment quality guidelines (SQG) indicating that biological health effects would rarely be observed. The dominant PAHs in the sediments were: Nap (13.26%), Flr (11.14%), DaA (11.14%), BaA (9.90%), Pyr (9.15%), and Acy (8.42%). However, the major contributors to sediments' toxicities were: DaA (65.32%), BaP (23.55%), BaA (5.80%), BbF (2.27%), and IcP (1.36%). The order of toxicities of the locations based on normalized toxicity equivalence quotients (TEQs) were; LT-B > LT-E > LT-D > LT-A > LT-C. Source diagnostic ratios of PAHs gave range of: ∑LMW/∑HMW (0.75–2.73), Ant/(Ant + Phe) (0.14–0.80), Flt/(Flt + Pyr) (0.10–0.77), BaA/ (BaA + Chr) (0.83–1.00), and IcP/ (IcP + BgP) (0.5); suggesting that the anthropogenic source of PAHs was of mixed origin–combustion (pyrogenic) and unburned petroleum. The result revealed the accumulation of PAHs in the sediments at elevated level, which need to be monitored regularly to avoid their negative impact on the living organisms.

**Capsule Summary:** This study was conducted to assess the distribution and source of polycyclic aromatic hydrocarbons (PAHs) along with their eco-toxicological risks assessment in the sediments of the Aba River.

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## INTRODUCTION

Pollution of River bodies hinders its capacity to support and sustain life; productivity, stability, species abundance and diversity, as well as the physiological conditions of indigenous organisms may be largely affected (Chokor, 2021a). Sediments represent natural parts of the aquatic habitats whose quantities and characteristics affect the physical, chemical, and biological quality of the aquatic ecosystems (US EPA, 2006). They generally act as sink for many pollutants – particularly Persistent Organic Pollutants (POPs). Also, they frequently act as a source for the presence of the same pollutants in the water column as processes such as desorption and re-suspension help to recycle pollutants through the aquatic environment. Sorbed pollutants such as PAHs released to the water as an extended source may threaten the aquatic ecosystem through bioaccumulation in food chains. Thus, sediment quality may impact the overall integrity of the aquatic ecosystem even long after the external sources of contaminants have been halted. The presence of persistent organic pollutants and their accumulations in River sediments had been a source of concern because of their mutagenic and carcinogenic potentials (Tongo *et al.,* 2017; Patel et al., 2020). Polycyclic aromatic hydrocarbons (PAHs) represent one of such group of persistent organic pollutants in the river sediments. PAHs are composed of two or more fused aromatic rings with no heteroatoms or substitutes attached. Their inherent properties such as: aromatic ring structures, hydrophobic nature, and thermo-stability have made them highly resistance to degradation hence their persistence in various environmental media where they exhibit wide range of biological toxicity.

PAHs have been identified to exhibit mutagenic, carcinogenic, teratogenic, and immunotoxic effects to various life forms (Burcheil and Gao, 2014; Rengarajan et al., 2015; Abdel-Shafy and Mansour, 2016; Bolden et al., 2017; Patel et al., 2020). The impacts of PAHs on organisms' health are dependent on various factors such as: mode of exposure, exposure duration, exposure dose, pre-health status and age of the organisms etc. However, the general acute effects of PAHs on human include: eye and skin irritations and inflammation, vomiting, diarrhoea, and confusion (Abdel-Shafy and Mansour, 2016; Patel et al., 2020). The chronic health effects include eye cataracts, kidney, liver, and lungs damages and malfunctions, breathing problems and asthma-like symptoms, as well as decreased immune function (Abdel-Shafy and Monsour, 2016). Long-term exposure to PAHs is also associated with tumour formation in skin, lung, oesophagus, colon, pancreas, bladder and women breasts (Yu, 2002; Rajpara et al., 2017). The concern for the environmental and health impacts of PAHs has necessitated the United States Environmental Protection Agency (US EPA) to enlist sixteen (16) PAHs in her list of priority pollutants, viz: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Acp), fluorine (Flr), anthracene (Ant), phenantrene (Phe), fluoranthene (Flt), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DhA), indeno[1,2,3-cd]pyrene (IcP), and benzo[g,h,i]perylene (BgP).

PAHs get into water bodies from petroleum spills, industrial and municipal waste water discharges, urban runoff, commercial shipping activities, atmospheric fallout of automobile exhaust and combustion activities, indiscriminate dumping of petroleum products on water ways, etc. (Adeniji et al., 2018; Chokor, 2021b). The source of PAHs may be natural or anthropogenic; Biogenic PAHs synthesized by biological species like microorganisms, phytoplankton, algae, and plants during conversion of organic materials account for most natural inputs of PAHs (Mojiri et al., 2019). Emissions from volcanic eruptions, natural forest fire and moorland fire caused by lightning flashes and diagenesis of organic matter in anoxic sediments are other natural sources but of less significant (Abdel-Shafy and Mansour, 2016; Patel et al., 2020). PAHs are found in coal and coal products, crude oil, and refined petroleum products. They are also formed during incomplete combustion of fossil fuels, woods, grasses, and other organic substances (Readman et al., 2002; Maioli et al., 2011). Incinerations of wastes, and industrial processes such as coal gasification, production of aluminium, iron, steel, and petroleum refining also results in the generation of PAHs as by-products (Igwo-Ezikpe et al., 2010; Esedafe et al., 2015). Generally, the anthropogenic sources of PAHs may be classified as either petrogenic (inputs from petroleum or related products such as oil spills, road construction materials like coke, carbon black, coal tar, and asphalt) or pyrogenic (due to combustion processes like fossil fuel combustion, electric power generation, refuse Incineration, home heating and industrial emissions) (Adeniji et al., 2018).

Various diagnostic indices have been used to distinguish the anthropogenic source of PAHs in environmental media. This distinction is anchored on the uniqueness of PAH composition and distribution as a function of the emission source (Soclo et al., 2000; Zhang et al., 2004). They include: Ant/(Ant + Phe), Flt/(Flt + Pyr), BaA/(BaA + Chr), IcP/(IcP + BgP) and ∑LMW/∑HMW (Yunker et al., 2002; Tolosa et al., 2004; Guo et al., 2007; Maioli et al., 2011). These ratios are based on the differences in heat of formation at the same molecular mass isomers. The pyrogenic processes involve formation of stable isomers from fast reactions (the kinetic product) whereas, digenesis favours the most stable isomers by increasing the time of reaction (the thermodynamic product) (Yunker et al., 2012; Brewster et al., 2019). Thus, petrogenic inputs will show lesser values for the above first four (4) ratios, and higher values for pyrolytic source. The reverse is the case for the ∑LMW/∑HMW ratio. Though, PAHs exist naturally in the environment, Industrialization and rapid urbanization with its attendant increased in anthropogenic activities is a major reason for their large presence in water bodies and consequent accumulation in sediments. The Ogbor Hill area of Aba coasting the Aba River popularly known as Waterside River by the indigenes is a densely populated community in Abia State. The inhabitants often depend on the waterside for their domestic water supply particularly in the dry season due to shortage in availability of potable public water. The river also serves as a source of recreational and commercial activities.

## MATERIAL AND METHODS

### Study area

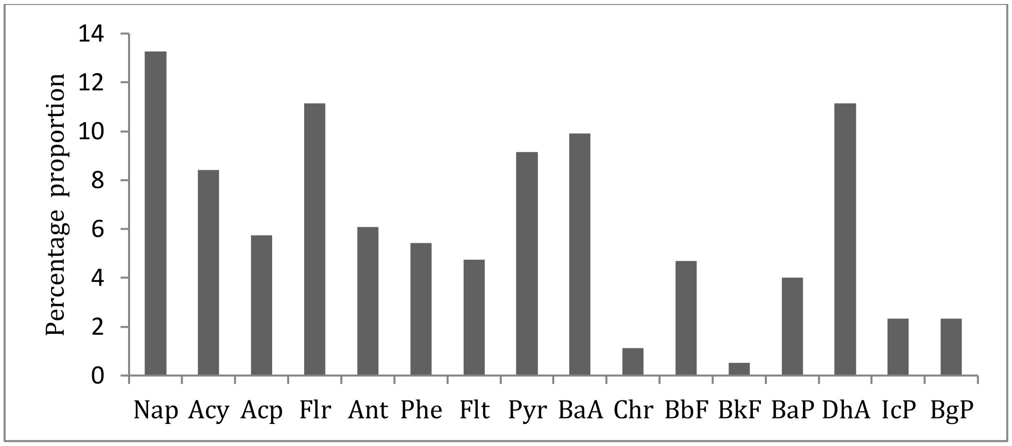

**Fig. 1:** Percentage proportions of individual PAH to the total mean PAHs (∑16PAHs) concentration in the sediments

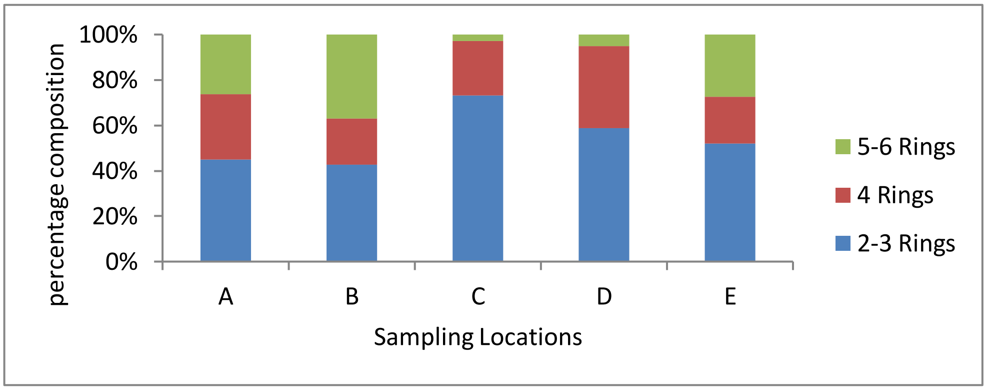

**Fig. 2:** PAHs composition in sediments showing percentage proportion of 2 and 3 rings, 4 rings, and 5 and 6 rings PAHs.

The Aba River also known as the Waterside River, takes it source from Okpu-Umuobu town, some kilometers from Aba town and flows North-South direction before joining the Imo River. It is recharged by precipitation and groundwater. Five composite sediments samples were collected at five different locations at some kilometres apart downstream beginning from LT-A., and their coordinates were properly recorded viz: LT-A (N05°07’20.74” E07°22’34.47”), LT-B (N05°06’45.67” E07°22’53.21”), LT-C (N05°06’30.87” E07°22’58.58”), LT-D (N05°06’09.65” E07°23’10.73”) and LT-E (N05°07’01.64” E07°23’19.11”). The area – both banks of the river is characterized by the presence of: factories (such as Nigerian Breweries, 7UP Bottling Company PLC, International Glass industry, Soap and Detergent factories etc), artesian workshops (like auto-mechanic workshops, iron welding and fabrications workshops), auto-refueling stations, abattoirs, and dumpsites. The major activities taking place at the river’s sides include: hide and skin burning (using vehicular tyres as fuel), commercial shipping, and regular vehicular transportation, as well as indiscriminate dumping of waste on the river. Effluents from these factories as well as run-off from the town also find their way into the river.

### Sample collection

Sediment samples were taken from the bottom surface (5–10cm) with a stainless steel Van Veen grab sampler aboard a fishing boat. At each location, three samples were collected randomly and composited to form a representative sample for each location. Samples were placed in pre-cleaned wide-mouth amber bottles and kept in ice chest at temperature below 4°C for onward transfer to the laboratory for analysis.

### PAHs extraction

Sediment samples were air dried in the dark for a period of five (5) days. The samples were homogenized and sieved through 0.5mm mesh. The aliquot of the sieved air-dried samples (10 g) was mixed with sufficient quantity of anhydrous sodium sulphate ($Na_2SO_4$) (about 5 g) to eliminate moisture, spiked with surrogate standard (10 µg/mL of σ-Terphenyl and 2-Fluorobiphenyl), wrapped in a filter paper, placed in a thimble and then loaded into the main chamber of the soxhlet extractor. Extraction was performed with dichloromethane (DCM) for 24 hr. Extracts were dried by passing through column of anhydrous sodium sulphate; and

**Table 1:** Concentrations (μg/Kg) of PAHs in the River sediments

| compound | Ring No | Concentrations (μg/Kg) | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | A | B | C | D | E | Average | ERL | ERM |
| Nap | 2 | 35.89 | 554.76* | 46.14 | 101.52 | 6.15 | 148.89 | 160 | 2100 |
| Acy | 3 | 63.58* | 184.58* | 65.63* | 103.57* | 55.37* | 94.54* | 44 | 640 |
| Acp | 3 | 52.30* | 59.47* | 65.63* | 121.00* | 23.58* | 64.40* | 16 | 500 |
| Flr | 3 | 169.20* | 169.20* | 89.21* | 84.09* | 113.82* | 125.10* | 19 | 540 |
| Ant | 3 | ND | 101.52 | 46.14 | 168.17 | 24.61 | 68.09 | 843 | 1100 |
| Phe | 3 | ND | 26.64 | 39.99 | 93.31 | 145.61 | 60.91 | 240 | 1500 |
| Flt | 4 | ND | 37.94 | 77.93 | 138.43 | 12.31 | 53.32 | 600 | 5100 |
| Pyr | 4 | ND | 360.95 | 23.58 | 90.24 | 38.97 | 102.75 | 665 | 2600 |
| BaA | 4 | 190.73 | 113.82 | 14.36 | 153.81 | 83.06 | 111.16 | 261 | 1600 |
| Chr | 4 | 14.36 | 5.13 | ND | 30.76 | 13.33 | 12.72 | 384 | 2800 |
| BbF | 5 | ND | 148.69 | ND | 58.45 | 55.37 | 52.50 | NA | NA |
| BkF | 5 | ND | ND | ND | ND | 29.74 | 5.95 | NA | NA |
| BaP | 5 | ND | 196.88 | 5.13 | ND | 23.58 | 45.12 | 430 | 1600 |
| DhA | 5 | ND | 603.98* | 2.05 | ND | 19.48 | 125.10* | 63.4 | 260 |
| IcP | 6 | 94.34 | ND | 3.08 | ND | 32.81 | 26.05 | NA | NA |
| BgP | 6 | 94.34 | ND | 3.08 | ND | 32.81 | 26.05 | NA | NA |
| ∑16PAHs | | 714.73 | 2562.55 | 481.95 | 1143.36 | 710.62 | 1122.64 | 4022 | 44792 |

*:concentrations higher than the ERL; ND: not detectable; ERL: Effects Range Low; ERM: Effects Range Median; Nap: naphthalene; Acy: acenaphthylene; Acp: acenaphthene; Flr: fluorine; Ant: anthracene; Phe: phenantrene; Flt: fluoranthene; Pyr: pyrene; BaA: benzo[a]anthracene; Chr: chrysene; BbF: benzo[b]fluoranthene; BkF: benzo[k]fluoranthene; BaP: benzo[a]pyrene; DhA: dibenzo[a,h]anthracene; IcP: indeno[1,2,3-cd]pyrene; and BgP: benzo[g,h,i]perylene.

with the aid of rotatory evaporator, the extracts were reduced to about 2mL (Iyang et al., 2018; Chokor, 2022).

### Sample clean-up and separation

A silica gel chromatographic column lined at the top with anhydrous $Na_2SO_4$ (2cm thick) was used for the clean-up of extracts. Concentrated extracts were transferred into the column (10mm i.d. X 30cm), and eluted first with 30mL of n-hexane to separate the hydrocarbon fraction and then with 30mL of DCM to obtain the aromatic fraction. The aromatic fractions were concentrated to about 2mL with rotatory evaporator at 30$^{o}$C; 1.5mL of it was transferred into chromatographic vial and stored at 4$^{o}$C awaiting gas chromatographic analysis. A blank sample was processed the similar way for the purpose of quality assurance (Maioli et al., 2011; Adeniji et al., 2017; Iyang et al., 2018; Chokor, 2022).

### Gas chromatographic analysis

Polycyclic aromatic hydrocarbons (PAHs) analyses were performed with the aid of gas chromatography coupled to a mass spectrometer as a detector (GC-MS); Agilent 6890N system equipped with DB-5 capillary column with dimension of 30m X 0.32mm X 0.25μm. pure helium gas at a flow velocity of 1mL/min was used as the carrier gas. The samples were injected into the GC via a pulsed split-less mode with an injection volume of 1μL. The initial chromatographic column temperature was 70°C, which was held for 20 min, and then increased at 25°C min$^{-1}$ to 150°C; it was further raised to 200°C at 3°C min$^{-1}$, and finally increased to 300°C at 2°C min$^{-1}$. The temperature of the injection port, ion source, quadrupole and transfer line were 250, 230, 150 and 280°C respectively.

### Identification and quantification

PAHs were identified by comparing their retentions time with those of corresponding standards, and quantification was done using response factors related to the respective internal standards based on five-point calibration curve for the individual PAH. Deuterated PAH internal standard solutions (naphthalene-d8, acenaphthene-10, phenanthrene-d10, chrysene-d12, and perylene-d12) and surrogate standard solutions (2-fluorobiphenyl and 4-terphenyl-d14) were used for sample quantification and quantifying procedural recovery.

### Determination of organic carbon

The organic carbon contents of the sediments were determined using Standard methods (Nelson and Sommer, 1982).

## RESULTS AND DISCUSSION

### PAHs concentrations and distributions

PAHs in the sediments of Aba River at Ogbor Hill section are as shown in table 1. Benzo(k)fluoranthene (BkF) was not detected in any other locations except LT-E (29.74 µg/Kg). There were also undetectable levels of Ant, Phe, Flt, Pyr, BbF, BkF, BaP, and DhA in LT-A. IcP and BgP were undetected in LT-B, while LT-C had Chr, BbF and BkF below detection limits. Samples taken at LT-D, had BkF, BaP, DhA, IcP and BgP also below detection limits. The mean concentrations of individual PAH ranged from 5.95–148.89 µg/Kg while, the total PAHs concentrations for the different locations ranged from 481.95–2,562.55 µg/Kg. Location B (LT-B) had the highest value (2,562.55 µg/Kg) while, the lowest was from LT-C (481.95 µg/Kg). Values in other locations were: LT-A (714.73 µg/Kg), LT-D (1,143.36 µg/Kg), and LT-E (710.62µg/Kg). The mean for total PAHs (∑16PAHs) in all locations was 1122.64±839.70 µg/Kg. The large variations in values for the various locations as indicated by the huge standard deviation tend to implied that the source of PAHs in the river bed may be largely anthropogenic. The predominant PAHs (Fig.1) in the studied area were: naphthalene (Nap), fluorene (Flr), dibenzo(a,h)anthracene (DhA), benzo(a)anthracene (BaA), pyrene (Pyr), and acenaphthylene (Acy), with an average concentrations of 148.89; 125.10; 125.10; 111.16; 102.75; and 94.54 µg/Kg. respectively. This corresponded to 13.26; 11.14; 11.14; 9.90; 9.15; and 8.42% of the total PAHs respectively and a summation of 63.01% of the total PAHs (Fig.1).

The mean values for the seven carcinogenic PAHs (∑7CPAHs) present in the river sediments were: benzo(a)anthracene (111.16µg/Kg), benzo(a)pyrene (45.12 µg/Kg), benzo(b)fluoranthene (52.50 µg/Kg), benzo(k)fluoranthene (5.95 µg/Kg), chrysene (12.72 µg/Kg), dibenzo(a,h)anthracene (125.10 µg/Kg), and indeno(1,2,3-cd)pyrene (26.05 µg/Kg), which amounted to 33.72% of the total PAHs concentration.

## Comparison of Total PAHs

Essien et al (2011) reported value range of 6,100–35,270 µg/Kg total PAHs for sediments of the Iko River estuary mangrove system, Nigeria. Oyo-Ita et al (2018) also reported total PAHs (∑PAHs) range of 1,670–20,100 µg/Kg with a mean of 9,370 µg/Kg for the River Calabar, Nigeria, while, a mean total PAHs value of 4,587.70 µg/Kg was reported for sediments of the Warri River at Ubeji, Niger Delta, Nigeria (Asagbra et al., 2015). The range (481.95–2,562.55 µg/Kg) and the mean (1122.64 µg/Kg) of these studies were comparably lower than those of these authors.

The range of total PAHs (∑16PAHs: 481.95–2562.55 µg/Kg) obtained in this study was within the 440-21,600 and 206–13,710 µg/Kg reported by Edokpayi et al (2016) respectively for the Mvudi and Nzhelele rivers in Vhembe District, South- Africa. Elsewhere around the world, the range of values were also comparable to: the 23–2,534 µg/Kg reported for the Donggang River, Taiwan (Hsieh et al., 2010), the 5.6–1,187 µg/Kg observed for the Negro River, Brazil (Souza et al., 2015), and the 5.24–3,722.87 µg/Kg recorded for Gomti River, India (Malik et al., 2011). It was however, lower than the 796.2–10,470 µg/Kg (mean: 2,713.6µg/Kg) obtained by Zhao et al (2017) for the sediments of River Qinhuai and lake Xuaniou, Nanjing, China, and the 774.81–255,371.9 µg/Kg reported for the Haihe River, in the same China by Jiag et al (2007). The range were on the other hand, much higher than the 27-418 µg/Kg recorded for sediments of Estero de Urias, Estuary in Mexico (Jaward et al., 2012), and the 97.2–04.8 µg/Kg reported for Yellow River Estuary, China (Hu et al., 2014).

The mean of this study was also lower than the 11,711 µg/Kg and 7,035 µg/Kg reported for respective sediments of Mutshundudi and Nzhelele Rivers in Vhembe District of South Africa by Nekhavambe et al (2014). It was also much lower than the 5,875.00 µg/Kg reported by Brewster et al (2019) for the sediments core of lower Fox River, Wisconsin, US, and the 52,000 µg/Kg recorded for the Porto Atlantic Coast of Portugal (Rocha et al., 2017). It was however higher than the 65.22 µg/Kg reported for sediment of the Nakdong River estuary, South Korea (Lee et al., 2017), and the 195 µg/Kg recorded by Bastami et al (2014) for sediment of Hormuz Striat, Persian Gulf.

## Compositions of PAHs in the sediments

The PAHs are normally classified as low molecular weight (LMW) and high molecular weight (HMW) PAHs. Two and three ring PAHs are regarded as LMW while, those with four or more rings are considered as HMW PAHs. Fig. 2 shows the distributions of various rings forms of PAHs in the sediments. There was much abundance of low molecular weight hydrocarbons (LMW) particularly in location C (LT-C) with 73.19% of it composed of 2 – 3 rings PAHs, two other locations – D and E - had 58.74 and 51.95% LMW hydrocarbons, while LT- A and LT-B were composed of 44.91 and 42.74% low molecular weight hydrocarbons (2 -3 ring PAHs). Four (4) rings PAHs represented 28.69; 20.21; 24.04; 36.14; and 20.78 percent of the total PAHs in locations A, B, C, D, and E respectively. The percentage contributions of five and six member rings PAHs were: LT-A (26.40), LT-B (37.05), LT-C (2.77), LT-D (5.11), and LT-E (27.27%).

## PAHs toxicological risks in the sediments

Baumard et al (1998) suggested classification of PAHs pollutant level as: low (0-100µg/Kg), moderate (100–1,000 µg/Kg), high (1,000–5,000 µg/Kg) and very high (>5,000 µg/Kg). Thus, sediments from this study area can be characterized based on this classification as having moderate to high PAHs pollution. However, sediments' PAHs potential toxicity may be assessed by comparing the values with the United State National Oceanic sediment quality guidelines–Effect range low (ERL) and Effect range median (ERM) target values. The values were produced by correlations of observed PAHs concentrations in sediments

**Table 2:** TEFs and TEQs values of the 16 priority PAHS in the sediments

| PAHs | TEFs | TEQs (µg/Kg dw) | | | | | | Mean Contribution (%) |
|---|---|---|---|---|---|---|---|---|
| | | LT-A | LT-B | LT-C | LT-D | LT-E | Ave | |
| Nap | 0.001 | 0.03589 | 0.55476 | 0.04614 | 0.10152 | 0.00615 | 0.148892 | 0.078 |
| Acy | 0.001 | 0.06358 | 0.18458 | 0.06563 | 0.10357 | 0.05537 | 0.094546 | 0.049 |
| Acp | 0.001 | 0.0523 | 0.05947 | 0.06563 | 0.121 | 0.02358 | 0.064396 | 0.034 |
| Flr | 0.001 | 0.1692 | 0.1692 | 0.08921 | 0.08409 | 0.11382 | 0.125104 | 0.065 |
| Ant | 0.01 | ND | 1.0152 | 0.4614 | 1.6817 | 0.2461 | 0.68088 | 0.356 |
| Phe | 0.001 | ND | 0.02664 | 0.03994 | 0.09331 | 0.14561 | 0.06111 | 0.032 |
| Flt | 0.001 | ND | 0.03794 | 0.07793 | 0.13843 | 0.01231 | 0.053322 | 0.028 |
| Pyr | 0.001 | 19.073 | 0.36095 | 0.02358 | 0.09024 | 0.03897 | 0.102748 | 0.054 |
| BaA | 0.1 | 0.1436 | 11.382 | 1.436 | 15.381 | 8.306 | 11.1156 | 5.804 |
| Chr | 0.01 | 0.1436 | 0.0513 | ND | 0.3076 | 0.1333 | 0.12716 | 0.066 |
| BbF | 0.1 | ND | 14.869 | ND | 5.845 | 5.537 | 5.2502 | 2.748 |
| BkF | 0.1 | ND | ND | ND | ND | 2.974 | 0.5948 | 0.311 |
| BaP | 1.0 | ND | 196.88 | 5.13 | ND | 23.58 | 45.118 | 23.56 |
| DhA | 1.0 | ND | 603.98 | 2.05 | ND | 19.48 | 125.102 | 65.326 |
| IcP | 0.1 | 9.434 | ND | 0.308 | ND | 3.281 | 2.6046 | 1.360 |
| BgP | 0.01 | 0.9434 | ND | 0.0308 | ND | 0.3281 | 0.26046 | 0.136 |
| ∑C7TEQs | | 28.6506 | 827.1623 | 8.924 | 21.5336 | 63.2913 | 189.9124 | 99.17 |
| ∑C16TEQs | | 29.91497 | 829.571 | 9.82431 | 23.94746 | 64.26131 | 191.5038 | 100 |

∑C7TEQs: Toxic equivalent quotient for the seven carcinogenic PAHs, ∑C16TEQs: TEQs value for the 16 PAHs, Nap: naphthalene; Acy: acenaphthylene; Acp: acenaphthene; Flr: fluorine; Ant: anthracene; Phe: phenantrene; Flt: fluoranthene; Pyr: pyrene; BaA: benzo[a]anthracene; Chr: chrysene; BbF: benzo[b]fluoranthene; BkF: benzo[k]fluoranthene; BaP: benzo[a]pyrene; DhA: dibenzo[a,h]anthracene; IcP: indeno[1,2,3-cd]pyrene; and BgP: benzo[g,h,i]perylene.

**Table 3:** Diagnostic Ratios and source apportionment of PAHs in Sediments

| Sites | ∑LMW/∑HMW | Ant/(Ant+Phe) | Flt/(Flt+Pyr) | BaA/(BaA+Chr) | IcP/(IcP+BgP) |
|---|---|---|---|---|---|
| A | 0.82 | ∞ | ∞ | 0.93 | 0.50 |
| B | 0.75 | 0.80 | 0.10 | 0.96 | ∞ |
| C | 2.73 | 0.54 | 0.77 | 1.00 | 0.50 |
| D | 1.42 | 0.64 | 0.61 | 0.83 | ∞ |
| E | 1.08 | 0.14 | 0.24 | 0.86 | 0.50 |

to the toxicity response of marine organisms or communities. Concentrations below ERL indicate a minimal-effect range in which effects should be rare.

Concentrations equal to or above the ERL, but below the ERM, connote the possible effects-range in which adverse biological effects would occur occasionally. Concentrations equal or above the ERM signify a probable-effects range in which negative health effects would occur frequently (Long et al., 1995; McGrath et al., 2019). Table 1 shows the concentrations of PAHs in the sediments in comparison with ERL and ERM target values. Values of individual PAH in all the sites were below their ERM. All the sites had Acy, Acp, and Flr above their ERL, while only site B had Nap and DhA above their ERL target values. The other individual PAHs values were below their ERL. On the average, Acy, Acp, Flr, and DhA were noted to be above ERL but below ERM. Thus, this individual PAH (Acy, Acp, Flr, and DhA) may occasionally exhibit adverse health effects at these locations. The total PAHS (∑16PAHs) range (481.95–2,562.55 µg/Kg) and mean total (1122.64 µg/Kg) were however, below the 4022 µg/Kg ERL target value tending to suggest that total PAHs level in the sediments would show only minimal-effects range that is, effects would rarely be observed at this level of concentrations. Johnson et al (2006) however, proposed a sediment quality guideline of 1,000 µg/Kg dw to protect estuarine fish against several important health effects. The results of this study show sites B and D to have exceeded this guideline while the other sites were less than it. The toxicity of seven carcinogenic and nine non-carcinogenic PAHs was also assessed using the toxicity equivalence factors (TEFs). Benzo(a)pyrene is regarded as one of the most carcinogenic PAHs and is usually used as an exposure marker for risk assessments (Nisbet and LaGoy, 1992; USEPA, 2003; Lee and Vu, 2010). The benzo(a)pyrene toxic equivalence quotient (TEQs) was calculated using Eq. 1.

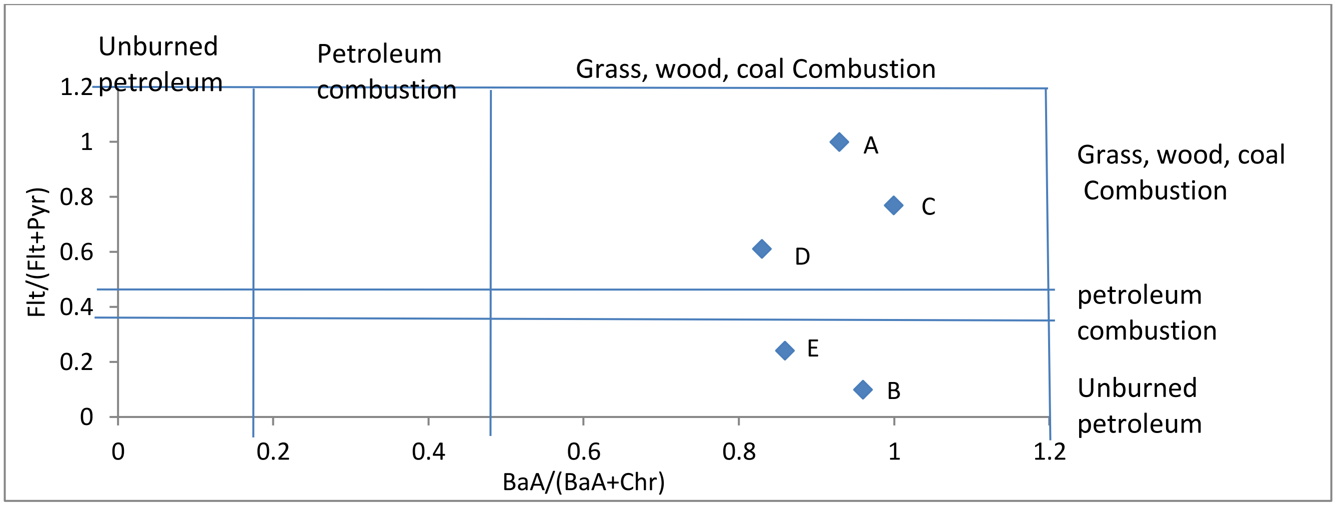

**Fig. 3:** PAHs cross plot for the ratios of Flt/(Flt+Pyr) versus BaA/(BaA+Chr).

$$\text{Total TEQ} = \sum C_i \times TEF_i \quad (1)$$

Where, TEQ = toxic equivalent quotient, $C_i$ = concentrations of individual PAHs, $TEF_i$ = toxic equivalent relative to benzo(a)pyrene. The values obtained for the 16 PAHs are presented in Table 2. The total TEQ values ($\sum C_{16}$TEQs) for the sites ranged from 9.82 – 829.57 µg/Kg dry wt., with a mean of 191.50 µg/Kg dw. While the range for the seven carcinogenic PAHs ($\sum C_7$TEQs) were from 8.92 – 827.16 µg/Kg. The United State Environmental Protection Agency (US EPA) recommended 137 µg/Kg $\sum C_7$TEQs value as clean-up level for a mixture carcinogenic PAH. The values for all locations except LT-B were less than the recommended toxic level. Thus, most locations in the study area are not likely to pose threat to the environment. The major contributors to the sediment toxicity was: DhA (65.32%), BaP (23.55%), BaA (5.80%), BbF (2.74%), and IcP (1.36%). The order of total PAHs concentrations in the locations viz: B > D > A > E > C, was different from that of total TEQs which was: B > E > A > D > C. This demonstrates the impacts of individual PAH on the overall toxicity of total PAHs concentrations at the sites.

The PAHs bind to the organic carbon, making them less available to aquatic life, thereby lessening their toxicity. Thus, it is necessary to factor in the amount of organic carbon in the sediment while assessing the toxicity of PAHs in the sediments. It is usual to normalized PAHs toxicity at 1% organic carbon sediment concentration in order to account for the differences in bioavailability due to it. The organic carbon concentrations for the different locations viz: A (1.76%), B (3.21%), C (1.02%), D (1.11%), and E (1.46%) were to use to normalized TEQs values at 1% organic carbon. The values obtained for the normalized $\sum$TEQs were: LT-A ($1.6997 \times 10^3$ µg/Kg dw/OC), LT-B ($2.5843 \times 10^4$ µg/Kg dw/OC), LT-C ($9.6316 \times 10^2$ µg/Kg dw/OC), LT- D ($2.1574 \times 10^3$ µg/Kg dw/OC), and LT-E ($4.4014 \times 10^3$ µg/Kg dw/OC). The order for the normalized $\sum$TEQs which was: B > E > D > A > C, was also different from the un-normalized $\sum$TEQ. This emphasised the impacts of organic carbon contents on bio-availabilities and thus toxicities of the PAHs. The toxicities of PAHs are highly cushioned by the presence of organic carbon in the sediments.

## PAHs source identification

Table 3 gives some source diagnostic indices of PAHs for sediments in the studied locations. The ratio of the sum of low molecular weight to high molecular weight PAHs ($\sum$LMW/$\sum$HMW) were lower than one (1) in locations A and B, but locations C, D, and E had value of this ratio greater than unity. This indicates that the source of PAHs in sites A and B is majorly due to petroleum contaminations whereas, locations C, D, and E showed much evidence of pyrogenic contribution. Petrogenic contamination is characterized by the prevalence of low-molecular-weight PAHs (PAHs with two and three aromatic rings), whereas the dominance of high molecular weight PAHs (four, five and six rings) are indication of pyrolytic origin (Soclo et al., 2002; Yunker et al., 2002; Zhou et al., 2003; Tolosa et al., 2004; Nasher et al., 2013; Tobiszewski, 2019).

The Ant/(Ant +Phe) ratio has been used to differentiate petrogenic source from pyrolytic one. When value of this ratio is greater than 0.1, pyrogenic source is implicated. Ratio less than 0.1 however, connote petrogenic inputs (Yunker et al., 2002; Brandi et al., 2007; Bastami et al., 2014). Except for location A where the ratio could not be computed because, neither anthracene (Ant) nor phenanthrene (Phe) was detected in the location, the ratios were greater than 0.1 in all the other locations. This is an indication of pyrogenic source (Table 3).

Source diagnosis could as well be performed, using the ratio of Flt/(Flt +Pyr). Ratio less than 0.4 is related to petrogenic origin, ratio greater than 0.5 implicate PAHs from combustion of wood, grass, and coal, whereas ratio in the range of 0.4 – 0.5, indicate a mixture of combusted and non-combusted petrol fuels in conjunction with biomass combustion and degradation (Yunker et al., 2002; Bastami et al., 2014). The ratio obtained in this study, fall within the range of 0.10 – 0.77, with a mean of 0.34. Locations C and D had values greater than 0.5 indicating pyrolytic source. The values obtained for locations B (0.10) and E (0.24) were however less than 0.5 suggesting petrogenic inputs. Again, location A had values of fluoranthene (Flt) and pyrene (Pyr) below detection limits. Thus, this ratio could not be determined.

The BaA/(BaA + Chr) ratio is useful for the identification of PAHs source. Ratio greater than 0.35 is implicative of pyrogenic source (Dvorska et al., 2011), ratio within 0.2 – 0.35 suggests combustion processes (Yunker et al., 2002; Tobiszewski, 2019), while ratio less than 0.2 typify petrogenic inputs (Yunker et al., 2002; Adeniji et al., 2019). The values obtained in this study viz: LT-A (0.93) LT-B (0.96), LT-C (1.00), LT-D (0.83), and LT-E (0.86) tend to suggest that the anthropogenic sources are majorly from pyrogenic inputs.

The values for the IcP/(IcP + BgP) ratios were not determinable in locations B and D, but the values in the other locations were each 0.5 respectively. This again gives credence to the pyrogenic source of PAHs in the sediments. IcP/(IcP + BgP) ratio above 0.5 indicates PAHs input of grass, wood and coal combustion, values in the range of 0.2 – 0.5 connotes petroleum combustion while values below 0.2 represents PAHs input from petroleum (Yunker et al., 2002; Chen et al., 2012; Sun et al., 2016; Ihunwo et al., 2019). Cross plot involving coupling of two ratios can be more useful in estimation of PAHs source. This is because, overlap of characteristic PAH profiles from each source and selective decay of more labile compounds effects are more pronounce on single ratios than double ratios (cross plot). A cross plots (Fig. 3) for the ratios of Flt/(Flt + Pyr) versus BaA/(BaA + Chr) shows that sources of PAHs in the sediments were majorly from combustion (pyrogenic) processes and unburned petroleum. One of the likely pyrogenic sources is the burning of hides and skins. This is evidence from the sooty fumes that daily rise from rubbles of burnt vehicular tyres used as fuel to roast hides and skins by butchers at the cattle market and abattoir at the vicinity of the river bank. The burning of grass, wood, and coal constitutes other pyrogenic sources.

## CONCLUSIONS

Total PAHs distribution in sediments of Aba River at Ogbor Hill region ranged from 481.95 – 2,562.55 µg/Kg with a mean of 1122.64±839.70µg/Kg. Naphthalene (Nap), fluorine (Flr), dibenzo(a,h)anthracene (DhA), benzo(a)anthracene (BaA), pyrene (Pyr), and acenaphtylene (Acy) constituted the dominant PAHs corresponding to 13.26, 11.14, 11.14, 9.90, 9.15, and 8.42% of the total PAHs, respectively. The total PAHs range and mean total however, were below the 4022 µg/Kg Effects Range Low (ERL) target value for sediment quality guidelines (SQG)suggesting that biological health effects would rarely occur. The total toxicity equivalence quotient (∑16TEQs) for the locations ranged from 9.82–829.57 µg/Kg dry wt., with a mean of 191.50 µg/Kg dw. The major contributors to sediments toxicity were: DhA (65.32%), BaP(23.58%), BaA (5.80%), BbF (2.74%), and IcP (1.36%). The order of total PAHs concentrations in sediments was: B > D > A > E > C., while that of total TEQs was: B > E > A > D > C., but after normalization of ∑16TEQs at 1% organic carbon (OC), the order changed to: B > E > D > A > C. PAHs diagnostic ratios reveal that the sources of PAHs were majorly from combustion (of grass, wood, coal, hides and skins) and unburned petroleum. Although, the concentrations of PAHs in the sediments did not constitutes much treats to the environment, the necessity to preserve aquatic lives and public health calls for concerted efforts to prevent further depositions and accumulations of PAHs in the sediments.

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