

# Concentration of Polycyclic Aromatic Hydrocarbons in Water and Sediment from Woji-Elelenwo Creek, Rivers State, Nigeria.

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## ABSTRACT

Concentration Of Polycyclic Aromatic Hydrocarbons in Water and Sediment from Woji-Elelenwo Creek in Rivers State Nigeria were determined, water and sediment samples were collected and analyzed using the standard methods for sample collection and analysis, PAHs in the surface water and sediment were analyzed using High-pressure Liquid Chromatography -Fluorescence Detection (HPLC – FLD). Results obtained were subjected to statistical analysis using descriptive analysis to determine their mean, range and standard deviation. The PAHs concentrations were grouped into individual PAHs, total low molecular mass PAHs and total high molecular mass PAHs. The low molecular mass PAHs in water were Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, and Anthracene which were from below detectable limit (BDL) to 0.20 mg/l, while the high molecular mass PAHs were Fluoranthene, Pyrene, Benzo (b) fluoranthene, Benzo (k)fluoranthene, Benzo (a)pyrene, Indeno (123-cd) pyrene (from BDL - 0.21mg/l). The total values recorded for the LMM PAHs were 1.52E-02 mg/l and HMM PAHs in surface water of this study were 3.27E.2 mg/l. Sixteen (16) PAHs were found in sediment, the PAH concentration in sediment were higher than those found in water with (low mol. mass 0.077 and high mol mass 0.39 mg/kg), with sediment acting as a sink and depository. Water ways should be monitored from time to time to discourage locals from illegal bunkering that introduces fossil fuel into the ecosystem.

**Keywords: PAH, Sediment, Surface Water**

## 1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic chemicals characterized by chemical stability, low volatility, and low solubility in water. PAHs are formed during the incomplete combustion of organic substances, are widespread in the environment, and typically occur in mixtures. PAH are formed from natural and anthropogenic processes such as energy

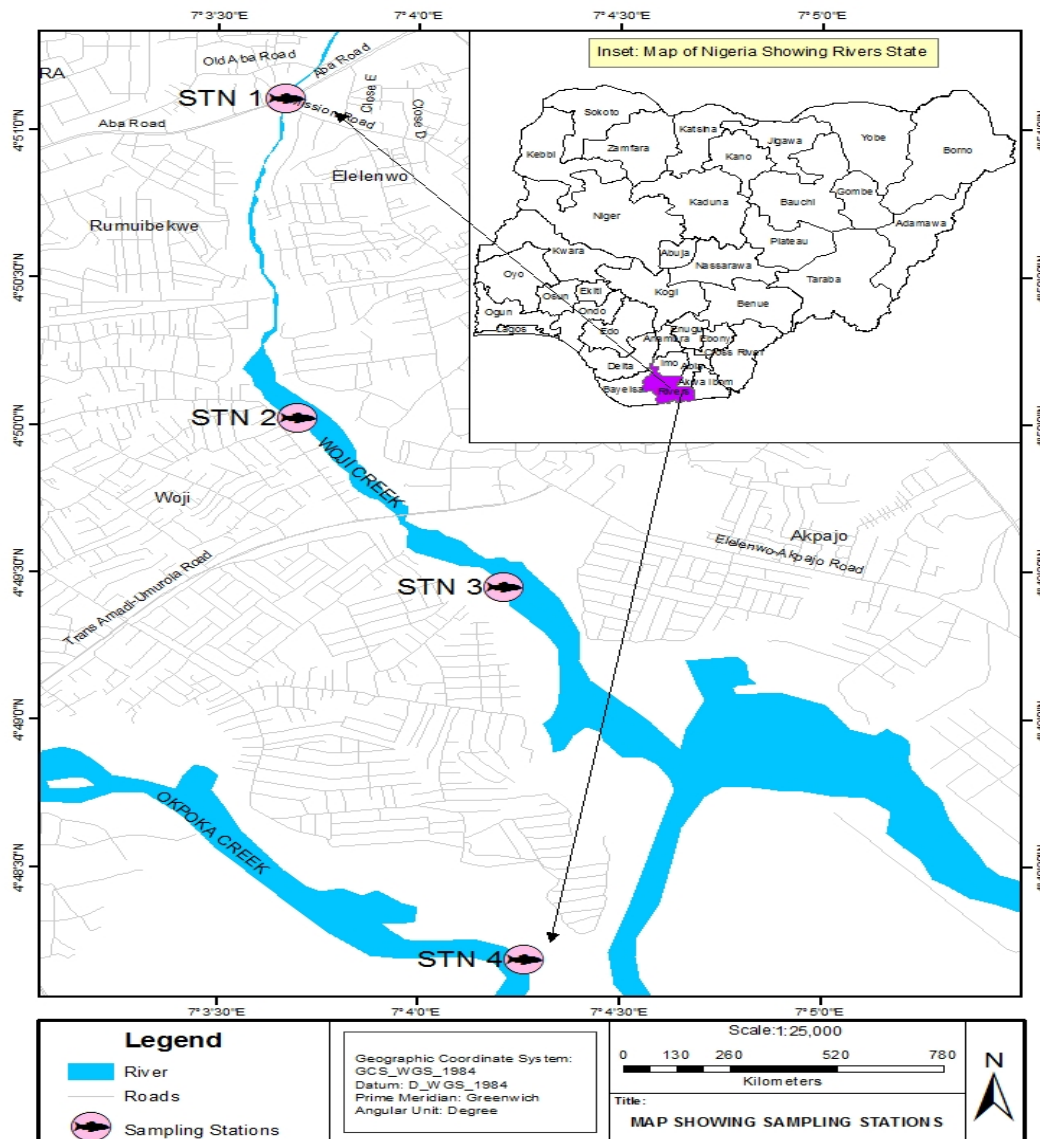
production, waste incineration, transport and industrial processes such as aluminum and coke production. (Olumuyiwa, 2012).

There are two categories of anthropogenic sources of PAHs which are pyrogenic and petrogenic. Pyrogenic PAH are those generated through the combustion of fossil fuel example coal, petroleum and biomass. These sources of combustion include power plants, automobiles, steel smelting and incinerators. Atmospheric polycyclic aromatic hydrocarbons that escape photo-degradation in the air are added to land surfaces through dry and wet deposition which becomes street dust due to hydrophobicity, they are also stored in soil. These street dust and surface soil PAHs are re-suspended back into the atmosphere by wind and volatilization (Manoli *et al.*, 2002, 2004). PAHs are found in the drainage network by storm runoff and are finally transported to aquatic environments during the rainy season, where they are absorbed into sediment (Mai *et al.*, 2003; Boonyayumanond *et al.*, 2006). Thus, soil and dust are natural sinks and environmental reservoirs for PAHs in the urban environment.

## 2. Methodology

### Study Area

The study area (sampling stations) were mapped out with GPS and the coordinates are recorded as follows; Station 1 (**Elelenwo Abattoir Waterfront**) latitude 4°51'2.94" and longitude 7° 3'35.84", station 2 (**Oil Servicing Company Waterfront**) latitude 4°50'2.16" and longitude 7° 3'40.55", station 3 (**ABEC International Secondary School Waterfront**) latitude 4°49'28.17" and longitude 7° 4'9.43", station 4 (**Elelenwo by Akpajo Waterfront**) latitude 4°48'12.53" and longitude 7° 4'10.84".



**Figure 1; Map of study area**

The study area is characterized by diverse industrial activities such as oil services company, abattoir, a “computer village” where electronics are sold and repaired, there is a domestic waste dump site at the bank of the Creek.

## **Water Sample Collection**

The surface water samples were collected at depth of 15-25cm, with pre-rinsed containers. One-liter plastic containers were used for the collection of samples for physico-chemical analysis. Samples for polycyclic aromatic hydrocarbons were collected with wide mouthed 500ml glass bottles fitted tightly with aluminum foil to prevent contamination of the samples. The samples were collected by lowering the container by hand until it was completely immersed, allowed to fill and corked under water. Each sample was treated according to the analysis to be carried out on them. Glass bottles were used in sampling for the analysis because hydrocarbons (oil) react with plastic containers.

## **Sediment Sample Collection**

Samples of sediment were collected from the four sampling stations. Sediment was collected using 0.1m square quadrant with the aid of a shovel taken to 0.1m depth on a mudflat (Ekweozor, 1985). The samples were sealed in a zip lock bag, labelled appropriately, and transported to the laboratory where they were air dried under room temperature for 4 days. 5g of air-dried sediment was ground with mortar and pestle, sieved with 2mm meshed sieve, sediment sample was preserved prior to analysis.

## **Extraction of PAH from Water Sample (Water – Liquid Matrix Sample)**

The extraction method used for surface water sample is the liquid-liquid extraction method, were 250ml of water sample was transferred into a separation funnel (500ml UI type), the container was rinsed with dichloromethane, 25ml of dichloromethane was added to 250ml part of the water sample and shaken vigorously to have the organic solvent and all the available organic material were extracted. The organic extract was collected into a receiving container by passing the extract through a column containing cottonwool, silica gel and anhydrous sodium sulphate and was later subjected to GC-MSD condition for identification of PAH.

## **Extraction of PAH from Sediment**

Sediment samples were collected with a stainless-steel columnar sampler. Samples of the sediment was mixed and stored in the washed glass bottles and transported back to the laboratory on crushed ice in a cooler and stored at 4°C before processing. The sediments were prepared for extraction of PAH based on the ultrasonic method; samples were air dried and screened through

a sieve with 1mm pore size. Sub samples of 2.0g dry mass were placed in 50ml borosilicate beakers with 10ml of acetone, 0.2g of copper powder, 5g anhydrous sodium sulphate, and extracted for 15mins in a 30°C ultrasonic water bath. Procedural blanks and reference controls spiked with 5µL PAH standard mixture were included for each batch of sample. The mixture was centrifuged at 400ppm for 5mins and the supernatant liquid was decanted. Another 10ml of acetone was added to the residue and both the extraction and decanting process were repeated. Extracts were combined and evaporated to dryness in a rotary evaporation.

### **Clean-up of PAHs Samples by Packed Solid Phase Extraction**

The sediments extracts were subjected to packed solid phase clean-up cartridge which was prepared in-house (plugged a glass wool on 10ml length column); then 1g of 20% deactivated silica gel and 0.4 anhydrous sodium sulphate ( $\text{Na}_2\text{SO}_4$ ) were weighed and conditioned with 5ml of dichloromethane. The sediment extracts were loaded to the cartridge using 10ml of elute (dichloromethane), fractions collected in a 50ml flask was evaporated on a rotatory evaporator at 40°C near to dryness and dissolved in 2ml of toluene and then GC-MS Model HP 6890 was applied for analysis of each sample.

## **3. RESULTS**

### **Polycyclic Aromatic Hydrocarbons (PAHs) in Surface Water**

PAH in this study were divided into high and low molecular mass compounds. The low molecular mass PAHs and high molecular mass PAH.

Low molecular mass PAH comprising two to three aromatic rings detected in the surface water of this study were Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, and Anthracene. Naphthalene had a mean value of 0.001mg/l during the wet season across the stations and highest concentrated mean value of  $0.19 \pm 0.005$  in station 1 during the study duration. Acenaphthylene recorded a mean value of 0.01mg/l during the wet season and highest mean value of  $0.03 \pm 0.05$  in station 1. Acenaphthene and Fluorene recorded a mean value of 0.0444mg/l. Phenanthrene recorded concentrated mean value of  $0.01 \pm 0.001$ . Fluorene had the highest concentrated mean value of  $0.05 \pm 0.08$  in station 1 for low molecular mass PAH

throughout the duration of the study as shown in (Table 1). Naphthalene, 2-methyl Naphthalene and Phenanthrene were record BDL in station 2 as shown in (Table 1). Fluorene and Acenaphthene had the highest percentage of 25% of low molecular mass PAH detected in the surface water during the study period while Naphthalene had 7% as the lowest LMM PAH detected. (Figure 2).

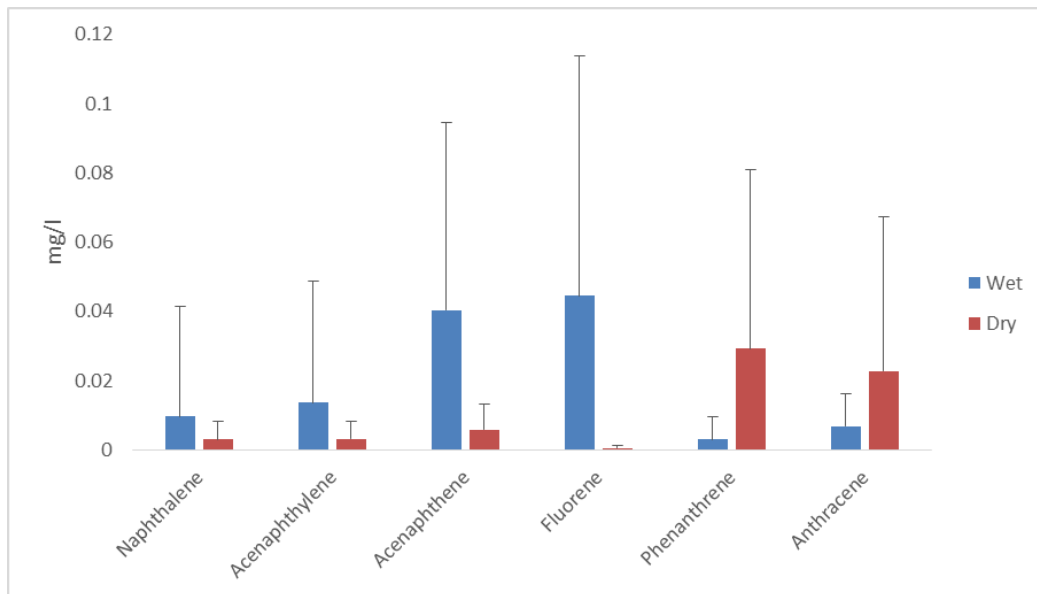
High Molecular mass PAH comprising four to six benzene rings detected in the surface water of this study (Figure 3) showed that Chrysene had the highest mean concentration of 0.09mg/l across the stations during the dry season. The nine HMM PAHs Fluoranthene, Pyrene, Benz (a) anthracene, Chyrene, Benzo (b) fluoranthene, Benzo (a) Pyrene, Indeno (1,2,3 Ocd)Pyrene and Dibenz (a,b) anthracene were all detected with highest mean concentration of  $0.09 \pm 0.08$  recorded in station 1 Fluoranthene and Pyrene showed the lowest mean concentration of  $0.001 \pm 0.001$  in station 2 and 4 respectively (Table 1). Chrysene recorded 18% concentration of high molecular mass PAH (Figure 4).

In comparison, High Molecular Mass PAH were all detected, and recorded the highest mean concentration value of  $0.091 \pm 0.139$  by Chrysene in the dry season during the study period (Figure 5). The Low Molecular Mass PAH had the lowest mean concentration of  $0.001 \pm 0.001$  by Acenaphthylene, Fluorene and Phenanthrene in stations 3 and stations 4. Naphthalene, 2-Methyl Naphthalene and Phenanthrene were not detected in stations 2 and 3 (Table 1).

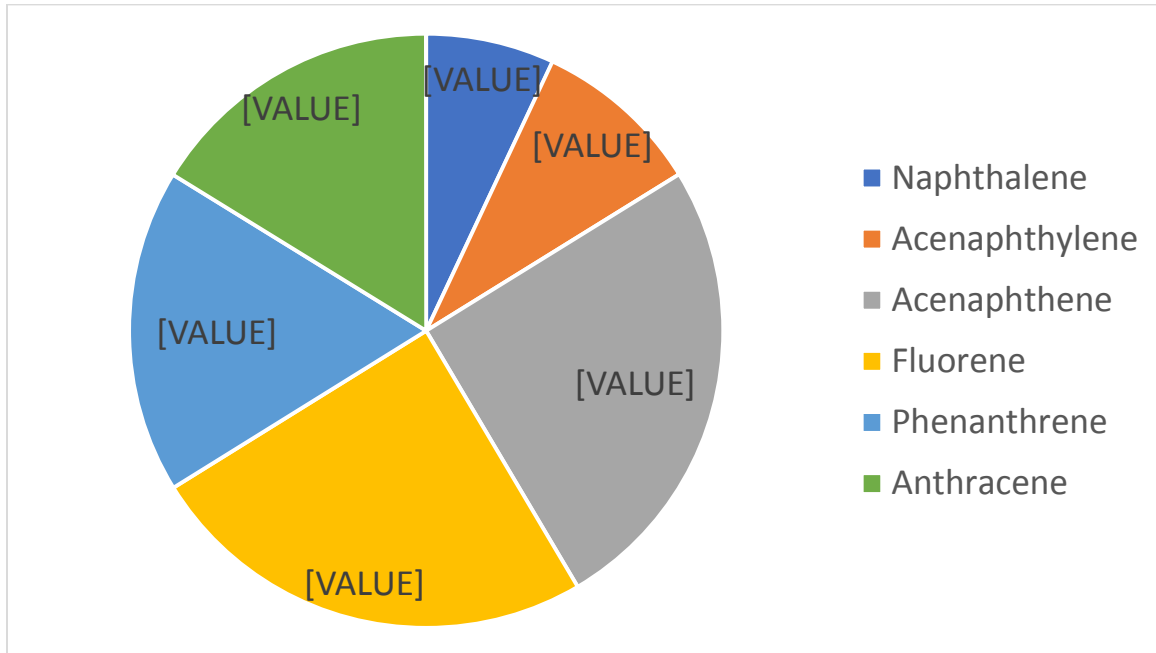
**Table 1 Mean and Standard Deviation of PAHs in Water by Stations**

PAHs	Station1	Station2	Station 3	Station4
Naphthalene	$0.19 \pm 0.01$	Not detected	$0.000 \pm 0.01$	$0.002 \pm 0.005$
2-methyl Naphthalene	$0.02 \pm 0.04$	Not detected	Not detected	$0.01 \pm 0.01$
Acenaphthylene	$0.026 \pm 0.05$	BDL	BDL	$0.01 \pm 0.01$
Acenaphthene	$0.04 \pm 0.05$	$0.04 \pm 0.60$	$0.01 \pm 0.01$	$0.01 \pm 0.01$

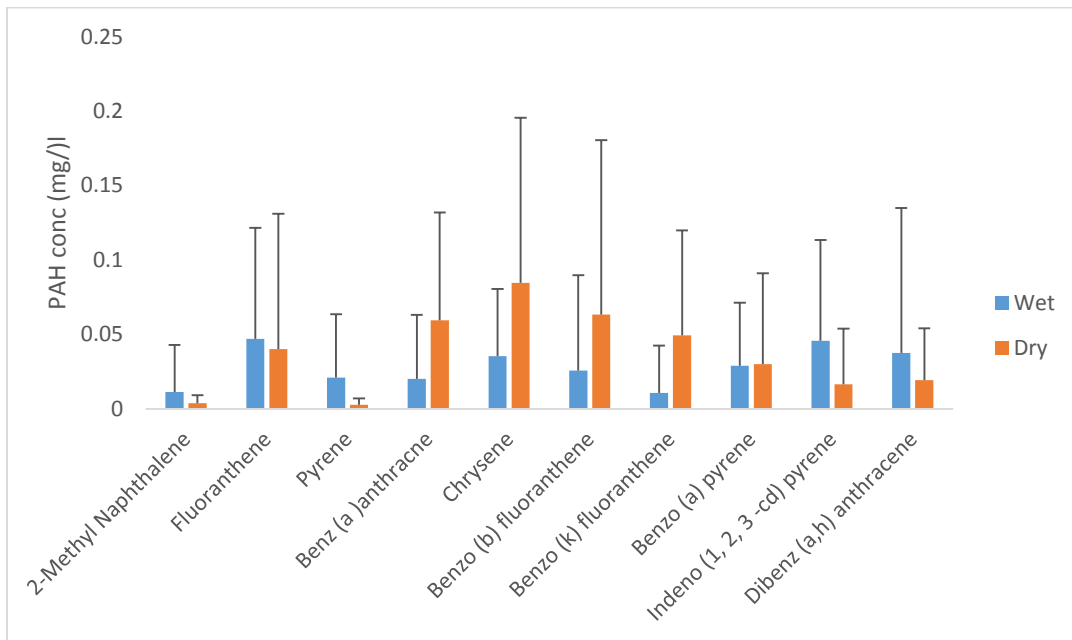
Fluorene	0.05±0.08	0.04±0.06	BDL	BDL
Phenanthrene	0.01±0.01 <sup>a</sup>	- <sup>b</sup>	BDL <sup>b</sup>	0.06±0.06 <sup>b</sup>
Anthracene	0.01±0.01	0.00±0.01	0.04±0.06	0.01±0.01
Fluoranthene	0.09±0.08	BDL	0.06±0.12	0.02±0.04
Pyrene	0.02±0.05	0.02±0.04	0.01±0.01	BDL
Benz(a) anthracene	0.04±0.06	BDL	0.04±0.06	0.08±0.09
Chyrene	0.05±0.05	0.02±0.04	0.09±0.12	0.08±0.09
Benzo(b) fluoranthene	0.01±0.01	0.04±0.09	0.06±0.06	0.07±0.16
Benzo(k) fluoranthene	BDL	0.02±0.05	0.03±0.05	0.07±0.09
Benzo(a) Pyrene	0.02±0.04	0.04±0.05	0.04±0.08	0.02±0.03
Indeno(1,2,3Ocd) Pyrene	0.03±0.05	0.05±0.08	0.02±0.05	0.01±0.03
Dibenz(a,b) anthracene	0.02±0.05	0.06±0.13	0.02±0.05	0.01±0.02



**Figure 1. Seasonal Variation (Mean ±SE) of low molecular mass PAHs in surface water.**

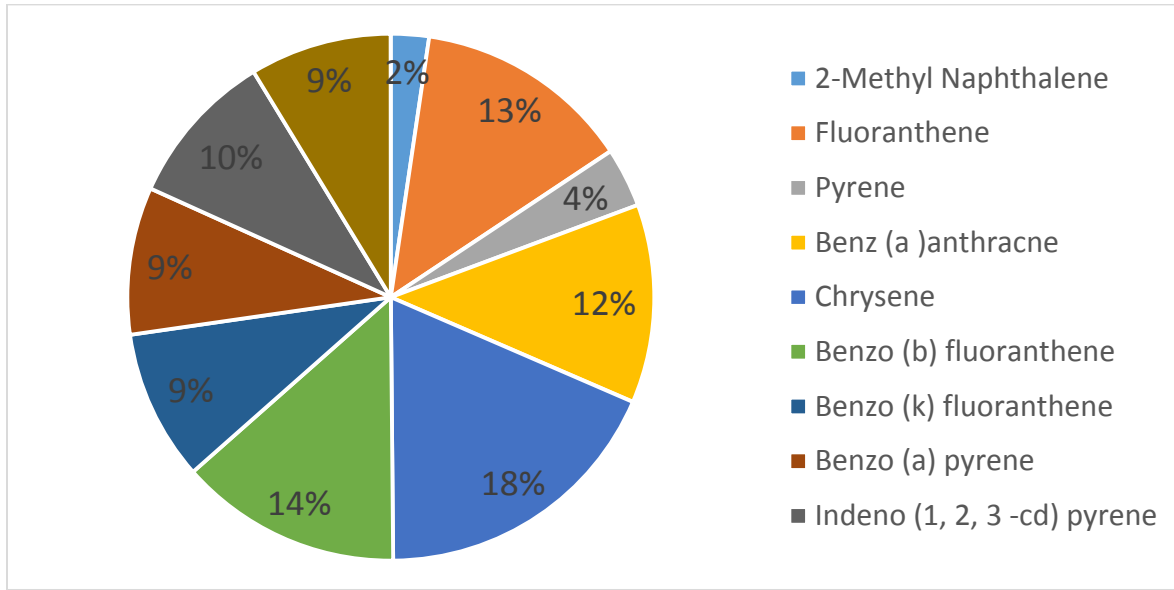


**Figure 2. Percentage Mean Concentration of low molecular mass PAHs in surface water of the study area.**

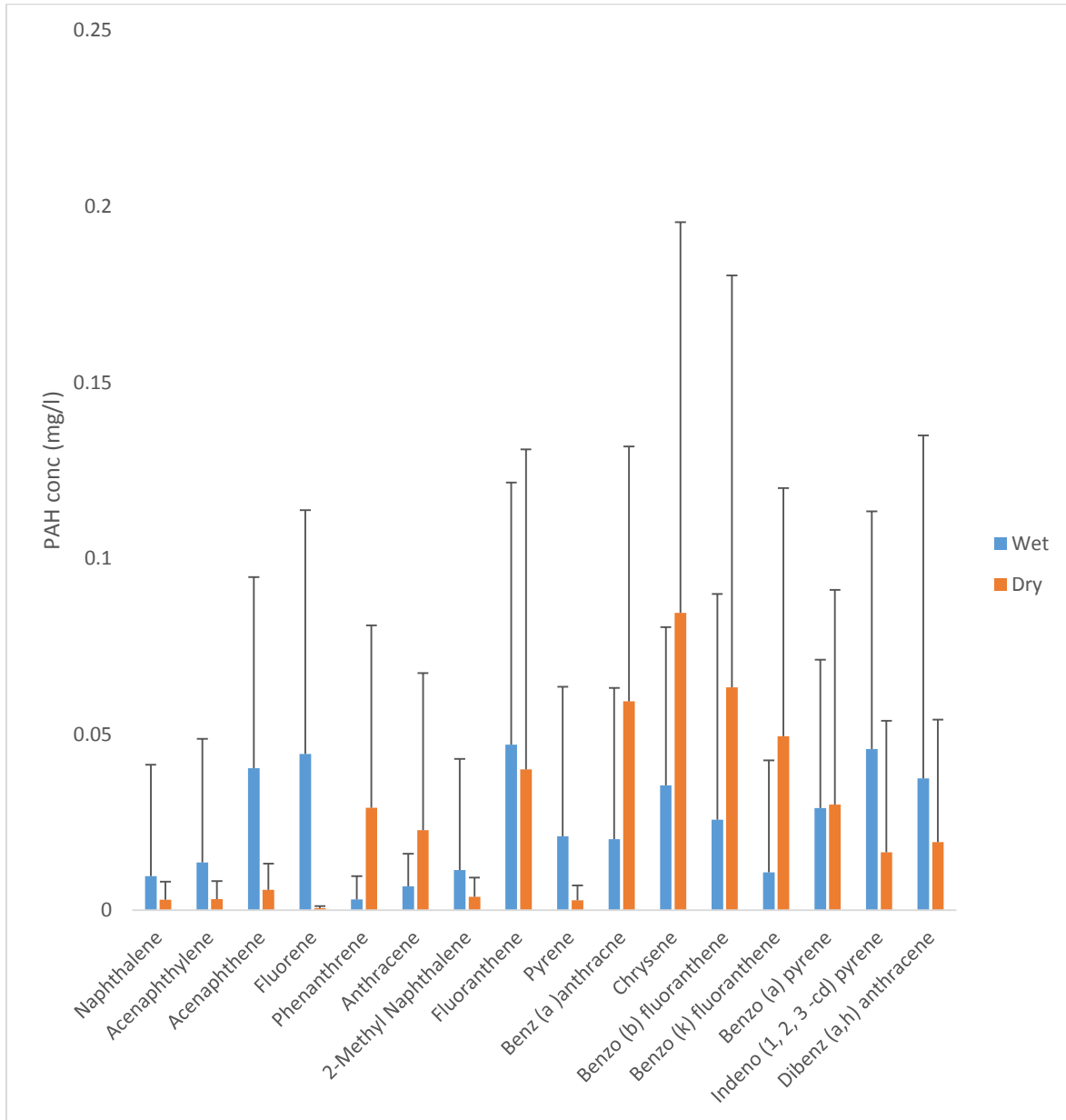


**Figure 3. Seasonal Variation (Mean ±SE) of High Molecular Mass PAHs in surface water area.**





**Figure 4. Percentage Mean Concentration of High Molecular Mass PAHs in surface water of the study area.**



**Figure 5. Seasonal Variation (Mean  $\pm$ SE) of high molecular and low molecular mass PAHs in surface water of the study area.**

## Polycyclic Aromatic Hydrocarbon (PAHs) in Sediment

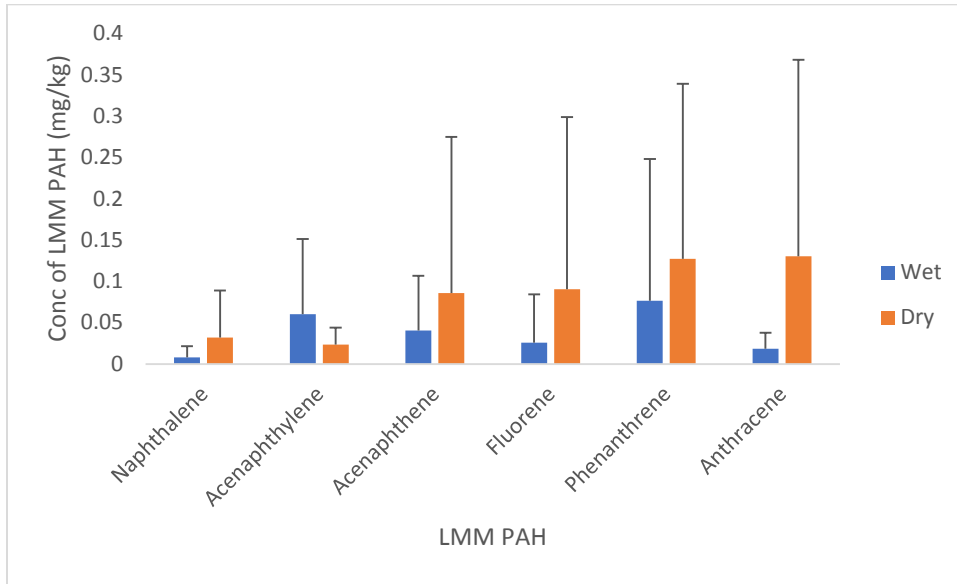
Low molecular mass PAH detected in the sediment of this study were Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, and Anthracene. Anthracene highest mean concentration value was 0.13mg/kg during the dry season (Figure 6). Naphthalene had the lowest mean value of 0.0007mg/kg during the wet season (Figure 6). Acenaphthylene mean concentration value was 0.040mg/kg during the wet season and 0.0859mg/kg during the dry season (Figure 6). LMM PAHs were higher in the dry season than the wet season (Figure 6). Phenanthrene had the highest percentage of 28% of low molecular mass PAH detected in the sediment during the study period. Naphthalene had 6% as the lowest LMM PAH detected during the period of the study. (Figure 7).

High molecular mass PAHs recorded high values during the wet season than dry season (Figure.8). Fluoranthene recorded the highest value in wet season with the highest mean concentration of  $0.49\pm 0.35$  in station 3 (Table 2) and total percentage of 19% (Figure 9). Pyrene and 2-Methyl Naphthalene had total percentage proportion of 5% (Figure 9). Benz[a] anthracene highest mean concentration value was  $0.531\pm 0.328$  in station 1 (Table 2) and percentage proportion of 17% (Figure 9). Chrysene highest mean concentration value was  $0.47\pm 0.18$  in station 4 (Table 2), Benzo(b) fluoranthene and Chrysene were both 16% (Figure 9). Benzo (k) fluoranthene recorded the highest mean concentration value of  $0.281\pm 0.227$  in station 3 and percentage proportion of 7% (Table 2 & Figure 9).

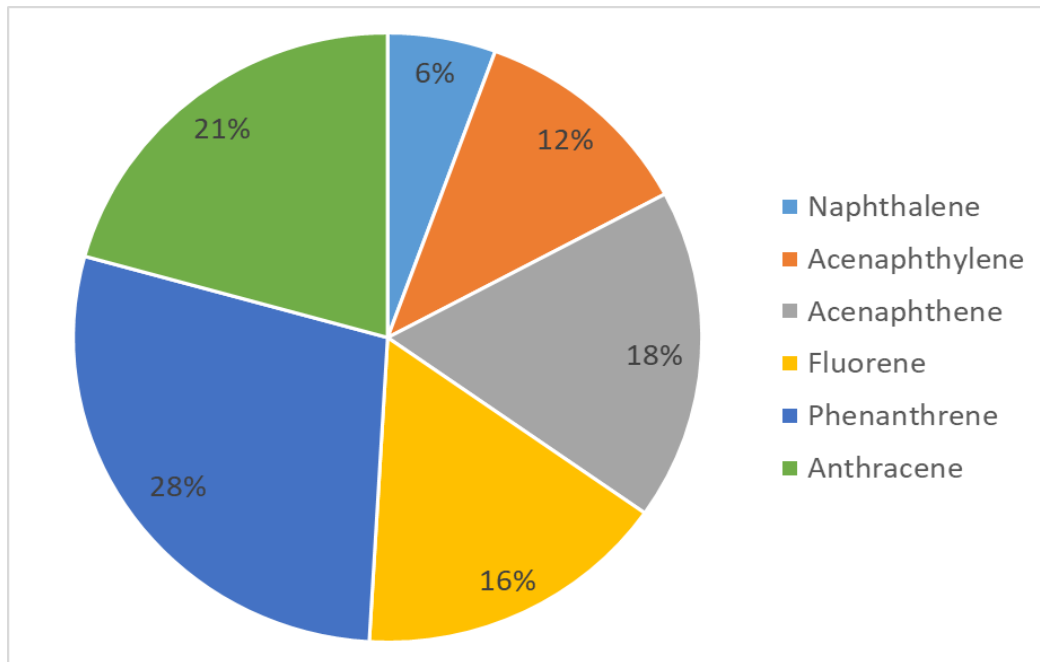
In comparison, High Molecular Mass PAH and Low Molecular Mass PAH were all detected, as shown in Figure 10.

**Table 2 Mean and Standard Deviation of PAHs in Sediment by Stations**

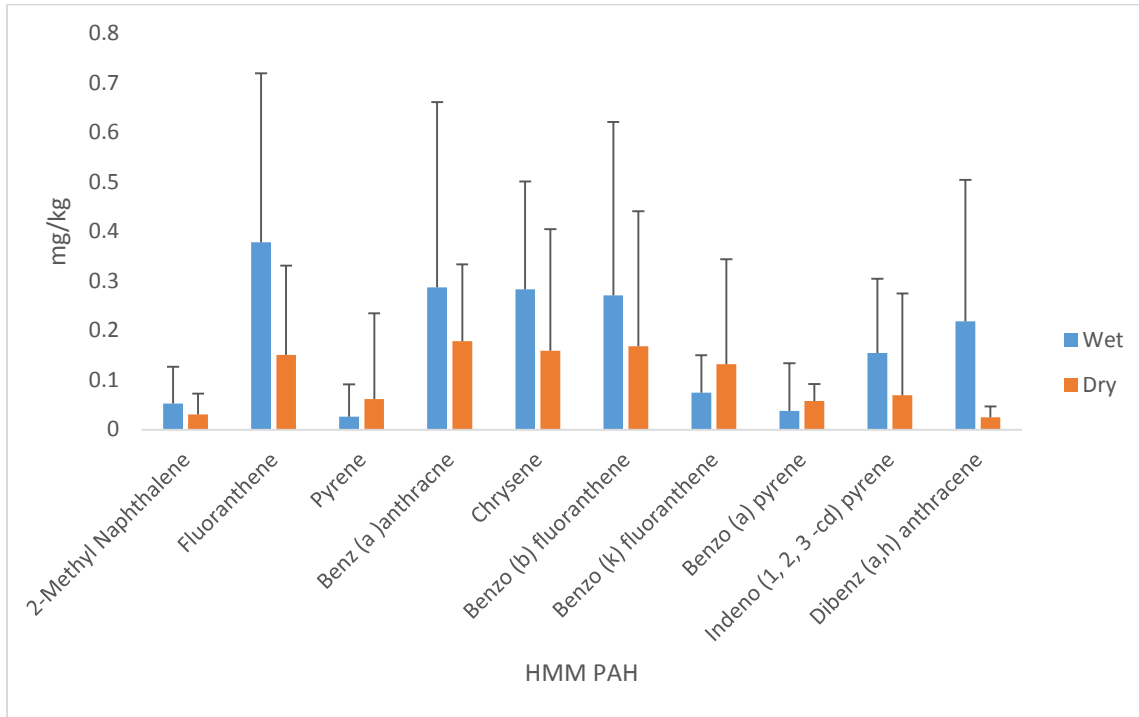
PAHs	Station1	Station2	Station 3	Station 4
Naphthalene	0.01±0.01	0.01±0.01	0.06±0.08	0.01±0.01
2-methyl Naphthalene	0.06±0.09	0.00±0.00	0.04±0.07	0.06±0.04
Acenaphthylene	0.08±0.12	0.28±0.03	0.03±0.03	0.02±0.02
Acenaphthene	0.05±0.08	0.13±0.27	0.05±0.05	0.03±0.02
Fluorene	0.04±0.08	0.18±0.29	0.09±0.28	0.00±0.00
Phenanthrene	0.04±0.04	0.01±0.03	0.01±0.02	0.34±0.27
Anthracene	0.03±0.01	0.01±0.06	0.23±0.32	0.02±0.01
Fluoranthene	0.43±0.19	0.02±0.02	0.49±0.35	0.12±0.20
Pyrene	0.05±0.09	0.12±0.24	0.01±0.00	0.01±0.00
Benz(a) anthracene	0.53±0.33	0.01±0.02	0.15±0.24	0.24±0.19
Chrysene	0.09±0.11	0.18±0.25	0.14±0.21	0.47±0.18
Benzo(b) fluoranthene	0.03±0.02	0.28±0.40	0.41±0.27	0.14±0.32
Benzo(k) fluoranthene	0.01±0.00	0.06±0.08	0.07±0.06	0.28±0.23
Benzo(a) Pyrene	0.06±0.05	0.08±0.13	0.03±0.02	0.01±0.01
Indeno(1,2,3Ocd) Pyrene	0.05±0.12	0.24±0.28	0.08±0.10	0.07±0.14
Dibenzo (a,b) anthracene	0.08±0.18	0.22±0.38	0.10±0.09	0.09±0.16



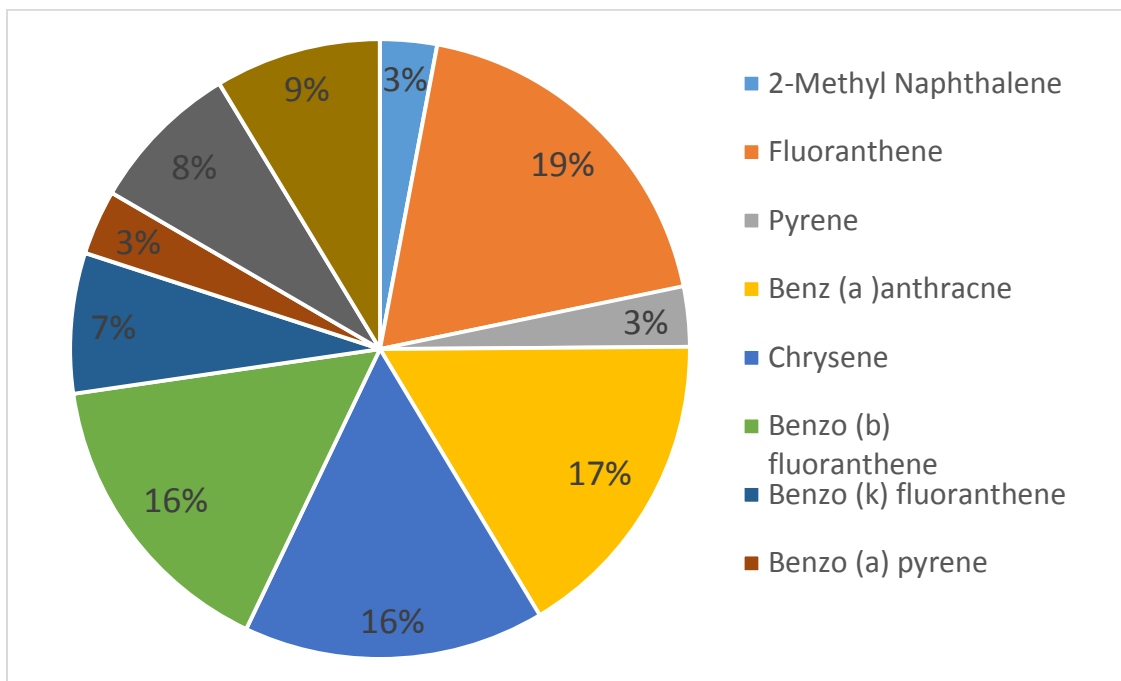
**Figure 6: Seasonal Variation (Mean  $\pm$ SE) of Low Molecular Mass PAHs in Sediment of the study area.**



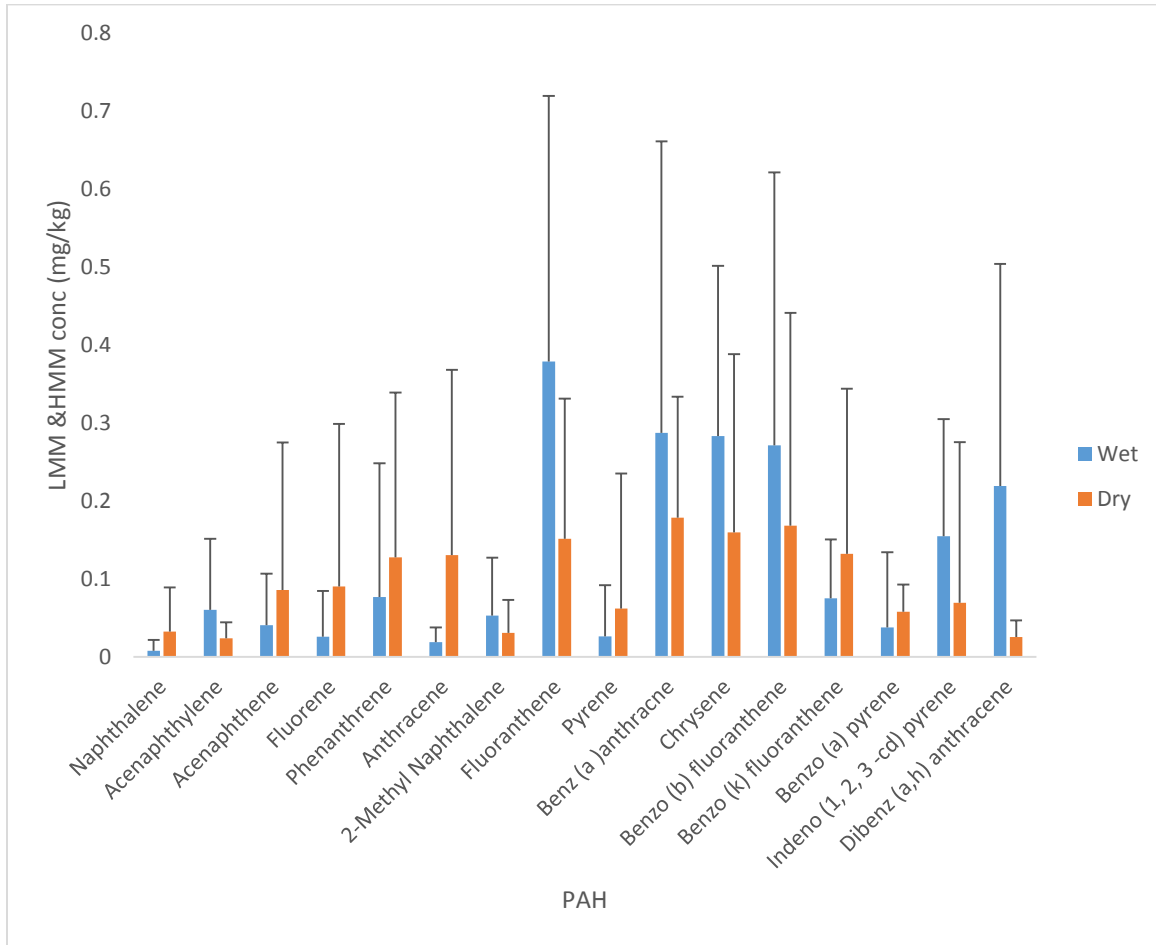
**Figure 7: Percentage Mean Concentration of Low Molecular Mass PAHs in Sediment of the study area.**



**Figure 8: Seasonal variation of HMM PAH in Sediment of the study area.**



**Figure 9: Percentage mean concentration of high molecular mass PAHs in sediment of the study area.**



**Figure 10: Seasonal Variation (Mean ± SE) of LMM and HMM PAHs in sediment during the study period.**

**Table 3 Concentrations of PAHs in Sediment Compared with US National Oceanic Sediment Quality Guidelines**

PAHs	ERL	ERM	Station1	Station2	Station 3	Station 4
Naphthalene	160	2100	0.006	0.008	0.056	0.009
2-methyl Naphthalene	-	-	0.064	0.003	0.044	0.056
Acenaphthylene	44	640	0.084	0.277	0.033	0.022
Acenaphthene	10	500	0.048	0.129	0.049	0.026
Fluorene	19	540	0.035	0.177	0.018	0.003
Phenanthrene	240	1500	0.043	0.013	0.013	0.339
Anthracene	85	1100	0.034	0.011	0.229	0.024
Fluoranthene	600	5100	0.434	0.023	0.489	0.115
Pyrene	665	2600	0.045	0.116	0.009	0.005
Benz(a) anthracene	260	-	0.531	0.013	0.145	0.243
Chrysene	380	-	0.090	0.182	0.141	0.473
Benzo(b) fluoranthene	430	-	0.033	0.282	0.416	0.148
Benzo(k) fluoranthene	280	-	0.005	0.058	0.067	0.281
Benzo(a) Pyrene	430	2800	0.064	0.079	0.035	0.013
Indeno(1,2,3Ocd) Pyrene	-	-	0.054	0.247	0.078	0.070
Dibenz(a,b) anthracene	-	-	0.076	0.217	0.101	0.094

**ERL -Effect Range Low, ERM – Effect Range Medium**



## 4. DISCUSSION

### Polycyclic Aromatic Hydrocarbons (PAHs) in Surface Water

PAHs dispersion through air, moves through terrestrial and aquatic environments. They can settle as soot on surface water like rivers, streams and lakes and integrate with the sediment. Sixteen (16) of the PAHs investigated in this study were all detected in surface water. The high molecular mass PAHs detected were Fluoranthene, Pyrene, Benz[a]anthracene, Chyrene, Benzo(9b) Fluoranthene, Benzo(k)fluoranthene, Benzo(a)Pyrene, Indeno(123Ocd)Pyrene, Dibenz(a,b)anthracene. The concentration of HMM PAHs were higher than that of the LMM PAHs, the values recorded for the LMM PAHs were  $1.52E-02$  mg/l and HMM PAHs were  $3.27E-2$  mg/l, which is in consistent with other researchers reports such as Edori, (2020) who recorded higher HMM from Elelenwo River. The total concentration of PAHs of surface water of this study was  $4.79E-2$ , which was higher than  $1.38E-03$  reported for SOKU oil field by Olu (2019). Fluoranthene had highest concentration of  $0.093 \pm 0.084$  in all PAH congeners across the sampling stations. Naphthalene, 2-Methyl Naphthalene and Phenanthrene were not detected in station 2. While 2- Methyl Naphthalene was not detected in station 3 during the study period. Generally, the higher molecular mass PAHs were higher in concentration when compared to the lower molecular mass PAHs. However, individual group concentrations showed the six-membered rings as the most abundant and the four membered rings were the least in abundance. The observed concentration ranges of the different classes of PAHs observed in the present work was different from the observation of Kalf *et al*, (1997) where the order of abundance of the different categories was four membered ring and Olanyika *et al.*, (2019) who reported 4 and 2 membered PAH rings as the most abundant. This study revealed that the high molecular mass components were more abundant than the corresponding low molecular mass PAHs. This variation in the concentrations of the different PAHs could be ascribed to the molecular mass of the individual class categories and degradation from bacteria. An extensive range of microbes comprising algae, fungi and bacteria have been identified to breakdown PAHs Olu (2019). The LMM PAHs are easily degraded when compared to the HMM. The degradation of the LMM PAHs is achieved through microbial activities, oxidation, easy penetration into tissues and organs of plants and animals and are easily carried along through drifts (wind and moving water current Edori and Iyama 2019; Obayori and Salam, 2010).

The detection of these PAHs could be a result of fishing activities and boat transportation used by the community within the Creek. Other sources could be from burning of firewood, spill from hydrocarbons, atmospheric deposition and run off from domestic waste. HMM PAHs detected in this study could be a result of their lipophilic nature and high molecular mass which makes them sparingly soluble in water. In this study the total concentration of PAHs in surface water was lower than WHO recommended maximum permissible limit of 10 mg/l in surface and coastal water.

### **Polycyclic Aromatic Hydrocarbons (PAHs) in Sediment.**

The predominance of LMM and HMM PAHs in the sediments reflects the presence of combustion products from pyrolytic or petrogenic sources Chen *et al.*, 2011. The presence of LMM and HMM PAHs in the sediments indicates the presence of significant combustion products from pyrolytic processes and/ or petrogenic sources (Chen *et al.*, 2011).

The sixteen (16) PAHs dictated in the sediment of the study area were also detected in the surface water. The low molecular mass congeners recorded a total value of 0.039mg/kg, while that of the high congeners recorded 0.136mg/kg.

Polycyclic aromatic hydrocarbon concentrations in sediment can be classified according to the following categories: >1.0 mg/kg, 0.001 – 1.0 mg/kg and < 0.001 mg/kg for high risk, medium risk, and low risk, respectively Ana *et al.*, 2009. In this study, the concentration of PAHs from all sampling points were less than >1.0 mg/kg. This study shows a low pollution range and could be compared to the PAH results from Saleem *et al.*, 2017. It can also be compared to the result from Olu (2019) in Soku Oil Field. The results were also comparative with the results from Dosunmu, *et al.*, 2012 in Imo River Sediment of the Niger Delta Region, it is also in line with the results from Asagbra *et al.*, (2015) who reported lower values of PAH in sediment.

The PAHs concentration in the sediments were also compared with US National Oceanic sediment quality guidelines Long and MacDonald (1998) as shown in (Table 3). The recommended effect range low (ERL) and effect range median (ERM) target values were used to determine toxic effects in the sampling locations. When PAH concentrations vary between ERL and ERM values, a mild toxic effect is expected. In addition, no negative effect is expected for PAH concentrations lower than ERL values. Acenaphthene, and Benz [a] anthracene exceeded

the ERL values, but were within ERM values in all sampling locations, indicating a mild toxic effect. Anthracene exceeded the ERL and ERM values at sampling locations 3, Fluorene exceeded ERL limit in station 2 but were within ERM value at other sample points. The value of Anthracene and Fluorene at these stations suggests that negative biological effects such as reproductive and physiological disorder may occur in biota at these stations Nyarko *et al.*, (2011). A sediment quality guideline of 1000 ng/g dry weight total PAHs was designed to protect estuarine fish against several important health effects Johnson *et al.*, (2002). Based on this guideline, the results of this study showed that the concentrations of total PAHs did not exceed 1000 ng/g dry weight at all sampling points, indicating that aquatic organisms are not at very high risk, but the presence of these congeners could pose a major risk in the future.

The low values recorded from PAH in this study could be a result of the security patrol on the water ways by armed forces against crude oil theft / bunkering during the period of this study. The concentration of high molecular mass PAHs that are probable human carcinogens according to the US Environmental Protection Agency (USEPA, 1993), were detected in all the stations with a total percentage of 67% against LMM PAHs with a percentage of 23%. However, the sixteen congeners recorded in this study is indicative of an imminent pollution if not properly monitored.

## 5. CONCLUSION

Polycyclic aromatic hydrocarbon levels in surface water were within the recommended permissible limits by international standards. The HMM PAHs were readily and more available in surface water than the LMM PAHs at the study area as stated in this research.

PAH availability in sediment indicated combustion activities within the study area resulting from pyrolytic sources. The sixteen (16) PAHs that were selected were found in detectable concentrations in the study area. The total concentrations of HMM PAHs were higher than the LMM PAHs. The concentration of PAHs across the sampling stations were less than 1.00mg/kg.

Water ways should be monitored from time to time to discourage locals from illegal bunkering that introduces fossil fuel into the ecosystem

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