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Investigation and Assessment of Polycyclic Aromatic Hydrocarbons Concentrations in Sediments at Drainage Discharge Points into the Mangrove Stretch of New Calabar River, Port Harcourt, Nigeria

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Abstract

Sediment samples were collected from surface of sediments from parts of the mangrove wetlands of the New Calabar River at points of effluents discharge. The samples were treated according to standard procedures and analyzed for the concentrations of polycyclic aromatic hydrocarbons (PAHs). The results showed the presence of ten PAHs at Iwofe Jetty and Police Post stations and eleven PAHs at the Minipiti station. The summation of the total PAHs in the stations showed 23.440, 33.694 and 51.094 mg/Kg in sediments from Iwofe Jetty, Minipiti and Police Post stations respectively. Pyrene was the most abundant PAH in sediment from Iwofe station, Dibenzo (a,h) anthracene was the most abundant PAH at the Minipiti Station and acenaphthylene was the most abundant PAH at the Police Post Station. The categorization of the PAHs classes showed that the high molecular weight PAHs (HMW PAHS) were more abundant than the low molecular weight (PAHs) in all the stations examined, although, at the Police Post station, the values were very close. The 4-membered rings were more abundant in sediments from Iwofe Jetty station. The 4 and 5-membered rings were more abundant at the Minipiti station and the 2-3 membered rings were dominant at the police Post station. The 6-membered rings were the least abundant or undetected in all the stations. Source and origin diagnosis showed that LMW/ HMW PAHs as pyrogenic in all the stations, An/(An+Phe) ratio revealed pyrogenic sources of PAHs, Fl/(Fl+Pyr) analysis in all the stations indicated petrogenic origin of PAHs, BaA/(BaA+Chr) evaluation in all the stations revealed pyrogenic sources of PAHs. The mixed sources of PAHs in the wetlands drainage discharge points calls for concern and therefore, efforts should be geared to identify the points at which they were introduced into the effluents and be adequately controlled to prevent harm to the environment.

Keywords: New Calabar River, PAHs, Pyrogenic, Petrogenic, Minipiti and Iwofe Stations

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are referred to as collection of organic substances which contains two or more aromatic rings with a complete absence of a heteroatoms or substituents attached to the ring [1]. They are very common in the environment. They are natural or anthropogenic and formed as a result of partial combustion processes from homes, forest fire, industrial burning and heating system that involves hydrocarbons, discharge from automobiles, industrial used water, spillages of different petroleum products and burning of grasses from agricultural farms and lawns [2,3]. The negative effects of PAHs are noticed all over the world and more pronounced in water surroundings [4]. They are associated with different toxicity issues which culminates in some identified diseased conditions which manifests in the form of erotogenesis, teratogenesis, carcinogenicity and mutagenicity [5,6].

Polycyclic aromatic hydrocarbons (PAHs) are used to describe a group of universal poly-ringed organic contaminants that are prevalent in water and sediment of rivers within urban settlements [7]. PAHs are naturally lipophilic. The different components of PAHs effectively partitions in octanol-water solvent mixture and are transported efficiently in water media over a long range of distance. PAHs when present in water environments, more especially the sediment, gradually increases in concentration and subsequently accumulate in tissues and organs of aquatic plants and animals [8].

One of the factors that changes the natural environment is urbanization. Through growth of urban centres and cities, the original features of some rivers are changed so that some human activities such as navigation, waste discharge, and drainages built to discharge wastes from industries and homes can be effectively performed along the river shore or course [9]. Due to urbanization, rivers that are found within close range of the metropolitan city suffer from input of contaminants from human induced sources [10]. Quantities of polluted areas have been observed in rivers (Navarro-[11]. Most of these points where discovery of heavy pollution occur are harbours, boat jetties, wastes, sewage and discharge channels being the major points of organic contaminants [12].

However, many studies have been put in place to study the concentrations, sources, pathways, toxicity and health implications of PAHs and also on different approaches to reduce the input sources and toxicity, yet the problem has continued unabated [13]. Therefore, this study was undertaken to examine the concentrations of PAHs in sediments at drainage discharge points into the New Calabar River with the view to determine the sources of input.

Materials and Methods

Collection of Samples

Sediment samples were collected bi-monthly between the months of January to May. The stations where sampling was done were at the Iwofe Jetty, Minipiti station and the Police Post station. All the stations where samples were collected were checked for geographic location with an android handset. Plastic hand trowel was used to scoop off the surface of the sediment to a depth not more than 10 cm and put into glass containers placed in ice-cold pack and moved to the laboratory and refrigerated pending time for analysis.

Extraction of PAHs from Sediment and Analysis

100 g of freeze dried sediment samples were placed in cellulose thimble and soxhlet extractor, which contains hexane and toluene. After the first extraction, tetrachloro methane was used to elute only PAHs [14,15]. The concentrations of the PAHs was achieved by the use of gas chromatographic (GC) technique coupled with a mass spectrometer (MS).

Source Evaluation and Origin of PAHs

Different methods were applied to determine the sources of the PAHs in the sediment and the values obtained were used to adopt a position whether the origin was petrogenic or pyrogenic. The five different parametric approach used and their interpretations were;

i) Low Molecular Weight (LMW-PAHs)/ High Molecular Weight (HMW-PAHs)

All the values observed in the 2-3 rings PAHs were divided by the values obtained in all the other rings (4-6). If the values (ratio) is > 1, the sources are considered to be of petrogenic origins, while values < 1 are considered pyrogenic sources [16].

ii) Anthracene/Anthracene + Phenanthrene. An/(An+Phe)

If the calculated ratio between An/(An+Phe) falls below 0.1, the origin of the PAHs is petrogenic, while ratio higher than 0.1 suggests pyrogenic sources.

iii) Fluoranthene/fluoranthene +pyrene (Fl/Fl + Py).

Calculated values > 1 identifies pyrogenic origin and values < 1 are ascribed to petrogenic sources (Qiu, 2009).

iv) Benzo(a)Anthracene/(Benzo(a)Anthracene + Chrysene). BaA/ (BaA+Chr).

The value obtained from division of BaA/(BaA+Chr) is considered to be of petrogenic origin if it is > 0.2 and when observed within the range of 0.2-0.35, it is an indication of mixed origin, but when the ratio value exceeds 0.35, then the origin is classified as pyrogenic [17].

v) Indeno (1, 2, 3-cd) pyrene/(Indeno (1, 2, 3-cd) Pyrene + Benzo (ghi) Perylene). IP/ (IP+ BgP).

For this group, classification of source was determined on the basis that values < 0.2 suggested petrogenic, but values between 0.2 - 0.5 suggests mixed sources of origin, that is petrogenic or pyrogenic and values > 0.5 are adjudged to be pyrogenic [18].

Results and Discussion

Concentrations of PAHs

The concentrations of the PAHs obtained from the different discharge points are shown in Table 1. The first four members of the group, Benzo (g,h,i) perylene and Indeno (1,2,3-cd) pyrene were not detected in sediment at the Iwofe Jetty station. The most concentrated member of the group observed in sediment from this station was pyrene, with a value of 20.226±4.69 mg/Kg. At the Minipiti station, naphthalene, acenaphthene, anthracene, Benzo (k) fluoranthene and Indeno (1,2,3-cd) pyrene were not detected. However, the most concentrated member of the group observed in the station was Dibenzo (a,h) anthracene, with a concentration of 11.827±4.21 mg/Kg. At the Police Post Station, naphthalene, phenanthrene, pyrene and all the six membered rings were not detected. The most abundant member of the group was acenaphthene with a concentration of 12.283±3.42 mg/Kg. The total content of polycyclic aromatic hydrocarbons in the stations showed 23.440, 33.694 and 51.094 mg/Kg for Iwofe Jetty, Minipiti and Police Post respectively.

The summation of the PAHs in the individual stations in the present work is lower than the observation of many authors [19,20,21]. The observation of PAHs in the present research work is at variance with the observations of [22]. In lower reaches of River Chaohu, China and [19]. In sediment of Alkahlaa River in Missan Province, Iraq. Observations from the nature of concentrations from the stations which showed the order as Iwofe Jetty < Minipiti < Police Post may be due to some factors such exposure to direct sunlight and nearness to the main river. The Iwofe Jetty location is directly on the New Calabar River course, while the Minipiti location is a bit covered by vegetation of different aquatic plants and a little far away from the river, but the Police Post station is covered by vegetation and very far from the river. It is only during the high tide that water spreads to these point as to remove whatever may have been deposited in the course of drainage. The openness of the surface of water to direct sunlight can affect the distribution, volatility and degradation of PAHs in an environment [23]. These concept is true with the observation made in the present work, where the first three members are completely absent in Iwofe Jetty station (the most exposed), which was followed by those of Minipiti, where two of the lower molecular weight PAHs were absent and then the Police Post where only one is absent. Generally, the concentration of PAHs in sediment or water is attributable to the amount of dis-

charge of organic matter in the environment, the presence of industries, discharge of wastes and effluents, the rate of evaporation and volatility, photo-oxidation processes, bacteria responsible for biodegradation and temperature of the environment [23,19,14]]. Another factor that may have promoted more PAHs in sediment at the Police Post and Minipiti stations over the Iwofe Jetty is reduced water movement, which allows resident time for the accumulation of PAHs in the surface sediments [22]. Another possible factor for PAHs concentration is based on the sediment water hydrodynamics that may have taken place at the three different stations. When the flow of water is slow and lamina (streamline flow) it promotes deposition of pollutants, but when the flow is turbulent (irregular), it gives rise to re-suspension of sediment bound pollutants which are then transported to other places and the actual volume that was originally collected could not be ascertained. Besides these situations that promoted more PAHs in the sediments at Minipiti and Police Post stations, is the nature and disturbance that occur at the Jetty point due to human activities, which will always unsettle pollutants of which PAHs are inclusive.

Table 1: Concentrations of PAHs from Sediments at the different locations

PAHs (mg/Kg)	Stations		
	Iwofe Jetty	Minipiti	Police Post
Naphthalene	ND	ND	ND
Acenaphthylene	ND	1.128±0.05	12.283±3.42
Acenaphthene	ND	ND	1.805± 0.36
Fluorene	ND	2.065±0.45	4.660±1.13
Phenanthrene	0.415±0.12	1.969±0.84	ND
Anthracene	0.710±0.30	ND	4.902±1.54
Fluoranthene	0.151±0.01	0.951±0.22	2.071±0.21
Pyrene	20.226±4.69	6.164±1.54	ND
Benz (a) anthracene	0.892±0.05	3.150±1.21	11.060±3.07
Chrysene	0.056±0.02	2.922±0.71	1.115±0.04
Benzo (b) fluoranthene	0.011±0.00	0.667±0.11	1.268±0.33
Benzo (k) fluoranthene	0.262±0.02	ND	10.453±2.68
Benzo (a) pyrene	0.541±0.06	0.091±0.00	1.477±0.25
Dibenzo (a,h) anthracene	0.176±0.01	11.827±4.21	ND
Benzo (g,h,i) perylene	ND	2.762±0.92	ND
Indeno (1,2,3-cd) pyrene	ND	ND	ND
Total	23.440	33.694	51.094

Concentrations of Different PAHs Categories

The concentrations of the different ring classifications are shown in Table 2. The concentrations of the low molecular weight PAHs varied from 1.125 to 25.650 mg/Kg, while the high molecular weight PAHs varied from 22.315 to 28.534 mg/Kg. The concentrations of the 2-3 ringed PAHs (also referred to as LMW PAHs) varied from 1.125 to 25.650 mg/Kg. The concentrations of 4-ringed PAHs varied from 21.324 to 13.187 mg/Kg. The values of the 5-membered rings ranged from 0.990 to 13.198 mg/Kg and the 6-membered rings had values of range, ND - 2.762 mg/Kg.

The observed higher values of HMW PAHs than the LMW PAHs in the present work is in consonance with the observation of [19] in sediment of Alkahlaa River in Missan Province, Iraq and also those of [20] in sediments along the Aurá River, Northern Brazil. The observed values of the various categories of molecular weight classes showed that the Iwofe Jetty station was dominated by the 4-membered rings as against the other ring classes. At the Minipiti station, the 4 and 5-membered rings were dominant over the other ring members while at the Police post; the 2-3 membered rings were dominant over the other groups. The least concentrated category in all the stations was the six-membered ring. The dominance of any ring type in the sediment as observed in this work may be from the sources, origin, fate, volatility, adsoptivity and absorptivity, transformation, solubility, degradation and mobility of the PAHs [24,22]. In any environment, the dominance of 2-3 membered rings PAHs can be attributed to the length of time the PAHs were deposited [25]. In other words, higher values of LMW PAHs are an indication that most of the PAHs were produced or formed recently. Low concentrations of LMW (2-3 rings) PAHS in sediments from some of the stations can further be explained on the basis of rapid degradation whereas the abundance of HMW PAHs (4-6 rings) is due to recalcitrant bahaviour towards degradation, hydrophobicity and hydrophilicity [26]. The LMW PAHs are more hydrophilic, while the HMW PAHs are more Hydrophobic [27]. which defines the reasons for the occurrence of more HMW PAHs in sediment over LMW PAHs.

However, the almost equal value of LMW PAHs to HMW PAHs at the Police Post station can be ascribed to spills from petroleum products which may have occurred recently and may have passed through the drains to the point of sampling.

Table 2: Concentrations (mg/Kg) and Classification of the Different Ring Categories of PAHs in Sediments from the Stations

Ring Category	Stations		
	Iwofe Jetty	Minipiti	Police Post
LMW	1.125	5.162	25.650
HMW	22.315	28.534	27.444
2-3	1.125	5.162	25.650
4	21.324	13.187	14.246
5	0.99	12.585	13.198
6	ND	2.762	ND

Source Diagnosis of PAHs

The diagnostic ratio and the sources of PAHs are shown in Table 3. The ratio of LMW/HMW PAHs in the stations showed that at Iwofe station, the value was 0.0594, at the Minipiti station, 0.181 while the Police Post station recorded value of 0.935. All the val-

ues observed from the ratio analysis of LMW/HMW gave values < 1. This is a suggestion that the PAHs were from pyrogenic origin. This observation is in agreement with those of [19]. who observed lower than 1 ratio between LMW/HMW PAHs in sediments from Alkahlaa River in Iraq, but at variance with the observation of [28] in sediments from River Jakarta, where the LMW/HMW PAHs ratio exceeded the value of 1, which suggested petrogenic sources of PAHs.

The diagnostic ratio of An/(An+Phe) gave values of 0.631, unresolved value (-) and 1 for Iwofe Jetty, Minipiti and Police Post respectively. The observed values showed that PAHs at Iwofe Jetty and Police Post stations were > 0.1, which suggested pyrogenic sources of origin. The above observation did not agree with those of [21]. That observed values lower than 0.1 in the ratio between An/(An+Phe) in sediments from Yinma River, China, but agrees with those of sediment from Pearl River, China and those of sediments Qinhuai River and Xuanwu Lake, Nanjing, China, where there was excessive coal and grass burning along the area [29,30].

The results of the diagnostic ratio for Fl/(Fl+Pyr) gave values of 0.007, 0.134 and 1 for Iwofe Jetty, Minipiti and Police post stations respectively. The observed values showed that the sources of PAHs in the wetland areas originated from petrogenic sources. However, combined sources may be the reason for the value observed at the Police Post station and values observed in the present work.

The above ratio analysis is used to establish different individual sources of PAHs contamination of sediment. When the ratio of Fl/(Fl+Pyr) gives values that are greater than 0.5, the pyrolytic or pyrogenic sources may have resulted from burning of plants or coal, but values that are lower than 0.4 is deemed to have originated from petroleum spill sources, but value in between 0.4 and 0.5 is a pyrogenic source from burning of fossil fuel [31,32].

The ratio of BaA/(BaA+Chr) in the stations were 0.941, 0.519 and 0.908 for Iwofe, Minipiti and Police Post stations respectively. All the values observed in the stations were greater than 0.35, which showed that the source and origin of the PAHs were pyrogenic. Besides, the ratio of BaA/(BaA + Chr) is mostly used in the diagnosis of coal incineration [33]. All the values observed in the stations were > 0.35, which is an indication that the pyrolytic sources of the PAHs in the sediments from the stations originated from the burning of firewood, coal, grass, and wood [31].

The ratio IP/ (IP+ BgP) used for source identification among the six membered ring could not be used to assess the origin of the PAHs. This is due to the fact that they were not identified or detected during the analysis. The non-detection of members of this category may be due to absence of large molecular weight hydrocarbons in the combusted materials.

Table 3: Diagnostic Ratios and source Identification of PAHs

Stations	LMW/ HMW	An/(An+Phe)	Fl/(Fl+Pyr)	BaA/(BaA+Chr)	IP/ (IP+ BgP)
Iwofe Jetty	0.0504	0.631	0.007	0.941	-
Minipiti	0.181	-	0.134	0.519	-
Police Post	0.935	1	1	0.908	-

The sign (-) in the table represents a situation where either the numerator or both were undetected

Conclusion

The study was able to provide information on the concentrations of polycyclic hydrocarbons in sediments from drainage discharge points into the marshy wetland areas of New Calabar River, along the Rumuolumeni axis of Port Harcourt. The results revealed the presence of 14 PAHs, where naphthalene and Indeno (1,2,3-cd) pyrene were not detected in any of the stations. The summation of the all the PAHs in the different stations showed that Iwofe Jetty < Minipiti < Police Post stations. It was also observed that there were mixed sources of origin of PAHs, although, pyrolytic (pyrogenic) PAHs were dominant over petrogenic PAHs. Therefore, relevant authorities should take steps to control the input sources now that the values are still at low levels.

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