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Sources and Health Risk Assessment of Polycyclic Aromatic Hydrocarbons at Uncontrolled Solid Waste Dumpsites in Port Harcourt City, Nigeria

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Abstracts:-

➢ ***Background:***

Household solid waste dumpsites located within the habitation on humans poses health risk via ingestion, dermal contact and inhalation when Polycyclic Aromatic Hydrocarbons are generated. This study investigated the concentration levels of PAHs in the soil at dumpsites for human health risk assessment at solid waste dumpsites located in Markets, Semi-industrial and Residential areas.

➢ ***Materials and Method:***

Fifteen soil samples were collected in each season and analyzed at the Rivers State University, Institute of Pollution Studies Research Laboratory Gas Chromatography. Each of the season laboratory data were subjected to Ecological Toxicity, Environmental Risk Quotient and Incremental lifetime Cancer Risk (ILCR) Analysis.

➢ ***Results:***

High Molecular weight (HMW) PAHs which are toxicologically relevant were detected in all the three classified dumpsites in both seasons with highest value of Chrysene in residential dumpsites during rainy season and Dibenez(a,h)anthracene in dry season. Each of classified dumpsites has average total PAHs values greater than 1mg/kg (>1) in Rainy season while in dry season, only market dumpsites has an average greater than 1mg/kg.. On health risk assessment, The PAHs generated at each classified dumpsite during Rainy season are from pyrogenic source since the Index (PI) values are all less the 1(<1), however residential dumpsites with PI > 1 in dry season indicates petrogenic source. The Risk Quotient (Negligible concentrations (RQNCs) values for the PAHs and the Risk Quotient Maximum Permissible concentration (RQMPCs) of individual PAHs were all less than 1.0, indicating that the listed PAHs pose a moderate level of ecological risk in these study sites since they are all less than the threshold ecological risk values of 0.8mg/kg. The Incremental Life Cancer Risk (ILCR) values of the PAHs on most sites during Rainy season are higher than 1 x10^{-6} which by USEPA standard, indicates risk of cancer. However, the Dry season shows lesser values.

➢ ***Conclusion***

The PAHs at most of the dumpsites have pyrogenic source and toxically contaminated with high molecular weight Polycyclic Aromatic Hydrocarbons. The ILCR values for children is higher than adults.

Keywords:- *Polycyclic Aromatic Hydrocarbons, Pyrogenic, Health Risk.*

I. INTRODUCTION

Polycyclic Aromatic Hydrocarbons are ubiquitous environmental pollutants generated primarily during the incomplete combustion of organic materials (e.g. coal, oil, petrol, and wood). Emissions from anthropogenic activities predominate; nevertheless, some PAHs in the environment originate from natural sources such as open burning, natural losses or seepage of petroleum or coal deposits, and volcanic activities. Many PAHs have toxic, mutagenic and/or carcinogenic properties. PAHs are highly lipid soluble and thus readily absorbed from the gastrointestinal tract of mammals.

The following three types: *pyrogenic*, *petrogenic*, and *biological* are the major PAH sources to the environment. In a process called pyrolysis, *Pyrogenic* PAHs are formed whenever organic substances are exposed to high temperatures under low oxygen or no oxygen conditions. The temperatures at which the pyrogenic processes occur are ranging from about (350 °C to more than 1200 °C). Pyrogenic PAHs are generally found in greater concentrations in urban areas and in locations close to major sources of PAHs. In addition, PAHs can also be formed at lower temperatures. It is worth mentioning that crude oils contain PAHs that formed over millions of years at temperatures as low as (100–150 °C). In this respect, PAHs formed during crude oil maturation and similar processes are called *petrogenic*. Such *petrogenic* PAHs are common due to the widespread transportation, storage, and use of crude oil and crude oil products. On the other hand, it is not well-known that PAHs can be produced *biologically*. For example, they can be synthesized by certain plants and bacteria or formed during the degradation of vegetative matter.

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Reference[3] examined the concentrations and compositional patterns of the United States Environmental Protection Agency (US EPA) 16 priority polycyclic aromatic hydrocarbons (PAHs) in surface soils of an urban environment in the Niger Delta of Nigeria with a view to providing information on the sources, extent of contamination and human health risks of PAHs in these soils. The analyses were performed by means of gas chromatography-mass spectrometry (GC-MS) after extraction of the soils with hexane/dichloromethane and clean-up of the extracts. The concentration of Σ16 PAHs in the urban soils ranged from 188 to 684 µg kg-1, while the ΣPAH7c (carcinogenic PAHs) ranged from 28.5 and 571 µg kg-1. The estimated carcinogenic and mutagenic potency factors for these sites ranged from 2.34 to 197 and 9.66 to 195 µg kg-1 respectively. The composition of PAHs in these soils follows the order: 5-rings>4-rings>3- rings>6-rings>2-rings, and higher molecular weight PAHs accounted for a significant proportion of the Σ16 PAH concentration in this study. The results indicated that there is a high potential risk of cancer development as a result of exposure of PAHs via ingestion, dermal contact and inhalation. The diagnostic ratios indicate that the PAHs in these soils originated mainly from pyrogenic processes, such as combustion of petroleum, fossil fuels and biomass such as woods, charcoal straw and grasses.

Most past studies on PAH are carried out on soil suspected to have been contaminated with either hydrocarbon products or arear where combustion has taken place. There are various domestic solid waste dumpsites that are in markets, semi-industrial and residential areas. These dumpsites are the first place where domestic solid waste is dumped. The presence of PAHs at these domestic solid waste dumpsites are generally not considered since combustion is not imagined at these sites most especially the hydrocarbon presence considered negligible. This study is set to primarily establish the presence, the type of PAHs as well as the health risk to environment and humans.

PAHs	Chemical Formula	Molecular weight	No of Rings
Low molecular weight			
Naphthalene	$C_{10}H_8$	128	2
Acenaphthylene	$C_{12}H_8$	152	3
Acenaphthene	$C_{12}H_{10}$	154.21	3
Fluorene	$C_{13}H_{10}$	166.2	3
Phenanthrene	$C_{14}H_{10}$	178.2	3
Anthracene	$C_{14}H_{10}$	178.2	3
Medium Molecular weight			
Fluoranthene	$C_{16}H_{10}$	202.16	4
Pyrene	$C_{16}H_{10}$	202.3	4
High Molecular weight			
Chrysene	$C_{18}H_{12}$	228.3	4
Benzo(a)anthracene	$C_{18}H_{12}$	228.89	4
Benzo(k)fluoranthene	$C_{20}H_{12}$	251.3	5
Benzo(a)pyrene	$C_{20}H_{12}$	251.3	5
Indono(1,2,3-Cd)pyrene	$C_{22}H_{12}$	276.3	6
Benzo(g,h,i)pevylene	$C_{22}H_{12}$	276.3	6
Dibenzo(a,h)anthracen	$C_{22}H_{12}$	278.35	6

Table 1:- Classification of Polycyclic Aromatic Hydrocarbon (PAHs) Based on Molecular Weight
Source: ATSDR (Agency for Toxic Substances and Disease Registry). Atlanta, GA, 1995;458

➢ *Ecological Toxicity & Risk Assessment for PAH*

The determination and distribution of potential sources of PAHs in the environment using diagnostic ratios[11] such as Phe/Ant, Flu/Pyr, BaA/Chr, Flu/Flu+Pyr, and Ant/Ant+Phe. It is also suggested that the pyrogenic index (PI), i.e., the ratio of LMW to HMW or vice versa, is applicable to determine the potential sources of PAHs . LMWPAHs are dominated by a homologous series of five petrogenic alkylated PAHs (naphthalene, phenanthrene, dibenzothiophene, fluorene, and chrysene), while the HMWPAHs are chiefly pyrogenic compounds[12]. . Therefore, using the PI can better reflect the potential sources of PAHs. In addition to the diagnostic ratio and PI, total index (TI) is also used to identify the high-temperature (combustion) or low-temperature (petroleum) sources of PAHs. TI, which is the ratio of (Flu/Flu+Pyr)/0.4 + (Ant/Ant+Phe)/0.1 + (BaA/BaA+Chr)/0.2 . This is used in this study. A TI > 4 indicates that PAHs have originated mainly from combustion, while lower values indicate petrogenic sources. For this study, identification of the potential sources of PAHs in soil was carried out using PAH diagnostic ratios, PI, and TI.

Evaluation of the ecological toxicity of the PAH compounds in the soil was carried out by comparing the risk quotient (RQ) of PAHs under investigation and their corresponding environmental quality values. There is insufficient toxicological data for PAHs in agricultural soil [4]. The environmental quality concentration values negligible concentration (NCs and maximum permissible concentration (MPSc) for Phe, Ant, Flu, BaA , Chr, and Bap were taken from the work of [6]. Researchers have agreed that PAHs with the same toxicity equivalence Factor (TEF)[8] have similar human and ecological health impacts

[14][2]. Therefore, the environment quality values of congeners with the same TEF were used to calculate the environmental risk quotient of Nap, Acy, Ace, Flr, and Pyr. The individuals and total environment risk quotient were determined using the following equations:

$$RQ = \frac{CPAHs}{CQV} \quad \text{..... (1)}$$

$$RQNC = \frac{CPAHs}{CQV(NCs)} \quad \text{..... (2)}$$

$$RQMPCs = \frac{CPAHs}{CQV(NCs)} \quad \text{..... (3)}$$

$$RQ \sum PAHs \sum_{n=1}^{11} RQi(RQi \geq 1) \quad \text{..... (4)}$$

$$RQ \sum PAH(NCs) = \sum (n=1) \wedge 11(RQi(NCs)(RQi)(NCs) \geq 1) \quad \text{..... (5)}$$

$$RQ \sum PAH(NCs) = \sum_{n=1}^{11} RQi(MPCs(RQi)(MPCs) \geq 1) \quad \text{.....(6)}$$

Where CPAHs is the concentration of certain PAHs in soil. CQV is the corresponding quality values of certain PAHs, NCs and MPCs are respectively, the negligible and maximum permissible concentrations of PAHs in soil, RQ is the risk quotient, and CQVMPSCs are quality values of the NCs of PAHs in soil, and CQVMPCs are the quality values of MPCs in soil.

The total summed environmental risk RQΣPAHs was calculated by considering the individual RQMPCs and RQMPCs $\geq 1.$ the environmental risk levels for individual and total PAHs are summarized in Table 2.. In addition to the risk quotients, the carcinogenic risk of individual PAHs was calculated from concentration of each PAHs and their corresponding toxicity equivalency factor [8].

Individual PAHs					**∑PAHs**	
Risk Grade	**RQ(NCs)**	**RQ(MPCs)**		**Risk Grade**	**RQ∑PAHs(NCs)**	**RQ∑PAHs(MPCs)**
Lower risk	< 1	<1		Risk - Free	<1	<1
Medium Risk	≥ 1	< 1		Low Risk	≥ 1, < 800	<1
High Risk	≥ 1	≥ 1		Medium Risk 1	≥ 800	<1
				Medium Risk 2	< 800	≥1
				High Risk	≥800	≥1

Table 2:- The Environmental Quality Values Negligible Concentration and Maximum Permissible Concentration of Polycyclic Aromatic Hydrocarbons
RQ: Risk Quotient. Source:[19][2]

Human health impacts of persistent organic pollutants can arise through dermal contact, ingestion, and inhalation. Human risk assessments of PAHs were evaluated by using the equations adopted from previous work[20][7]. The toxicity equivalency quotient (TEQ) is an incremental lifetime cancer risk (ILCR) of each PAH through direct ingestion dermal contact, and inhalation for adults and children were calculated using Equations (7-10):

$$TEQ = \sum_{n-1} PAHs * TEF_i \quad \text{..... (7)}$$

$$ILCRSingestion = \frac{CS\, x \left(CSFingestion\, x \sqrt[3]{\frac{BW}{70}}\, x\, IRsoil\, x\, EF\, x\, ED \right)}{BW\, x\, AT\, x\, cf} \quad \text{.....(8)}$$

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$$ILCRdermal = \frac{CS\,x\left(CSFdermal\,x\sqrt[3]{\frac{BW}{70}}\;x\;SA\,x\,AF\,x\,ABS\,x\,EF\,x\,ED\right)}{BW\,x\,AT\,x\,cf} \quad \ldots\ldots\ldots (9)$$

$$ILCRinhalation = \frac{CS\,x\left(CSFinhalation\;x\;\sqrt[3]{\frac{BW}{70}}\;x\;IRair\,x\,EF\,x\,ED\right)}{BW\,x\,AT\,x\,PEF\;x\;cf} \quad \ldots\ldots\ldots (10)$$

Where TEQ is the toxicity equivalence quotient, TEF is the toxicity equivalence factor,[8] CS is the PAH concentration of soils (μg kg^{-1}), CSF is the carcinogenic slope factor (μg kg^{-1}) $^{-1}$, CSF was based on the cancer-causing ability of BaP, and the CSfingestion, CSFdermal, and CSFinhalation of BaP were 7.3, 25, and 3.85 (μg kg^{-1} $day^{-1})^{-1}$, respectively. BW is body weight (70kg), AT is average life span (70 years), EF is exposure frequency (350 days $years^{-1}$), ED is the exposure duration (30 years), IRsoil is an intake rate (100mg day^{-1}), IRair is inhalation rate ($20m^3$ day^{-1}), cf is conversion factor (10^6), AF is the dermal adherence factor ($10mgcm^{-2}$), ABS is the dermal absorption fraction unitless (0.1), and PEF is the soil dust produce factor (1.32 x 10^9) m^3 kg^{-1}). The total risks were the sum of risks of the ILCRs in terms of direct ingestion, dermal contact, and inhalation. For children, BW (15kg), EF (189 days) ED (6 years), IRair (10 m^3day^{-1}), SA (2800 cm^2 day^{-1}), and AF (0.2 $kgcm^{-1}$) were assumed[18].

II. MATERIALS AND METHODS

Port Harcourt is the capital and largest city of Rivers State, Nigeria. It lies along the Bonny River and is located in the Niger Delta. As of 2016, the Port Harcourt urban area has an estimated population of 1,865,000 inhabitants, up from 1,382,592 as of 2006. The urban area (Port Harcourt metropolis), on the other hand, is made up of the local government area itself and parts of Obio-Akpor and Eleme accordingly. Port Harcourt, which is the current capital of Rivers State, is highly congested as it is the only major city of the state.

The area of study is in Port Harcourt metropolis, Rivers State. The area is bounded geographically by latitudes 4°46'N to 5°00'N and longitudes 6°55' E to 7°03' E. Open dump sites are the most common waste disposal methods in Port Harcourt and many cities in Nigeria. Open dump sites are found in several residential, Markets and semi – industrial locations around the city, for example, Rukpokwu village, Rumuokoro, Rumuomasi, Diobu, Marine base, and Borokiri, to mention a few. The dumpsites within the study area and the locations of all the sampling points were recorded with the aid of a garmin Global Positioning System (GPS). include; Residential, Markets & Industrial as stated below:

Soil samples for PAH determination were collected in aluminum foil from the various dumpsites. An auger was used for collecting soil samples at shallow depth of about 1 - 15cm. For each sampling point, three samples were taken from the same area and mixed thoroughly to form a composite homogenous sample. Sampling tools were washed with water and dried before the next sample was collected. About 1 kg of soil sample were collected at each sampling site in order to ensure that enough fine-grained material would be available for analysis. Fifteen (15) soil samples were collected for each season, from a different dumpsite (Table 3). Samples were labeled properly including date of collection, location and code number of soil samples.

2g samples were weighed into a clean extraction container. 20 ml of extraction solvent (hexane) was added into the sample, mixed thoroughly and allowed to settle. The mixture was carefully filtered into solvent-rinsed extraction bottles using filter paper fitted into Buchner funnels. The extracts were concentrated to 2 ml and then transferred for clean-up/separation.1cm of moderately packed glass wool was placed at the bottom of 10 mm I.D (internal diameter) x 250 mm long chromatographic column. Slurry of 2 g activated silica in 10ml dichloromethane was prepared and placed into the chromatographic column. To the top of the column was added 0.5 cm of sodium sulphate. The column was rinsed with additional 10 ml of dichloromethane. The column was pre-eluted with 20 ml of hexane. This was allowed to flow through the column at the rate of about 2 minutes until the liquid in the column was just above the sodium sulphate layer. Immediately, 1 ml of the extracted sample was transferred into the column. The extraction bottle was rinsed with 1 ml of hexane and the dissolved extract was added to the column as well. The stop cork of the column was opened and the eluent was collected in a 10 ml graduated measuring cylinder. Just prior to exposure of the sodium sulphate layer to air, hexane was added to the column in 1 – 2 ml increments. Accurately measured volume of 8 – 10 ml of the volatile aromatics (BTEX) as applicable.

Gas Chromatographic analysis: The concentrated aliphatic or aromatic fractions were transferred into labelled glass vials with Teflon or rubber crimp caps for GC analysis. 1 ul of the concentrated sample was injected by means of an hypodermic syringe through a rubber septum into the column. Separation occurs as the vapour constituent partitions between the gas and liquid phases. The sample was automatically detected as it emerged from the column by the FID detector whose response is dependent upon the composition of the vapour.

S/no		Location Name	Codes	GPS
1	Market Areas (MA)	Okija Market	MA01	N 04º 47' 50.5''
				E 006º 59' 54.6''
2		Creek Road Market	MA02	N 04º 45' 31.5''
				E 007º 01' 33.0''
3		Rumokuta (Market area)	MA03	N 04º 51'' 58.2''
				E 007º 00' 02.8''
4		Water side (Creek road)	MA04	N 04º 45' 31.2''
				E 007º 01' 27.4''
5		Mile 1 Market	MA05	N 04º 47' 37.2''
				E 006º 59' 43.8''
6	Semi Industrial area. (SIA)	Trans Amadi	SIA01	N 04º 49' 22.7''
				E 007º 02' 22.1''
7		Stadium road	SIA02	N 04º 49' 30.9''
				E 007º 01' 29.2''
8		Odili Road	SIA03	N 04º 48' 14.2''
				E 007º 02' 57.7''
9		Elekohia	SIA04	N 04º 49' 07.8''
				E 007º 01' 35.0''
10		D/Line	SIA05	N 04º 48' 10.1''
				E 007º 00' 00.8''
11	Residential Areas (RA)	Borokiri	RA01	N 04º 44' 47.4''
				E 007º 02' 29.5''
12		Marine base	RA02	N 04º 46' 17.4''
				E 007º 01' 29.0''
13		Elekohia Residence	RA03	N 04º 49' 06.2''
				E 007º 01' 49.4''
14		Diobu residential	RA04	N 04º 47' 33.6''
				E 007º 00' 03.0''
15		Rukpokwu village	RA05	N 04º 54' 11.7''
				E 006º 59' 19.0''

Table 3:- Locations for soil sampling

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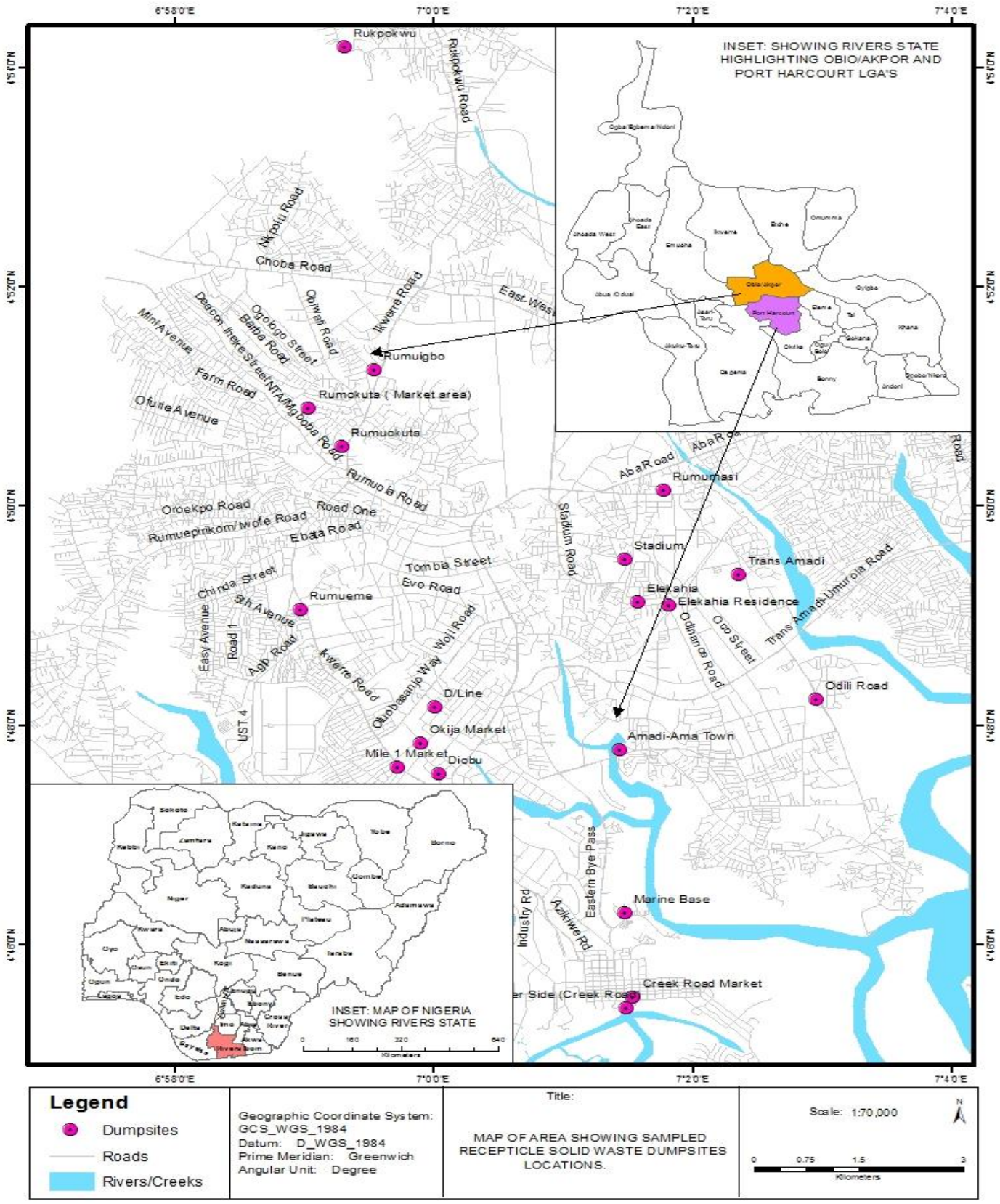

Fig 1:- Map of study area, Portharcourt City , Nigeria

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III. RESULTS

			Market Area					Semi - Industrial					Residential				
			Okija Market	Creek Road Market	Rumokuta (Market area)	Water side (Creek road)	Mile 1 Market	Trans Amadi	Stadium Road	Odili Road	Elekohia	D/Line	Borokiri	Marine Base	Elekohia	Diobu	Rukpokwu village
			N 04o 47' 50.5'' E 006o 59' 54.6''	N 04o 45' 31.5'' E 007o 01' 33.0''	N 04o 51'' 58.2'' E 007o 00' 02.8''	N 04o 45' 31.2'' E 007o 01' 27.4''	N 04o 47' 37.2'' E 006o 59' 43.8''	N 04o 49' 22.7'' E 007o 02' 22.1''	N 04o 49' 30.9'' E 007o 01' 29.2''	N 04o 48' 14.2'' E 007o 02' 57.7''	N 04o 49' 07.8'' E 007o 01' 35.0''	N 04o 48' 10.1'' E 007o 00' 00.8''	N 04o 44' 47.4'' E 007o 02' 29.5''	N 04o 46' 17.4'' E 007o 01' 29.0''	N 04o 49' 06.2'' E 007o 01' 49.4''	N 04o 47' 33.6'' E 007o 00' 03.0''	N 04o 54' 11.7'' E 006o 59' 19.0''
S/no	**PAH name**	**No of rings**	**MA001**	**MA002**	**MA003**	**MA004**	**MA005**	**SIA001**	**SIA002**	**SIA003**	**SIA004**	**SIA005**	**RA001**	**RA002**	**RA003**	**RA004**	**RA005**
1	Naphthalene	Two	0.005	0.055	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.048	0.000	0.000	0.000	0.000	0.001
2	Acenaphthylene	Two	0.039	0.019	0.003	0.003	0.000	0.000	0.079	0.000	0.000	0.006	0.000	0.000	0.012	0.000	0.026
3	Acenaphthene	Two	0.209	0.026	0.013	0.001	0.009	0.017	0.014	0.002	0.004	0.007	0.003	0.007	0.003	0.004	0.005
4	Fluorene	Two	0.120	0.007	0.009	0.004	0.007	0.019	0.093	0.002	0.008	0.005	0.008	0.002	0.005	0.005	0.005
5	Phenanthrene	Three	0.042	0.007	0.013	0.007	0.013	0.229	0.075	0.010	0.011	0.012	0.006	0.011	0.019	0.004	0.007
6	Anthracene	Three	0.015	0.010	0.007	0.002	0.012	0.036	0.274	0.009	0.009	0.024	0.005	0.007	0.012	0.004	0.003
7	Fluoranthene	Three	0.010	0.023	0.025	0.023	0.011	0.036	0.379	0.018	0.014	0.044	0.009	0.027	0.016	0.010	0.011
8	Pyrene	Four	0.113	0.007	0.011	0.001	0.004	0.003	0.022	0.002	0.005	0.024	0.002	0.002	0.010	0.018	0.001
9	Benz(a)anthracene	Four	0.009	0.006	0.031	0.006	0.003	0.010	0.032	0.004	0.004	0.014	0.002	0.009	0.016	0.106	0.009
10	Chrysene	Four	0.141	0.212	0.641	0.365	0.741	0.296	0.054	0.434	0.775	0.455	0.678	0.656	0.765	2.839	0.418
11	Benzo(b)Fluoranthene	Four	0.156	0.268	0.420	0.078	0.180	0.165	0.180	0.216	0.065	0.238	0.342	0.156	0.265	0.235	0.035
12	Benzo(k)fluoranthene	Four	0.090	0.145	0.072	0.083	0.045	0.079	0.033	0.009	0.058	0.076	0.073	0.109	0.094	0.144	0.092
13	Indeno (1,2,3-cd)Pyrene	Five	0.371	0.224	0.360	0.321	0.265	0.212	0.105	0.005	0.070	0.185	0.448	0.458	0.267	0.156	0.172
14	Dibenez(a,h)anthracene	Five	0.552	0.531	0.283	0.243	0.264	0.369	0.189	0.180	0.089	0.377	0.381	0.234	0.397	1.100	0.810
	Total PAH (Mg/kg)		**1.872**	**1.54**	**1.888**	**1.137**	**1.556**	**1.471**	**1.529**	**1.012**	**1.112**	**1.516**	**1.958**	**1.679**	**1.879**	**4.625**	**1.595**

Table 4:- RAINY season PAH Results (mg/kg)

No of samples	Location name	Nap	Acy	Ace	Flr	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkFlu	Ipyr	DAnt	PAH (mg/kg)
5	**Market sites**	0.012	0.013	0.052	0.03	0.017	0.009	0.018	0.027	0.011	0.42	0.22	0.087	0.308	0.375	1.599
5	**Semi Indusrial Sites**	0.01	0.017	0.009	0.025	0.068	0.07	0.098	0.011	0.013	0.403	0.173	0.051	0.115	0.241	1.304
5	**Residential sites**	0	0.008	0.005	0.005	0.009	0.006	0.015	0.006	0.029	1.071	0.206	0.103	0.3	0.584	2.347
5	**Control Sites**	0	0	0	0	0	0.0004	0.0004	0.0003	0.0001	0.0019	0.0104	0.0263	0.0037	0.0082	0.0517

Table 5:- RAINY season Mean / Average PAH(mg/kg)

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			Market Area					Semi - Industrial					Residential				
			Okija Market	Creek Road Market	Rumokuta (Market area)	Water side (Creek road)	Mile 1 Market	Trans Amadi	Stadium Road	Odili Road	Elekohia	D/Line	Borokiri	Marine Base	Elekohia	Diobu	Rukpokwu village
			N 04o 47' 50.5'' E 006o 59' 54.6''	N 04o 45' 31.5'' E 007o 01' 33.0''	N 04o 51'' 58.2'' E 007o 00' 02.8''	N 04o 45' 31.2'' E 007o 01' 27.4''	N 04o 47' 37.2'' E 006o 59' 43.8''	N 04o 49' 22.7'' E 007o 02' 22.1''	N 04o 49' 30.9'' E 007o 01' 29.2''	N 04o 48' 14.2'' E 007o 02' 57.7''	N 04o 49' 07.8'' E 007o 01' 35.0''	N 04o 48' 10.1'' E 007o 00' 00.8''	N 04o 44' 47.4'' E 007o 02' 29.5''	N 04o 46' 17.4'' E 007o 01' 29.0''	N 04o 49' 06.2'' E 007o 01' 49.4''	N 04o 47' 33.6'' E 007o 00' 03.0''	N 04o 54' 11.7'' E 006o 59' 19.0''
S/no	PAH name	No of rings	MA001	MA002	MA003	MA004	MA005	SIA001	SIA002	SIA003	SIA004	SIA005	RA001	RA002	RA003	RA004	RA005
1	Naphthalene	Two	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
2	Acenaphthylene	Two	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000-	0.000	0.000
3	Acenaphthene	Two	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.051	0.000
4	Fluorene	Two	0.000	0.000	0.000	0.000	0.000	0.140	0.003	0.000	0.000	0.000	0.000	0.000	0.012	0.002	0.013
5	Phenanthrene	Three	0.115	0.000	0.000	0.000	0.000	0.054	0.020	0.000	0.000	0.081	0.000	0.000	0.000	0.016	0.002
6	Anthracene	Three	0.024	0.002	0.000	0.000	0.000	0.002	0.001	0.000	0.000	0.100	0.001	0.000	0.001	0.008	0.003
7	Fluoranthene	Four	0.052	0.007	0.000	0.001	0.000	0.000	0.010	0.015	0.000	0.075	0.001	0.000	0.005	0.024	0.004
8	Pyrene	Four	0.005	0.156	0.522	0.000	0.000	0.000	0.000	0.000	0.000	0.147	0.000	0.000	0.002	0.012	0.002
9	Benz(a)anthracene	Four	0.005	0.060	0.074	0.248	0.000	0.000	0.000	0.008	0.000	0.114	0.001	0.000	0.003	0.008	0.002
10	Chrysene	Four	0.012	0.053	0.034	0.157	0.210	0.000	0.001	0.060	0.000	0.027	0.003	0.000	0.005	0.003	0.000
11	Benzo(b)Fluoranthene	Four	0.025	0.201	0.125	0.108	0.404	0.000	0.003	0.069	0.000	0.048	0.003	0.000	0.004	0.005	0.000
12	Benzo(k)fluoranthene	Four	0.014	0.014	0.206	0.069	0.076	0.063	0.064	0.000	0.000	0.016	0.001	0.000	0.000	0.000	0.000
13	Indeno(1,2,3-cd)Pyrene	Six	0.018	0.083	0.241	0.311	0.402	0.036	0.005	0.091	0.029	0.056	0.032	0.000	0.000	0.000	0.000
14	Dibenez(a,h)anthracene	Five	0.009	0.152	0.195	0.995	0.740	0.146	0.002	0.050	0.000	0.119	0.003	0.000	0.000	0.000	0.000
	Total PAH (Mg/kg)		0.279	0.729	1.4	1.89	1.834	0.44	0.11	0.426	0.03	0.783	0.045	0.000	0.034	0.129	0.003

Table 6:- DRY season PAH Results (mg/kg)

No of samples	Location name	Nap	Acy	Ace	Flr	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkFlu	Ipyr	DAnt	PAH (mg/kg)
5	Market sites	0	0	0	0	0.023	0.0052	0.012	0.1366	0.0774	0.0932	0.1726	0.0758	0.211	0.4182	1.225
5	Semi Industrial sites	0	0	0	0.03	0.031	0.0206	0.02	0.0294	0.0244	0.0176	0.024	0.0286	0.0434	0.0634	0.3324
5	Residential sites	0	0	0.01	0.01	0.0036	0.0026	0.0068	0.0032	0.0028	0.0022	0.0024	0.0002	0.0064	0.0006	0.0508
5	Control sites	0.0046	0.003	0.0067	0	0	0.0008	0.0018	0.0013	0.0008	0.0009	0.0011	0.0001	0	0.0019	0.023

Table 7:- DRY Season Mean / Average PAH (mg/kg)

➢ *Seasonal Variation of Polycyclic Aromatic Hydrocarbons Levels in Soil.*

	Market		Residential		Semi- Industrials		Control	
	Rainy	Dry	Rainy	Dry	Rainy	Dry	Rainy	Dry
Naphthalene	0.012	0.000	0.010	0.000	0.000	0.000	0.0000	0.0046
Acenaphthylene	0.013	0.000	0.017	0.000	0.008	0.000	0.0000	0.0030
Acenaphthene	0.052	0.000	0.009	0.000	0.005	0.010	0.0000	0.0067
Fluorene	0.030	0.000	0.025	0.029	0.005	0.006	0.0000	0.0000
Phenanthrene	0.017	0.023	0.068	0.031	0.009	0.004	0.0000	0.0000
Anthracene	0.009	0.005	0.070	0.021	0.006	0.003	0.0004	0.0008
Fluoranthene	0.018	0.012	0.098	0.020	0.015	0.007	0.0004	0.0018
Pyrene	0.027	0.137	0.011	0.029	0.006	0.003	0.0003	0.0013
Benz(a)anthracene	0.011	0.078	0.013	0.024	0.029	0.003	0.0001	0.0008
Chrysene	0.420	0.093	0.403	0.018	1.071	0.002	0.0019	0.0009
Benzo(b)Fluoranthene	0.220	0.173	0.173	0.024	0.206	0.002	0.0104	0.0011
Benzo(k)fluoranthene	0.087	0.076	0.051	0.028	0.103	0.000	0.0263	0.0001
Indeno(1,2,3-cd)Pyrene	0.308	0.211	0.115	0.043	0.300	0.006	0.0037	0.0000
Dibenez(a,h)anthracene	0.375	0.418	0.241	0.063	0.584	0.001	0.0082	0.0019

Table 8: - Average PAHs Concentration at the Dumpsites in RAINY and DRY seasons

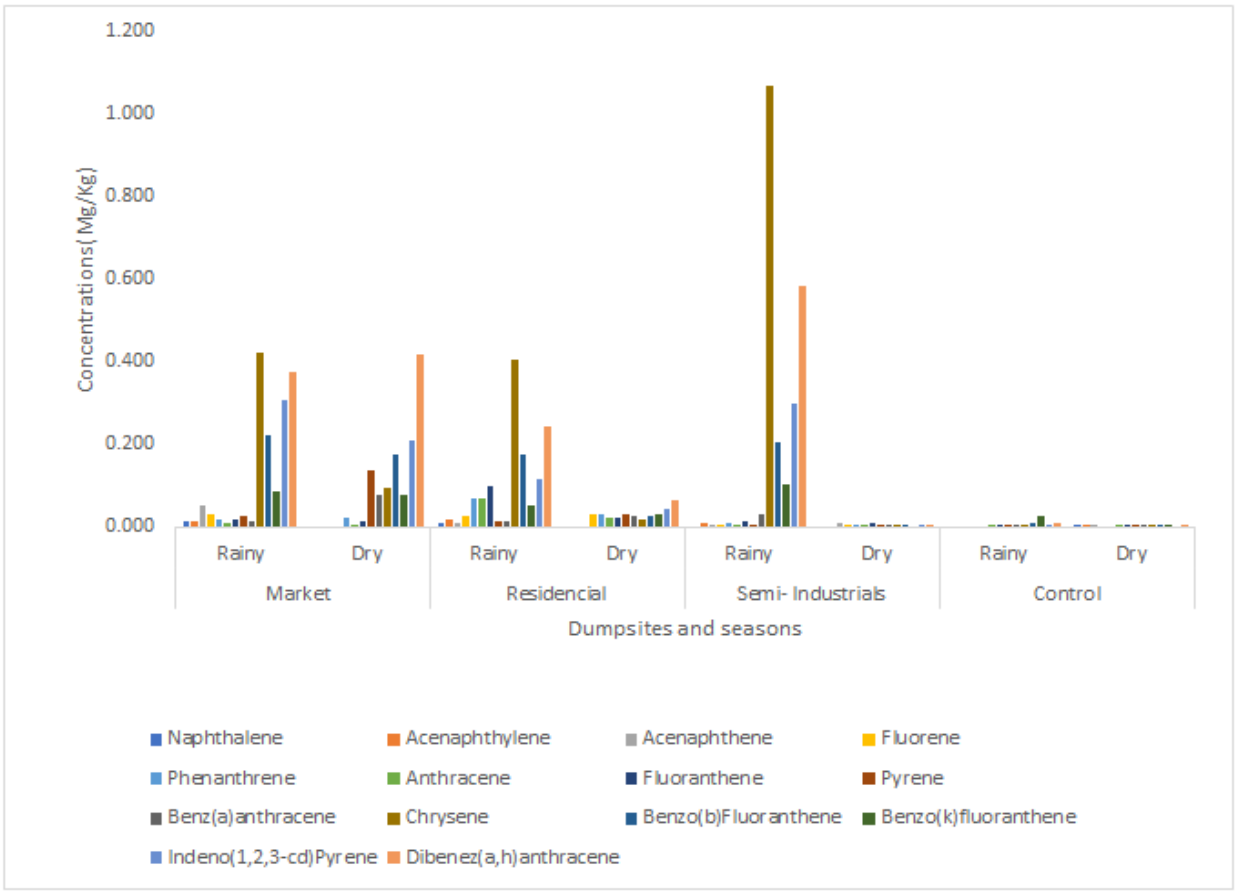

Fig. 2:- General graphical comparative analysis of PAHs Concentration at the dumpsites in RAINY and DRY seasons

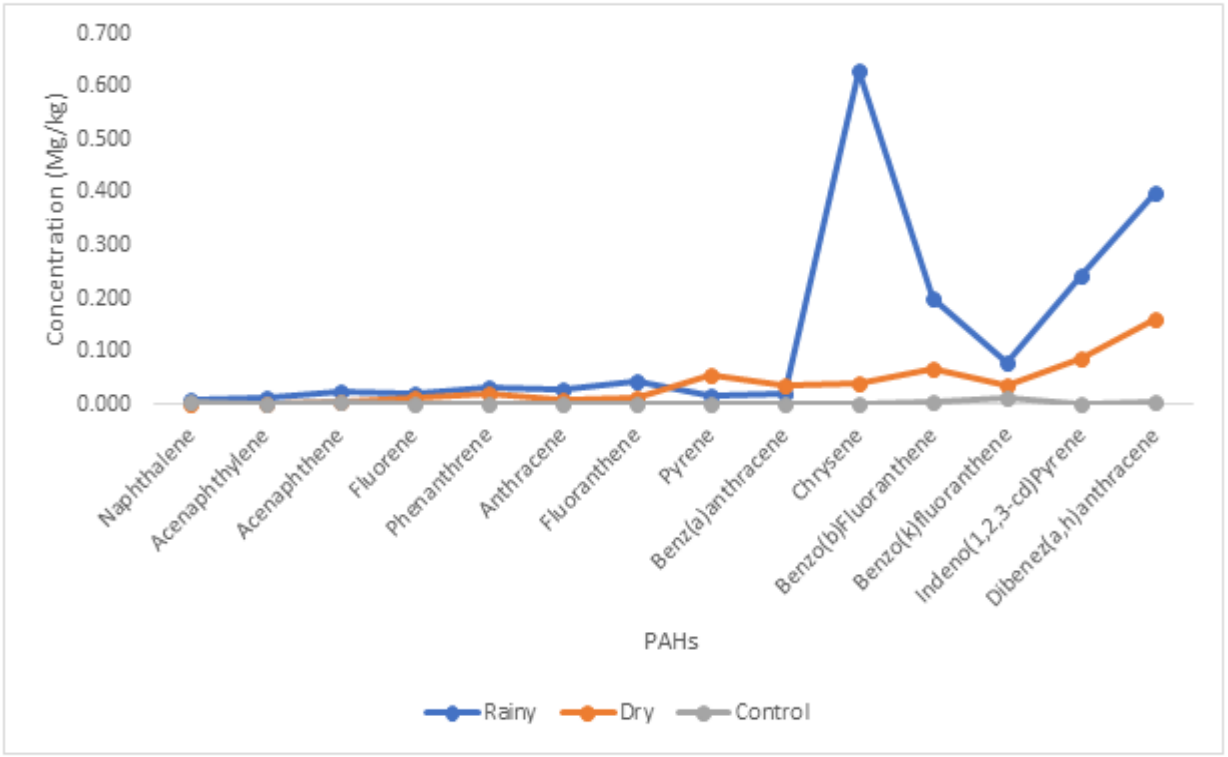

Fig. 3:- General Graphical Comparative Analysis of PAHs Concentration at the Dumpsites in RAINY and DRY seasons

➢ *Sources of PAHs*

To determine the distribution and potential sources of PAHs in the environment using diagnostic ratios [10] such as Phe/Ant, Flu/Pyr, BaA/Chr, Flu/Flu+Pyr, and Ant/Ant+Phe.

PI is Pyrogenic Index used to determine the potential sources of PAHs.

TI is Total Index used to identify the high-temperature (combustion) or low-temperature (petroleum) sources of PAHs. TI, which is the ratio of (Flu/Flu+Pyr)/0.4 + (Ant/Ant+Phe)/0.1 + (BaA/BaA+Chr)/0.2 . was employed in this study. A TI > 4 indicates that PAHs have originated mainly from combustion, while lower values indicate petrogenic sources.

The seasonal variations of PIs and TIs for this study are computed below in Table 9 are shown on Table

When PI is <1 and TI is > 4 : the PAHs source is Pyrogenic due to high temperature combustion.

When PI is >1 and TI is > 4: The PAHs source is Petrogenic (Presence of hydrocarbon products) with Low temperature combustion.

	Rainy		Dry	
Dumpsites	**PI**	**TI**	**PI**	**TI**
Markets	0.091	4.588	0.024	4.309
Semi- Industrial	0.18	7.475	0.325	7.907
Residential	0.014	5.917	1.065	8.693
Control	0.007	11.678	1.911	13.804

Table 9:- Seasonal Variation of Pyrogenic Index (PI), and Total Index (TI) of the Investigated PAHs

Table 9 shows the calculated values of Pyrogenic Index (PI) and Total Index (TI) of the Polycyclic Aromatic Hydrocarbons(PAHs) for solid waste dumpsites located in market areas to be 0.091 and 4.588 for rainy season and 0.024 and 4.309 for dry season. Since the PI is < 1 and TI is >4, it clearly shows that the PAHs source is Pyrogenic and occur as a result of high temperature combustion. This is also similar to dumpsites located at Semi- industrial areas with values PIs and Tis 0.18, 7.475 for rainy season and 0.325 ,7.907 for dry season. However, dumpsites located at residential areas shows pyrogenic source for rainy season with values PI: 0.014 and TI: 5.917 while in dry season, it shoe that the PAHs sources is petrogenic with PI:1.065 and TI: 8.693.

➢ *Environmental Risk Quotient*

(RQMPCs)

	Market				Semi - Industrial				Residential			
	Rainy		Dry		Rainy		Dry		Rainy		Dry	
PAHs	RQ(NCs)	RQ(MPCs)	RQ(NCs)	RQ(MPCs)	RQ(NCs)	RQ(MPCs)	RQ(NCs)	RQ(MPCs)	RQ(NCs)	RQ(MPCs)	RQ(NCs)	RQ(MPCs)
Nap	8.57	0.09	0	0	6.86	0.07	0	0	0.14	0.00	0	0
Acy	10.83	0.11	0	0	14.17	0.14	0	0	6.33	0.06	0	0
Ace	43.33	0.43	0	0	7.33	0.07	0	0	3.67	0.04	8.50	0.09
Flr	11.15	0.11	0	0	9.77	0.10	11.00	0.11	1.92	0.02	2.08	0.02
Phe	3.14	0.03	4.51	0.05	13.22	0.13	6.08	0.06	1.84	0.02	0.71	0.01
Ant	7.67	0.08	4.33	0.04	58.67	0.59	17.17	0.17	5.17	0.05	2.17	0.02
Flu	6.92	0.07	4.62	0.05	37.77	0.38	7.69	0.08	0.01	0.06	2.62	0.03
Pyr	22.5	0.23	113.83	1.14	9.33	0.09	24.50	0.25	5.50	0.06	2.67	0.03
BaA	4.4	0.04	30.96	0.31	5.12	0.05	9.76	0.10	11.36	0.11	1.12	0.01
Chr	3.9	0.04	0.87	0.01	3.76	0.04	0.16	0.00	10.01	0.10	0.02	0.00
Sum	**122.44**	**1.23**	**159.12**	**1.59**	**166.00**	**1.66**	**76.36**	**0.76**	**51.56**	**0.52**	**19.87**	**0.20**

Table 10:- Seasonal Variation Risk Quotient (Negligible Concentration (RQNCs) and Maximum Permissible Concentration

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	Market		Semi- industrial		Residential	
PAH	**Rainy**	**Dry**	**Rainy**	**Dry**	**Rainy**	**Dry**
Nap	0.01	0	0.01	0	0.00	0
Acy	0.01	0	0.02	0	0.01	0
Ace	0.05	0	0.01	0	0.00	0.01
Flr	0.03	0	0.03	0.03	0.01	0.01
Phe	0.02	0.023	0.07	0.03	0.01	0.00
Ant	0.09	0.052	0.70	0.21	0.06	0.03
Flu	0.02	0.012	0.10	0.02	0.01	0.01
Pyr	0.03	0.137	0.01	0.03	0.01	0.00
BaA	1.10	7.74	1.28	2.44	2.84	0.28
Chr	4.20	0.932	4.03	0.18	10.71	0.02
Sum	**5.56**	**8.89**	**6.25**	**2.93**	**13.66**	**0.35**

Table 11:- Seasonal Variation Toxicity Equivalency Quotient (TEQ) Values of PAHs using [8]

			Adult				Children		
Location	**Average**	**Ingestion**	**Dermal**	**Inhalation**	**Total ILCR**	**Ingestion**	**Dermal**	**Inhalation**	**Total ILCR**
1	133.69	2.36E-04	4.60E-05	2.29E-14	**2.82E-04**	3.30E-03	4.92E-05	9.48E-15	**3.35E-03**
2	109.98	1.94E-04	3.79E-05	1.88E-14	**2.32E-04**	2.71E-03	4.05E-05	7.80E-15	**2.75E-03**
3	134.84	2.38E-04	4.64E-05	2.31E-14	**2.84E-04**	3.33E-03	4.97E-05	9.56E-15	**3.38E-03**
4	81.23	1.43E-04	2.80E-05	1.39E-14	**1.71E-04**	2.00E-03	2.99E-05	5.76E-15	**2.03E-03**
5	111.17	1.96E-04	3.82E-05	1.90E-14	**2.34E-04**	2.74E-03	4.09E-05	7.87E-15	**2.78E-03**
6	105.08	1.85E-04	3.62E-05	1.80E-14	**2.21E-04**	2.59E-03	3.87E-05	7.45E-15	**2.63E-03**
7	109.22	1.93E-04	3.76E-05	1.87E-14	**2.31E-04**	2.70E-03	4.02E-05	7.74E-15	**2.74E-03**
8	63.65	1.12E-04	2.19E-05	1.09E-14	**1.34E-04**	1.57E-03	2.34E-05	4.51E-15	**1.59E-03**
9	79.04	1.40E-04	2.73E-05	1.36E-14	**1.67E-04**	1.96E-03	2.93E-05	5.63E-15	**1.99E-03**
10	108.28	1.91E-04	3.72E-05	1.85E-14	**2.28E-04**	2.67E-03	3.98E-05	7.67E-15	**2.71E-03**
11	139.82	2.46E-04	4.81E-05	2.39E-14	**2.94E-04**	3.45E-03	5.15E-05	9.91E-15	**3.50E-03**
12	119.91	2.11E-04	4.13E-05	2.05E-14	**2.52E-04**	2.96E-03	4.41E-05	8.50E-15	**3.00E-03**
13	134.18	2.37E-04	4.62E-05	2.30E-14	**2.83E-04**	3.32E-03	4.95E-05	9.53E-15	**3.37E-03**
14	330.35	5.83E-04	1.14E-04	5.65E-14	**6.97E-04**	8.15E-03	1.22E-04	2.34E-14	**8.27E-03**
15	113.93	2.01E-04	3.92E-05	1.95E-14	**2.40E-04**	2.81E-03	4.20E-05	8.08E-15	**2.85E-03**
Sum	**1874.37**	**3.31E-03**	**6.46E-04**	**3.21E-13**	**3.95E-03**	**4.63E-02**	**6.91E-04**	**1.33E-13**	**4.70E-02**

Table 12:- RAINY Season Incremental Lifetime Cancer Risk (ILCR) Values

			Adult				Children		
Location	**Average**	**Ingestion**	**Dermal**	**Inhalation**	**Total ILCR**	**Ingestion**	**Dermal**	**Inhalation**	**Total ILCR**
1	19.93	3.51E-05	6.87E-05	3.41E-15	**1.04E-04**	4.91E-05	7.35E-05	1.41E-15	**1.23E-04**
2	52.00	9.17E-05	1.79E-04	8.89E-15	**2.71E-04**	1.28E-04	1.92E-04	3.69E-15	**3.20E-04**
3	99.79	1.76E-04	3.44E-04	1.71E-14	**5.20E-04**	2.46E-04	3.68E-04	7.08E-15	**6.14E-04**
4	134.93	2.38E-04	4.65E-04	2.31E-14	**7.03E-04**	3.32E-04	4.98E-04	9.57E-15	**8.30E-04**
5	130.86	2.31E-04	4.51E-04	2.24E-14	**6.82E-04**	3.22E-04	4.83E-04	9.28E-15	**8.05E-04**
6	31.50	5.55E-05	1.09E-04	5.39E-15	**1.65E-04**	7.76E-05	1.16E-04	2.23E-15	**1.94E-04**
7	7.79	1.37E-05	2.69E-05	1.33E-15	**4.06E-05**	1.92E-05	2.87E-05	5.52E-16	**4.79E-05**
8	20.93	3.69E-05	7.22E-05	3.58E-15	**1.09E-04**	5.16E-05	7.72E-05	1.48E-15	**1.29E-04**
9	2.07	3.65E-06	7.14E-06	3.54E-16	**1.08E-05**	5.10E-06	7.64E-06	1.47E-16	**1.27E-05**
10	55.93	9.86E-05	1.93E-04	9.56E-15	**2.92E-04**	1.38E-04	2.06E-04	3.97E-15	**3.44E-04**
11	3.21	5.67E-06	1.11E-05	5.49E-16	**1.68E-05**	7.92E-06	1.18E-05	2.28E-16	**1.97E-05**
12	0.00	0.00E+00	0.00E+00	0.00E+00	**0.00E+00**	0.00E+00	0.00E+00	0.00E+00	**0.00E+00**
13	2.29	4.03E-06	7.90E-06	3.92E-16	**1.19E-05**	5.63E-06	8.45E-06	1.62E-16	**1.41E-05**
14	9.21	1.62E-05	3.18E-05	1.57E-15	**4.80E-05**	2.27E-05	3.40E-05	6.53E-16	**5.67E-05**
15	1.86	3.27E-06	6.41E-06	3.18E-16	**9.68E-06**	4.57E-06	6.86E-06	1.32E-16	**1.14E-05**
Sum	572.30	1.01E-03	1.97E-03	9.79E-14	**2.98E-03**	1.41E-03	2.11E-03	4.06E-14	**3.52E-03**

Table 13:- DRY Season Incremental lifetime Cancer Risk (ILCR) Values

IV. DISCUSSION

The seasonal variation of pyrogenic index (PI) and Total index (TI) using [10] formulae is shown on Table 9. The LMW/HMWPAHs ratios (PI values) for all the sampling sites were <1. The Pyrogenic index (PI) values in this study for rainy and dry season for Markets and semi-industrial dumpsites, which ranged between 0.004-0.5 indicated a pyrogenic origin of the PAHs. This is agreeing with the work of [15] and [3]. However, the PI for the residential dumpsites during dry season is 1.065 and this indicated petrogenic source. It is due to incomplete combustion of some hydrocarbon containers like aerosol been dumped in the residential dumpsites.

The seasonal variations for the RQMPCs and RQNCs are shown on Table. 10 and the seasonal variation for TEQs are shown on Table. 11. In Rainy season, the RQ(MPCs) value for ∑PAHs for Market and Semi-industrial dumpsites are less than 1(<1) and also the RQ(NCs) are greater than 1 but less than 800ug/kg. From Table 2, using [18][2], it signified that the PAHs at these sites posed a moderate level of contermination or medium Ecological risk. While the RQ(MPCs) and RQNCs values for the PAHs for residencial dumpsites indicated low ecological risk. In Dry season the RQ(MPCs) and RQ(NCs) values for ∑PAHs for Market only indicated medium ecological risk while the semi- industrial and residencial dumpsites indicated low ecological risk. Sumarily the PAHs at the market dumpsites indicated moderate ecological risk both in rainy and dry season. This is due to the fact the dumpsite has high molecular weight PAHs in both season.

The respective TEQ(Toxicity Equivalency Quotient) value of a PAH is a measure of its ability to modify human DNA in such a way that it can result to cancer[8] . TEQ does not have threshold however, the PAH that has the highest TEQ value at a perticular site is termed as highest contributor to cancernogenic ability of PAHs at that site.In Rainy season (Table 11), it is obeserved that Chrysene and Benz(a)anthracene has the highest TEQ at all the sites while for Dry season, only Benz(a)anthracene has the highest. This means that the presence of these two PAHs at these dumpsites generated an harmful impact human health for the people living around these dumpsites.

The incremental lifetime cancer risk (ILCR) values calculated using formula of [17][19][9], ILCR < 10^{-6} indicates negligible potential cancer and human health risks, while values from 10^{-6} to 10^{-4} indicate low cancer risk. Thus, the obtained cancer risk values for adults in Rainy Season, revealed that the PAHs can bring a Low carcinogenic impact through ingestion and inhalation, whereas the ILCR dermal (1.36×10^{-14}–5.65×10^{-14}) indicates negligible potential cancer risk. In the same Rainy season, for children, the ILCR ingestion range (1.93×10^{-3}–8.15×10^{-3})and ILCR dermal range (2.93×10^{-5}–1.22×10^{-4})values for children indicated moderate cancer risk, respectively while range (4.51×10^{-15}–2.34×10^{-14}) ILCR inhalation indicated negligible potential cancer risk.

In Dry Season, ILCR values for adults ranged as 3.2×10^{-6} –4.6×10^{-4}, 6.4×10^{-6} –4.5×10^{-4}, for ILCR ingestion, ILCR dermal, , respectively indicated Low Cancer risk while 3.9×10^{-16}–2.2×10^{-14} for ILCR inhalation indicated negligible potential cancer risk. For children were ranged from 4.5×10^{-6}–3.3×10^{-4}, 6.8×10^{-6}–4.9×10^{-4}, for ingestion, dermal respectively indicated Low cancer risk while 1.32×10^{-16}–9.57×10^{-15} inhalation, indicated negligible cancer risk.

V. CONCLUSIONS

The potencial source of PAHs at all the dumpsites in Rainy season is Pyrogenic. In Dry season the PAHs in Markets and Semi- Industrial has pyrogenic origin while that of Residential and control is likely Petrogenic Source which may be due to some hydrocarbon containers like aerosol been dumped at dumpsites.

The PAHs at each dumpsite poses a moderate level of ecological risk since the RQNCs(Risk Quotient Negligible concentration values for the PAH and the RQMPCs (Risk Quotient Maximum Permissible Concentration) of individual PAHs were all less than 1.0 and are all less than the threshold ecological risk values of 0.8mg/kg.[19] [2],

According to USEPA, the PAHs present at each of the dumpsite poses risk of Cancer since the total ILCR (incremental Life Cancer Risk) values of each PAHs on most sites are higher than 1 $x10^{-6}$.

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