Polycyclic Aromatic Hydrocarbons Concentrations in Soils from Power Plant Stations in Universities in Port Harcourt, Rivers State, Niger Delta, Nigeria

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Soil contamination as a result of human interferences with nature is a common occurrence all over the world. Soil samples were collected from three universities (Ignatius Ajuru University of Education (IAUE), University of Port Harcourt (UNIPORT) and the Rivers State University (RSU) in Port Harcourt, Rivers State. The soils were taken at the points where the different institutions generate electricity with diesel powered generators. The soil samples were collected from the surface to a dept of 30 cm and were put into tightly closed glass containers. They were transported to the laboratory and prepared by standard methods of extraction and purification. The extracts were analyzed with a gas chromatography – mass spectrophotometry instrument to obtain the various fractions of PAHs present in the samples. It was observed that all the 16 priorities PAHs were present in all the samples from the station. The total concentrations of PAHs showed that IAUE>UNIPORT>RSU with respective values of 18.18, 16.64 and 16.53 mg/Kg. Benz(a)anthracene was observed to be the most abundant PAH in all the stations with a value of 2.11±0.14, 2.01±0.51 and 2.10±0.57 mg/Kg for IAUE<UNIPORT and RSU respectively. Fingerprinting and source diagnosis identified only one source of PAHs being pyrogenic sources of PAHs in the three stations. Ring size analysis showed the order 2-3>4>5>6 rings. The concentrations of carcinogenic and non-carcinogenic PAHs were almost in equal proportions in the soils examined. Therefore, the workers involved directly with the electrical generation on these campuses should adequately protected and informed on the consequences of contact with the soil and the gases that come out of the exhaust.

Keywords: *polycyclic aromatic hydrocarbons, soil, fingerprinting, power generating stations, pollution, human influence*

Introduction

The evolution of industrial revolution and subsequent industrialization and mechanization of different human activities has led to serious negative implications in the environment. These activities have resulted in the contamination of different

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sites which might be within the immediate vicinity or far away from the main point of the activity. Besides, when these manufacturing sites are no more in use, the contaminants and their effects (associated health conditions) are not adequately removed from the environment so contaminate (Guarino et al. 2019).

Soil being contaminated by chemicals of organic and inorganic contaminants is a fast-rising issue, which is a consequence of increased industrial development, exhaustive agriculture, and general application of xenobiotics into the natural environment (Harms et al. 2011). Contact to these contaminants positions humans and the ecology at very high risk and health imbalance (Singh et al. 2009). The pollution of the surface soil is a well-recognized and grave problematic issue all around the globe. It has been identified to negatively affect food chain and also portend great danger to the already scarce water resources, being a source of its contamination. The contamination of surface soil with tenacious organic chemicals is of utmost concern because of the fact that when they are present in the environment, it takes a very long time before they are abated (Roskam and Comans 2009).

Polycyclic aromatic hydrocarbons (PAHs) are one amongst the different tenacious organic based compounds that has been given serious consideration presently, because of the health challenges and implications associated with this group of compounds (Cai et al. 2017). PAHs are resistant to biological degradation and highly hydrophobic and are composed of fused hydrocarbons of 2-6 benzene rings (Ellenhorn and Barceloux 1988, Ekpete et al. 2019). Their sources of origin are either natural are anthropogenic. In most cases, human influenced sources of PAHs are the major contributors of PAHs in an environmental media (Wick et al. 2011, Edori and Iyama 2019). PAHs are very common in the environment (Zhang et al. 2017). Majority of PAHs found in the environmental are produced from the partial burning or pyrolysis of vestige fuels (Holbrook 1990). Exposure routes of humans to PAHs may be oral (food and water), dermal (skin) and inhalation (air) (EFSA 2008).

In Nigeria, like many other developing countries, electricity to power industrial and research activities are irregular. Therefore, power stations are built and are powered by heavy duty diesel generators. The way of filling the fuel tank and discharging of fuel from the tank is a cause for concern. The soil environment within the immediate vicinity of the generating are at the receiving end of the hydrocarbon waste discharged from the generating house. Pollution rising due to the discharge of the tank content, engine oil and petrol used during servicing and repair works has cause pollution of the immediate soil environment (Odjegba and Sadiq 2002, Edori et al. 2020). Therefore, there is the need to examine the contamination status of the immediate soil environment, using the concentration of polycyclic aromatic hydrocarbons in soil within the immediate vicinity of the power stations in three selected universities in Port Harcourt, Rivers State, Nigeria.

Materials and Methods

Samples were collected from the surface to a depth of 20 cm from soils within the area of heavy-duty generators used to power electricity in three universities in

Port Harcourt Rivers State, Nigeria. Soil samples were randomly collected with soil auger and pooled together to form representative sample for each of the sample points. The three universities where samples were collected were; The Rivers State University (RSU), Ignatius Ajuru University of Education (IAUE) and University of Port Harcourt (UNIPORT). The samples were immediately put into glass containers and well corked and then transported to the laboratory and refrigerated for further treatment.

The determination of the concentrations of PAHs in the soil samples, was achieved by weighing out 8 grams of soil into a Soxhlet extractor and 30 mL of acetone and n-hexane solvent in a 50:50 ratio. The temperature of the extraction system was put at 100°C for 1200 seconds. Then were allowed to cool to 25°C and the extracting solvent were removed and put into an inverted sample vessel. The extraction container was washed three times with n-hexane and thereafter removed to the inverted sample vessel to avoid loss of PAHs present in the original sample. Subsequently (after the extraction), each of the samples were cleansed in the chromatographic column, which contain 10 cm long activated silica gel, 2 cm of anhydrous Na_2SO_4, which compacted with a cotton absorbent. Thereafter the solvent used for the extraction was added to the chromatographic column and eluted using 30 mL of dichloromethane and 30 mL of n-hexane to separate out the PAHs. The solvents were volatilized and the sample concentrated to between 2.0-0.3 mL through a rotary evaporator. The final volume obtain was stored frozen pending time for analysis.

The eluted PAHs were analyzed by a gas chromatographic instrument model Agilent 6890 gas chromatograph (GC) furnished with a tube support model HP-5 of length 30 m, whose inner radius is 0.125-mm inner and the film is 0.25 m thick, manufactured by J & W Scientific Inc., Folsom, CA, USA) and a 5975C mass selective detector (MS). The carrier gas used in the gas chromatographic process was helium, which was inserted at a rate of 1 mL/min with care to avoid splitting. The operating temperature was set at 60°C and maintained for 60 seconds and thereafter increased to 110°C at 20 C/min. Then, the temperature was increased to 290°C at an increment rate of 4°C/minutes and allowed to stand for 1200 seconds. The temperature of the detector was set at 290°C. The individual components were identified by ion selection mode, based on the scan and retention time on the chromatogram. The quantification and confirmation of the ions were read from the chromatogram chart from the mass detector.

The sources and origin of the PAHs were examined using different diagnostic ratios and the sources were identified on the basis of the value obtained for that particular ratio. The ratios used were: (a) low molecular weight/high molecular weight (LMW/HMW), for source identification in this, if the ratio gives a value >1, the probable source is petrogenic, while a value <1 is pyrogenic. (b) anthracene/ anthracene+phenanthrene (An/(Ant+Phe), for source classification and identification, when the ratio value is <0.1, it is petrogenic, but > when >0.1, it indicates pyrogenic source (c) Fluoranthene/Fluoranthene+pyrene (Fl/(Fl+Pyr)), for this grouping, if the value of the ratio is >0.5, then pyrogenic source is suggested, but if the ratio <0.4, then petrogenic source is inferred. (d) benzo (a) anthracene/benzo(a)anthracene+chrysene (BaA/(BaA+Chr)), for the identification

of source and origin in this class, when the ratio <0.2, petrogenic source is inferred, but when >0.35, pyrogenic source is suggested and (e) Indeno(1,2,3-ed) pyrene/ Indeno(1,2,3-ed) pyrene/Benzo (g,h,i) perylene (Ip/(I+Bgp)), when the value of the ratio <0.2, it is proposed petrogenic, values ranging from 0.2-0.5 is proposed to be mixed petrogenic and pyrogenic sources and values >0.5 are termed pyrogenic (Tolosa et al. 2004).

Results and Discussion

Concentrations of PAHs in the Power Generating Stations

The results of polycyclic aromatic hydrocarbons (PAHs) concentrations in the soil samples from the power generating stations are shown in Table 1. The results showed that the total concentrations of PAHs in the selected stations was 18.18, 16.64 and 16.53 mg/Kg at IAUE, UNIPORT and RSU respectively. It was also observed that the most abundant PAH in all the examined stations was Benz (a) anthracene, which was 2.11±0.14, 2.01±0.51 and 2.10±0.57 mg/Kg at IAUE, UNIPORT and RSU respectively, which was followed by the observed values of Dibenz (a,h) anthracene, anthracene, chrysene, Benzo(k) fluoranthene, phenanthrene , naphthalene etc. The least abundant PAH in all the stations was O-terphenyl.

The total concentrations of PAHs observed in the stations in the present is lower than the values observed in soils in Lagos, Nigeria (Fatunsin et al. 2019), higher than the values of PAHs observed in urban and natural forest soils in the Atlantic Forest, (São Paulo State), Brazil (Bourotte et al. 2009) and also lower than the total values of PAHs observed in contaminated sites of Hisar, India (Bishnoi et al. 2009). However, the values observed in the various university stations were higher than the 10 mg/Kg for recreational land use recommended by the Italian legislation (Guarino et al. 2019). The observed concentrations of PAHs in soils of the different stations investigated may be related to the equilibrium amongst contributing and discharge sources and the length of time that the generation has existed (Holoubek et al. 2009). From the continuous discharge of generator emissions in the three universities, it will be expected that the concentrations of PAHs in the soils within the immediate vicinity will be higher than observed, but it was not so. The reason may not be far-fetched from the observation of Nam et al. (2008), that concentrations of PAHs in soils are connected majorly with the nature of the emissions, atmospheric transport and land cover. Looking at the different levels of concentrations of the PAHs in soils in each of the universities, the order is IAUE>Uniport>RSU. Taking into consideration, IAUE generates more electricity, than all the other universities, while RSU gets electricity from the national supply line more than the others, which probably may have emitted less of hydrocarbons than the others.

Table 1. *Concentration (mg/Kg) of Polycyclic Aromatic Hydrocarbon (PAHs) in Soil from Power Generating Stations in the Universities*

Concentrations of PAHs (mg/Kg)	University Location		
	IAUE	UNIPORT	RSU
Naphthalene	1.10±0.12	1.09±0.01	1.00±0.00
Acenaphthylene	0.55±0.02	0.40±0.01	0.42±0.01
Acenaphthene	0.28±0.00	0.25±0.01	0.20±0.00
Fluorene	0.68±0.03	0.66±0.00	0.67±0.19
Phenanthrene	1.50±0.10	1.39±0.13	1.36±0.58
Anthracene	1.81±0.04	1.75±0.25	1.72±0.07
O-Terphenyl	0.05±0.00	0.03±0.00	0.01±0.00
Fluoranthene	0.70±0.01	0.61±0.03	0.58±0.02
Pyrene	0.56±0.02	0.54±0.02	0.50±0.00
Chrysene	1.75±0.11	1.74±0.16	1.74±0.14
Benz(a) anthracene	2.11±0.14	2.01±0.51	2.10±0.57
Benzo(b) fluoranthene	0.65±0.00	0.65±0.03	0.68±0.06
Benzo(k) fluoranthene	1.86±0.21	1.54±0.20	1.46±0.00
Benzo(a) pyrene	0.80±0.09	0.67±0.02	0.60±0.00
Dibenz (a,h) anthracene	2.01±0.31	1.60±0.14	1.47±0.16
Benzo(g,h,i) perylene	0.81±0.00	0.81±0.06	1.10±0.14
Indeno (1,2,3-cd) pyrene	0.96±0.06	0.90±0.02	0.92±0.05
Total	18.18	16.64	16.53

PAHs do occur in soils in a diverse fraction which are related to appropriation and maturing developments that increases with time (Harmsen 2007). Also, owing to the excessive sorption and ability of the soil to retain the absorbed PAHs. Thus, coarse soils have low capacity to retain pollutants which are easily leached to inner depth of the soil or quickly washed away.

The contamination of the environment with PAHs have different health effects on both humans and the environment. The effects depend on the degree of contact, dosage, distinctive toxicity characteristics and the exposure pathway (Ekpete et al. 2019). Although, the effects might not be noticed on the workers now, both the chances of being affected later may be there. On the other hand, the present health condition and the age of the workers in this unit can enhance or promote the effects of PAHs on them (Adedosu et al. 2015, Edori and Iyama 2019). In other words, taking into consideration the levels of PAHs in the soil, not considering the amount inhaled during the period at work, the artisans are at a risk of acute and chronic effects due to exposure (ATSDR 1995). The most notable and considered effects of PAHs is related to their carcinogenicity and teratogenicity, inducing of dioxin-like activity and weak estrogenic response (Villeneuve et al. 2002, Nwineewii and Marcus 2015).

The PAHs observed from the sites examined in this research majorly originated from the partial burning of diesel, spills during refueling of the generator, precipitation of discharged fumes by rain, engine oil and petrol used during maintenance and servicing of the generators. This observation is in agreement with an earlier one by Korosi et al. (2013) who observed that PAHs in soils originated majorly from fume waste and dumps within that particular point. When PAHs is

present in soil, it possesses the ability to contaminate ground water through percolation down the soil profile and also surface water through running water after heavy down pour of rain (Al-Delaimy et al. 2014). The consequences of incidence of PAHs in water to living organisms is an issue that demands utmost concern because the direct and indirect effects either through individual personal contact of through food chain transmission (Ifemeje et al. 2014).

Fingerprinting and Diagnosis of PAHs from the Power Stations

The results of the different analytical ratios and sources of the PAHs is shown in Table 2. Different tools have been used to judge the origin of Polyaromatic hydrocarbons in environmental media by different authors (Jiao et al. 2017, Zeng et al. 2019, Ekpete et al. 2019, Edori et al. 2020). The outcome is used to apportion the supposed causes and the particular origin of the source type, whether natural or anthropogenic. The ratios of LMW/HMW in the different sites were 0.48, 0.5 and 0.48 at IAUE, UNIPORT and RSU respectively. All the values observed in the examined stations were less than 1, which signifies that the PAHs origin were of pyrogenic or combustion sources. This observation is in consonance with the observation of Jiao et al. (2017), where they observed that concentrations of the ratio of LMW/HMW revealed pyrogenic sources of PAHs in soils around a chemical plant in Shanxi, China. However, this observation is at variance with the observation of Ekpete et al. (2019), that observed values of the ratio LMW/HMW higher greater than 1 at dumpsites situated at psychiatric Road and Rumuokwuta in Obio/Akpor Local Government Area, Port Harcourt, Rivers State, Nigeria, which was ascribed to petrogenic sources of PAHs.

The ratio of An/(Ant+Phe) in the stations showed that in IAUE, UNIPORT and RSU were 0.55, 0.56 and 0.56 respectively. All the ratio values from the stations were >0.1, which suggested pyrogenic sources of PAHs. This observation corroborates the findings of Jiao et al. (2017), where they observed that PAHs sources from a chemical plant sited in Shanxi, China, originated from combustion of organic materials such as coal, wood, grass and petroleum. Also, Cao et al. (2019), while investigating the concentrations of PAHs in reconstructed land from a coking plant, Beijing, China, observed that finger printing analysis for source identification of PAHs using An/(Ant+Phe) gave values >0.1 and concluded that the PAHs present in the top soil in the coking area were principally from incineration that resulted from the burning of coal and biomass burning, but little contribution came from petroleum source.

The ratio of Fl/(Fl+Pyr) in the stations showed that in IAUE, UNIPORT and RSU were 0.56, 0.53 and 0.54 respectively. All the ratio values from the stations were >0.50, which suggested that pyrolytic sources were responsible for the observed PAHs. The observations from this work is in agreement with those of Ekpete et al. (2019) in selected dumpsites within Obio/Akpor local Government Area, Rivers State, whose Fl/(Fl+Pyr) values were all greater than 0.5. According to Yunker et al. (2002), when ratios of Fl/(Fl+Pyr) falls within the value range of 0.40-0.50, the PAHs origin are chiefly a consequence of petroleum combustion,

but less than 0.40 is an indication that the sources are from oil origin, while values that are >0.50 are alluded to the burning of coal and biomass sources.

The ratio of BaA/(BaA + Chr in the stations showed that in IAUE, UNIPORT and RSU were 0.55, 0.54 and 0.55 respectively. All the ratio values were >0.35, which a consequence of pyrogenic sources (Salih et al. 2015). Cao et al. (2019), observed that the classifications of the origin of PAHs based on BaA/(BaA + Chr) ratio are; values between 0.2-0.35 is due to petroleum combustion, if it is less than 0.2, then the source is from lower petroleum fractions, but greater than 0.35values is due to biomass combustion from grass, wood and coal.

The ratio of Ip/(Ip+Bgp) in the stations showed that in IAUE, UNIPORT and RSU were 0.54, 0.53 and 0.46 respectively. Guarino et al. (2019), suggested that Ip/(Ip+Bgp) ratio above 0.50 is a result of biomass and coal incineration. Whereas, pyrolytic products that originated from gas, petrol, paraffin, diesel and crude oil produces ratios that are less than 0.50 and discharges from automobiles fall within 0.24 and 0.40.

**Table 2.** *Diagnostic Ratios and Source Apportionment of PAHs in Power Stations*

Power Generating Stations	LMW	HMW	LMW/ HMW	An/(Ant+Phe)	Fl/(Fl+Pyr)	BaA/(BaA+Chr)	Ip/(Ip+ Bgp)
IAUE	5.92	12.26	0.48	0.55	0.56	0.55	0.54
UNIPORT	5.54	11.10	0.5	0.56	0.53	0.54	0.53
RSU	5.37	11.16	0.48	0.56	0.54	0.55	0.46

Ring Size Analysis of PAHs in the Power Generating Stations

The concentrations of the PAHs ring types in the various power generating stations is shown in Figure 1. The results from the stations revealed that in IAUE, the values were 5.92, 5.17, 5.32 and 1.77 mg/Kg for 2-3 rings, 4, 5 and 6 rings respectively. At the UNIPORT station, the values were 5.54, 4.93, 4.46 and 1.71 mg/Kg for 2-3 rings, 4, 5 and 6 rings respectively and at the RSU station, the values were 5.37, 4.93, 4.21 and 2.02 mg/Kg for 2-3 rings, 4, 5 and 6 rings respectively. The stations values showed the predominance of 2-3 membered rings in all cases, which was followed by the 4-membered rings, then the five membered rings and the least was the six membered rings. The high levels of the LMW PAHs is due to the continuous use of generators to supply electricity to the various units of the universities' establishments. Highly contaminated soil with LMW PAHs as was observed in the present work is an indication of recent deposition. This because they are highly hydrophobic, hydrophilic, volatile and soluble, therefore can easily be removed under slight environmental changes (Wu et al. 2019, Edori et al. 2020). Despite the volatility of the LMW PAHs yet they were still present in very high proportions. The HMW PAHs were probably formed due to combination reactions of the LMW PAHs under very high temperature. They are not easily biodegradable nor volatile, so the fairly high proportions of 4 and five membered rings in the soils might be a result of deposition or accumulation over a long time, putting into considerations the length of time generators have been used to supply energy needs of the universities. It is and established fact that LMW PAHs are primarily formed from petroleum contamination, whereas HMW PAHs are chiefly

due to partial pyrolysis of coal at high temperatures (Li and Duan 2015). This is the case with the observations made in the present work, where the presence of the PAHs is linked to the use of diesel-powered generators (heavy duty generators), which operate at very high temperatures. Further to this is the nature of discharge of spent oil by the staff and the nature of solvents used during servicing, which is gasoline.

Figure 1. *Ring Size Analysis of PAHs in the Power Generating Stations*
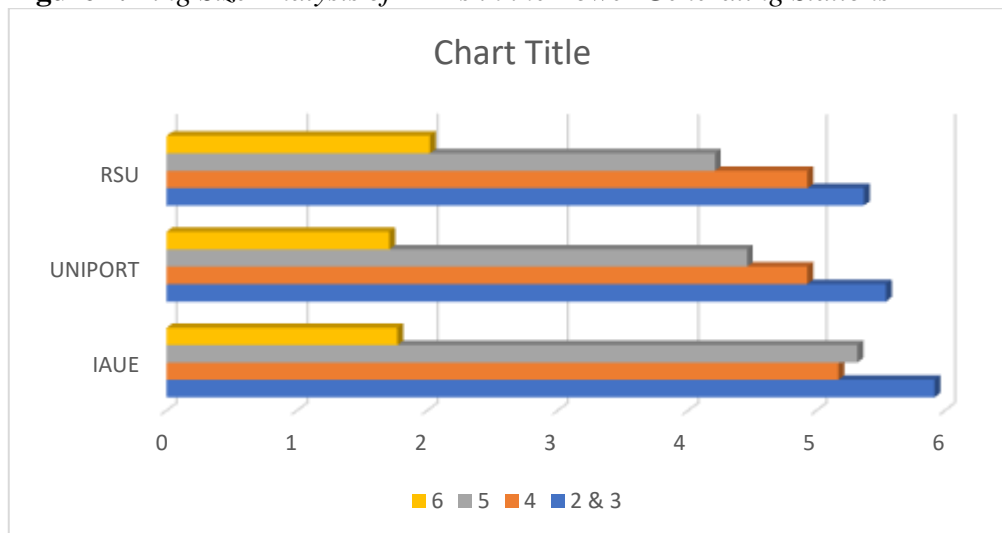

Carcinogenic and Non-Carcinogenic PAHs in Soils from Power Generating Stations

   The concentrations of the carcinogenic and non-carcinogenic are shown in Figure 2. The assessment of the values of non-carcinogenic and carcinogenic PAHs in the examined stations revealed that at IAUE the values were 8.98 mg/Kg and 9.20 mg/Kg for non-carcinogenic and carcinogenic PAHs respectively. At UNIPORT, the respective values for non-carcinogenic and carcinogenic PAHs were 8.46 and 8.18 mg/Kg, while that of RSU were 8.20 and 8.33 mg/Kg for non-carcinogenic and carcinogenic PAHs. The values of carcinogenic PAHs were slightly higher than the non-carcinogenic PAHs in soils from IAUE and UNIPORT, but slightly lower in soil from RSU. Highest values for both medically classified categories were observed at the IAUE station. This means that staff from the IAUE institution are more likely predisposed to health effects that might be associated with the work when compared to the other institutions. Although all the examined sixteen priority PAHs have negative health consequences on man, yet, six PAHs have been identified as having constituted highest risk when contacted for a very long period. All in this category fall within four to six membered rings. These six PAHs are namely benzo(a)anthracene, chrysene, benzo(b)fluoranthrene and benzo(k) fluoranthrene are known to be carcinogenic in nature (Obini et al. 2013). The implication of the high values of these PAHs on the workers is exposure to cancer causing agents which have a negative toll on them after protracted

exposure through contact with the soil (ATSDR 1999) and inhalation of gases which are released from the generators exhaust.

Figure 2. *Concentrations (mg/Kg) of Carcinogenic and Non-Carcinogenic PAHs in the Power Generating Stations*
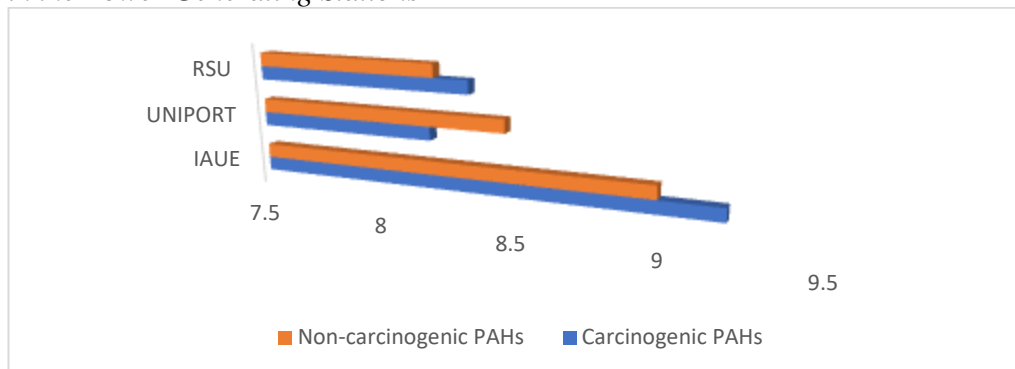
The nature of toxicity of PAHs are categorized and the basis of benzo[a] pyrene because its toxicological profile and cancer-causing characteristics have been well understood (Lee and Shim 2007). The carcinogenicity and mutagenicity of benzo[a]pyrene is mostly pronounced when it is converted to its metabolites at very high temperatures ranging from 300-600°C (Aygün and Kabadayi 2005).

Conclusions

Due to the educational development and the energy requirements of universities, power stations were built to meet up with such requirements seeing the failure of the national grid to supply electricity. This has resulted in some sort of environmental pollution of the soils within the immediate environment. This study is on the level of pollution and the origin of PAHs in the soil surface of power stations in three universities in Port Harcourt Rivers State, Nigeria. The total concentrations of 16 priority PAHs in the stations ranged from 16.53-18.18 mg/Kg. The profiling of the different ring types showed that the low-ring aromatics (2-3 rings) were most dominant, which was followed by the 4 membered rings, then 5 membered rings and the least was 6 membered rings, which indicated that fuel combustion was the major sources of PAHs. The finger printing and diagnosis of the origin of the PAHs indicated anthropogenic pyrolytic sources in all the stations, which assessment of the total carcinogenic and non-carcinogenic PAHs revealed that almost equal proportions of the two types, although carcinogenic PAHs dominated at the IAUE and RSU stations. The level of PAHs in the soils examined is alarming and therefore caution should be taken to avoid further increase so as to stem any emergence of health effects in the near future.

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