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Investigation of Presence and Levels of Polycyclic Aromatic Hydrocarbons around An Asphalt Plant in Imo State, Nigeria

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Abstract:- This study was carried out to investigate the presence and concentrations of PAHs around an asphalt plant in Owerri, South East Nigeria. Soil samples were collected from three different locations around the asphalt plant, while a control sample was collected some kilometres away from the asphalt plant. The samples were collected using a hand auger; after which they were put into a sterile polythene bag and transported to the laboratory for analysis. At the laboratory, the samples were oven dried and sieved to remove large particles. Edaphic variables were determined using approved Standard Operations Procedures (SOP), while PAHs were determined using gas chromatography interphased with Flame Ionization Detector (GC-FID. Narrow variations were noticed for benzo (a) anthracene (0.29 – 0.32 mg/kg) with a mean and standard error of 0.40 and 0.13 respectively. Maximum concentrations of 0.82mg/kg and 0.72mg/kg were recorded for benzo(a)anthracene and chrysene respectively at sampling location B (SLB). Nitrate, phosphate and sulphate had minimum concentrations of 28.90, 8.65 and 58.20mg/kg, respectively, with maximum concentrations of 31.00, 10.80 and 66.50mg/kg, respectively. At $p<0.05$, chrysene had a positive correlation with pH ($r=0.970$) and sand composition ($r=0.977$). Also, benzo(k)flouranthene, diabenzo(a,h)anthracene and benzo(g,h,i)perylene each correlated positively with loam component ($r=0.974$, $r= 0.973$and $r=0.970$), respectively. PAHs [benzo(a)anthracene, chrysene, benzo(b)flouranthene, benzo(k)flouranthene, benzo(a)pyrene, indeno-1,2,3-cd pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene] were identified in the soil samples, at a relatively high concentrations. The high concentration of PAHs present in the soil samples can be attributed to high organic carbon content of the soil, as well as low pH value, high sand composition and high moisture contents.

Keywords:- *Asphalt; Asphalt Plant; Concentration; Gas Chromatography; Edaphic variables; Polycyclic Aromatic Hydrocarbons (PAHs)*

I. INTRODUCTION

There is a growing demand for road expansion in Nigeria, and this has led to a huge increase in asphalt production. Asphalt is a dark fur-like substance used in surfacing roads or water proofing buildings. Hot mix Asphalt Facility, is an assembly of mechanical equipment, where aggregates (i.e. inert materials such as sand, gravel, crushed stones, slag, rock dust or powder) are blended, heated, dried and mixed with bitumen (a black sticky substance obtained naturally or from petroleum refining), also known as asphalt cement (EPA, 2000). Most of the pollutants emitted from Hot Mix Asphalt facility are; particulates (dust, smoke stone particles), sulphur dioxide (SO_2), oxides of carbon (CO and CO_2), oxides of Nitrogen (NO, N_2O and NO_2), Volatile organic compounds (VOCs), aliphatic hydrocarbons and Polycyclic Aromatic Hydrocarbons (PAHs) (EPA, 2000; WHO, 2000).

Polycyclic aromatic hydrocarbons (PAHs) which may be generated during asphalt operations are toxic to human health (NIOSH, 2001). They are a class of diverse organic compounds containing two or more fused aromatic rings made up of carbon and hydrogen atoms (Gu et al., 2016;). Generally, PAHs are produced from incomplete combustion of organic materials, fossil fuels, petroleum product spillage and various domestic and industrial activities (Johnson et al., 2005; Jiang et al., 2016 [2,3]. Once emitted, PAHs can be widely dispersed in air, water, soil and sediment. Due to the hydrophobicity and lipophilicity of PAHs, soil is the most important sink for PAHs in natural environment (Yin et al., 2008; Kaamal et al., 2015).

Due to PAHs documented mutagenic, carcinogenic and persistent properties, there is currently growing global concern about their emerging environmental and human health effects (US-DHHS, 1995, Zenzes, 2000; Luch, 2005; Gammon and Santela, 2006). To date, several studies have reported environmental PAHs concentrations and human health risk arising from occupational exposure. Similarly, there are several reported studies on PAHs concentrations in auto-mechanic workshops, road-side and urban soils (Dong & Lee, 2009; Emoyan, Agbaire & Akporido, 2015); However, there are no reported studies on the

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concentrations of PAHs in soils around an asphalt plant in the study area.

Researchers have confirmed that pollutants in the atmosphere finally settle on soil surfaces, and finally find their way into the deeper soils through leaching and percolation water (Van Jaarsveld, Van Pul & De Leeluwa, 1997; Maliszewska-Kordybach, 1999). The Asphalt plant is surrounded by farmlands, as inhabitants in the area are mainly agrarians. Staple foods such maize, yam, cassava and plantain are cultivated on the farmland. Research has also shown evidence plant uptake of PAHs through the roots (Li & Chen, 2008 and Collins & Finnegan, 2010). Therefore, the focus of this study was to determine the presence and levels of PAHs in soils around the asphalt plant in the study area.

II. MATERIALS AND METHODS

- **Study Design:** This is an experimental study that was conducted to investigate the presence and concentration of PAHs around as asphalt plant in Owerri. the study was conducted during the rainy season (between March-May, 2018). Soil samples around the asphalt plant were investigated against a control.

- **Description of Study Area:** This study was carried out in Umunebo Oforola, along port-Harcout Owerri road, in Owerri, Imo State, where New Idea Asphalt Plant is Located.

Owerri lies between Latitude $4^{0}15^{1}$ and $7^{0}05^{1}$ North and Longitude $5^{0}50^{1}$ and $9^{0}30^{1}$ East. The geology is dominated by deeply weathered coastal plain sands (Benin formation) of the Oligocene Miocene era. The watershed is a typical humid environment. The average precipitation in the area is 2500mm with three distinct months of dryness, while the annual temperature ranges from 29-29^{0}C. Soils of the study area are very sandy and acidic (Soil survey staff, 2003). Farming is the major socio economic activity of the area. The soil samples used for the study were gotten from the area around the asphalt plant in Umunebo Village while the control sample was taken from Umuelu, in Umuagwo Ohaji-Egbema L.G.A of Imo State.

- **Sample Collection Method:** Soil samples were collected using the random survey method, that is, the samples were collected at irregular positions but with a known distance from each other. Soil samples were taken from four different points. The first point of collection, known as SLA was collected 100meters from a plant. The second point of collection (SLB) was 100m away from the first point while the third sample (SLC) was taken 100m away from the second point of collection. The control sample was taken 8km away from the asphalt plant. The sample location, coordinate, elevation and distances are given in table 3.1 below. Within each of the sample location, three samples were randomly collected and were well mixed. Then a representative sample of that particular location was then collected. The depth of the soil at which the samples were taken is thirty centimeter (30cm).

- **Sample Handling:** The soil samples were put in a sterile polythene bag and tied very well. It was then put in a cooler to preserve its moisture content and other edaphic variables, as well as the PAHs concentration. The soil samples were then sent to the laboratory (Transcontinental Petrotech Nigeria Limited, Port Harcourt) for analysis.

- **Sample Preparation:** The procedures involved in soil sample preparation include:
  - **Air or Oven Drying**: freshly collected soil samples are often moist. The first step is to dry it as soon as possible, to ceases all biological activities/ transformation in the soil. The soil samples collected, were spread out on a flat sheet of paper and left on the oven for three days at low temperature to fully eliminate all moisture present.
  - **Sieving:** This involves the use of sieve to remove gravel, stone, plant debris, etc. from the soil sample. The sieve is usually 2mm and the materials that are able to pass through its mesh are called fine earth materials and are kept for laboratory analysis

- **Soil Edaphic Variables Determination:** Determination of edaphic variables such as Textural Classification, Nitrate, Soil pH, Phosphate, Moisture Content, Total Nitrogen (TN), Organic Carbon in Soil and Sulphate in Soil was done using approved Standard Operating Procedures (SOPs).

- **Determination of PAHs in Sample:** 10g of each soil sample was taken and 60ml of mixture was added. The mixture was warmed for 3mins at 30˚C in a water bath. The extract was decanted into clean dried round bottom flask and taken to the rotator evaporator to be concentrated to 3ml. this was fractionated by a fractionating column of silica gel into aromatic portion using 60ml dichromethane. The dichromethane fractionates were transferred to a rotator evaporator and reconcentrated to 3ml, transferred to 3ml Gas chromatography vials, corked and ultimately analyzed with Gas chromatography (G.C 6890 series).

- **Data Presentation and Analysis:** Results of the experiment were presented using descriptive statistics, while SPSS version 17.0 was used to analyze the data as was required.

III. RESULTS

- *Variations in the Concentration of Polycyclic Aromatic Hydrocarbons (PAHs) In Soil Samples*

Narrow variations were noticed in the concentration of PAHs measured in the soil samples. benzo (a) anthracene varied from 0.29 – 0.32 mg/kg with a mean and standard error of 0.40 and 0.13 respectively, while chrysene varied between 0.34 – 0.72 (0.54 ± 0.08) mg/kg in the soil samples (Table 1). Benzo(b)flouranthene varied from 0.16 – 0.42 (0.28 ± 0.008) mg/kg, while benzo(k)flouranthene varied from 0.10 – 0.96 (0.60 ± 0.21) mg/kg. also, benzo(a)pyrene varied between 0.18 and 0.85mg/kg with a mean and standard error of 0.36 and 0.16 respectively.

However, Indeno-1,2,3-cd pyrene varied between 0.09-0.26 (0.17±0.04) mg/kg, while dibenzo(a,h)anthracene and benzo(g,h,i) pyrene varied between 0.00-0.26(0.15±0.06)mg/kg and 0.00-0.19(0.11±0.04)mg/kg, respectively.

- *Spatial Variation in PAHs Concentration in Soil Sample*

There were observed numerical variations in the levels of PAHs in the soils sampled during the study (Appendix 2). Maximum concentrations of 0.82mg/kg and 0.72mg/kg were recorded for benzo(a)anthracene and chrysene respectively at sampling location B (SLB), whereas, minimum concentrations of 0.29mg/kg and 0.34mg/kg were recorded for them respectively at the control sampling location (SLcontrol) (fig. 1).

Benzo(b)flouranthene, benzo(k)flouranthene and benzo(a)pyrene recorded their highest concentrations of 0.42, 0.96 and 0.85mg/kg respectively at sampling locations B, A and C, respectively (fig. 2 and 3). However, at sampling location A (SLA), indeno-1,2,3-cd pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene recorded their maximum concentrations of 0.26, 0.26 and 0.19mg/kg respectively, with minimum concentrations of 0.088, 0.12 and 0.071mg/kg respectively, at the control sampling location (fig. 4).

A test of homogeneity in variance using the single factor ANOVA, revealed that, there was a significant spatial heterogeneity of the PAHs concentration in the soil samples [$F_{(202.02)} > F_{(3.92)}$] at $P < 0.05$ (Appendix 1).

A further structure detection of group means using the post-ANOVA means plot revealed that, between the control sampling location and sampling location A, benzo(k)flouranthene was most responsible for the observed spatial heterogeneity in PAHs concentrations (fig 5). Similarly, between the control sampling location and sampling location B(SLB), benzo(k)flouranthene, benzo(a)anthracene and chrysene were most responsible for the observed spatial heterogeneity in the PAHs (fig. 6). Also, between the control sampling location and sampling location C (SLC), benzo(a)pyrene and chrysene were responsible for the observed spatial heterogeneity in the PAHs (fig. 7).

- *Variations in Concentrations of the Edaphic Variables in Soil Samples*

There were wide variations in the concentrations of nitrate, phosphate, sulphate and textural class (sand, silt and clay) of the soils sampled during the study period, while narrow variations were observed in pH, moisture content, total nitrogen and total organic carbon concentrations (table 4.2). Nitrate, phosphate and sulphate had minimum concentrations of 28.90, 8.65 and 58.20mg/kg, respectively, with maximum concentrations of 31.00, 10.80 and 66.50mg/kg, respectively.

- *Relationship between PAHs and Edaphic Variables*

At $p<0.05$, chrysene had a positive correlation with pH (r=0.970) and sand composition (r=0.977) (table 3). Also, benzo(k)flouranthene, diabenzo(a,h)anthracene and benzo(g,h,i)perylene each correlated positively with loam component (r=0.974, r= 0.973and r=0.970), respectively.

However, at $p<0.01$, chrysene and dibenzo(a,h)anthracene had a negative correlation with sulphate (r= -0.972) and total nitrogen (r= -0.976), while benzo(g,h,i)perylene and benzo(k)flouranthene each correlated negatively with total nitrogen (r= -0.985 and r= -0.992), respectively.

PAHs (mg/kg)	Minimum Value	Maximum value	Range	Mean	Standard Error
Benzo(a)anthracene	0.29	0.82	0.53	0.48	0.13
Chrysene	0.34	0.72	0.38	0.54	0.08
Benzo(b)flouranthene	0.16	0.42	0.26	0.28	0.08
Benzo(k)flouranthene	0.10	0.96	0.86	0.60	0.21
Benzo(a)pyrene	0.18	0.83	0.67	0.36	0.16
indeno-1,2,3-cd pyrene	0.09	0.26	0.17	0.17	0.04
Dibenzo(a,h)anthracene	0.00	0.26	0.26	0.15	0.06
Benzo(g,h,i)perylene	0.00	0.19	0.19	0.11	0.04

Table 1:- Descriptive statistics of the PAHs in soil samples around the asphalt plant

Edaphic Variables	Minimum Value	Maximum Value	Range	Mean	Standard Error
Nitrate(mg/kg)	28.90	31.00	21.10	30.05	0.51
Phosphate (mg/kg)	8.65	10.80	2.15	10.02	0.51
Sulphate (mg/kg)	58.20	65.50	8.30	61.58	1.78
pH	5.00	5.66	0.60	5.33	0.13
Sand (%)	65.00	72.00	7.00	69.25	1.49
Clay (%)	18.00	30.00	12.00	22.50	2.63
Loam (%)	5.00	10.00	5.00	8.25	1.18
Moisture content (%)	2.00	3.00	1.00	2.50	0.29
Total Nitrogen	0.14	0.16	0.02	0.15	0.01
TOC	1.30	1.80	0.50	1.50	0.11

TOC= Total Organic Carbon

Table 2:- Descriptive statistics of the Edaphic variables of soil samples around the asphalt plant.

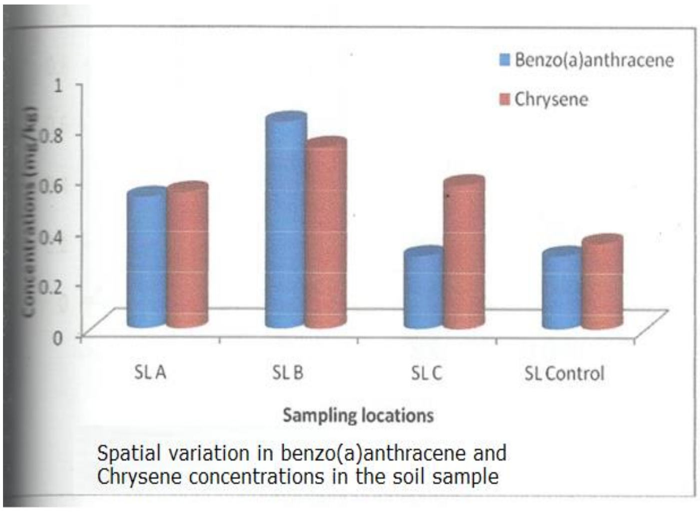

Fig 1:- Spatial variation in benzo(a)anthracene and chrysene concentrations in the soil sample

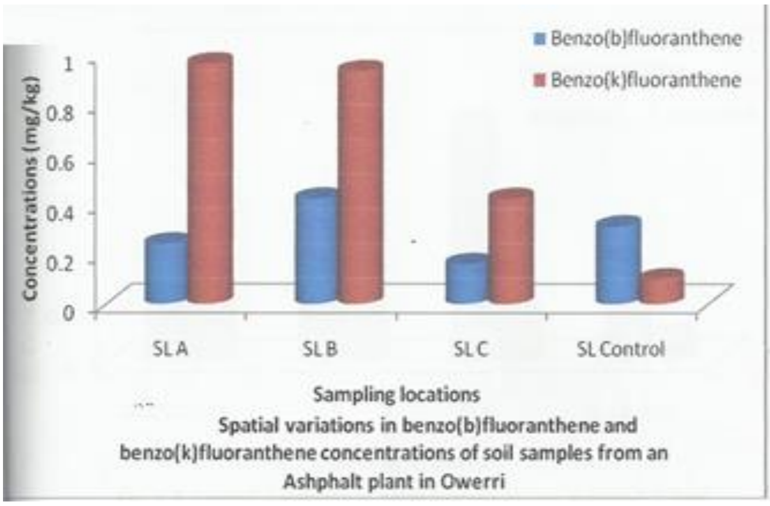

Fig 2:- Spatial variation in Benzo(b)flouranthene and benzo(k)flouranthene concentrations in the soil sample

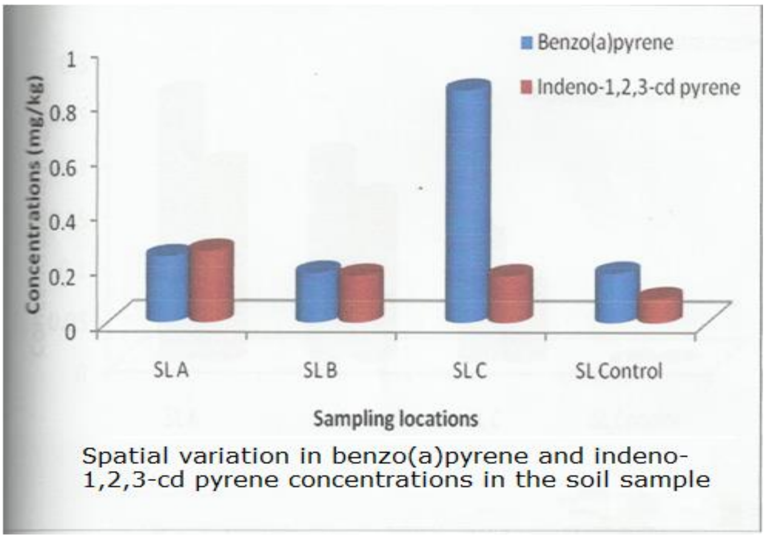

Fig 3:- Spatial variation in benzo(a)pyrene and indeno-1,2,3-cd pyrene concentrations in the soil sample

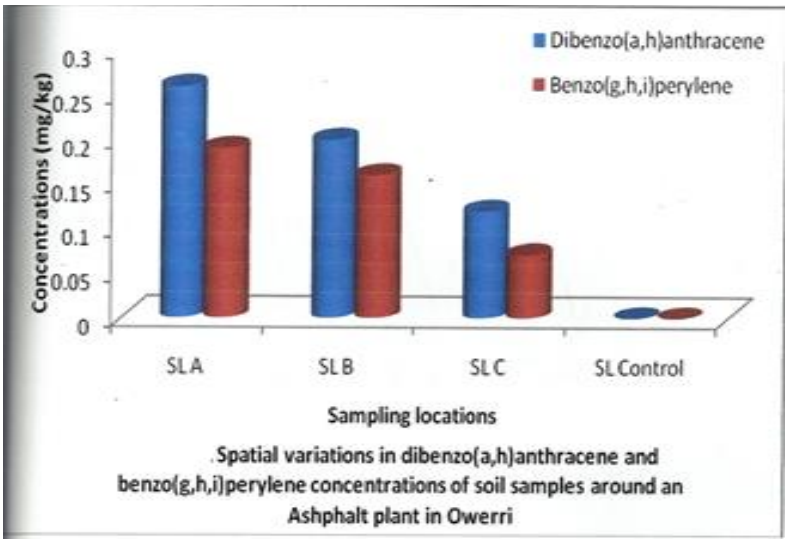

Fig 4:- Spatial variation in dibenzo(a,h)anthracene and benzo(g,h,i)perylene concentrations in the soil sample

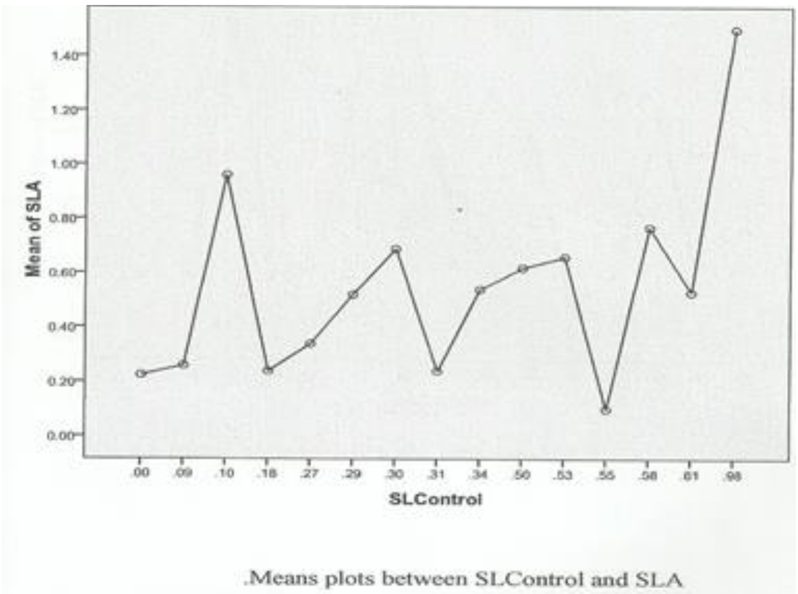

Fig 5:- Mean plots between SLControl and SLA

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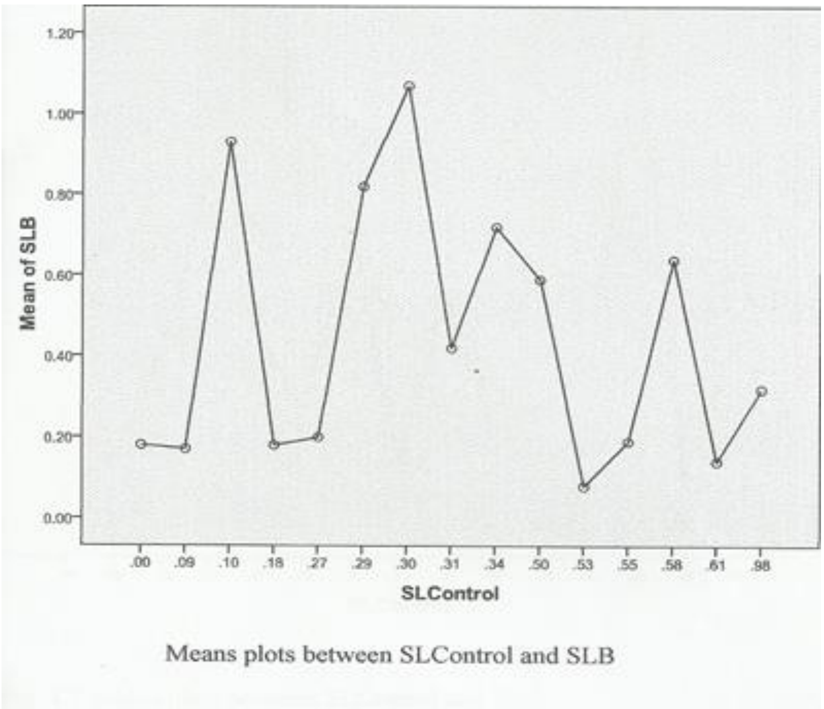

Fig 6:- Mean plots between SLControl and SLB

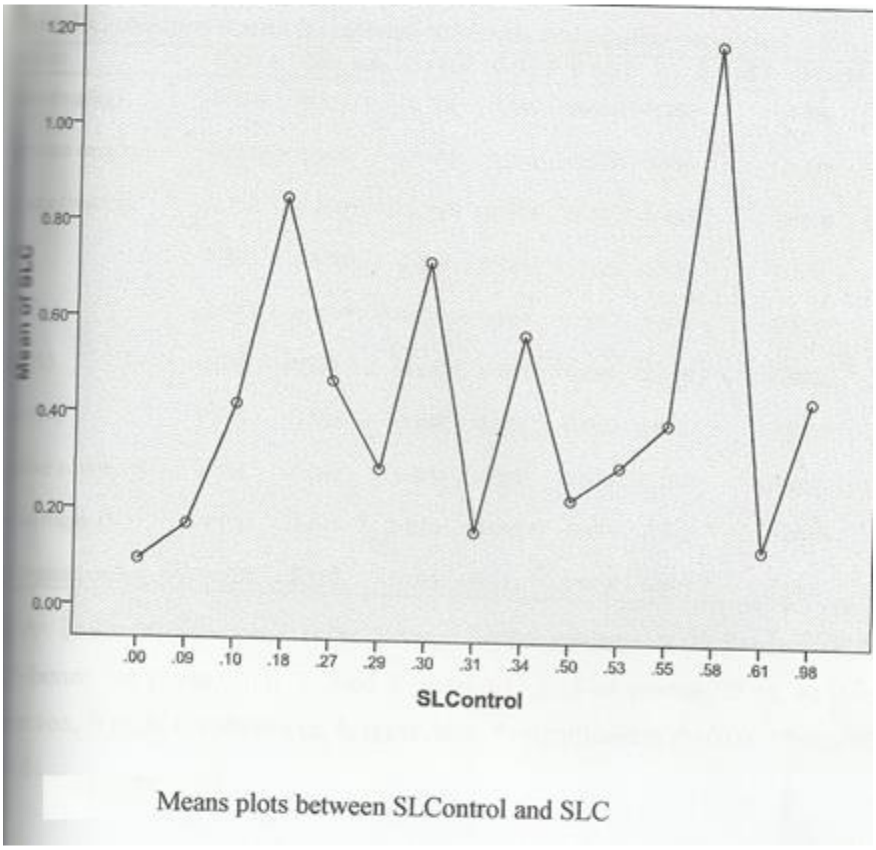

Fig 7:- Mean plots between SLControl and SLC

Variables	B(a)A	Chrysene	B(b)F	B(k)F	B(a)P	I-1,2,3-cd P	D(a,h)A	B(g,h,i)P
Nitrate (mg/kg)	-0.156	0.335	-0.761	0.388	0.686	0.756	0.582	0.462
Phosphate (mg/kg)	0.098	0.546	-0.573	0.594	0.571	0.846	0.748	0.651
Sulphate(mg/kg)	-0.776	-0.972*	0.200	-0.894	-0.093	-0.687	-0.864	-0.865
pH	0.865	0.970*	0.348	0.911	0.050	0.634	0.848	0.871
Sand(%)	0.742	0.977*	0.139	0.849	0.187	0.661	0.829	0.821
Clay(%)	-0.747	-0.943	-0.141	-0.919	-0.094	-0.758	-0.908	-0.902
Loam(%)	0.047	0.864	0.138	0.974*	-0.027	0.851	0.973*	0.970*
Moisture content(%)	0.345	-0.092	0.864	-0.242	-0.646	-0.707	-0.462	-0.337
Total Nitrogen(%)	-0.790	-0.841	-0.213	-0.992**	0.131	-0.842	-0.976*	-0.985*
Total Organic Carbon (%)	0.098	-0.237	-0.028	0.411	-0.549	0.593	0.467	0.478

B(a)A= benzo(a)anthracene, B(b)F= benzo(b)flouranthene, B(k)F= benzo(k)flouranthene, B(a)P = benzo(a)pyrene, I-1,2,3-cd P = Indeno-1,2,3-cd pyrene, D(a,h)A = dibenzo(a,h)anthracene, B(g,h,i)P= benzo(g,h,i)perylene, *= significant at $P<0.05$. **= significant at $P<0.01$.

Table 3:- Correlation matrix (r) between the PAHs and Edaphic variables

IV. DISCUSSION

The narrow variations in the concentration of the measured PAHs can be attributed to the fact that the soil samples in all sampling locations had the same pollution source. The presence of the same pollution source was also responsible for the small range in the concentration of the PAHs.

The relatively low volatility and presence of less than six benzene fused ring in the structure of benzo(a)anthracene, chrysene and benzo(b)flouranthene (Kookana *et al*., 1990; Kieron and Doick, 2005), made it possible for them to adsorb on particulates in the plume from the company. They finally settled soil surfaces through gravitational settling and deposition (Al-Hadad, 2005; Nam *et al*., 2008[a]), thus, accounting for their maximum concentration in sample location B (SLB). Nam *et al*., 2008[b] have shown that "the concentration of the PAHs showed significant spatial heterogeneity, which is an implication that, across each sampling location, the concentration of the individual PAHs is significant enough to make a difference.

Benzo(a)pyrene however, recorded highest concentrations in sample location C (SLC) and this, is due to dispersion and subsequent deposition (Lee and Kim, 2007; Srogi 2007). Sample location control (SLcontrol), recorded least concentration in all the measured PAHs and it can be attributed to volatilization, leaching, bioaccumulation by plants and dispersion by wind (Li *et al*., 2008; Zohair *et al*, 2006).

Nitrate and Sulphate had high concentrations with sulphate being higher. Nitrate and Sulphate exist in the atmosphere as oxides of Nitrogen (NOx) and sulphur dioxide (SO2), respectively. Both oxides are produced when fuel is burnt. In the atmosphere, the combine with rain to form acid rain and then, fall to the soil (Harvey, 1997).

The concentration of chrysene increases with increasing pH and sand composition and vice versa. This implies that, at lower pH and lower sand composition, the concentration of chrysene in the soil sample will decrease. High loam concentration on the other hand, has direct relationship with the concentrations of benzo(k)flouranthene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene, this means that, an increase in the loam composition of the soil, results to an increase in the concentration of the said PAHs (Abbas *et al*., 2007).

However, indirect relationships exist between chrysene and sulphate and between dibenzo(a,h)anthracene and Total Nitrogen. This implies that, at a higher concentration of chrysene, the pH of the soil sample will be low due to the low concentration of sulphate. This low pH will however, not favor the proper growth of crops as a result of its high acidity. The concentration of benzo(g,h,i)perylene and benzo(k)flouranthene each have inverse relationship with the total nitrogen of the soil.

The fate of PAHs in the soil is dependent on the edaphic variables of the soil, such as organic matter, textural class, moisture content and pH (Nam *et al.*, 2008[a]; Abbas *et al.*, 2007 and Yang, Zhang and Korenga, 2002). The high concentration of PAHs in the soil samples can be attributed to high organic carbon content of the soil, as well as low pH value, high sand composition and high moisture contents (Kookana *et al.*, 1990).

V. CONCLUSSION

The result of the analysis of soil samples around the Asphalt Plant showed the presence and a relatively high concentration of PAHs, thus, making it responsible for the primary input of PAHs into the neighboring environment (Srogi, 2007). Due to the fact that the location of the asphalt plant is inhabited majorly by agrarians, the crops grown and harvested within this area would have some concentration of PAHs in them (Li *et al*., 2008, Li & Chen, 2008 and Collins & Finnegan, 2010) and the consumption of such crops will become a route of exposure to the PAHs by the inhabitants. This becomes very dangerous to their health as PAHs have been proven to be carcinogenic, mutagenic, teratogenic and have other known health implications (Ramesh *et* al, 2010, ATSDR, 1995; Luch, 2005; WHO, 2010).

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➤ *Appendix 1: Homogeneity in spatial mean variance*

Anova: Single Factor

SUMMARY

Groups	*Count*	*Sum*	*Average*	*Variance*
Column 1	64	27.288	0.427938	0.096688
Column 2	64	160	2.5	1.269841

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	134.3902	1	137.3902	201.079	7.16528	3.916325
Within Groups	86.09137	126	0.683265			
Total	223.4815	127				

Table 4

➢ *Appendix 2: Edaphic variables measured in soils around an asphalt plant in Owerri.*

S/N	PHYSICOCHEMICAL PROPERTIES	SLA	SLB	SLC	SLCONTROL
1	Nitrate (mg/kg)	30.8	29.5	31.0	28.9
2	Phosphate (mg/kg)	10.8	9.82	10.8	8.65
3	Sulphate (mg/kg)	60.4	58.2	61.2	66.5
4	pH	5.4	5.6	5.3	5.0
5	Textural class				
	Sand	70	72	70	65
	Clay	20	18	22	30
	Loam	10	10	8	5
6	Moisture content (%)	2.0	3.0	2.0	3.0
7	Total Nitrogen (%)	0.14	0.14	0.15	0.16
8	Total Organic Carbon (%)	1.8	1.4	1.3	0.15
9	Carbon – Nitrogen Ratio	12.86	10	8.67	9.38

Table 5