

A Measurement of Selected Polycyclic Aromatic Hydrocarbons in Petroleum Product Contaminated Soils Using a Gas Chromatograph

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Oil contaminated soils around refined petroleum products depot and retailing stations were investigated for selected polycyclic aromatic hydrocarbons (PAHs) to determine their levels in the soils. Soil samples were collected using standard analytical procedures and the petroleum hydrocarbons extracted using standard method, while the PAHs (benzo[a]pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, and anthracene) contents of the extracted petroleum hydrocarbons were determined using SNR 8126 gas chromatograph equipped with a flame ionization detector (FID). The results indicated that benz[a]anthracene has the highest concentrations in both the oil depot contaminated soils (59.1 ± 3.3 mg/kg) and oil retailing station contaminated soils (20.8 ± 2.1 mg/kg). The results also showed that the oil depot contaminated soils were burdened with the analyzed PAHs than oil retailing station contaminated soils; this was confirmed by the results of *t* test analysis. Comparison of the values of the PAHs in the oil impacted soils with their background levels showed that their values in the two sets of oil impacted soils were excessive; this was corroborated with their high pollution (PI) and geoaccumulation (I_{geo}) index values, except chrysene, which *moderately polluted* the soils (low PI and I_{geo} values); this was clear indication of indiscriminate discharge of petroleum fuels and petrochemicals into the soils.

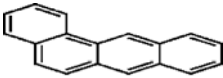
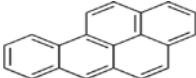
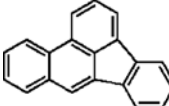
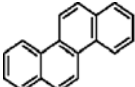
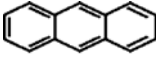
Keywords: GC, oil depot, oil retailing station, PAH, petroleum, soil contamination

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a set of varied organic compounds containing more than one aromatic ring of carbon and hydrogen atoms chemically combined together. They are mostly produced from the combustion of fossil fuels and are mostly colorless, white, or pale yellow-green solids. Most of them are not readily soluble in water, but easily evaporate into the air and are often adhere to dust particles. They are usually adsorbed strongly to the organic fraction of sediments and soils. They are one of the potent persistent organic compounds (POPs) common globally and can bioaccumulate. Due to this reason, sediments and soils are usually considered as the main sinks for PAHs in the environment (U.S. Environmental Protection Agency, 1984; Jones and Voogt, 1999; Maldonado et al., 1999; Sitaras et al., 2004). The U.S. Environmental Protection Agency (1984) has classified seven PAHs viz. benzo[a]pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, dibenz[a,h]anthracene and indeno[1,2,3-cd]pyrene as probable human carcinogens. Several PAHs and their derivatives are carcinogenic and/or mutagenic in nature. They

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TABLE 1
Physical-chemical Properties of the Analyzed PAHs of Interest

PAH	Structure/Molecular Formula	Molecular Weight	Aqueous Solubility (Room Temp.), mg/L	Vapor Pressure (Room Temp.), mm Hg	Henry's Law Constant, atm.m ³ .mol ⁻¹	Log Kow	Log Koc
Benzo(a)anthracene	 C ₁₈ H ₁₂	228	9.4 × 10 ⁻³	3.05 × 10 ⁻⁸ to 1.05 × 10 ⁻⁷	3.35 × 10 ⁻⁶	5.7 ^A	5.3
Benzo(a)pyrene	 C ₂₀ H ₁₂	252	1.6 × 10 ⁻³	5.49 × 10 ⁻⁹	1.13 × 10 ⁻⁶	5.97, 6.11 ^A , 6.58 ^B	6.0 to 6.7
Benzo(b)fluoranthene	 C ₂₀ H ₁₂	252	1.5 × 10 ⁻³	5.0 × 10 ⁻⁷	1.11 × 10 ⁻⁴	6.2 ^A	5.2
Chrysene	 C ₁₈ H ₁₂	228	2.0 × 10 ⁻³ to 6.3 × 10 ⁻³	6.23 × 10 ⁻⁹	9.46 × 10 ⁻⁵	5.7 ^A	5.1
Anthracene	 C ₁₄ H ₁₀	178	0.043 to 0.075	2.67 × 10 ⁻⁶	1.93 × 10 ⁻⁵ to 6.5 × 10 ⁻⁵	4.45, 4.55 ^A	4.2 to 4.4

Source: Canadian Council of Ministers of the Environment (2008).

have been recognized as primary pollutants, significantly influence public health, and are considered to be responsible for a number of respiratory diseases such as pharynx cancer. PAHs have moderate to high acute toxicity to aquatic life and birds. Some cause damage and death to agricultural and ornamental crops. Several studies on their fate in natural environments, source, occurrence, health effects as well as transport have been carried out by many workers (Wilson and Jones, 1993; Manoli and Samara, 1999; Xu and Lee, 2001; Oviasogie et al., 2006).

In Nigeria, several works have been conducted on environmental impacts of petroleum; these works were concentrated on crude oil spillages and air quality in oil producing areas of Niger Delta, Nigeria (Osuji and Adesina, 2005; Osuji and Onojake, 2006; Oviasogie et al., 2006; Osuji and Nwoye, 2007), but until now there are little or no literature/work on PAHs pollution impacts on petroleum products contaminated soils around petroleum products retailing stations and depots areas.

Table 1 presents a brief review of the structural, physical, and chemical properties of the PAHs of interest. The octanol-water partition coefficient (K_{ow}), organic carbon-water partition coefficient (K_{oc}), Henry's law constant, vapor pressure, and aqueous solubility are the specific chemical properties that are direct significance in foretelling the environmental fate of any substance,

including multi-media partitioning behavior, bioavailability and resistance to biodegradation (Canadian Council of Ministers of the Environment, 2008).

This study thus aimed at the determination of the levels of some selected polycyclic aromatic hydrocarbons in petroleum products contaminated soils of the aforementioned areas in Nigeria.

Mosimi Petroleum Products and Marketing Company (PPMC) depot located at Shagamu and petroleum products retailing stations in Ile-Ife, Southwestern Nigeria, were studied. Mosimi depot is one of the principal depots in Nigeria; it accepts refined petroleum products primarily from Warri refinery and Atlas Cove Jetty in Lagos, Nigeria, by the use of pipelines, stores them and distributes them to marketers from Ogun, Lagos, and some Northern states of Nigeria. Most of the inhabitants of the area are peasant farmers who do their farming activities around the depot and they depend on ground and surface waters as sources of domestic water and irrigation. The majority of the petroleum products retailing stations in Nigeria are located inside residential and commercial districts. Petroleum products that contain PAHs can contaminate soils and leach into ground and surface waters, which are major sources of water for drinking and domestic uses. Due to resultant adverse effects of PAHs, it is vital to assess their levels in petroleum products contaminated soils of the areas.

2. EXPERIMENTAL

2.1 Sample Collection and Treatment

Simple random sampling method was used to collect 10 oil contaminated soil samples from different locations within the study areas (Mosimi Petroleum Products and Marketing Company [PPMC] depot located at Shagamu and petroleum products retailing stations in Ile-Ife, Southwestern Nigeria) at a depth of 0–5 cm using a hand trowel and was wrapped in aluminum foils to prevent contamination. The samples were preserved at -4 to 0°C and were immediately taken to the laboratory for analysis.

2.2 Analysis

The petroleum hydrocarbons of the soil were extracted by Intergovernmental Oceanographic Commission (1982) method. The extract was then subjected to cleanup with a short silica column. After elution with purified n-hexane, the petroleum hydrocarbons were isolated by evaporation of the n-hexane and concentration of the extract (petroleum hydrocarbons) by rotary evaporator to a constant weight.

All the reagents used were of BDH Analar grade and the solvents (methanol, n-hexane, water) used were triply distilled. The glass wares used were cleaned using the procedure of Laxen and Harrison (1981). Three replicate measurements of each of the test samples were carried out.

The concentrations of selected polycyclic aromatic hydrocarbons viz. benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthrene, chrysene, and anthracene were determined using SNR 8126 gas chromatograph with a flame ionization detector (FID) in a splitless mode at Analytical Service Laboratory, International Institute of Tropical Agriculture (IITA), Ibadan, Southwestern, Nigeria. Equipped with the chromatograph was a capillary column (dimethylpolysiloxane) of length 30 m and 0.25 mm i.d., 2.5 μm film thickness, while the flow rate of the carrier gas (nitrogen) was 12.7 mL min^{-1} . Temperature of the oven was maintained at 40°C for $9^{\circ}\text{C min}^{-1}$ rate; while the injector and detector temperatures were 210 and 310°C , respectively, and 1 mL was used as the injection volume.

Measurement of the PAHs was carried out from GC profiles by means of the external standard technique that comprises 16 PAH compounds (U. S. EPA 610 PAH mixture) including the analyzed PAHs. Measurement of the analyzed PAHs was carried out by the linear regression modus operandi

TABLE 2
Statistical Summary and Pollution Index (PI) of the PAH Concentrations in the Contaminated Soils

PAH, mg/kg	Background Level Jones et al. (1987)														
	Petroleum Product Depot			Petroleum Product Retailing Station			Rural Soil			Agriculture Soil			Urban Soil		
	56.3–61.8 (59.1 ± 3.3)	49.1–52.4 (50.7 ± 3.2)	38.2–41.2 (39.7 ± 2.7)	19.2–22.4 (20.8 ± 2.1)	19.0–20.1 (19.5 ± 1.3)	18.8–21.4 (20.1 ± 1.9)	0.005	0.056	0.169	11,820.0	1,055.4	349.7	4,160.0	371.4	123.1
Benzo(a)anthracene	56.3–61.8 (59.1 ± 3.3)	49.1–52.4 (50.7 ± 3.2)	38.2–41.2 (39.7 ± 2.7)	19.2–22.4 (20.8 ± 2.1)	19.0–20.1 (19.5 ± 1.3)	18.8–21.4 (20.1 ± 1.9)	0.005	0.056	0.169	11,820.0	1,055.4	349.7	4,160.0	371.4	123.1
Benzo(a)pyrene	49.1–52.4 (50.7 ± 3.2)	38.2–41.2 (39.7 ± 2.7)	55.1–62.6 (58.8 ± 3.9)	15.6–16.0 (15.8 ± 1.8)	15.6–16.0 (15.8 ± 1.8)	17.4–21.6 (19.5 ± 1.8)	0.002	0.0046	0.165	25,350.0	11,021.7	307.3	9,750.0	4,239.1	118.2
Benzo(b)fluoranthene	38.2–41.2 (39.7 ± 2.7)	55.1–62.6 (58.8 ± 3.9)	48.5–59.9 (54.2 ± 3.1)	0.0383	0.078	0.251	0.020	0.058	15.0	1,985.0	684.5	2.7	1,002.0	346.6	1.34
Chrysene	55.1–62.6 (58.8 ± 3.9)	48.5–59.9 (54.2 ± 3.1)		0.0383	0.078	0.251	0.0383	0.078	1,535.2	753.9	234.3	412.5	202.6	63.0	
Anthracene	48.5–59.9 (54.2 ± 3.1)			17.4–21.6 (19.5 ± 1.8)			N.A	0.011	N.A	—	4,927.3	—	—	1,772.7	—

N.A = not available.

TABLE 3
Statistical Summary and Geoaccumulation Index (I_{geo}) of the PAH Concentrations In The Contaminated Soils

PAH, mg/kg	Petroleum product depot	Petroleum product retailing station	Background Level Jones et al. (1987)						Petroleum Product Depot (I_{geo})						Petroleum Product Retailing Station (I_{geo})					
			Rural Soil		Agriculture Soil		Urban Soil		Rural Soil		Agriculture Soil		Urban Soil		Rural Soil		Agriculture Soil		Urban Soil	
Benzo(a)anthracene	56.3–61.8 (59.1 ± 3.3)	19.2–22.4 (20.8 ± 2.1)	0.005	0.056	0.169	2,372.1	211.8	70.2	834.9	74.5	24.7									
Benzo(a)pyrene	49.1–52.4 (50.7 ± 3.2)	19.0–20.1 (19.5 ± 1.3)	0.002	0.0046	0.165	5,087.4	2,211.9	61.7	5,087.4	850.7	23.7									
Benzo(b)fluoranthene	38.2–41.2 (39.7 ± 2.7)	18.8–21.4 (20.1 ± 1.9)	0.020	0.058	15.0	398.4	137.4	0.531	398.4	69.6	0.269									
Chrysene	55.1–62.6 (58.8 ± 3.9)	15.6–16.0 (15.8 ± 1.8)	0.0383	0.078	0.251	308.1	151.3	47.0	308.1	40.7	12.6									
Anthracene	48.5–59.9 (54.2 ± 3.1)	17.4–21.6 (19.5 ± 1.8)	N.A	0.011	N.A	—	988.8	—	—	355.8	—									

N.A = not available.

TABLE 4
Results of the *t* Test Comparing the Concentrations of the PAHs in the Petroleum Product Depot and Petroleum Product Retailing Station Contaminated Soils

PAH	<i>t</i> Test		Remark
	<i>t</i> _{exp}	Observation	
Benzo(a)anthracene	26.0	<i>t</i> _{exp} > <i>t</i> _{theor}	SD
Benzo(a)pyrene	21.8	<i>t</i> _{exp} > <i>t</i> _{theor}	SD
Benzo(b)fluoranthene	16.3	<i>t</i> _{exp} > <i>t</i> _{theor}	SD
Chrysene	24.7	<i>t</i> _{exp} > <i>t</i> _{theor}	SD
Anthracene	25.1	<i>t</i> _{exp} > <i>t</i> _{theor}	SD

*t*_{exp} = *t*_{experimental}; *t*_{theor} = *t*_{theoretical} at 95% confidence level = 4.303. SD = significant difference.

(*r*² > 0.99) using five-point calibration curves recognized between the reliable standard concentration and corresponding peak areas. Blanks levels of analyzed PAHs were low and in a good number of cases not detectable. Analysis of serial dilutions of PAHs standard indicated that chromatographic method detection limit ranged between 0.007 and 0.16 ng for PAH compounds. The recovery efficiency of the technique was assessed by the analysis of known concentrations of spiked standard PAH compounds. The compounds presented high recoveries with average values ranging between 75% and 85%.

3. DATA TREATMENT

3.1 *t* Test Analysis

Comparison of the concentrations of the PAHs in the oil depot and retailing station soil samples was done using *t* test analysis. The value of *t* was computed by employing the expression:

$$t = (\mu - x) / \sqrt{N} / S.D \quad (1)$$

and then compared with the theoretical value of *t*(4.303) from statistical table at 95% confidence interval and the degree of freedom. Where *N* = number of samples; S.D = standard deviation; *t* = *t* test value; μ = concentration of a particular PAH in oil depot soils, and *x* = concentration of the same PAH in oil retailing soils respectively. The *t* values were calculated to indicate apparent significant difference from statistical point of view, between the concentrations of the PAHs in the two sets of soil samples. If *t*_{experimental} > *t*_{theoretical}, it is concluded that there is a *significant difference* between μ and *x*, otherwise it is *insignificant* (Hadjiioannou et al., 1988).

3.2 Pollution Index

The pollution index (PI) values of the PAHs in the soils were determined. The PI is the quotient of the concentration of the PAH *x* in the sample to the background levels of the PAH. $PI_{(x)} = \text{Concentration in the sample} / \text{background level}$. It is agreed in principle that if the value of PI of the analyte is greater than 1.0, it implies that the contamination of the sample by the analyte is high and may be toxic at the level it is present in the sample (Onianwa and Adoghe, 1997). The background levels of the PAHs in agriculture, and urban and rural soils as reported by Jones et al. (1987), were used.

TABLE 5
Comparison of the Concentrations of the PAHs in the Soils With other Nigerian Soils and Soils of Other Nations

PAH, mg/kg	Nigerian Petroleum Contaminated Soils										Canadian Soils (CANADIAN COUNCIL OF MINISTERS OF THE ENVIRONMENT, 2008)	
	This Study					Refinery and Niger Delta Soils (Okoro and Ikolo, 2007)					Rural Soils	Agricultural Soils
	Petroleum Product		Petroleum Product		Refinery	Niger Delta Soils		English Soils (Bradley <i>et al.</i> , 1994)	Rural Soils	Agricultural Soils		
	Depot	Product	Station	Product		Enerthen	Ekpan					
Benzo(a)anthracene	56.3–61.8 (59.1 ± 3.3)	19.2–22.4 (20.8 ± 2.1)	0.553	0.435	0.464	0.005–0.02	0.056–0.11					
Benzo(a)pyrene	49.1–52.4 (50.7 ± 3.2)	19.0–20.1 (19.5 ± 1.3)	0.731	0.908	0.562	0.002–1.3	0.005–0.9					
Benzo(b)fluoranthene	38.2–41.2 (39.7 ± 2.7)	18.8–21.4 (20.1 ± 1.9)	2.068	0.697	0.718	0.02	0.06–0.22					
Chrysene	55.1–62.6 (58.8 ± 3.9)	15.6–16.0 (15.8 ± 1.8)	4.685	1.911	1.716	0.04	0.08–0.12					
Anthracene	48.5–59.9 (54.2 ± 3.1)	17.4–21.6 (19.5 ± 1.8)	0.526	0.122	0.219	N.A	0.011–0.013					

N.A = Not available

3.3 Geoaccumulation Index (I_{geo})

Geoaccumulation index (I_{geo}) values for the PAHs were calculated using the equilibrium equation $\{I_{geo} = \text{Log}_2(C_n/1.5B_n)\}$ according to Diatta (2008) and background levels of PAHs in noncontaminated soils as reported by Jones et al. (1987). Where C_n is the measured mean concentration of the PAH in the soil samples; B_n is the background value and 1.5 is the background matrix correction factor due to lithogenic effects.

4. RESULTS AND DISCUSSION

Table 2 presents and compares the concentrations of the analyzed PAHs (benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthrene, chrysene, and anthracene) in the oil contaminated soils collected from the oil depot and retailing station areas. The results indicated that benz[a]anthracene has the highest concentrations in both the oil depot contaminated soils (59.1 ± 3.3 mg/kg) and oil retailing station contaminated soils (20.8 ± 2.1 mg/kg). It is observed that all the PAHs have higher concentrations in the oil contaminated soils collected from the depot than the soils collected from the retailing stations; indicating that the impacts of the petroleum products are higher in the depot than the retailing stations. This was corroborated by the t test values of the analyzed PAHs in Table 4 which indicated that statistically all the analyzed PAHs showed apparent *significant* differences in their values between the contaminated oil depot and retailing station soils as a result of high concentrations of the PAHs in the oil-impacted soils collected from the oil depot.

Table 2 also compares the values of the analyzed PAHs in the oil depot and retailing soils with their background levels in agriculture, urban and rural soils as reported by Jones et al. (1987). It is observed that the concentrations of all the analyzed PAHs exceeded their background levels. The high levels of PAHs observed are clear indications of indiscriminate discarding of petroleum products onto the soils of the study areas.

The PI results indicated that all the analyzed PAHs in the two set of soils have values greater than 1.0; indicating that the soils were contaminated with the PAHs and may pose health problem to the residents of the study areas.

Table 3 presents the I_{geo} values for the analyzed PAHs. The I_{geo} values of the analyzed PAHs in the of soils were calculated using the equation according to Diatta et al. (2008) and their background levels as reported by Jones et al. (1987). It is observed that the two sets of oil contaminated soils were *very heavily polluted* with respect to all the PAHs (I_{geo} value > 6), except chrysene which *moderately polluted* the soils.

The results of the t test comparing the concentrations of the PAHs in the petroleum product depot and petroleum product retailing station contaminated soils are provided in Table 4. All the t test values using the averages of the analyzed PAHs in the two set of soils as variables are considered *significant* ($t_{\text{experimental}} > t_{\text{theoretical}}$, 4.303 at 95% confidence level) and this is due to the considerably higher concentrations of the PAHs in the soils of the petroleum product depot than petroleum product retailing station soils.

Table 5 presents the comparison of the concentrations of the PAHs in the analyzed soils in this study with other Nigerian soils and soils of developed nations. It is observed that the values of all the analyzed PAHs are more in the soils than other Nigerian petroleum contaminated soils (Okoro and Ikolo, 2007) and other nations viz. Canadian soils (Canadian Council of Ministers of the Environment, 2008) and English soils (Bradley et al., 1994).

The elevated levels, I_{geo} and PI values obtained for these toxic PAHs in the soils are worrisome due to their possible undesirable effects on the health of living organisms including human beings and can also alter the ecosystem components of the areas if the oils are washed into the ground and surface waters of the areas by run-off or emitted and pollute the ambient air. The pollutants may also

get into the food web as a result of their bioaccumulation by food crops via the contaminated soils as a result of animal and crop farming activities around the depots by the villagers living around the area who are majorly peasant farmers. The bulk of the petroleum products retailing stations in Nigeria are located within residential and commercial centers. Petroleum products that contain PAHs can contaminate soils and leach into ground and surface waters, which are major sources of water for drinking and domestic uses. These can cause health problems for the inhabitants of the areas.

5. CONCLUSION

The concentrations of selected PAHs (benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthrene, chrysene, and anthracene) in soils around one of the biggest petroleum products depot and petroleum products retailing stations in Nigeria were measured using SNR 8126 gas chromatograph. The oil-impacted soils were burdened with these potential environmental pollutants due to the fact that their values were higher than their background values that were corroborated with their high PI values. The results also showed that oil-impacted soils collected from oil depot contained higher concentrations of the analyzed PAHs than the oil-impacted soils obtained from oil retailing stations; this was confirmed by their *t* test values that showed significant differences. It is also observed that the PAHs have elevated levels of I_{geo} and PI in the soils.

As it has been established by this study that the oil impacted soils have extreme levels of the potential toxic PAHs that can cause different kinds of ill health, it is recommended that indiscriminate discharge of petroleum oils and petrochemicals should be discouraged; locating of petroleum products depots and retailing stations close to human settlements, and farming activities around petroleum depots should also be discouraged. The environmental regulatory bodies should be active in order to eschew environmental degradation.

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