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Assessment of polycyclic aromatic hydrocarbons in the soil around River Owan in Edo State, Nigeria: Occurrence, distribution, source, and health risk

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Abstract

Environmental pollutants called polycyclic aromatic hydrocarbons (PAHs) are common contaminants with human health and environmental concerns. The 16 priority, 1-methylnaphthalene, and 2-methylnaphthalene PAHs in soil samples from agricultural farmland around River Owan, Edo State, Nigeria was determined by deploying a flame ionization detector with gas chromatography. The total concentration ∑16PAHs of the soil samples is in the range of 0.198 – 0.518µg/kg, ∑18PAHs 0.23 - 0.56 µg/kg, ∑LMW PAHs 0.095 – 0.205 µg/kg while the ∑HMW PAHs 0.087 - 0.348 µg/kg. The Concentration of ∑PAHs is in this order: SO1>SO4>SO5>SO6>SO2>SO7>SO3. The ratio of the LMW/HMW PAHs percentage is 43% - 57%. The ∑7cPAHs were in the range of 0.032 - 0.245 µg/kg with a mean value of 0.133 µg/kg. The ∑TEQ range is 0.004 - 0.139 while the BaP-EQ of the soil samples is 0.285 µg/kg indicating no risk. The diagnostic ratio showed more of the pyrogenic source. The percentage of petrogenic to pyrogenic in the ratio of Ant/(Ant + Phe) is 43% to 57%. The ratio of Flt/(Flt + Pry) is above >0.1 meaning they are from pyrogenic sources. The value of the children's incremental lifetime cancer risk (ILCR) ranged from 6.43×10^{-8} - 6.46×10^{-6}, 2.58×10^{-8} - 2.59×10^{-6}, and 1.09×10^{-12} - 1.10×10^{-10} for dermal, ingestion and inhalation while for adult: 2.43×10^{-8} - 2.44×10^{-6}, 1.37×10^{-8} - 1.37×10^{-6}, and 9.27×10^{-13} - 9.32×10^{-11} for same routes of exposure respectively showing a negligible risk.

Keywords: Ecology, Risk assessment, Owan, PAHs, Soil toxicity

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are chemical compounds with two or more benzene rings fused together, which are part of the major group called persistent organic pollutants (POPs). There are two major classes of PAHs, those with 2-3 benzene rings called low molecular weight (LMW) such as naphthalene, acenaphthylene, fluorene, etc., and those that contain 4-6 benzene rings which are fused together, known as high molecular weight (HMW) they include pyrene, chrysene, benzo[a]anthracene, benzo[a]pyrene and so on [1-4]. They are usually produced and released as a result of artificial (anthropogenic activities) and natural occurrences. The natural sources include emissions from forest fires, volcanic eruptions, fossil fuels, crude petroleum, and so on, while the anthropogenic sources are from any human action(s) which include oil spillage, gas flaring, exhaust from vehicles and automobiles, effluents from industries, asphalt particles, incinerators, emissions from man-made combustion processes, accidental releases, agricultural and municipal runoff and so on. [5-7]. Polycyclic aromatic hydrocarbons have attracted the attention of environmental researchers and policymakers as a result of their toxicity, carcinogenic, mutagenic, and teratogenic effects. A total of 16 PAHs have been designated as priority pollutants by the United States Environmental Protection Agency (USEPA). Among the list of 16 are seven (07) PAHs majorly classified as carcinogenic PAHs (cPAHs) which are: Chrysene (Chr), benzo[a]anthracene (BaA), dibenzo[a,h]anthracene (DB), benzo[ghi]perylene (Bghi), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), they all have negative health implications [4, 8-11].

They tend to bio-accumulate and bio-magnify in organisms, are persistent, and are found everywhere even in pristine areas [6, 12]. Since they have a propensity to bind endogenous receptors, they are categorized as endocrine-disrupting substances [13-15]. Researchers across the globe have found them in the atmospheric air [16-18], water [19, 20], sediments [7, 21, 22], soil [6, 20, 23-25], agricultural produce and products [26-30], daily needs such as cosmetics [31-34], biota [35-37], and in biological samples such as human blood, including breast milk [11, 38, 39].

Soil is the sink of most environmental pollutants including POPs and heavy metals. Researchers across the world examined PAHs in soil from various land use (Table 1) with varying concentrations. Man depends directly and or indirectly on the usage of soil for various purposes most importantly – agriculture [10, 20, 40]. Produce and commodities from agriculture are the major sources of food depended upon by man and livestock. Plant crops can uptake environmental pollutants such as PAHs from the soil, bio-accumulate, and bio-magnify them in their system, thereby making food crops from contaminated or polluted soil a health concern for consumers

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and food quality regulators [41]. Pollutants from the soil are usually leached and or washed to nearby rivers by erosion thereby polluting the water bodies around such land or agricultural sites. The water body and the aquatic organisms in the river are exposed to these pollutants, they also ingest the pollutants, bio-accumulate, and bio-magnify them. All these processes take place at varying trophic levels. The three main ways by which individuals become vulnerable to environmental pollutants like PAHs are by means of ingestion, inhalation, and dermal absorption [11, 28, 34].

Ovia North East Local Government in Nigeria's Edo State is where Owan is located. It is a community known for agricultural activities such as lumbering, cultivation of food and cash crops, and fishing from the river in the community. Owan is one of the largest producers of plantain and other agricultural produce such as cocoa, and palm oil in Nigeria. The environment is chosen for its various human activities including vehicular movement of trucks across two major States of Nigeria (Ondo and Edo States, see Figure 1a). This study area is known for several agricultural practices. Bush burning producing fly ash, deforestation, and heavy use of various agrochemicals are some of the human activities [25]. There is a need to determine the concentration of environmental pollutants like PAHs in soil from this agricultural zone. There has not been any scientific report on the concentration of PAHs in the soil from this area except in the preliminary studies carried out by our research group [25]. Thus, to further provide baseline data, this study aims to further deter PAHs in the soil samples from various farms around River Owan, while the specific objectives are to determine the occurrence, and distribution, identify possible sources, and risk assessment of PAHs in the soil surrounding the Owan community.

Table 1 Levels of PAHs in some soil land use around the world

Location/Country/Region	**Land use**	**∑16PAHs (ug/kg)**	**Reference**
Daye Lake, Yangxin County, and Huangshi City	Farming	8.06 – 3233.80	[20]
Himalayan region, Pakistan	Riverine	62.79 – 1080	[42]
Coastal and estuarine areas of Northern Bohai and Yellow Seas, China	Coastal/Industrial	66-920	[43]
Isfahan metropolis, Iran	Urban soil	57.70 – 11,730.08	[44]
Coal-fired power plants, South Africa	Industrial	9.73 – 61.24	[23]
Central South Africa	Residential, industrial, and agricultural	44 – 39000	[45]
Poland	Agricultural soils	3 – 685	[46]
Megacity Shanghai, China	Urban soils	83.3 – 7220	[47]
Hangzhou, China	Urban soils	180.77 – 1981.45	[21]
Jilin Province, China	Agricultural soil	602.12 – 1271.87	[6]
Gyeonggi-do, Ulsan, and Pohang, Korea	Agricultural soil	19.53–672.93, 125.01–3106.27, and 51.94–8106.21	[24]
Shanghai, China	Agricultural soil	223 – 8214	[10]
Midway Atoll, North Pacific Ocean	Industrial and agriculture	3.55 – 3200	[48]
Owan community. Nigeria	Agricultural soil	0.23 – 0.56	This study

2. Materials and methods

2.1 Research location

Owan community belongs to Ovia North East Local Government of a popular State in Nigeria Edo State. It falls at latitudes 5° N and 6° N, and longitudes 7° E and 7° E (Figure 1) [49]. Two major climatic seasons exist in Nigeria: the dry and the wet seasons. The dry season is usually experienced between November to March while the wet or rainy season is from April to October. There are several towns, communities, villages, and camps around River Owan including Agbenikaka, Odei, Ogbigbi Okpokhumi, Sabongida, etc. Owan community is known for the cultivation of cash and food crops. Some of the crops are cocoa, palm tree, plantain, banana, yam, cassava, etc. Fishing is another occupation of the people in this community from the rivers which is also used for irrigation purposes of farm plantations close to the river. The river serves various uses including domestic utilisation such as drinking, washing, bathing, cooking, etc. In addition to the plantain market along the river banks, other activities include the building, repair, and maintenance of canoes and other automobiles such as speedboat engines [25, 49].

2.2 Sampling and sampling period

Aggregate soil samples were taken by collecting soil samples from three points and composited by quartering method for a location as one, this was done for all the sampling stations. Seven (07) farm locations were selected which are along River Owan. The types of crop(s) on the farms were noted (Table 2). A stainless-steel soil auger was utilized to collect the soil samples from 0 to 30 cm depth.

Co-ordinates of farm locations were taken using GPSMAP76S [50]. All soil samples collected were assigned codes (SO1 to SO7). Clean 1-liter glass jars with Teflon-lined screw caps were used to collect the soil samples. These containers were then transferred to the lab while being kept at <4°C in an ice chest. Samples were taken in February 2019 during the dry season. In the Chemistry lab of the Department of Science Laboratory Technology at Rufus Giwa Polytechnic, Nigeria, soil samples were dried at ambient temperature.

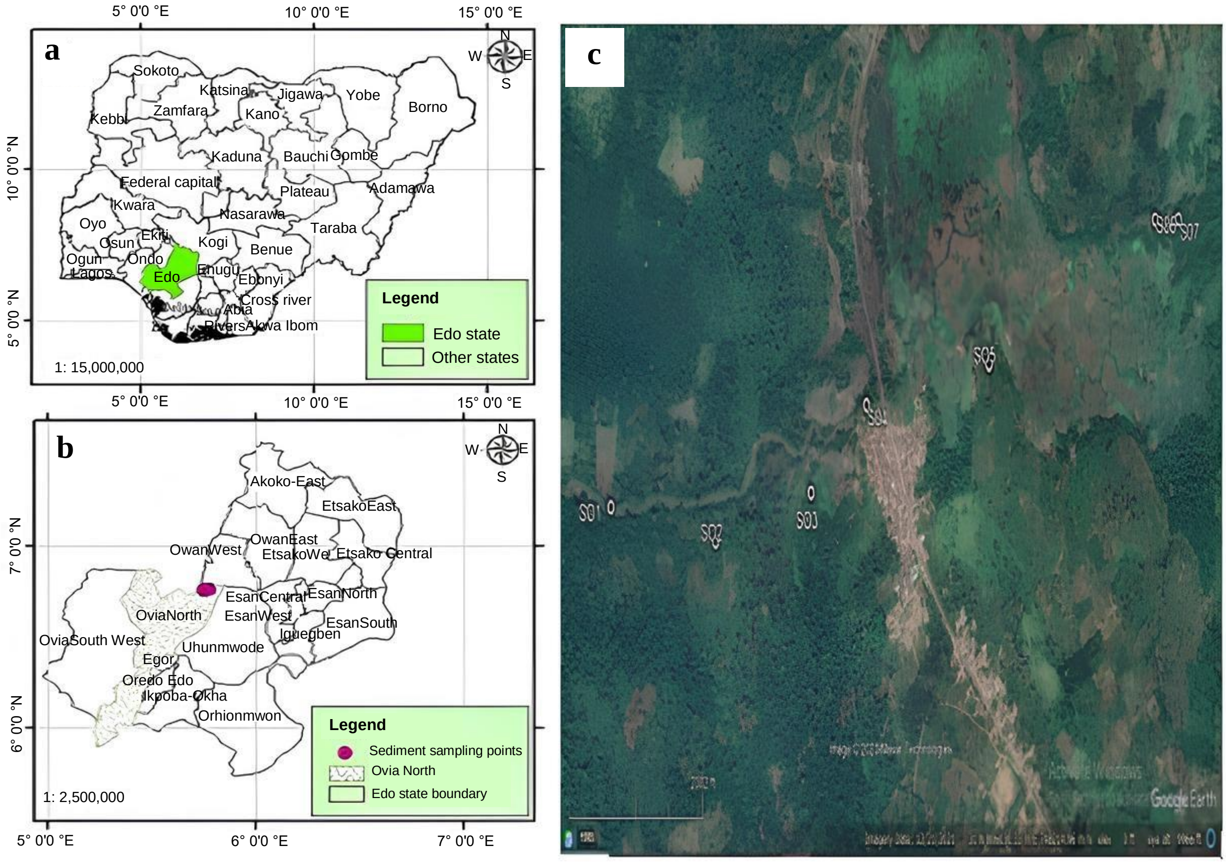

Figure 1 Locations of the study area and sampling; (a) Map of Nigeria; (b) Administrative map of Edo State; (c) Sampling locations

Table 2 Co-ordinates and description of farmland use where soil samples were collected

Farm stations	GPS Readings	Location	Type of crop(s) on the farm
1	N 06.76605° E 005.78208°	±5m	Palm tree plantation only.
2	N 06.76604° E 005.78096°	±5m	Cocoa, palm trees, and plantain plantations.
3	N 06.76280° E 005.77303°	±5m	Cassava, Cocoa, and plantain plantations.
4	N 06.76189° E 005.76719°	+4m	Palm tree plantation only.
5	N 06.75961° E 005.75499°	±5m	Cocoa plantation only.
6	N 06.75884° E 005.75999°	±5m	Cocoa, plantain, rubber plantations.
7	N 06.75993° E 005.76454°	+5m	Palm tree, Cassava, plantain, and pineapple plantations.

Source: This study

2.3 Sample preparation, treatment, and analysis

We adopted all of the steps laid out by Chen et al. [6]. In a nutshell, dichloromethane and acetone were used at a 1:1 (v/v) ratio employing an ultrasonic bath to extract 5.0 g of soil samples and 5.0 g of sodium carbonate. Using silica chromatography columns (Cleanert Florisil 1000 mg/6-mL cartridge, Agela Technologies Inc., USA), the extracts were cleaned. Using a rotary evaporator (RE–52A), the extracts were concentrated to almost dryness before being redissolved in 10 mL of n-hexane. The columns were initially eluted using a 7:3 (v/v) mixture of n-hexane and DCM. The elutes were transferred to 2-mL Teflon-lined vials with caps for gas chromatography (GC) analysis after being reduced to 1 mL using a rotary evaporator, and refrigerated at - 4ºC for analysis.

Agilent 7890A Gas Chromatography (GC) with a flame ionization detector (FID) was used for the analysis, using a micro syringe type (Exmire), and a rubber septum. One microlitre (1.0 µL) of concentrated sample was injected into the column. The following describes the operating setting utilized for GC analysis:

2.4 Gas chromatography calibration

Equipment calibration was carried out with PAH standard from Accu Standards containing the 16 priority PAHs, 1-methylnaphthalene, and 2-methylnaphthalene. According to the manufacturer's instructions, the standards were prepared, then setting the gas chromatography (GC) to the optimal temperature, injecting 1.0 µL of the standards prepared for calibration, and then run the GC as usual. The quantification and calibration of PAH were carried out.

2.5 Gas chromatograph analysis conditions

Helium gas was the carrier gas used with a percentage purity of 99.999%, flowing at a steady rate of 1.0 mL per minute. After being originally set at 50 °C for 2 minutes, the GC oven temperature was raised to 200 °C for another 2 minutes at a rate of 19 °C per minute, 240 °C for another 2 minutes at a rate of 4.5 °C/minute, and 300 °C for 5 minutes at a rate of 2.5 °C/minute.

2.6 Source apportionment using diagnostic ratio

Diagnostic ratio (DR) is one of the methods available to identify the source(s) of PAHs in various environmental matrices qualitatively by using the ratios of the PAH components [51]. It is applied based on the fact that isomers of PAHs possess similar chemical properties and their reactions in the environment are similar in terms of transformation and degradation. The ratios of the isomers remain the same from the time of emission to analysis or determination [51, 52]. The ratios of Ant/(Ant+Phe) and Fla/(Fla +Pyr) are usually used to differentiate between pyrogenic and petrogenic sources. A ratio of Ant/(Ant+Phe) less than 0.1 (< 0.1) points to a petroleum source, while a ratio of Ant/(Ant+Phe) greater than 0.1 (> 0.1) indicates the dominance of combustion. Additionally, Fla/(Fla+Pyr) 0.4 is indicative of combustion from a petroleum source, > 0.5 of biomass or coal, and ratios between 0.4 and 0.5 of petroleum sources, according to Yunker et al. [53]. LMW to HMW PAH ratios below 1 imply pyrogenic sources, while above 1 indicates petrogenic sources [52]. Additionally, values between 0.2-0.35 imply a petroleum source, particularly for liquid fossil fuel, vehicles, and crude oil, while values above 0.35 reflect the combustion of coal, grass, and wood. A BaA/(BaA+Chr) value below 0.2 indicates the presence of a petroleum source [6, 8, 54]. It is important to note that DRs were only calculated if the concentrations of the concerned PAHs were quantifiable.

2.7 Toxicity evaluation

The Toxic equivalent quantity (TEQ) and Toxic equivalent factor (TEF) are two terms used to describe the toxic equivalent of a substance. The toxic equivalent factor is the carcinogenic strength of each (C_n) PAH relative to that of Benzo[a]pyrene (BaP) and toxic equivalent quantity (TEQ).

$$\mathrm{TEQ} = \sum C_n \times \mathrm{TEF}_n$$

2.8 Carcinogenicity of Benzo[a]pyrene-equivalent (BaP-EQ)

The toxicity of the seven (7) carcinogenic PAHs in the agricultural soil samples around River Owan, Edo State, Nigeria based on BaP-EQ was determined using their relative potency factor (RPFs) because they have a similar mechanism but differing levels of carcinogenicity. All of the carcinogenic PAH in the soil have been reduced using this technique to a single concentration, known as BaP-EQ. By dividing the concentration of each PAH by its RPF based on BaP-EQ, it is possible to quantify the risk of direct exposure to PAHs in soil [55-57].

$$\sum 7\mathrm{cPAHs} = [\mathrm{DhA}] + [\mathrm{BaP}] + 0.001\,[\mathrm{CHY}] + 0.01\,[\mathrm{BkF}] + 0.1\,[\mathrm{BbF}] + 0.1\,[\mathrm{IP}] + 0.1\,[\mathrm{BaA}]$$

2.9 Lifetime risk assessment

Human health risk lifetime assessment is usually subject to several variables like age, lifestyle, health condition, and contact time with the pollutant(s) under consideration. In most cases, incremental lifetime cancer risks (ILCRs) were used to evaluate the estimated lifetime risk of PAHs. Dermal, ingestion, and inhalation are the major routes of impact exposure pathways. Equations 1–4 were used considering the children (0–18 years) and the adults (19–70 years) age groups (2 age groups) in this study. After evaluation, ILCRs $\leq 10^{-6}$ represent a negligible risk, between 10^{-6} and 10^{-4} show low risk, from 10^{-4} to $\leq 10^{-3}$ represents moderate, while $10^{-3} \leq$ ILCRs $\leq 10^{-1}$ shows high and $\geq 10^{-1}$ represents very high risk [58-60].

$$BaPeq = \sum \mathrm{Ci} \times \mathrm{TEF} \tag{1}$$

$$ILCRs(Dermal) = \frac{\mathrm{CS} \times \left(\mathrm{CSF\ (Dermal)} \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)}\right) \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{BW\ x\ AT\ x\ 10^6} \tag{2}$$

$$ILCRs(Ingestion) = \frac{\mathrm{CS} \times \left(\mathrm{CSF}\ (Ingestion) \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)}\right) \times \mathrm{IR(Ingestion)} \times \mathrm{EF} \times \mathrm{ED}}{BW\ x\ AT\ x\ 10^6} \tag{3}$$

$$ILCRs(Inhalation) = \frac{\mathrm{CS} \times \left(\mathrm{CSF\ (Inhalation)} \sqrt[3]{\left(\frac{\mathrm{BW}}{70}\right)}\right) \times \mathrm{IR(Inhalation)} \times \mathrm{EF} \times \mathrm{ED}}{BW\ x\ AT\ x\ PEF} \tag{4}$$

The incremental lifetime cancer risk assessment uses the following parameters: CS, which is the sum of converted PAHs concentrations for 7 carcinogenic PAHs based on toxic equivalents of BaP using the toxic equivalency factor (TEF) as per Nisbet and LaGoy [61], CSF, which is the carcinogenic slope factor $(\mathrm{mg/kg/day})^{-1}$, BW, which is the body weight of the exposed resident or person (kg), AT, which is the average lifespan in years, SA is the surface area of dermal exposure (cm^2), $\mathrm{IR}_{\mathrm{Ingestion}}$ is the soil ingestion rate (mg/day), $IR_{\text{Inhalation}}$ is the inhalation rate (m^3/day), dermal adherence factor for soil is AF measured in mg/cm^2/h, dermal adsorption factor is ABS, and particle emission factor (PEF) measured in m^3/kg. The value for CSF ingestion is 7.30 $(\text{mg kg}^{-1}\ \text{day}^{-1})^{-1}$, CSF dermal is 25 $(\text{mg kg}^{-1}\ \text{day}^{-1})^{-1}$, and CSF inhalation is 3.85 $(\text{mg kg}^{-1}\ \text{day}^{-1})^{-1}$ of BaP based on the cancer-causing potency of BaP according to Bhutto et al. [20].

2.10 Quality control and assurance (QC/QA)

After startup, the GC was air-flushed to clean the column and get ready for new analysis, while autotune and air-water checks were carried out and passed to ensure that there are no leakages. A QC standard is usually incorporated into every batch of ten (10) samples set up for analysis along with a solvent blank. Acetone (99.8% purity), dichloromethane (99.5%), n-hexane (99.8%), and acetonitrile, were all GC grade solvents. Sodium chloride and anhydrous sodium sulfate were of analytical grade and of high purity. The result of the blanks did not show the presence of target compounds. The calibration curves for the standards showed correlations that were between 0.997 to 1.000 co-efficient (r^2) translating to a perfect linearity. The PAHs surrogate recovery studies were in the range of 80.7 to 97.8%, while the spiked samples were in the range of 76.7 to 93.5%. The limits of detection and quantitation (LOD and LOQ) were calculated by multiplying the signal-to-noise ratio of the examined blanks by 3 for LOD and 10 for LOQ.

2.11 Statistical analysis

Minitab statistical package was used for descriptive analysis of the data generated while the Excel package was used for the generation of various distribution charts.

3. Results and discussion

3.1 Soil samples PAHs concentration and distribution

Table 3 shows the levels of the various PAH components in the soil samples (LMW, HMW, 1-methylnaphthalene, and 2-methylnaphthalene). The concentration of the LMW PAHs which are naphthalene, acenaphthene, fluorene, phenanthrene, and anthracene are in this range: 0.036 – 0.093, 0.005 – 0.032, below the detection limit (BDL) – 0.035, 0.028 – 0.046, BDL – 0.010µg/kg respectively. Their mean values are 0.070, 0.018, 0.018, 0.033, and 0.005 µg/kg. Acenaphthalene indicated concentration in the soil samples from all the sampling locations to be below the instrument's detection threshold (BDL). Anthracene was below the limit of detection from sampling location 1. The ∑LMW PAHs' highest concentration is from sampling location 4 (SO4) at 0.205 µg/kg while the least is from sampling location 6 (SO6) at 0.094 µg/kg (Figure 2). A study conducted by Chen et al. [6] evaluated the health risk and the contamination levels of PAHs in farmland soil located at China, the Yinma River Basin, which revealed PAHs in varying concentrations 15.13 – 81.06, 7.13 – 51.30, and 21.50 – 75.40 ng/g for naphthalene in May, August, and November 2016 while anthracene was in the range of 8.01 – 57.10, 15.20 – 157.23 and 12.50 – 42.90 ng/g for the same specific months. Other LMW PAHs such as fluorene, acenaphthylene, and phenanthrene were detected in varying concentrations from month to month with all their concentrations higher than that of this study. High concentration of pollutants in the soil or environmental samples generally is a function of so many factors such as regulatory policies in the region under study, awareness of such pollutants, degradation process, previous usage, and or continuous usage of facilities that produce and release these pollutants into the environment [8, 62, 63].

Table 3 PAHs in soil samples

PAHs/Locations	SO1	SO2	SO3	SO4	SO5	SO6	SO7	SD	TEF
Naphthalene	0.083	0.084	0.064	0.092	0.093	0.038	0.036	0.002	0.001
Acenaphthylene	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.001
Acenaphthene	0.026	0.007	0.005	0.032	0.032	0.015	0.006	0.001	0.001
Fluorene	0.023	0.023	0.006	0.035	0.021	BDL	0.018	0.001	0.001
Phenanthrene	0.038	0.028	0.028	0.046	0.030	0.031	0.030	0.001	0.001
Anthracene	BDL	0.006	0.008	BDL	0.008	0.010	0.005	0.001	0.01
∑ LMW	**0.17**	**0.148**	**0.111**	**0.205**	**0.184**	**0.094**	**0.095**	-	-
Pyrene	0.021	0.010	0.013	0.024	0.022	0.013	0.011	0.001	0.001
Fluoranthene	BDL	BDL	BDL	0.031	BDL	BDL	BDL	BDL	0.001
Benzo(a)anthracene*	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.1
Chrysene*	BDL	BDL	BDL	BDL	0.012	BDL	BDL	BDL	0.01
Benzo(k)fluoranthene*	0.039	0.008	0.021	0.054	0.024	0.027	0.034	0.001	0.1
Benzo(b)fluoranthene*	0.029	0.004	0.004	0.009	0.008	0.014	0.013	0.003	0.1
Benzo(a)pyrene*	0.087	0.031	BDL	0.079	BDL	0.001	0.001	0.004	1
Benzo(g,h,i)perylene*	0.054	0.026	0.007	0.093	0.086	0.057	0.073	0.004	0.01
Dibenzo(a,h)anthracene*	0.036	BDL	BDL	BDL	BDL	BDL	BDL	BDL	1
Indeno(1,2,3-cd)pyrene	0.082	0.022	0.042	BDL	BDL	0.141	BDL	0.005	0.1
∑ HMW PAHs	**0.348**	**0.101**	**0.087**	**0.29**	**0.152**	**0.253**	**0.132**	-	-
∑16 PAHs	**0.518**	**0.249**	**0.198**	**0.495**	**0.336**	**0.347**	**0.227**	-	-
1-methylnaphthalene	0.026	0.039	0.025	0.034	0.034	BDL	0.021	0.002	-
2-methylnaphthalene	0.014	0.019	0.012	0.019	0.026	0.010	0.014	0.003	-
∑1&2-methylnaphthalene	**0.04**	**0.058**	**0.037**	**0.053**	**0.06**	**0.01**	**0.035**	-	-
∑7cPAHs	**0.245**	**0.069**	**0.032**	**0.235**	**0.13**	**0.099**	**0.121**	-	-
Total ∑PAHs(ug/kg)	**0.56**	**0.31**	**0.23**	**0.55**	**0.40**	**0.36**	**0.26**	-	-
∑TEQ	**0.139**	**0.035**	**0.007**	**0.087**	**0.004**	**0.020**	**0.007**	-	-

PAHs* - carcinogenic PAHs, TEQ – Toxicity equivalent quantity, TEF – Toxicity equivalent factor, SD – Standard deviation.

The HMW PAHs which include pyrene, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene, benzo[g,h,i]perylene, and indeno[1,2,3-cd]pyrene which are in the range of: 0.010 – 0.024, 0.008 – 0.054, 0.004 – 0.029, BDL – 0.087, 0.007 – 0.093 and BDL – 0.082 µg/kg respectively. Their mean concentration for pyrene, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene, and benzo[g,h,i]perylene are 0.016, 0.030, 0.012, 0.028, and 0.057 µg/kg. Benzo[a]anthracene was below the detection limit in all the soil sampling locations. Fluoranthene was only detected in sampling location 4 (SO4) with 0.031µg/kg as the concentration value. Chrysene was only detected from sampling location 5 (SO5) having a value of 0.012 µg/kg. Dibenzo[a,h]anthracene was detected at a concentration of 0.036 µg/kg from sampling location 1 (SO1) out of all the seven sampling stations. The ∑HMW PAHs of the soil samples from the seven locations are in the range of 0.018 – 0.348 µg/kg. The highest ∑HMW PAHs are from sampling location 1 (SO1) while the least is from sampling location 3 (SO3) (Figure 3). HMW PAHs were determined by Chen et al. [6] in soil samples from the farmland of Yinma River Basin, China in concentrations range: pyrene (42.06 – 152.37, 35.00 – 113.25, 52.30 – 156.20 ng/g), BaA (21.11-129.15, 9.31 – 103.20, 21.30 – 117.30 ng/g) and BkF (8.36 – 31.28, 8.15- 30.21, 7.20-35.40 ng/g) these are for May, August, and November. The concentration of 1-methylnaphthalene and 2-methylnaphthalene were in the range of BDL – 0.039 and 0.01 – 0.019 µg/kg while their summation (∑1-methylnaphthalene and 2-methylnaphthalene) range is 0.01 – 0.058 µg/kg. The highest is from sampling location 2 (SO2) and the least is from sampling location 6 (SO6) (Figure 4). Considering the total concentration of all the PAHs components (LMW, HMW, 1-methylnaphthalene, and 2-methylnaphthalene), the highest concentration is from sampling location 1 (SO1) and the least from sampling location 3 (SO3). The concentration ∑PAHs of other sampling locations are 0.310 µg/kg (SO2), 0.550 µg/kg (SO4), 0.400 µg/kg (SO5), 0.360 µg/kg (SO6) and 0.260 µg/kg (SO7) respectively. The Concentration of ∑PAHs is in this order: SO1>SO4>SO5>SO6>SO2>SO7>SO3 in soil samples from the locations. Figure 5 show the distribution of ∑LMW, ∑HMW, ∑1and2-methylnaphthalene, ∑16PAHs, ∑7cPAHs, and Total ∑PAHs.

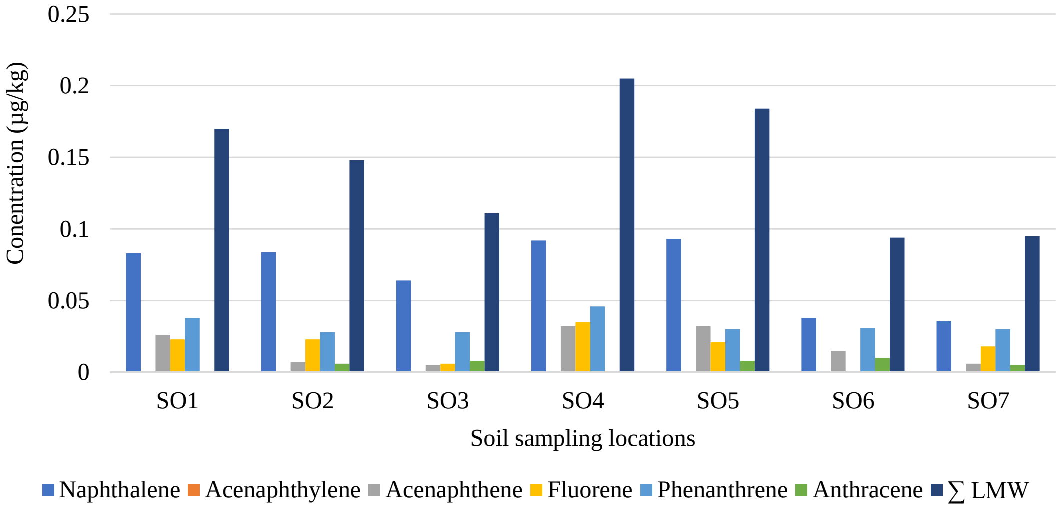

Figure 2 Concentration and summation of ∑LMW PAHs

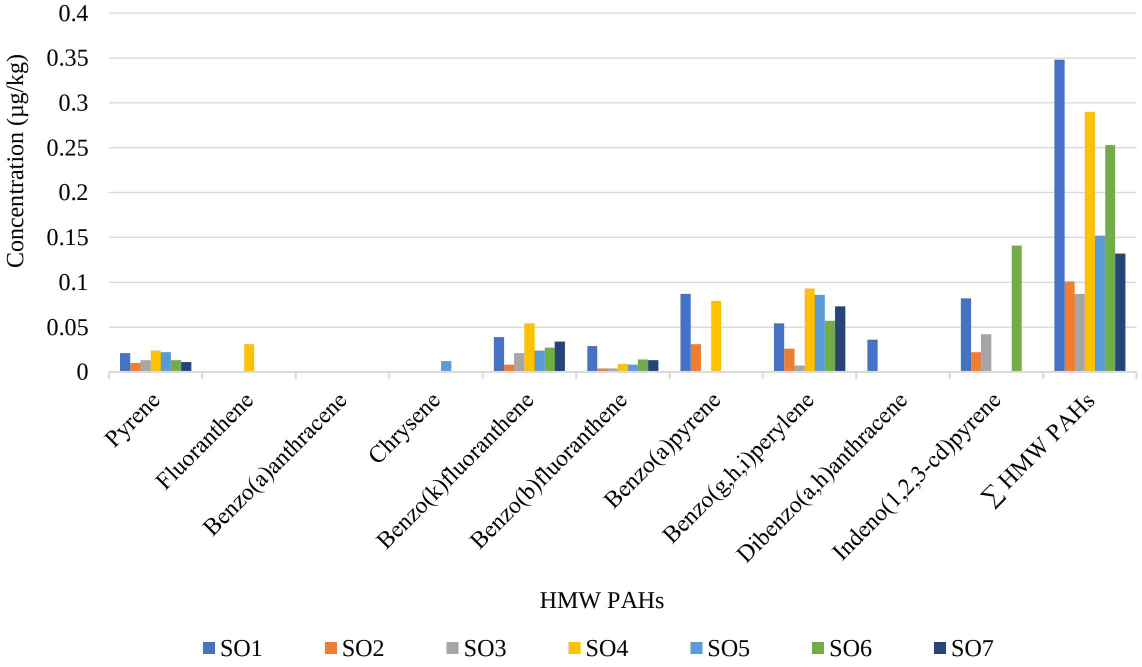

Figure 3 Concentration and ∑HMW PAHs

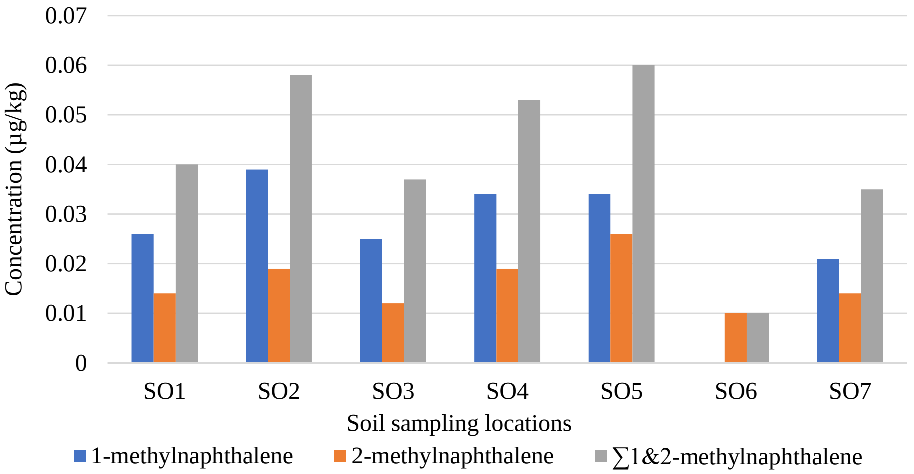

Figure 4 Concentration and ∑ 1&2 methlynapthalene

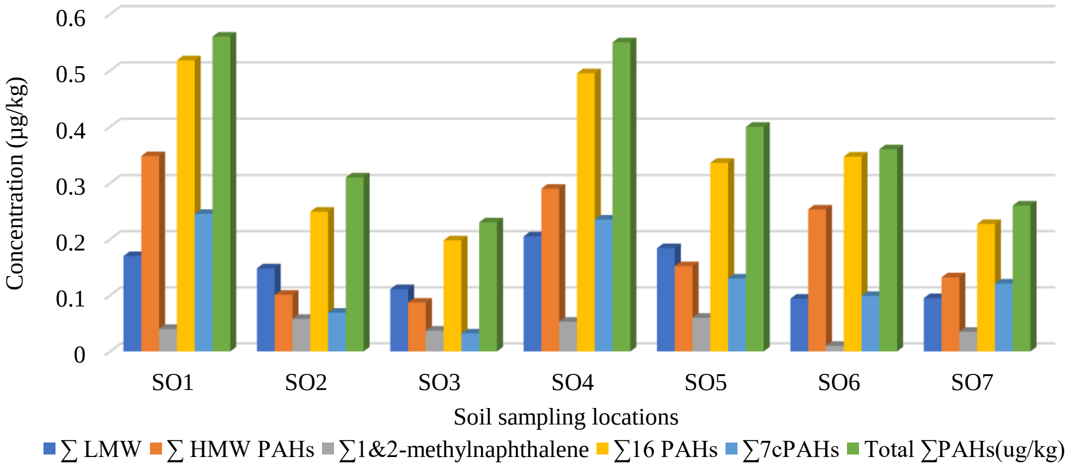

Figure 5 ∑LMW, ∑HMW, ∑1&2-methylnaphthalene, ∑16PAHs, ∑7cPAHs, and Total ∑PAHs

3.2 Toxicity evaluation

The mean concentration of 7cPAHs is 0.133 g/kg, and the concentrations range from 0.032 to 0.245 g/kg. The BaP-EQ of the soil samples from the locations is 0.285 µg/kg is lower than 14.95 µg/kg which was reported by Ding et al. [64] of agricultural soils from Northwest Fujian, Southeast China. The % ratio of ∑7cPAHs to ∑16PAHs is 37%, meaning that the 7cPAHs contribute less % to the total 16 priority PAHs indicating negligible toxicity.

Table 4 Incremental Life Cancer Risk (ILCR) Assessment

Routes/Age groups	SO1	SO2	SO3	SO4	SO5	SO6	SO7	Mean	Max.	Min.	RIs
Dermal (Children)	6.46E-06	1.67E-06	6.43E-08	4.40E-06	4.74E-07	4.11E-07	4.88E-07	2.00E-06	6.46E-06	6.43E-08	Negligible
Dermal (Adult)	2.44E-06	6.31E-07	2.43E-08	1.66E-06	1.79E-07	1.55E-07	1.84E-07	7.54E-07	2.44E-06	2.43E-08	Negligible
Ingestion (Children)	2.59E-06	6.71E-07	2.58E-08	1.77E-06	1.90E-07	1.65E-07	1.96E-07	8.01E-07	2.59E-06	2.58E-08	Negligible
Ingestion (Adult)	1.37E-06	3.55E-07	1.37E-08	9.36E-07	1.01E-07	8.73E-08	1.04E-07	4.24E-07	1.37E-06	1.37E-08	Negligible
Inhalation (Children)	1.10E-10	2.84E-11	1.09E-12	7.47E-11	8.03E-12	6.96E-12	8.27E-12	3.38E-11	1.10E-10	1.09E-12	Negligible
Inhalation (Adult)	9.32E-11	2.41E-11	9.27E-13	6.35E-11	6.83E-12	5.92E-12	7.03E-12	2.88E-11	9.32E-11	9.27E-13	Negligible

SO1 to SO7 – Soil sampling locations, Min. – Minimum, Max. – Maximum, RIs – Risk Implications

The ILCR results are shown in Table 4. The value for the children is 6.43 x 10^{-8} to 6.46 x 10^{-6}, 2.58 x 10^{-8} to 2.59 x 10^{-6}, and 1.09 x 10^{-12} to 1.10 x 10^{-10} respectively while for the adult is 2.43 x 10^{-8} to 2.44 x 10^{-6}, 1.37 x 10^{-8} to 1.37 x 10^{-6}, and 9.27 x 10^{-13} to 9.32 x 10^{-11} for dermal, ingestion and inhalation exposure routes respectively. The risk implication evaluated showed negligible risk. A study by Bhutto et al. [20] evaluated the ILCRs of sediment soil samples of Daye Lake, China reported values in the range of 10^{-6} to 10^{-12} implying negligible risk in both children and adult age groups. The PAHs ILCRs of this study do not pose any risk at present. According to Zhang et al. [18], it has been reported that children are more susceptible to cancer than adults.

3.3 The diagnostic ratio and possible sources of PAHs in the soil

Table 5 showed the diagnostic ratio of soil samples from the study site, along with their likely sources (petrogenic or pyrogenic source). Looking at the ratio of Ant/(Ant + Phe), the percentage of petrogenic to pyrogenic ranges from 43 to 57 percent. The ratio of Flt/(Flt + Pry) is higher than the 0.1 value, revealing that they are of pyrogenic origin. LMW PAHs account for 43 percent to 57 percent of total PAHs. Petrogenic PAHs are distinguished by a high proportion of 2-3 ring LMW PAHs, whereas pyrogenic PAHs are distinguished by a high percentage of 4-ring HMW PAHs [47]. According to Wu et al. [65] and Tavakoly Sany et al. [66], if the ratio of LMW to HMW is lower than one (<1), pollution of pyrolytic origin is likely. According to Tavakoly Sany et al. [66], microbial impact and degradation are responsible for HMW PAH resistance, which results in a low LMW/HMW ratio. They also discovered that the high volatility and solubility of LMW could contribute to a low LMW/HMW ratio. The Flt/(Flt + Pry) ratio can also be employed as a PAH origin indicator. The petrogenic source has a Flt/(Flt + Pry) ratio of less than 0.4, but pyrogenic sources such as wood and coal combustion have a ratio greater than 0.5 [37, 66-68]. The ratio of Flt/(Flt + Pry) >0.5 in this investigation implies a pyrogenic source.

Table 5 PAH diagnostic ratios from various locations

Locations/Ratios	**Ant/(Ant + Phe)**		**Flt/(Flt + Pry)**		**LMW /HMW**	
SO1	0	Petrogenic	0.6	Pyrogenic	0.5	Pyrogenic
SO2	0.2	Pyrogenic	0.7	Pyrogenic	1.5	Petrogenic
SO3	0.2	Pyrogenic	0.7	Pyrogenic	1.3	Petrogenic
SO4	0	Petrogenic	0.7	Pyrogenic	0.7	Pyrogenic
SO5	0.2	Pyrogenic	0.6	Pyrogenic	1.2	Petrogenic
SO6	0.2	Pyrogenic	0.7	Pyrogenic	0.4	Pyrogenic
SO7	0.1	Petrogenic	0.7	Pyrogenic	0.7	Pyrogenic

Ant – Anthracene, Phe - Phenanthrene, Flt – Fluoranthene, Pry – Pyrene, LMW – Low molecular weight, and HMW – High molecular weight, SO1 to SO7 – Soil sampling locations.

4. Conclusion

As a result of the peculiarity of polycyclic aromatic hydrocarbons in the environment and its characteristics to include persistency and toxicity to human, and the contamination of the food chain, this study was carried out. Our study revealed the presence of the LMW and HMW PAHs in the soil samples from the study area. The concentration of the HMW component is higher than the LWM PAHs. The highest value of the LMW PAHs is from sampling location 4 while the least is from sampling location 6. Sampling location 1 contains the highest concentration values of HMW PAHs while the least is from location 3. 1-methylnaphthalene and 2-methylnaphthalene were the most predominant from the sampling locations. The % ratio of ∑7cPAHs to ∑16PAHs is 37%, meaning that the 7cPAHs contribute less % to the total 16 priority PAHs indicating negligible toxicity. This shows that no toxic effect occurrence in this area at present. ILCR assessment implication revealed a negligible risk. The diagnostic ratio suggests more of a pyrogenic source than petrogenic.

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