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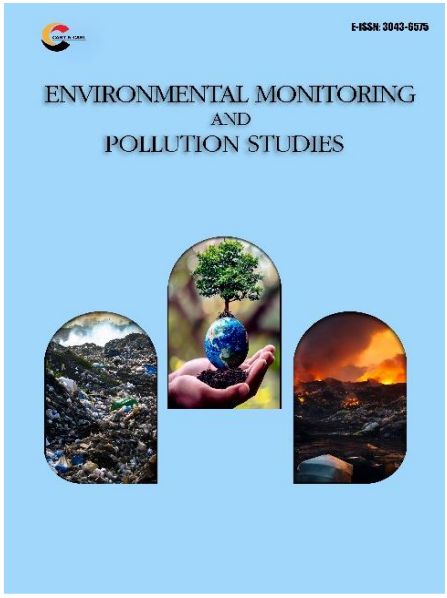

Polycyclic Aromatic Hydrocarbon Concentrations in Environmental Media in Nigeria's Niger Delta Region

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Abstract

The release of pollutants such as Polycyclic Aromatic Hydrocarbons (PAHs) due to oil and gas activities in the Niger Delta region of Nigeria is known to have calamitous environmental and inadvertent human health effects. This study assessed the concentrations of PAHs in soil, food (garri & pumpkin leaves) and water in two selected communities in the Niger Delta. The study was conducted in Eleme, an oil-producing Local Government Area (LGA) in Rivers State and Aboh Mbaise, a non-oil-producing LGA in Imo State. Soil, food and water samples were collected using pre-existing environmental media collection guidelines and sent to the laboratory for extraction and quantification of PAHs using a Gas Chromatography Flame Ionization Detector (GC-FID). The data obtained was analyzed using the Statistical Package for Social Sciences (SPSS) software and statistical significance was set at 0.05. The sample mean concentration of PAHs in soil was 0.31±0.26 (5.03) mg/kg and 0.19±0.18 (2.95) mg/kg, garri was 0.56±0.45 (8.98) mg/kg and 0.26±0.18 (4.17) mg/kg, pumpkin leaf was 0.03±0.02 (0.48) mg/kg and 0.04±0.02 (0.58) mg/kg and water was 0.02±0.02 (0.39) mg/l and 0.05±0.04 (0.72) mg/l for Eleme and Aboh Mbaise respectively. There was a significant difference in the PAHs concentrations in water (p-value: of 0.035) and pumpkin leaves (p-value: 0.018) were also identified. The obtained ΣPAHs from the two communities exceeded the acceptable limits for ΣPAHs set by the United States Environmental Protection Agency. There is a need for the elimination of pyrolytic and petrogenic sources of PAH pollution by concerned environmental health stakeholders.

Keywords: Human Health Risk, Contaminated Food Crops, Farmlands, Artisanal Refining Areas, Niger Delta

Introduction

In the African continent, one of the regions where crude oil is explored is the Niger Delta region of Nigeria which has seen years of oil and gas activities that have served as a huge source of revenue and foreign exchange to Nigeria (Odalonu & Eronmhonsele, 2015). The Niger Delta region of Nigeria has played host to more than 80% of these oil and gas activities (Odalonu & Eronmhonsele, 2015). The exploration and exploitation of crude oil in Nigeria has gone on for years and has largely contributed to the economy and development of the country, at the expense of depletion of the environment and occurrence of health problems(Adeniji et al., 2018; Obenade & Amangabara, 2014). This is a result of the formation and release of various hazardous organic products such as volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), and other inorganic pollutants during oil and gas exploitation activities (Yakubu, 2017).

Polycyclic aromatic hydrocarbons (PAHs) are a large subset of ubiquitous organic compounds that possess the capacity to be widely distributed in

terrestrial and aquatic ecosystems (Boisa et al., 2019; Lawal, 2017). They are highly lipophilic and hydrophobic compounds that are environmentally persistent, hardly biodegradable, carcinogenic, mutagenic and environmentally toxic. The level of toxicity of these organic compounds is largely dependent on their molecular weight as the larger molecular weight PAHs (high MW PAHs) having four to seven aromatic rings are not acutely toxic but possess a greater capacity for carcinogenicity. Examples include fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene, dibenz(a,h)anthracene and inden0(1,2,3-c,d)pyrene. Conversely, the small molecular weight PAHs having two or three aromatic rings are more acutely toxic in and are more volatile, soluble and relatively environmentally mobile than the larger PAHs. Examples include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene and anthracene (Nwaichi et al., 2017; Singh et al., 2016).

Whenever PAHs are released into the environment, they contaminate the water, soil and air of affected areas with the resultant degradation of the environment (Nriagu et al., 2016; Obenade & Amangabara, 2014; Ordinioha & Brisibe, 2013). A Netherland-based review which was conducted by Shell Global Solutions, reported that hydrocarbons released into the environment during illegal oil exploration activities, have the capacity to cause soil and water contamination (Brown et al., 2017). Contamination of vegetables and fruits can occur and is reported to be generally greater on plant surfaces than on the internal components of the plant. Foods of aquatic origin can also be exposed to contamination from PAHs present in water and sediments obtained from atmospheric pollution or oil spills (Srogi, 2007; Zelinkova & Wenzl, 2015).

They can also accumulate in animals, and humans indirectly through the food chain and can cause various health problems (Dum-Awara et al., 2023; Ogbuagu et al., 2011; Okuroemi et al., 2023; Olayinka et al., 2019). Bioaccumulation of PAHs has been reported in fish and invertebrates obtained from the Lagos Lagoon in Lagos, Nigeria. Typically, high molecular weight PAHs are known to bioaccumulate more than Low molecular weight PAHs, and can exert their harmful effects in the body (Rose et al., 2012). Thus, the extensive exposure to these compounds is an issue of great public health concern requiring a holistic approach to tackling it (Almeda et al., 2013; Brandt & Einhenkel-Arle, 2016; Ozoani et al., 2020). Health risks associated with PAHs are also dependent on the route of entry, intensity of exposure, age, duration of exposure, gender, individual susceptibility and immune system capabilities (Brandt & Einhenkel-Arle, 2016; Onanuga & Onanuga, 2014).

In Rivers State, especially in Ogoniland, PAHs have been identified to be 1000 times higher than permissible limits in air, water, and soil (Anyanwu et al., 2018; Onanuga & Onanuga, 2014; Yabrade & Tanee, 2016). Inhabitants of these communities are thus predisposed to high and chronic exposure to PAHs through ingestion, inhalation and adsorbtion portals (Ephraim-Emmanuel & Ordinioha, 2021; Kponee et al., 2015; Lawal, 2017). Though previous research has shown problems relating to concentrations of PAHs in environmental media, it is essential to constantly monitor its levels to ensure that it doesn't exceed safe limits for environmental safety, human habitation and consumption (Amangabara & Njoku, 2012; Ephraim-Emmanuel et al., 2022b; Kadafa, 2012; Odalonu, 2015; Odalonu & Eronmhonsele, 2015). To provide an up-to-date assessment of PAHs concentrations in environmental media within the Niger Delta region of Nigeria, it became necessary to conduct this study.

Materials and Methods

This study adopted a descriptive, comparative design and was conducted in Eleme, an oil-producing Local Government Area (LGA) in Rivers State and Aboh Mbaise a non oil-producing LGA in Imo State; both within the Niger Delta region of Nigeria.

The formula for the sample size for comparing two means was used in calculating the sample size for environmental samples used for this study (Pandis, 2012). The mean and standard deviation (2.12±3.53 µg/l) of the attribute of interest (PAHs concentration in water) in a non-Niger Delta community was obtained from the study by Adekunle et al. (2020) at Ife North in Osun state (Adekunle et al., 2020). Also, the mean and standard deviation (0.27±0.32 µg/l) of the attribute of interest (PAH concentration in water) in a Niger Delta community was obtained from the study conducted by Aigberua (2020) in Bayelsa state (Aigberua, 2020). These were used to arrive at a total sample size of 44 used in this study.

Environmental assessment of the PAHs levels in surface soil, food (garri & pumpkin leaves) and water was done using the gas chromatography/flame ionization detector (GC/FID) method as described by Aigberua (Aigberua, 2020). Eleven samples of each of these four environmental media were obtained and involved in this assessment with 6 samples obtained from communities

in Eleme, Rivers state and 5 samples from communities in Aboh Mbaise, Imo state. Topsoil samples were collected from the communities using a stainless soil auger and placed in pre-labeled bags for transport to the laboratory, pumpkin leafs samples were obtained from different farm locations while garri was obtained from local garri producers within communities in the study sites. Water samples were collected from different wells and taps in the communities as well, using previously cleaned one litre capacity glass bottles. These samples were immediately delivered to Endpoint Laboratories & Equipment Limited, Port Harcourt, Rivers State, Nigeria; for gas chromatographic analysis of the 16 priority PAHs. This was done using the Gas Chromatographic System - (HP Agilent 7890A Gas Chromatography) equipped with a Flame Ionization Detector based on the USEPA method 8100 (United-States-Environmental-Protection-Agency, 1998).

Extraction and quantification of PAHs from soil and food samples

The extraction method described in the operating guide of the Agilent 7890A Gas Chromatograph was applied for the extraction of PAHs in the soil, garri and vegetable samples (Agilent Technologies, 2010). The vegetable samples were washed with distilled water and chopped into pieces while the soil and garri samples were properly ground using the laboratory grinder. Ten grammes of each well ground sample was transferred into a closed glass vial; whose mouth was wiped with a clean tissue paper and covered. For the samples in each batch, a matrix spike sample was selected and 1ml of the matrix spiking standard was added. One ml of appropriate surrogate spiking solution was also added to selected samples, quality control samples and blanks. Drying of the wet samples which did not have a free-flowing sandy texture was done using anhydrous sodium sulphate. Immediately Dichloromethane (DCM): Acetone mixture (1:1 v/v) was added to bring the total volume to 10ml considering the initial volumes of surrogate and matrix spiking solutions added. Extraction was then done by sonicating the samples and the solvent for 15 minutes. Also, filtration of the extract was carried out using a Whatman filter paper into a glass vial. The procedure was then repeated using 10ml of DCM: Acetone mixture (1:1) on the same extracted sample until the sample was exhaustively extracted. The extract was then concentrated to 2ml by passing it through a gentle stream of nitrogen gas, after which the extract was now ready for clean-up.

Extraction was done using the Extract Clean-up (3630C)-Silica Gel Technique. Firstly, the extract solvent was exchanged to cyclohexane by adding 4ml of cyclohexane following the concentration of the extract to 1-2mL. A slurry of 10g activated silica gel in DCM was placed into a chromatographic column; which was tapped to settle the silica gel and elute the dichloromethane. One to two centimeters of anhydrous sodium sulphate was then added to the top of the silica gel and the column was pre-eluted with 40mL of pentane and the eluate discarded.

Just before exposure of the sodium sulfate layer to the air, 2ml cyclohexane sample extract was transferred into the column using an additional 2ml cyclohexane to complete the transfer. Also, 25 ml of pentane was added and elution continued into a flask. This was then concentrated into 2ml volume by passing through a gentle stream of nitrogen gas collected into a vial bottle and capped as PAH extract.

With the use of a hypodermic syringe, exactly 1 µL portion of the reconstituted extract was introduced into the injection port of the gas chromatograph-flame ionization detector (GC-FID). PAH components were eluted through the capillary column based on their boiling points (BP) and molecular weights (MW). Standard pre-set operating conditions of the GC-FID included an initial oven temperature of 65ºC, final oven temperature of 320ºC, injector temperature of 275ºC, an inlet pressure of 14.8 psi as well as an inlet condition set to split. Others include setting the detector temperature at 310ºC, nitrogen flow amount of 30 mL/ min, hydrogen flow amount of 35 mL/min, and an airflow rate of 250 mL/min (Aigberua & Seiyaboh, 2021; Ephraim-Emmanuel et al., 2022a).

Extraction and clean-up of PAHs from water samples

The liquid-liquid extraction technique was applied to extract PAHs in the surface water samples using the method described in the operating guide of the Agilent 7890A Gas Chromatograph and that applied in a previous study (Agilent Technologies, 2010; Ephraim-Emmanuel et al., 2022a). Firstly, the 250 mL water sample was homogenized before emptying the entire volume into a 500 mL separating funnel. Afterward, PAHs were extracted by a three-batch extraction process using 20 mL of dichloromethane (DCM)/n-hexane (1:3 v/v) mixed solvents at each time. The sample-solvent mixture in the separating funnel was vigorously agitated with intermittent ejection of built-up pressure from the tap of the glass funnel. This was done to eliminate the risk of blowing up the glass material. Thereafter, the organic extract was dehydrated by filtering through anhydrous sodium sulfate. Organic contaminants in filtered extracts were cleaned by eluting through a 10 mm I.D (internal diameter) x 250 mm long chromatographic column

packed with glass wool, a slurry of silica gel, and anhydrous sodium sulfate. The cleaned-up extract was reconstituted to about 1.0 mL, after being concentrated in a temperature-regulated water bath at 35 – 40°C. Finally, sample extracts were transferred into glass vials with rubber-crimped caps. Another 250 mL portion of the water sample was transferred into a separating funnel and spiked with a pre-deuterated PAHs mixture (naphthalened8, phenanthrene-d10, chrysene-d12 and perylene-d12) as internal standards, to establish the efficiency of the extraction protocol. The recovery rates ranged between 92% and 107%. Exactly 20 mL of dichloromethane (DCM)/n-hexane (1:3 v/v) mixed solvents were added to the sample mixture, thoroughly mixed and kept standing to allow for adequate phase separation before dehydration and filtration, followed by clean-up and elution through a chromatographic column. Afterward, the eluted extracts were concentrated to 1.0 mL volume and stored in air-tight rubber-crimp cap glass vials (Aigberua, 2020; Ephraim-Emmanuel et al., 2022a).

Quantification of PAHs from water samples

Exactly 1µL portion of the reconstituted extract was injected into the gas chromatograph-flame ionization detector (GC-FID) using a hypodermic syringe. Nitrogen served as the carrier gas while a combination of hydrogen and air was used to create an ionization environment at the detector head. The various fractions of the aromatic compounds were automatically detected at the FID (whose response is dependent on the composition of the eluted vapor) as they emerged from the column. Results were expressed in µg/L units. Standard pre-set operating conditions of the GC-FID were ensured (Aigberua, 2020; Ephraim-Emmanuel et al., 2022a).

The instrument conditions above are based on manufacturer recommendations and PAHs method suitability for repeatability of analytical data on the HP 6890 Plus GC-FID, version A.03.08. Quality assurance/quality control (QA/QC) parameters applied during GC-FID analysis included the spike concentration, concentration obtained, percentage recovery, limit of detection (LOD), and limit of quantification (LOQ) (Aigberua, 2020). The surface water (SW) matrix was used to calculate extraction recovery efficiency for different PAHs. The instrument limit of detection (LOD) and limit of quantification (LOQ) were also estimated and ranged between 0.001 – 0.04µg/mL and 0.004 – 0.10µg/mL respectively. The acceptable recovery range of the equipment was stipulated between 90 and 110% (Aigberua, 2020; Ephraim-Emmanuel et al., 2022a).

The Statistical Package for Social Sciences (SPSS) version 23 was used to perform both descriptive and inferential analyses. The student's t-test was used to compare the concentrations of PAHs in the soil, garri, vegetable and water samples obtained from the oil-producing and non-oil-producing study sites. All analyses were conducted at a 95% confidence level and a p-value ≤ 0.05 was considered as being statistically significant. Ethics approval for the research was obtained from the Research Ethics Committee of the University of Port Harcourt (Approval number: UPH/CEREMAD/REC/MM80/006). During the collection of samples from the environment, it was ensured that the appropriate techniques were applied and that no harm came to the environment in the course of doing so.

Results and Discussion

Polycyclic Aromatic Hydrocarbons (PAHs) concentrations in soil

Assessment of the concentrations of the 16-priority PAHs in soil obtained from the two study locations showed that the mean concentration of PAHs in the oil-producing area was 0.31±0.26 mg/kg and total PAHs concentration (ΣPAHs) of 5.03 mg/kg. The mean concentration in soil samples obtained from the non-oil-producing area was 0.19±0.18 mg/kg with ΣPAHs of 2.95 mg/kg. Assessment of the concentrations of the 16-priority PAHs in the garri samples showed that the mean concentration from the oil-producing area was 0.56±0.45 mg/kg and ΣPAHs of 8.98 mg/kg. The mean concentration in garri samples obtained from the non-oil-producing area was 0.26±0.18 mg/kg with ΣPAHs of 4.17 mg/kg. Assessment of the concentrations of the 16-priority PAHs in pumpkin leaf samples showed that the mean concentration in samples obtained from the oil-producing area was 0.03±0.02 mg/kg and ΣPAHs was 0.48 mg/kg. The mean concentration in samples obtained from the non-oil-producing area was 0.04±0.02 mg/kg with ΣPAHs of 0.58 mg/kg. Assessment of the concentrations of the 16-priority PAHs in water samples showed that the mean concentration from the oil-producing area was 0.02±0.02 mg/l and ΣPAHs was 0.39 mg/l. The mean concentration in samples obtained from the non-oil-producing area was 0.05±0.04 mg/l with ΣPAHs of 0.72 mg/l. These are shown in Tables 1 to 4.

Table 1: Mean and total concentrations of PAHs in Soil

PAHs (mg/kg)	Oil-producing community	Non-oil-producing community
Naphthalene	0.00	0.00
Acenaphthylene	0.03	0.01
Acenaphthene	0.00	0.00
Fluorene	0.12	0.12
Phenanthrene	0.43	0.13
Anthracene	0.37	0.23
Fluoranthene	0.24	0.15
Pyrene	0.38	0.18
Benzo(a)anthracene	0.24	0.11
Chrysene	0.69	0.45
Benzo(b)fluoranthene	0.35	0.23
Benzo(k)fluoranthene	0.93	0.66
Benzo(a)pyrene	0.59	0.39
Indeno(1,2,3-cd)pyrene	0.24	0.18
Dibenz(a,h) anthracene	0.15	0.06
Benzo(g,h,i)perylene	0.27	0.07
Total PAHs (ΣPAHs)	**5.03**	**2.95**
Mean ± S.D	**0.31± 0.26**	**0.19 ± 0.18**

USEPA Maximum Contaminant Level for ΣPAHs: ≤ 0.2 mg/kg (Ofori et al., 2020)

Table 2: Mean and total concentrations of PAHs in Garri

PAHs (mg/kg)	Oil-producing community	Non-oil-producing community
Naphthalene	0.00	0.00
Acenaphthylene	0.18	0.15
Acenaphthene	0.05	0.03
Fluorene	0.19	0.13
Phenanthrene	1.76	0.20
Anthracene	0.47	0.26
Fluoranthene	0.81	0.28
Pyrene	0.71	0.26
Benzo(a)anthracene	0.36	0.14
Chrysene	0.62	0.35
Benzo(b)fluoranthene	0.60	0.24
Benzo(k)fluoranthene	1.19	0.72
Benzo(a)pyrene	0.54	0.44
Indeno(1,2,3-cd)pyrene	0.77	0.34
Dibenz(a,h) anthracene	0.26	0.17
Benzo(g,h,i)perylene	0.46	0.44
Total PAHs (ΣPAHs)	**8.98**	**4.17**
Mean ± S.D	**0.56 ± 0.45**	**0.26 ± 0.18**

USEPA Maximum Contaminant Level for ΣPAHs: ≤ 0.2 mg/kg (Rengarajan et al., 2015)

Table 3: Mean and total concentrations of PAHs in Pumpkin Leaves

PAHs (mg/kg)	Oil-producing community	Non-oil-producing community
Naphthalene	0.00	0.00
Acenaphthylene	0.01	0.02
Acenaphthene	0.00	0.01
Fluorene	0.01	0.01
Phenanthrene	0.02	0.03
Anthracene	0.02	0.05
Fluoranthene	0.02	0.04
Pyrene	0.05	0.05
Benzo(a)anthracene	0.02	0.03
Chrysene	0.06	0.07
Benzo(b)fluoranthene	0.05	0.06
Benzo(k)fluoranthene	0.07	0.08
Benzo(a)pyrene	0.03	0.04
Indeno(1,2,3-cd)pyrene	0.04	0.04
Dibenz(a,h) anthracene	0.04	0.04
Benzo(g,h,i)perylene	0.04	0.02
Total PAHs (ΣPAHs)	**0.48**	**0.58**
Mean ± S.D	**0.03 ± 0.02**	**0.04 ± 0.02**

USEPA Maximum Contaminant Level for ΣPAHs: ≤ 0.2 mg/kg (Rengarajan et al., 2015)

Table 4: Mean and total concentrations of PAHs in Water

PAHs (mg/l)	Oil-producing community	Non-oil-producing community
Naphthalene	0.00	0.00
Acenaphthylene	0.00	0.00
Acenaphthene	0.01	0.01
Fluorene	0.01	0.02
Phenanthrene	0.05	0.11
Anthracene	0.05	0.11
Fluoranthene	0.03	0.05
Pyrene	0.05	0.09
Benzo(a)anthracene	0.01	0.02
Chrysene	0.04	0.06
Benzo(b)fluoranthene	0.02	0.06
Benzo(k)fluoranthene	0.01	0.06
Benzo(a)pyrene	0.03	0.05
Indeno(1,2,3-cd)pyrene	0.03	0.05
Dibenz(a,h) anthracene	0.03	0.02
Benzo(g,h,i)perylene	0.01	0.02
Total PAHs (ΣPAHs)	**0.39**	**0.72**
Mean ± S.D	**0.02 ± 0.02**	**0.05 ± 0.04**

USEPA Maximum Contaminant Level for ΣPAHs: ≤ 0.2 mg/kg (Aigberua, 2020; World Health Organization, 2022)

Comparison of Polycyclic Aromatic Hydrocarbons (PAH) concentrations in oil-producing and non-oil-producing areas

Assessment of the PAHs concentrations in the environmental media from the oil-producing and non-oil-producing areas revealed statistically significant differences in the PAHs concentrations in water (p=0.035) and pumpkin leaves (p=0.018). This is shown in Table 5.

Source diagnostic ratios of the PAHs concentration in tested samples

Also, in soil samples obtained from the oil-producing area, source diagnostic ratios gave a Flt/(Flt+Pyr) ratio of 0.39; Ant/(Ant+Phe) ratio of 0.46, and a BaA/(BaA+Chr) ratio of 0.26. In samples obtained from the non-oil-producing area, Flt/(Flt+Pyr) ratio was found to be 0.36; Ant/(Ant+Phe) ratio was 0.51 and BaA/(BaA+Chr) ratio was 0.59. Likewise, in the garri samples obtained from the oil-producing area, source diagnostic ratios gave a Flt/(Flt+Pyr) ratio of 0.53; Ant/(Ant+Phe) ratio of 0.21, and a BaA/(BaA+Chr) ratio of 0.37. In samples obtained from the non-oil-producing area, Flt/(Flt+Pyr) ratio was found to be 0.32; Ant/(Ant+Phe) ratio was 0.63 and BaA/(BaA+Chr) ratio was 0.39. Also, in pumpkin leaves samples obtained from the oil-producing area, source diagnostic ratios gave a Flt/(Flt+Pyr) ratio of 0.34; Ant/(Ant+Phe) ratio of 0.44, and a BaA/(BaA+Chr) ratio of 0.21. In samples obtained from the non-oil-producing area, Flt/(Flt+Pyr) ratio was found to be 0.31; Ant/(Ant+Phe) ratio was 0.79 and BaA/(BaA+Chr) ratio was 0.50. Finally, in water samples obtained from the oil-producing area, source diagnostic ratios gave a Flt/(Flt+Pyr) ratio of 0.80; Ant/(Ant+Phe) ratio of 0.39, and a BaA/(BaA+Chr) ratio of 0.63. In samples obtained from the non-oil-producing area, Flt/(Flt+Pyr) ratio was found to be 0.79; Ant/(Ant+Phe) ratio was 0.25 and BaA/(BaA+Chr) ratio was 0.35. These are shown in Table 6

Table 5: Levels of PAH in food (garri and vegetables), water and soil in the Niger Delta communities in oil and non-oil producing sites

Variables	Site		Student t-test (p-value)
	Oil-Producing (n=24)	Non-oil producing (n=20)	
	Mean ± SD	Mean ± SD	
Water (mg/l)			
Mean ± SD	0.02 ± 0.02	0.05 ± 0.04	2.207 (0.035) *
Pumpkin Leaves (mg/kg)			
Mean ± SD	0.48 ± 0.22	0.58 ± 0.24	2.512 (0.018) *
Garri (mg/kg)			
Mean ± SD	0.56 ± 0.45	0.26 ± 0.18	1.648 (0.110)
Soil (mg/kg)			
Mean ± SD	0.31 ± 0.26	0.19 ± 0.18	0.792 (0.434)

***Statistically significant (p≤0.05)**

Table 6: Source identification of PAHs in environmental media

	Oil-producing	Non-oil-producing
PAHs (soil)	**mg/kg**	**mg/kg**
Flt/(Flt+Pyr) ratio	0.39	0.36
Ant/(Ant+Phe) ratio	0.46	0.51
BaA/(BaA+Chr) ratio	0.26	0.59
Source	More petrogenic	More pyrolytic
PAHs (garri)		
Flt/(Flt+Pyr) ratio	0.53	0.32
Ant/(Ant+Phe) ratio	0.21	0.63
BaA/(BaA+Chr) ratio	0.37	0.39
Source	More pyrolytic	More pyrolytic
PAHs (pumpkin leaves)		
Flt/(Flt+Pyr) ratio	0.34	0.31
Ant/(Ant+Phe) ratio	0.44	0.79
BaA/(BaA+Chr) ratio	0.21	0.50
Source	More petrogenic	More pyrolytic
PAHs (water)		
Flt/(Flt+Pyr) ratio	0.80	0.79
Ant/(Ant+Phe) ratio	0.39	0.25
BaA/(BaA+Chr) ratio	0.63	0.35
Source	More pyrolytic	More pyrolytic

Key: Flt/(Flt+Pyr) ratio > 0.5 shows more input of PAHs from pyrolytic sources, Ant/(Ant+Phe) ratio > 0.1 shows more input of PAHs from pyrolytic sources, BaA/(BaA+Chr) ratio > 0.35 shows more input of PAHs from pyrolytic sources

Polycyclic Aromatic Hydrocarbons (PAHs) were found to exist in the four environmental media assessed in this study from both the oil-producing and non-oil-producing areas. The total PAHs (∑PAHs) concentration in all the media was found to exceed the guideline limits of PAHs for each tested medium as stipulated by environmental

regulatory agencies (Aigberua, 2020; Honda & Suzuki, 2020; Oyedeji et al., 2016; Patel et al., 2020).

In the soils, the total concentration of the 16-priority PAHs obtained from both the oil-producing and non-oil-producing areas exceeded the stipulated safe limits of PAHs in soil.(Rao & Kumar, 2015) Applying source diagnostic ratios however revealed that the PAHs in the soil from the oil-producing area were more from petrogenic sources whereas the soil PAHs present in the non-oil-producing area were more from pyrolytic sources, for example bush burning, coal use and so on (Aigberua, 2020; Ephraim-Emmanuel et al., 2022a). The finding of petrogenic PAHs contamination in this study is similar to the findings of other authors who have reported elevated PAHs levels in soils from areas where oil exploratory activities are carried out (Aigberua, 2020; Chikere et al., 2020; Kuch & Bavumiragira, 2019; Nwaichi et al., 2014). Very high concentrations of PAHs have also been reported in crude oil-polluted soils obtained from an illegal oil exploratory site in Tombia in Rivers State (Chikere et al., 2020). Other studies are also in agreement with the finding of elevated PAHs levels of mostly pyrolytic sources of PAHs made in this study (Ephraim-Emmanuel et al., 2022a; Nieuwoudt et al., 2011). The use of biomass fuels for cooking has also been described as a potential source of PAHs release and subsequent contamination of the environment (Alexander et al., 2018). The implication of these findings of PAHs-polluted soils is the consequent adverse effect on soil biodiversity, fertility, and vitality and a resultant depletion of the soil and vegetation in affected areas (Douglas & Cornelius, 2018; Yabrade & Tanee, 2016). Furthermore, in areas where the major source of livelihood is agriculture; this pollution of the soil can result in the loss of livelihoods as well as a reduction of food production (Bede-Ojimadu & Orisakwe, 2020; Ephraim-Emmanuel et al., 2022b; The Strategic Partnership on Lobby & Advocacy Programme in Nigeria, 2020). These biomass fuels from wood, animal dung, and so on are used by a large proportion of populations in Africa despite the supplementation by fossil fuels and electricity in this modern era (Bede-Ojimadu & Orisakwe, 2020). They also remain a major source of energy for heating, cooking and other purposes in developing countries (Sola et al., 2017). It is crucial to provide alternatives to the use of these forms of energy in areas where they are known to be frequently used to avoid the risk of potential health effects that could arise when foods obtained from PAHs contaminated soils are ingested and metabolized in the human body (Adeniji et al., 2019).

The concentration of ∑PAHs compounds in foods in this study was also identified to exceed the normal stipulated limits (Aigberua, 2020; Honda & Suzuki, 2020; Oyedeji et al., 2016; Patel et al., 2020) The PAHs in garri were however found to be predominantly from pyrolytic sources in both oil-producing and non-oil-producing areas. This finding is also in agreement with the findings of other authors whom have reported high levels of PAHs contamination in staple foods taken in Nigeria (Abiona et al., 2019; ATSDR, 2005; Ogbuagu & Ayoade, 2012). This kind of contamination from pyrolytic sources of PAHs in food has been described to be related to the use of biomass fuels for cooking, bush burning activities on farms which exposes surrounding vegetation (including unharvested pumpkin leaves) to PAHs contamination (Alexander et al., 2018; Okonkwo et al., 2018). Contamination of the waxy surface of vegetables and fruits can occur when low molecular mass PAHs attach and accumulate on them via surface adsorption (Zelinkova & Wenzl, 2015). Food processing methods have also been implicated as possible routes of exposure of food to PAHs contamination (Ingenbleek et al., 2019; Tiwo et al., 2019). An example, is the processing of cassava to produce garri, which is usually fried by local means that utilize wood or charcoal stoves (Bede-Ojimadu & Orisakwe, 2020; Owili et al., 2017). Smoking of food also has the propensity of causing the release of carcinogenic PAHs into the environment which have the potential to cause cancers in humans and animals (Orodu & Sunny, 2018).

The PAHs in pumpkin leaves assessed in the present study were however identified to be predominantly sourced from petrogenic sources in the oil-producing area but from pyrolytic sources in the non-oil-producing area. Food substances can thus also become polluted by PAHs from petrogenic sources as a result of crude oil exploratory activities as well as products of these activities. These PAHs compounds can be present in the air, soil, or water and have the ability to affect the nutritional potential of such food products (Ipeaiyeda et al., 2015; Itodo et al., 2018).

The concentration of ∑PAHs compounds in water obtained from the oil-producing and non-oil-producing areas in this study was also found to exceed the required maximum contaminant level (MCL) of total PAHs to be found in water (Aigberua, 2020; United States Environmental Protection Agency, 2001). It was further revealed that the PAHs in the water samples were mostly from pyrolytic sources. This finding agrees with the findings of other authors who reported PAHs compounds from mostly pyrolytic sources, which were present in

surface waters assessed in Imiringi, Bayelsa State, and in Shitou Koumen, China (Aigberua, 2020; Sun et al., 2015) The predominant contamination of pyrolytic-sourced PAHs in water bodies has been described to mainly occur among households in rural areas in developing countries. This is due to their exclusive reliance on burning wood for their cooking and heating needs (Bede-Ojimadu & Orisakwe, 2020). Other sources of pyrolytic-sourced PAHs compounds have also been reported to include waste incineration, dumping unsegregated waste along river banks, and biomass combustion among others (Aigberua, 2020; Lawal, 2017; Sun et al., 2015) The environmental and epidemiologic effects of pollution from pyrolytic sources can thus not be overlooked considering that they are also capable of evoking adverse consequences on environmental life and human health.

Conclusion

Polycyclic Aromatic Hydrocarbons (PAHs) were found to exist in the four environmental media assessed in this study from both the oil-producing and non-oil-producing areas. The total PAHs concentration in each of the assessed media was found to exceed the guideline limits stipulated by environmental regulatory agencies. The PAHs concentrations in water and pumpkin leaves from both areas also differed significantly.

Recommendations made for policymakers included that when conducting their oversight functions, there is the need for rigidly ensuring adherence to regulations promoting environmental health and safety during oil and gas exploration and exploitation in the Niger Delta. It was also recommended for policymakers to formulate policies that encourage the use of environmentally safe methods for transportation, cooking, energy and waste management. For real-life applications, the need for regular environmental monitoring by the concerned stakeholders in oil-producing regions was recommended. It was also advised that more effort be put into tackling the pyrolytic sources of PAHs pollution of the environment by environmental regulatory agencies alongside action to tackle the petrogenic sources of these compounds. There is a need for the government to provide an enabling environment that promotes the availability and accessibility of environmentally safe options for transportation e.g. electric vehicles, power supply e.g. solar energy, cooking as well as waste management.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Credit Authorship Contribution Statement

Beauty, MT. and Ephraim-Emmanuel, B. C.: Conceptualization, Methodology, Formal analysis, Investigation, Resources, Data curation, Visualization, Project administration, Writing - original draft, Validation, Review & Editing.

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