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Research paper

Distribution of polycyclic aromatic hydrocarbons in some selected tubers grown in farmlands in Alakahia and Eleme communities, Rivers, Nigeria

Okereke Chioma Joy *, Okonkwo Chinedu Joseph

Department of Biochemistry, University of Port Harcourt, Nigeria
**Corresponding author E-mail: chioma.okereke@uniport.edu.ng*

Abstract

The present study investigated the level of polycyclic aromatic hydrocarbon (PAH) in three commonly consumed tubers grown in farmlands in two communities- Alakahia and Eleme, Rivers state, Nigeria. The total PAH concentrations in the tubers ranged from 2.94 ± 0.26 to 70.83 ± 1.56 -µg/kg. Pyrene was the predominant PAH in most of the tuber samples from Alakahia and Eleme communities. The PAH profile in the tuber samples were dissimilar at both communities when compared. The Low molecular weight PAH to High molecular weight PAH ratio (LMW-PAH/HMW-PAH) showed that the high molecular PAH were predominant in the tubers thus suggesting that the source of PAH in the tubers are of pyrogenic origin.

Keywords: *PAH; Tubers; Farmlands; Rivers; Nigeria.*

1. Introduction

Land pollution particularly the contamination of agricultural lands with environmental pollutants such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls etc have far reaching implication on human health. Serious health challenges can develop from consumption of PAHs and/or heavy metal contaminated food. Ingesting such food can seriously deplete some essential nutrients in the body causing a decrease in immunological defenses, intrauterine growth retardation, impaired psycho-social behaviours, disabilities associated with malnutrition and a high prevalence of upper gastrointestinal cancer [1].

PAHs generally occur in complex mixtures which may consist of hundreds of compounds. They are potent atmospheric pollutants, mostly found in oil, coal, and tar deposits, and are also produced as byproducts from the incomplete combustion of these compounds. As a pollutant, they are of concern because some of their compounds have been identified as carcinogenic, mutagenic, and teratogenic [2]. Incomplete combustion, pyrolysis of organic materials by industry, agriculture and traffic, diagenetic alteration of natural organic matter, long-term wastewater irrigation, reused sewage sludge and fertilizer use in agricultural production result in high concentrations of Polycyclic aromatic hydrocarbons (PAHs) in farmland soil [3,4,5]. The United State Environmental Protection Agency (USEPA) and International Agency for Research on Cancer classify seven PAHs (benzo[a]pyrene, benz[a]anthracene, chrysene, benz[b]fluoranthene, benzo[k]fluoranthene, dibenz[a,h]anthracene and indeno[1,2,3-cd]pyrene as probable human carcinogens (USEPA Class B2). Other PAHs may also contribute to carcinogenic risk and should not necessarily be assumed to be non-carcinogens.

According to the report released in 2012 by the Nigerian Bureau of Statistics [6] for consumption pattern in Nigeria, in the South-South region where Rivers state is situated tuber and plantain consumption is 35.09%. Together, they represent approximately one-third of the total foods consumed in the region. There is paucity of data on the polycyclic aromatic hydrocarbon contents of locally consumed tubers in the region.

In view of this, the present study was designed to provide an insight into the current scenario of polycyclic aromatic hydrocarbons contamination in selected tubers consumed by the population at Alakahia and Eleme communities in Rivers state Southern Nigeria.

2. Materials and method

2.1. Study areas

The study areas namely Alakahia and Eleme communities are situated in Obi-Akpor and Eleme Local Government Areas respectively. Obio-Akpor is in the metropolis of Port Harcourt, one of the major centres of economic activities in Nigeria, and one of the major cities of the Niger Delta, located in Rivers State. The Local Government Area covers 260 km^2 and at the 2006 Census held a population of 878,890. Eleme is located east of the Port Harcourt Local Government area. It is in the greater Port Harcourt metropolis. It covers an area of 138 km^2 and at the 2006 Census had a population of 190,884. The Eleme community is home to a number of industries including petrochemicals, oil refining, paint and fertilizer industries.

2.2. Collection of test samples

Fresh samples of three commonly consumed tubers: Cassava (Manihot esculenta), Cocoyam (Colocasia esculenta), Yam (Dioscorea rotundata) were collected from farms in Alakahia and Eleme (precisely Ogale) communities situated in Obi-Akpor and Eleme Local Government Area (LGA) respectively in Rivers state

Nigeria. At each site, three tubers were collected, cleaned and wrapped in aluminum foils. Samples were identified at the Plant and Soil Biotechnology Department in University of Port Harcourt, Nigeria before they were transported to the laboratory for analysis.

2.3. Sample preparation

The peeled tubers were cleaned. The tubers (Cassava, Yam and Cocoyam) samples were chopped into small pieces and then oven dried before they were crushed in a laboratory mortar and sieved using 0.5mm sieve. The cleaned vegetable samples were then ground with blender (National, MX 795N, Japan) and kept in air tight containers prior to extraction process.

2.4. Extraction of samples for PAH determination

2g of samples were weighed into a clean extraction container (50ml beaker). Ten grams of Sodium Sulphate was added and mixed together with the sample through wrist action. 10ml of extraction solvent (dichloromethane) was then added into the sample and mixed thoroughly and allowed to settle. The mixtures were carefully filtered into clean solvent rinsed extraction bottle, using filter paper fitted into Buchner funnels. The extraction were concentrated to 2ml and then transferred for cleanup/separation. The dichloromethane extract was cleaned up by passing through a column packed with anhydrous Na_2SO_4 salt. The resulting extract was concentrated on a rotary evaporator to give an oily residue; which was again dissolved in 1ml CH_2Cl_2 and 1µL was injected into the GC for analysis.

2.5. Gas chromatography analysis

The concentrated hydrocarbon fractions were transferred into labeled glass vials with Teflon rubber clip cap for gas chromatography analysis. 1ul of the concentrated sample was injected by means of hypodermic syringe through a rubber septum into the column. Separation occurred as the vapor constituent partition between the gas and liquid phase. The sample was automatically detected as it emerges from the column (at constant flow rate) by the FID detector whose response is dependent upon the composition of the vapor.

2.6. Chromatographic conditions

The gas chromatography was Hewlett Packed 6890N series, gas chromatography apparatus, coupled with flame ionization detector (FID) (Hewlett Packard, Wilmington, DE, USA), powered with HP Chemstation Rev. A 09:01 (10206) software to identify and quantify compounds. The GC operating conditions were as follow: fused silica column [30m*0.25µmfilmof HP-5(thickness)]; the inlet and injection temperature was set at 275ºC to 310^{0}C. Split injection was adopted with a split ratio of 8:1. Using rubber septum and volume injected was 1ul. The column temperature was programmed as follow; held at 65^{O}C for 2min; 65-260^{O}C at 12^{O}C /min; 260-320^{O}C at 15^{O}C /min and maintained at 310^{O}C for 8minutes and oven temperature was set at 65ºC. Nitrogen was used as carrier gas. The hydrogen and compressed air pressure was 30psi. The oven initial temperature was at 65^{0}C. Verification of peaks was carried out based on retention times compared to those of external PAHs. Procedural blank and solvent blanks were analyzed and quantified, but no PAHs were found in these blanks.

3. Result

Table 1: PAH Concentrations (µg/Kg Wet Wt.) in M. Esculenta Collected from Alakahia and Eleme

PAH	ALAKAHIA	ELEME
Naphthalene$^{*,\Delta}$	0.16 ± 0.03^{a}	0.36 ± 0.05^{b}
Acenaphthylene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
2-Methyl naphthalene$^{*,\Delta}$	0.27 ± 0.03^{a}	0.08 ± 0.02^{b}
Acenaphthene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
Fluorene$^{*,\Delta}$	0.06 ± 0.01^{a}	0.18 ± 0.03^{b}
Phenanthrene$^{*,\Delta}$	0.02 ± 0.01	0.10 ± 0.01
Anthracene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
Fluoranthene$^{*,\Delta}$	0.34 ± 0.06^{a}	0.11 ± 0.01^{b}
Chyresene$^{*,\beta}$	0.03 ± 0.01	0.03 ± 0.01
Pyrene$^{\gamma,\Delta}$	1.23 ± 0.04^{a}	2.36 ± 0.15^{b}
Benzo[b]fluoranthene$^{\gamma,\beta}$	0.05 ± 0.01	0.03 ± 0.00
Benzo[k]fluoranthene$^{\gamma,\beta}$	0.55 ± 0.05^{a}	0.25 ± 0.05^{b}
Dibenz[a,h]anthracene$^{\gamma,\beta}$	0.05 ± 0.01	0.01 ± 0.00
Benzo[a]pyrene$^{\gamma,\beta}$	0.02 ± 0.01	0.01 ± 0.00
Indeno[1,2,3-cd]pyrene$^{\gamma,\beta}$	0.12 ± 0.02	0.16 ± 0.03
Benzo[g,h,i]perylene$^{\gamma,\beta}$	0.04 ± 0.00	0.03 ± 0.01
Benzo[a]anthracene$^{\gamma,\beta}$	0.00 ± 0.00	0.00 ± 0.00
Total PAH	2.94 ± 0.26^{a}	3.71 ± 0.37^{b}
LMW-PAH	0.85 ± 0.11	0.83 ± 0.12
HMW-PAH	2.09 ± 0.15	2.88 ± 0.25
LMW-PAH/HMW-PAH ratio	0.41	0.29

*denotes low molecular weight (LMW) PAH, γ- high molecular weight (HMW) PAH $^{\Delta-}$ Non-carcinogenic PAH, β- Carcinogenic PAH.

Table 2: PAH Concentrations (µg/Kg Wet Wt.) in C. Esculenta Collected from Alakahia and Eleme

PAH	ALAKAHIA	ELEME
Naphthalene$^{*,\Delta}$	0.26 ± 0.02^{a}	0.00 ± 0.00
Acenaphthylene$^{*,\Delta}$	0.00 ± 0.00	1.76 ± 0.05^{b}
2-Methyl naphthalene$^{*,\Delta}$	0.50 ± 0.05^{a}	0.00 ± 0.00
Acenaphthene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
Fluorene$^{*,\Delta}$	0.59 ± 0.16^{a}	0.00 ± 0.00
Phenanthrene$^{*,\Delta}$	0.19 ± 0.03^{a}	0.00 ± 0.00
Anthracene$^{*,\Delta}$	0.01 ± 0.01	0.00 ± 0.00
Fluoranthene$^{*,\Delta}$	0.70 ± 0.23^{a}	0.00 ± 0.00
Chyresene$^{*,\beta}$	0.56 ± 0.07^{a}	0.00 ± 0.00
Pyrene$^{\gamma,\Delta}$	1.14 ± 0.04^{a}	0.00 ± 0.00
Benzo[b]fluoranthene$^{\gamma,\beta}$	0.19 ± 0.03^{a}	53.75 ± 0.03^{b}
Benzo[k]fluoranthene$^{\gamma,\beta}$	1.28 ± 0.06^{a}	10.70 ± 0.78^{b}
Dibenz[a,h]anthracene$^{\gamma,\beta}$	0.30 ± 0.02^{a}	0.00 ± 0.00
Benzo[a]pyrene$^{\gamma,\beta}$	0.05 ± 0.01^{a}	2.68 ± 0.05^{b}
Indeno[1,2,3-cd]pyrene$^{\gamma,\beta}$	0.34 ± 0.02^{a}	1.94 ± 0.08^{b}
Benzo[g,h,i]perylene$^{\gamma,\beta}$	1.28 ± 0.06^{a}	0.00 ± 0.00
Benzo[a]anthracene$^{\gamma,\beta}$	0.00 ± 0.00	0.00 ± 0.00
Total PAH	7.39 ± 0.81^{a}	70.83 ± 1.56^{b}
LMW-PAH	2.25 ± 0.50	1.76 ± 0.05
HMW-PAH	5.14 ± 0.31	69.07 ± 1.51
LMW-PAH/HMW-PAH ratio	0.44	0.03

*denotes low molecular weight (LMW) PAH, γ- high molecular weight (HMW) PAH $^{\Delta-}$ Non-carcinogenic PAH, β- Carcinogenic PAH.

Table 3: Pah Concentrations (µg/Kg Wet Wt.) in D. Rotundata Collected from Alakahia and Eleme

PAH	ALAKAHIA	ELEME
Naphthalene$^{*,\Delta}$	0.72 ± 0.21^{a}	0.08 ± 0.02^{b}
Acenaphthylene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
2-Methyl naphthalene$^{*,\Delta}$	0.10 ± 0.01^{a}	0.00 ± 0.00
Acenaphthene$^{*,\Delta}$	0.00 ± 0.00	0.00 ± 0.00
Fluorene$^{*,\Delta}$	0.12 ± 0.02	0.04 ± 0.01
Phenanthrene$^{*,\Delta}$	0.03 ± 0.01	0.00 ± 0.00
Anthracene$^{*,\Delta}$	0.01 ± 0.00	0.00 ± 0.00
Fluoranthene$^{*,\Delta}$	0.26 ± 0.02	0.19 ± 0.03
Chyresene$^{*,\beta}$	0.11 ± 0.01^{a}	0.40 ± 0.06^{b}
Pyrene$^{\gamma,\Delta}$	1.95 ± 0.18^{a}	1.62 ± 0.06^{b}
Benzo[b]fluoranthene$^{\gamma,\beta}$	0.06 ± 0.01	0.02 ± 0.01
Benzo[k]fluoranthene$^{\gamma,\beta}$	1.97 ± 0.10^{a}	0.08 ± 0.02^{b}
Dibenz[a,h]anthracene$^{\gamma,\beta}$	0.18 ± 0.04	0.30 ± 0.02
Benzo[a]pyrene$^{\gamma,\beta}$	0.02 ± 0.01	0.03 ± 0.01
Indeno[1,2,3-cd]pyrene$^{\gamma,\beta}$	0.09 ± 0.01	0.25 ± 0.05
Benzo[g,h,i]perylene$^{\gamma,\beta}$	0.08 ± 0.02	0.03 ± 0.01
Benzo[a]anthracene$^{\gamma,\beta}$	0.00 ± 0.00	0.00 ± 0.00
Total PAH	5.70 ± 0.65^{a}	3.04 ± 0.30^{b}
LMW-PAH	1.24 ± 0.27	0.31 ± 0.06
HMW-PAH	5.14 ± 0.31	2.73 ± 0.24
LMW-PAH/HMW-PAH ratio	0.28	0.11

*denotes low molecular weight (LMW) PAH, γ- high molecular weight (HMW) PAH $^{\Delta-}$ Non-carcinogenic PAH, β- Carcinogenic PAH.

Table 4: Distribution of Total Carcinogenic and Non-Carcinogenic Pahs in the Tubers

	M. esculenta		C. esculenta		D. rotundata	
	Alakahia	Eleme	Alakahia	Eleme	Alakahia	Eleme
Σ Carcinogenic PAH	0.86 ± 0.11	0.52 ± 0.10	4.00 ± 0.27	69.07 ± 1.51	2.51 ± 0.20	1.11 ± 0.18
Σ Non-carcinogenic PAH	2.08 ± 0.15	3.19 ± 0.27	3.39 ± 0.54	1.76 ± 0.05	3.19 ± 0.45	1.93 ± 0.12
Total PAH	2.94 ± 0.26	3.71 ± 0.37	7.39 ± 0.81	70.83 ± 1.56	5.70 ± 0.65	3.04 ± 0.30
% Carcinogenic PAH	29.25	14.02	54.13	97.52	44.04	36.51

4. Discussion

The distribution of Polycyclic Aromatic Hydrocarbons in Manihot esculenta (Cassava) collected from Alakahia and Eleme shown in Table 1 revealed that a total of 17 PAHs were detected in the M. esculenta from both sites. Average concentrations of these PAHs ranged from below detection limit of 0.0001 µg/kg to 2.36 ± 0.15 -µg/kg. The total concentrations of PAHs in the M. esculenta were 2.94 ± 0.26 and 3.71 ± 0.37- µg/kg from Alakahia and Eleme respectively. The total PAHs concentration in the M. esculenta collected from Alakahia was markedly lower than the Eleme site. In this tuber, the highest average concentrations of 1.23 ± 0.04 and 2.36 ± 0.15 were recorded for Pyrene from both sites. Pyrene's release to the environment is ubiquitous since it is a product of incomplete combustion. It is largely associated with particulate matter, soils and sediments. Pyrene has also been identified as one of the dominant PAHs in auto emission [7, 8]. Thus, the presence of Pyrene, a high molecular weight PAH in the cassava may be attributed to auto emissions as some farmlands in both communities are situated close to roads.

There was significant difference observed (at $p<0.05$) when the concentrations of Naphthalene, Fluorene, Pyrene, 2-methylnaphthalene, Fluoranthene and Benzo[k]fluoranthene in M. esculenta collected from Alakahia was compared to that of Eleme community. The difference in both communities may be due to difference in the degree of anthropogenic pressure.

The Low Molecular Weight-PAH/High Molecular Weight-PAH (LMW-PAH/HMW-PAH) ratios in Manihot esculenta were 0.41 and 0.29 from Alakahia and Eleme respectively. The LMW-PAH/HMW-PAH ratio in Alakahia was markedly higher when compared to ratio found in Eleme. The LMW-PAH/HMW-PAH ratios were less than 1 at the Alakahia and Eleme site further suggesting that the PAH in the tuber are of pyrogenic origin.

Table 2 above shows the mean concentrations of Polycyclic Aromatic Hydrocarbons in Colocasia esculenta collected from Alakahia and Eleme. The highest average concentrations of 1.28 ± 0.06 and 53.75 ± 0.60 were recorded for benzo[k]fluoranthene and benzo[b]fluoranthene from Alakahia and Eleme respectively. Fluoranthene, Benzo[a]fluoranthene, Indeno[1,2,3-c,d]pyrene, Phenanthrene and Chrysene were identified as the predominant PAH emitted from a municipal and medical waste incinerator [8]. The predominance of benzo[b]fluoranthene and Benzo[k]fluoranthene in C. esculenta grown in both communities is an indication that emissions from the waste incinerators may be transported into the farmlands. This warrants a sensitization action in both communities as there are several farmlands within close vicinity of industrial areas.

The LMW-PAH/HMW-PAH ratios in Colocasia esculenta (cocoyam) were 0.44 and 2.84 from Alakahia and Eleme respectively. The LMW-PAH/HMW-PAH ratio in Alakahia was markedly lower when compared to ratio found in Eleme. The LMW-PAH/HMW-PAH ratio of 2.84 at the Alakahia site suggests that the source of PAH in the tuber is of petrogenic origin. There are two anthropogenic sources of PAHs: petrogenic and pyrogenic. Petrogenic sources include crude oil and petroleum products. Pyrogenic sources from by the incomplete combustion of organic matter in industrial operation, garbage incinerators, power plants, vehicle engines and forest fires [9]. The Eleme community in Rivers, Nigeria is home to a number of industries including Petrochemicals, Paint, and Fertilizer production industry. The presence of these industries at Eleme increases the anthropogenic pressure thereby increasing health risk for the exposed population.

According to European Union scientific committee on Food [10], Benzo[a]Pyrene BaP can be used as indicator of occurrence, concentration and effect of carcinogenic PAHs in foods. The concentration of Benzo[a]pyrene analyzed in the C. esculenta from Alakahia and Eleme were 0.05 ± 0.01 and 2.68 ± 0.05 respectively. The concentration of the C. esculenta from Alakahia was below the European Union (EU) limit of 2.0µg/kg while C. esculenta from Eleme exceeded the limit thus suggesting that exposed population may be at risk of PAH toxicity.

The average PAH concentrations (mean ± S.E.M, µg/kg.) and Total mean PAH concentrations (µg/kg.) in D. rotundata are shown in Table 3. Average concentrations of these PAHs ranged from below detection limit of 0.0001 µg/kg to 1.97 ± 0.10 µg/kg. The total concentrations of PAHs in the D. rotundata were 5.70 ± 0.65 and 3.04 ± 0.30 from Alakahia and Eleme respectively. The total PAHs concentration in the D. rotundata collected from Alakahia was significantly higher ($p<0.05$) than the Eleme site. In this tuber, the highest average concentrations of 1.97 ± 0.10 and 1.62 ± 0.06 were recorded for benzo[k]fluoranthene and Pyrene from Alakahia and Eleme respectively.

The LMW-PAH/HMW-PAH ratios in Dioscorea rotundata were 0.28 and 0.11 from Alakahia and Eleme respectively. The LMW-PAH/HMW-PAH ratio in Alakahia was markedly higher when compared to ratio found in Eleme. The LMW-PAH/HMW-PAH ratios were less than 1 at the Alakahia and Eleme site respectively.

The concentration of Benzo[a]pyrene analyzed in the D. rotundata from Alakahia and Eleme were 0.02 ± 0.01 and 0.03 ± 0.01 respectively. Both concentrations were below the European Union (EU) limit of 2.0µg/kg.

In the present study, the percentages of carcinogenic PAHs as shown in Table 4, M. esculenta from the Eleme community had the least percentage (14.02) while . The tubers recorded variations in percentage of carcinogenic PAHs ranging from 29.25 – 54.13 at Alakahia and 14.02 – 97.52 at Eleme community. The high values reported for carcinogenic PAHs in the C. esculenta from Eleme community compared to that from Alakahia community may be attributed to higher PAH deposition resulting from vehicular emission and industrial activities at the Eleme community. The concentration of PAHs in the tubers in the present study were higher than recorded data for food crops collected from Owode and Aroje in Lagos, South-West Nigeria [11].

5. Conclusion

The present study provides insight on the distribution of PAH in tubers grown in farmlands in two notable communities in Rivers state. With the exception of C. esculenta from the Eleme farmlands, the current levels of PAH in the tubers are below the WHO maximum permissible limit of 10 ppb for PAH in food crops. However, considering the frequency of consumption of the tubers by the populace at Alakahia and Eleme, constant monitoring of the PAH levels in farm soils and food crops over time is required. Also cultivation of food crops close to industrial areas and major roads should be discouraged as tubers collected from farmlands close to major roads tend to have higher PAH content.

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