

# POLYCYCLIC AROMATIC HYDROCARBONS, PAHs CONTAMINATION LEVELS AND HEALTH RISKS IN FOODS CONSUMED IN NIGERIA: A SYSTEMATIC REVIEW

**Comment [NU1]:** Put PAH in bracket. Remove 's' and 'r' from the written one and abbreviation.

## Abstract

The rate of increase in the number of cancer patients in Nigeria is alarming and calls for constant investigations into polycyclic aromatic hydrocarbons, PAHs and other pollutants. PAHs are described as a group of organic compounds which are formed and released into the natural environment during incomplete combustion of organic materials such as crude oil, coal, wood, and are carcinogenic and genotoxic in nature. They are frequently present across various components of the natural environment (foods, soil, air, water). This study aimed at developing a comprehensive report on PAH pollution and its human health risks recorded in the Nigeria. Fifteen studies were selected on PAHs contamination levels and health risk assessment in the following food categories: grains and grain products; seafoods; protein foods; leafy and fruit vegetables. The following information describing each study were extracted: authors, year of publication, aim of study, area of study, period of sampling, type(s) and number of samples collected, analytical technique, number and concentrations of PAHs identified, risk assessment and potential sources (in some of them) of PAH pollution in the study area. All the papers reviewed reported detection of PAHs in their samples but majority recorded the values of margin of exposure, MOE higher than 10,000 which according to European food security authority (EFSA) indicate low concern for human health and considered low priority for risk management actions. While few recorded MOE values less than 10,000 indicating concern for human health. Some studies reported mean values of PAHs lower than the permissible limit by some regulatory bodies while the PAHs of some samples were higher. It is recommended that prompt action be taken by the policy makers and stakeholders to ensure human health protection and future studies should focus on PAH pollution in farmlands, soils, water, ambient air and the associated human and ecological health risks.

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**Comment [NU10]:** Put in bracket and remove r in exposure.

**Comment [NU11]:** Clarify how and why higher levels of MOE will give low concern and vice versa. I also suggest you restructure it this way; All the papers reviewed reported detection of PAHs in the samples, majority recorded values of margin of exposure (MOE) higher than 10,000 indicating low concern for human health and low priority for risk management according to EFSA. Remember to write EFSA in full before putting it bracket.

**Keywords:** Polycyclic aromatic hydrocarbons, Food, Contaminations, Health risk, Review

## 1.0 Introduction

Polycyclic aromatic hydrocarbons (PAH) are organic compounds that possess two or more fused aromatic rings of carbon and hydrogen atoms which are produced and released quotidianly into the environment during incomplete combustion of organic materials [1,2,3]. The primary sources of PAHs are identified as anthropogenic in origin such as the exhaust of motor vehicles, petroleum refineries, heating in power plants, combustion of refuse, deposition from sewage, oil/gasoline spills, tobacco smoke, barbeque smoke, and coke production.

There are over 100 PAH congeners (including parent PAHs and alkylated derivatives) identified in the environment. The United States Environmental Protection Agency [4] has, however, indicated 16 of them as priority pollutants under its Clean Water Act due to the possible risks they pose to human and ecological health. They are chrysene, acenaphthylene, acenaphthene, phenanthrene, anthracene, fluoranthene, fluorene, pyrene, naphthalene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*a*]anthracene, dibenzo[*a,h*]anthracene, benzo[*g,h,i*]perylene, and indeno[1,2,3-*cd*]pyrene[4].

The properties of PAHs are determined by the number of benzene rings and their molecular mass [5]. PAH congeners that possess four or more benzene rings are described as high molecular weight (HMW) PAHs and they include fluoranthene, pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indeno[1,2,3-*cd*]pyrene, dibenzo[*a,h*]anthracene, benzo[*g,h,i*]perylene. While congeners with two or three benzene rings are described as low molecular weight (LMW) and they include naphthalene, acenaphthylene, acenaphthene, fluorine, phenanthrene and anthracene. [5,2,3]. Heavy PAHs are more stable and more toxic than the light PAHs [6]. Benzo(*a*)pyrene [B(a)P], a well-known PAH congener, is used to represent PAHs as it is considered by the International Agency for Research on Cancer[7] as a known carcinogen. They suggested the use of benzo[*a*]pyrene as a marker of occurrence and effect of the carcinogenic PAHs.

The CONTAM Panel concluded that the risk characterization should be based on the PAHs for which oral carcinogenicity data were available i.e. for benzo[*a*]pyrene and the other PAHs that were measured in the two coal tar mixtures used in the carcinogenicity studies of [8]: benzo[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*g,h,i*]perylene, chrysene, dibenzo[*a,h*]anthracene and indeno[1,2,3-*cd*]pyrene. The CONTAM Panel concluded that these eight PAHs (PAH8), either individually or in a combination, are currently the only possible indicators of the carcinogenic potency of PAHs in foods. Besides, the sum of the above mentioned eight PAHs (PAH8), the sum of benzo[*a*]pyrene, chrysene, benzo[*a*]anthracene and benzo[*b*]fluoranthene (PAH4) as well as the sum of benzo[*a*]pyrene and chrysene (PAH2) were calculated. It has been suggested that MOE approach, using PAH4 (BaA, BaP, BbF& Chy) and PAH8 (BaA, Chy, BaP, BbF, BkF, DahA, BghiP&IndP), is better for the risk assessment, since BaP alone is not a suitable indicator of occurrence and effects of PAHs in foods [9].

**Comment [NU12]:** Write in full. You don't start a sentence with abbreviation.

**Comment [NU13]:** Replace with International Agency for Research on Cancer. Minimize the use of pronoun.

For different food categories and subcategories, the data on PAH8, PAH4 and PAH2 were used for the exposure calculation as well as the estimation of margins of exposure (MOEs) based on the benchmark dose lower confidence limit for a 10% increase in the number of tumour bearing animals compared to control animals (BMDL<sub>10</sub>)[10]

PAHs have been reported to cause carcinogenic and mutagenic effects and are potent immune suppressants. They can interfere with the normal function of DNA.[11]. Human beings may be exposed to these substances at home, outside or at workplace through inhalation, ingestion or dermal contact. Oncogenic, teratogenic effects, genotoxicity, an increased level of cholesterol in the blood or reproduction defects, biochemical disruption and cell damage were observed after longterm PAHs exposure which was confirmed by toxicological experiments.[12]

The widespread characteristic of PAHs and their ability to cause cancer or toxic effects on living organisms in the environment constitute a significant risk to human health. The release of PAHs can result in their uptake into the body by humans and other mammals through ingestion of contaminated food or water, inhalation of polluted air, and dermal contact with polluted soil. The toxic effects which may occur as a result of public exposure to PAHs have short- and long-term health effects commonly associated with breathing and cardiovascular disorders. [13,3]. The two highest contributors to the dietary exposure were cereals and cereal products, and sea food and sea food products. Foods can be contaminated by environmental PAHs present in air, soil or water, by industrial food processing methods (e.g heating, drying and smoking processes) and by home food preparation (e.g grilling and roasting processes).

Several studies reported PAHs contaminations and health risk assessment in some foods commonly consumed in Nigeria – grains and its products, [14,15,16,17] vegetables and vegetable oils[18,19,20] sea foods [21,22,23]edible mushroom[24],meat [25,26,27], fish [28,29].Some studies reported PAHs contamination levels lower than the established permissible limit while some reported higher. For risk assessment, some studies revealed that the values of margin of exposures (MOEs) obtained for all the indicators were higher than 10,000 which according to EFSA indicate low concern for human health and considered low priority for risk management actions. While some reported that the values of margin of exposures (MOEs) obtained for all the indicators were lower than 10,000 which according to EFSA indicate concern

for human health and considered priority for risk management actions. This situation limits the ease of access and use of these studies by the public and even the scientific community, and further breeds a lack on general understanding of PAH pollution in Nigeria. It is, therefore, timely and important to address the above issues through a systematic review. The current study serves as a comprehensive report on the degree of PAH pollution in various foods commonly consumed in Nigeria and its associated human health risks. It reviewed critically the quality of data presented by the various selected studies, as well as provided recommendations necessary for tackling this environmental issue in the Nigeria

## **2.0 Method.**

### **2.1 Criteria for Search and Selection of PAH-related Studies.**

The search and selection process extracted studies that have been published from 2013 to 2023. Twenty studies were selected. The keywords used in the search process were “Polycyclic Aromatic Hydrocarbons contamination and risk assessment in foods consumed in Nigeria”. Based on the different studies that were selected, the following categories of studies were used to aid in the selection process: PAHs in grains and grain products; PAHs in seafoods; PAHs in protein foods; PAHs in vegetable and vegetable oils. The selected studies were then assessed for relevance. Concerning studies that were published twice, researchers decided to choose the one that was most recent with adequate information on identified PAHs.

### **2.2 Data Extraction**

After selection of suitable studies, a set of information describing each study was extracted: authors, year of publication, aim of study, area of study, period of sampling, type(s) and number of samples collected, analytical technique, number and concentrations of PAHs identified, risk assessment and potential sources of PAH pollution in the study area.

### 3.0 Contamination levels and risk assessment of PAHs in the Foods in Nigeria

#### 3.1 Grains and grain products:

The study carried out by [14] on risk assessment of polycyclic aromatic hydrocarbons in foreign and local rice consumed in South East Nigeria aimed at determining the quantity of polycyclic aromatic hydrocarbons in foreign and local rice consumed in the South East of Nigeria; estimating the daily intake and the health risks associated with the consumption among adult male and female individuals in South East, Nigeria. Eighteen (18) samples of foreign rice (Indian rice, Royal Stallion, Thailand rice) and local rice (Abakiliki, Lafia, Adani.) were purchased from some major markets in Enugu and Anambra states of Nigeria. Extraction of PAHs from the samples was by sonication followed by clean-up. Recovery experiments to optimize PAH extraction from grain samples were carried out. Following a good recovery, the samples were analyzed for sixteen PAHs contamination levels using gas chromatography coupled with flame ionization detector. The authors reported contamination of the samples with 16 PAHs but the contamination levels were much lower than the permissible limit 1.0 µg/kg established by EFSA. From the estimated daily intake, total dietary exposure of male was less than that of female indicating that female daily intake of rice is higher.

Comment [NU14]: Name the markets

They also reported that the values of margin of exposures (MOEs) obtained for all the indicators (PAH2, PAH4 and PAH8) were much higher than 10000 which according to EFSA indicate low concern for human health and considered low priority for risk management actions. The result of their study provided base values for future monitoring of contamination levels of rice grains and equally revealed that the safety of consumption of both local and foreign rice obtained from markets in South East, Nigeria can be ascertained. They recommended that other foods should be analyzed regularly to ensure their safety with respect to PAHs.

Another study by [15] on health risk of polycyclic aromatic hydrocarbons (PAHs) from wheat (*Triticum specie*), bambara nut and pigeon peas (*Cajanus cajanifolia*) aimed at determining the PAH contamination levels in types of wheat (*Triticum specie*), bambara groundnut (*Vigna subterranea*) and pigeon peas (*Cajanus cajanifolia*) commonly consumed in the Eastern part of

Nigeria and assessing the health risk associated with their consumption. Eighteen (18) samples which included different types of wheat, (*Triticum specie*)(Hard and Soft), bambara groundnut (*Vigna subterranean*)(Pure white and Mixed white) and pigeon peas (*Cajanus cajanifolia*) (White and Red) were purchased from some major markets in Enugu and Anambra states of Nigeria. The grain samples were analyzed for sixteen priority PAHs using gas chromatography coupled with flame ionization detector (GC-FID) after extraction by sonication. Estimation of daily intakes were carried out using adult male and female consumers while margin of exposure was used to assess the health risk applying bench mark dose levels for the indicators-BaP, PAH2, PAH4 and PAH8. The three analyzed grains contained the sixteen PAHs but at the level lower than 1.0 µg/kg permissible limit established by EFSA for cereals and cereal based products. Estimation of daily intake revealed that adult female individuals are more exposed when compared to their male counterparts. The values of margin of exposures (MOEs) obtained for all the indicators were higher than EFSA (10,000) indicating low concern for human health and considered low priority for risk management actions. This study guaranteed safety of consumption of these grains. Data from this study can be used by regulatory bodies to establish limits for grain legumes (bambara groundnut and pigeon peas). The authors recommended that the environmental substances like foods, soil, water and air should be on regular chemical analysis to ensure their safety with respect to PAHs and other hazardous compounds.

Comment [NU15]: Name the markets

A study by [16] on risk assessment of polycyclic aromatic hydrocarbons on Pasta products consumed in Nigeria was aimed at quantitative determination of polycyclic aromatic hydrocarbons in pastas consumed in Nigeria in view of estimating the daily intake and the possible risks to consumers. Sixty samples of different brands of locally manufactured and imported pastas were collected in November 2014 from two major cities in Nigeria (Abuja and Enugu). The samples were categorized into three: *Noodles*, *Spaghetti* and *Macaroni*. The extractions were carried out, using 100 ml mixture of n-hexane and dichloromethane (1:1 v/v) for 16h in Soxhlet extractor. After clean-up, the eluted extracts were combined and evaporated to 1ml and analyzed for PAHs using gas chromatography coupled with mass spectrometer (GC-MS). The study found sixteen polycyclic aromatic hydrocarbons (PAHs) in the locally produced and imported pastas. Estimation of daily intake was done on generally exposed (low) and typically exposed (high) consumers. The margin of exposure was used to assess the risk to

consumers. The concentration of  $\Sigma$  16 PAHs in Nigerian and imported brands of pastas ranged from 9 $\mu\text{g}/\text{kg}$  to 800 $\mu\text{g}/\text{kg}$  and 2 $\mu\text{g}/\text{kg}$  to 7 $\mu\text{g}/\text{kg}$ , respectively. The Margin of Exposure (MOE) based on PAH8 for generally exposed children was less than 10,000 in 25% of Nigerian brands while it was 38% for typically exposed children. For imported brands of pastas, the MOE values were far higher than 10,000 for generally and typically exposed children and adults. The result of their study revealed a potential concern for individuals who consume certain Nigerian brands of pastas especially children and a low concern for consumers (children and adults) of imported brands of pastas. They recommended the need for setting allowable limits for PAHs in Nigerian foods and continuous analytical monitoring and increased research on Nigerian brands of pasta to ensure adequate protection of human health and the risk management actions.

**Comment [NU16]:** Why don't you use significantly instead of much, far etc

**Comment [NU17]:** I prefer safety of the products.

A review of a study on health risk assessment and dietary exposure to polycyclic aromatic hydrocarbons (PAHs) (lead and cadmium) from bread consumed in Nigeria by (17) which aimed at assessing the dietary exposure and health risk associated with consumption of bread from bakeries in Nigeria. Sixty samples of bread were collected on June 2015 from different types of bakeries where the heat is generated from wood (42 samples) and by electricity (18 samples) from twenty bakeries located in Gusau Zamfara (B1-B14) and Port Harcourt Rivers States (B15-B20) in Nigeria. Extraction of poly aromatic hydrocarbons from the bread samples was done with a Ultrasonic bath, Elmsonic S40H) in accordance with US SW-846 Method 3550 while analysis of PAHs was carried out using GC-FID. The study observed that the non-carcinogenic PAHs pyrene (13.72  $\mu\text{g}/\text{kg}$ ) and genotoxic PAHs (PAH8), benzo[a]anthracene (9.13  $\mu\text{g}/\text{kg}$ ) had the highest concentrations. Total benzo[a]pyrene concentration of 6.7  $\mu\text{g}/\text{kg}$  was detected in 100% of tested samples. Dietary intake of total PAHs ranged between 0.004-0.063  $\mu\text{g}/\text{kg}$  body weight  $\text{day}^{-1}$  (children), 0.002-0.028  $\mu\text{g}/\text{kg}$   $\text{day}^{-1}$  (adolescents), 0.01-0.017  $\mu\text{g}/\text{kg}$   $\text{day}^{-1}$  (male), 0.002-0.027  $\mu\text{g}/\text{kg}$   $\text{day}^{-1}$  (female), and 0.002-0.025  $\mu\text{g}/\text{kg}$   $\text{day}^{-1}$  (seniors). The study reported that PAHs concentrations detected in all bread samples were within the acceptable and the intake limit indicating that health risk associated with exposure to B[a]P may not be significant. The maximum acceptable concentration of 1  $\mu\text{g}/\text{kg}$  for B[a]P set by European regulations for cereal processed foods was not exceeded in any of the samples. The contamination profile of PAHs, Pb and Cd in wood and electric baked bread showed no significant difference.

## 3.2. Protein foods

### 3.2.1. Fish

[28] studied the levels and risk assessment of polycyclic aromatic hydrocarbons in water and fish of rivers Niger and Benue confluence Lokoja, Nigeria. Twenty fish samples namely *Clarias spp* (Catfish) and *Oreochromis spp* (Tilapia) were purchased from fishermen at the bank of the river confluence. The study was carried out at the confluence of rivers Niger and Benue in Lokoja, Kogi State, Central Nigeria. The state lies between the coordinate of latitude 7°30'N/6°42'E and longitude 7°50'N/6°70'E and Lokoja lies between 7°45'N, 7°52'N of the equator and longitude 6°45'E of the Greenwich meridian. Extraction of PAHs from the samples was done using Soxhlet apparatus with appropriate solvent mixtures (mixture of acetone and *n*-hexane). Extracted PAHs were cleaned up and concentrated to 1 ml using rotary evaporator. Following a good recovery result, the samples were analyzed for PAHs using GC-MS. The result of the study revealed the following: recovery test result ranged from 99.90 to 104% for the PAHs, limit of detection ranged from 0.0001–0.0002 µg/kg while limit of quantitation ranged from 0.0003–0.0007 µg/kg of the polycyclic aromatic hydrocarbons. Six out of the sixteen United States Environmental Protection Agency (US EPA) priority polycyclic aromatic hydrocarbons were detected in the water and fish samples respectively. The PAHs detected were naphthalene, phenanthrene, anthracene, benzo [b] fluoranthene, benzo [k] fluoranthene and benzo [a] pyrene. The concentrations of the six detected PAHs in water ranged from Nap (Not Detected {ND} to 0.543), Ph (ND to 0.083), Ant (ND to 0.083), BbF (0.080 to 0.093), BkF (0.083 to 0.093) and BaP (0.083 to 0.113) mg/l with distribution pattern of Nap > BaP > BbF = BkF > Ant = Ph. While the mean concentration value of PAHs in Catfish and Tilapia were Nap (2.383 and 1.947), Ph (0.050 and 0.057), Ant (0.057 and 0.057), BbF (0.043 and ND), BkF (0.043 and ND) and BaP (0.050 and ND). The health risk assessment showed that the concentration of Benzo[a]pyrene, a known indicator of carcinogenic PAHs was of health risk concern. The level of PAHs in water and fish were not significantly different ( $p > 0.05$ ). The correlation studies showed that the PAHs were from the same source. Benzo[a]pyrene concentration values in water and Catfish was high compared to the concentration limit of 0.01 µg/l for water recommended by Environment Canada and should be of concern. Immediate attention was recommended to reduce improper waste disposal and improve on measures against indiscriminate dumping of petroleum

Comment [NU18]: Reference correctly.

products and domestic waste in the water as well as burning of tyres, organic and petroleum products which are the major sources of these PAHs.

Another study by [29] on Human health risk assessment of polycyclic aromatic hydrocarbons (PAHs) in smoked fish species from markets in Southern Nigeria was reviewed. The researchers determined polycyclic Aromatic Hydrocarbon (PAH) levels in four commonly consumed smoked fish species from markets in Southern Nigeria and assessed the possible human health risks associated with their consumption. Samples of *Clarias gariepinus*, *Tilapia zilli*, *Ethmalosa fimbriata*, and *Scomberscombrus* were randomly collected from three major markets (Oreogbeni (6°21' 0.09"N and 5° 39' 32.67"E), New Benin (6°23' 59.96"N and 5° 36' 67.37"E) and Santana market (6°17' 44.6"N and 5° 38' 8.9"E) (Where?) in Southern, Nigeria. Samples were collected every month for four months (June–September). Extraction of PAHs was carried out based on the method described by [30]. Extracts were then cleaned up, concentrated and collected in 2 ml vials and analyzed for PAHs using GC-MS. The result of the study showed that the values of PAH4 observed in smoked fish species were 0.45, 0.22, 0.26, 0.47, 0.16 mg/kg for *C. gariepinus*, *T. zilli*, *E. fimbriata*, and *S. scombrus* respectively. These observed values were above the recommended limit (0.03 mg/kg) set by the European Union[31] for PAHs in smoked fish and smoked fishery products. The result, therefore, implied that consumption of these fish species could be a potential health hazard to humans. The high values of PAHs observed in the smoked fish samples might be attributed to the smoking process during preparation and preservation. Most PAHs in smoked foods, especially fish, comes from wood smoke [32] and wood smoke has been reported to contain a large number of PAHs[33]. Carcinogenic human health risk assessment using carcinogenic toxic equivalents (TEQ) indicated that consumption of *E. fimbriata* had higher carcinogenic risk potential. TEQ values for all the fish species were however, below the estimated screening value (SV) of 3.556 mg/kg, while the estimated cumulative excess cancer risk (ECR) for *E. fimbriata* and *C. gariepinus* and PAH4 index for all the assessed fish species exceeded threshold values indicating potential carcinogenic risk on consumption. The study, therefore, deduced that there were substantial exposure and possible carcinogenic human health risk from consumption of smoked fish species from Southern Nigeria and recommended education of fish mongers and vendors on safer processing and preservative alternatives.

Comment [NU19]: Write in full.

### 3.2.2 Meat

A study by [25] on health risk assessment of polycyclic aromatic hydrocarbons in singed *Capra aegagrus* meat from Uyo Municipal Abattoir in Southern Nigeria aimed at evaluating the concentrations of PAHs in singed and unsinged liver and kidney samples of *C. aegagrus* obtained from Uyo Municipal Abattoir in Southern Nigeria and the associated health risks on adult and children population. The study revealed the potential carcinogenic risk in humans associated with the consumption of *C. aegagrus* meat singed with scrap car tyres and condemned plastics. Liver and kidney samples of goat singed with used scrap car tires, condemned plastics and woods were procured in August, 2019, from Uyo Municipal Abattoir in Uyo Metropolis, Akwa Ibom State, Southern Nigeria. Liver and kidney samples of unsinged goat in the same abattoir were also obtained and used as control for this study. Solid phase extraction was used for sample purification, PAHs were extracted from the samples with n-hexane/dichloromethane, the extracts were cleaned up and concentrated. Gas chromatography-mass spectrometry was used for the quantification of the levels of PAHs in the liver and kidney samples. After extraction with n-hexane/dichloromethane and concentrated. The total PAH concentrations for singed liver samples ranged from 44.70-48.23 mg kg<sup>-1</sup> while 23.53 mg kg<sup>-1</sup> was recorded for unsinged liver sample. Similar quantification for singed kidney samples yield ranged from 66.89-71.51mg kg<sup>-1</sup> and 34.63 mg kg<sup>-1</sup> for unsinged kidney sample. The hazard quotient and index values of singed and unsinged samples indicated no substantial non-carcinogenic risk relative to exposure to either individual polycyclic aromatic hydrocarbons or complex PAHs mixture through consumption of liver and kidney of *C. aegagrus*. The study also revealed potential carcinogenic risk in humans from consumption of singed liver and kidney of *C. aegagrus*. The result of the study provided reasonable evidence on the need to evaluate the risks of PAHs in singed meat to safeguard the health of consumers.

**Comment [NU20]:** Repetition

**Comment [NU21]:** Not clear. I think it will be no substantial carcinogenic risk.

Another study on health risk assessment of polycyclic aromatic hydrocarbons in charbroiled meat commonly consumed in Port Harcourt Metropolis by [26] was reviewed. The study aimed

at assessing polycyclic aromatic hydrocarbons (PAHs) concentrations in four selected commonly consumed charbroiled meats (croaker fish (*Micropogonias undulatus*), chicken (*Gallus gallus domesticus*), pork (*Sus scrofa domestica*) and beef (*Bos taurus*) and the potential health risks associated with their consumption. The eight samples were bought from various roadside barbeque stands at Mile 4 and Chobain Obio-Akpor local government area, Port Harcourt. The PAHs were extracted from the samples by solvent extraction while the analysis was carried out using gas chromatography coupled with mass spectrometer (GC-MS). The result revealed that the levels of PAHs in croaker fish, chicken, pork and beef collected from Mile 4 were significantly lower ( $p < 0.05$ ) than the levels of PAH in croaker fish, chicken, pork and beef collected from Choba. The highest mean concentration was recorded for Benzo (a) pyrene in croaker fish ( $0.733 \pm 0.015 \text{ mg/kg}$ ) and pork ( $0.733 \pm 0.021 \text{ mg/kg}$ ) from Choba and Mile 4 respectively. These values were above the maximum permissible limits recommended for Benzo (a) pyrene by USEPA. Benzo(a) pyrene levels in the samples were significantly higher ( $p < 0.05$ ) and above the European Union (EU) limit of  $2.0 \mu\text{g/kg}$  ( $0.0002 \text{ mg/kg}$  for meat). Data from the study highlighted a potential health concern for the indigenes of Choba and Mile 4 as the estimated daily intake of PAHs in four commonly consumed charbroiled meats exceeded the tolerable daily intake level. Moreover, the Hazard Indexes across the study areas were less than one (1) suggesting no potential adverse health risk. The carcinogenic potency equivalent of benzo(a) pyrene B(a)P of charbroiled meat in both study sites exceeded the critical allowable limit for carcinogenic PAHs thus suggesting potential adverse health hazard for Choba and Mile 4 populace. The researchers with respect to estimated health risk in their study strongly recommended that subsequent dietary intake of the commonly consumed meats around industrial areas or close to the main roads should be discouraged.

**Comment [NU22]:** Is croaker fish meat?

**Comment [NU23]:** Croaker fish and pork from Choba and Mile 4 recorded the highest individual mean concentrations with similar values ( $0.733 \text{ mg/kg}$ )

**Comment [NU24]:** Based on the estimated health risks, the researchers recommended that consumption of charbroiled meat sold, processed or prepared around industrial areas or close to the main roads should be discouraged.

### 3.2.3 Mushroom

Study by [24] on Polycyclic Aromatic Hydrocarbons in Edible Mushrooms from Niger Delta, Nigeria: Carcinogenic and non-carcinogenic health risk assessment carried out in oil-rich Niger Delta. The study involved assessment of PAH levels in wild and cultivated edible mushroom species consumed by the general populace from the oil producing Niger Delta, Nigeria. Ten mushroom samples were collected in triplicates from different locations in Port Harcourt,

Rivers State, Nigeria in October 2015. The extraction of PAHs from the mushroom was by sonication using Ultrasonic bath-Elmsonic S40H. The extracts were cleaned up and concentrated and the PAHs determined using GC-MS. The concentrations of USEPA-16 PAHs were determined by gas chromatography and carcinogenic and non-carcinogenic health risks were calculated. The concentrations of USEPA-16 PAHs ranged from 0.02 mg/kg – 3.37 mg/kg. The dietary intake of carcinogenic and non-carcinogenic USEPA-16 PAHs for adults, adolescents and seniors ranged from 0.00 – 0.05 mg/kg/day, 0.00 – 0.06 mg/kg/day and 0.00 – 0.07 mg/kg/day respectively. The BaPeq ranged from 0.02 – 2.76 with margin of exposure MOE values of BaP ranging from 3,500,000 to 700,000, 3,500,000 and 3,500,000 to 7,000,000 for adults, adolescents and seniors respectively indicating very insignificant health risk concern. The incremental lifetime cancer risk was within the safe range of  $1.56 \times 10^{-8}$  –  $1.73 \times 10^{-6}$  with the highest calculated risk found in wild *Pleurotus ostreatus* mushroom species from the study area. The researchers concluded from the result of the study that mushrooms studied seemed relatively safe for consumption and recommended need for regular studies on the PAHs concentration levels and health risk assessment in other agricultural produce within the region with a view of determining the extent of contamination and the health risk associated with the natural product consumption by the general populace.

A study by [27] on the level and health risk assessment of polycyclic aromatic hydrocarbons in protein foods from Lagos and Abeokuta, Southwestern, Nigeria assessed levels and health risk of polycyclic aromatic hydrocarbons (PAHs) in protein foods collected from selected locations in Lagos and Abeokuta. A total of forty-eight protein food samples (meat, cow skin, fish and crayfish) were collected between July and September 2018. The protein food samples were meat (raw and smoked beef (12 samples)), cow skin (raw and smoked (12 samples)), fish (raw and smoked catfish (12 samples)) and crayfish (raw and smoked (12 samples)). The protein foodstuffs (0.25 kg) were collected in triplicates across the sampling sites. The samples were subjected to chemical analysis of polycyclic aromatic hydrocarbons (PAHs) using standard method. Data collected were subjected to simple descriptive statistics of mean and standard deviation using SPSS for Windows (22.0). The health risk assessment was evaluated for average daily dose (ADD), hazard quotient (HQ), hazard index (HI) and cancer risk (CR) using the United States Environmental Protection Agency model. Results revealed higher concentrations

of  $\Sigma$ PAHs in protein food samples from Abeokuta than those from Lagos (except smoked cowskin). Indenol[1,2,3-cd]pyrene ( $96.817 \pm 65.922 \text{ mg kg}^{-1}$ ) was the highest PAH congener measured in protein foodstuffs (raw fish samples from Abeokuta). The  $\Sigma$ CR values of PAHs in Abeokuta fish (smoked) and crayfish (raw and smoked) samples were higher than the priority risk level of  $1.0 \times 10^{-4}$  indicating possible risk of developing cancer through consumption of protein foodstuffs. The smoked meat samples were generally dominated by high molecular weight PAHs, while the smoked cowskin samples were dominated by low molecular weight PAHs. The researchers concluded from the result of their study that PAH levels were generally higher in smoked than raw protein food samples.

Comment [NU25]: Clarify

### 3.3 Sea foods

( ) [23] on Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Commonly Consumed Shellfishes (whelk, oyster and periwinkle) from Kula, Rivers State, Nigeria revealed bioaccumulation of PAHs in the sea organisms (shellfish) due to anthropogenic activities going on in the study region. Samples of the sea foods were randomly collected from communities in Kula Kingdom in the month of November 2017-May 2018. PAHs were determined in the sample according to established protocol by USEPA (1986). The PAHs determination was done using gas chromatography coupled with flame ionization detector (GC-FID). Human health risk assessment models based on United States Environmental Protection Agency (USEPA) was used to characterize risks of PAHs exposure to non- cancer (Hazard Index) and excess cancer risk (ECR). From the results, Benzo [a] Anthracene (BaA) had highest concentrations in whelk ( $0.689 \pm 0.003$ ) and Periwinkle ( $0.930 \pm 0.001$ ) while Naphthalene had highest concentration in oyster ( $2.000 \pm 0.000$ ). The Total concentration of PAHs in  $\mu\text{g/kg}$  for whelk, oyster and periwinkle were  $1.797 \pm 0.013$ ,  $3.977 \pm 0.024$  and  $1.564 \pm 0.017$  respectively while the estimated daily intake (EDI) of PAHs ( $\text{mg/kg/day}$ ) via consumption of shell fish ranged from  $2.00 \times 10^{-4}$  to  $6.40 \times 10^{-2}$ ,  $7.0 \times 10^{-4}$  to  $1.86 \times 10^{-1}$  and 0 to  $8.64 \times 10^{-2}$  respectively which was far above oral reference dose (RFD). The toxic equivalent (TEQ) values were  $1.276 \times 10^{-4}$ ,  $1.252 \times 10^{-4}$  and  $4.034 \times 10^{-4}$  for whelk, oyster and periwinkle respectively. These values were significantly ( $p < 0.05$ ) higher than the screening value (SV)  $1.81 \times 10^{-5} \text{ mg/kg}$  for shellfish. The estimated

excess cancer risk (ECR) obtained for whelk was  $(3.0 \times 10^{-4})$ , oyster  $(2.00 \times 10^{-4})$  and periwinkle  $(3.24 \times 10^{-4})$ . These values were far above the USEPA acceptable limit  $(1 \times 10^{-4})$ . The authors deduced from their study that bioaccumulation of PAHs in the sea organisms (shellfish) due to anthropogenic activities going on in the study region (illegal oil refining, kpoos fire or bunkering) was a potential health hazard to consumers. Carcinogenic indices indicated that daily Intake of contaminated shellfish exposes the local populace to cancer risks. The researchers in the light of their findings recommended need for policymakers and other stakeholders to regulate anthropogenic activities resulting from increased emission of PAHs in the study area and protect local residents from impending health risk associated with exposure.

[22] analyzed polycyclic aromatic hydrocarbons in sediments and health risk of fish, crab and shrimps around Atlas Cove, Lagos Nigeria. The study determined the PAHs concentrations in sediments and two species of fish (*Drepaneaficana* and *Pomadasysjubelini*), crabs (*Callinectes amnicola*) and shrimps (*Penaeus notialis*) around Atlas Cove, Jetty, Lagos, Nigeria. A total of 45 surface sediment samples were collected in five different locations, at 5 cm depth, around Atlas Cove jetty between June and August 2016. Sediment samples were collected from five different sites, all of which were impacted by anthropogenic activities such as ship traffic and offloading. Sediment samples were collected using sediment grab and transferred onto aluminum foil papers. Twelve (12) samples of each fish species and 20 samples of shrimps and crabs were purchased from local fishermen at the landing site of the study area and transported to the laboratory on ice packs. The PAHs were extracted from the samples using Soxhlet extractor with solvent mixture of hexane and dichloromethane in the 1:1 ratio. The extracts were cleaned up, concentrated and analyzed for PAHs using gas chromatography-mass spectrometry. Human health risk assessment was calculated from biota using dietary daily intake and carcinogenic potencies of individual PAH concentrations. A total of 17 PAH congeners were detected in sediment samples and ten were detected in biota samples. Concentrations of total PAHs obtained in sediment and fish samples ranged from 2.15 - 36.46 mg/kg and 11.89 - 71.06 mg/kg respectively. Concentrations of total PAHs were higher in whole fish than in fillet samples (muscle) in both fish species. High values of PAHs were recorded in the dietary intake (0.10 - 2.33 mg/kg body weight/day) of the organisms. Toxic equivalent quotient values (0.01 to 0.10 mg/kg) observed were higher than the screening values (0.0014 to 0.0599 mg/kg). In the muscle of *Drepaneaficana* and *Pomadasysjubelini*, splitting and atrophy of the muscle bundles were observed. The result from

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the study showed that the concentrations of PAHs in analyzed sediment and organisms were higher than the maximum permissible limit of the United State Environmental Protection Agency (USEPA). The high concentration of total PAHs in the organisms indicated that the compounds have bioaccumulated in their tissues and organs over a period of time. The calculated TEQ values were higher than the screening values, indicating potential health risks. Histopathological examination revealed that both fish and shellfish were exposed to high concentrations of PAHs which brought about changes in morphologic structure of these organs, necrosis of the muscle bundles and cellular degeneration. Most of the detected PAHs were of petrogenic origin, which is an indication that anthropogenic activities influenced PAH concentrations

### 3.4 Vegetables

A study by [19] on Distribution and risk assessment of polycyclic aromatic hydrocarbons in vegetables and agricultural soils from two communities in Rivers State, Nigeria was also reviewed. Fresh samples of three commonly consumed vegetables: *Telfairia occidentalis* (*Fluted Pumpkin*), *Ocimum gratissimum* (Scent leaf) and *Vernonia amygdalina* (Bitter leaf) and 3 tubers: Cassava (*Manihot esculenta*), Cocoyam (*Colocasia esculenta*) and Yam (*Dioscorea rotundata*) were collected from farms in Alakahia and Eleme (Ogale) communities situated in Akpor and Eleme local government area respectively, Rivers State, Nigeria. At each site, six samples comprising of three vegetables and three tubers. Also, farm soil samples were collected from the 2 study areas namely Alakahia and Eleme communities. Soil samples were collected in aluminum foils and transported to the laboratory for analysis. The samples were prepared and extracted using Soxhlet extractor with a mixture of solvents. The extracts were cleaned, concentrated to 1  $\mu\text{L}$  and analyzed for PAHs with gas chromatography coupled with flame ionization detector (GC-FID). The result of the study revealed that a total of 14 and 16 PAHs were detected in the farm soil from Alakahia and Eleme respectively. The PAH concentration at both sites ranged from 0.00 – 47.45  $\mu\text{g}/\text{kg}$ . The researchers warned that although, the total PAH concentration in the vegetables were generally lower than the 10ppb recommended for foods of plant origin that there is still need for health concern as increased anthropogenic pressures and frequency of vegetable consumption by the populace in Alakahia

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and Eleme may increase the body burden of PAHs in the local populace. The result of the study also reported that carcinogenic Potency equivalent concentration ( $\mu\text{g}/\text{kg}$ ) were estimated to be 0.31- 1.51 and 0.37 - 0.97 for vegetables collected from Alakahia and Eleme communities respectively. These values exceeded the screening value ( $0.23\mu\text{g}/\text{kg}$ ) for vegetables, thus an indication that consumption of such vegetables is risky for the population of the study area.

The current levels of PAH in the vegetables and farm soils were not yet alarming as they are below the World Health Organisation(WHO) maximum permissible limit (put the value) for PAH in vegetables and soil. Considering the fact that these vegetables are still frequently consumed by the populace at Alakahia and Eleme, the authors advised that there is dire need to constantly monitor the PAH levels in farm soils and food crops over time as well as develop strict strategies to prevent PAH accumulation in food crops to minimize chronic health risk for the population.

Another study on the determination of polycyclic aromatic hydrocarbons in some foods from industrialized areas in South Eastern, Nigeria on human health risk impact by [18] was reviewed. Five samples each of twelve different food crops which include (i)vegetables: bitterleaf (*Vernonia amygdalina*), water leaf(*Talinum triangulare*) and pumpkin leaf (*Telfairia occidentalis*) (ii) tubers: yam (*Dioscoreaalata*), cocoyam (*Colocasia esculenta*) and cassava (*Manihot esculenta*); (iii) fruits: orange (*Citrus sinensis*), paw paw (*Carica papaya*) and star apple (*Chrysophyllum albidum*) (iv) nuts: kola nut (*Cola acumulata*), palm kernel nut (*Elaeisqueensiacq*) and coconut (*Cocos nucifera*) were harvested from farmlands closeto the industries (study sites) at Osisioma, Akwuuru, Ishiagu, Ngwo, and Irete. Michael Okpara University of Agriculture, Umudike farmland devoid of industries was used as control for this study. The samples were prepared for extraction. The PAHs were extracted from the samples using solvent extraction followed by clean up. The determination of PAHs was carried out using gas chromatography coupled with flame ionization detector (GC-FID). Recovery test was used to validate the extraction and analytical methods. The result revealed that the mean concentration of PAHs in food samples ranged from below detection limit ( $< 0.01$ ) to  $2.64 \pm 0.02$ ,  $5.27 \pm 0.04$ ,  $0.96 \pm 0.02$ ,  $8.94 \pm 0.01$  and  $1.95 \pm 0.06$  mg/kg respectively for Star apple collected from Osisioma, pumpkin leaf from Ishiagu, and bitter leaf from Irete, Akwuuru and Ngwo respectively. The authors attributed this to the uptake of PAHs by plant through gaseous (aerial)

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and particle bound deposition[34,35]. Total PAHs concentrations in most crop samples had highest values 14.49, 36.29, 4.59, 23.36 and 21.8 in mg/kg for *Chrysophyllum albidum*, *Telferia occidentalis*, *Vernonia amygdalina*, *Talinum triangulare* and *Elaiesguinnensis* for Abia (Osisioma), Anambra (Akwuuru), Imo (Irete), Ebonyi (Ishiagu) and Enugu (Ngwo) respectively. These values exceeded the European Union regulatory limit of 0.2 mg/kg body weight in foods suggesting possible health risk of exposure to PAHs through the food chain for the populace in these areas. The study revealed that vegetables from all the areas accumulated higher levels of PAHs significantly ( $p \leq 0.05$ ) compared to other classes of food crops analyzed. The authors attributed the abundance of PAHs in vegetables analyzed in the study to their relatively high solubility, volatility and bioavailability [36,37] as well as their large surface area which is constant in contact with air laden with dust and pollutants like PAHs [36]. The distribution of PAHs in this study had higher concentrations in *Vernonia amygdalina* (a vegetable with broad and rough-surface leaves) more than the other two vegetables *T. triangulare* and *T. occidentalis*, vegetables often consumed by the Igbos. The accumulation of PAHs in foods could be as a result of nearness of agricultural lands to the industrial/urban areas where high levels of anthropogenic activities occur [38]. Furthermore, differences in the concentration levels of PAHs generally could be as a result of differences in the bioaccumulation of plants and the proximity to the pollution source [39]. Human health risk assessment of benzo(a)pyrene (BaP) in the analyzed samples gave values above the DPR intervention limit of 0.01 mg/kg of B(a)P in some foods [39]. The incremental life time cancer risk has predicted permissible lifetime risks for carcinogens as  $10^{-6}$  (1 in 10,000,000) and  $10^{-4}$  (1 in 10,000) range [Ohiozebanu et al 2017; Zhang et al 2015; Nkpaa et al 2013; Chawda et al 2017; Wei et al 2011]. Values above these ranges indicate carcinogenic risk [9]. Considering study population, the carcinogenic risk resulting from consumption of the varieties of food crops for adults and children were 1.39 E-06, 1.54 E-06; 3.43 E-06, 3.79 E-06; 6.19 E-07, 6.87 E-07; 1.09 E-06, 1.21 E-06; and 9.71 E-07, 1.05 E-06 for Osisioma, Akwuuru, Irete, Ishiagu and Enugu industrial locations respectively. From the above, CR values were within the predicted life time risks for carcinogens for adults and children of Osisioma, Akwuuru and Ishiagu while CR values obtained for Irete and Enugu were within safe limits. These values suggested that CR exposure through the intake of food cultivated around the industrialized areas may be attributed to the type and activities peculiar to each industry such as petrochemical and mining activities at Osisioma Akwuuru and Ishiagu.

**Comment [NU29]:** PAHs levels significantly higher ( $p < 0.05$ ) than other classes of food crops analyzed.

**Comment [NU30]:** The distribution concentration of PAHs in this study was higher in .... relative to those of other one ..... often consumed .....

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The authors reported that adults and children residing at industrial areas in South Eastern, Nigeria may be at serious risk due to PAHs intake in foods overtime due to bioaccumulation of this toxic carcinogen. The researchers, therefore, recommended need for prompt action by policy makers and other concerned stakeholders to make regulations and policies mitigating measures for environment and human health protections.

#### 4.0. Conclusion

This study investigated on relevant information on the occurrence, concentrations, sources and associated potential human health risks of PAHs in foods consumed in Nigeria using reported results from selected studies. Majority of the selected studies reported PAH pollution occurring in various food consumed in Nigeria. While some studies reported PAHs values above permissible limits others reported PAHs values below the limit. From the studies reviewed, majority recorded values of margin of exposure (MOE) above 10000 indicating no human health risk concern while few recorded MOE values lower than 10000 indicating human health concern according to EFSA. The PAHs identified in samples from the environments were of both fuel combustion, petrogenic and pyrogenic sources. Researchers recommended mainly on the need for regular studies on the PAHs concentration levels and health risk assessment in other agricultural products as well as for policymakers and stakeholders to ensure environmental sanity. The reviewers of the present study hereby recommend further studies on other PAH groups such as alkylated PAHs and chlorinated PAHs considering their environmental relevance.

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**Comment [NU32]:** References are not numbered, remove the numbers in all the references. The authors of the second and third references are not the same so remove the 'a and b' in them. People's surname not only in references is followed by comma where the surname is written first ie before first name or initials. Do same in all the references.

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**Comment [NU34]:** Be consistent in your referencing. Use your own reference pattern. Study references 1 to 16 I did, that is the correct format. Whenever you bring out data from the body of the reference, remember to delete it from there.

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