Original Article

Risk Assessment of Polycyclic Aromatic Hydrocarbons in Pasta Products Consumed in Nigeria

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ABSTRACT

Background: The present study aimed to quantitatively determine polycyclic aromatic hydrocarbons in pastas consumed in Nigeria with the view of estimating the daily intake amount and the possible risks to consumers.

Methods: Sixteen polycyclic aromatic hydrocarbons (PAHs) were determined in locally produced and imported pasta using a GC-Mass Spectrometer. 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.

Results: The concentration of Σ 16 PAHs in Nigerian and imported brands were in the range of 9 to 800μg/kg and 2 to 7μg/kg, respectively. The benzo[a]pyrene (BaP) concentrations in 25% of Nigerian samples were above the maximum allowable level in processed cereal-based food. The concentrations of Σ8 carcinogenic PAHs in the Nigerian and imported brands ranged from 1 to 10μg/kg and 1 to 4μg/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.

Conclusions: The MOE values indicate serious concern particularly for children who are the major consumers of Nigerian pastas. This study is the first of its kind in Nigeria and can serve as a useful baseline for continuous monitoring of PAHs in the Nigerian pastas in order to ensure protection of human health in the country.

Key words: Estimated Daily Intake, Human Health, Margin of Exposure, PAHs, Pasta.

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INTRODUCTION

Contamination of foods by chemical toxins has become a major public health concern worldwide. Food contamination can stem from the environment, industrial food processing and certain home food preparation methods (1-3). Polycyclic aromatic hydrocarbons (PAHs) are one of the major organic contaminants in foods (4). Humans are exposed to PAHs through various pathways. For non-smokers, the major route of exposure is consumption of foods, which account for more than 90% of the total PAHs exposures of the general pollution in various countries (5-7). Food contamination may also occur during periods of atmospheric pollution, in which PAHs are deposited on seeds, fruits and vegetables, which are later consumed by people (1-3). Many studies have shown that cereals, vegetables (8) oil and fat (9-10) are amongst the contributors to PAHs intake by humans. Most food items and produce could be contaminated from soil polluted air and water (11). Some aquatic foods like fish, can be exposed to PAHs present in water and sediments, and the PAH content greatly depends on the ability of the aquatic organisms to metabolize them (12).

Furthermore, in food processing industry, such additives as smoke flavoring products (SFP), lubricants, solvents, propellants, glazing agents and protective coatings contribute to food contaminations by PAHs (10). The occurrence of PAHs in human foods reflects the environmental conditions and consequences of some thermal treatments that are used during the preparation and manufacturing of foods (13). The quantity and type of PAHs compounds produced by the thermal processing of foods depend on the temperature, oxygen availability, type of fuels used, treatment duration, fat contents and distance from the energy source (14). In remote areas, the levels of PAHs found in unprocessed foods reflect the background contamination, which originates from long distance airborne transportation of particles and natural emission from volcanoes and forest fires. In industrial areas or along highways, the contamination of vegetation can be ten-fold higher than that in rural areas (15).

Pastas are hard wheat product formed from unleavened dough, formed into different shapes (such as thin strips, tubes, or shells), and are usually boiled. The pastas that are commonly consumed in Nigeria include noodles, macaroni and spaghetti. They are quite popular because of the minimum time needed to prepare them, ease of preparation and low cost. These commodities serve as quick foods for children and adults in more than one third of homes in Nigeria and beyond (16). The import ban, changing consumption patterns, increasing demand for more nutritious and easy-to-cook foods, and the more expensive local substitutes have contributed to the growing demand for pasta products (17). The high demand for foods, due mainly to population growth and urbanization, severe shortage of time on the part of single people, and working mothers, and changes in feeding habits and way of life style have all combined to

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make the consumption of pastas very popular in Nigeria (17).

Using margin of exposure (MOE), toxic equivalency factor (TEF), benzo [a]pyrene (BaP) and several others have been proposed as markers for the risk assessment of PAHs present in foods (1,18). 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 (1). Toxic equivalency factor has been judged as being not scientifically valid because of the carcinogenic potency, the mode of action and lack of data from the oral carcinogenicity studies on various PAHs, based on the current proposed TEF values (1,5,11).

A review of the literature indicates that there is shortage of information on PAHs in pastas. Therefore, the objectives of this study were: *a)* to determine the levels of PAHs in some brands of locally produced and imported pastas in Nigerian market, and, *b)* to estimate the potential health risk involved in their consumption by adults and children. The findings will provide data and insight on formulating guidelines and standards for PAHs in foods in the Nigerian society.

MATERIALS AND METHODS

Sample Collection and Preparation

Different brands of locally manufactured and imported pastas were collected in November 2014 from two major cities in Nigeria (Abuja and Enugu). The choice of the samples was carefully made to reflect different origins and popular brands consumed by different income classes. The samples were categorized into three: *Noodles*, *Spaghetti* and *Macaroni*. A total of 60 samples were randomly collected. Each sample was ground into powder with a porcelain mortar and pestle, and sieved through thin fine cotton and labeled for analysis. The samples were stored at 4^{o}C prior to analysis.

Chemicals and Materials

All solvents were of high purity grade. n-Hexane and dichloromethane were supplied by JHD Co. (China). A chromatographic column was used to clean up of the extract, and was filled with 5g of 60-120 mesh silica gel obtained from Qualikems & Fine Chemicals Private Ltd. (India) and anhydrous sodium sulphate (BDH Chemicals, Poole, England). The Soxhlet extractor was purchased from Quick Fit (England) and the rotary evaporator from Senco Technology (China). The PAHs standard mixture (catalog #: Z-014G-R) containing 16 priority PAH compounds namely, Ace, Acy, Ant, BaA, BaP, BbF, BghiP, BkF, Chy, DahA, Flt, Flu, IndP, Nap, Phe and Pyr were obtained from a major supplier in New Haven, USA. The helium gas was obtained from Air Liquid Gas Company (France).

Samples Extraction and Clean-Up

Fifteen grams of the sample was homogenized with 5g of Na_2SO_4 until a completely dry homogenate was obtained. The homogenate was carefully transferred into the thimble and place in the extraction chamber of the Soxhlet extractor. The extractions were carried out, using 100 mL mixture of n-hexane and dichloromethane (1:1 V/V) for 16h. The extracts were evaporated to 2-3 mL, using a rotary evaporator and purified by passing through a column packed with silica gel. The column was pre-rinsed with mL of n-hexane and subsequently eluted with 50mL mixture of n-hexane and dichloromethane (1:1 V/V). The eluted extract were combined and evaporated to 1mL (19). The identification of PAHs was based on the comparison of the retention times of peaks with those obtained from standard mixture of PAHs. Quantification was based on external calibration curves prepared from the standard solution of each PAHs sample.

Gas Chromatography & Mass Spectrometry Conditions

PAHs analysis was carried out with an Agilent 7890, series A, gas chromatography unit (Avondale, USA) interfaced with a mass selective detector (5975 series MSD, Agilent, Avondale, USA). Separation of PAHs was performed using a 5% phenyl-methylsilicone (DB-5MS) bonded-phase fused-silica capillary column (30m x 250µm ID., film thickness 0.25µm; part #:19019J-413; Agilent, USA) at a temperature range of -60 to 350^{o}C. The injector port was run in splitless mode. The oven temperature was 65^{o}C for 1min, and rose finally to 290^{o}C at a rate of 10^{o}C/min and maintained at this temperature for 11min. The transfer line was maintained at 300^{o}C. Stock solutions were used to establish the retention time of each analyte. Detection of PAHs was carried out using scan mode, which was designed for preselected ion peaks; non-selected peaks were not identified and quantified. Helium was used as the GC carrier gas. The carrier helium was maintained at a constant pressure of 9.0855 Psi with a linear flow rate of 37.604 cm/sec.

Validation Method

To evaluate the extraction efficiency of the 16 PAH compounds, recovery analysis was performed. This was done by fortifying some samples with standard PAH mixture solution (New Haven, USA) before extraction was carried out as described earlier. The percent recoveries for the 16 PAH compounds are presented in Table 1. The uncertainty calculated for the PAHs values was ± 5%. The limit of detection (LOD) and limit of quantification (LOQ) were determined by the signal-to-noise method. The peak-to-peak noise for the analyte retention time was measured, and subsequently, the concentration of the analyte that yielded a signal equal to the noise-to-signal ratio *was* estimated. A signal-to-noise ratio of three is generally accepted for estimating LOD and signal-to-noise ratio of ten is used for estimating *LOQ (20).* The LOD (0.2µg/kg) and LOQ (0.7µg/kg) were obtained for acenaphthene, anthracene and phenanthrene while LOD (0.1µg/kg) and LOQ (0.3µg/kg) were obtained for the remaining PAH samples (Table 1).

Table 1. Mean % Recovery, LOD and LOQ of the PAHs.

PAHs	Certified concentrations (µg/mL)	% Mean Recovery	LOD (µg/kg)	LOQ (µg/kg)
Acenaphthene	2001	100±0.00	0.2	0.7
Acenaphthylene	2001	99.3±0.14	0.1	0.3
Anthracene	2001	99.4±0.28	0.2	0.7
Benz[a]anthracene (BaA)	2002	100±0.00	0.1	0.3
Benzo[a]pyrene (BaP)	2002	99.9±0.14	0.1	0.3
Benzo [b] fluoranthene (BbF)	2002	99.5±0.64	0.1	0.3
Benzo[g,h,i]perylene (BghiP)	2001	98.3±0.14	0.1	0.3
Benzo[k] fluoranthene (BkF)	2001	100±0.14	0.1	0.3
Chrysene (Chy)	2002	98.7±0.35	0.1	0.3
Dibenz[a,h]anthracene (DahA)	2002	98.1±0.07	0.1	0.3
Fluoranthene	2059	97.4±0.21	0.1	0.3
Fluorene	2002	99.2±0.14	0.1	0.3
Indeno[1,2,3- c,d]pyrene (IndP)	2061	97.1±0.35	0.1	0.3
Naphthalene	2002	99.6±0.07	0.1	0.3
Phenanthrene	2001	99.4±0.21	0.2	0.7
Pyrene	2002	99.1±0.07	0.1	0.3

Data Analysis

Analysis of variance (ANOVA) was used to determine whether the concentrations of the PAHs varied significantly within and between the groups, with values less than 0.05 ($P<0.05$) considered to be statistically significant. Data were analyzed using SPSS version 16.0 for Windows.

Daily Intake & Risk Assessment

The dietary intakes of the 16 PAHs were estimated, using a deterministic approach. A fixed value for the consumption of an individual food was multiplied by a fixed value for the contaminant concentration in that food (21). The total exposure was obtained by summing the intakes from all foods, using the following equation:

Estimated daily Intake (EDI) =

$$\frac{\sum(consumption\ rate \times occurrence)}{Body\ weight}$$

Body weight for children = 15 Kg and body weight for adults = 60 Kg were assumed (22).

Since the general population cannot consume the same portion size of pastas, the intake was estimated for two categories of exposures: generally exposed individuals (average consumers) and typically exposed individuals (high consumers). For generally exposed individuals, a small portion size was used while for typically exposed individuals, a higher portion size was used depending on the pack size of the pasta. The risk was estimated using the Margin of Exposure (MOE) approach according to the following equation (23):

$$MOE = \frac{BMDL_{10}}{EDI}$$

Where $BMDL_{10}$ is the benchmark dose lower confidence limit at 10% incidence level. Considering a $BMDL_{10}$ of 0.07, 0.17, 0.34 and 0.49 all in mg/kg bw per day for BaP, PAH2, PAH4 and PAH8, respectively, for adult and children scenario, where:

BaP = Benzo[a] pyrene

PAH2 = Benzo [a]pyrene and chrysene

PAH4 = Benzo [a]anthracene, benzo[a] pyrene, benzo [b] fluoranthene and chrysene

PAH8 = The sum of eight carcinogenic PAHs: benzo [a] anthracene; benzo [b]fluoranthene; benzo [k] fluoranthene; benzo[g,h,i]perylene; benzo [a]pyrene; chrysene; dibenz[a,h]anthracene; and indeno[1,2,3-C,d] pyrene.

RESULTS

The mean concentrations of the individual PAH compounds in different brands of Nigerian pastas were presented in Table 2. The concentrations of Σ16 PAHs in these pastas ranged from 9 to 800µg/kg. The lowest mean concentration of Σ16 PAHs was found in brand F spaghetti and the highest mean concentration of Σ16 PAHs was found in brand B noodles. The concentrations of Σ16 PAHs varied significantly ($p<0.05$) among different brands of pasta as well as within a particular brand. Benzo [a]pyrene (BaP) was detected in all of the eight brands of pastas examined at concentrations of 0.4 to 3µg/kg. The highest BaP concentration was observed in brand C noodles, and BaP constitutes 0.8% of the Σ16 PAHs in the sample. The concentrations of PAH2, PAH4 and PAH8, suitable indicators for the occurrence of total PAHs, are also presented in Table 1. The concentrations of PAH2, PAH4 and PAH8 in these brands of pasta ranged from 0.4 to 4µg/kg, 1 to 100µg/kg, and 1 to 10µg/kg, respectively. Brands A and B noodles had the highest concentrations of the 8 carcinogenic PAH compounds than other brands. Non-carcinogenic PAHs (napthalene, acenanthracene, fluoranthene, fluorene, phenanthrene, anthrancene, fluoranthene and pyrene) were present in high proportions, ranging from 1 to 300µg/kg in different brands analyzed. Benzo (g,h,i) perylene, Dibenz [a,h] anthracene and Indeno [1,2,3-C,d] pyrene were not detected in the Nigerian pastas.

The mean concentrations of the individual PAH compounds in different brands of imported pastas are presented in Table 3. The concentrations of Σ16 PAHs in these pastas ranged from 0.002 to 0.007 mg/kg. The lowest mean concentration of Σ16 PAHs was found in brand M macaroni. Brand L spaghetti and brand O macaroni equally had the highest mean concentration of the Σ16 PAHs. Benzo [a]pyrene (BaP) was detected in all of the imported brands of pastas examined at concentrations of 0.0002 to 0.0007 mg/kg, which was lower than those obtained in the Nigerian pastas (0.0002 to 0.003 mg/kg). The highest BaP concentration was found in brand O macaroni, and constituted 10% of the Σ16 PAHs in this brand of pastas. The concentrations of PAH2, PAH4 and PAH8 are also presented in Table 3. The concentrations of PAH2, PAH4 and PAH8 in the imported brands ranged from 0.0002 to 0.003 mg/kg, 0.0004 to 0.002, and 0.001 to 0.004 mg/kg, respectively. Brand O macaroni's had the highest concentration of the eight carcinogenic PAH compounds than did other brands analyzed in this study. Benzo [g,h,i] perylene, Dibenz [a,h] anthracene and Indeno [1,2,3,- C, d] pyrene were not detected in different brands of imported pastas.

Table 4 presents the daily intakes of suitable indicators for the occurrence of total PAH such as BaP, PAH2, PAH4, and PAH8 of generally exposed children and adults who consumed Nigerian and imported pastas while Table 5 reflects the estimates of BaP, PAH2, PAH4 and PAH8 daily intakes of typically exposed individuals who consumed Nigerian and imported pastas. Tables 6 and 7 present the margin of exposure (MOEs) for typically exposed persons consuming Nigerian pastas, respectively.

Table 2. Mean concentrations (µg/Kg) of polycyclic aromatic hydrocarbons in different brands of Nigerian pastas.

	NOODLES				**SPAGETTI**		**MACARONI**	
	A	**B**	**C**	**D**	**E**	**F**	**G**	**H**
PAH	**Mean±SD**	**Mean ±SD**	**Mean ±SD**	**Mean±SD**	**Mean ±SD**	**Mean ± SD**	**Mean ±SD**	**Mean±SD**
Naphthalene	30.0±30.0	100.0±10.0	50.0 ± 10.0	20.0 ± 0.0	6.0 ± 0.7	5.0 ± 1.0	30.0±20.0	7.0 ± 0.0
Acenaphthylene	30.0 ± 0.0	40.0 ± 0.0	10.0 ± 10.0	6.0 ± 1.0	0.3 ± 0.2	0.4 ± 0.0	0.1 ± 0.0	7.0 ± 1.0
Acenaphthene	20.0± 10.0	20.0± 10.0	5.0 ± 0.7	2.0 ± 1.0	ND	ND	ND	0.3 ± 0.1
Fluorene	60.0 ± 7.0	50.0 ± 0.0	50.0 ± 20.0	6.0 ± 6.0	3.0 ± 0.7	ND	0.4 ± 0.2	2.0 ± 1.0
Phenanthrene	100.0±130.0	300.0±30.0	100.0 ± 7.0	30.0 ±40 .0	20.0 ± 20.0	ND	0.4 ± 0.0	1.0 ± 0.0
Anthracene	100.0±130.0	300.0±20.0	200.0 ± 0.0	30.0 ± 40.0	2.0 ± 0.0	ND	0.5 ± 0.1	3.0 ± 2.0
Flouranthene	3.0 ± 1.0	1.0 ± 0.7	4.0 ± 0.0	0.2 ± 0.1	1.0 ± 0.0	0.5 ± 0.1	0.5 ± 0.2	0.4 ± 0.0
Pyrene	30.0 ± 7.0	50.0± 20.0	3.0 ± 0.7	5.0 ± 1.0	5.0 ± 1.0	0.2 ± 0.0	0.2 ± 0.1	0.6 ± 0.1
Benzo [a]anthracene	7.0 ± 1.0	5.0 ± 0.0	4.0 ± 1.0	0.6 ± 0.0	3.0 ± 3.0	0.6 ± 0.1	1.0 ± 0.0	0.3 ± 0.3
Chrysene	2.0 ± 1.0	2.0 ± 10.0	1.0 ± 0.0	0.1 ± 0.0	0.7 ± 0.0	0.2 ± 0.1	0.2 ± 0.0	0.5 ± 0.1
Benzo [b]flouranthene	2.0 ± 0.7	1.0 ± 0.0	0.7 ± 0.2	0.2 ± 0.1	0.7 ± 0.1	0.1 ± 0.0	2.0 ± 3.0	0.5 ± 0.0
Benzo [k]fluoranthene	1.0± 0.0	2.0± 0.7	0.2 ± 0.1	0.3 ± 0.1	0.8 ± 0.0	0.5 ± 0.1	0.6 ± 0.1	ND
Benzo [a]pyrene	0.3 ± 0.0	0.7 ± 0.1	3.0 ± 2.0	0.4 ± 0.2	2.0 ± 2.0	0.7 ± 0.1	0.2 ± 0.0	0.2 ± 0.1
Benzo [g,h,i]perylene	ND	ND	ND	ND	ND	ND	ND	ND
Dibenz [a,h]anthracene	ND	ND	ND	ND	ND	ND	ND	ND
Indeno[1,2,3 – c,d]pyrene	ND	ND	ND	ND	ND	ND	ND	ND
$\sum$ 16 PAHs	300.0±40.0	800.0±30.0	400.0 ±40.0	200.0 ±10.0	80.0 ± 40.0	9.0 ± 2.0	30.0±30.0	60.0 ± 10.0
PAH2	2.0 ± 1.0	3.0 ± 1.0	4.0 ± 2.0	0.5 ± 0.2	3.0 ± 1.0	0.9 ± 0.1	0.4 ± 0.0	0.7 ± 0.0
PAH4	10.0 ± 0.0	9.0 ± 1.0	8.0 ± 3.0	1.0 ± 0.0	7.0 ± 1.0	2.0 ± 0.7	4.0 ± 2.0	2.0 ± 0.7
PAH8	10.0 ± 0.0	10.0 ± 2.0	8.0 ± 3.0	1.0 ± 0.0	7.0 ± 4.0	2.0 ± 1.0	5.0 ± 2.0	2.0 ± 0.7
$\sum LMW - PAHs$	300.0±400.0	700.0±60.0	400.0 ±40.0	200.0 ± 7.0	60.0 ± 30.0	5.0 ± 1.0	7.0 ± 4.0	40.0 ± 20.0
$\sum HMW - PAHs$	40.0 ± 0.0	60.0± 20.0	100.0 ± 100.0	5.0 ± 7.0	10.0 ± 8.0	4.0 ± 0.7	0.0±0.0	30.0± 40.0

ND = Not Detectable

Table 3. Mean concentrations (µg/kg) of polycyclic aromatic hydrocarbons in different brands of imported pastas.

	NOODLES		**SPAGETTI**		**MACARONIE**		
	I	**J**	**K**	**L**	**M**	**N**	**O**
PAH	**Mean ±SD**	**Mean ±SD**	**Mean ±SD**	**Mean ±SD**	**Mean ±SD**	**Mean ± SD**	**Mean ±SD**
Naphthalene	4.0 ±0.7	ND	3.0 ± 1.0	0.5 ± 0.2	0.2 ± 0.0	4.0 ± 0.7	0.2 ± 0.1
Acenaphthylene	0.2 ± 0.0	ND	ND	0.4 ± 0.2	ND	ND	0.1 ± 0.0
Acenaphthene	ND	ND	ND	ND	ND	0.4 ± 0.3	ND
Fluorene	0.1 ± 0.1	ND	ND	3.0 ± 0.7	ND	ND	ND
Phenanthrene	ND	1.0 ± 0.0	ND	0.7 ± 0.0	0.5 ± 0.1	ND	0.3 ± 0.1
Anthracene	ND	2.0 ± 1.0	ND	0.7 ± 0.1	0.2 ± 0.0	ND	0.3 ± 0.1
Flouranthene	ND	0.3 ± 0.1	ND	0.4 ± 0.1	0.6 ± 0.1	ND	0.4 ± 0.3
Pyrene	ND	0.1 ±0.0	ND	0.2 ± 0.0	0.3 ± 0.0	ND	0.2 ± 0.1
Benzo [a]anthracene	0.2 ± 0.0	0.5 ± 0.1	ND	0.3 ± 0.0	1.0 ± 0.0	0.2 ± 0.1	0.4 ± 0.3
Chrysene	ND	0.1 ± 0.0	ND	ND	ND	ND	0.3 ± 0.0
Benzo [b]flouranthene	0.7 ± 0.2	0.5 ± 0.1	0.4 ± 0.1	0.2 ± 0.1	0.6 ± 0.1	0.6 ± 0.1	0.5 ± 0.1
Benzo [k]fluoranthene	0.3± 0.1	0.4 ± 0.0	0.8 ± 0.0	0.8 ± 0.2	0.6 ± 0.0	0.7 ± 0.0	3.0 ± 1.0
Benzo [a]pyrene	0.3 ± 0.0	0.3 ± 0.1	0.5 ± 0.1	0.2 ± 0.1	0.6 ± 0.3	0.4 ± 0.2	0.7 ± 0.3
Benzo [g,h,i]perylene	ND	ND	ND	ND	ND	ND	ND
Dibenz [a,h]anthracene	ND	ND	ND	ND	ND	ND	ND
Indeno [1,2,3 – c, d] pyrene	ND	ND	ND	ND	ND	ND	ND
$\sum$ 16 PAHs	6.0 ± 0.7	5.0 ± 1.0	5.0 ± 1.0	7.0 ± 0.7	2.0 ± 3.0	6.0 ± 2.0	7.0 ± 2.0
PAH2	0.3 ± 0.0	0.4 ± 0.1	5.0 ± 0.1	0.2 ± 0.1	0.6 ± 0.3	0.4 ± 0.2	1.0 ± 03
PAH4	0.9 ± 0.1	1.0 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	2.0 ± 0.0	0.8 ± 0.1	0.9 ± 01
PAH8	1.0 ± 0.0	1.0 ± 0.0	1.0 ± 0.0	1.0 ± 0.0	3.0 ± 0.0	2.0 ± 0.7	4.0 ± 1.0
$\sum LMW - PAHs$	0.4 ± 0.1	3.0 ± 1.0	3.0 ± 1.0	5.0 ± 0.7	0.6 ± 0.4	3.0 ± 3.0	0.8 ± 0.3
$\sum HMW - PAHs$	0.4 ± 0.4	2.0 ± 0.0	2.0 ± 0.0	2.0 ± 0.0	4.0 ± 0.7	2.0 ± 3.0	6.0 ± 2.0

ND = Not Detectable

Table 4. Estimated daily intakes (ng/kg bw/day) of generally exposed individuals on Nigerian and imported pasta.

Pasta	Brand	Portion size (g)	CHILDREN*				ADULTS**			
			Bap	PAH2	PAH4	PAH8	Bap	PAH2	PAH4	PAH8
Nigerian										
Noodles	A	70	1.4	11	50	5.5	0.4	2.7	13	14
	B	70	3.3	13	41	48	0.8	3.2	10	12
	C	70	9.3	5.6	27	28	0.2	1.4	6.8	7.0
	D	70	1.6	2.1	6.1	7.5	0.4	0.5	1.5	1.9
Spaghetti	E	62.5	8.3	11	27	30	2.1	2.8	6.7	7.5
	F	62.5	2.9	4.0	6.9	9.0	0.7	10	1.7	2.2
Macaroni	G	62.5	0.8	1.7	14	16	0.2	0.5	3.5	4.1
	H	62.5	0.6	2.9	18	18	0.2	0.7	4.4	4.4
Imported										
Noodles	I	100	2.0	2.0	8.3	10	0.5	0.5	2.1	2.6
(Thailand)	J	75	1.3	1.8	6.5	8.5	0.3	0.4	1.6	2.1
Spaghetti	K	62.5	1.9	1.9	3.5	6.9	0.5	0.5	0.9	1.7
(Turkey)	L	62.5	0.8	0.8	2.7	5.8	0.2	0.2	0.7	1.5
Macaroni	M	62.5	2.5	2.5	5.4	7.9	0.6	0.6	1.4	2.0
(France)	N	62.5	1.9	1.9	5.0	7.9	0.5	0.5	1.3	2.0
	O	62.5	3.1	4.4	7.7	9.0	0.8	1.1	1.9	2.2

* **Body weight** (bw) of a child (15 Kg); **Body weight (bw) of an adult (60 Kg)
PAH 2 = Benzo [a] pyrene + chrysene
PAH 4 = PAH2 + Benzo [a]anthracene + benzo [b]flouranthene
PAH 8 = PAH4 + benzo [k]flouranthene + benzo [g,h,i]perylene + dibenz [a,h]anthracene + indno [1,2,3,- C,d]perylene

Table 5. Estimated daily intakes (ng/kg bw/day) of typically exposed individuals on Nigerian and imported pasta

Pasta	Brand	Portion size (g)	CHILDREN				ADULTS			
			Bap	PAH2	PAH4	PAH8	Bap	PAH2	PAH4	PAH8
Nigerian										
Noodles	A	120	2.4	18	86	9.4	0.6	4.6	22	24
	B	120	5.6	22	70	82	1.4	5.4	17	20
	C	120	1.6	9.6	46	48	0.4	2.4	12	12
	D	120	2.8	3.6	10	13	0.7	0.9	2.6	3.2
Spaghetti	E	125	17	23	5.3	60	4.2	5.6	13	15
	F	125	5.8	7.9	1.4	18	1.5	2.0	3.4	4.5
Macaroni	G	125	1.7	3.3	28	33	0.4	0.8	7.1	8.3
	H	125	1.3	5.8	35	35	0.3	1.5	8.8	8.8
Imported										
Noodles	I	200	4.0	4.0	17	21	1.0	1.0	4.2	5.2
(Thailand)	J	150	2.5	3.5	13	17	0.6	0.9	3.3	4.3
Spaghetti	K	125	3.8	3.8	7.1	14	0.9	0.9	1.8	3.4
(Turkey)	L	125	1.7	0.2	5.4	12	0.4	0.4	1.4	2.9
Macaroni	M	125	5.0	5.0	13	16	1.3	1.3	2.7	4.0
(France)	N	125	3.8	3.8	10	16	0.9	0.9	2.5	4.0
	O	125	6.3	8.8	15	18	1.6	2.2	3.9	4.5

Table 6. Margin of Exposure (MOE) of generally exposed individuals on Nigerian and imported pastas.

Pasta	Brand	CHILDREN				ADULTS			
		Bap	PAH2	PAH4	PAH8	Bap	PAH2	PAH4	PAH8
Nigerian									
Noodles	A	50000	15455	6800	8909	200000	62963	26154	35000
	B	12112	13077	7083	10208	85366	53125	34000	40833
	C	7527	30357	12593	17500	304348	121429	50000	70000
	D	43750	80952	55738	65333	333333	320755	226667	257895
Spaghetti	E	8434	15455	12593	16333	33333	60714	50746	65333
	F	24138	42500	49275	54444	95890	171717	200000	222727
Macaroni	G	24337	100000	24286	30625	333333	346939	97143	119512
	H	111111	58621	18889	27222	437500	232877	77273	111364
Imported									
Noodles	I	35000	85000	40964	49000	140000	340000	161905	188462
(Thailand)	J	53846	94444	52308	57647	225806	386364	212500	233333
Spaghetti	K	36842	89474	97143	71014	148936	361702	382022	288235
(Turkey)	L	84337	204819	125926	84483	333333	809524	500000	326667
Macaroni	M	28000	68000	62963	62025	111111	269841	242857	245000
(France)	N	36842	89474	68000	62025	148936	361702	261538	245000
	O	22581	38636	44156	54444	89744	1545455	178947	222727

Table 7. Margin of exposure (MOE) of exposed individuals using Nigerian and imported pastas.

Pasta	Brand	CHILDREN				ADULTS			
		Bap	PAH2	PAH4	PAH8	Bap	PAH2	PAH4	PAH8
Nigerian									
Noodles	A	29167	9444	3953	5213	116667	36957	15455	20417
	B	12500	7727	4857	5976	50000	3148	20000	24500
	C	43750	17708	7391	10208	175000	70833	28333	40833
	D	25000	47222	34000	37692	100000	188889	130769	153125
Spaghetti	E	4118	7391	64151	8167	16667	30357	26154	32667
	F	12069	21519	242857	27222	46667	85000	100000	108889
Macaroni	G	41176	51515	12143	14848	166667	204819	52113	59036
	H	53846	29310	9714	14000	225806	113333	42045	55682
Imported									
Noodles	I	17500	42500	20000	23333	70000	170000	80952	94231
(Thailand)	J	28000	48571	26154	28824	111111	193182	103030	113953
Spaghetti	K	18421	44739	47887	35000	74468	180851	188889	144118
(Turkey)	L	41176	100000	62963	40833	166667	404762	242857	168966
Macaroni	M	14000	34000	26154	30625	53846	130769	125926	122500
(France)	N	18421	44737	34000	30625	74468	180851	136008	122500
	O	11111	19318	22667	27222	43750	77273	87179	108889

DISCUSSION

The differences observed in the concentrations of Σ16 PAHs (Table 2) among the different brands of pasta as well as within a particular brand could be attributed to variations in temperature, treatment duration (time), distance from the energy (heating) source, oxygen availability, fat content and the fuel type used during the production of these pastas (14). Benzo[a] pyrene is the most carcinogenic PAH compound and makes for the occurrence of PAHs in foods (5). European Community (EC) regulation has set the maximum allowed level (ML) for benzo[a]pyrene in processed cereal-based food as 1µg/kg (1). In this study, 75% of the brands examined had BaP concentrations below the maximum allowable level for BaP in processed cereal-based foods while 25% had BaP concentrations above the maximum allowable limit. EFSA (1) reported the upper bounds for the mean concentration of Bap in processed cereal food as 0.26 µg/kg while BaP concentrations of 0.1 to 0.4 µg/kg have been reported in biscuits (22). Also, BaP (0.03-0.49 µg/kg) and PAH4 (0.07-3.3 µg/kg) has been reported in cereal products, bread and flour in UK (24). The total PAHs in locally produced pasta in this study (9-800 µg/kg) were higher than the value (14.454µg/kg) reported in cereals in Catalonia (25). The predominance of non-carcinogenic PAHs in the pastas is in agreement with previously published reports (26,27). Low molecular weight PAHs were dominant in locally produced pastas. The non-detection of Benzo[g,h,i]perylene, Dibenz[a,h]anthracene and Indeno[1,2,3,C,d]pyrene in the locally produced pastas was not consistent with what was reported in imported biscuits, where the above PAH compounds were detected (22).

In this study, all of the imported pastas (Table 3) we examined, had Bap concentrations at levels below 0.001mg/kg, the maximum allowable level in processed cereal based foods (1). Non-carcinogenic PAHs were present in low concentrations ranging from undetectable to 0.004 mg/kg compared to what was reported in the Nigerian brands of pasta (0.001 to 0.3 mg/kg). Low molecular weight PAHs have almost the same concentrations compared with high molecular weight PAHs. This differs from what was obtained in the Nigerian pastas where the low molecular weight PAHs were approximately 10-fold higher than that for the high molecular weight PAHs.

As seen in Table 4, brand D noodle was the most important source of BaP, contributing 33% of the estimated total daily intake of children followed by brand E spaghetti (29%). For PAH2, brand B noodle was the major contributor (25%) followed by brand A noodle and brand E spaghetti, whith an equal contribution (21%). Brand A noodle was the major contributor (26%) of PAH4 followed by brand B noodle (22%), brand E spaghetti and brand C noodle, with equal contribution (14%) for PAH4. The most important contributors of daily PAH8 intake were brand B noodles (30%), followed by brand E spaghetti (19%) and brand C noodle (17%) of the total daily intakes. As for adult scenario, brand E spaghetti (42%) was the major contributor of BaP for the total daily intakes of 5 x 10^{-6} mg/kg bw/day, followed by brand B noodle (16%) and brand F spaghetti (14%). Brand F spaghetti was the major source of PAH2, contributing to 46% of the total daily intakes. Also, brands A and B noodles were the major contributors with 27% and 21% of PAH4, respectively. This was similar to what has been reported in cookies and short breads, as being the major contributors of PAH4 (22). Brands A and B noodles were also the major contributors of PAH8.

For the imported pastas (Table 4), the children scenario for generally exposed persons, brand L spaghetti was the least important source of BaP, PAH2, PAH4 and PAH8, contributing 6%, 5%, 7% and 10%, respectively. For the adults' scenario, brand O macaroni was the most important source of BaP, PAH2, PAH4 and PAH8, contributing 24%, 29%, 19% and 16%, respectively.

The estimated daily intakes obtained for typically exposed individuals (Table 5) showed that for the children scenario in Nigerian pastas, brand E spaghetti was the most important source of BaP with the daily intake of 17x 10^{-6} mg/kg bw/day, contributes 44% of the total daily intakes of 38.2 x 10^{-6} mg/kg, which was about four folds the daily intakes of generally exposed individuals (8.3 x 10^{-6} mg/kg bw/day) who consumed Nigerian pastas. For PAH2, brand A noodles (19%), brand B noodles (23%) and brand E spaghetti (24%) were the major contributors. The total daily intakes of PAH4 were 281.7x 10^{-6}mg/kg bw/day, which was higher than that for generally exposed individuals (1.9 x 10^{-4} mg/kg bw/day) who consumed Nigerian pastas. Brand A noodles (30%) and B noodles (25%) were the major contributors of PAH4. Brand B noodles (27%) and brand E spaghetti (20%) were the most important sources of PAH8. For the adults' scenario, brand E spaghetti was the major contributor of BaP. The major contributors to PAH2 were brand E spaghetti (24%), followed by brand B noodle (23%) and brand A noodle (19%). The most important contributors of PAH4 were brand A noodle (26%), brand B noodle (19%), followed by brand E spaghetti (15%) and brand C noodle (13%). Brands A (25%) and B (21%) noodles were the major contributors of PAH8. In children's case for imported pastas, brand L spaghetti was the least contributor of BaP (6%), PAH2 (1%), PAH4 (7%) and PAH8 (11%), with total daily intakes of 2.7 x 10^{-5} mg/kg bw/day, 2.9 x 10^{-5} mg/kg bw/day, 8.0 x 10^{-5} mg/kg bw/day and 1.1 x 10^{-5} mg/kg bw/day, respectively.

Reports have shown that children risks are always higher than those for adults (28, 29). The dietary intakes of PAHs are usually expressed in micrograms per kilogram of body weight per day and thus are age dependant. Falco *et al* (25) have reported a higher intake of total PAHs from food for children (0.307µg/kg/day) than that for senior adults (0.102µg/kg/day). Reports have also shown that cereals make the largest contribution to the total dietary intake of PAHs compared to other food products (25,30). The source of PAHs in foods, especially cereals, could come from anthropogenic inputs into agricultural soils, air and water. Also, certain types of processing such as smoking, roasting and drying by wood fire or oven can lead to the formation of PAHs in foods (25).

As shown in Tables 6 and 7, children consuming Nigerian brands A-E pastas are at risk of exposure to PAH4 and PAH8 since these brands have MOE values below 10,000. However, for generally exposed adult consumers of Nigerian pastas, the MOE values were far higher than 10,000, suggesting that their daily intake of PAHs is of low health concern.

Based on the BaP content and MOE values of 13% of the pasta brands being below 10,000 (Table 6), the consumption of these foods is a cause for public health concern in Nigeria. However, using PAH2, PAH4 and PAH8 as indicators for occurrence of PAHs in foods, 38%, 50% and 50% of the pastas, respectively, had their MOE values below 10,000. For the adults' scenario, 13% of the brands had MOE values less than 10,000 for PAH2 while none of the brands had an MOE value less than 10,000 for BaP, PAH4 and PAH8. These values are less than that reported for the children and adults scenarios for biscuits (22). The Food Standard Agency (24) reported the mean MOE value of all PAHs in cereal and vegetables ranging from 267700-323800 (adults) and 119700-145900 (children) in the UK. They concluded that exposure assessments for cereals and vegetables for different groups provided a minor contribution to the total exposure of PAHs and the MOE values indicated low health concerns. Generally, exposed children and adult consuming imported pastas in this study showed low health concern as the observed MOE values were above 10,000. From the MOE values for typically exposed children and adults, all the different brands of pastas had MOE value greater than 10,000 for all the indicators of risk assessment, suggesting that the exposure to BaP, PAH2, PAH4 and PAH8 is of low public health concern.

CONCLUSION

The concentrations of Σ16 PAHs, ΣLMW-PAHs and ΣHMW – PAHs in different brands of Nigerian pastas were generally higher than that in imported pastas. However, 75% of the Nigerian brands and 100% of imported brands had BaP concentrations less than 0.001mg/kg permissible limit specified for processed cereal foods by the European Commission. PAH2, PAH4 and PAH8 which are also markers of carcinogenicity were found in higher amounts in Nigerian brands of pastas compared to imported brands. The MOE values for generally exposed individuals were lower than 10,000 in 25% of children and none of the adults for Nigerian brands, based on PAH8 concentration while for typically exposed individuals, it were 38% and 0% for child and adult scenarios, respectively. For imported brands of pastas, the MOE values were found to be far higher than 10,000 for generally and typically exposed individuals in both children and adults scenarios. According to the European Food Safety Authority (EFSA), a MOE value less than 10,000 indicates a potential concern for the consumer's health. Therefore, there is a potential concern for individuals who consume certain Nigerian brands of pastas especially for children, and a low concern for consumers (children and adults) of imported brands of pastas. The levels of PAHs found in these samples demonstrate the need for setting allowable limits for PAHs in Nigerian foods. Continuous demand for analytical monitoring and increased research efforts concerning Nigerian brands should be advocated to ensure adequate protection of human health and the risk management actions.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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