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Polycyclic Aromatic Hydrocarbon and Metal Concentrations in Imported Canned Maize

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A B S T R A C T

Concentrations and profile of polycyclic aromatic hydrocarbons(PAHs) and metals (Cd, Pb, Ni, Cr, Fe and Mn) were determined in selected brands of canned maize in the Nigeria market with a view to providing information on the hazards associated with the consumption of these products. The measurement of the concentrations of PAHs was carried out by using a gas chromatography equipped with flame ionization detector (GC-FID) after extraction by ultra-sonication with acetone/dichloromethane and clean-up. The 16 PAH concentrations varied between 45.1 and 335.7 µg/kg. The concentrations of the indicators for occurrence and effects of PAHs in food varied from 3.6 to 114.5 µg/kg for BaP, 6.4 to 168.2 µg/kg for PAH2, 11.8 to 232.7 µg/kg for PAH4 and 19.4 to 327.3 µg/kg for PAH8. The concentrations of metals were determined by using atomic absorption spectrometry after acid digestion. The concentrations of metals in these samples ranged from <0.05 to 0.9 µg/g for Cd; 5.0 to 8.0 µg/g for Pb, 0.8 to 1.7 µg/g for Fe while Cr and Mn were less than the limits of quantification (<0.05 µg/g). The concentrations of Cd and Pb in these canned maize samples were above their permissible limits for foods.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a diversified group of over 100 organic compounds with two or more aromatic and/pentacyclic rings in a linear, angular or cluster formation (Tuteja et al., 2011; Martorell et al., 2011), and a number of which have proven carcinogenic, mutagenic and genetoxic properties. PAHs are ubiquitous environmental contaminants which are mainly produced from the incomplete combustion or pyrolysis of organic materials and natural combustion such as volcanic eruptions and forest fires. Other anthropogenic sources include emissions from the industries, waste incineration, road traffic and heating system (Naccari et al., 2011). PAHs have relatively low water solubility but are highly lipophilic, low biodegradability and are capable of undergoing long-range atmospheric transportation and deposition. Because of these characteristics, PAHs are likely to cause adverse human health or environmental effects near to and distance from their source (Protocol on Persistent Organic Pollutant, 1999; Naccari et al., 2011).

PAHs occur in foodstuffs as a consequence of environmental contamination and the thermal processes to which the foods are subjected to during processing and manufacturing. Food processing methods such as drying, boiling, cooking, frying, grilling, roasting and smoking are recognised sources of PAHs in foods (Naccari et al., 2011; Marteroll et al., 2010; Ciecierska and Obiedzinski, 2007).

The United States Environmental Protection Agency (USEPA) and European Union have identified the following 16 most frequently occurring/or dangerous PAHs as priority pollutants that needed to be monitored in environmental matrices. They include naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chy), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)perylene (IndP), dibenzo(a,h)anthracene (DahA) and benzo(ghi)perylene (BghiP). The International Agency for Research on Cancer (IARC) has further classified benzo(a)pyrene as group 1A (carcinogenic to humans), dibenzo(a,h)anthracene group 2A (probably carcinogenic to humans) and benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene and indeno(1,2,3-cd)perylene as group 2B (possibly

carcinogenic to human), while other are not classifiable as their carcinogenicity to humans (IARC, 2010).

Metal ions play significant roles in human health and diseases which ranged from the dietary requirement of essential trace elements and toxicity associated with the overload of essential and toxic metals (Hague et al. 2008). These reasons have prompted research in the determination of metal concentrations in food items. Some metals such as Hg, Cd, and Pb cannot be tolerated even at low levels because they are exceptionally toxic to humans (Suppin et al., 2006). For example, cadmium accumulates in human body and may induce kidney dysfunction, skeletal damage, hypertension, tremor and hepatic and reproductive deficiencies. Also, Cadmium is a cell poison that causes different types of damages, including cell death or an increase in cell proliferation. For this reason, the International Agency for Research in Cancer (IARC) has categorized Cd as a group 2A carcinogen (FDA, 2001). Lead absorption may constitute a serious risk to public health. Lead can inhibit cognitive development and intellectual performance in children, and increased blood pressure and cardiovascular disease in adult (Suppin et al. 2006). Exposure to lead has been associated with slow growth, hyperactivity, antisocial behaviours and impaired learning and hearing (Dahiya et al. 2005). On the other hand, chromium, cobalt, copper, iron, zinc and manganese are essential for human health. However, for metal micronutrients, there are fixed allowed levels for adequate dietary intake. At high concentrations, chromium, zinc cause nephritis anuria and extension of lesions in the kidney (Iwegbue et al. 2009; Iwegbue et al. 2010). In addition to the endogenous metal ion content of foodstuffs, numerous steps during processing and packing may add to the metal load. This is exemplified by authorised used of many metal containing additives such as the 'anti-oxidant' stannous chloride (Hague et al. 2008).

Canning makes food available for consumption far away from production sites. Trans-boundary movement of food products requires proper scrutiny. Thus constant monitoring of food products is needed to ensure the levels of these contaminants does not exceed their stipulated maximum limits. A survey of literature reveals that limited information are currently available on the concentrations and profiles of PAHs and heavy metals in the different brands of canned maize available in the Nigerian market. The objective of this study was to determine the concentrations of PAHs and metals in canned maize commonly consumed in Nigeria.

Materials and Methods

Reagents

All chemicals and reagents used were of analytical grade. Dichloromethane (LC grade), Acetone (Rieldel-de Haën, Seelze, Germany, purity 99.8%), anhydrous sodium sulfate (purity 99%), silica gel was obtained from BDH (Poole, UK). A PAH standard mixture containing the 16 priority PAHs, namely naphthalene (Nap) 1000 µg/mL, acenaphthylene (Acy) 2000 µg/mL, acenaphthene (Ace) (1000 µg/mL), fluorene (Flu) (199.9 µg/mL), phenanthrene (Phe) 99.8 µg/mL, anthracene (Ant) (100.0 µg/mL), fluoranthene (Flt) 200.1 (µg/mL), pyrene (Pyr) 99.9 µg/mL, benzo(a)anthracene (BaA) (100.1 µg/mL), chrysene (Chy) (100.0 µg/mL), benzo(b)fluoranthene (BbF) (200.2 µg/mL), benzo(k)fluoranthene (BkF) (99.9 µg/mL), benzo(a)pyrene (BaP) (100.0 µg/mL), dibenzo(a,h) anthracene (DahA) 200.0 µg/mL, indeno(1,2,3-cd)perylene (IndP) 100.1 µg/mL and benzo(g,h,i)perylene (BghiP) was purchased from Supelco (Bellefonte, PA, USA). Working mixed standard solutions containing all the PAHs were prepared by the dilution of the stock solution with acetone and store at -20°C in darkness to avoid volatilization and photo degradation. Nitric acid, perchloric acid and sulphuric acids were obtained from BDH (Poole, United Kingdom). Working standards of the metals (Cd, Pb, Ni, Cr, Fe and Mn) were prepared by diluting concentrated stock solution (Merck, Darmstadt, Germany) of 1000 mg L^{-1} with 0.25 mol L^{-1} nitric acid.

Sampling

A total of 5 brands of canned maize were collected from retail operation in Warri and Benin City, Nigeria. The choice of the samples was carefully made to reflect various brands consumed by the different income classes. Within a brand five to seven samples were collected and mixed together. From this a sub sample was obtained for PAHs analyses.

Sample preparation, extraction and clean-up

A mass of 10 g of the canned maize sample was weighed into a beaker and extracted with 50 mL of acetone and dichloromethane (DCM) in a ratio of 1:1by ultra-sonication at 35°C for 30 minutes. The process was repeated times on the residue in fresh mixture of DCM/Acetone each time. The solvent extracts were combined and passed through a column packed with anhydrous sodium sulphate by using 50 mL of a 1:1 mixture of DCM/acetone. The eluted fractions were transferred into a rotary evaporator to evaporate the volume to 2 mL. The extracts were subjected to purification by using solid phase with activated silica gel and eluted with 20 mL of dichloromethane (DCM). The extracts were collected and concentrated to 2 mL by using a rotary evaporator. The concentrated extract was transferred into a vial bottle using a pipette and store at -4°C before analysis.

Chemical Analysis

Samples were analysed for 16 PAHs by a gas chromatography (Hewlett Packard 6890) equipped with a HP5 capillary column (Cross linked PHME Siloxane) (0.25 µm film thickness × 0.25 µm × 30 m) and Flame ionization detector (FID). A 0.5 µL aliquot of the extract was manually injected with a syringe. The injector temperature was 250°C. The temperature program was initially set at 60°C, held at 60°C for 1 min followed by a 20°C/min ramping to a final temperature of 300°C. Helium was used as a carrier gas at a flow rate of 1.36 mL/min. Quantification was carried out by external standard method.

Metal Analysis

A mass of 1 g of the sample was weighed into a beaker and 20 mL of the acid mixture (perchloric acid, nitric acid and sulphuric acid in the ratio of 1:2:2) was

added to the beaker containing the sample. Thereafter, the sample was placed in the fume cupboard and heat to 120°C for 1 h on regulated hotplate until white fumes were observed. After which the sample was allowed to cool and filtered by using a Whatman No. 42 filter paper, and made to 25 mL with ultra-pure water. The samples were analysed for metal contents by using atomic absorption spectrophotometry (Perkin Elmer Analyst 200, USA).

Quality Control/Assurance

To evaluate the extraction efficiency for the PAH compounds, recovery studies were carried out by spiking selected already analysed samples with known standards of the individual PAH compounds and re-analysing the samples. The recoveries for the PAH compounds, r^2 values for the calibration lines for the PAH compounds, limits of detection and quantification are displayed in Table 1 while the recoveries for metals for analysis of certified reference material DORM-2 are displayed in Table 2.

Results and Discussion

Polycyclic Aromatic hydrocarbons

The concentrations of PAHs in these brands of canned maize are showed in Table 3 while Figure 1 depicts the PAH profiles in the canned maize. The concentration and profile of PAHs in these brands of canned maize ranged between 45.1 and 441.3 µg/kg. The highest concentration of the 16 PAH was observed in M-5. The concentrations and profile of PAHs varied significantly among the different brands of canned maize. In these brands naphthalene was not detected in any of the brands.

In these samples, naphthalene was below the limit of quantification. The three-ring PAHs (Acy +Ace+ Flu+ Phe+ Ant) varied from 12.7 to 114 µg/kg, which constituted 7.8% to 57.2% of the 16 PAHs content. Phenanthrene and anthracene were not detected in these brands of canned maize. Acenaphthylene is the dominant three-ring PAHs in these brands of canned maize and was detected at concentrations of 6.4 to 54.8 µg/kg. The highest acenaphthylene and Acenaphthene was detected in M-5. Acenaphthylene constituted 3.9% to 51.4% of the ∑16PAHs. Acenaphthylene was detected in 4 out of the 5 brands examined at concentrations varying from 6 µg/kg to 59.2 µg/kg, which constituted 3.9% to 13.4% of the 16 PAHs contents of these brands of canned maize. Flourene was detected in M-1 and M-2 at concentrations of 2.6 µg/kg which constituted 3.5% and 5.8% of the ∑ 16PAHs in brands M-1 and M-2 respectively.

The four-ring PAHs (Flt+Pyr+BaA+Chy) concentration ranged from 4.1 to 127.1 µg/kg. The highest concentration of four-ringed PAHs were observed in brand M-5. The four-ringed PAHs constituted 5.5% to 28.8% of the 16 PAHs in these brands of canned maize. Benzo(a)anthracene was detected in three samples at concentrations of 5.4 to 83.6 µg/kg. In these 3 samples (M-1, M-2 and M-5), BaA constituted 3.5% to 18.9% of the ∑16 PAHs. Chyresene is the dominant four-ringed PAH compound in these samples of canned maize in terms of concentration and frequency of occurrence. Chrysene was detected at concentrations of 2.8 to 53.7 µg/kg which constituted approximately 20% to 16% of the ∑16 PAHs.

Table 1 Percent recoveries, correlation coefficients for calibration lines, limits of detection (LOD) and limits of quantification (LOQ) for each of the 16 PAH standards.

PAH	Percent Recovery (%)	R^2	LOD (µg kg^{-1})	LOQ (µg kg^{-1})
Acenaphthene	97.2	0.9999	0.02	0.2
Acenaphthylene	93.4	0.9993	0.02	0.2
Anthracene	78.5	0.9996	0.02	0.2
Benzo(a)anthracene	77.6	0.9995	0.04	0.4
Benzo(a)pyrene	98.7	0.9993	0.02	0.2
Benzo(b)anthracene	90.6	0.9993	0.03	0.3
Benzo(g,h,i)perylene	98.4	0.9996	0.03	0.3
Benzo(k)fluoranthene	93.2	0.9991	0.02	0.2
Chrysene	92.4	0.9996	0.01	0.1
Dibenzo(a,h)anthracene	93.4	0.9994	0.03	0.3
Fluoranthene	93.6	0.9995	0.02	0.2
Fluorene	87.6	0.9998	0.06	0.6
Indeno(1,2,3-cd)perylene	87.6	0.9998	0.07	0.7
Naphthalene	69.6	0.9995	0.01	0.1
Phenathrene	79.7	0.9998	0.01	0.1
Pyrene	83.3	0.9999	0.01	0.1

Table 2 Validation Method with Standard Reference Material Dorm-2 (Dogfish Muscle) (µg/g dry weight)

Elements	Certified value	Measured value
Cd	0.043 ±0.08	0.042±0.04
Pb	0.065± 0.007	0.059±0.06
Ni	19.45±3.1	17.7±3.6
Cr	34.7± 5.5	31.9±4.8
Fe	142 ±10	126.4±8.2
Mn	3.66 ± 0.34	3.55±0.28

Table 3 Concentration of PAHs in µg/kg in canned maize

PAH COMPONENT	M-1	M-2	M-3	M-4	M-5
Naphthalene	<0.1	<0.1	<0.1	<0.1	<0.1
Acenaphthylene	9.3	23.2	6.4	27.5	54.8
Acenaphthene	6.0	<0.2	6.3	41.0	59.2
Fluorene	2.6	2.6	<0.6	<0.6	<0.6
Phenanthrene	<0.2	<0.2	<0.2	<0.2	<0.2
Anthracene	<0.2	<0.2	<0.2	<0.2	<0.2
Fluoranthene	<0.2	<0.2	<0.2	<0.2	<0.2
Pyrene	<0.1	<0.1	<0.1	<0.1	<0.1
Benzo(a)anthracene	5.4	<0.4	5.7	<0.4	83.6
Chrysene	2.8	4.1	3.2	53.7	43.5
Benzo(b)fluoranthrene	<0.3	5.0	<0.3	26.4	25.3
Benzo(k)fluoranthrene	42.2	<0.2	75.2	30.7	69.9
Benzo(a)pyrene	3.6	10.2	65.8	114.5	80.3
Indeno(1,2,3-cd)perylene	2.7	<0.7	<0.7	42.0	24.7
Dibenzo(a,h)anthracene	<0.3	<0.3	<0.3	<0.3	<0.3
Benzo(g,h,i)perylene	<0.3	<0.3	<0.3	<0.3	<0.3
TOTAL	74.6	45.1	162.6	335.7	441.3
2-ring	-	-	-	-	-
3-ring	17.9	25.8	12.7	68.5	114
4-ring	8.2	4.1	8.9	53.7	127.1
5-ring	45.8	15.2	141	171.6	175.5
6-ring	2.7	-	-	42	24.7
PAH2	6.4	14.3	69	168.2	123.8
PAH4	11.8	19.3	74.7	194.6	232.7
PAH8	56.7	19.3	149.9	267.3	327.3

PAH2 (Chry + BaP); PAH4 (PAH2 + Bbf + BaA); PAH8 (PAH4 + BkF + IndP + DahA + BghiP)

Table 4 Concentrations of metals (µg/g) in canned maize samples

METALS	M-1	M-2	M-3	M-4	M-5
Cd	0.7	<0.05	<0.05	<0.05	0.9
Pb	6.0	8.0	8.0	5.0	5.0
Ni	1.4	0.8	1.7	1.1	1.0
Cr	<0.05	<0.05	<0.05	<0.05	<0.05
Fe	163.4	193.6	181.7	92.4	114.1
Mn	<0.05	<0.05	<0.05	<0.05	<0.05

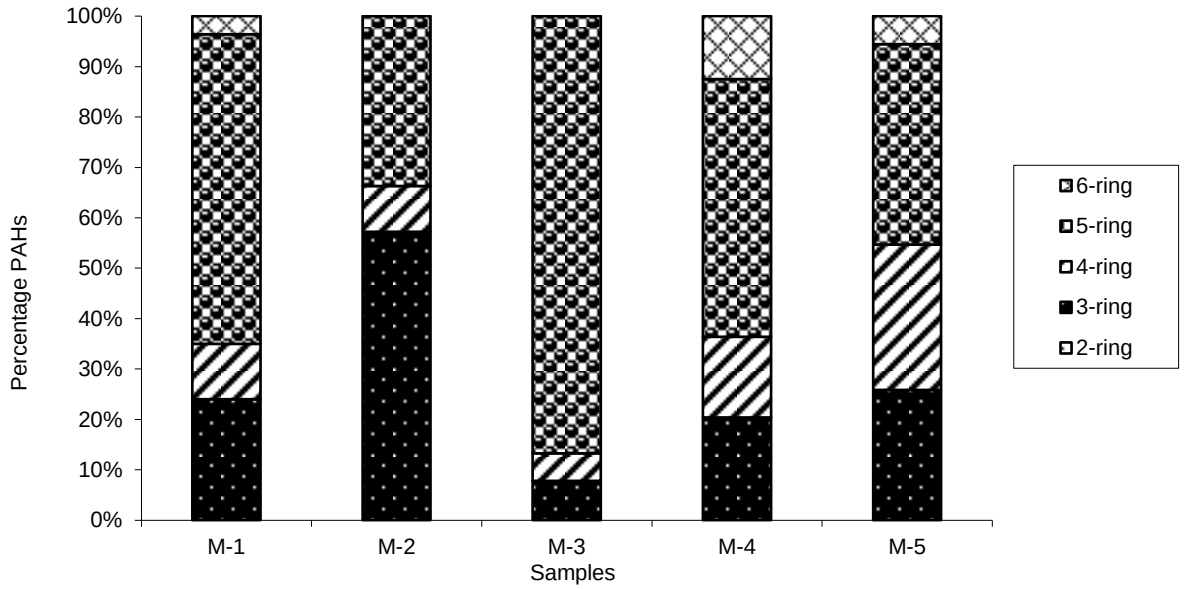

Figure 1 PAHs profiles in canned maize samples

The concentration of five-ring PAHs (Bbf + Bkf +BaP + DahA) ranged from 15.2 to 175.5 µg/kg which constituted 33.7% to 86.7% of the 16 PAHs in these brands of canned maize. The five-ring PAHs are the most abundant PAH compounds in these brands of canned compared with 2, 3, 4 and 6-ring PAHs. Benzo(a)pyrene was the dominant five-ring PAH compound in these canned maize and occurred at concentrations of 3.6 µg/kg to 114.5 µg/kg. The concentrations of BaP in these products were above the permissible limit of BaP in cereal products. BaP constituted 4.8% to 40.5% of the ∑16 PAHs content. The concentrations of benzo(b)fluoranthrene and benzo(k)fluranthrene were 5 µg/kg to 26.4 µg/kg in three of the brands respectively. Benzo(b) fluoranthrene and benzo(b)fluoranthrene constituted 5.75 to 11.1% and 9.2% to 56.5% of the ∑16 PAHs in these brands of canned maize.

Indeno(1,2-3 cd)perylene was the only six-ringed PAHs compound found in these products. Indeno(1,2-3 cd) perylene was found in M-1, M-5 and M-6 at concentrations of 2.7, 42.0 and 24.7µg/kg respectively. In these samples, IndP constituted 3.6 to 12.5 % of the ∑16 PAHs.

In 2008, European Food Safety Authority (EFSA) established that the following PAHs: BaA, Chy, BbF, BbF, BkF, BaP, IndP, DahA and BghiP have known oral carcinogenity and suggested that these PAHs (PAH8) or the subgroup of PAH4 (Chy+BaA+BbF+BaP) or the subgroup of PAH2 (Chy+ BaP) are the best indicators for the occurrence and effects of PAHs in food rather than the use of BaP alone as indicator. The concentrations of PAH2, PAH4 and PAH8 in these brands of canned maize ranged from 6.4 to 168.2 µg/kg, 11.8-232.7 µg/kg and 19.3 to 367.3 µg/kg respectively. The highest concentrations of PAH4, PAH8 were found in brand M-5 while M-2 had the highest concentration of PAH2. In this study, PAH2, PAH4 and PAH8 constituted 8.6-50.1%, 15.8-58.0%, 42.8 to 92.2% of the 16 PAHs respectively.

Metals contents in canned maize

The concentrations of metals showed significant inter-brand variations ($P<0.05$). The concentrations of Cd in samples ranged from <0.05 to 0.9 µg/g (Table 4). The concentration of Cd in brands M-1 and M-5 were above the 0.5 µg/g Codex Committee on Food Additives and Contaminants (CCFAC, 2001) permissible limit of Cd in food.

The concentrations of Pb in these samples of canned maize ranged between 5.0 and 8.0 µg/g. The highest concentrations were observed in brand M-2 and M-3. The permissible level of Pb in canned food is 1 µg/g. The concentrations of Pb in these brands of canned were 5 to 8 times above the permissible limit. The major source of Pb in canned food is leaching from the soldering of the can and in addition to the background Pb content of the maize. The concentrations of Ni in these samples of canned maize ranged from 0.8 µg/g in brand M-2 to 1.7 µg/g in brand M-3. There is no specific guideline for Ni in food in Nigeria. However, excessive intake of Ni could lead to contact dermatitis in pre-sensitized individuals. Chromium and manganese in the food were below detection limit and therefore does not pose any health risk to the consumer. However, Fe was detected at concentrations that varied between 92.48 µg/g and 193.6 µg/g. The highest Fe concentration was observed in M-2. Exposure to excess Fe can lead to numerous pathological events (Ponka et al., 2007; Mol, 2011). A maximum limit of 15 µg/g Fe in canned foods was recommended (Anonymous, 2002). In this study, the canned maize samples had Fe concentrations above the recommended levels of Fe in canned foods.

Conclusion

The concentrations of Cd, Pb and Fe in these samples of canned maize were above permissible limits allowed in canned food while metals such as Mn and Cr were below the limits of detection. The concentration and Profile of PAHs indicated that these samples of canned maize contained high levels of the suggested indicators for occurrence and effects of PAHs in foods such as BaP, PAH2, PAH4 and PAH8 far above permissible limits. Excessive consumption of these brands of canned maize could constitute serious health problem. The study demonstrates the urgent need for screening of trans-boundary food items for possible implementation risk management action. It also demonstrates the need to establish guidelines and standards for metals in canned foods products in Nigeria.

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