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Effects of Roasting on the Proximate Composition and Levels of Polycyclic Aromatic Hydrocarbons in Some Roasted Nigerian Delicacies

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are carcinogenic pollutants which get into foods during processing. In spite of this, only a few studies have been carried out on Nigerian delicacies. Raw and roasted food samples of corn (*Zea mays*), ripe plantain, unripe plantain (*Musa paradisiaca*) and yam (*Dioscorea sagittifolia*) of southwest Nigeria were evaluated for nutritional (proximate analysis and macro nutrients) and anti-nutritional qualities PAHs. The starchy food samples consist mainly of carbohydrate ranging from 35.93 – 55.52%, percentage crude fibre, protein, crude fat and ash content ranged between 0.82 – 14.21%, 3.15 – 8.27%, 1.12 – 19.56% and 1.12 – 2.59% respectively. The gross energy content of the food samples ranged from 777 – 1678 KJ/100g. The concentrations of minerals (macro nutrients) were as follows: Copper (7.05–20.60 ppm), Iron (3.15-28.3 ppm), Magnesium (1.15–4.95 ppm), Manganese (0.70–3.80 ppm) and Zinc (1.05–9.60 ppm). Higher levels of PAHs were found in the roasted samples than the raw samples. The values of total PAHs for raw food samples ranged from 0.19 - 0.53 ppm with unripe plantain having the least concentration. For roasted food samples the total PAHs concentration ranged from 3.62 – 40.33 ppm, with ripe plantain having the highest concentration. There was no significant difference between the proximate composition of raw and roasted food samples (P<0.05, t-test). These results from this data can be used to determine any potential risk associated with the ingestion of these foods.

Keywords: HPLC, macro nutrients, PAHs, proximate analysis, roasted food.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a class of ubiquitous environmental pollutants consisting of two or more fused aromatic rings which can be generated during the preparation of food (Agerstad and Skog, 2005). A number of PAHs have been found to have carcinogenic and mutagenic effects while some of them may act as synergists (Wenzl *et al.*, 2006). One of the major routes of human exposure to PAHs in non-smoking people is food. These compounds can reach the food chain by different ways as PAHs have been found in different food products, such as dairy products, vegetables, fruits, oils, coffee, tea, cereals and smoked meat, therefore the analysis of PAHs in food is a matter of concern (Plaza-Bolanos *et al.*, 2010). Over the years, different sources of PAH contamination of food have been found. Food items and products could be contaminated by soils, polluted air and water (WHO, 2005). Some aquatic food products, such as 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 (Plaza-Bolanos *et al.*, 2010). On the other hand, PAHs are also found in foods as a result of certain industrial food processing methods such as smoke curing, broiling, roasting and grilling over open fires or charcoal which permit the direct contact between food and combustion products (Silva *et al.*, 2011). Furthermore, in the food processing industry, food additives such as smoke flavouring products (SFP), lubricants, solvents, propellants, glazing agents and protective coatings contribute to contamination of food items by PAHs (Moret and Conte, 2000).

It has been found that raw foods do not usually contain high levels of PAHs, presence of PAHs in uncooked food, such as vegetables, seeds and grains have been found to accumulate on the waxy surface of many vegetables and fruits. In areas remote from urban or industrial activities, the levels of PAHs found in unprocessed foods reflect the background contamination, which originates from long distance airborne transportation of contaminated particles and natural emissions from volcanoes and forest fires. Nevertheless, other studies show the possibility of vegetables to take PAHs from soil and water and metabolize them. Another example of possible PAH contamination in food is due to traffic, i.e. crops or livestock close to urban roads could be exposed to

PAHs and nitro-PAHs (derivates from PAHs). In the neighborhood of industrial areas or along highways, the contamination of vegetation can be ten-fold higher than in rural areas (Larsson and Sahlberg, 1982).

In general, PAHs are not present individually but in mixtures. Sixteen PAHs that have been extensively monitored are the compounds included in the United States Environmental Protection Agency (USEPA) list of priority organic pollutants (USEPA, 1994). Of these 16 PAHs, Benzo(a)pyrene (BaP) is probably the most studied and has been described by the International Agency for Research on Cancer (IARC) as probable human carcinogen in 1987 (IARC, 1987). Thus, the determination of BaP has been widely used in environmental analysis as marker for the entire PAH content.

Several researchers have investigated the presence of PAHs in food samples. In 1964, the presence of benzo(a)pyrene (BaP) and other related PAHs were first reported to be present in charcoal broiled beef (Lijinsky and Shubik, 1964). PAHs have been detected at different concentrations in liquid fatty matrices e.g. edible oils (Barranco *et al.*, 2004), olive oil (Bogusz *et al.*, 2004) and milk (Kishikawa *et al.*, 2003), liquid non fatty matrices e.g. coffee (García-Falcón *et al*., 2005) and tea (Lai *et al.*, 2004), solid fatty matrices e.g. smoked meat (Chiu *et al.*, 1997), fish (Pensado *et al.*, 2005; Silva *et al.*, 2011) and infant milk and cereal (Rey-Salgueiro *et al.*, 2009) as well as solid non fatty matrices e.g. bread and potato (Nieva-Cano *et al*., 2001), fruits and vegetables (Rojo Camargo and Toledo, 2003) and tea leaves (Lin and Zhu, 2004).

Yam tubers are edible, starchy root of certain plant of genus *Dioscorea* which are used as staple food and they are probably the oldest cultivated food in Nigeria. Plantain (*Musa paradisiaca*) is an important source of carbohydrate to man; cooked or roasted green (unripe plantain) is one of the most staple food in Delta region of Nigeria. Ripe plantain and maize is eaten and enjoyed as delicacies throughout Nigeria. Maize (Zea mays), yam (*Dioscorea Sagittifolia*) ripe and unripe plantain (*Musa paradisiaca)* are usually processed by roasting. The commonest mode of roasting yam, maize and plantain in our environment is by open flame roasting. This work seeks to identify and quantify the levels of PAHs in these commonly consumed delicacies in Nigeria.

A thorough search of the literature reveals that there is no information on the level of PAHs in roasted yam, corn, ripe and unripe plantain even though these food samples are consumed by many in Nigeria. The present study was therefore designed to evaluate the levels of PAHs in these roasted foods and correlate their values with their nutritional qualities (proximate analysis and macro nutrients).

MATERIALS AND METHODS

Food sampling and processing

Samples of corn (*Zea mays)*, yam (*Dioscorea Sagittifolia*) ripe and unripe plantain (*Musa paradisiaca)* were bought from the Yaba market in Lagos state, south west Nigeria. Raw yam samples were peeled to remove the bark, cut into one quarter inches thick and roasted by the open flame roasting method. Where a grid was placed over a pot of lighted charcoal and the samples were roasted with occasional fanning was used to regulate the flame with temperature ranging between 140 – 200 ºC. The corn and plantain samples were roasted in a similar way as that of yam. Raw and roasted food samples were grinded, blended and stored in the refrigerator at 4 ºC prior analysis. Proximate analyses of the samples were carried out using the Association of Official Analytical Chemist method (AOAC, 1990).

NUTRITIONAL FEATURES

Proximate Analysis

The moisture content of the various food samples was determined on drying at 100 ºC in an oven until a constant weight was attained. The difference in initial and final weight of flour was expressed in percentage moisture. Micro-kjeldahl method was employed to determine the total nitrogen and the crude protein was calculated based on nitrogen conversion factor of 6.25. Crude lipid (Soxhlet extraction), crude fibre and ash contents (gravimetric) were determined based on methods outlined in AOAC (1990).

In order to calculate energy, total crude carbohydrate was estimated as follows:

Total crude carbohydrates (%) = 100 – (Crude protein + Crude lipid + Crude fibre + Ash) (Muller and Tobin, 1980).

While gross energy values were calculated based on the formula:

Gross energy (kJ/100 g) = (protein x 16.7) + (lipid x 37.7) + (carbohydrates x 16.7)

(Ekanayake *et al.*, 1999).

Mineral Composition

The levels of the macro minerals viz., copper, iron, magnesium, manganese and zinc were determined after digestion with aqua regia and analysed using Aanalyst Perkin Elmer 200-(2) flame atomic absorption spectrometer (AOAC, 1990).

Analysis of PAHs

PAHs in roasted and raw yam, maize, ripe and unripe plantain were extracted using solvent extraction by ultrasonication and the extracts were analysed for the 16 USEPA PAHs using high performance liquid chromatography (HPLC) with an ultraviolet (UV) detector (Silva *et al.*, 2011). The combined extracts

were centrifuged at 2500 rpm for 10 min and the supernatant was decanted. The supernatant was cleaned-up using the Whatman nylon filter membrane with further clean-up using the solid phase extraction (SPE) cartridges. The sorbent of the SPE cartridges were first conditioned with *n*-hexane, after which the filtered extracts were loaded on to the cartridges, the analytes were eluted with dichloromethane. The volume of the dichloromethane was blown down to dryness and extract was reconstituted in of acetonitrile. The quantification of PAHs was performed using an Agilent 1100 model HPLC system with a quaternary pump, vacuum degasser, a temperature controlled column oven and a UV diode-array detector. Separation of the PAHs was performed on a monomeric type octadecyl silica column, Supelcosil LC PAH 2 cm × 4.6 mm i.d containing 5 µm particles at ambient temperature (25 ±1°C) at a flow rate 1.0ml/min.

Statistical Analysis

The t-test was employed to ascertain the difference between raw and roasted food samples for proximate composition, mineral components and PAHs. Source groupings and the association of the parameters were determined using principal component analysis (PCA) and applying varimax with Kaiser Normalization rotation method to facilitate easier interpretation of data. Principal components having eigenvalues >1 of the complete data set were retained. Factor analysis in this study was carried out using the statistical analysis SPSS 15.0 software package.

RESULTS AND DISCUSSION

Proximate analysis (percentage crude fibre, protein, carbohydrate, were carried out on the food samples; yam, maize, ripe and unripe plantain and the result is as shown in Table 1.

Table 1: Proximate Analysis of Raw and Roasted Food samples

	% Crude Fibre	% Protein	% Carbohydrate	% Water	% Crude Fat	% Ash
Raw Unripe Plantain	11.83	3.88	40.13	41.93	1.12	1.12
Roasted Unripe Plantain	2.32	3.15	49.22	39.57	4.23	1.52
Raw Corn	1.58	5.81	35.93	50.70	4.75	1.23
Roasted Corn	0.82	4.73	48.03	38.84	6.07	1.51
Raw Ripe Plantain	12.95	8.27	55.52	18.70	2.80	1.76
Roasted Ripe Plantain	7.11	5.25	51.12	15.39	19.56	1.57
Raw Yam	14.21	4.94	47.37	32.95	1.40	1.90
Roasted Yam	10.07	4.91	43.95	30.63	1.20	2.59

All the raw food samples had higher percentage of crude fibre than the roasted samples. Raw yam had the highest percentage crude fibre of 14.21 %, followed by raw ripe plantain with a composition of 12.95 %, while raw unripe plantain and raw corn had percentage crude fibre compositions of 11.83 % and 1.58 % respectively. The percentage compositions of crude fibre in the roasted food samples ranged from 0.82 % - 10.07 %, with roasted corn having the least percentage composition. Percentage composition of protein and carbohydrate were also found to be higher in the raw food samples than in the roasted food samples. The proximate analysis result for roasted maize and yam was comparable with that of similar researches on proximate composition of street snacks purchased from selected motor parks in Lagos, Nigeria. The fibre content of roasted yam was found to be 8.49 g/100g and the carbohydrate content of roasted plantain was found to be 61.58 g/100g (Oboh and Ogbebor, 2010).

There was no significant difference between the ash content of the raw and roasted samples as seen in Table 1. The ash content which is the inorganic residue remaining after the organic matter has been burnt away is a very useful parameter in assessing the quality of edible material (Ojokoh and Gabriel, 2010). Water content in the roasted sample was less than in the raw sample. This is obviously due to the drying effect of the open flame roasting method.

The analysis of the macro nutrient in the raw and roasted maize, yam and plantain samples showed they were all rich in copper, manganese, magnesium, iron and zinc with the values shown in Table 2. There were no significant differences in the amount of nutrient in the raw and roasted samples and they were not found to follow any trend. The differences might be due to washing and handling of the sample.

Table 2: Mineral composition of raw and roasted food samples

	Cu (ppm)	Mn (ppm)	Mg (ppm)	Fe (ppm)	Zn (ppm)
Raw Unripe Plantain	10.65	2.4	1.95	28.3	1.05
Roasted Unripe Plantain	14.3	3.8	3.4	5.05	3.85
Raw Corn	12.45	0.7	4.05	6.85	6.2
Roasted Corn	12.2	0.8	4.7	3.15	9.6
Raw Ripe plantain	20.05	2.05	4.4	10.7	3.6
Roasted Ripe plantain	7.05	3.2	2.2	16.2	3.7
Raw Yam	17.3	1	4.95	16.1	1.3
Roasted Yam	20.6	2.45	1.15	4.9	5.6

PAHs were analysed with the results shown in Table 3. The levels of PAHs in the roasted samples were found to be higher than in the raw samples. However, roasted plantain was found to have the highest level of total PAHs with a concentration of 40.33 ppm. There is no limit yet for PAHs in plantain, maize and yam but the result showed that Benzo (a) pyrene (BaP), a bio-indicator was not detected in the raw and

roasted samples except in the roasted maize. BaP is the most studied carcinogenic polycyclic aromatic hydrocarbon and one of the most potent and it is often used as a toxicological prototype or surrogate for all carcinogenic polycyclic aromatic hydrocarbons (Collins *et al.*, 1991). The EU has established a maximum permissible level for BaP of 5 μg kg^{-1} wet weight for smoked meat and smoked meat products although a legal limit of 1 μg kg^{-1} had previously been adopted by some European countries. The EU has also set a maximum limit for BaP present in foodstuffs as a result of the use of smoking-flavour agents at 0.03 μg kg^{-1} (Lorenzo *et al.*, 2011).

Table 3: Concentration of PAHs in some Raw and Roasted foods

	PAHs	ROUP (ppm)	RAUP (ppm)	ROC (ppm)	RAC (ppm)	RORP (ppm)	RARP (ppm)	ROY (ppm)	RAY (ppm)
1	Naphthalene	nd	nd	2.73	2.63	4.49	nd	0.62	nd
2	Acenaphthylene	0.50	nd	1.70	nd	3.01	nd	nd	nd
3	Acenaphthene	nd	nd	5.12	nd	1.70	nd	nd	nd
4	Fluorene	0.78	nd	0.85	nd	4.94	nd	1.51	0.12
5	Phenanthrene	0.26	nd	0.16	nd	2.25	0.14	0.14	nd
6	Anthracene	0.66	0.11	nd	nd	7.56	0.05	0.35	0.08
7	Fluoranthene	0.67	nd	0.61	nd	5.55	nd	nd	nd
8	Pyrene	0.65	nd	0.35	nd	1.34	0.11	0.45	0.13
9	Benzo(a)anthracene	0.61	nd	0.07	nd	4.53	0.05	0.16	nd
10	Chrysene	0.61	nd	0.05	nd	2.77	nd	0.07	nd
11	Benzo(b)Fluoranthene	1.31	0.08	0.37	nd	1.10	nd	0.31	0.20
12	Benzo(k)fluoranthene	0.67	nd	0.19	nd	nd	nd	nd	nd
13	Benzo(a)pyrene	nd	nd	0.09	nd	nd	nd	nd	nd
14	Dibenz(a,h)anthracene	2.40	nd	1.56	0.54	0.54	nd	nd	nd
15	Benzo(g,h,i)perylene	0.56	nd	nd	nd	nd	nd	nd	nd
16	Indeno(1,2,3,c)pyrene	0.60	nd	nd	nd	0.55	nd	nd	nd
	Total PAHs (ppm)	10.28	0.19	13.85	3.17	40.33	0.35	3.62	0.53

Key: ROUP - Roasted unripe plantain, RAUP - Raw unripe plantain, ROC - Roasted corn, RAC - Raw Corn, RORP – Roasted ripe plantain, RARP – Raw ripe plantain, ROY – Roasted yam, RAY – Raw yam.

Total PAH concentrations in raw food samples ranged from 0.19 ppm in a raw unripe plantain to 3.17 ppm in raw corn. As expected, higher concentrations were found in roasted food samples, ranging from 3.62 ppm in roasted yam to 40.33 ppm in roasted ripe plantain. It was also found that the unripe food samples whether they were raw or roasted had lower concentrations of total PAHs than the ripe counterparts, raw unripe plantain had a concentration of 0.19 ppm of total PAHs while the ripe raw plantain had a concentration of 0.35 ppm. Similar trend was also observed in the roasted unripe plantain, total PAHs in roasted unripe plantain was found to be 10.28 ppm while the roasted ripe plantain had a concentration of 40.33 ppm. Individual concentrations of the carcinogenic PAHs (benzo[a]anthracene, benzo[b]fluoranthene, benzo[a]pyrene, dibenz [a,h]anthracene, and indeno [1,2,3-c,d]pyrene) ranged from non-detectable in several of the food samples to 4.53 ppm in roasted ripe plantain.

Table 4: Eigen values of factor for proximate composition, macronutrients and total PAHs in the food samples

Component	Initial Eigen values		
	Total	% of variance	Cumulative %
1	3.341	27.841	27.841
2	3.104	25.863	53.704

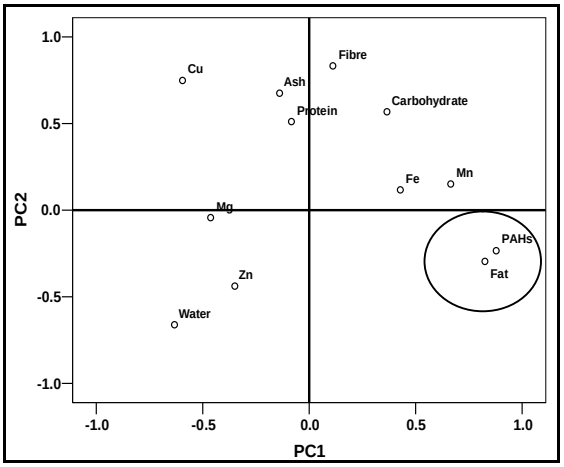

Figure 1: Plots of loadings of proximate composition, macronutrients and total PAHs in the food samples

The data were subjected to principal component analysis (PCA) to elucidate the correlation between the various parameters in both the raw and roasted food samples. Eigen values were used to determine the number of principal components that should be retained for further study. For both raw and roasted food samples, the first two principal components had eigen values greater than 1 which explained 54% of the variance. As seen in Table 4, PC1 accounts for about 28% of the variation in the data while PC2 accounts for 28% of the variation. The loadings for PC1 suggest that it is correlated with sum of the PAHs and percentage crude fat content in the roasted

food samples, but to a lesser degree, the concentrations of iron and manganese. PC2 was found to be strongly correlated with percentage fibre, ash, protein, carbohydrate and copper. The rest of the PCs do not describe as much variation in the data analysis. This is clearly illustrated in Figure 1.

The score plot (Figure 2), shows that roasted ripe plantain has a high correlation with PC1 which in turn is correlated with high total PAHs and percentage fat content. The high fat content of roasted ripe plantain (19.56%) probably suggests the increase in the amount of total PAHs that can be adsorbed by the food sample on roasting.

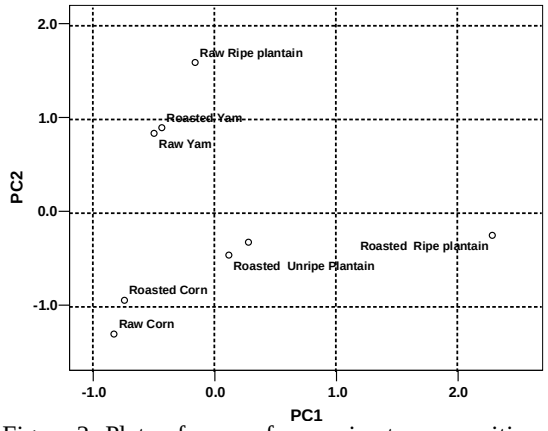

Figure 2: Plots of scores for proximate composition, macronutrients and total PAHs in the food samples

CONCLUSION

The proximate analysis of the nutrients in the samples of raw and roasted corn (*Zea mays)*, yam (*Dioscorea Sagittifolia*) ripe and unripe plantain (*Musa paradisiaca)* show that all the samples have significantly different nutritional compositions. There are indications that all the food samples are good sources of nutrients, however, the results show that both the raw and the roasted food samples contain certain levels of PAHs, with the roasted food samples having greater levels of total PAHs as a result of the food processing. Except for roasted corn, benzo[a]pyrene was not detected in raw and roasted food samples; therefore, it can be assumed that both raw and roasted foods do not represent a health risk for human, since benzo[a]pyrene is considered as a marker of carcinogenic PAHs. But, it should be stated that these food products are contaminated in total PAHs after roasting.

In conclusion, as a result of roasting of the food samples, it was obvious that the concentration of total PAHs increased and for this reason an alternative process should be introduced. Considering the carcinogenic potential of the PAHs, the reduction of these contaminants in the diet is highly desirable and special attention must be given to the intake of roasted foods since considerable amount of PAHs can be ingested in a single meal.

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