



Effect of home processing methods on the levels of heavy metal contaminants in four food crops grown in and around two mining towns in Ghana

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ABSTRACT

Unregulated small-scale mining activities, by young untrained men using some poisonous chemicals, occur in several agricultural forest belts in Ghana. These activities contaminate water bodies in these areas, which happen to be the main farming sites where food crops are intensively cultivated. The presence of these heavy metal contaminants in popular food staples is therefore worrying because of its adverse health implications. Previous studies have shown that processing is able to decrease the concentrations of heavy metals in foods. This study investigated the effectiveness of home processing methods (boiling, frying and roasting) in significantly reducing the levels of heavy metal contaminants in food crops grown in and around two main mining centers in Ghana. The heavy metals contaminants analyzed for, were Arsenic (As), Cadmium, (Cd), lead (Pb), Manganese (Mn), and Mercury (Hg), determined using atomic absorption spectrometry (AAS) and inductively-coupled plasma mass spectrometry (ICP-MS). From the data, the average daily intakes of the heavy metals and the associated long-term health risks to consumers were assessed. Unprocessed samples from Akwatia had higher levels of contaminants than those from Obuasi. Levels of Mn, Pb and As recorded in all unprocessed samples were higher compared to WHO permissible limits in foods. The levels showed a decreasing trend in the processed samples; with the lowest As and Pb content recorded after frying and boiling. The study showed that roasting allowed for the least reduction in the heavy metal contaminations in the four food crops. The levels of Cd in both processed and unprocessed samples were within safe WHO specifications. Except for Pb in unprocessed cassava, boiled cassava and unprocessed plantain and Hg (unprocessed yam and roasted yam), the hazard indices of all metals in all food crops were less than one and posed no risk to consumers. The study therefore reveals that the normal home processing methods are able to reduce the levels of heavy metal contaminants found in cassava, cocoyam, plantain and yam considerably.

1. Introduction

Cassava, cocoyam, plantain and yam are the most used food crops in Ghanaian food culture. They are mainly cultivated in the forest agricultural ecozones of Ghana, very much close to several natural river bodies. More recently, some parts of the country and along some of these natural water bodies, there has been an upsurge in the activities of illegal small-scale mining in the forest agricultural ecozones where these food crops are intensively cultivated. Most of these mining activities are

unregulated and carried out by young men, most of whom are illiterates, without the requisite modern equipment for proper mining and have very little regard to acceptable conventional mining practices. Their activities therefore impact negatively on the environment and communities, especially because of the regular use of chemicals such as potassium cyanide in the extraction of gold [1,2]. Several of these environments are polluted with heavy metals, which usually found their way into water bodies in these communities [3–5]. Therefore, there is a possible presence of heavy metal contaminants in the food chain, fresh

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foods and food products used by inhabitants in such mining areas [6].

Heavy metals are naturally occurring compounds that have high density compared to other metals and usually are five times denser than water [7] and non-biodegradable [8,9]. Even small doses when consumed can be toxic to humans [10,11]. They cause toxicity by forming complexes with sulphur, oxygen or nitrogen-containing cell compounds [12], when entering our bodies through food, drinking water and air. These complexes inactivate or modulate critical enzymes or protein structures resulting in cellular dysfunction and necrosis [13]. Lead, for example, can cause cancer to human internal organs because of its ability to cause disruption to the biosynthesis of hemoglobin and anemia and can also cause increase in blood pressure and kidney damage.

Food crops are able to absorb several heavy metals from the soils they are cultivated in depending on the physicochemical properties of the soils such as pH, organic matter content, clay fraction, mineral composition and the binding capacity of the soil [14–16]. Dheri et al. [17] have explained that heavy metals dissolved in waste water and water bodies are easily transported to suspended solids. In addition, several agricultural lands have witnessed many residue deposits either directly or indirectly from the mining companies [1,18]. These depositions have caused a rise in pollutants such as heavy metals in the soil [19,20].

Two major towns where both legal and illegal mining activities occur in Ghana are Akwatia (Eastern Region) and Obuasi (Ashanti Region). Akwatia is a major diamond extraction center in Ghana. Mining and farming along the Birim river are the main occupation of the inhabitants of Akwatia. Obuasi, on the other hand, is noted for gold mining. Apart from the recognized major mining companies, like AngloGold Ashanti, there are several illegal small-scale mining operators, operating outside the statutory concession areas of the major mining companies within the Pra and Tano rivers' basins [6]. Apau et al. [21] reported that levels of copper, zinc, and cadmium in yam, cocoyam, sweet potato, and cassava from farms along the banks of these rivers exceed the WHO specifications.

The Government of Ghana has unfortunately not been able to regularise the activities of these illegal mining in ways that can help assure the safety of food crops from these mining centers. This means the problem of heavy metal contamination of food crops especially is bound to be with us for some time to come. Probably, the problem of heavy metal contamination of local food produced is systemic. Massoud et al. [22] have discussed the potential of using *Saccharomyces cerevisiae* in bio mediation of heavy metals in the food industry. However, such technologies will be far from the reach and ability of developing countries. Hajeb et al. [23] and Morgan [24] have however explained that food processing methods can be used to reduce the levels of toxic heavy metals found in foods. Therefore, it will be interesting to find out whether the levels of heavy metal contaminants in these popular food crops could be reduced significantly during food preparations, using typical home processing methods of foods in Ghana. Such information should assist in intervention programs that might be undertaken to help mitigate the adverse effect of heavy metal contaminants in local foods. The objective of this study therefore was to determine the effect of home processing methods such as boiling, roasting and frying on the levels of heavy metal contaminants found in cassava, cocoyam, plantain and yam from Akwatia and Obuasi.

2. Methodology

2.1. Solvents, reagents and materials

Nitric acid was purchased from BDH chemicals (Gyeonggi-do, Korea), hydrogen peroxide from BDH chemicals (Leuven, Belgium), multi-element standard from BDH chemicals (Fomlenay-Sous-Bios, France), sodium chloride anhydrous from BDH chemicals (Tolderlundsvej, Denmark) and formic acid, ethyl acetate, acetonitrile,

Table 1

Fresh crop samples purchased from farms in Obuasi and Akwatia.

Commodity	Total samples analysed	Average weight (g)	Individuals
Cassava	40	800	150 Tubers
Cocoyam	40	500	225 Tubers
Yam	40	1500	75 Tubers
Plantain	40	240	465 Pieces

disodium hydrogen citrate sesquihydrate, tri-sodium citrate dehydrate, magnesium sulphate were all from BDH Chemicals (Caton, United Kingdom).

2.2. Source of samples

A total of three-hundred and sixty (360) fresh and processed yam, cassava, cocoyam and plantain samples were used for this study. Yam, cassava, cocoyam and plantain samples were purchased from two mining towns, Akwatia and Obuasi in Ghana. Akwatia is a major diamond extraction center in Ghana, and occupies an area of 240 km², lying between latitude 6°-3' North and longitude 0°-48' West and has a total population of 23,766 people. Along the Birim river, which passes through Akwatia Township, there are several farms and sites for illegal mining activities [25]. Obuasi is a gold mining community which occupies an area of 162.4 km², lies between latitudes 6°-12' North, and longitude 1°-41' West and has a total population of 175, 043 people. Two major rivers, Pra and Tano, are found close to this town. Along the banks of these two rivers both intensive farming and illegal mining are carried out [26]. There were two sources of samples as described in sections 2.2.1 and 2.2.2

2.2.1. Fresh crop samples from farms in Akwatia and Obuasi

Fresh yam, cocoyam, cassava and plantain were purchased from fifteen randomly selected farms, five farms along each of the three rivers, along the Birim river of Akwatia and Pra and Tano rivers of Obuasi. Purchases were done on five occasions between the months of March and June 2019. An average of 1.5 kg of each food crop was purchased each time from each of the five farms. Fresh crop samples analysed are shown in Table 1. These samples were thoroughly washed under running tap water and rinsed with laboratory processed deionized water using Millipore Elix (SAS 67120 Mosheim, France). They were peeled, washed and rinsed again with deionized water and packed into different acid-cleaned polypropylene zip-lock bags and were kept in the fridge at temperature of 4 °C.

2.2.2. Processed food samples from sellers in markets at Akwatia and Obuasi

Two-hundred (200) processed yam, cassava, cocoyam and plantain samples were analysed. Locally prepared foods on sale; boiled (yam, cassava, cocoyam and plantain), fried (yam, cocoyam, cassava and plantain), and roasted (yam, cocoyam, cassava and plantain) were purchased from three regular sellers each at *Akwadum* and *Central* markets at Akwatia and Obuasi, respectively. Purchases of these samples were done five times over a period of 10 weeks, from March to June of 2019. An average of 2 kg of each prepared food was purchased from each seller each time. Large quantities of processed samples were purchased to enable sorting for a representative samples. These prepared foods were cleaned gently using a moist kitchen tissue paper to remove any adhering dirt from the markets. They were then packed into different acid-cleaned polypropylene zip-lock bags and kept in the fridge at temperature of 4 °C until laboratory analysis. When ready for analysis, different portions of each food item, purchased from the three sellers, were pooled together, thawed at room temperature and homogenized to a pulp and powder mixtures with a blender before aliquots were weighed out for analysis.

2.2.3. Laboratory sample preparations from fresh crops

Fresh cassava, cocoyam, plantain and yam samples from farms, as described in Section 2.2.1, were further divided into four longitudinal sections, using a stainless steel kitchen knife. Sub-samples containing a section each of food piece were selected and again further divided into four equal portions for the various sub-samples as explained in Sections 2.2.4 to 2.2.7.

2.2.4. Unprocessed samples

The first of these four portions was labeled as sub-sample to be used for the analysis of the heavy metals for the unprocessed laboratory sample. 1 kg sub-samples of this portion was milled in a blender (Grindomix Retsch GM 200 blender) at a speed of 1000 rpm for 15 s, divided into 250 g, transferred into glass sample cups, appropriately labeled and kept in a fridge at a temperature of 4 °C prior to digestion.

2.2.5. Boiled samples

The second portion (from Section 2.2.3.) was boiled using deionized water (in ratio, 1 food portion:2 water portions, in w/v) in a 2.8 L stainless steel sauce pan for 30 min on a laboratory hot plate (Gestigeit) set at a temperature 300 °C. Excess water was drained off, as usually done during household cooking. Samples were left to cool in glass plates after boiling. The cooled samples were milled and transferred into glass sample cups and stored as explained in 2.2.4.

2.2.6. Fried samples

The third portion (from Section 2.2.3) was fried using a laboratory-certified oil of known metallic concentration (500 g sample: 1000 L oil) in 2.8 L stainless sauce pan for 25 min, on the hot plate, set at a temperature of 200 °C. Accredited commercial cooking oil, with known composition as indicated by the test results of the Food and Drugs Authority of Ghana, was used for this study. This was done to ensure cooking oil did not contain any trace of heavy metals. The frying was done in batches and the oil was changed after each batch of frying. After cooling, each fried sample was milled and transferred into sample cups and stored as explained in 2.2.4.

2.2.7. Roasted samples

The fourth portion of each unprocessed sample of yam, cocoyam, cassava and plantain was cut into chips, averagely with thickness of about 3 cm, on a separate glass plate (1 kg) and were spread on the plate and roasted at 120 °C for 15 min in a microwave grill (Samsung Grill Microwave MC32K7055CK). The grilled samples were allowed to cool. They were then milled and transferred into sample cups and stored as explained in 2.2.4.

2.3. Analyses of heavy metals

Dry ashing method was used for Atomic Absorption Spectrometry (AAS) analysis of manganese (Mn) as it was readily available [27]. All glassware were washed using 1% nitric acid, followed by deionized water. Approximately 3 g of each prepared sample was weighed into cleaned pre-weighed crucibles with corresponding labels and placed in a Muffle furnace at a temperature of 540 °C for 3 h. The ashed samples were mineralized with 100 mL of 2% nitric acid and filtered through a 541 ash-less filter paper into a 100 mL volumetric flask. The filter papers were rinsed with deionized water after each drain till it got close to the 100 mL mark. The resultant solution was topped up to the mark with deionized water. Blank solutions were treated same way as the sample. The flasks with the digested samples were then corked and labelled. Atomic Absorption Spectrometry (Varian AA240FS) was used to read the absorbance values at appropriate wavelength of interested metal in the sample solution, Mn (wavelength 279.5 nm). The content of metal was derived from a calibration graph made up of five standards. Standards were prepared from a 100 mg/L multi-element stock solution via a serial dilution to produce 0.5 mg/L, 1.0 mg/L, 2.0 mg/L, 3.0 mg/L and

5.0 mg/L using 2% HNO₃.

Microwave digestion was used for Inductively Coupled Plasma Mass Spectrometry (ICP-MS) analysis of cadmium (Cd), lead (Pb), arsenic (As) and mercury (Hg). Roughly, 1 g of each sample was weighed into Teflon vessels that have been acidified, washed in a microwave digester for 1 h and cooled. Five milliliter (5 mL) concentrated nitric acid (63.01 % analytical grade) and 3 mL of 30 % concentrated hydrogen peroxide were added. The vessels were tightened into their respective shields and packed accordingly with their numbers into the microwave digester (Milestone) and digested for 1 h at a temperature of 170 °C and a pressure of 5 MPsi. After digestion the samples were allowed to cool completely and were poured into a 50 mL centrifuge tubes and topped up to 25 mL mark with deionized water. Chemical analyses of Cd, Pb, As and Hg were carried out using Agilent ICP-MS 7700. The ICP-MS was calibrated using 0.5 µg/L, 1.0 µg/L, 5.0 µg/L and 10.0 µg/L standards solution prepared from a multi-element stock solution of concentration 100 mg/L.

2.4. Estimation of dietary exposure and health risk assessment

The dietary exposure and health risk associated were calculated using the estimated average daily intakes (EADI) of the heavy metals in these four food-crops and their associated hazard indices (HI). The EADI was estimated from the average food consumption assumption used to determine long term health risks to consumers [28]. The average food consumption rates in Ghana for cassava, cocoyam, plantain and yam are 154.0, 38.0, 101.8 and 50.0 kg/person/year, respectively [29]. For each type of exposure, the EADI was calculated using the formula below:

$$EADI = \frac{(C \times F)}{(W \times D)}$$

where C is the concentration of metal in each commodity (mg/kg), F is mean annual intake of food per person, D is number of days in a year (365) and W is the mean body weight (60 kg).

The health risk indices were obtained by dividing the estimated average daily intake (EADI) by the acceptable daily intakes (ADI) established by Codex Committee [30] using the equation:

$$HI = \frac{EADI}{ADI}$$

When the HI is more than 1; the food involved is considered a risk to the concerned consumers. When the HI is less than 1, the food involved is considered as acceptable (no concern) to the concerned consumers [31–33].

2.5. Quality control of results

Samples were handled carefully to avoid contamination as part of measures to ensure reliability of results. The recovery test of the total analytical procedures was also carried out for the metals analysed in the selected samples by spiking analysed samples with aliquots of metal standards and then reanalysed the samples. Acceptable recoveries of 98.29 ± 0.71, 89.56 ± 0.29, 99.84 ± 0.07, 99.72 ± 0.09 and 99.79 ± 0.14 % were recorded for As, Cd, Hg, Mn and Pb respectively.

2.6. Data analysis

Mean metal concentrations of 360 sample solutions of yam, cocoyam, cassava and plantain purchased from farms and markets across Akwatia and Obuasi were compared using one-way analysis of variance (Robust Welch, ANOVA, Duncan test, $p \leq 0.05$) via Statistical Package for Social Sciences (IBM SPSS, version 26).

Table 2

Mean heavy metal contents in unprocessed and laboratory-processed cassava from Akwatia and Obuasi farms.

Sites	Food sample	Mn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	As (mg/kg)	Hg (mg/kg)
Obuasi	Boiled cassava	0.58 ± 0.34 ^a	0.61 ± 0.23 ^a	<0.01 ^a	0.01 ± 0.00 ^{ab}	0.06 ± 0.01 ^a
	Fried cassava	0.50 ± 0.19 ^{ab}	0.07 ± 0.02 ^b	ND	<0.01 ^a	0.02 ± 0.01 ^a
	Roasted cassava	0.70 ± 0.37 ^a	1.61 ± 0.09 ^c	ND	0.01 ± 0.02 ^{ab}	0.11 ± 0.03 ^b
	Unprocessed	0.80 ± 0.46 ^a	2.11 ± 0.61 ^d	<0.01 ^a	0.02 ± 0.02 ^b	0.12 ± 0.02 ^b
	Boiled cassava	0.42 ± 0.29 ^b	0.60 ± 0.26 ^a	ND	0.01 ± 0.02 ^{ab}	0.05 ± 0.02 ^a
Akwatia	Fried cassava	0.42 ± 0.25 ^b	0.08 ± 0.02 ^b	ND	<0.01 ^a	0.03 ± 0.01 ^a
	Roasted cassava	0.56 ± 0.34 ^b	0.10 ± 0.19 ^c	<0.01 ^a	0.01 ± 0.01 ^{ab}	0.11 ± 0.04 ^b
	Unprocessed	0.62 ± 0.58 ^{ab}	2.11 ± 0.68 ^d	<0.01 ^a	0.02 ± 0.02 ^b	0.16 ± 0.05 ^b
	LSD (5%)	0.27	0.07	<0.01	0.01	0.04
	CV (%)	19.32	20.1	14.34	11.21	12.4

*ND means not detected. Figures are presented as means ± standard deviations. Superscripts to figures in the same column implies significant or not significant difference at $p \leq 0.05$ (ANOVA, Duncan test). Limit of detection (LOD) for Cd = 0.0000 ppb.

3. Results and discussion

3.1. Variations in heavy metals in unprocessed and laboratory-processed cassava from Akwatia and Obuasi farms

Table 2 shows that the concentrations of Mn in both unprocessed and laboratory-processed cassava from Obuasi and Akwatia farms, in the range 0.42 ± 0.25 – 0.80 ± 0.46 mg/kg, were far above the WHO specification of 0.05 mg/kg Mn content in foods [31]. Interestingly, the cassava samples from Obuasi farms, except for the roasted sample, there was no significant difference between the level of Mn content obtained by unprocessed cassava and other processed cassava samples, boiled and fried (at $p \leq 0.05$), whilst cassava samples from Akwatia showed more variations in their Mn contents. The levels of Pb, Cd, As and Hg recorded for unprocessed and processed cassava from Obuasi and Akwatia farms were in the range, 0.07 ± 0.02 – 2.11 ± 0.68 mg/kg (Pb), ND– <0.01 mg/kg (Cd), <0.01 – 0.02 ± 0.02 mg/kg (As) and 0.02 ± 0.01 – 0.16 ± 0.05 mg/kg (Hg). The levels of Pb recorded for boiled cassava, fried cassava and roasted cassava were lower compared to those of unprocessed cassava from both Obuasi and Akwatia farms. This result is an indication that home processing method can reduce the levels of Pb during boiling, frying and roasting. Ofori et al. [34] did not detect Pb content in high quality cassava flour, a product from cassava obtained through a combination of several processing steps including drying. Oluyemi et al. [35], on the other hand, reported Pb content ranging from 57.00 ± 0.02 – 87.50 ± 0.01 mg/kg for unprocessed cassava from farm lands close to areas where mining companies had previously dumped waste. The higher concentrations of Pb, reported by Celik and Oehlenschlager [36], compared to those obtained in this study, may be due to different geographical locations where samples were collected and their mining activities. Previous studies by Akabzaa et al. [1], Dube et al. [14] and Dheri et al. [17] have all reported that farm lands with heavy deposits of mining waste tend to have high levels of pollutants. Both Cd and As concentrations recorded in the present study were far lower compared to what were obtained by Tuzen [38], which were in the ranges 5.20 ± 0.01 – 14.00 ± 0.01 and 4.18 ± 0.01 – 6.61 ± 0.02 mg/kg respectively. Ofori et al. [34] did not detect As in high quality cassava flour, but found Hg content of <0.01 mg/kg which is lower compared to Hg content of 0.02 – 0.16 mg/kg recorded for both unprocessed and processed cassava

Table 3

Mean heavy metal contents in unprocessed and laboratory-processed cocoyam from Akwatia and Obuasi farms.

Sites	Food sample	Mn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	As (mg/kg)	Hg (mg/kg)
Obuasi	Boiled cocoyam	0.61 ± 0.53 ^{ab}	0.40 ± 0.62 ^a	ND	0.03 ± 0.01 ^a	0.15 ± 0.05 ^a
	Fried cocoyam	0.55 ± 0.46 ^b	0.20 ± 0.24 ^b	ND	0.02 ± 0.01 ^b	0.14 ± 0.03 ^a
	Roasted cocoyam	2.00 ± 0.53 ^c	0.95 ± 0.60 ^c	<0.01 ^a	0.06 ± 0.03 ^c	0.18 ± 0.06 ^b
	Unprocessed	2.04 ± 0.27 ^c	1.00 ± 0.64 ^c	<0.01 ^a	0.07 ± 0.01 ^d	0.19 ± 0.06 ^b
	Boiled cocoyam	0.78 ± 0.49 ^a	0.30 ± 0.48 ^{ab}	ND	0.03 ± 0.01 ^a	0.15 ± 0.04 ^a
Akwatia	Fried cocoyam	0.52 ± 0.52 ^b	0.21 ± 0.42 ^b	ND	0.02 ± 0.01 ^b	0.14 ± 0.04 ^a
	Roasted cocoyam	1.99 ± 0.27 ^d	0.65 ± 0.58 ^d	<0.01 ^a	0.07 ± 0.03 ^c	0.19 ± 0.06 ^b
	Unprocessed	2.01 ± 1.42 ^d	0.73 ± 0.56 ^d	<0.01 ^a	0.07 ± 0.08 ^c	0.19 ± 0.06 ^b
	LSD (5%)	0.17	0.12	<0.01	<0.01	0.12
	CV (%)	15.2	13.01	12.34	6.3	13.2

*ND means not detected. Figures are presented as means ± standard deviations. Superscripts to figures in same column imply significant or not significant difference at $p \leq 0.05$ (ANOVA, Duncan test). LOD for Pb = 0.0000 ppb.

from Obuasi and Akwatia farms in the present study.

3.2. Variations in heavy metals in unprocessed and laboratory-processed cocoyam from Akwatia and Obuasi farms

A general decrease in all the heavy metal contents was recorded when cocoyam was boiled, fried or roasted (Table 3). The reductions in the Mn contents of the cocoyam are more pronounced than for the remaining heavy metals. The highest Mn concentration of 2.04 ± 0.27 mg/kg was recorded for unprocessed cocoyam from Obuasi farm with the lowest of 0.52 ± 0.52 mg/kg Mn content obtained by fried cocoyam and the difference was statistically significant. The general reductions in Mn contents were observed in both samples sourced from Obuasi and Akwatia farms. However, there was no significant difference between the level of Mn content recorded for unprocessed cocoyam and roasted cocoyam for samples from Obuasi and Akwatia farms. Even though Mn content reduced drastically using home processing methods, both the highest (2.04 ± 0.27 mg/kg) and lowest (0.52 ± 0.52 mg/kg) Mn content recorded for unprocessed and processed cocoyam, from Obuasi and Akwatia farms were far above the WHO specification of 0.05 mg/kg Mn content in foods [34]. The unprocessed and processed cocoyam samples from Obuasi and Akwatia farms obtained Pb levels ranging between 0.20 ± 0.24 mg/kg and 1.00 ± 0.64 mg/kg. The difference between the highest Pb content of 1.00 ± 0.64 mg/kg recorded for unprocessed cocoyam from Obuasi farm and the lowest of 0.20 ± 0.24 mg/kg obtained by fried cocoyam from Obuasi farm was statistically significant. The range of 0.20 ± 0.24 – 1.00 ± 0.64 mg/kg Pb concentrations recorded for unprocessed cocoyam, boiled cocoyam, fried cocoyam and roasted cocoyam was far below the WHO recommend limit of 2.00 mg/kg Pb content in foods [31]. The Pb levels recorded for both unprocessed and processed cocoyam from Obuasi and Akwatia farms therefore pose no public health threat. A previous study by Oluyemi [35] reported Pb content of 10.00 ± 0.01 – 50.00 ± 0.01 mg/kg for unprocessed cocoyam when they investigated variation in heavy metal content in soils and some selected crops at a land fill site in Nigeria. The Pb concentration of 10.00 ± 0.01 – 50.00 ± 0.01 mg/kg reported by [35] was above the Pb content of 0.20 ± 0.24 – 1.00 ± 0.64 mg/kg recorded for both unprocessed and processed cocoyam in the present study.

The levels of Cd, As, and Hg determined in unprocessed and processed cocoyam from Obuasi and Akwatia farms were in the range ND–

Table 4

Mean heavy metal contents in unprocessed and laboratory-processed plantain from Akwatia and Obuasi farms.

Sites	Food sample	Mn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	As (mg/kg)	Hg (mg/kg)
Obuasi	Boiled plantain	0.38 ± 0.32 ^a	0.06 ± 0.03 ^{ab}	<0.01 ^a	0.01 ± 0.00 ^a	0.11 ± 0.05 ^a
	Fried plantain	0.30 ± 0.23 ^a	0.04 ± 0.02 ^a	ND	<0.01 ^a	0.10 ± 0.05 ^a
	Roasted plantain	0.50 ± 0.36 ^b	0.13 ± 0.06 ^b	ND	0.01 ± 0.01 ^a	0.11 ± 0.05 ^a
	Unprocessed	0.60 ± 0.71 ^b	1.02 ± 0.65 ^c	<0.01 ^a	0.02 ± 0.01 ^d	0.12 ± 0.07 ^a
	Boiled plantain	0.32 ± 0.26 ^a	0.07 ± 0.01 ^a	ND	0.01 ± 0.02 ^a	0.05 ± 0.01 ^b
	Fried plantain	0.22 ± 0.19 ^d	0.05 ± 0.01 ^a	ND	<0.01 ^a	0.04 ± 0.02 ^b
	Roasted plantain	0.36 ± 0.31 ^a	0.75 ± 0.04 ^b	<0.01 ^a	0.01 ± 0.01 ^a	0.12 ± 0.07 ^a
	Unprocessed	0.42 ± 0.47 ^b	1.11 ± 0.65 ^c	<0.01 ^a	0.02 ± 0.01 ^b	0.13 ± 0.05 ^a
	LSD (5%)	0.12	0.05	<0.01	0.01	0.05
	CV (%)	20.32	18.1	17.34	11.65	19.86

*ND means not detected. Figures are presented as means ± standard deviations. Superscripts to figures in the same column implies significant or not significant difference at $p \leq 0.05$ (ANOVA, Duncan test). LOD for Cd = 0.0000 ppb.

<0.01 mg/kg (Cd), $0.02 \pm 0.01 - 0.07 \pm 0.08$ mg/kg (As) and $0.14 \pm 0.03 - 0.19 \pm 0.06$ mg/kg (Hg). The highest Cd content of <0.01 mg/kg obtained for unprocessed cocoyam from Obuasi and Akwatia farms was slightly below the WHO recommended limit of 0.02 mg/kg Cd content in foods of root and tuber [31]. However, when the fresh cocoyam was processed into boiled and fried cocoyam, Cd was not detected (ND) in these products. Even though As content in unprocessed cocoyam was significantly higher compared to boiled cocoyam and fried cocoyam; the highest As concentrations 0.07 ± 0.08 mg/kg recorded for both unprocessed and processed cocoyam from Obuasi and Akwatia farms were above the WHO specification of 0.05 mg/kg As content in foods [31]. Oluyemi et al. [35] reported As content of $3.30 \pm 0.01 - 8.26 \pm 0.01$ mg/kg for unprocessed cocoyam from Nigeria which is higher compared to As content of $0.02 \pm 0.01 - 0.07 \pm 0.08$ mg/kg obtained by unprocessed and processed cocoyam. The difference in As content between these two studies may be related to the differences in geographical location and human activities that have gone on at the sampling points. Mercury (Hg) concentrations recorded for unprocessed and processed cocoyam in the present study was $0.14 \pm 0.03 - 0.19 \pm 0.06$ mg/kg and were far below the WHO recommended Provisional Tolerable Weekly Intake (PTWI) of 1.6 µg/kg Hg per body weight [31].

3.3. Variations in heavy metals of unprocessed and laboratory-processed plantain from Akwatia and Obuasi farms

Levels of the four heavy metals, in Table 4, are in the following respective ranges $0.22 \pm 0.19 - 0.60 \pm 0.71$ mg/kg (Mn), $0.04 \pm 0.02 - 1.11 \pm 0.65$ mg/kg (Pb), ND - <0.01 mg/kg (Cd), $<0.01 - 0.02 \pm 0.01$ mg/kg (As) and $0.04 \pm 0.02 - 0.13 \pm 0.05$ mg/kg (Hg). Manganese (Mn) concentration of $0.22 \pm 0.19 - 0.60 \pm 0.71$ mg/kg obtained by both unprocessed and laboratory-processed plantain from Akwatia and Obuasi was far lower compared to Mn content of $10.90 \pm 0.01 - 15.75 \pm 0.01$ mg/kg reported for unprocessed plantain by Oluyemi [35]. Oluyemi et al. [35] collected their plantain samples from a landfill site in Nigeria, whilst the plantain samples used in the present study have been collected from farms within Obuasi and Akwatia in Ghana; indicating possible differences in load of pollutants and therefore reasons for the differences in Mn contents [18]. Lead (Pb) contents ranging from $0.04 \pm 0.02 - 0.07 \pm 0.01$ mg/kg obtained by boiled and fried plantain from Obuasi and Akwatia farms were far lower compared to the Pb content of

Table 5

Mean heavy metal contents in processed and unprocessed yam from Akwatia and Obuasi farms.

Sites	Food Sample	Mn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	As (mg/kg)	Hg (mg/kg)
Obuasi	Boiled Yam	0.21 ± 0.10 ^a	<0.01 ^a	<0.01 ^a	0.01 ± 0.00 ^a	0.11 ± 0.06 ^a
	Fried Yam	0.15 ± 0.24 ^a	<0.01 ^a	ND ^a	<0.01 ^a	0.05 ± 0.02 ^a
	Roasted Yam	0.95 ± 0.56 ^b	0.01 ± 0.01 ^b	0.01 ± 0.00 ^a	0.07 ± 0.03 ^b	0.85 ± 0.11 ^b
	Unprocessed	1.44 ± 0.67 ^c	1.00 ± 0.64 ^c	0.01 ± 0.01 ^a	0.08 ± 0.03 ^b	1.34 ± 0.23 ^c
	Boiled Yam	0.18 ± 0.22 ^d	<0.01 ^a	<0.01 ^a	0.01 ± 0.00 ^a	0.08 ± 0.03 ^b
	Fried Yam	0.12 ± 0.09 ^b	ND ^a	<0.01 ^a	<0.01 ^a	0.02 ± 0.01 ^a
	Roasted Yam	0.89 ± 0.54 ^c	0.01 ± 0.01 ^b	0.01 ± 0.00 ^a	0.09 ± 0.01 ^b	0.79 ± 0.09 ^b
	Unprocessed	1.35 ± 0.43 ^f	0.99 ± 0.80 ^c	0.07 ± 0.02 ^b	0.09 ± 0.02 ^b	1.25 ± 0.24 ^c
	LSD (5%)	0.02	<0.01	<0.01	0.02	0.11
	CV (%)	12.7	13.3	17.34	10.7	12.5

*ND means not detected. Figures are presented as means ± standard deviations. Superscripts to figures in same column implies significant or not significantly different at $p > 0.05$ (ANOVA, Duncan test). LOD for Pb = 0.0000 ppb, Cd = 0.0000 ppb.

$1.02 \pm 0.65 - 1.11 \pm 0.65$ mg/kg obtained by unprocessed plantain from Obuasi and Akwatia farms and the difference was statistically ($p \leq 0.05$) significant. Arsenic (As) levels ($1.67 \pm 0.01 - 4.36 \pm 0.01$ mg/kg) and Pb levels ($1.02 \pm 0.65 - 1.11 \pm 0.65$ mg/kg) reported by Oluyemi [35], for unprocessed plantain, were all far higher than what were obtained for both unprocessed and processed plantain in this study. The highest Hg content recorded in the present study was 0.13 ± 0.05 mg/kg and was obtained by unprocessed plantain from Akwatia farm. WHO has recommended a Provisional Tolerable Weekly Intake (PTWI) of Hg as 1.6 µg/kg body weight [36].

3.4. Variations in heavy metal contents of unprocessed and laboratory-processed yam from farms in Obuasi and Akwatia

The mean concentrations of Mn recorded for unprocessed and laboratory-processed yam samples were in the range of $0.12 \pm 0.09 - 1.44 \pm 0.67$ mg/kg with the latter obtained by unprocessed yam samples (Table 5). Both values were above the recommended WHO specification of 0.05 mg/kg Mn content in foods [31]. The difference between the minimum and maximum Mn concentrations recorded for unprocessed and processed yam from Obuasi and Akwatia farms was statistically significant at $p \leq 0.05$. There was a sharp decrease in the level of Mn moving from unprocessed yam to boiled yam and fried yam and it was statistically significant. However, there were no significant differences between the level of Mn obtained by boiled yam and fried yam. Even though Mn is considered as essential metal because of its role as a co-factor in metabolic and biosynthetic processes, when high, it can be worrying because they become toxic when accumulated in human body tissues and are not metabolized [37,38]. Unprocessed and processed yam samples from Obuasi and Akwatia farms gave Pb concentrations ranging from <0.01 to 1.00 ± 0.64 mg/kg, respectively, and were far below the WHO specification of 2.00 mg/kg Pb content in root and tuber crops [31]. Statistically, there was no significant difference between Pb content recorded for boiled yam and fried yam samples from Obuasi and Akwatia farms. The Pb content obtained by boiled yam (<0.01 mg/kg), fried yam (<0.01 mg/kg) and roasted yam (0.01 ± 0.01 mg/kg) from Obuasi and Akwatia farms were all lower compared to Pb concentration of ($0.99 \pm 0.80 - 1.00 \pm 0.64$ mg/kg) recorded for unprocessed yam from both Obuasi and Akwatia farms. Ofori et al. [34]

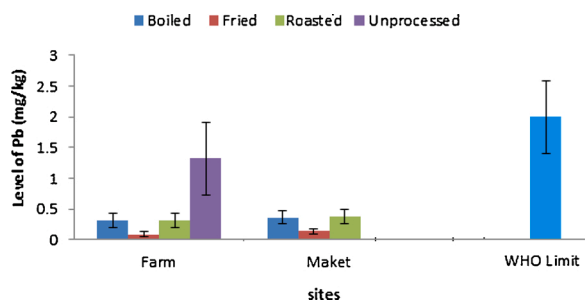


Fig. 1. Comparison of Mn levels in farm and market samples to WHO specification.

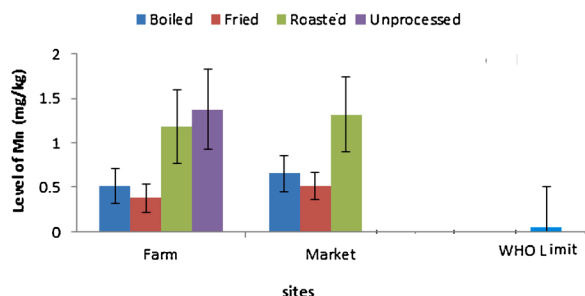


Fig. 2. Comparison of Pb concentrations in farm and market samples to WHO specification.

reported Pb concentration of <0.01 mg/kg in water yam flour and was in agreement with Pb content of <0.01 mg/kg recorded for boiled yam, and fried yam samples in the present study. The highest Cd content of 0.07 ± 0.02 mg/kg was obtained by unprocessed yam from a farm in Akwatia, whilst the lowest of < 0.01 mg/kg Cd content was recorded for fried yam prepared out of fresh yam bought from a farm in Akwatia. The highest Cd concentration recorded for both unprocessed and processed yam samples was slightly above the WHO specification of 0.02 mg/kg Cd content in foods of root and tuber [31]. Also, Table 4 indicates that compared to the boiling and frying, roasting did not cause more reduction in the As content of yam. This is worrying given that WHO minimum specification for As is 0.05 mg/kg and also the fact that As is toxic and long term exposure to it has been linked to increased risk of cancer [39]. The mean concentrations of Hg obtained by unprocessed and processed yam samples from farms in Obuasi and Akwatia were in the range 0.02 ± 0.01 – 1.34 ± 0.23 mg/kg. The difference between the highest Hg content of 1.34 ± 0.23 mg/kg recorded for unprocessed yam from Obuasi farm and the lowest of 0.02 ± 0.01 mg/kg Hg content, obtained by fried yam from Akwatia farm, was statistically significant and both values were below WHO recommended weekly minimum tolerable level of 1.6 µg/kg body weight [31].

3.5. Comparison of heavy metals in laboratory-processed foods to already prepared foods purchased from Akwatia and Obuasi Markets

The order of increasing Mn content determined in processed food

samples as roasted > boiled > fried for both farm and market samples from Obuasi and Akwatia (Fig. 1). The highest mean Mn content of 1.38 mg/kg was recorded for unprocessed samples and the lowest mean Mn content of 0.38 mg/kg was achieved by fried samples from farm. The difference between the highest (1.38 mg/kg) and the lowest Mn contents (0.38 mg/kg) recorded for farm and market samples were statistically significant and unfortunately far above the WHO specification of 0.05 mg/kg Mn content in foods [31].

The highest mean Pb content of 1.32 mg/kg was recorded for unprocessed sample, and lowest of 0.09 mg/kg obtained by fried sample, and were all below the WHO specification of 2.00 mg/kg Pb content in foods (Fig. 2). Lead (Pb) content in the prepared commodities from the markets were in the range <0.01 to 1.07 mg/kg and were above the Pb levels of <0.01 – 0.05 mg/kg reported by Tortoe et al. [40] when they determined trace metal concentrations in three pastry product from roots and tuber and cereal composite flour.

In general, the heavy metal contents determined in fried yam samples were lower compared to boiled and roasted samples from both farm and market. Lead levels determined in unprocessed and processed yam samples were higher compared to other commodities across all locations. The Pb contamination in the unprocessed samples may be related to high vehicular traffic and car exhaust emissions on Obuasi and Akwatia roads. However, these sources are impossible to discriminate, and it cannot be determined whether environmental or a postharvest process contaminated the cocoyam.

3.6. Effect of Processing Methods on the Extent of Reduction of Heavy metal Concentration in the four food crops

Differences were recorded in the percentage reductions in heavy metal contents of the four food crops when processed by the three methods (Table 6). Reductions in heavy metal contents occurred best in frying, followed by boiling and then roasting. Reductions in Pb contents were more pronounced than the rest of the heavy metals. In terms of the food crops, the order of decreasing percentage reduction in heavy metal contents, as a result of the processing, was Yam > Plantain > Cassava > Cocoyam. All these heavy metals are held within food matrixes by various surface forces phenomena [41]. All three methods of processing involve complex mechanisms involving mass and heat transfers and considerably alter the plant cell walls of the food crops [42]. Boiling, roasting and frying, as cooking methods, have different mechanisms. Boiling involves wet processing in which there is considerable mass movement, involving a two-phase heat transfers, the latent of vaporization of the liquid is used for dissipating large amounts of heat flux [43]. This must account for the relatively high percentage reduction in all four heavy metals when all four food samples were boiled, as shown in Table 5. Frying is considered a dry processing cooking method in which there is also removal of water, oil substitutes the water in the tissues as vapour into the oil [44]. Roasting is essentially drying, in which the heat elements applied are much closer to the food than in ordinary drying. This results in much severe physical changes such as shrinkage than in drying [45]. Prothon et al. [46] further explain that during roasting there is pronounced destruction of the epidermal cell wall, resulting in more shrinkage of the tissues. This will certainly result in reduced ability of the heavy metals from moving out from the food

Table 6

Percentage reductions in contents of four heavy metals during boiling, frying and roasting of cassava, cocoyam, plantain and yam.

Home Processing Methods	Percentage reduction (%) in Heavy Metal Content of Processed Cassava, Cocoyam, Plantain and Yam															
	Cassava				Cocoyam				Plantain				Yam			
	Mn	Pb	As	Hg	Mn	Pb	As	Hg	Mn	Pb	As	Hg	Mn	Pb	As	Hg
Boiling	30.7	71.3	50	59.4	65.6	59.5	76.4	21.1	30.2	93.9	50	34.9	86	99	88.2	92.7
Frying	50.3	96.4	50	82.3	73.6	75.6	84.3	26.3	48.8	95.8	50	42.9	90.3	99	100	97.3
Roasting	17.1	33.4	50	19.3	1.5	8	52.9	5.3	15.5	59.8	50	8	34.1	99	6.3	36.7

Table 7
Estimated Average Dietary Intake (EADI) and Hazard Index (HI) for Laboratory-Processed and Unprocessed Food Samples from Akwatia.

	EADI				Mean EADI	HI				Mean HI
Heavy metal	Boiled Cassava	Fried Cassava	Roasted Cassava	Unprocessed Cassava		Boiled Cassava	Fried Cassava	Roasted Cassava	Unprocessed Cassava	
As	0.0001	0.0000	0.0001	0.0001	0.0001	0.0350	0.0000	0.0356	0.0713	0.0355
Hg	0.0004	0.0002	0.0008	0.0011	0.0006	0.2493	0.1496	0.5484	0.7977	0.4363
Mn	0.0044	0.0030	0.0040	0.0044	0.0039	0.0044	0.0030	0.0040	0.0044	0.0039
Pb	0.0043	0.0006	0.0007	0.0150	0.0052	1.4259	0.1901	0.2377	5.0145	1.7171
Total	0.0092	0.0038	0.0056	0.0206	0.0098	1.7146	0.3427	0.8257	5.8879	2.1927
	Boiled Cocoyam	Fried Cocoyam	Roasted Cocoyam	Unprocessed Cocoyam		Boiled Cocoyam	Fried Cocoyam	Roasted Cocoyam	Unprocessed Cocoyam	
As	0.0001	0.0000	0.0001	0.0001	0.0001	0.0260	0.0174	0.0521	0.0607	0.0391
Hg	0.0003	0.0002	0.0003	0.0003	0.0003	0.1820	0.1699	0.2114	0.2305	0.1985
Mn	0.0014	0.0009	0.0035	0.0035	0.0023	0.0014	0.0009	0.0035	0.0035	0.0023
Pb	0.0005	0.0004	0.0011	0.0013	0.0008	0.1735	0.1215	0.3760	0.4222	0.2733
Total	0.0023	0.0015	0.0050	0.0052	0.0035	0.3829	0.3097	0.6430	0.7169	0.5131
	Boiled Plantain	Fried Plantain	Roasted Plantain	Unprocessed Plantain		Boiled Plantain	Fried Plantain	Roasted Plantain	Unprocessed Plantain	
As	0.0005	ND	0.0005	0.0009	0.0006	0.0232	ND	0.0232	0.0465	0.0309
Hg	0.0002	0.0002	0.0006	0.0006	0.0004	0.1625	0.1300	0.3901	0.4226	0.2763
Mn	0.0015	0.0010	0.0017	0.0021	0.0016	0.0015	0.0010	0.0017	0.0021	0.0016
Pb	0.0017	0.0020	0.1085	0.0052	0.0294	0.0930	0.6198	0.2014	1.7200	0.6586
Total	0.0039	0.0032	0.1113	0.0088	0.0319	0.2802	0.7508	0.6164	2.1912	0.9674
	Boiled Yam	Fried Yam	Roasted Yam	Unprocessed Yam		Boiled Yam	Fried Yam	Roasted Yam	Unprocessed Yam	
As	0.0000	ND	0.0002	0.0002	0.0001	0.0114	ND	0.1027	0.1027	0.0723
Hg	0.0002	0.0000	0.0018	0.0029	0.0012	0.1277	0.0319	1.2613	1.9957	0.8542
Mn	0.0004	0.0003	0.0020	0.0031	0.0015	0.0004	0.0003	0.0020	0.0031	0.0013
Pb	ND	ND	0.0000	0.0023	0.0012	ND	ND	ND	0.7610	0.7610
Total	0.0006	0.0003	0.0040	0.0085	0.0039	0.1395	0.0322	1.3640	2.8625	1.6887

*ND = Not Detected, EADI = Estimated Average Daily Intake (mgkg⁻¹bw⁻¹ d⁻¹) and HI= Hazard Index.

Table 8
Estimated Average Dietary Intake (EADI) and Hazard Index (HI) for Laboratory-Processed and Unprocessed Food Samples from Obuasi.

	EADI				Mean EADI	HI				Mean HI
Heavy metal	Boiled Cassava	Fried Cassava	Roasted Cassava	Unprocessed Cassava		Boiled Cassava	Fried Cassava	Roasted Cassava	Unprocessed Cassava	
As	0.0001	ND	0.0001	0.0001	0.0001	0.0356	ND	0.0356	0.0713	0.0475
Hg	0.0004	0.0001	0.0008	0.0009	0.0006	0.2991	0.0997	0.5484	0.5983	0.3864
Mn	0.0027	0.0021	0.0036	0.0043	0.0032	0.0027	0.0021	0.0036	0.0043	0.0032
Pb	0.0043	0.0005	0.0011	0.015	0.0052	1.4497	0.1664	0.3802	5.0145	1.7527
Total	0.0075	0.0027	0.0056	0.0203	0.0091	1.7871	0.2682	0.9678	5.6884	2.1898
	Boiled Cocoyam	Fried Cocoyam	Roasted Cocoyam	Unprocessed Cocoyam		Boiled Cocoyam	Fried Cocoyam	Roasted Cocoyam	Unprocessed Cocoyam	
As	0.0001	0.0000	0.0001	0.0001	0.0001	0.0260	0.0174	0.0521	0.0607	0.0391
Hg	0.0003	0.0002	0.0003	0.0003	0.0003	0.1820	0.1699	0.2114	0.2305	0.1985
Mn	0.0011	0.0010	0.0035	0.0035	0.0023	0.0011	0.0010	0.0035	0.0035	0.0023
Pb	0.0007	0.0003	0.0016	0.0017	0.0011	0.2314	0.1157	0.5495	0.5784	0.3688
Total	0.0022	0.0015	0.0055	0.0056	0.0037	0.4405	0.3040	0.8165	0.8731	0.6085
	Boiled Plantain	Fried Plantain	Roasted Plantain	Unprocessed Plantain		Boiled Plantain	Fried Plantain	Roasted Plantain	Unprocessed Plantain	
As	0.0005	ND	0.0005	0.0009	0.0006	0.0232	ND	0.0232	0.0465	0.0309
Hg	0.0005	0.0005	0.0005	0.0001	0.0004	0.3576	0.3251	0.3557	0.3901	0.3571
Mn	0.0018	0.0014	0.0023	0.0030	0.0022	0.0018	0.0014	0.0023	0.0030	0.0022
Pb	0.0003	0.0019	0.0006	0.0047	0.0019	0.0930	0.6198	0.2014	1.5804	0.6237
Total	0.0031	0.0038	0.0039	0.0087	0.0050	0.4756	0.9463	0.5826	2.1100	1.0364
	Boiled Yam	Fried Yam	Roasted Yam	Unprocessed Yam		Boiled Yam	Fried Yam	Roasted Yam	Unprocessed Yam	
As	0.0000	ND	0.0002	0.0002	0.0001	ND	ND	0.0799	0.0913	0.0856
Hg	0.0003	0.0001	0.0019	0.0031	0.0014	0.1756	0.0798	1.3571	2.1394	0.9379
Mn	0.0005	0.0003	0.0022	0.0033	0.0012	0.0005	0.0003	0.0022	0.0033	0.0016
Pb	ND	ND	0.0000	0.0023	0.0012	ND	ND	ND	0.7610	0.7610
Total	0.0008	0.0004	0.0043	0.0089	0.0042	0.1761	0.0801	1.4392	2.9950	1.7862

*ND = Not Detected, EADI = Estimated Average Daily Intake (mgkg⁻¹bw⁻¹ d⁻¹) and HI= Hazard Index.

matrix as readily as in boiling. This, together with the arguments made earlier, might explain the differences in the relative percentages reductions in the heavy metal amounts in the four food crops as they were processed by the three methods.

3.7. Dietary exposure and health risk assessment

The health risk assessment for systemic effects associated with heavy metals determined in the food samples from Akwatia and Obuasi are shown in Tables 7 and 8. The estimated average daily intake (EDAI) calculated was in the range of 0.000 and 0.021 mg kg⁻¹bw⁻¹ d⁻¹ with corresponding hazard indices ranging from 0.00–5.00 for the tested metals in yam, cocoyam and cassava from Obuasi and Akwatia. Both Tables 6 and 7 show that reductions in EADI and HI, across all heavy metals, from the two study sites, as a result of the processing methods used for the food crops. The reductions occurred more through boiling and frying than with roasting. These are consistent with the results obtained as discussed in Section 3.6. According to the Joint FAO/WHO Expert Committee on Food Additives [36], the tolerable acceptable daily intake levels of As, Hg, Mn and Pb are 0.002, 0.5, 1.0 and 0.5 mg/kg/bw/day, respectively. Even though the HI of slightly more than 5 for unprocessed cassava, from the two study sites, is worrying, overall the study clearly indicate that all three home processing methods are able to reduce the concentrations of heavy metals in the four food crops to tolerable levels.

4. Conclusion

The study revealed that the concentrations of As, Cd, Hg, Mn and Pb in cassava, cocoyam, plantain and yam can be reduced significantly to tolerable levels, through boiling, frying and roasting. The most effective method was through boiling and least through roasting. In general, the heavy metal concentrations determined in fried yam samples were lower compared to boiled and roasted samples from both farm and market. Lead contents determined in unprocessed and processed yam samples were higher compared to other commodities across all locations. The hazard indices recorded for all metals in food crops, except for Pb in unprocessed cassava, boiled cassava and unprocessed plantain and Hg (unprocessed yam and roasted yam), were below one and posed no health risks to consumers.

Author statement

Rebecca Adjei-Mensah: Conceptualisation, design of experiment, methodology.

Hayford Ofori: Methodology, editing.

Charles Tortoe: Conceptualisation, design of experiment, editing.

Paa-Nii Torgbor Johnson: Conceptualisation, design of experiment, editing.

David Aryee: Data and statistical analysis.

Samuel Kofi Frimpong: Data and statistical analysis.

Rebecca Adjei-Mensah and Hayford Ofori: Wrote first draft of the paper.

Declaration of Competing Interest

The authors declare no conflict of interest.

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