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ANALYTICAL CHEMISTRY | RESEARCH ARTICLE

Trace metal concentrations in three pastry products prepared from root and tuber and cereal crops composite flours

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Abstract: The concentrations of Arsenic (As), Lead (Pb), Mercury (Hg), Copper (Cu), Iron (Fe), and Zinc (Zn) in doughnuts, cookies, and bread prepared from root and tuber and cereal crops composite flours were quantitatively measured by Atomic Absorption Spectrophotometry. Varying concentrations of trace metals were identified in three pastry products. Concentration of Hg was <0.01 mg/kg wet weight in all three pastry products. The As levels in all three pastry products ranged between <0.01 and 0.03 mg/kg. Concentrations of Pb was <0.01–0.05 mg/kg, far lesser than 10 mg/kg recommended limit by World Health Organization (WHO). The highest Fe level of 66.3 mg/kg occurred in cookies made from millet composite flour, whereas the lowest value of 10.4 mg/kg was in doughnuts made from water yam composite flour. Concentrations of Cu (1.03–1.83 mg/kg) and Zn (5.49–13.72 mg/kg) were lower than the tolerance limit of 40 mg/kg set by WHO. The presence of trace metals at varying concentrations in all three pastry products demonstrates the need for observing food safety controls in sourcing for raw materials and during processing of flours from root and tuber and cereal crops.

Subjects: Food Chemistry; Preservation; Processing; Product Development

Keywords: trace metals; composite flour; bread; doughnuts; cookies; root and tuber; cereal crops

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PUBLIC INTEREST STATEMENT

Pastry and confectionary products made from root and tuber and cereal crops composite flours are gaining popularity due to their health benefits and low production cost. However, knowledge on trace metals contamination of these products are limited to inform their preparations. This study explores the trace metal concentrations in doughnuts, cookies, and bread prepared from root and tuber, cereal and legume composite flours. Interestingly, the three pastry products had varied trace metal concentrations. The concentrations of Cu, Hg, Pb, and Zn were lower than tolerance limits set by WHO. However, concentrations of As and Fe were sometimes above tolerance limits. Clearly, the variations in the trace metal concentrations in the three pastry products requires adhering to food safety controls in acquiring the raw materials, processing of the flours, and preparation of the pastry products.

1. Introduction

Food safety is generally an important issue for every country in order to guarantee the safety of its citizens. Trace metal contamination of food is one of the food safety concerns of public health. Trace metals are broadly categorized into essential and non-essential trace metals. The essential trace metals such as cupper (Cu), iron (Fe), and zinc (Zn) are important for growth within tolerable limits but become toxic in the body beyond the tolerable limits, whereas the non-essential trace metals including arsenic (As), lead (Pb), and mercury (Hg) may pose a health risk even at very low concentrations. Trace metals are naturally occurring with high atomic numbers and cannot be easily degraded. As a result, their accumulation has deleterious effect on the body. They often find their way into the food chain through the environment they are exposed into.

There are public health concerns from trace metals because they can breakdown the functions of the nervous system and kidney, cause lung irritation, alter blood composition, and also reduce energy levels (*1,2*). Exposure to lead (Pb) has been associated with hypertension, neurological disorders, and interference in the metabolism of vitamin D (*3*). Chronic exposure to arsenic (As) resulted in weakness, mental abnormalities, and debility (*3*). Additionally, mercury toxicity resulted in nervous breakdown, digestive and immune system malfunction, skin and eye lesions (*3*).

The presence of trace metals in many food products have been reported. High levels of arsenic (As) have been reported in rice and vegetables from areas where fields were irrigated with water contaminated with high amounts of arsenic (As) (*4,5*). Fruits and vegetables sampled from Nigeria (*6*), Egypt (*7*) were found to contain some levels of trace metals. Previous works by Ofori et al. (*8*), Shin et al. (*9*), Nedzarek et al. (*10*), and Hoha et al. (*11*) also independently established the presence of trace metals such as lead (Pb), nickel (Ni), arsenic (As), and cadmium (Cd) in cereal and tuber flours, yam flour products, coffee, beverages, and processed meat. Subsequently, Hajeb et al. (*1*) observed that the key factor in susceptibility of contamination by a particular trace metal depends on the structure, chemical composition, and kind of food.

In many countries, commercial utilization of flours from crops such as cassava, cocoyam, sweetpotato, sorghum, and millet for bakery and confectionery products is on the ascendency. This is especially so as these crops provide alternative cheaper, easy to produce flours suitable as composite flours for a wide range of bakery and confectionery products including, bread, cookies, doughnuts, cakes, meat pies, chips, and noodles (*12–14*). Interestingly, most of these bakery and confectionery products have been assessed for their physiochemical parameters, proximate composition, and nutritional properties by these authors. However, there is a dearth in information concerning potential toxicity of these products due to trace metal contamination. There is, therefore, the need to critically examine the potential trace metal contaminants of products prepared from the flours of these crops. The aim of this study was, therefore, to determine the concentration of trace metals in 3 pastry products made from root and tuber and cereal composite flours.

2. Materials and methods

2.1. Raw materials

Matured cream flesh sweetpotato (*Ipomoea batatas*) and water yam (*Dioscorea alata*) were sourced from a root and tuber supplier at Medina market in Accra, Ghana. Pearl millet (*Pennisetum glaucum*) and red sorghum (*Sorghum bicolor*) were purchased from a local cereal supplier at Medina market in Accra, Ghana. These raw materials were sorted and cleaned of sand and other foreign materials and stored at room temperature (28°C) prior to processing into flours. Wheat flour, margarine, eggs, and milk were purchased from a local shop in Accra, Ghana.

2.2. Preparation of root and tuber flour

The sweetpotato or water yam flours were produced according to the method of Tortoe et al. (*15,16*). The roots or tubers were washed in potable water, hand-peeled and washed again. The roots or tubers were then manually cut into slices (2–4 mm thick) into 0.01% Sodium metabisulphite solution to prevent browning and subsequently spread thinly on trays and dried at 60°C for 14 h in a mechanical dryer (Apex Dryer, CSIR-FRI, Accra, Ghana). The dried sweetpotato or water yam slices were milled into flour using a hammer mill (Christy and Norris Ltd., Surrey, UK) to pass through a 250 µm sieve. The flour was packaged in transparent polypropylene bags, sealed airtight impulse sealer (Oalink, QNS-3200HI) and stored at room temperature until use later.

2.3. Preparation of cereal flour

Cereals were processed into flour using the method described by Addo et al. (*17*). The sorghum or millet grains were sorted, winnowed, washed, spread on a clean black polythene sheet, and solar dried to a moisture content of about 11%. The dried grains were milled into fine flour to pass through a 250 µm sieve using hammer mill.

2.4. Composite flour

Wheat flour (80%) was mixed with either 20% sweetpotato flour, water yam flour, sorghum flour, or millet flour using a high-speed blender to form a uniform composite flour. The resulting flour was used in the production of the pastry products.

2.5. Composite flour pastries

The pastry products (doughnut, cookies, and bread) were produced according to methods described by Tortoe et al. (*15*). The doughnuts were processed by rubbing-in margarine into previously mixed dry ingredients before milk and eggs were added. The mixture was whipped, scooped into smaller portions before deep frying in moderately hot oil. For cookies, margarine and sugar were creamed until light and fluffy. Flour and baking powder were gradually added to the cream, mixed uniformly, piped out and baked at 175°C for 25 min. The bread was processed by mixing composite flour, margarine, sugar, salt, and instant baking yeast thoroughly for 3 min at low speed before kneading into a soft dough. The dough was allowed to rise for 30 min before panning and proofing at room temperature for 120 min. The loaves were baked at 175°C for 20 min. Products were cooled to room temperature before chemical analysis.

2.6. Determination of trace metal

The dry ashing method was used for the Atomic Absorption Spectrometry (AAS) analysis (*18*). Buck Scientific 210VGP Flame Atomic Absorption Spectrophotometer (Buck Scientific, Inc. East Norwalk, USA) was used to read the absorbance values at appropriate wavelength of the interested metal in sample solution. Cathode lamps used were As (wavelength 193.7 nm, lamp current 4.0 mA), Cu (wavelength 324.8 nm, lamp current 1.5 mA), Fe (wavelength 248.3 nm, lamp current 7.0 mA), Hg (wavelength 253.7 nm, lamp current 0.7 mA), Pb (wavelength 217.0 nm, lamp current 3.0 mA), and Zn (wavelength 213.9 nm, lamp current 2.0 mA). Using a calibration graph consisting of a minimum of three standards, the metal content of the sample was derived.

2.7. Quality control of results

In order to ensure reliability of results, samples were handled carefully to avoid contamination. Recovery test for the analytical procedures was also carried out for the metals analyzed as described by Ofori et al. (*8*). Samples were spiked with aliquots of metal standards and analyzed for the selected metals. Acceptable recoveries of 95 ± 1%, 97 ± 1%, 95 ± 1%, 94 ± 1%, 95 ± 1%, and 96 ± 1% were obtained for As, Cu, Fe, Hg, Pb, and Zn, respectively.

2.8. Data analysis

The mean values of duplicate determinations were compared by ANOVA and Duncan Multiple Range Test (SPSS 21.0, SPSS Inc., USA) was used to separate significantly different means. Level of significance was set at 95% confidence interval.

3. Results and discussion

3.1. Trace metal concentration of doughnut

Arsenic (As) and lead (Pb) levels ranged between 0.01 and 0.03 mg/kg wet weight of product (Table 1). Comparing the levels of As obtained and the Provisional Tolerable Weekly Intake (PTWI) of 0.015 mg/kg body weight for As (*19*), only water yam composite flour doughnut fell below the tolerable limit. Doughnuts from the different composite flours did not vary significantly ($p > 0.05$) in their As and Pb levels. However, there were significant differences ($p < 0.05$) in Cu, Fe, and Zn levels of doughnuts made from all the different composite flours. According to Dobaradaren, et al. (*20*), Pb at trace levels are toxic and only 10 mg/kg is allowed even in raw materials by World Health Organization (*21*). The Pb levels obtained for doughnuts were far below the tolerable limit. The concentrations of Cu in the doughnuts ranged from 1.06 to 1.42 mg/kg (Table 1). Copper (Cu) was highest in doughnuts in descending order of water yam > sweetpotato > sorghum > millet composite flours, but these values were far less than the acceptable limits of 40 mg/kg in foods by World Health Organization (*21*). Although accumulation of Fe in the body causes colorectal cancer its deficiency causes anemia (*22*). The required amount recommended in food by World Health Organization is 15 mg/kg (*21*). Only millet composite flour doughnut had Fe (46.38 mg/kg) above the recommended level. The Hg in all the composite flour doughnuts was less than 0.01 mg/kg, comparable to the Provisional Tolerable Weekly Intake (PTWI) of 1.6 µg/kg body week (*3*). Additionally, peripheral vision, disturbances in sensations, muscle weakness, and lack of movement coordination are some toxicity associated with Hg according to Xiong et al. (*23*). The doughnuts have Zn concentrations ranging from 6.09 to 8.25 mg/kg, far lesser than the recommended limit of 60 mg/kg (*21*).

3.2. Trace metal concentration of cookies

Arsenic (As), Cupper (Cu), Mercury (Hg), Lead (Pb), and Zinc (Zn) levels in the cookies followed a similar trend as observed for doughnuts (Table 2). Among cookies made from the different composite flours, significant differences ($p \leq 0.05$) were only established in Cu, Fe, and Zn levels. The range of As in the composite flour cookies was 0.02–0.03 mg/kg and only sorghum composite flour cookies of As (0.02 mg/kg) was comparable to the Provisional Tolerable Weekly Intake (PTWI) of 0.015 mg/kg (*23*). The highest concentration of Pb in the cookies was observed in water yam and millet composite flours (0.03 mg/kg), less than 10 mg/kg recommended (*21*). The cookies contained lesser concentrations of Cu in the range of 1.03–1.42 mg/kg lesser than 40 mg/kg in foods (*21*). Comparing the level of Fe in the cookies with 15 mg/kg recommended (*21*), only sweetpotato composite flour cookie (13.22 mg/kg) was within acceptable limit. The Fe concentrations were generally higher. Iron (Fe) concentrations in sorghum composite flour cookies was nearly three times higher whereas millet composite flour cookies was nearly four times higher compared to WHO standard of 15 mg/kg (*21*). All the composite flour cookies had similar Hg concentration as observed in doughnuts. However, the levels of Zn in the cookies were higher (6.93–13.25 mg/kg) compared to levels observed for doughnuts but lesser than 60 mg/kg recommended (*21*) (Tables 1 and 2).

3.3. Trace metal concentration of bread

In the bread, As concentration was <0.01–0.03 mg/kg and water yam composite flour bread recorded the lowest (<0.01 mg/kg) (Table 3) comparable to 0.015 mg/kg, which is Provisional Tolerable Weekly Intake (PTWI) set (*19*). The levels of the trace metals of As, Cu, Fe, and Zn were significantly different ($p < 0.05$) among the composite flours (Table 3). The lowest Pb level (<0.01 mg/kg) was recorded in water yam composite flour bread whereas the highest Pb (0.05 mg/kg) was recorded for millet composite flour bread, which are all lesser than 10 mg/kg recommended (*21*). A range of 1.05–1.83 mg/kg for Cu was obtained for bread, which was lesser than the recommended limits of 40 mg/kg in foods (*21*). The concentrations of Fe in the bread were highest (40 mg/kg) for sorghum composite flour bread (Table 3). Only water yam and millet composite flour bread had Fe levels within range of the recommended limit of 15 mg/kg recommended (*21*). The concentrations of Hg (<0.01 mg/kg) in the bread were similar to both doughnuts and cookies. Varying concentrations of Zn were recorded in bread made from the different composite flours, ranging between 5.5 and 13.7 mg/kg, however, the levels were lower than the recommended limit by WHO (*21*) of 60 mg/kg.

Table 1. Trace metal concentration in mg/kg wet weight of doughnuts made from different composite flours

Doughnuts	As	Cu	Fe	Hg	Pb	Zn
Sweetpotato	0.03 ± 0.01^{a}	1.38 ± 0.01^{bc}	11.96 ± 0.02^{b}	<0.01	0.03 ± 0.01^{a}	6.79 ± 0.02^{b}
Water yam	0.01 ± 0.01^{a}	1.42 ± 0.02^{c}	10.37 ± 0.02^{a}	<0.01	0.03 ± 0.01^{a}	6.09 ± 0.02^{a}
Sorghum	0.02 ± 0.00^{a}	1.37 ± 0.00^{b}	11.95 ± 0.00^{b}	<0.01	0.01 ± 0.01^{a}	6.78 ± 0.00^{b}
Millet	0.03 ± 0.01^{a}	1.06 ± 0.03^{a}	46.38 ± 0.04^{c}	<0.01	0.02 ± 0.01^{a}	8.25 ± 0.01^{c}

Note: Superscripts within the same column imply significant differences at $p \leq 0.05$.

Table 2. Trace metal concentration in mg/kg wet weight of cookies made from different composite flours

Cookies	As	Cu	Fe	Hg	Pb	Zn
Sweetpotato	0.03 ± 0.01^{a}	1.03 ± 0.04^{a}	13.22 ± 0.01^{a}	<0.01	<0.01	6.93 ± 0.02^{a}
Water yam	0.03 ± 0.02^{a}	1.42 ± 0.01^{c}	22.09 ± 0.01^{b}	<0.01	0.03 ± 0.01^{a}	7.11 ± 0.01^{b}
Sorghum	0.02 ± 0.01^{a}	1.25 ± 0.01^{b}	41.55 ± 0.01^{d}	<0.01	<0.01	13.25 ± 0.01^{d}
Millet	0.03 ± 0.01^{a}	1.28 ± 0.01^{b}	66.25 ± 0.0^{c}	<0.01	0.03 ± 0.02^{a}	12.46 ± 0.02^{c}

Note: Superscripts within the same column imply significant differences at $p \leq 0.05$.

Table 3. Trace metal concentration in mg/kg wet weight of bread made from different composite flours

Bread	As	Cu	Fe	Hg	Pb	Zn
Sweetpotato	0.03 ± 0.01^{b}	1.83 ± 0.02^{c}	16.62 ± 0.02^{a}	<0.01	0.03 ± 0.02^{a}	11.6 ± 0.02^{a}
Water yam	<0.01	1.08 ± 0.02^{a}	14.42 ± 0.03^{d}	<0.01	<0.01	10.4 ± 0.02^{d}
Sorghum	0.02 ± 0.02^{ab}	1.62 ± 0.01^{b}	40.00 ± 0.01^{c}	<0.01	0.03 ± 0.02^{a}	13.72 ± 0.01^{c}
Millet	0.03 ± 0.01^{b}	1.05 ± 0.01^{a}	11.09 ± 0.02^{b}	<0.01	0.05 ± 0.04^{b}	5.49 ± 0.03^{b}

Note: Superscripts within the same column imply significant differences at $p \leq 0.05$.

Generally, As, Hg, and Pb are toxic contaminants and tolerable only at very low concentration by the body, whereas Cu, Fe, and Zn are regarded as essential elements within acceptable limits and toxic contaminants above tolerable limits. Acceptable limits of Cu, Fe, and Zn are required for the body's proper function. Interestingly, Fe plays a major role in the formation of hemoglobin, Cu is needed for proper metabolic function, and Zn is essential in strengthening the immune system stabilizing DNA and for gene expression (*24*). However, above recommended levels they may distort proper functionality. According to Oyaro et al. (*25*) accumulated amounts of Cu may result in nausea or vomiting and excessive amounts of Zn may damage the pancreas or interfere with protein metabolism.

Lin et al. (*26*) reported As levels between 0.2 and 1.2 mg/kg in rice gain, suggested that some grains may have been contaminated from heavily polluted fields. In a study by Ofori et al. (*8*), the authors reported As levels below allowable limit in some processed tuber and cereal composite flours, which is an indication that these flours may serve as a raw material in various food applications. The As levels in all three pastry products ranged between <0.01 and 0.03 mg/kg, comparable to the 0.015 mg/kg Provisional Tolerable Weekly Intake (PTWI) (*19*).

Lead (Pb) in all the products examined was between <0.01 and 0.05 mg/kg lesser than 10 mg/kg recommended limit (*21*). According to Chary et al. (*27*) accumulation of Pb occur when there is a high concentration of this element in the soil. This suggests that the fields in which the cereals and root tubers were cultivated were not likely contaminated by Pb. In all the products examined, Hg was below the tolerable limit of 1.6 μg/kg stipulated by World Health Organization (*3*). Exposure to doses of Hg beyond these tolerable limits may cause neurotoxicity (*28*).

Among the three composite flour products, the highest Fe levels of 66.3 mg/kg (Table 3) was obtained in cookies made from millet composite flour whereas the lowest value of 10.4 mg/kg (Table 2) was observed in doughnuts made from water yam composite flour. The Fe levels in millet and sorghum products in the present study were generally lower than the amounts reported for millet (201.4 mg/kg) and sorghum (116.8 mg/kg) flours by Ofori et al. (*8*). Levels of Cu in the three composite flour products were lower than the tolerance limit of 40 mg/kg, and therefore, consumption of these products does not pose any risk of Cu toxicity (*3*). However, accumulation of high amounts above tolerable limits of Fe and Cu in the body has been associated with colorectal cancer (*22*). Similarly, Zn levels of the pastry products were below the allowable maximum of 40 mg/kg (*21*).

The difference in trace metal levels observed in this study may be ascribed to differences in composition of food products from root/tubers and cereals and environmental conditions under which these crops were cultivated. These elements essentially contaminate plants through the soil or polluted air, which precipitates and falls as rain (*1*). A key possibility for the high Fe contamination could be as a result of processing machinery. This is buttressed by reports from Perello et al. (*29*) and Cubadda et al. (*30*) indicating some processing methods and their machinery affect trace metal levels in food products.

4. Conclusion

All the products studied showed some extent of trace metal contamination. Apart from Fe in the millet and sorghum products, the concentrations involved were below the maximum limits stipulated by World Health Organization. The high Fe concentrations in all the three products above the tolerable amount of 15 mg/kg suggested contamination of soils in which the root and tubers or cereals were grown. Additionally, it may be due to the effect of processing methods and machinery, which may possibly contributed to the trace metal levels in these products. Consumption of these products, therefore, does not pose any health threat to the consumer. Therefore, the trace metal levels may not be directly ascribed to differences observed among the various composite flour products.

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Competing interests
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