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Effect of Milling on the Level of Heavy Metal Contamination of Some Nigerian Foodstuffs

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Abstract

The study investigated the metallic composition of the GUK and PREMIER milling plates by x-ray fluorescence (XRF) method. The plates were used in processing of some Nigerian foodstuffs such as maize, millet, sorghum and cowpea. Metallic contamination of the foodstuffs by milling with the two milling plates was determined using atomic absorption spectroscopy (AAS). Wet ashing of the dry and wet milled foodstuffs were employed for isolating the metals in food from their complex matrix before evaluation with AAS. The presence of iron, nickel and lead metals were detected in the unprocessed grains in the ranges of 5.95-13.10 mg/kg, 3.89-5.54 mg/kg, and 0.98-2.78 mg/kg, respectively. The level of metal contamination was found to be a function of their composition in the milling plate used.

Keywords: Heavy metals, PREMIER milling plate, GUK milling plate, unmilled-foodstuffs, milled-foodstuffs

1. Introduction

Food contamination by metals is an inevitable product of modern technology. Nriangu and Azcue [1] and WHO [2] reported that the diet of man is a major exposure route for most of the unknown toxic contaminants in the environment. Among these contaminants, heavy metals have contributed markedly to the contamination of foodstuffs. It has been reported that Hg, Pb, Cd, Ni, Cr, Co, V, Mn, Fe, Zn, Sn and several other metals appears in processed food everyday [3-6], but concentrations vary greatly [7-12]. Food processing equipment has long been recognized as a source of metal contamination of foodstuffs [13-16]. The concern of the food processor is to see that the product is free of toxic or harmful metals or even of essential metals in quantities great enough to cause poisoning [17]. However with high quality steel, Cheftel [18] and Reilly [17] pointed out that metals are unlikely to migrate from processing equipment into food. Low levels of heavy metals can often be reasonably achieved by using good manufacturing and processing practices [19].

Hernadez *et al*. [20], found by atomic absorption spectroscopy (AAS) that metal contaminants (Cu, Zn, Fe, Mg, Mn) originating from grinding tools prevailed over uptake of same metals from the soil in cocoa. The metal contaminants exhibit a spectrum of health hazards [21], and have long been associated with acute and sub-acute food borne intoxicants [22-23]. Lead toxicity was reported by ancient Greek and Arab physicians [24-26] and can trigger both acute and chronic symptoms of poisoning [27]. Lead is a typical cumulative poison [28-31]. The danger of chronic intoxications is the greater problem as reported by Adeniyi and Anetor [32]. Increasing awareness of the toxicity and widespread environmental distribution of metals has prompted many investigations into their levels in general food supply [33- 34].

In Nigeria, the use of milling machines to locally process foodstuff is all over the place and indeed it has become an economically attractive activity both in the urban and rural settings. The machines have already taken over the place of the

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old traditional methods of milling using stones. The machine is operated by a 6-horse power electric motor and uses milling plates at a time. When in operation the plates revolve and rob against each other as the foodstuff is being crushed into powder or paste. The milling plates eventually wears away into the milled foodstuffs as they rob against each other and after a while the plates have to be replaced. Currently, there is no regular testing of heavy metals in foodstuffs by the designated health authorities.

The interest of this work is to investigate the extent of contamination of processed foodstuffs by Fe, Ni and Pb from the milling plates, and the possible effect of consuming such foods, particularly by the metals that are introduced into the food during milling.

2. Materials and Methods

2.1. Reagents and Apparatus

All chemicals and reagents used were of the analytical grade and all solutions used were prepared with distilled water. All analyses were carried out in replications of fives. All analytical measurements were made with a Pre-Unicam Model SP 1900 AAS, equipped with Fe, Ni and Pb metallic hollow cathode lamp, and operated under conditions recommended by the manufacturer.

2.2. Sample Preparation and Metals Determination

Two types of milling plates GUK and PREMIER were employed for this research to investigate the contribution of each type to the level of metal in the milled foodstuffs. Four foodstuffs (maize sorghum, cowpea and millet) commonly consumed in Nigeria were carefully selected for this experiment. Wet ashing was employed for isolating the metals in the food samples from their complex matrix before evaluating them with AAS according to the method reported by AOAC [35-36]. This was achieved by heating 1.0 g of each of the food samples with a digestion mixture containing concentrated nitric, perchloric and sulphuric acids in a Kjedahl flask for some hours, where all the organic matter is oxidized to water and carbon (IV) oxide, while the metal ions are left behind as uncomplexed ions.

The clear digest was diluted with distilled water to the 500 mL mark. Appropriate dilutions were then made for each metal and aliquots of the diluted clear digest used for the AAS determination of the metals. This was repeated for each portion of the milled foodstuffs (dry and wet milled), and the level of Fe, Ni, and Pb contamination were determined.

Calibration curves were prepared for each (Fe, Ni, and Pb) metal element using standard solutions. The appropriate lamps and correct wavelength (248.3 nm for Fe, 232 nm for Ni and 283.3 nm for Pb) were selected on the instrument.

2.3. Analysis

The metallic content of the milling plates were first determined by x-ray fluorescence (XRF). The aliquot of the milled and unmilled samples were subjected to analysis of the metals (Fe, Ni, and Pb) using AAS. All analyses were performed using an air-acetylene flame [37-38]. The concentrations of individual metals were calculated from standard readings using the following relationship in Equation 1.

$$M(mg/100\ g\ sample) = \frac{F(mg/kg)\ x\ Solution\ volume\ x\ 10}{Aliquot\ volume\ x\ weight\ of\ sample} \quad 1$$

where *M* is the metal to be determined and *F* is the metal from the calibration curve.

3. Results and Discussions

3.1. The Metal Composition of the Milling Plates.

The result of the XRF analysis of the milling plates revealed that they were made up of mainly Fe, Sn, and Cr, other metals are Mn, Pb, and Ni (Table 1). Metals such as copper, zirconium, niobium, and molybdenum were in trace amounts.

3.2. Level of Fe, Ni and Pb Metals in Unmilled Foodstuffs

The results of the AAS determination of the metals in the unprocessed foodstuff are recorded in Table 2. There is variation in the level of the metals in the unmilled foodstuffs.

Table 1. Metal Composition of Milling Plates

Element	GUK plate (mg/g)	PREMIER Plate (mg/g)
Iron	969.00 ±6.84	942.10 ±6.21
Tin	21.00 ±0.33	94.80 ±0.94
Chromium	3.13 ±0.24	14.40 ±0.20
Manganese	4.52 ±0.20	9.76 ±0.19
Lead	2.48 ±0.05	2.79 ±0.05
Copper	2.26 ±0.22	0.55 ±0.18
Nickel	1.16 ±0.26	1.85 ±0.30
Cobalt	<0.15 ±0.01	<0.05 ±0.04
Zirconium	0.08 ±0.01	<0.03 ±0.02
Niobium	0.06 ±0.01	0.02 ±0.01
Molybdenum	0.3 ±0.03	0.05 ±0.05
Germanium	ND	0.07 ±0.01

*ND = Not detected

Table 2. The Mean Concentration Levels of Metals in Unmilled, Dry Milled and Wet Milled Foodstuff (mg/kg)

			FOODSTUFFS			
			Maize	**Millet**	**Soghum**	**Cowpea**
Metals in Unmilled Foodstuff		Iron	13.10±0.09	12.30±0.13	5.95±0.17	8.81±0.19
		Nickel	5.54±0.07	4.34±0.06	3.89±0.09	4.55±0.14
		Lead	2.78±0.25	1.54±0.14	2.47±0.18	0.98±0.10
Iron in milled foodstuff	GUK	Dry milled	33.67±0.12	31.75±0.06	26.00±0.05	28.97±0.04
		Wet milled	28.54±0.07	28.12±0.08	23.80±0.05	31.22±0.05
	PREMIER	Dry milled	31.22±0.04	23.32±0.05	20.18±0.04	22.82±0.06
		Wet milled	27.70±0.06	25.30±0.05	18.84±0.10	24.57±0.06
Nickel in milled foodstuff	GUK	Dry milled	7.29±0.08	4.98±0.07	4.28±0.14	4.66±0.18
		Wet milled	6.93±0.02	4.76±0.08	4.06±0.11	4.87±0.10
	PREMIER	Dry milled	7.43±0.06	5.55±0.04	4.68±0.12	5.33±0.09
		Wet milled	6.91±0.31	5.00±0.11	4.38±0.13	4.97±0.10
Lead in milled foodstuff	GUK	Dry milled	3.47±0.30	2.79±0.03	2.78±0.05	2.06±0.03
		Wet milled	3.09±0.08	2.25±0.02	2.67±0.05	2.90±0.03
	PREMIER	Dry milled	7.70±0.04	4.64±0.03	4.54±0.04	2.97±0.04
		Wet milled	4.58±0.05	3.62±0.05	3.55±0.03	3.42±0.03

In the unprocessed foodstuffs, the mean concentration of iron metal is highest in maize (13.10 mg/kg, closely followed by that in millet grain with a concentration of 12.30 mg/kg. The least concentration of iron metal was in sorghum (5.95 mg/kg). The level of iron in the foodstuffs was found to be relatively high compared to other metals determined. Wheat grains have been reported by Schroeder [39-41] to contain as much as 40 mg/kg iron. The iron content of the major cereal grains in Nigeria has been reported in mg/100g dry weight as 5.30 for maize, 12.70 (soghum), 9.00 (millet), 24.30(rice), 3.60 (wheat) and 13.36 (acha) [42-46]. All these levels are within the Acceptable Dietary Intake.

The concentration of nickel is lower than iron in all the foodstuffs under investigation. The concentration of nickel was found to be least in sorghum (3.89 mg/kg) and highest in dry maize grains. This result agreed with the findings reported by Lisk [47] and others [48-51], Lisk reported the level of nickel in foodstuffs as within the range of 0.00-6.40 mg/kg in cereals and 0.00-0.34 mg/kg in fruits.

The AAS determinations of the concentration of lead in the unmilled foodstuffs are shown in Table 2. The mean concentration of lead is highest in maize (2.78 mg/kg) with its lowest value recorded in cowpea (0.98 mg/kg). Dry unprocessed millet grain gave a mean concentration of lead as 1.54 mg/kg. Values of 0.17 mg/kg in cereals have been reported by the Ministry of Agriculture, Fisheries and Food of the United States [52] and Lisk [47]. Osagie and Eka [42], however, did not detect lead in the major cereal grains grown in Nigeria except in acha (0.02 mg/kg). It is evident therefore that certain foodstuff can contribute more than others to the total lead intake in man's diet.

3.3. Level of Metal (Fe, Ni and Pb (mg/Kg)) Contamination in Milled Foodstuffs

Wet milling of the foodstuffs appears to have less iron metal contaminants than dry milling; the result of the analysis is as displayed in Table 2. With both GUK and PREMIER milling plates, dry milled foodstuffs recorded higher level of iron contamination (except in millet and cowpea). The highest iron metal contamination was determined in dry milled maize while the least (11.02 mg/kg) was in dry milled millet. In wet milled foodstuffs, cowpea recorded the highest level of iron contamination of 22.41 mg/kg while others are within the average of 15.00 mg/kg.

Table 2 shows the level of nickel metal in the milled foodstuffs determinations. Nickel has already been indicted in the unprocessed foodstuffs. It further contaminated foodstuffs during milling, whether it is wet or dry milling. Dry milled maize with PREMIER plate released 1.89 mg/kg nickel metal while wet milling with GUK plate released 1.39 mg/kg nickel metal. For the other milled foodstuffs the level of nickel released into them was low, for instance only 0.17 mg/kg nickel metal was determined in sorghum after wet milling and 0.11 mg/kg in dry milled cowpea, from GUK plate. This correlates with the low values of the metal in the milling plates. The mean concentration of nickel metal released is high in mills from PREMIER plate.

The level of lead metal contamination of milled foodstuffs as determined by the AAS (Table 2) ranged from 0.20 mg/kg in wet milled sorghum using GUK plate to 7.70 mg/kg in dry milled maize with PREMIER plate. In this determination, the highest level of lead metal contamination from GUK is recorded in wet milled cowpea with a concentration of 1.92 mg/kg. PREMIER milled foodstuffs recorded its highest lead concentration as 3.10 mg/kg in dry milled millet and lowest (0.20 mg/kg) in wet milled sorghum.

3.4. Comparison of Results from the Milling Methods

The metals released into the dry milled foodstuffs with GUK plates falls between the mean concentration of 0.11 mg/kg Ni in cowpea to 20.56 mg/kg Fe in maize. Dry millings with PREMIER plates introduced more lead into the foodstuffs (2.97 mg/kg in cowpea to 4.92 mg/kg in maize). Wet milled foodstuffs with GUK plates recorded a mean concentration of the metals under investigation between 0.17 mg/kg nickel metal in millet to 22.41 mg/kg iron in cowpea. With PREMIER plates, wet milling released metals ranging from 0.42 mg/kg nickel in cowpea to 15.76 mg/kg iron in cowpea; these values are higher than those recorded in GUK mills.

The contamination of foodstuffs by these metals in wet mills are lower than in dry mills, it can therefore be inferred that wet milling is “safer” than dry milling in terms of metal contamination.

3.5. Statistical Comparison of Results from the Milling Methods

A statistical comparison of the mean concentration of the metals in the control against each of the milling methods were performed using the student t-test for the pair data experiment at 95% confidence limit ($_t$0.05). All pair data of the milled techniques at F=4 degrees of freedom indicated a significant difference in the level of metal contamination of foodstuffs. The level of metal contamination of milled foodstuffs depends on the method of milling.

The levels of lead and nickel metals released into foodstuffs milled with PREMIER plates are generally higher than those from GUK plates. GUK is leading in food contamination with iron metal. This is because the PREMIER plate has higher lead and nickel content than the GUK plate (Table 1), and will tend to contaminate the foodstuffs more with these metals during milling. A student t-test for the level of metal contamination by the two milling plates indicated a high level of significant difference in the release of metals into the milled foodstuffs.

4. Conclusions

Milling plates, PREMIER and GUK, commonly employed in the processing of most Nigeria cereals grain foodstuffs consist of a variety of metals. The findings revealed that milling foodstuffs using the milling plates released lead, nickel and iron metal into the foodstuffs. Dry milling was found to introduce higher level of iron, lead and nickel contamination into foodstuffs than wet milling; thus making wet milling a “safer” method than dry milling. The level of metal contamination of foodstuffs is a function of the composition of the metal in the milling plate; hence, mills from PREMIER were highly contaminated with lead than mills from GUK. The level of metals in the foodstuffs commonly consumed falls within the range of the Average Daily Intake [53-56]. However, one cannot totally dismiss its possible potential threat to the health of man as a result of cumulative effect.

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