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Assessment of Cr, Cu, Fe, Ni and As Contamination/Pollution Load Indices in Milled Millet: Evaluation of the Impact of Grinding Plates in Agro-Food Milling Processes

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Abstract— The study sought to investigate the contribution of grinding plates to heavy metal levels in milled millet within the Wa Municipality of Ghana. The levels of selected heavy metals including Cr, Cu, Fe, Ni and As in milled millet were determined using the Atomic Absorption Spectrophotometer model AA 220. The levels of these elements in the milled millet samples exceeded the levels found in the control sample indicative that grinding plates used in milling millet samples potentially contributed to the higher levels of these metals recorded. The abundance of these elements followed the order- Fe > Cu > Cr > Ni > As across the various milled millet samples tested. Contamination indices of 1.46 - 2.32, 1.14 - 4.21, 1.03 - 34.17, 1.16 - 2.12 and 1.00 - 9.00 were recorded for Cr, Cu, Fe, Ni and As respectively. Recorded contamination indices essentially reflect varying degrees of pollution rather than contamination. Pollution load indices across milled millet samples on the other hand ranged from 5.13 - 1017.74. Pollution load indices attained in the current study suggest very highly polluted milled millet samples. These indices invariably suggest that milled samples in general were contaminated/polluted with the aforementioned heavy metals and potentially threaten the health of consumers. The present findings corroborate the findings of several other studies on the impact of grinding plates in the milling of agro-foods such as maize, cassava among others.

Keywords— Agro-food Grinding/Milling Machine; Grinding Plates; Heavy Metal Contamination; Millet Flour; Cast Iron; Wa Municipality.

I. INTRODUCTION

Food grinding/milling (corn mill) machine is a unit operation designed for the pulverization and or processing of foods including cereals, spices, nuts, tubers, etc. into powdered form or paste. It is an indispensable tool in food processing in Ghana and many other developing nations [1, 2]. Milling machines are found in cities, towns and villages across Ghana due to the vast dependence of the country folks on cereal food products. Maize milling for instance is a prime activity in Ghana as over 95 % of Ghanaians enjoy delicacies prepared from milled maize including *banku*, *kenkey*, *akple*, *aprenprensa* and maize porridge [2]. In a typical milling machine, grinding occurs by means of a pair of (circular) metal plates, often made of cast iron, one stationary and the other rotating. As the grain passes between the plates, they are crushed to powder [1].

Both locally made and foreign made grinding plates can be found on the Ghanaian market. The only two foreign made plates available are the Nigerian and the Indian made which come with brand names like radget, amuda, rex, premier, bin, England and bamford [2]. According to Kwofie et al. [1], unpublished market survey suggests that locally manufactured grinding plates dominate the Ghanaian market and are the most patronized. High patronage has been primarily due to its availability and relatively cheaper cost. Cast iron used in the manufacture of grinding plates are often mixed or alloyed with metals such as nickel (Ni), chromium (Cr), copper (Cu), silicon (Si) and other elements to enhance physical properties such as tensile strength [3].

In Ghana, the production of grinding plates is by small scale artisans. They do this by sand-casting using old vehicle engine blocks and other scrap metals and castings [4]. These locally-manufactured plates as emphasized by Kwofie et al. [1], wear out 3 to10 times faster than their foreign counterparts, lasting a maximum of four (4) months. The wearing of these plates in service invariably introduces metal debris into milled food products which could present health hazards to consumers.

II. RELATED WORK

Kwofie and Chandler [4] studied the potential health effects of locally manufactured corn-mill grinding plates

and reported varied hardness and wear resistances for the different grinding plates in relation to their microstructures. Kwofie et al. [1] in a follow up study on the quality of locally manufactured corn-mill grinding plates reported that iron particles from milling plates contaminated milled maize whether milled wet or dry.

Adeti [2] similarly in his study compared the rate of wear of a Ghana made, Nigerian made and an Indian made grinding plate. Maize samples milled using the aforementioned plates were found to contain varying levels of heavy metal contaminants. The Ghana made plate imparted the highest levels of heavy metal contaminants followed by the Nigerian plate with the Indian plate contributing the least levels of metal contaminants. The study also reported decreasing heavy metal levels with time for all the milling plates studied.

On the premise of the above findings, wearing of grinding plates during milling processes is inevitable and compromises food safety [1, 4]. The gravity of any possible health issue resulting from the use of locally made grinding plates is heightened by the fact that a greater number of Ghanaians depend extensively on milled cereals and grains for the preparation of various kinds of delicacies. The situation may even be more prevalent in the northern part of Ghana such as Wa as indigenes within this part of Ghana are well noted for their extensive dependence on milled cereals and grains for the preparation of their chief foods including millet porridge, maize porridge and *tuo-zaafi*. The present study was conducted at the Wa Polytechnic with the Wa Municipality as the study area. The study determined the levels of selected heavy metal contaminants in milled millet within the municipality to drum home the problem of heavy metal contamination resulting from the use of locally made milling/grinding plates and its prevalence within the municipality. The study is expected to inform policy direction on the need for locally made milling/grinding plates to conform to recommended standards.

III. MATERIALS AND METHODS

Study Area

The study was conducted in Wa, the Regional capital of Upper West Region of Ghana. The Wa Municipality shares administrative boundaries with, Nadowli District to the North, Wa East District to the East and South and Wa West District to the West and South. It lies within latitudes $1^{o}40$'N to $2^{o}45$'N and longitudes $9^{o}32$'W to $10^{o}20$'W. It has an estimated landmass area of 234.74 square kilometres, representing approximately 6.4 % of the region [5]. The Wa Municipality according to the Ghana Statistical Service [6] has a total population of about 107,214. The vegetation and the climatic conditions found in Wa are very suitable for livestock production owing to the abundance of pasture and non-availability of tsetse flies [7]. The inhabitants of Wa are chiefly crops and livestock farmers as well as traders [5, 8].

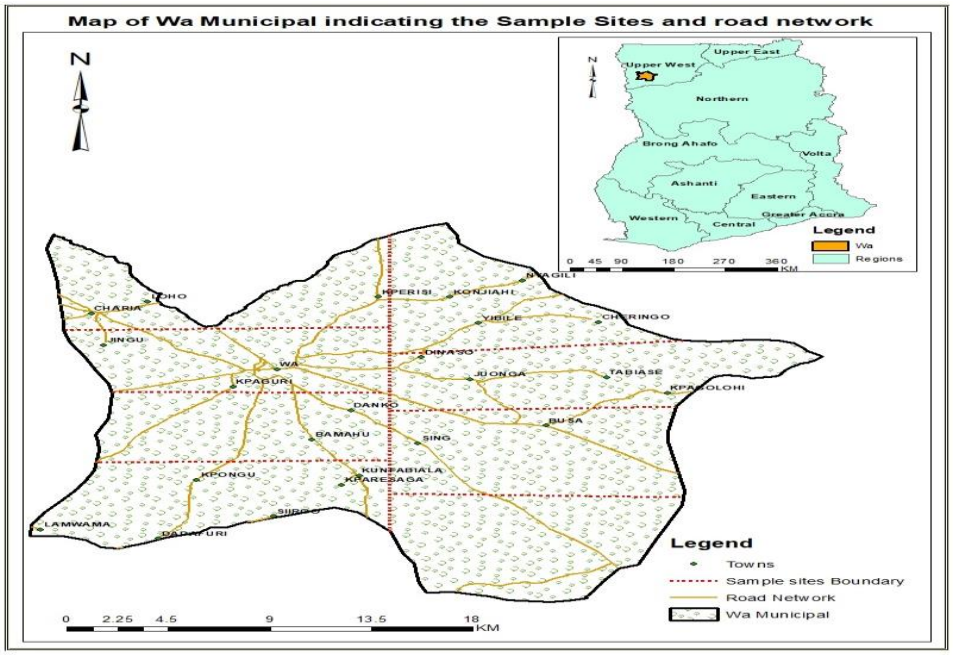

Figure 1: Wa Municipal Map [Source: Mensah *et al*. [5]]

Sample Preparation

A 25 kg bag of dried millet was purchased from the Wa Municipal market for the study. The dried millet grains were soaked in water (tap water) in a suitable plastic container and left to stand for 24 hours. The water was poured off after 24 hours of soaking.

A total of twelve (12) milling plants were patronized for milling of the wet millet grains. Milling plants patronized were scattered across eight (8) townships located within the Wa Municipality. The townships included the Wa Township (Wa T), Kpaguri (KP), Bamahu (BH), Kpongu

(KG), Konta (KT), Danko (DK), Konfabiela (KF) and Kparesaga (KS). For each of the townships- Wa, Kpaguri, Bamahu and Kpongu, two milling plants were accessed whereas for the townships Konta, Danko, Konfabiela and Kparesaga, a milling plant each was accessed. About 2.0 kg of wet millet was milled at each milling plant. Milled samples were transported to the laboratory for subsequent treatment (digestion). About a handful of the wet millet grains was pounded to paste/powder using porcelain mortar and pestle to serve as the control (CT) for the study.

Sample Digestion

Control and test samples were digested using nitric-perchloric acid combination [2, 8, 9, 10]. For each milled/powdered sample, 0.5 g of it was weighed into a 200 mL beaker. 10 mL of a binary acid mixture of HNO_3 and $HClO_4$ in the ratio of 9:4 was added to the sample and the content mixed thoroughly by swirling. The mixture was heated on a hot plate between 85 - 150 ^oC until the evolution of NO_2 fumes ceased. Heating continued until the volume was about 3 - 4 mL and appeared yellowish but not dried. The digested sample was allowed to cool to room temperature. The sides of the beaker were washed with de-ionized distilled water and the solution filtered into a 50 mL volumetric flask using a glass funnel and a Whatman 1 acid-washed filter paper. The sample in the volumetric flask was topped up with de-ionized distilled water to make up the volume (50 mL). The digest was then transferred into a clean sample bottle and stored at 4 ^oC for subsequent elemental analysis using the Atomic Absorption Spectrophotometer (AAS) model AA 220. Samples were analyzed at the Environmental Quality Laboratory of Anglo Gold Ashanti- Obuasi.

Preparation of Standards

Commercial stock solutions of chromium (Cr), copper (Cu), iron (Fe), nickel (Ni) and arsenic (As) were employed in preparing the respective standard solutions that were used in calibrating the AAS machine in each instance. A series/range of concentrations was prepared in each instance for the calibration of the AAS [5].

Analysis of Samples

A blank was initially run through the AAS machine followed by a series of calibration (standard) solutions and their respective responses measured to obtain a calibration graph. The AAS was adjusted to read zero (0) for the blank solution. For each element, a calibration graph was plotted after which the test samples were atomized and their responses measured. The level of each element in each digest was determined in triplicate. Metal concentrations were obtained from the calibration curve in relation to the absorbance obtained for the test solutions [5].

Analysis of Data

Concentrations of elements measured were expressed as mean ± standard deviation (SD) using Minitab (17) statistical software. ANOVA was employed in comparing mean concentrations of elements among and between milled samples (milling plants) from the various townships. Minitab (17) statistical software was used in performing ANOVA computations. For each element, Dunnett simultaneous tests were performed between the control and each milled sample using Minitab (17) statistical software.

IV. RESULTS AND DISCUSSION

Cr, Cu, Fe, Ni and As concentrations measured for the different milled samples are presented in Table 1. The levels of all elements monitored across the various milled samples were higher than the levels present in the control. Table 2 shows the contamination index (CI) for each element assessed and the pollution load index (PLI) owing to the milling process. Table 3 shows the computed percentage increases in the levels of these elements (Cr, Cu, Fe, Ni, As) across the various milled samples.

CI was calculated as a function of the concentration of each element to the respective control concentration. Thus

$$CI = Ce/Cc \quad (1)$$

Where Ce= measured metal concentration and Cc= elemental concentration in control sample [12, 13].

The PLI gives a generalized assessment of the level of elemental contamination. PLI was calculated using the relation

$$PLI = [CI_1 \times CI_2 \times CI_3 \times \ldots \times CIn]^{1/n} \quad (2)$$

Where CI = contamination index and n = number of metals [11, 12].

Table 1: Cr, Cu, Fe, Ni and As concentrations for milled samples

Milled Sample	Metal (Mean concentrations in mg/kg dry weight ± SD)				
	Cr	Cu	Fe	Ni	As
Wa T_1	4.13 ± 0.13	6.89 ± 0.07	29.67 ± 0.42	1.58 ± 0.00	0.06 ± 0.01
Wa T_2	4.15 ± 0.15	9.06 ± 0.05	88.98 ± 0.95	1.50 ± 0.03	0.07 ± 0.02
KP_1	4.88 ± 0.25	5.82 ± 0.08	523.08 ± 7.72	1.53 ± 0.06	0.15 ± 0.01
KP_2	4.31 ± 0.01	6.35 ± 0.07	144.62 ± 4.03	1.62 ± 0.02	0.02 ± 0.00
BH_1	3.98 ± 0.18	8.20 ± 0.11	27.03 ± 1.22	1.47 ± 0.06	0.05 ± 0.01
BH_2	3.92 ± 0.11	4.70 ± 0.16	34.10 ± 0.95	1.18 ± 0.01	0.03 ± 0.00
KG_1	5.21 ± 0.27	10.51 ± 0.45	234.22 ± 3.19	1.43 ± 0.01	0.08 ± 0.01
KG_2	3.70 ± 0.09	5.81 ± 0.18	15.76 ± 0.06	1.22 ± 0.11	0.04 ± 0.00

KT	4.25 ± 0.28	7.23 ± 0.07	41.15 ± 0.17	1.55 ± 0.03	0.06 ± 0.01
DK	4.77 ± 0.21	5.02 ± 0.03	47.28 ± 0.51	1.50 ± 0.11	0.09 ± 0.01
KF	3.29 ± 0.15	17.32 ± 1.16	30.41 ± 0.37	1.82 ± 0.05	0.02 ± 0.00
KS	3.95 ± 0.22	6.48 ± 0.06	28.42 ± 0.30	2.16 ± 0.14	0.04 ± 0.01
CT	2.25 ± 0.14	4.11 ± 0.08	15.31 ± 0.10	1.02 ± 0.02	0.01 ± 0.00

Table 2: CI and PLI for milled samples

Milled Sample	CI					PLI
	Cr	Cu	Fe	Ni	As	
Wa T_1	1.84	1.68	1.94	1.55	6.00	242.01
Wa T_2	1.84	2.20	5.81	1.47	7.00	631.75
KP_1	2.17	1.42	34.17	1.50	4.00	89.43
KP_2	1.92	1.55	9.45	1.59	2.00	25.71
BH_1	1.77	1.14	1.77	1.44	5.00	5.13
BH_2	1.74	1.14	2.23	1.16	1.00	1017.74
KG_1	2.32	2.56	15.30	1.40	8.00	11.43
KG_2	1.64	1.41	1.03	1.20	4.00	81.61
KT	1.89	1.76	2.69	1.52	6.00	105.73
DK	2.12	1.22	3.09	1.47	9.00	43.54
KF	1.46	4.21	1.99	1.78	2.00	43.86
KS	1.76	1.58	1.86	2.12	4.00	55.77

Table 3: Percentage increase in Cr, Cu, Fe, Ni and As concentrations across milled samples

Element/Metal	Mean percentage increase in concentration ± SD
Cr	46.5 ± 2.5 - 132.0 ± 5.9
Cu	14.5 ± 4.3 - 321.3 ± 26.4
Fe	3.0 ± 1.0 - 3317.6 ± 67.9
Ni	15.7 ± 1.3 - 111.3 ± 9.8
As	100.0 ± 0.0 - 1366.7 ± 115.5

Figures 2a, 3a, 4a, 5a and 6a respectively represent the concentrations of Cr, Cu, Fe, Ni and As across milled samples with figures 2b, 3b, 4b, 5b and 6b respectively representing the multiple comparison plots for each element. Multiple comparison tests for each element were performed between the various means for the different milled samples and the control mean.

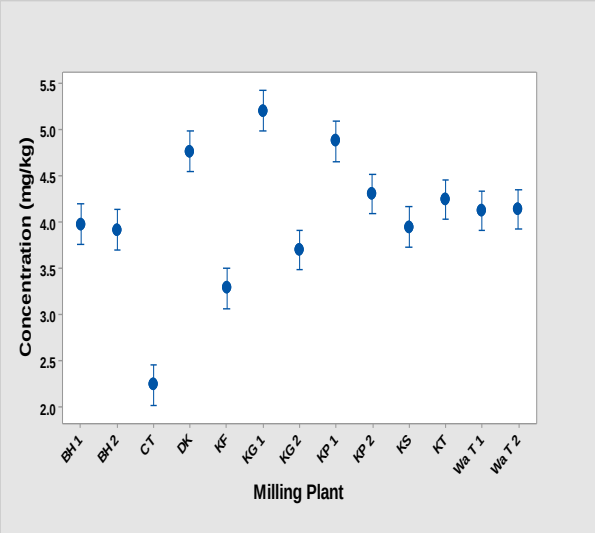

Figure 2a: Cr concentration for milled samples

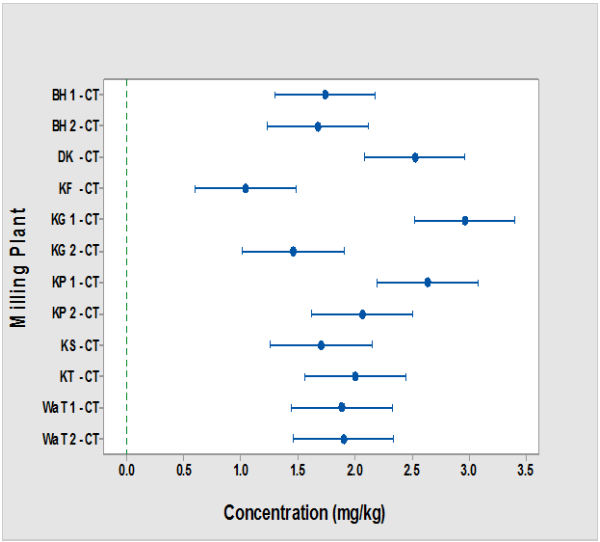

Figure 2b: Dunnett simultaneous tests for Cr

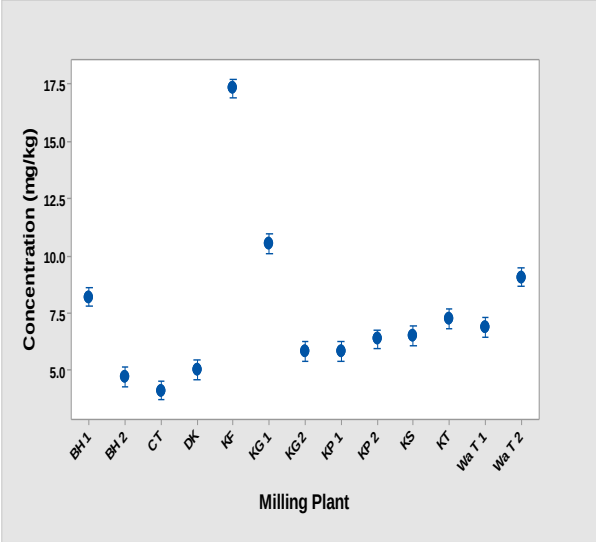

Figure 3a: Cu concentration for milled samples

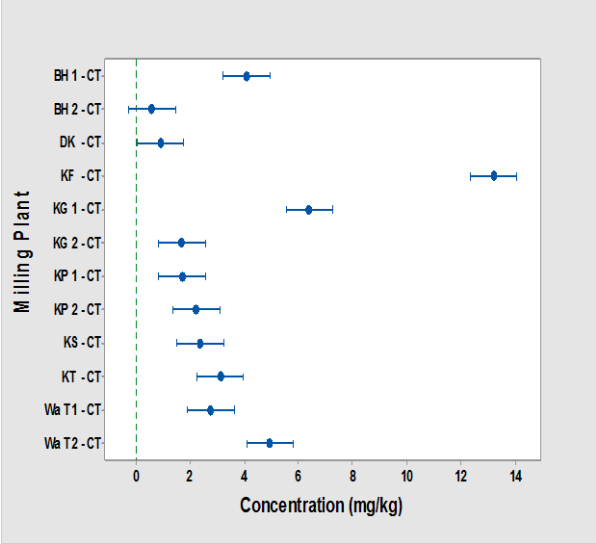

Figure 3b: Dunnett simultaneous tests for Cu

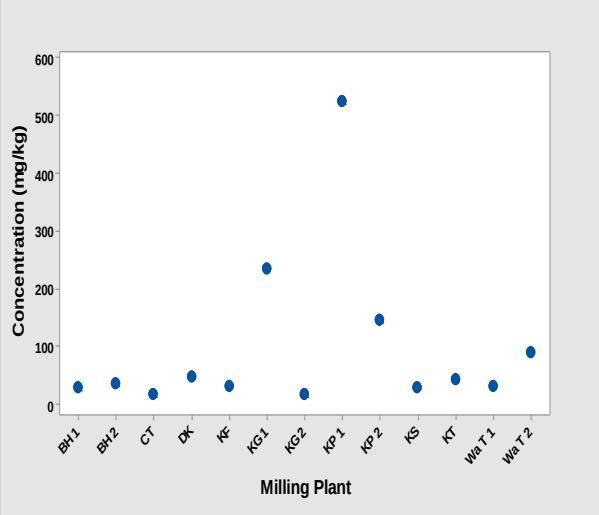

Figure 4a: Fe concentration for milled samples

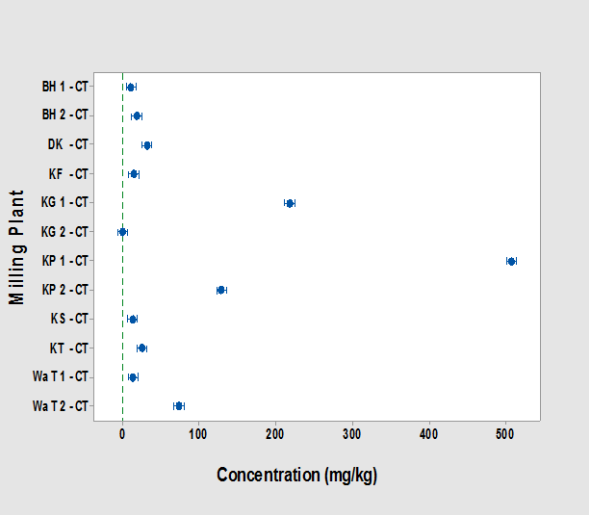

Figure 4b: Dunnett simultaneous tests for Fe

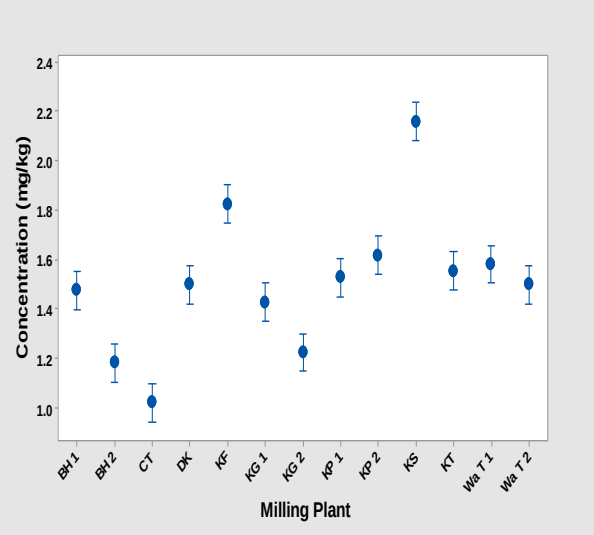

Figure 5a: Ni concentration for milled samples

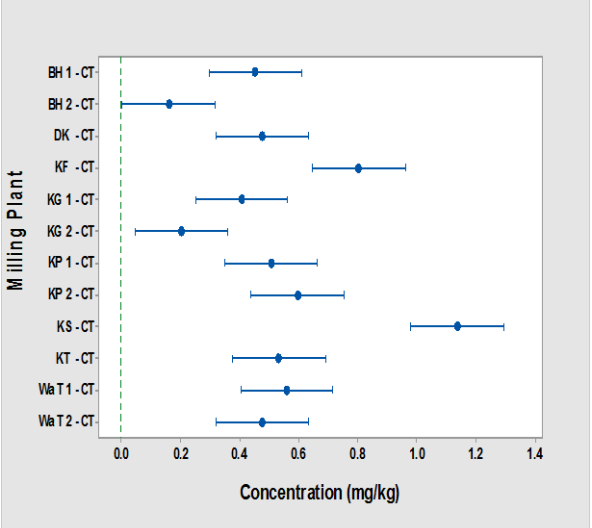

Figure 5b: Dunnett simultaneous tests for Ni

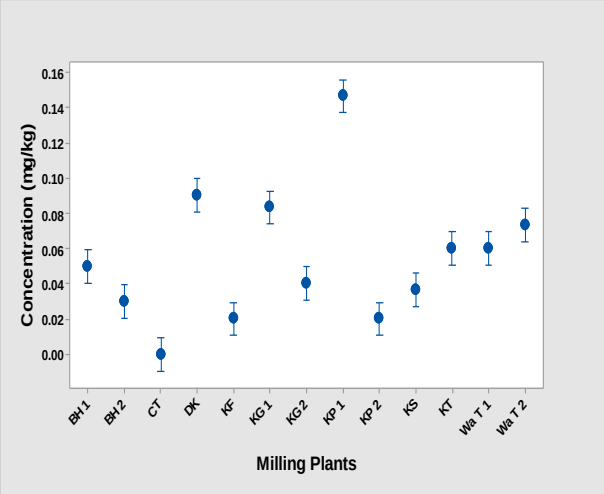

Figure 6a: As concentration for milled samples

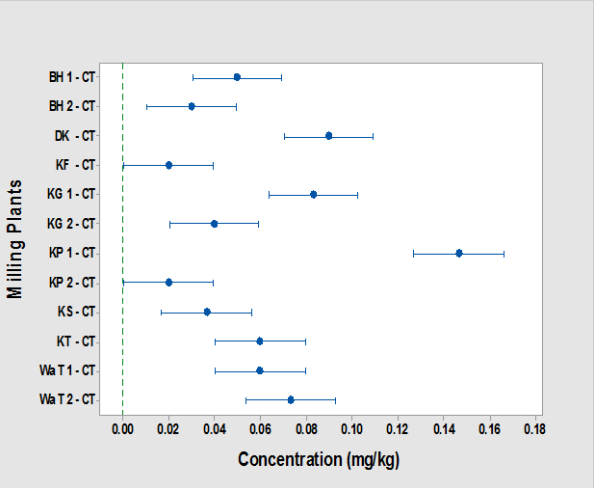

Figure 6b: Dunnett simultaneous tests for As

One way ANOVA results revealed significant differences ($p < 0.05$) in mean concentrations for all elements studied across all milled samples. For Cr, Ni and As, Dunnett multiple comparison tests revealed all means to be significantly different from the control mean ($p < 0.05$) (Figure 2b; Figure 5b; Figure 6b). However, Dunnett multiple comparison tests for Cu showed all means to be significantly different from the control mean ($p < 0.05$) except BH_2 (Figure 3b). In a similar manner, multiple comparison tests for Fe showed all means to be significantly different from the control mean ($p < 0.05$) except KG_2 (Figure 4b).

Mean Concentration and Percentage Increase

The practice of agro-food milling cut across length and breadth of Ghana taking into account the fact that a greater percentage of Ghanaians depend extensively on milled cereals and grains for various native delicacies. Metal contamination of milled agro-foods is essentially a function of the conjoint action of corrosion and friction/abrasive wear [1, 14]. The wearing of grinding plates during milling processes is inevitable and compromises food safety [1, 4]. Milling as the present study revealed, invariably introduced considerable levels of Cr, Cu, Fe, Ni and As into milled millet in comparison to the control. From the present study, milling essentially introduced 3.29 ± 0.15 - 5.21 ± 0.27 mg/kg of Cr, 4.70 ± 0.16 - 17.32 ± 1.16 mg/kg of Cu, 15.76 ± 0.06 - 523.08 ± 7.72 mg/kg of Fe, 1.18 ± 0.01 - 2.16 ± 0.14 mg/kg of Ni and 0.02 ± 0.00 - 0.15 ± 0.01 mg/kg of As into the milled millet (Table 1). Cr, Cu, Fe, Ni and As levels were respectively 46.5 ± 2.5 - 132.0 ± 5.9 %, 14.5 ± 4.3 - 321.3 ± 26.4 %, 3.0 ± 1.0 - 3317.6 ± 67.9 %, 15.7 ± 1.3 - 111.3 ± 9.8 %, 100.0 ± 0.0 - 1366.7 ± 115.5 (Table 3) higher than the levels found in the control. Cast iron, the chief material used in the manufacture of grinding plates is often mixed with other metals to enhance the physical properties [3] hence the presence of the abovementioned metals/elements were anticipated. In the case of As however, though it is not technically a "metal," that may be added on purpose to achieve any desirable properties/characteristics, this metalloid may potentially have come from old engine blocks commonly used for the casting as well through the use of other scrap metals and castings. The order of abundance of measured elements with respect to their concentrations was Fe > Cu > Cr > Ni > As across the milled samples. The levels of all elements in the control sample were well below their respective WHO permissible limits except for Cr whose concentration exceeded the recommended WHO permissible limit. Cr exists in nature as a chemical compound and may have found its way into the food crop (millet) via uptake from soil or water (used for irrigation) that possibly contained higher concentrations of Cr. It is also possible that water used in soaking the millet may have contained significant levels of this metal thereby contributing appreciably to the high levels of Cr in the control sample. Fe levels recorded in the present study were in agreement with the findings of Kwofie et al. [1] and Adeti [2]. Kwofie et al. [1] in their study found the average concentration of Fe in wet milled maize to be 50.00 mg/kg. Adeti [2] similarly recorded Fe levels of 61.24 - 175.34 mg/kg in his study. With reference to the present study, recorded Fe levels inevitably are attributable to cast iron- the chief component of grinding plates. Despite its good mechanical properties, cast iron is also susceptible to corrosion attack, a phenomenon that influences wearability of cast iron. The effect of corrosion on the wearability of cast iron has been estimated to be 10 - 90 % [15]. Adeti [2] in his study also recorded Cr, Cu, Ni and As levels of 0.96 - 1.51 mg/kg, 10.21 - 15.04 mg/kg, 10.43 - 23.23 mg/kg and < 0.01 mg/kg respectively. Whereas the levels of Cr and As recorded for the present study were comparatively higher, the levels of Ni recorded were lower than that reported by Adeti [2]. In the case of Cu, reported levels by Adeti [2] were consistent with the findings of the present study. The observed difference in Cr, Ni and As levels is possible taking into account the fact that the production of these grinding/milling plates by local artisans are not regulated to conform to any local or international standards. As such variations in the physical/mechanical and chemical properties of these locally made grinding plates are bound to occur to a greater

extent from one artisan to the other. These physico-chemical properties as it were, define the degree and rate of wear of these plates. Kwofie and Chandler [4] in their study were able to establish differences in physico-chemical properties of locally made grinding plates from same manufacturer.

The levels of Cr, Cu, Fe, Ni and As monitored across the milled samples were higher than that found in the control. This potentially constitutes a public health threat and raises toxicological concerns with reference to recorded levels that exceeded permissible thresholds. In respect of Fe, only KG_2 and the control had Fe levels well below the World Health Organization's safe limit of 20.00 mg/kg [16]. The remaining eleven (11) milled samples obviously raise toxicological concerns. Fe is an essential element in humans as it helps in oxygen transport and also regulates cell growth and differentiation [1, 17]. Fe deficiency would thus limit oxygen delivery to cells and cause fatigue, poor work performance and decreased immunity. In excess, Fe intake can cause toxicity and Fe overload as the human body can excrete very little Fe [1]. In the case of Cu, only two (2) (KG_1 and KF) (Table 1) out of the twelve (12) milled samples, recorded levels in excess of the WHO permissible threshold of 10.00 mg/kg [16] and potentially pose a health threat. Cu, though essential in trace amounts is responsible for hyperactivity in autistic children and can cause anaemia, liver and kidney damage, as well as stomach and intestinal irritation in high doses [5, 18]. On the other hand, Cr levels recorded for all milled samples including the control exceeded the WHO permissible limit of 1.50 mg/kg [16]. Chromium comes in two forms-trivalent (Cr^{3+}) and hexavalent (Cr^{6+}). Trivalent Cr in trace amounts is vital to human health and can be found in dietry supplements. Hexavalent Cr is also of biological significance and is more readily absorbed across cell membranes. Hexavalent Cr is associated with allergic dermatitis in humans. Higher concentrations of Cr in milled agro-foods present a health risk on consumption. Excessive levels of Cr (especially the hexavalent) form can damage the liver, kidneys and nerves, and may as well cause cancer and irregular heart rhythm [2]. In comparison to the WHO permissible threshold of 1.50 mg/kg, five (5) out of the twelve (12) milled samples in addition to the control had Ni levels below or equal to the WHO permissible limit posing no health threat. The remaining seven (7) milled samples potentially present harmful health effects, such as chronic bronchitis, reduced lung function, cancer of the lung and nasal sinus [2, 19]. In respect of As, all milled samples recorded levels below the WHO safe limit of 1.00 mg/kg. As levels recorded essentially presents no adverse effects health wise. That notwithstanding, as a known carcinogen, As can cause cancer of the skin, lungs, liver and bladder over prolonged exposure. Low level exposure can cause decreased production of red and white blood cells, abnormal heart rhythm, damage to blood vessels etc. Ingestion of very high levels can possibly result in death [5].

Contamination Indices and Pollution Load Indices

The baseline value of CI is 1.0. Values below 1.0 show varying levels of contamination and defines the contamination range whereas values above 1.0 indicate varying levels of pollution and defines the pollution range. CI essentially depicts the levels of individual metal contaminants in relation to levels in the control/background. PLI reflects the combined elemental contamination. CI and PLI levels attained in the present study were respectively compared to that of Lacutusu [13] and Thomilson et al. [11]. CI ranges of 1.46 - 2.32, 1.14 - 4.21, 1.03 - 34.17, 1.16 - 2.12 and 1.00 - 9.00 were recorded for Cr, Cu, Fe, Ni and As respectively. Recorded CI values essentially reflect varying degrees of pollution rather than contamination. Compared to the CI ranges as interpreted by Lacutusu [12], the degree of Cr and Ni pollution was slight to moderate, that of Cu was slight to severe with that of Fe being slight to excessive. That of As depicted slight to very severe pollution. PLI across milled samples ranged from 5.13 - 1017.74 far above the range of 0 - 5 put forth by Thomilson et al. [11]. PLI values attained in the current study invariably reflect very highly polluted milled samples. These indices invariably suggest that milled samples in general were polluted with the aforementioned heavy metals and potentially threaten the health of consumers.

V. CONCLUSION

The wearing of grinding plates during milling processes is inevitable and compromises food safety. Cr, Cu, Fe, Ni and as levels monitored across milled samples in the present study were higher than that found in the control. The order of abundance of these elements with respect to their concentrations was Fe > Cu > Cr > Ni > As across milled samples. The presence of these elements is suggestive of the wearing of grinding plates during the milling process. Recorded CI and PLI values recorded essentially portray varying degrees of pollution that raises health concerns. The present study corroborates the findings of several other studies on the impact of grinding plates in the milling of agro-foods such as maize, cassava among others. The gravity of any possible health issue resulting from the use of locally made grinding plates is heightened by the fact that milling has become an indispensable tool in food processing in most Ghanaian homes. There is as such the need for the production of local milling/grinding plates to be regulated to conform to highest standards. The present study was limited in scope as it covered only a selected number of heavy metal contaminants. Future studies can be expanded in scope to capture a wider number of heavy metal contaminants to give a better perspective of the CI and PLI estimations.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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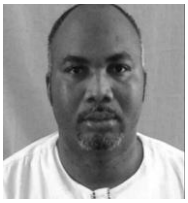

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