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**RESEARCH ARTICLE**

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# Selected Heavy Metal Levels in Cereals in Fluorspar Mining Belt, Elgeyo Marakwet County in Kenya

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**Abstract**

*Minerals in the earth crust get exposed to the environment through mining. Food crops grown on the contaminated soils bio-accumulate the metals posing adverse effects to the consumers. The study was aimed at investigating the concentrations of Arsenic (As), Cadmium (Cd), Chromium (Cr) and Lead (Pb) in cereals (maize, millet and beans) collected from three zones in Fluorspar mining belt in Kimwarer sub-catchment. A total of 60 food samples were collected from the households and analyzed using Flame Atomic Absorption Spectrophotometer (AAS) for Cd, Cr and Pb while As was analyzed using Atomic Absorption Spectrophotometer coupled with Vapour Generating component. Data was analyzed using ANOVA and Students New Kaules (SNK). The levels of Arsenic, Cadmium and Lead varied significantly ($P<0.05$, $\alpha=0.05$) in millet between Upper and Lower Kimwarer and also between Middle and Lower Kimwarer. .The metal mean concentrations of Arsenic, Cadmium and Lead in food were above levels recommended by CODEX Alimentarius International Food Standards in all the zones. Ingestion of the selected heavy metals above the recommended levels poses health risks to the consumers living within the Fluorspar mining area. It's therefore recommended that the Ministry of Public Health should advise farmers to avoid planting and consuming food grown within the contaminated soils in Kimwarer mining belt.*

**Key Words**: Mining, Heavy Metals, Cereals

## INTRODUCTION

Heavy metals which occur in various forms and concentrations are toxicants that impact the environment negatively (Bystricka and Tomas, 2009). Human exposure to heavy metals through food items account for at least 90% of overall human exposure (Llobet *et al.*, 2008).The contamination of the living environment with potentially toxic heavy metals is considered as a very important health concern, which may result in accumulation of the elements in many food items (Arianejad *et al.*, 2015). Heavy metals such as As, Cd, Cr, and Pb have been considered as the most toxic elements in the environment and are included in the list of priority pollutants declared by US Environment Protection Agency (USEPA) (Cameron, 1992). Therefore, knowing the metal concentrations in food crops that are grown where the soil is contaminated with heavy metals is crucial for risk prevention (Islam *et al*., 2018).

The greatest threat to public health from Arsenic originates from contaminated groundwater. Drinking-water, crops irrigated with contaminated ground water and food prepared with contaminated water are the sources of exposure. Cereals can also be dietary sources of Arsenic, although exposure from these foods is generally much lower compared to exposure through contaminated groundwater (Singh and Ghosh, 2012). Previous studies in Lake Victoria and River

Nyamasaria showed that Arsenic content in the maize and beans samples collected ranged from 5.21 to 7.03 µg/100 g. respectively which was within the safe WHO Arsenic standard of 0.14 mg/l limit (Makokha *et al*., 2012).

Food crops grown on Cadmium-containing soils or on soils naturally rich in Cadmium heavy metal constitute a major source of non-workplace exposure to Cadmium. The other work place exposure is from cigarette smoking (Satarug and Moore, 2004). Previous studies on cereal sampled from an open market in north-eastern China indicated substantially higher concentrations of Cadmium in beans (55.7 ng/g) than in cereals (maize and millet) (9.2 ng Cd/g which were above WHO standard of 0.03 ng/g(Zhang *et al*., 1998).Maize, millet and beans samples in North-eastern Chinawere collected from open markets in North-eastern China and analyzed for Pb the result of the Studies carried outshowed that the average Pb levels were 25.7ng/g, 54.3ng/g and 35.4 ng /g for foxtail millet and maize respectively. These values were above the recommended WHO standard of 0.01ng/g (Zhang *et al*., 1998).

The Codex Alimentarius Commission being a joint intergovernmental body of the Food and Agriculture Organization of the United Nations and WHO harmonizes international food standards to protect the health of consumers (FAO & WHO, 2003). The minimal risk levels (MRLs) which are the estimated daily intake of the individual metal per day through ingestion of food is an important source of information for risk assessment. The minimal risk levels depicts the daily human exposure to a hazardous substance that is likely to be without appreciable risk of adverse, non-cancer health effects over a specified duration of exposure. The minimal risk levels are determined from the amount of food ingested and the amount of the heavy metal contaminant in food. The information in this MRL serves as a screening tool to help public health professionals decide where to look more closely to evaluate possible risk of adverse health effects from human exposure (Farsalinos *et al*., 2015). Therefore, this study sets out to determine selected heavy metal levels in cereals in Fluorspar Mining Belt, Elgeyo Marakwet County in Kenya.

## MATERIALS AND METHODS

### Sample Collection and Preparation

The three common cereals maize, beans and millet were collected in the three zones in Fluorspar Mining belt, Kimwarer sub-catchment in Elgeyo Marakwet, County Kenya (Figure 1). These zones were selected basing on the distance from the mines and the topography of the area. The three zones included the Upper Kimwarer (UK) zone located in the highlands which is about 20-25km on the windward direction from the mining site. Middle Kimwarer (MK) is about 10-20 Kms from the mining area and forms the part of the escarpment. Lower zone (LK) is where the mining company is located. The villages which were sampled in Upper Kimwarer included Kibonge, Kewabsui, Hz, Tambul, and Kiparus. The villages in Middle Kimwarer were Turesia, Kowochi, Kapchi, Tarita and Kapchepno. Samples were collected from Muskut, Kabokbok, Kimwarer, Kewabmwen and Chepsirei in Lower Kimwarer.

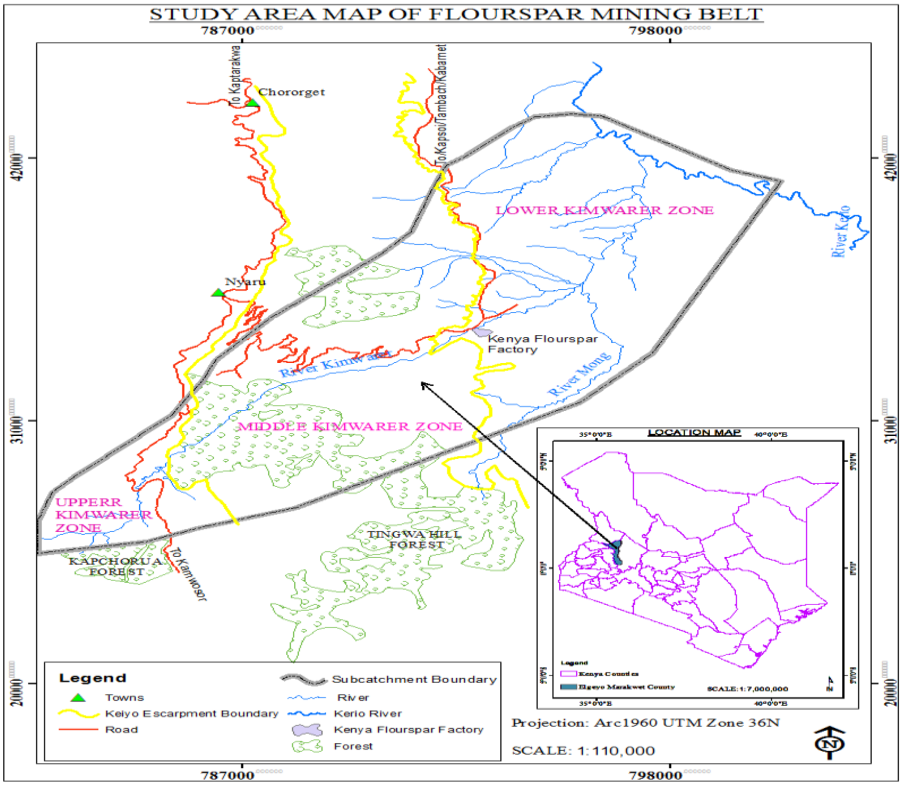


Figure 1. Study Area - Fluorspar Mining Belt in Elgeyo Marakwet 2018

**Sample Preparation and Analysis**

A total of 60 food samples were collected from the farms within the homesteads close to where the soil samples were collected and stored in polythene bags. Duplicate samples of each cereal namely maize, millet and beans were collected in the study period and taken to University of Eldoret Biochemical Laboratory for analysis. The digestion of cereals was done using a block Digester (Digestion System 20 1015 Digester. SNO. 3846, Sweden) following the stated procedure by (Okalebo et al., 2002). (Okalebo et al., 2002) 0.3±0.001g of finely ground homogenous oven dried (70$^0$C), ground cereal (<0.25mm, 60 mesh) into a labeled, dry and clean digestion tube. Digestion mixture 4.4 ml was added to each tube and also to 2 reagent blanks for each batch of samples. The mixture and sample was heated at 360$^0$C for 2 hours till the solution turned colorless and any remaining sand white. The solution was further heated for 1 hour to ensure all the colors had turned colorless. The contents were then allowed to cool and 25ml of deionized water was added and mixed well until no more sediment dissolved. It was then allowed to cool and topped up with distilled water to 50ml in a volumetric flask. The digests was allowed to settle, and then sieved using a 0.4 sieve into sample bottles for analysis. Analysis followed by aspirating Cadmium, Chromium and Lead digests and standards into Flame Atomic Absorption Spectrophotometer while Arsenic was analyzed using Atomic Absorption Spectrophotometer coupled with hydride vapor generator.

**Statistical Analysis**

The data collected was entered, organized, managed and analyzed using MS EXCEL spreadsheet. Statistical analysis of the data such as mean, standard deviation, one-way analysis of variance (ANOVA) was performed by SPSS software version 21.The data on the concentrations of the heavy metals; Arsenic, Cadmium, Chromium and Lead in cereals was calculated as a mean (±S.D) for each sampling site from each of the three zones sampled. One way ANOVA was used to determine whether there was any significant difference between the means of three independent variables (zones).The significant difference between the zones was determined by a post hoc test –Students Newman Kaules (SNK) Test.

## RESULTS AND DISCUSSIONS

### Concentration of Selected Heavy Metals in Maize

Maize (Zea mays L.) is an edible flowering plant in the gramineae family and is a warm-season crop that serves as the main food source for humans and animals around the world (Lu et al., 2015). Arsenicresults showed no significant variation between the Upper, Middle Kimwarer zones and Lower Kimwarer zone ($p<0.001$, 95%CL).Cadmium, Chromium and Lead concentrations in maize differed significantly between the Upper and Lower Kimwarerand also between Upper Kimwarer with Lower Kimwarer zone ($p<0.001$, 95%CL). The significant difference in Cd, Cr and Pb concentration in the different zones is a reflection of the concentration of the soils where the plants are drawing their nutrients.

Heavy metal levels in maize showed that the lowest heavy metal concentration was Chromium 0.95 ±0.50µg/g in Upper Kimwarer and highest was Cadmium19.17±2.82µg/g both in Lower Kimwarer. Upper Kimwarer had the lowest levels in all the heavy metals except for Lead levels whose lowest concentration was in the Middle Kimwarer zone. Lower Kimwarer had the highest levels of the heavy metal except Arsenic which had highest levels in Middle Kimwarer. Hence the heavy metal concentration in descending order as per the zones was Lower Kimwarer>Middle Kimwarer>Upper Kimwarer. The order of concentrations from the highest in Upper Kimwarer was Pb>Cd>As>Cr, Middle Kimwarer was Cd>As>Cr>Pb and Lower Kimwarer was Cd>Cr>Pb>As. The overall order of heavy metal concentrations from the highest in all the zones was Cd>Cr>Pb>As. The order of heavy metal concentrations from the highest in Upper Kimwarer was Pb>Cd>As>Cr, Middle Kimwarer was Cd>As>Cr>Pb and Lower Kimwarer was Cd>Cr>Pb>As. Concentration of selected heavy metals in maize is shown in Table 1.

Table 1. Concentration of Selected Heavy Metals in Maize

| | UK (µg/g) | MK (µg/g) | LK (µg/g) | p-value | Reference (mg/kg) |
|---|---|---|---|---|---|
| As | 2.31 ±1.12 | 4.36±0.96 | 2.13±0.12 | 0.107 | 0.1 |
| Cd | 3.41 ±0.81$^{a}$ | 4.57±0.38$^{a}$ | 19.17±2.82$^{b}$ | <0.001 | 0.1 |
| Cr | 0.95 ±0.50$^{a}$ | 3.81±0.78 $^{a}$ | 15.23±1.63$^{b}$ | <0.001 | Not available |
| Pb | 3.71±0.88$^{a}$ | 2.31±0.82$^{a}$ | 10.83±0.79$^{b}$ | <0.001 | 0.2 |

Mean values followed by the same small letter within the same row do not differ Significantly from one another (one-way ANOVA, SNK, $\alpha=0.05$)

The mean concentrations of As, Cd and Pb in maize in Upper, Middle and Lower Kimwarer were above recommended level of As and Cd by CODEX Alimentarius Commission. The CODEX Alimentarius International Food Standards levels for maize are 0.1mg/kg, 0.1mg/kg and 0.2 mg/kg for As, Cd and Pb respectively (FAO and WHO, 2015). Previous studies in China showed concentration of Cr and Pb in maize with mean values of 0.23 and 0.49 mg/kg respectively. However, Cr and Pb concentrations were generally higher in the current study. The same study showed that maize is a potential accumulator plant, which can be used to decrease the pollution level by harvesting, disposing of, and recovering the plant material. A similar study done by (Akenga *et al.* 2017) on heavy metals uptake in maize grains in Uasin Gishu County indicated elevated levels of Cd in maize grains which were above the WHO standard.

**Concentration of Selected Heavy Metals in Beans**

Beans are one of the common foods that are being consumed by residents in the study area. The results showed no significant variation in Arsenic, Cadmium and Lead concentrations in beans between Upper and Lower Kimwarer and also between Middle and Lower Kimwarer (p >0.05, One way ANOVA, SNK). However, Chromium concentrations in beans varied significantly between Upper and Lower Kimwarer and also between Middle and Lower Kimwarer (p=0.033, One way ANOVA, SNK).

Heavy metal levels in beans showed that the lowest heavy metal concentration was Chromium 1.28±0.64 µg/g in Middle Kimwarer and highest was Lead 17.24±2.30 µg/g both in Lower Kimwarer. Upper Kimwarer had the lowest levels in all the heavy metals except for Cadmium levels whose lowest concentration was in the Middle Kimwarer zone. Lower Kimwarer had the highest levels in all heavy metals in the study area. Therefore, Lower Kimwarer had the highest level of heavy metals followed by Middle Kimwarer then Upper Kimwarer with the least levels. The order of heavy metal concentrations from the highest in Upper Kimwarer was Pb>Cd>As>Cr, Middle Kimwarer was Cd>As>Pb>Cr and Lower Kimwarer was Pb>Cr>Cd>As. The overall order of heavy metal concentrations from the highest in all the zones was Pb>Cd>Cr>As. Concentration of selected heavy metals in beans is shown in Table 2.

Table 2. Concentration of Selected Heavy Metals in Beans

| | UK (µg/g) | MK (µg/g) | LK (µg/g) | p-value | Reference (mg/kg) |
|---|---|---|---|---|---|
| As | 2.56±1.23 | 4.39±0.70 | 9.09±2.78 | 0.122 | 0.1 |
| Cd | 2.98±1.07 | 5.66±1.52 | 12.33±8.45 | 0.561 | 0.1 |
| Cr | 2.44±1.37[ab] | 1.28±0.64[a] | 14.10±4.81[b] | 0.033 | Not available |
| Pb | 3.33±1.07 | 4.34±0.76 | 17.24±2.30 | 0.205 | 0.1 |

Mean values followed by the same small letter within the same row do not differ Significantly from one another (one-way ANOVA, SNK, α=0.05)

The lowest level of heavy metal concentration in beans was Chromium (1.28±0.64µg/g) in Upper Kimwarer and highest level was Lead (17.24±2.30 µg/g) in Lower Kimwarer. The concentrations of As, Cd and Pb in beans in the Upper, Middle and Lower Kimwarer were above the recommended levels by CODEX Alimentarius International Food Standards levels (FAO and WHO, 2003). This implies that the beans are contaminated with Arsenic, Cadmium, Chromium and Lead in Upper, Middle and Lower Kimwarer. Therefore, consumption of beans in the study area is a health risk to the people.

**Concentration of Selected Heavy Metals in Millet**

The results showed that the levels of Arsenic, Cadmium, Chromium and Lead were significantly higher in millet between Upper and Lower Kimwarer and also between Middle and Lower Kimwarer (p<0.05, One way ANOVA, SNK). The lowest heavy metal level in millet was Chromium 1.74±0.16 µg/g in Upper Kimwarer and highest was Cadmium 17.19±2.48 µg/g in Lower Kimwarer. Upper Kimwarer had the lowest levels in all the heavy metals except for Arsenic levels whose lowest concentration was in the Middle Kimwarer zone. Lower Kimwarer had the highest levels in all heavy metals in the study area. Hence the order of concentration in an ascending order per zones was; Upper Kimwarer followed by Middle Kimwarer the Lower Kimwarer. The order of heavy metal concentrations from the highest in Upper Kimwarer was

Pb>As>Cd>Cr, Middle Kimwarer was Pb>Cr>Cd>As and Lower Kimwarer was Cd>As>Pb>Cr overall order of concentrations from the highest in all the zones was Cd>Pb>Cr>As. Concentration of selected heavy metals in millet is shown in Table 3.

Table 3. Concentration of Chromium in Cereals Millet

| | UK (µg/g) | MK (µg/g) | LK (µg/g) | p-value | Reference (mg/kg) |
|---|---|---|---|---|---|
| As | 3.75±0.47[a] | 1.99±0.45[a] | 10.26±2.30[b] | 0.007 | 0.1 |
| Cd | 3.51±0.83[a] | 3.21±0.67[a] | 17.19±2.48[b] | <0.001 | 0.1 |
| Cr | 1.74±0.16[a] | 4.63±0.67[ab] | 10.20±2.77[b] | 0.035 | Not available |
| Pb | 5.58±1.51[a] | 5.02±0.58[a] | 10.21±0.42[b] | <0.001 | 0.2 |

Mean values followed by the same small letter within the same row do not differ significantly from one another (one-way ANOVA, SNK, T-test, α=0.05)

Results on heavy metal concentrations in millet were lowest in Chromium 1.74±0.16µg/g in Upper Kimwarer (1.92±0.44µg/g) and highest in Cadmium (17.19±2.48µg/g) in Lower Kimwarer. The mean concentrations of As, Cd and Pb were above the recommended CODEX Alimentarius International Food Standards of 0.1 mg/kg/g, 0.1mg/kg 0.2 mg/kg respectively in millet of Upper, Middle, and Lower Kimwarer. The CODEX Alimentarius International Food Standards levels for maize are 0.1mg/Kg, 0.1mg/Kg and 0.2 mg/kg for As, Cd and Pb respectively (FAO and WHO, 2015). The people within the study area are at risk of cancers and non-carcinogenic diseases caused by Arsenic, Cadmium, Chromium and Lead as they ingest contaminated millet.

In summary, cereals in the current study had the lowest concentration in Chromium (0.34±0.095µg/g) in Upper Kimwarer and highest was 19.17 µg/g) in Lower Kimwarer during both in maize. The highest levels for As, Cd, Cr and Pb in maize were, 4.36±0.96µg/g, 19.17±2.82µg/g, 15.23±1.63µg/g and 10.83±0.79 respectively. In beans the concentrations of As, Cd, Cr and Pb were 9.09±2.78µg/g, 12.33±8.45µg/g, 14.10±4.81µg/g and 17.24±2.30µg/g respectively. For millet, the levels of As, Cd, Cr and Pb were 10.26±2.30 µg/g, 17.19±2.48, 10.20±2.77 and 10.21±0.42 respectively. The order of concentrations from the highest for As was Millet>maize>beans, Cd: maize>millet>beans, Cr: maize>beans>millet and the overall order for all the metals was millet>maize >beans. The observed variation in metal concentrations for analyzed cereals might be due to variable capabilities of absorption and accumulation of metals by the cereal crops (Pandey & Pandey, 2009).

Maize, beans and millet results showed a spatial significant difference in As, Cd, Cr and Pb levels between the zones in the study area (P<0.05, 95%CL).All the levels in maize, beans and millet were above the CODEX Alimentarius International Standards of food. This shows that the people consuming these ccontaminated cereals are at risk of suffering from diseases which are associated with the selected heavy metals. Previous studies on Cadmium showed concentrations above the recommended CODEX level in cereals hence a potential hazard to the consumers (Sadovska, 2012). In Poland, Krelowska (1991) found lead levels of 0.07 µg/g in cereals, this result was lower than the results found in the study area. A study done by (Makokha *et al.*, 2012) in Kisumu revealed that Arsenic content in the maize and beans samples ranged between 5.21 to 7.03 µg/100 g. These values are lower than the results obtained in the current study.

## Correlation of Cereals and Soil and Water

### *Correlation of Cereals and Soil*

The heavy metal results in soil showed a positive correlation with maize, beans and millet. Cadmium and Lead in maize had a strong positive significant correlation(r = 0.725) with maize and a weak positive correlation with Lead (r = 0.379) (P < 0.01). Lead in beans and millet had a positive correlation with soil (p < 0.01) while soil had a weak positive correlation with Cadmium and Chromium in millet (p<0.05). The relationship of the concentrations of As, Cd, Cr and Pb in maize, beans millet and soil is shown in Table 4.

Table 4. The Correlation of Heavy Metals in Maize, Beans, Millet and Water

| | Heavy Metal Concentration in Soil (mg/kg) | | | |
|---|---|---|---|---|
| **Media** | **As** | **Cd** | **Cr** | **Pb** |
| Maize | -0.028 | 0.725** | 0.189 | .379** |
| Beans | 0.235 | 0.170 | 0.032 | .441** |
| Millet | 0.198 | 0.333* | .353* | .399** |

** Correlation is significant at the 0.01 level (2-tailed)
* Correlation is significant at the 0.05 level (2-tailed).

In the study area Pb in soil had a significant ($P < 0.01$) positive correlation with maize, beans and millet.

### *Correlation of Cereals and Water*

In the study area As in water had a significant positive correlation ($p < 0.05$) with beans and millet Cadmium in water had a weak significant positive correlation with maize and millet ($p < 0.05$). Chromium in water had a significant positive correlation with maize and millet, while Lead in water had a significant positive correlation with maize, beans and millet ($p<0.05$). The relationship of the concentrations of As, Cd, Cr and Pb in maize, beans, millet and water is shown in Table 4.

Table 4. The Correlation of Heavy Metals in Maize, Beans, Millet and Water

| | Heavy Metal Concentration in Water (mg/kg) | | | |
|---|---|---|---|---|
| **Media** | **As** | **Cd** | **Cr** | **Pb** |
| Maize | 0.117 | 0.138 | 0.457** | 0.348** |
| Beans | 0.497** | -0.028 | 0.026 | 0.727** |
| Millet | 0.449** | 0.631** | 0.645** | 0.643** |

** Correlation is significant at the 0.01 level (2-tailed)
* Correlation is significant at the 0.05 level (2-tailed).

The relation of the concentration between the media was strong in Cadmium (r = 0.631), Chromium (r = 0.645) and Lead in water and millet. Lead in beans also had a strong correlation with water (r = 0.727). Previous studies have established that cereal grain yields were correlated significantly and positively with the topsoil and subsoil cation exchange capacity and topsoil water content (Usowicz & Lipiec, 2017). Studies by (Zhao *et al*., 2015) showd a clear spatial correlation ranges for heavy metals in rice in the study area. This shows that the levels of heavy metal in the cereals are a reflection of the contamination in soil and water in the study area.

Maize, beans and millet are the most common stable foods consumed by the people living in Kimwarer sub-catchment. The current study has established that the cereals were contaminated with Arsenic, Cadmium, Chromium and Lead. The accumulation of all the studied metals is related to mining activities. Previous studies by (Jarup, 2003; Dorne, *et al*., 2011) confirm that humans are exposed to heavy metals as well as metalloids in the environment through ingesting contaminated foods like the studied metals. (Olero *et al*., 2018) found that the selected heavy metal concentrations were found in the soils and water in the study area. The same study also determined that there are potential non-carcinogenic and carcinogenic risks associated with Arsenic, Cadmium, Chromium and Lead in Fluorspar mining belt.

## CONCLUSIONS

1. The study established that there was a significant spatial variation in concentration of metals in the sampled media in all the zones. The levels of Arsenic, Cadmium and Lead varied significantly ($P<0.05$, $\alpha=0.05$) in millet between Upper and Lower Kimwarer and also between Middle and Lower Kimwarer respectively.

2. The metal mean concentrations in food were above the CODEX Alimentarius International Food Standards levels of As (0.1 µg/g), Cd (0.1 µg/g) and Pb (0.2 µg/g) in all the zones. The high heavy metal concentration levels in the selected metals puts people at risk in consuming maize, beans and millet in the study area.

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