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Heavy Metals Contamination Levels in the Vegetables Grown around Riruwai Mining Area, Kano State, Nigeria

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ABSTRACT

Vegetables grown in mining areas can accumulate significant amounts of heavy metals (HMs), which can cause serious developmental disorders and have long-term negative effects on public health. In the present study, the HMs contamination level in vegetables grown around the Riruwai mining area in Kano State, Nigeria, was investigated. Fifteen (15) vegetable samples were collected, including lettuce (*Lacuta sativa* L.), tomato (*Solanum lycopersicum* L.), and bean (*Phaseolus vulgaris* L.), as well as their corresponding soils. The levels of As, Cd, Cr, Hg, Mn, Ni, Pb, and Zn in all the samples were determined using Microwave Plasma Atomic Emission Spectrometry, and the measured concentrations were used to calculate the bioaccumulation factor (BAF). The results of the study revealed that HMs concentrations in the investigated vegetables were found to be significantly high, with the majority of levels exceeding the WHO/FAO (2007) recommended limit, and the concentration of HMs in the soil decreased in the order of Zn > Mn > Cr > Pb > As > Ni > Cd > Hg, with As, Pb, and Zn exceeding the WHO/FAO (2001) recommended limit. Pollution levels were found to significantly differ between HMs and vegetable types. BAF results revealed that cadmium is an accumulator of all the studied vegetables (BAFs > 1), while mercury was found to be an accumulator of L. sativa. Higher concentrations of these metals in vegetables and soil, particularly arsenic, cadmium, lead, and zinc, necessitate immediate scientific attention and further research to determine the optimum concentration required for human health. Planting of vegetables for human and animal consumption should be stopped until this is accomplished.

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Introduction

Heavy metals (HMs) contamination of soils and vegetables have remained a threat to public health around the world, particularly in developing countries (Bayissa and Gebeyehu, 2021). Vegetables grown in mining areas can accumulate significant amounts of HMs, which can cause serious developmental disorders and have long-term negative effects on public health (Cao et al., 2010; Gebeyehu and Bayissa, 2020; Guo et al., 2020). Mining operations have historically played a significant role in national and regional economies. However, mining operations are usually carried out uncontrollably, resulting in profound ecological problems, particularly HMs pollution (Zhuang et al. 2014). Short and long-term exposure to HMs could result in numerous human health issues, including dermal, cardiovascular, gastrointestinal, hematological, renal, and hepatic effects (Lin et al., 2013; Zhuang et al. 2014). HMs contamination of soils is a major pathway for HMs exposure to humans, as they can enter the human body through ingesting contaminated vegetables (Makanjuola et al., 2019). Vegetables are important edible crops that are a necessary segment of the human nutrition regimen. They are high in nutrients that are essential for good health and provide carbohydrates, vitamins, minerals, and fiber (Khan et al., 2015; Sevindik et al., 2017; Akgül et al., 2022). HMs are easily absorbed by vegetable root systems and can accumulate at high levels in the edible portions of vegetables even at low levels in the soil (Hu et al., 2013; Liu et al., 2013). It is clearly evident that more than 70% of the dietary intake of HMs is contributed to the food chain (Liu et al., 2013).

Riruwai, located in the southern part of Kano State, Northwestern Nigeria, is principally a mining community. A large-scale mining operation began in 1979 and was expanding rapidly with close to nine hundred (900) tons of zinc and tin ore production every day. The mine was stopped after five years of continued mining. However, small-scale mining has continued regularly in the area. There is growing evidence across the globe that HMs pollution in mining areas is causing health problems for the local residents (Li et al., 2014; Irzon et al., 2018). The residents of the mining communities are constantly at risk of being exposed to these metals through the consumption of vegetables. There is, therefore, a need to investigate the HMs contamination levels in the vegetables grown in the study area in order to protect the environment and public health. To the best of our knowledge, this is the first scientific study that investigated the HMs contamination levels in the vegetables cultivated on the farmland of the study area.

Materials and Methods

Description of the Study Area

Riruwai covers an area of 129 km^2 and is located between latitude 10°43"97"N - 10°45'01"N and longitude 8°43"3"E - 8°47'39" E (Fig. 1). The climate was classified as tropical savanna by KÖppen, with mean annual rainfall ranging from 400 to 1200 mm/year (Alhaji et al., 2017). Riruwai is one of Nigeria's younger granite complexes. The complex is an example of decayed roots of an alkaline volcano that formed as component of an early Jurassic chain of anorogenic centers. It is surrounded by a collection of metamorphic and calc-alkaline meta-igneous rocks that transitioned from Precambrian to Cambrian ages (Olasehinde et al., 2012). According to the 2006 census, the population of Riruwai is estimated to be 150,645 people. The projected population is 257479 people as at 2015 at 3.2 % growth rate (NPC, 2006). The occupation of majority of people of the community is farming. Riruwai is one of the promising historical sites in Kano State that is well-known for its beautiful natural and man-made sites (Yakubu et al., 2019).

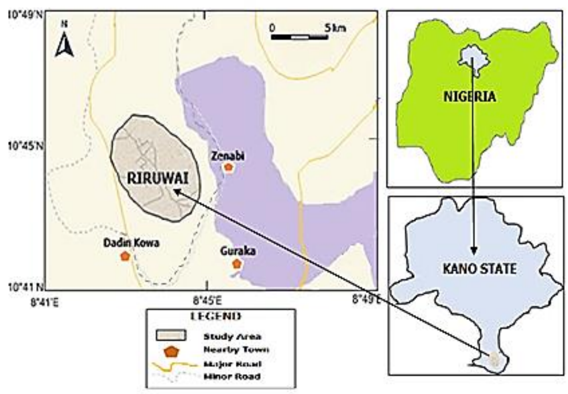

Figure 1. Map showing the study area

Vegetables Sampling and Pre-treatment

A total of fifteen (15) vegetables that represent the species commonly grown in the area were collected directly from the farms in the study area. The vegetables include lettuce (*Lacuta sativa* L.), tomato (*Solanum lycopersicum* L.) and beans (*Phaseolus vulgaris* L.). The specific sampling quantities were *L. sativa (4)*, *S. lycopersicum (6)*, and *P. vulgaris (5)*. The collected vegetable samples were washed three times with tap water to remove dust particles, blotted with tissue paper to dryness, and separated into edible and non-edible parts. The edible parts were oven-dried at 30 °C until they reached a consistent weight, then ground into powder with a blender (Jamali et al., 2009).

Determination of Heavy Metals in Vegetables

A 0.50 g sample of each treated vegetable was digested in a 5:1:1 tri-acid mixture of HNO_3, H_2SO_4, and $HClO_4$. The mixture was heated to 80°C until clear and then filtered into a 50 cm^3 volumetric flask using Whatman filter paper number 42. The filtrate was diluted with deionized water to mark. A blank sample was also prepared using similar acid mixtures into empty digestion vessels. Microwave Plasma Atomic Emission Spectrometer (MP-AES, Model: 4200) was used for the determination of HMs concentrations at their respective wavelengths in the soil and vegetable samples.

Determination of Heavy Metals in the Soils

The sample was air-dried at room temperature and visible materials such as stones, and plants residues were removed. The sample was pulverized, passed through a 0.15 mm nylon screen, sieved and homogenized. The soil sample was digested with a mixture of three concentrated acids (HNO_3, HF and $HClO_4$). Soil sample (0.50 g) was placed in a 200 cm^3 Teflon beaker. 10.00 cm^3 of HNO_3, 10.00 cm^3 of HF and 10.00 cm^3 of $HClO_4$ were added and the mixture was heated on an electric hot plate at 180 °C until the solution was almost dried. The beaker was removed and cooled to room temperature. 5.00 cm^3 of concentrated HNO_3, 5.00 cm^3 of HF and 5.00 cm^3 of $HClO_4$ were added and the mixture was boiled until dense white fumes appeared. The beaker was removed, cooled and 15.00 cm^3 of deionized water was added and boiled for 5 minutes. The digest was filtered using Whatman filter paper no. 42 into a 50 cm^3 volumetric flask. The wall of the beaker was washed three times with 10.00 % dilute HNO_3. The solution was made up to the mark with the HNO_3 for HMs analysis. Blank solution was treated and prepared in a similar way. The concentrations of As, Cd, Cr, Hg, Mn, Ni, Pb and Zn were analysed at their respective wavelengths using Microwave Plasma Atomic Emission Spectrometer (MP-AES, 4200). The concentrations of heavy metals were reported in mg/kg.

Determination of Bioaccumulation Factor (BAF)

The bioaccumulation factor (*BAF)* is a ratio of heavy metals present in vegetables to those found in the soil (Hossen et al., 2021):

$$BAF = \frac{C_{vegetable}}{C_{soil}} \quad (1)$$

Where: $C_{vegetable}$ and C_{soil} are the HMs concentrations in vegetables edible portions and soils on a weight basis, respectively (Halim et al., 2015).

Quality Control

Analytical grade reagents were used with no further purification. All glassware and plastic containers were soaked in 10% (v/v) nitric acid overnight before being washed three times with distilled water and three times with distilled deionized water. They were oven dried overnight at 50–60 °C. After oven drying, the containers were desiccated for about 20 minutes before being analyzed. All samples were analyzed three times, the standard solution for each metal was prepared by sequential dilution of certified standards (1000 mg/L) obtained from Sigma Aldrich, and a calibration curve for each metal was created. Blanks were determined using similar procedure.

Statistical Analysis

All data was transformed into the mean standard deviation of three replicates. One-way Analysis of Variance (ANOVA) was used to compare heavy metal concentration levels in different vegetable samples. A statistically significant difference was established using the Tukey HSD post-hoc test ($p \leq 0.05$). Statistical analyses were carried out using SPSS 23.0 (Chicago, USA), while Microsoft Excel® 2010 was used in plotting the graph.

Results and Discussion

Concentrations of Heavy Metals (HMs) in Vegetables

The concentrations of arsenic (As), cadmium (Cd), chromium (Cr), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn) in vegetables cultivated around the Riruwai mining area are presented in Table 1. The levels of As, Cd, Cr, Hg, Mn, Ni, Pb, and Zn in lettuce (*L. sativa*) samples are 0.20, 1.21, 3.66, 0.50, 1.46, 0.57, 6.43, and 64.25 mg/kg, respectively. Zinc accumulated the highest in *L. sativa*, while arsenic was found to be the lowest accumulated metal. The HMs in *L. sativa* accumulated in decreasing order of Zn > Pb > Cr > Mn > Cd > Ni > Hg > As. The values of all the HMs in *L. sativa* are above the threshold values established by WHO/FAO (2007), except for zinc. This indicates that *L. sativa* cultivated in the study area is contaminated with HMs due to mining activities. High concentrations of HMs in *L. sativa*, especially zinc, lead, and chromium, might be due to their corresponding high concentrations in the soils. This was consistent with the findings of Monterroso et al. (2014), who investigated the HMs distribution in plants grown in a Pb and Zn mining area in northwestern Spain. The concentrations of HMs (mg/kg) in tomato (*S. lycopersicum*) samples were: arsenic (0.19), cadmium (0.27), chromium (2.80), mercury (0.04), manganese (0.25), nickel (0.21), lead (0.40), and zinc (106.23). Zinc accumulated the most, while mercury had the lowest uptake. The uptake of HMs in *S. lycopersicum* follows a decreasing order of Zn > Cr > Pb > Cd > Mn > Ni > As > Hg. The concentrations are above the WHO/FAO (2007) maximum reference standards, indicating contamination of *S. lycopersicum* by heavy metals. The levels of arsenic, cadmium, chromium, mercury, manganese, nickel, lead, and zinc in bean (*P. vulgaris*) samples are 1.05, 1.15, 2.56, 0.09, 0.33, 0.28, 5.94, and 77.06 mg/kg, respectively. A high level of zinc accumulation was found in *P. vulgaris*, followed by lead, chromium, cadmium, arsenic, manganese, and nickel, with mercury having the lowest accumulation. Except for zinc, the levels of all HMs in beans exceed the WHO/FAO (2007) threshold values. Generally, the findings of this study revealed that the investigated vegetables are polluted with HMs and might pose a significant health risk to consumers. Similar reports of heavy metal overload in edible plants grown on contaminated sites are well documented (Gebeyehu and Bayissa, 2020). *L. sativa* accumulated the most heavy metals. This is because *L. sativa* is a leafy vegetable, and leafy vegetables grow faster and have higher transpiration rates than non-leaf vegetables. Consequently, leafy vegetables can increase the uptake of HMs by plant roots, causing metal to move from roots to other vegetable tissues (Luo et al., 2011; Chang et al., 2014). Moreover, due to their broad leaf area, leafy vegetables are more prone to HMs contamination by the dust from soil (Chang et al., 2014). The high level of HMs accumulated by vegetables is clearly consistent with the fact that plants in contaminated areas develop mechanisms to absorb greater amounts of the contaminants (Liang et al., 2017). HMs concentrations varied significantly ($p \leq 0.05$) between vegetables. This could be due to differences in their accumulation abilities caused by physiological and genetic differences as well as differences in soil properties (Khan et al., 2015; Liu et al., 2013). Consumption of HMs-contaminated foods for an extended period may result in an unending accumulation of toxins in the liver and kidneys of humans, thereby causing disruptions in biochemical processes such as liver, kidney, cardiovascular, nervous, and bone disorders (WHO, 2006). Lin et al. (2013) recommended that leafy vegetables should be cultivated in soil with low HMs content and non-leaf vegetables in soil with comparatively high HMs content in order to reduce HMs pollution in the vegetables.

Table 1. Concentrations of Heavy Metals (mg/kg) in Vegetables Grown around Riruwai Mining Area

Heavy Metals	Vegetables			WHO/FAO (2007)
	L. sativa	S. lycopersicum	P. vulgaris	
As	$0.20^{a} \pm 0.03$	$0.19^{a} \pm 0.01$	$1.05^{b} \pm 0.04$	0.15
Cd	$1.21^{a} \pm 0.01$	$0.27^{b} \pm 0.03$	$1.15^{a} \pm 0.09$	0.2
Cr	$3.66^{a} \pm 0.04$	$2.80^{b} \pm 0.11$	$2.56^{b} \pm 0.05$	2.3
Hg	$0.50^{a} \pm 0.03$	$0.04^{b} \pm 0.03$	$0.09^{b} \pm 0.03$	0.03
Mn	$1.46^{a} \pm 0.01$	$0.25^{b} \pm 0.04$	$0.33^{a} \pm 0.01$	0.2
Ni	$0.57^{a} \pm 0.03$	$0.21^{b} \pm 0.02$	$0.28^{a} \pm 0.00$	0.1
Pb	$6.43^{a} \pm 0.06$	$0.40^{b} \pm 0.01$	$5.94^{a} \pm 0.09$	0.3
Zn	$64.25^{a} \pm 0.09$	$106.23^{b} \pm 0.55$	$77.06^{c} \pm 0.04$	100

Values are mean ± standard deviation (n = 3). The values on the same row with same superscript letters are statistically the same ($P > 0.05$), whereas values on the same row with different superscript letters are statistically different ($P \leq 0.05$), as revealed by one-way ANOVA and the Tukey HSD post-hoc test.

Concentrations of Heavy Metals in the Soils

The concentrations of arsenic, cadmium, chromium, mercury, manganese, nickel, lead, and zinc in the farmland of the Riruwai mining area are 21.54 ± 1.81, 1.51 ± 0.06, 96.44 ± 3.75, 0.50 ± 0.09, 148.11 ± 4.56, 87.21 ± 2.15, and 426.51 ± 3.69 mg/kg (Figure 2.). The concentration of HMs decreases in the following order: Zn > Mn > Cr > Pb > As > Ni > Cd > Hg, with As, Pb, and Zn exceeding the recommended limit set by WHO/FAO (2001). High concentrations of Pb, Zn, and As observed in the farm area beyond their respective safe limits indicate the spread of heavy metal pollution from the mining sites. Acidic drainage and dust transport by wind may be the primary mechanisms causing pollution dispersion (Rodr guez et al., 2009).

Bioaccumulation Factor (BAF)

The results of Bioaccumulation Factor (*BAF*) in vegetables grown around the Riruwai Mining Area are presented in Figure 3. The *BAFs* of arsenic, cadmium, chromium, mercury, manganese, nickel, lead, and zinc in lettuce (*L. sativa*) are 0.02, 13.43, 0.05, 1.40, 0.02, 0.01, 0.10, and 0.18 , respectively. Cadmium had the highest *BAF*, whereas manganese had the lowest. According to Monterroso et al. (2014) and Bhatti et al. (2016), metal excluders have a *BAF* value of less than one, while accumulators have a *BAF* value of greater than one. The *BAF* of arsenic, chromium, manganese, nickel, lead, and zinc are less than one, indicating that the heavy metals are not accumulators of *L. sativa*. Cadmium and mercury, on the other hand, had *BAF* values of greater than one, signifying that mercury and cadmium are accumulators of *L. sativa*. The *BAF*s of HMs in tomato (*S. lycopersicum*) are arsenic (0.01), cadmium (3.00), chromium (0.04), mercury (0.11), manganese (0.003), nickel (0.01), lead (0.01), and zinc (0.29). With the exception of cadmium, the *BAF* values of all the studied HMs in *S. lycopersicum* were less than one. This shows that only cadmium is the accumulator of the plant, while the rest of the metals are excluders. *BAF* values of HMs in *S. lycopersicum* are in decreasing order of cadmium > zinc > mercury > chromium > arsenic > nickel > lead > manganese. The *BAF* values of arsenic, cadmium, chromium, mercury, manganese, nickel, lead, and zinc in beans (*P. vulgaris*) are 0.08, 12.76, 0.04, 0.25, 0.003, 0.02, 0.09, and 0.21, respectively.

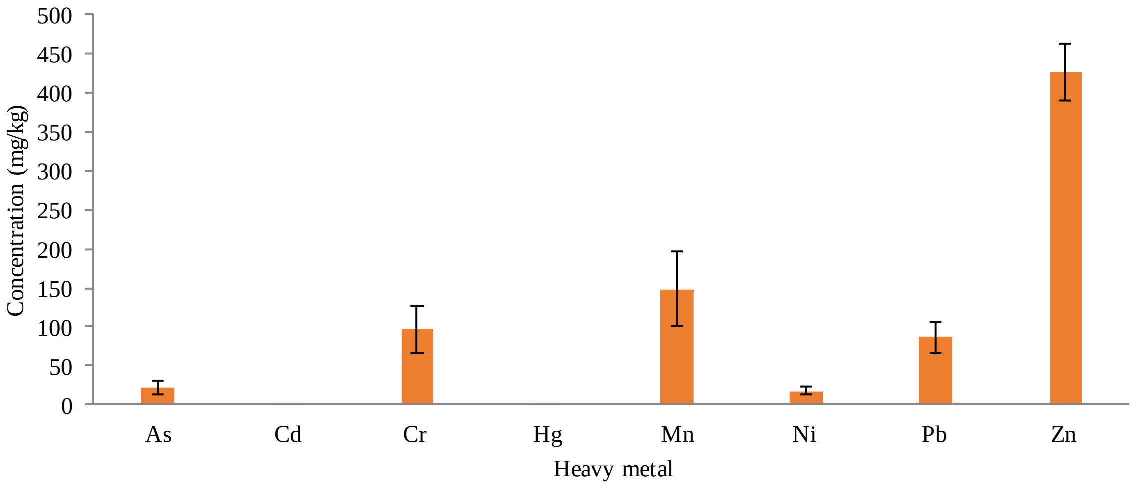

Figure 2. Heavy metals concentrations in the soils of Riruwai mining area

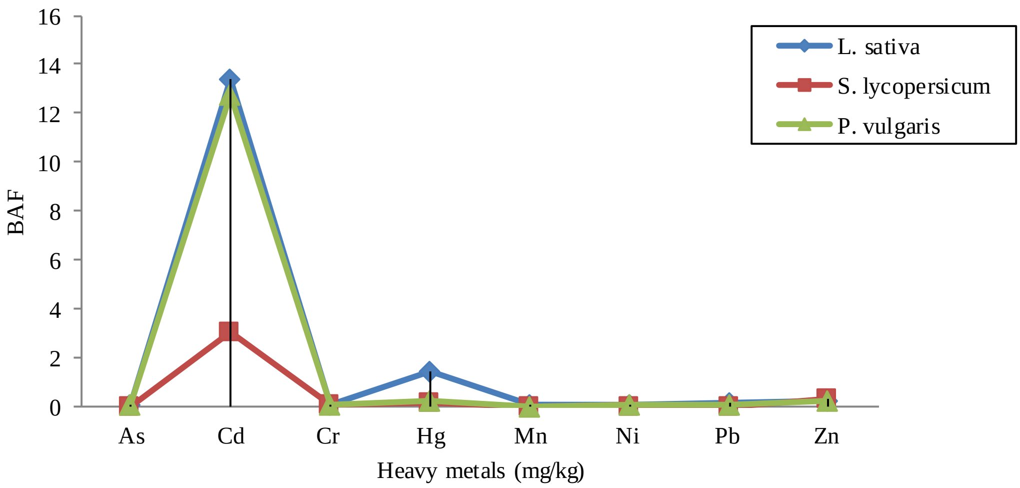

Figure 3. Bioaccumulation factor (BAF) vegetables grown around Riruwai mining area

The *BAF* of cadmium was the highest among the HMs. This is followed by mercury, zinc, lead, arsenic, chromium, and nickel, while manganese was recorded the lowest. The *BAF* of cadmium was significantly greater than one, demonstrating that cadmium had a considerably high accumulation capacity in *P. vulgaris*. High *BAF* value in cadmium could be ascribed to competition between Cd^{2+} and Ca^{2+} than other metals due to similarities in their ionic radius and valence state (Chang et al., 2014). Generally, cadmium can be considered as accumulator of all the studied vegetables (BAFs > 1.00), while mercury is regarded as accumulator of *L. sativa*.

Conclusion

In this study, the HMs contamination levels in vegetables grown around the Riruwai mining area in Kano State, Nigeria, and their corresponding soils were investigated. The findings of the study revealed that HMs concentrations in the investigated vegetables and soils were found to be significantly high, with the majority of levels exceeding the WHO/FAO (2007) threshold limit. Cadmium is an accumulator in all of the vegetables investigated (BAFs > 1.00), whereas mercury is an accumulator of *L. sativa*. The study recommends that planting and consumption of vegetables be stopped in the area until further research is conducted to determine the optimum concentration levels of HMs suitable for human consumption.

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