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The Fate of Selected Heavy Metals in River Water, Fruits and Vegetables to Potential Human Health Risks: The Case of Rau River in Moshi District, Tanzania

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Abstract
The presence of heavy metals in our environment significantly contributes to pollution in our food supply, particularly in fruits and vegetables. To analyze the levels of Cd, Cu, Fe, Pb, and Zn in green leafy vegetable (Amaranthus sp), fruit Citrullus lanatus (watermelon) and the Rau river water, an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) was used. Three sampling sites were selected, including one site with the least impact, Mawela and two highly impacted sites, Majengo and Msaranga. The order of heavy metal concentrations in the river water was found to be Fe > Cu > Zn > Pb > Cd, with the majority of the levels exceeding the acceptable standards set by the World Health Organization (WHO). There was significant variation in the concentrations of heavy metals in fruit. For example, the range of concentrations (mean ± standard deviation) observed were as follows: Fe (70.700 ± 1.532 to 221.010 ± 0.661 mg/kg), Cu (1.372 ± 0.047 to 2.540 ± 0.195 mg/kg), Zn (41.072 ± 0.519 to 45.718 ± 0.651 mg/kg), Pb (below detection limit to 0.398 ± 0.012 mg/kg), and Cd (0.031 ± 0.013 to 0.243 ± 0.112 mg/kg). The heavy metal concentration in Citrullus lanatus (watermelon) did not correlate with the levels found in the water samples. This disparity could be attributed to the watermelon's tendency to hyper-accumulate Cd from the river water. The hazard index, which assesses the potential health risks, indicates that the ingestion of vegetables exceeds a value of 1, while that of fruits remains below 1. Despite the heavy metal contamination levels in the tested vegetables and fruits being below the safe limit, there is a significant health concern regarding long-term exposure to even low doses of toxic elements. Therefore, it is crucial to conduct regular monitoring of heavy metals in all types of food items to accurately assess the health risks associated with heavy metal exposure in the human food chain.

Keywords: *Heavy metals, watermelon, Vegetables contamination, hazard index, Moshi*

1.0 INTRODUCTION
Toxic heavy metals are metallic elements that possess a higher density than water, as defined by Duffus (2002). In recent times, there has been growing

concern worldwide, both ecologically and in terms of public health, regarding the contamination of the environment by these metals (Bradl, 2002). Furthermore, human exposure to these metals has significantly increased due to their extensive utilization in various industrial, agricultural, domestic, and technological applications (He *et al*., 2005). The impact of heavy metals on our environment is substantial, particularly in relation to our food supply, specifically fruits and vegetables (Chauhan and Chauhan, 2014). Various sources of heavy metals in the environment have been reported, including geogenic, industrial, agricultural, pharmaceutical, domestic effluents, and atmospheric sources (Goyer, 2001).

Developing countries such as Tanzania, as highlighted by Alam *et al*. (2018), experience elevated levels of heavy metals in urban areas due to rapid industrialization, wastewater irrigation, and various other human activities. Given that the agricultural sector utilizes a significant portion, approximately seventy percent, of the available water (FAO, 2017); the reuse of treated wastewater plays a crucial role in promoting agricultural sustainability.

The release of industrial waste into rivers carries the risk of contaminating the water. This polluted water has direct repercussions on the soil, agricultural fields, and rivers, leading to the generation of multiple pollution sources, as noted by Butt *et al*. (2005). The presence of heavy metals in both soil and water raises significant concerns regarding public health, agricultural productivity, and ecological integrity, as emphasized by Fergusson and Kim (1991).

The contamination of surface water with toxic heavy metals represents a significant threat to human health, as stated by Yu *et al*. (2020). These metals can persist in the ecosystem as relatively stable and resistant contaminants (Redwan and Elhaddad, 2020), causing pollution in both surface and groundwater resources (Saria, 2016). Given the growing demand for water and the dwindling water supplies, coupled with increasing environmental pollution, it is imperative to monitor water quality, especially for drinking water supplies (Haghnazar *et al*., 2021).

The nutritional value of fresh fruits and vegetables is highly important for human health, as they contain vitamins, mineral salts, water, calcium, potassium, sulfur, and iron, as mentioned by Saria (2020). Furthermore, fruits and vegetables play a crucial role in maintaining health, as well as preventing and treating various diseases (Gwana *et al*., 2014). However, it is important to note that fruits and vegetables can contain both essential and toxic heavy metals, exhibiting a wide range of concentrations, as noted by Veazie and Collins (2004). Watermelon (*Citrullus lanatus*), a prominent fruit from the *curcurbitaceae family*, holds significant nutritional and medicinal value, as mentioned by Gwana *et al*. (2014). This fruit is known to be rich in carotenoids, vitamin C, citrulline, carbohydrates, water, sugar, and dietary fiber, as highlighted by Bruton *et al*. (2009). Additionally, Ogunbanwo *et al*. (2013) reported that regular consumption of watermelon can contribute to proper kidney function and may provide protection against cancer. In Tanzania, sliced watermelon fruits are typically sold by vendors in various locations such as marketplaces, roadside stalls, schools, and public bus stations.

In recent times, there has been a notable increase in public awareness regarding the health risks associated with the contamination of vegetables and fruits by heavy metals. This has made the risk assessment of heavy metal contamination a significant global concern, as highlighted by Jan *et al*. (2010). Prolonged consumption of vegetables and fruits contaminated with toxic levels of heavy metals can lead to the accumulation of these metals in the liver, kidneys, and bones of individuals, potentially resulting in kidney, cardiovascular, nervous, and bone diseases, as emphasized by Anwar *et al*. (2016). Furthermore, heavy metals may also contribute to the occurrence of congenital disabilities, low birth weight in babies (<2.5 kg), and premature births (<37 weeks of completed gestation), as reported by Taylor *et al*. (2015).

The objective of the present study was to assess the levels of Cd, Pb, Cu, Fe, and Zn in vegetable (*Amaranthus sp*), edible parts of watermelon (*Citrullus lanatus*), and the river water used for irrigation. Additionally, a non-carcinogenic risk assessment was conducted to evaluate the potential impact of consuming *Amaranthus sp* (vegetable) and *Citrullus lanatus* (watermelon) on human health. The study aimed to ensure the safety of the local population in the vicinity of Moshi Municipal.

2.0 MATERIALS AND METHODS

2.1 Study design

The research utilized a quantitative cross-sectional method to collect, compile, and analyze data. Sampling was conducted between October 2021 and June 2022 at three designated study points to facilitate comparisons. The study points, namely Mawela, Majengo, and Msaranga, were conceptually categorized as upper stream, middle, and downstream locations, respectively, based on the direction of water flow.

2.2 Sampling Site

Investigated Rau river is located in the northern-eastern part Kilimanjaro region, Tanzania mainland between 037º30’’E and 03º4’59’’S. The Lower Moshi irrigation Scheme relies on the Rau river water as its primary source for irrigation. Located approximately 20 km from Moshi town, the Scheme is situated in the Moshi Rural District of the Kilimanjaro region. Excessive extraction of water from the river for irrigation purposes has resulted in the degradation of the riverside wetland (Stuart, 1990). To ensure the preservation of the riverside wetlands in the Rau River, it is essential to maintain a healthy vegetation cover, which will provide habitats for fish and wildlife, aid in water purification, control soil erosion, and offer recreational opportunities. The Rau River courses its way southward, passing through the Njoro and Kahe forests, ultimately reaching its endpoint where it discharges into Lake Jipe (Odada and Olago, 2006). Three sampling stations were designated for the study, including one site with minimal impact known as Mawela, and two sites with significant impact, namely Majengo and Msaranga. The intensive land use practices in these areas, such as the cultivation of cash and food crops utilizing fertilizers and pesticides, animal grazing, and construction activities, have led to the near-total depletion of riparian vegetation.

2.3 Sampling

2.3.1 Water Sampling

Water samples were collected from three distinct points, namely Mawela, Majengo, and Msaranga. A total of 10 water samples were randomly obtained from each point, resulting in a grand total of 30 samples. Prior to collection, the sampling bottles were conditioned with 5% HNO3 and thoroughly rinsed with distilled de-ionized water, following the guidelines provided by APHA (2012). To ensure proper sampling, the polyethylene bottles were rinsed with river water at each sampling site a minimum of three times. Subsequently, the pre-cleaned polyethylene sampling bottles were immersed approximately 10 cm below the water surface, and approximately 0.5 L of water samples were collected from each site. The collected samples were then stored at temperatures below 4°C and sent to the Chief Chemistry Laboratory for further analysis.

2.3.2 Sampling of Vegetables and Fruits

Three individual 1 kg portions of African green leafy vegetable, specifically Amaranthus sp (commonly known as mchicha), were obtained from local gardens located in Mawela, Majengo, and Msaranga wards. To ensure hygienic conditions, the samples were carefully placed in pre-cleaned polyethylene bags. The collection of samples took place subsequent to removing the stalks from each sample, followed by rinsing them with de-ionized water. After allowing the excess moisture to naturally evaporate at room temperature, the samples were exposed to sunlight for a period of 2-3 days on a clean surface, with regular turning to prevent the growth of fungi.

A total of fifteen watermelon samples were collected from gardens located in Mawela, Majengo, and Msaranga wards, with five samples obtained from each ward. These watermelon samples were cultivated in gardens that were irrigated using water from the Rau River. To maintain cleanliness, the samples were stored in pre-cleaned polyethylene bags and subsequently sent to the Chief Government Chemistry Laboratory for analysis.

2.4 Sample Preparations and Analysis

Prior to determining the metal content, the instruments used were calibrated by utilizing standard solutions of metal ions prepared from salts. These standard solutions, obtained from Merck (Germany), were of the highest purity level (99.98%). To create the working standard solutions of Fe, Cu, Pb, Zn, and Cd, the commercial annular grade 1000 ppm stock solutions were diluted in 25cm3 standard flasks and then filled to the mark with deionized water, resulting in concentrations of 2.0 ppm, 3.0 ppm, and 4.0 ppm for each metal ion. These solutions were analyzed using an atomic absorption spectrophotometer.

To enable accurate analysis using the Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES), digestion was performed beforehand. This process served to eliminate interference from organic matter and convert the metals into a form suitable for analysis. Quality control measures were implemented by employing standard laboratory measurements and techniques, including replication as outlined in the APHA (2012) guidelines.

The test portions were subjected to a drying process in a drying oven set at 105°C until a constant weight was achieved. Afterward, they were cooled to room temperature and crushed using a clean pestle and mortar to obtain homogenized samples. These ground samples were then stored in airtight sealed polyethylene bags at room temperature.

Approximately 2.0g of each processed sample was weighed and underwent dry ashing in a well-cleaned porcelain crucible within a muffle furnace, specifically at 550°C. The resulting ash was dissolved in 5.0 mL of a mixture consisting of HNO3, HCl, and H2O (in a ratio of 1:2:3). The solution was gently heated on a hot plate until the disappearance of brown fumes.

To the remaining material in each crucible, 5.0 mL of de-ionized water was added and heated until a colorless solution was obtained. The mineral solution from each crucible was then transferred into a 100.0 mL volumetric flask through filtration using Whatman No.42 filter paper. The volume was adjusted to the mark using de-ionized water. This resulting solution was subsequently used for heavy metal analysis through Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES).

2.4.1 Fruit sample preparation and chemical analysis

The watermelon samples underwent a thorough rinse with de-ionized water. They were then sliced and homogenized using an electric blender equipped with stainless steel rotor knives. The resulting homogenized samples were dispensed into clean sterile sample bottles. To extract the desired components, the homogenized samples followed the acid digestion method described by Cui et al. (2010). Three portions, each measuring 30 mL, were accurately measured and transferred to a 200 mL beaker. Subsequently, 30 mL of 10% concentrated HNO3 was added, and the mixture was allowed to settle for 15 minutes. Wet acid digestion was then conducted using 10 mL of a 1:3 mixture of concentrated

HCl and HNO3 (obtained from Merck) on a hotplate until a clear solution was obtained. The digested samples were allowed to cool to room temperature.

The digested samples were filtered through Whatman No. 1 filter paper into a 50 mL volumetric flask. The final volume was adjusted to 50 mL using distilled water. The resulting solution was transferred to clean and dry plastic bottles for further analysis. Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) was employed to analyze the digestate and determine the concentration of heavy metals present.

2.4.2 Analysis of heavy metal parameters

From stock standard solutions containing 1000 mg/L of Cd, Pb, Cu, Zn, and Fe, intermediate standard solutions with a concentration of 100 mg/L for each metal were prepared. To create appropriate working standards, the intermediate solutions were diluted using distilled water in 2M HNO3. To optimize sensitivity, the working standards were aspirated sequentially into the Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) following the instructions provided in the instrument's operation manual. The absorbance of each working standard was recorded. Calibration curves were generated for each metal standard by plotting absorbance against concentration (mg/L), using different points on the curve. Once calibration was completed, the sample solutions were aspirated into the spectrometer, and the metal concentrations were directly determined by reading the results (as shown in Table 1).

Table 1: Calibration graph absorbance against the concentration of heavy metals (mg/kg)

S/N	Metal	Model for Absorbance vis Concentration	R^2
1	Zn	y = 0.3127x	0.9935
2	Cu	y = 0.0802x	0.9952
3	Cd	y = 0.057x	0.9906
4	Pb	y = 0.0032	0.9917
5	Fe	y = 0.018	0.9929

2.5 Assessment of Potential Human Health Risk

The heavy metals' non-carcinogenic risk to humans was determined by assessing the estimated daily intake (EDI) and hazard quotient (HQ).

2.5.1 Estimated daily intake (EDI)

The daily intake of metals from green leafy vegetables or from fruits depends on both the metal concentration in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance of contaminants. The EDI are a concept introduced to consider these factors. The EDI is calculated using equation 1.

$$EDI = \frac{CxIRxEFxED}{BWxAT} \quad \text{……………………………………………………………(1)}$$

Where EDI denotes estimated daily intake of heavy metals (mg/person/day); BW stands for adult body weight considered as 70 Kg (USEPA, 1989; FAO/WHO, 2019); C is the trace element concentration in the exposure medium (mg/kg); IR is the ingestion rate (kg/d), for the presents study the ingestion rate for vegetable was 0.32 kg/day and fruit was 0.22kg/day (Pipoyana et al., 2018; Storelli, 2008); EF is the exposure frequency (for investigated fruit and vegetable is 183 d/year); ED is the exposure duration 66 years (average Tanzania life expectancy) (UN, 2022). AT is the time period over which the dose is averaged (365 d x number of exposure years).

2.5.2 Hazard Quotient (HQ)

The risks to human health via the consumption of metal contaminated vegetables were estimated on the basis of the hazard quotient (HQ), which is defined as the health risk due to exposure of a pollutant with respect to the estimated daily intake (EDI). The HQ was calculated by dividing the estimated daily intake (mg/kg/day) of the contaminant through vegetable ingestion by the reference oral dose, as follows (equation 2).

$$HQ = \frac{EDI}{RfD} \qquad (2)$$

Where: EDI is the estimated daily intake; RfD (mg/kg/day) is The reference oral dose. This is an estimated value of tolerable daily ingestion of pollutants (maximum permissible risk) by human beings during a lifetime. The values of RfD used in this study were taken from De Miguel et al. (2007) and Sharma et al. (2016) where Pb = 0.004. Cd 0.001, Cu = 0.04, Fe = 0.7, Zn = 0. If the hazard quotient of a particular contaminant is less than one (<1), there would be no obvious adverse effects expected on the exposed population.

2.5.3 Hazard Index (HI)

Hazard Index (HI) measures aggregated non-cancerous risks because of heavy metals intake via regular ingestion of contaminated vegetables or fruits. To estimate the risk to human health through more than one heavy metal, the target hazard index (HI) has been developed (US EPA, 2013). The hazard index is the sum of the hazard quotients for all HMs, which was calculated by equation 3.

$$HI = \sum_{j=1}^{N} HQ_j = \sum_{j=1}^{N} \frac{EDI_j}{RfD_j} \qquad (3)$$

If the value of HI >1, it is considered to have possibility of substantial health hazard (Schaefer et al., 2000). If HI < 1, the non-carcinogenic adverse effect due to a particular route of exposure or chemical is assumed to be insignificant.

3.0 RESULTS AND DISCUSSION

3.1 Concentration of Heavy Metals in River Water

A comprehensive analysis was conducted on the total water samples to identify and quantify five heavy metals, namely Fe, Pb, Cu, Zn, and Cd. The levels of these heavy metals, based on the results obtained, followed the order Fe > Cu > Zn > Pb > Cd in the water samples collected from Rau River (Table 2).

Table 2: Heavy metal contents in water samples from Rau River (mg/L)

Metal	Mawela	Majengo	Msaranga	WHO (2008)	TBS (2005)
Zn	0.427±0.030	0.179±0.024	0.286±0.054	3.00	5.00
Cu	2.623±0.244	3.023±0.108	2.571±0.062	2.00	2.00
Fe	3.974±0.124	4.7393±0.0823	3.741±0.0956	3.00	3.00
Pb	0.320±0.018	0.2313±0.006	0.2602±0.027	0.01	0.01
Cd	0.110±0.01	0.2102 ±0.019	0.1193±0.045	0.003	0.002

The concentration of Fe varies from 3.741±0.096 mg/L (at Msaranga ward) to 4.739±0.082 mg/L (at Majengo ward). Similarly, the concentration of Cu ranges from 2.571±0.062 mg/L (at Msaranga ward) to 3.023±0.108 mg/L (at Majengo ward). These values exceed the limits set by WHO (World Health Organization) and TBS (Tanzania Bureau of Standards), which are 3.0 mg/L and 2.0 mg/L, respectively. The increased concentration of iron in river water could stem from various sources, such as runoff from small-scale industrial waste, natural geological influences, agricultural practices introducing fertilizers or pesticides, and the corrosion of iron-based structures along the riverbanks (Togibasa *et al*. 2018). Additionally, variations in pH levels or changes in the water's oxygen content could also influence the solubility and concentration of iron in the water, leading to elevated levels beyond recommended thresholds. These findings are consistent with a previous study by Parihar *et al*. (2020) which reported copper levels in river water ranging between 3.579 and 4.968 mg/L and iron levels between 2.578 and 2.690 mg/L. Although iron is an essential element, its high concentration in river water can lead to various problems, such as incrustation, clogging of water treatment facilities, acidification of water leading to pipe corrosion, decreased soil productivity, and unpleasant taste in drinking water (Parihar *et al*., 2019). Additionally, high iron intake by humans can result in "haemochromatosis," a disorder that necessitates frequent blood transfusions (Parihar *et al*., 2020). Copper is a naturally occurring element that is commonly found in water even at low concentrations, and it can be toxic. In mammals, it is known to cause brain damage (Fatoki *et al*., 2002).

The concentration of Pb varies from 0.231±0.006 mg/L (at Majengo ward) to 0.320±0.018 mg/L (at Mawela ward), while the concentration of Cd ranges from 0.110±0.014 mg/L (at Mawela ward) to 0.210±0.019 mg/L (at Majengo ward). Both of these values exceed the permissible limits for drinking water set by the WHO (2008), which are 0.01 mg/L for Pb and 0.003 mg/L for Cd. These results are lower compared to the values reported in a previous study by Nkinda *et al*. (2021) on water samples from the tributaries of the Mara River in Tanzania, which showed concentrations of Pb to be 0.76±0.09 mg/L and Cd to be 0.74±0.1 mg/L.

The presence of Cd is of major concern due to its potential health risks to both humans and aquatic organisms. The detected levels of cadmium may be attributed to emissions through air and water from hazardous waste sites and nearby factories along the river. High levels of Cd have been associated with various diseases, including memory loss, reproductive defects, cancer, as well as damage to the lungs, kidneys, and immune system, which can ultimately lead to death (WHO/UNICEF, 2015). Similarly, elevated levels of Pb exceeding 0.01 mg/L have been linked to anaemia, memory loss, anorexia, brain damage, and even fatality (DWAF, 2002).

The concentration of zinc ranges from 0.18 mg/L (at Majengo ward) to 0.43 mg/L (at Mawela ward). These levels are below the permissible limits set by the WHO (3.00 mg/L) and TBS (5.00 mg/L). Zinc is an essential element for the human body, serving as a catalytic or structural component in numerous enzymes involved in energy metabolism (Roohani *et al*., 2013). However, high doses of zinc can lead to health complications such as fatigue, dizziness, and neutropenia (Laura *et al*., 2010).

3.2 Concentration of Heavy Metals in Water Mellon (Citrullus lanatus)

Table 3 present the concentrations of Fe, Cu, Zn, Pb, and Cd in watermelon samples collected from three sites along the Rau River.

Table 3: Heavy Metals Concentration in Water Mellon (Citrullus lanatus) (mg/kg)

Metal	Mawela	Majengo	Msaranga	JECFA (2002)
Zn	44.860±0.631	41.072 ±0.519	45.718±0.651	99.4
Cu	2.540±0.195	1.372±0.047	2.169±0.115	73.3
Fe	70.700±1.532	221.010±0.661	208.187±0.625	425.5
Pb	BDL	0.321±0.081	0.398±0.012	0.3
Cd	0.031±0.013	0.243 ±0.112	0.230±0.072	0.2

The concentrations of Fe, Cu, Zn, Pb, and Cd in the watermelon samples showed significant variations. The range of concentrations observed were as follows: Fe (70.700±1.532 to 221.010±0.661 mg/kg), Cu (1.372±0.047 to 2.540±0.195 mg/kg), Zn (41.072±0.519 to 45.718±0.651 mg/kg), Pb (BDL to 0.398±0.012 mg/kg), and Cd (0.031±0.013 to 0.243±0.112 mg/kg).

These values were found to be higher compared to the concentrations reported in watermelon samples from Saudi Arabia by Ali and Khairia (2012), where Fe was 112.8 mg/kg, Cu was 3.21 mg/kg, Pb was 3.67 mg/kg, and Cd was 1.13 mg/kg. However, the concentration of Zn was lower in this current study compared to the findings from Saudi Arabia (29.78 mg/kg).

With the exception of Cd in Majengo and Msaranga, the concentrations of the other metals in the watermelon samples were below the maximum acceptable limits set by JECFA (2002). The elevated concentrations of Fe and Zn could potentially be attributed to the use of fertilizers, pesticides, organic waste dumping, and the utilization of sludge in the area.

The study revealed that the concentrations of selected heavy metals in watermelon were not directly proportional to those found in the water samples. Specifically, the highest concentration of Cd in the water samples was measured at 0.12 mg/kg, whereas in the watermelon samples, it was found to be 0.24 mg/kg. This observation could be attributed to the watermelon's tendency to hyper-accumulate Cd from the soil or water, as suggested by Hussain *et al*. (2021). Additionally, other potential sources of Cd exposure, such as wastewater irrigation, might also contribute to the elevated levels detected in the watermelon samples.

The relatively low levels of copper observed in all the sliced watermelon fruits may be attributed to minimal copper deposition in the soils (Akinola and Ekiyoyo, 2006) and minimal contamination during processing. Ashish *et al*. (2013) reported that the ingestion of copper beyond its permissible limit can lead to liver and gastrointestinal problems, while Flora *et al*. (2007) highlighted that exceeding the upper limit of copper intake can result in anemia, anxiety, insomnia, and cardiovascular diseases. Therefore, it is crucial to monitor copper levels in food to prevent them from exceeding the maximum permissible limit.

Zinc (Zn) is a vital element for both plants and animals, but even a slight increase in its concentration can disrupt physiological processes. Interestingly, the presence of Zn appears to play a crucial role in counteracting the toxic effects of cadmium (Cd). In the present study, the highest quantity of zinc was detected in the melon samples, with a concentration of 45.718±0.651 mg/kg. This finding deviates from the results reported earlier in Libya by Elbagermi *et al*. (2012), where the zinc concentration was reported to be 8.24 mg/kg.

3.3 Concentration of Heavy metal in Amaranthus sp (Mchicha)

The average concentrations of heavy metals (Fe, Cu, Zn, Pb, and Cd) in Amaranthus sp (Mchicha) collected from three sites along Rau River are presented in Table 4.

Table 4: Heavy Metal Concentration in *Amaranthus sp* (Mchicha)

Metal	Mawela	Majengo	Msaranga	WHO/FAO, 2007
Cd	0.220±0.007	0.142±0.012	0.238±0.007	0.2
Cu	0.279±0.032	1.922±0.143	1.230±0.055	73.3
Fe	13.500±0.370	31.820±1.059	28.269±0.446	425.5
Pb	0.140±0.021	0.020±0.014	0.130±0.008	0.3
Zn	48.560±0.855	51.647±0.867	45.718±0.687	99.4

The concentration of iron (Fe) in Amaranthus sp was found to be below the permissible limit set by WHO/FAO (2007). These findings align with the results reported by Zahir *et al*. (2009), who analyzed various vegetable samples and observed high concentrations of Fe (7.9-24.8 mg/kg) in Pakistan. Similarly, Waheed *et al*. (2003) conducted a study on raw foodstuffs grown in wastewater industrial areas and reported Fe concentrations ranging from 17.0 to 35.60 mg/kg.

The concentration of zinc (Zn) in Amaranthus sp varied from 45.718±0.687 mg/kg at Msaranga ward to 51.647±0.867 mg/kg at Majengo ward. These results were notably higher than those reported by Singh *et al*. (2004), who found Zn concentrations ranging from 3.56 to 4.59 mg/kg in vegetables. Conversely, our findings were consistent with the results obtained by Al Jassir *et al*. (2005), who reported Zn levels ranging from 14.14 to 76.28 mg/kg in certain vegetables. Overall, the present study demonstrated that the concentration of Zn in Amaranthus sp was below the international standards limits (99.4 mg/kg, WHO/FAO, 2007).

The Amaranthus sp (Mchicha) samples collected from Mawela and Msaranga wards exhibit the highest recorded concentration of Pb, with values of 0.140±0.021 and 0.130±0.008 mg/kg respectively. These concentrations fall below the acceptable limits set by the WHO/FAO (2007). Despite the fact that the Pb concentrations in the vegetable are within permissible levels according to FAO/WHO standards, it is crucial to consider the quantity consumed and the frequency of intake in the long term (Chove *et al*., 2006). The cumulative effect of sustained intake of heavy metals is significant since they tend to persist in the body and are not easily eliminated.

At Majengo ward, the Amaranthus had the highest recorded Cu concentration of 1.922±0.143mg/kg. The present findings indicate lower levels of Cu than those reported previously by Sharma *et al*. (2006) for vegetables grown in wastewater areas of Varanasi, India, which ranged from 2.25-5.42 mg/kg.

The reduced Cu concentration in vegetables can be attributed to the relatively lower release rates of copper from contaminated soils compared to other heavy metals, as indicated by Sukreeyapongse *et al*. (2002). The high tendency of added copper to persist in the soil acts as a significant controlling factor for its uptake by plants. Additionally, this phenomenon is likely due to the absence of industrial areas and areas with intense human activity along the Rau River, which runs near the sampled sites.

The highest concentration of cadmium was detected at Msaranga ward, (0.238±0.007 mg/kg) was exceptionally lower compared to values found in vegetables from Titagrah, West Bengal, India (17.79 mg/kg) (Gupta *et al*., 2008). Furthermore, it was lower than values reported in China (0.73 mg/kg) (Liu *et al*., 2005), but significantly higher than values observed in vegetables from Egypt (0.008 mg/kg) (Doghein *et al*., 2004). The lower Cd values in Msaranga ward can be attributed to soil pH levels. Soil pH plays a role in the uptake of cadmium by vegetables, with pH values below 5.5 increasing Cd uptake, while pH values above 6 reduce Cd uptake in crops. In the study area, the soil pH ranges from 6.6 to 7.3 (TARI, 2020), resulting in lower Cd uptake by the vegetables.

3.4 Non-carcinogenic Health Risk Assessment

Table 5 present results of the assessment conducted to determine the health risk posed to the residents of Moshi Municipality and its surrounding areas due to the intake of heavy metals from consuming Amaranthus sp and watermelon. The study calculated the daily intake of metals (DIM), health risk index (HRI), and hazard quotient (HQ) using equations 1, 2, and 3, respectively.

3.4.1 Daily intake of heavy metals from fruit and vegetable (DIM)

The results of the DIM analysis in Table 5 were compared to the recommended daily intake of metals and the upper tolerable daily intake level (UL) set by the Institute of Medicine for individuals aged 19 to 70 years (FDA, 2001; Garcia-Rico, 2007).

Table 5: Average Daily Intake of Heavy metals in Vegetable and Fruits (mg/day)

Metal	Sample	Mawela	Majengo	Msaranga	Upper Limit*
Cd	*Amaranthus*	5.04×10^{-4}	3.25×10^{-4}	5.47×10^{-4}	0.064
	Citrullus lanatus	3.47×10^{-4}	2.24×10^{-4}	3.76×10^{-4}	
Cu	*Amaranthus*	6.39×10^{-4}	4.41×10^{-3}	2.819×10^{-3}	10.0
	Citrullus lanatus	4.40×10^{-4}	3.03×10^{-3}	1.94×10^{-3}	
Fe	*Amaranthus*	3.09×10^{-2}	7.29×10^{-2}	6.48×10^{-2}	45
	Citrullus lanatus	2.13×10^{-2}	5.01×10^{-2}	4.45×10^{-2}	
Pb	*Amaranthus*	3.21×10^{-4}	4.60×10^{-5}	2.98×10^{-4}	0.24
	Citrullus lanatus	BDL	3.20×10^{-5}	2.05×10^{-4}	
Zn	*Amaranthus*	1.11×10^{-1}	1.18×10^{-1}	1.05×10^{-1}	40.0
	Citrullus lanatus	7.65×10^{-2}	8.14×10^{-2}	7.20×10^{-2}	

** (UL) Upper limit levels of heavy metals in foodstuffs (FDA, 2001; Garcia-Rico, 2007)*

Table 5 clearly shows that the vegetable species Amaranthus had the highest estimated daily intake of metals for Zn (8.14×10^{-2} mg/day), Pb (3.21×10^{-4} mg/day), Fe (7.29×10^{-2} mg/day), Cu (4.41×10^{-3} mg/day), and Cd (5.04×10^{-4} mg/day). On the other hand, the fruit species Citrullus lanatus had the highest estimated daily intake of metals for Zn (8.14×10^{-2} mg/day), Pb (2.21×10^{-4} mg/day), Fe (5.01×10^{-2} mg/day), Cu (4.40×10^{-4} mg/day), and Cd (3.76×10^{-4} mg/day).

In this study, the detected daily intake values of heavy metal were found to be lower than those reported in a previous study by Elbagermi *et al*. (2012). Elbagermi *et al*. (2012) found that the average contribution of heavy metal intake from a fruit-based diet for an average human being was 0.03689 mg/day for Pb, 0.0554 mg/day for Cd, 0.205 mg/day for Zn, and 0.288 mg/day for Cu. In the same study, the estimated daily intake levels of Pb, Cd, Zn, and Cu from vegetable consumption were 0.25 mg/day, 0.14 mg/day, 8.15 mg/day, and 3.36

mg/day, respectively, which are higher than the values found in this current study.

Both studies' results are significantly lower than the recommended daily intake of metals and the upper tolerable daily intake level (UL). It is noteworthy that 60% of the highest estimated daily intake of the analyzed heavy metals in vegetables and fruits was observed in Majengo ward, which is considered a middle-ranking area. The elevated levels of Cd and Zn measured in Majengo ward could potentially be attributed to the excessive use of fertilizers and pesticides in Mawela ward, given that crop production dominates the land use in this catchment area. Additionally, it is possible that some individuals in this area consume more than twice the average amount of rice, resulting in their daily dietary intake of heavy metals surpassing the DIM. The high daily intake of heavy metals from vegetables and fruits in Majengo ward is suspected to originate from industrial activities, traffic emissions, and hospital waste.

3.4.2 Hazardous Quotient (HQ)

Figure 2 shows that the hazard quotient (HQ) of the tested heavy metals in the selected leafy vegetables and fruits from the Rau river area are all below 1.0. Based on the results, the consumption of these vegetables and fruits can be deemed safe, with no significant risk to human health. In both the vegetable (Amaranthus sp) and the fruit (Citrullus lanatus), the hazard quotient (HQ) followed a descending order of Cu < Fe < Pb < Zn < Cd. It is important to note that while the HQ-based risk assessment method does not provide a quantitative estimate of the probability of adverse health effects in an exposed population, it does offer an indication of the risk level associated with pollutant exposure (Chary *et al*., 2008). The reliability of the risk estimation method has been acknowledged by many researchers (Chary *et al*., 2008; Khan *et al*., 2009; Wang *et al*., 2005), and its validity and usefulness have been demonstrated.

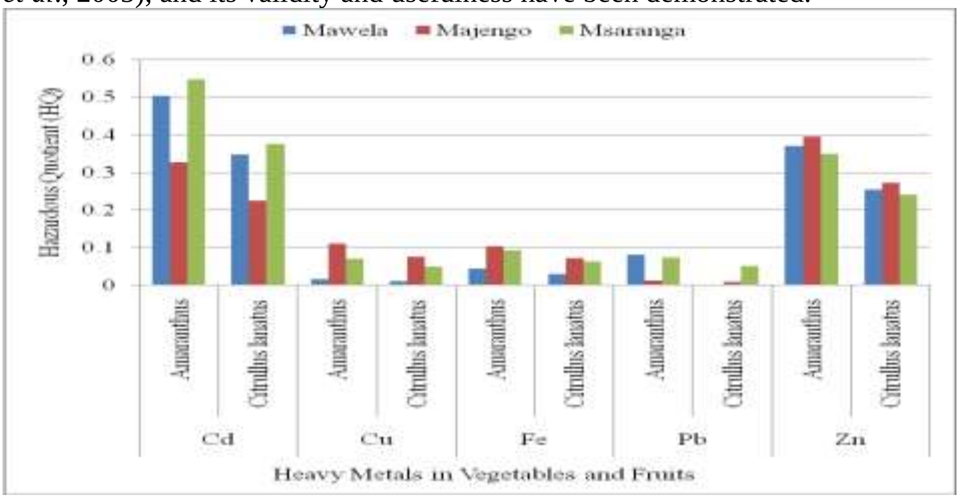

Figure 2: Hazard Quotient (HQ) of the Tested Heavy Metals in Vegetable and Fruit

3.4.3 Hazard Index

The hazard index calculated on the basis of ingestion of vegetable is higher than 1, indicating an axiomatic adverse health impact to residents (Figure 3).

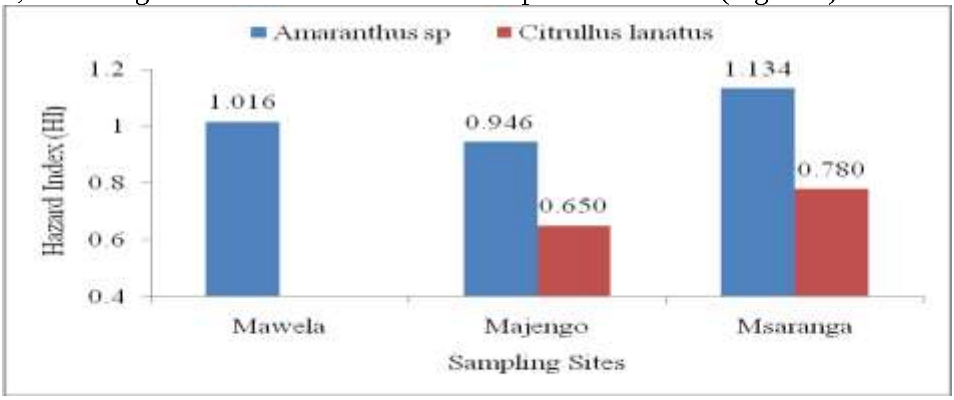

Figure 3: Hazard Index for heavy metals resulting from vegetable and fruit analyzed

Nevertheless, the health risk associated with fruit ingestion was found to be below 1, indicating that the potential health risk posed by consuming fruits in the Rau River and its surroundings is not significant.

4.0 CONCLUSION AND RECOMMENDATIONS

Several research studies have established a clear connection between the excessive bio-accumulation of heavy metals and various health abnormalities. These heavy metals not only present immediate environmental health risks but also pose long-term threats. Leafy vegetables and fruits that are cultivated in open-fields or irrigated with contaminated water have been found to contain high concentrations of heavy metals, thereby posing a significant risk to overall health and well-being. The hazard index calculated for vegetable ingestion exceeds 1, indicating a potential health risk. On the other hand, the hazard index for fruit ingestion is below 1, suggesting a lower health risk. However, it is important to note that when considering all other routes of heavy metal exposure, the potential health risks for the residents of Moshi might actually be higher.

The findings of this study suggest several recommendations. Firstly, raising awareness about the benefits of organic farming and promoting the long-term application of fertilizers, pesticides, and sewage effluents should be prioritized. Discouraging practices such as the direct release of untreated sewage water into agricultural fields or river bodies is crucial. Furthermore, Tanzanian food and health agencies should take a proactive approach in providing the public with information on permissible limits (both minimum and maximum) for contaminants in food. Regular monitoring of heavy metals in vegetables and other food items is essential to prevent their excessive accumulation in the food chain. It is imperative to emphasize the need for continuous monitoring of heavy metals in all types of food, not just fresh produce, to accurately assess the potential health risks associated with heavy metal contamination in the human food chain.

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