Heavy Metal Levels in Soil, Tomatoes and Selected Vegetables from Morogoro Region, Tanzania

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To cite this article:
Lugwisha Esther Hellen, Othman Chande Othman. Heavy Metal Levels in Soil, Tomatoes and Selected Vegetables from Morogoro Region, Tanzania. International Journal of Environmental Monitoring and Analysis. Vol. 4, No. 3, 2016, pp. 82-88. doi: 10.11648/j.ijema.20160403.13

Received: March 18, 2016; Accepted: March 28, 2016; Published: April 20, 2016

Abstract: The concentrations of heavy metals of Cd, Cu, Cr, Pb and Zn were determined in cauliflower (Brassica oleracea L. var. butrytis L.), carrot root (Daucus carota L.), tomato fruit (Lycopersicon esculenta Mill.), onion bulb (Allium cepa L.) and leafy cabbage (Brassica oleracea var. capital capitata L.), and the respective soils from Morogoro region, Tanzania. The accumulated heavy metals were quantified and levels compared to FAO/WHO (2003) and TZS (2007) safety limits for such produce. The methodology involved random sampling, extraction of the metals from the tomatoes, vegetables and soil and determination of heavy metals by using ICP-OEC and GFAAS. The results showed that levels of Cd, Cu, Cr, Pb and Zn at 90% of the sites in vegetables and tomatoes were above the FAO/WHO limit. The mean concentrations of Cd, Cr, Pb and Zn in all vegetables and tomatoes except Cu were also found to be above this limit therefore advocating a health risk for consumers. Levels of heavy metals in the soils were below the limits of the Tanzanian standard (TZS 972: 2007) and were lower than levels found in vegetables. The bioconcentration factor for Cu recorded 1 for tomatoes and cabbage at Towelo and below 1 for tomatoes and cauliflowers from Mgeta. Cd in onions at Mgeta as well recorded below 1. However, the bioconcentration factor for these heavy metals in vegetables and tomatoes in all other sites were found to be above 1 in all heavy metals an indication of high uptake of heavy metals in the vegetables and tomatoes from the soil. These results suggest that such vegetables and tomatoes might place the consumers at health risk; therefore they are not safe for consumption in their raw state.

Keywords: Heavy Metals, Vegetables, Tomatoes, Soil, Bioconcentration Factor, Tanzania

1. Introduction

Heavy metals are found naturally in the earth, and become concentrated as a result of human activities such as industrial production, mining, agriculture and transportation [1, 2]. These metals have both positive and negative roles in human life [1-4]. For instance, heavy metals such as copper (Cu), chromium (Cr), cobalt (Co), manganese (Mn) and zinc (Zn) are essential micronutrients for higher animals and for plant growth [5]. However, at higher concentrations they can lead to poisoning. On the other hand, Lead (Pb), cadmium (Cd), Arsenic (As) and nickel (Ni) are significant environmental pollutants [6]. Studies have revealed that fruits and leafy vegetables are vulnerable to heavy metal contamination from soil, wastewater and air pollution [7]. Heavy metals such as Cd, Cu, Pb, Cr, Zn, Ni, As, Co and Hg cannot be degraded or destroyed and can be accumulated in living tissues through the food chain, causing various diseases and disorders [7]. This fact necessitates for frequent determination of heavy metals in fruits, vegetables and soil to ensure that their levels meet the agreed international standards for the safety of consumers. Fruits, vegetables and other foods are among pathways by which heavy metals enter the human tissues leading to deterioration of health [8]. Tanzania has several regions where fruits, vegetables and other horticultural products are grown. These regions among others include Lushoto, Iringa, Mbeya, Morogoro, Kilimanjaro and Arusha [9, 10]. Morogoro Region is well known for the growing of tomatoes and green vegetables throughout the year. These fruits and vegetables are sold through different chains of markets, from local villages, regional to national markets and supermarkets while others are exported to neighboring countries [10]. Several studies conducted in Tanzania, on heavy metals in
fruits, vegetables and soils have indicated the presence of heavy metal contamination at different concentrations [11-15]. A similar study was conducted in Morogoro for pumpkin and chinese cabbage by Chove et al. [16]. Lugwisha and Othman [17] studied the contents of heavy metals in tomato fruits, vegetables and respective soil in Lushoto district and found that the vegetables and fruits were not safe for consumption. Determination of heavy metals in edible portions of amaranth (Amaranthus viridis), chinese cabbage (Brassica chinensis), cowpea leaves (Vigna unguiculata), leafy cabbage (Brassica oleracea var. capitata) and pumpkin leaves (Cucurbita moschata) in Dar es Salaam has shown the presence of Cd, Ni, Pb, Cr, Cu and Zn [18]. A direct correlation between Zn and Pb levels in soils with the levels in vegetables was also observed [12]. Consuming such vegetables maylead to severe health problems, such as acute/chronic chemical poisoning of body tissues. Knowledge of the contamination of soil, fruits and vegetables with heavy metal from some areas in Morogoro region, Tanzania has not yet been established; therefore, the present study was undertaken with the aim to investigate the concentration of some specific heavy metals (Cd, Cu, Cr, Pb and Zn) in cauliflower (Brassica oleracea var. botrytis L.), onion bulb (Allium cepa L.) carrot root (Daucus carota L.), leafy cabbage (Brassica oleracea var. capitata L.), tomato fruits (Lycopersicum esculentum Mill.), and the respective soils from this region.

2. Material and Methods

Samples of tomatoes, cauliflowers, cabbages, carrots and onions and of their respective soils were collected from 9 different locations (Table 1) in Morogoro region. About two kilograms of each sample of tomatoes and vegetables were randomly collected at each site, wrapped in pre-cleaned aluminum foil, packed in polyethylene bags and transported to Chemistry Department, University of Dar es Salaam for pre-treatment. Simultaneously, soil was sampled from the same locations of vegetables and tomatoes collection from a depth of 2 to 15 cm by using a non-steel sampler (coop sampling). Samples were stored in sealed pre-cleaned polyethylene bags and transported to the Chemistry Department for investigation.

Table 1. Locations, soil pH and samples from Morogoro Region, Tanzania.

| Location  | Soil pH | Sample name | Scientific name |
|-----------|---------|-------------|-----------------|
| Kihonda   | 7.22    | Onion       | Allium cepa L.  |
| Mgeta     | 5.84    | Tomatoes    | Lycopersicon esculentum Mill. |
| Mgeta     | 6.30    | Cabbages    | Brassica oleracea var. capital capitata L. |
| Mgeta     | 5.56    | Cauliflower | Brassica oleracea var. botrytis L. |
| *Mgeta sokoni* | NA     | Onion       | Allium cepa L.  |
| *Mgeta sokoni* | NA     | Tomatoes    | Lycopersicon esculentum Mill. |
| Towelo    | 6.13    | Tomatoes    | Lycopersicon esculentum Mill. |
| Towelo    | 5.53    | Carrots     | Daucus carota L. |
| Towelo *  | 6.13    | Cabbages    | Brassica oleracea var. capital capitata L. |

* Soil sample was not collected as samples were collected from the market

2.1. Sample Preparation

2.1.1. Vegetable and Tomato Samples

Chove et al. [16] method which was followed with minor modifications was used for vegetable and tomato samples collection. The collected samples were first washed with tap water and rinsed three times with distilled water to remove surface pollutants and any items adhering to the surfaces. The washed samples were sliced into small pieces and dried in open air on paper for about 2 hours to eliminate excess moisture. Each sample was weighed, dried in an oven at 80 °C for several hours and reweighed to constant weight. The dried sample was then ground in a mortar until it could pass through a 2 mm mesh sieve and stored in clean and dry polyethylene bags.

2.1.2. Soil Sample Extracts

Larger particles and other debris were removed from the soil samples. The samples were then air dried, ground, and sieved. Approximately 5.0 g samples were weighed by using a digital weighing balance and placed in a flask. 20 mL of extracting acid solution (0.05N HCl + 0.025N H2SO4) were added to the samples and shaken vigorously by a mechanical shaker for 15 minutes. The solution was then filtered through a Whatman filter paper No. 42 into a 50 mL volumetric flask and diluted to the mark prior to analysis of soil parameters.

2.1.3. Sample Digestion - Dry Ashing

About 2 g of ground tomatoes and vegetables were placed in a clean porcelain crucible and kept in a cool muffle furnace. The sample was then ashed at 450 - 500°C overnight (12 hours). The ashed sample was cooled to room temperature in a desiccator. The ash was then dissolved in 5 mL of 20% (v/v) hydrochloric acid. The solution was warmed slowly to dissolve any residues. The solution was filtered through an acid-washed Whatman filter paper No 42 into a 50 mL volumetric flask. The filter paper was washed with distilled water and washings collected in the volumetric flask. The resulting solution was diluted to the mark with distilled water, well mixed and used for determination of the heavy metals in vegetable samples [12].

2.2. Determination of Soil pH

The soil pH was determined by the use of pH-meter (RS Components Ltd, stock No 610-540, serial No.082316, UK) with a combined glass electrode. The pH meter was calibrated over the proper range using standard buffer solutions of pH 4 and pH 7. Adjustment was done for temperature and potential required by the instrument.

About 5 g of each soil samples were accurately weighed using the digital weighing balance (Mettler Toledo model B 301-S) and sieved. 50 mL distilled water was added to the sample [19]. The mixture was stirred vigorously for 15 seconds in a mechanical shaker and left to stand for 30 minutes. The electrode removed from the buffer solution was rinsed in the sample, placed in the slurry and swirled carefully. The pH values were recorded after each reading had stabilized. The electrode was always rinsed with distilled water between successive measurements.
2.3. Determination of Heavy Metals

Determination of heavy metals from the filtrate of tomatoes, vegetables and soil samples were carried out by using the GFAAS (Analytik Jena AG/Konrad-Zuse-Straße 1/07745/Germany) for Cu and ICP-OES (Ultma2, HORIBA Jobin Yvon S.A.S, version 3, 2001, France) for Cd, Cr, Pb and Zn. The analytical procedure is as described in [12].

2.4. Quality Assurance

Quality control tests were conducted on soil, tomatoes and vegetable samples in order to evaluate the experimental procedures and efficiency of the GFAAS and ICP-OES methods. The quality assurances was done by spiking each of the pre-digested samples of vegetable and soil with diluted solution of heavy metals standard solution (20 µg/kg of Cd, Cr, C, Pb, and Zn) prepared from several dilution of the 1000 µg/g stock standard solution. The spiked samples were then digested and extracted using the same procedure as that of soil and vegetable samples.

2.5. Statistical Analysis

Unpaired student’s t-test was used to determine and compare the statistical differences between the mean concentrations of two groups of samples. A probability level of P < 0.05 was considered statistically significant. The analysis was performed by feeding the raw data into a special program for unpaired student’s t-test determination as described by Kothari [20].

2.6. Bioconcentration Factor (BCF)

The transfer of trace elements from soil to plant edible parts is best described by considering the bioconcentration factor. BCF is calculated as the ratio of the concentration of heavy metals in the vegetables to that in the corresponding soil where vegetables were obtained, all based on (dry weight) for each vegetable separately.

The bioconcentration factor = [M] plant / [M] soil where [M] plant and [M] soil represent the heavy metal concentration in extracts of plants and soils on a dry weight basis respectively.

3. Results and Discussion

3.1. Soil pH

The pH of the soils from the investigated locations (Table 1) ranged between mildly acidic (pH 5.53) at Towelo to moderately alkaline (pH 7.22) at Kihonda, with a mean of pH 6.06 ± 0.60 (i.e. 6.06 ± Std Deviation). Akinola et al. [19] conducted a study in a non-industrial area and recorded pH 6.02. He concluded that pH recorded near neutral cause low uptake of heavy metals by vegetables, thereby leaving a higher concentration in the soil. In our study, in soil where pH was recorded near neutral (7.22), low concentrations of heavy metals were recorded in the soil contrary to [19]. This might be due to vegetables and tomatoes absorbing heavy metals through leaves exposed to air from polluted environment [16].

3.2. Concentrations of Heavy Metals in Vegetables, Tomatoes and Soils

3.2.1. Cadmium

Cadmium (Cd) is one of the most toxic heavy metals, because it bioaccumulates and has a long half life of about 30 years thereby causing health disorders such as lung cancer, kidney damage (necrotic protein precipitation), metabolic anomalies caused by enzyme inhibitions, reproductive failure, damage of central nervous system and DNA [21, 22, 24].

The concentrations of Cd in tomatoes, vegetables and soils at different sites of Morogoro are presented in Fig. 1. The concentrations of Cd in tomatoes and vegetables were recorded below detection limit in all samples except in onions from Mgeta sokoni (0.09 µg/g-dw) and in cabbages from Towelo (0.14 µg/g-dw), both values being above the FAO/WHO [23] permissible limit of 0.05 µg/g. At Towelo (cabbage), the soil recorded the pH of 6.13 which meant the soil was slightly acidic, thus facilitating solubility and availability of Cd for vegetable uptake [19]. The Cd uptake by vegetables from soil is greater at low pH of soil [19, 23].

In the respective soils, Cd was recorded below detection limit in all samples except in tomatoes (0.02 µg/g-dw) and cabbage (0.03 µg/g-dw) at Mgeta with pHs 5.84 and 6.30 (slightly acidic), respectively, both being recorded below the TZS [25] permissible limit of 1.0 µg/g.

Similar to the present study, studies [21, 26, 27] also recorded the concentration of Cd at below the FAO/WHO permissible limit in cauliflowers, cabbages, onions and tomatoes. On the other hand, Ray et al. [28] conducted studies in non-polluted areas and recorded high concentrations of Cd (1.1 µg/g-dw) in cauliflower, cabbage and tomatoes, than in their respective soils (0.014 µg/g-dw to 0.721 µg/g-dw).

3.2.2. Copper

Copper is an essential micronutrient element, which functions as a biocatalysts, required for body pigmentation, maintain a healthy central nervous system, prevents anemia and interrelated with the function of Zn and iron in the body [29]. The FAO/WHO [30] acceptable limit of Cu for human consumption in vegetables is (5 µg/g). When copper exceeds its safe concentration, it causes hypertension, sporadic fever
In this study the concentration of Cu varied between (0.06 µg/g-dw) in cauliflowers at Mgeta and (7.19 µg/g-dw) in onions at Kihonda (Fig. 2). The concentrations of Cu in vegetables and tomatoes were recorded below the FAO/WHO [30] permissible limit of 5 µg/g, except in onions at Kihonda (7.19 µg/g-dw) and carrots at Towelo (6.11 µg/g-dw). The mean concentration of Cu in vegetables and tomatoes was 1.80 ± 2.80 µg/g-dw. The level of Cu was high in vegetables and tomatoes compared to soil, indicating uptake to vegetables.

The results of Cu (7.19 µg/g-dw) in onion at Kihonda could have been contributed by industrial effluents and indiscriminate disposal of domestic or sewages directed to the stream of Ngerengere, which are untreated or partially treated [16]. At Towelo, where there are no industrial activities apart from horticultural activities, the concentration of Cu could have been contributed by the nature of rocks, fertilizer, atmospheric deposition and other sources [22].

The concentrations of Cu in soils from the respective locations ranged between 0.08 µg/g-dw at Mgeta and 1.5 µg/g-dw at Towelo. All Cu levels were below TZS [25] permissible limit of 200 µg/g. The mildly acidic to nearly neutral pH values of soil (5.65-7.21) could have contributed to the heavy metals uptake of vegetables from the soil at Kihonda and Towelo. The mean concentration was (0.436± 0.28). Unpaired t-test of the concentration of Cu in vegetables, tomatoes and soils indicated no significant differences (t =1.15, P = 0.269).

Similar studies [16, 27, 31, 32] recorded the concentration of copper below permissible limit in cabbages, carrots and tomatoes. With exception of onions at Kihonda (7.19 µg/g-dw) and carrots at Towelo (6.11 µg/g-dw), vegetables are safe from Cu contamination in both areas. The availability of Cu in soil is linked with soil pH. Malik et al. [33] has found that increased soil pH may also reduce the availability of Cu to vegetables through increased adsorption at cation exchange sites.

The concentration of Cr in vegetables and tomatoes ranged between (0.38 µg/g-dw) in cabbages at Mgeta and (20.40 µg/g-dw) in onion at Kihonda (Fig. 3). The results were above the FAO/WHO [30] permissible limit (0.06 µg/g). Health effects of consuming Cr contaminated vegetables include kidney and liver damage, skin rashes, stomach upset and ulcer, respiratory problems and lung cancer and alteration of genetic materials. The mean concentration of Cr in vegetables and tomatoes was 4.42 ± 6.71 µg/g-dw.

The respective soils recorded Cr between (0.07 µg/g-dw) and (0.53 µg/g-dw), all values being below TZS [25] permissible limit of 100 µg/g. The mean concentration recorded as 0.193 ± 0.185 µg/g-dw and unpaired t-test indicated the concentration in vegetables, tomatoes and soil was statistically not showing significant differences (t = 0.291, P = 0.774). BCF ratio recorded above 1, indicating the uptake of heavy metals to vegetables [19]. Variation of soil pH between mildly acidic (pH 5.53) and moderately alkaline (pH 7.22) could have facilitated Cr solubility and availability for absorption by vegetables [19, 34].

Similar studies by Alamin et al. [35] indicated higher Cr concentrations in cabbages (1.39 µg/g-dw). Ray et al. [28] studies in non-polluted areas recorded Cr (0.03 µg/g-dw) in cauliflower and (2.45 µg/g-dw) in cabbage with respective concentration in soil of cauliflowers (0.016 µg/g-dw) and cabbages (2.947 µg/g-dw).

The concentration of Pb ranged between non-detectable limit in tomatoes, cabbages, and cauliflowers at Mgeta and (18.17 µg/g-dw) in tomatoes at Mgeta sokoni (Fig. 4). The mean concentration for these vegetables and tomatoes being 6.20 ± 7.05 µg/g-dw. With exception of the non-detectable limits of Pb in some vegetables and tomatoes which are safe for consumption, all the other concentrations recorded for Pb were above FAO/WHO [30] permissible limit (0.3 µg/g). Consumption of these vegetables would certainly result in health problems such as cognitive dysfunction, neurobehavioral disorders, hypertension and renal impairment [36].

High levels of Pb (7.19 µg/g-dw and 18.17 µg/g-dw) at Mgeta sokoni in vegetables and tomatoes could be contributed by high traffic of vehicles loading vegetables to Dar es Salaam and other regions. Most of vegetables marketed outside Morogoro, are loaded at Mgeta sokoni.

In the respective soils levels of Pb were below detection limits.
limit in all locations except at Towelo (1.16 µg/g-dw) and at Mgeta (0.16 µg/g-dw), values which are below TZS [25] permissible limit (200 µg/g).

A study by Al-Chaarani et al. [21] reported Pb concentrations in vegetables ranged from non-detectable to 2.695 µg/g-dw while the concentration of Pb reported by Naser et al. [6] in cauliflower ranged between 1.027 and 1.968 µg/g-dw (tomatoes) and between 0.486 and 1.119 µg/g-dw (cauliflower). Moreover, Bhuuto et al. [26] reported that Pb concentration in onions was 0.022 µg/g-dw and in cabbages, cauliflowers and tomatoes ranged from (0.009 µg/g-dw) to (0.084 µg/g-dw). Similarly, Ray et al. [28] recorded the range of Pb concentration in soil was between (0.74 µg/g-dw) and (3.88 µg/g-dw). Pb levels measured in vegetables and tomatoes in this study were higher than the above reported values. These values can easily add up to exceed the maximum allowable values especially in the case of children. This becomes even more pronounced knowing that food is not the only source of Pb intake into the human body [24]. The soil pH recorded (mildly acidic to mild alkaline) contributed to increasing heavy metal solubility and uptake into vegetables and tomatoes.

3.2.5. Zinc

Zn is the least toxic among all heavy metals, and is an essential element in the human diet as it is required to maintain the proper functioning of the immune system, normal brain activity and is fundamental in the growth and development of the foetus. Excessive Zn in the diet e.g. prolonged daily intake of zinc ranging at 150 - 450 mg/day is also detrimental to human health [37].

In the present study, Zn was recorded high in onions, tomatoes, cabbages and cauliflowers. It ranged between 27.40 and 106 µg/g-dw (Fig. 5) values which were above the FAO/WHO [30] permissible limit of 5 µg/g. The mean concentration recorded was (51.90 ± 25.40 µg/g-dw). Similar studies [27, 32] also reported levels of Zn in onions, carrots, cabbages, and tomatoes above permissible levels of FAO/WHO.

The soil samples recorded Zn levels between (1.38 µg/g-dw) and (24.80 µg/g-dw), that were lower than in the respective vegetables and lower than the TZS [25] limit. The mean concentration recorded was 7.59 ± 8.81 µg/g-dw. Unpaired t-test indicated statistically significant differences of Zn in vegetables, tomatoes and soil (t = 4.39, P = 0.001). Since there was no major industry existing in the study areas, it was assumed that the sources of Zn were probably from motor vehicle tire rubber exacerbated by poor road surfaces and the lubricating oils in which Zn is found as part of many additives such as zinc dithiophosphates [38].

3.3. Bioconcentration Factors (BCF) of Heavy Metals

The BCF of above 1.0 indicates higher uptake of heavy metals in vegetables than in the soil. An area which recorded BCF below 1.0 indicates high heavy metals concentration in soil in relation to vegetables and therefore low uptake of heavy metals to vegetables [39]. The calculated BCFs for the heavy metals in Morogoro region are presented in Table 2. The BCFs of Cd, Cu, Cd, Pb and Zn for all vegetables and at all locations were found to be, in general, above one (1.0) except for Cu which recorded one (1.0) for tomatoes and cabbage at Towelo and below one (<1.0) for tomatoes and cauliflowers from Mgeta. Cd (onions) at Mgeta as well recorded below one (<1.0). Such high BCF values suggest that these leafy vegetables and tomatoes should not be eaten raw.

The overall BCF observation of heavy metals showed that BCF values of Cr and Zn were the highest as compared to Cu, Cd and Pb values from all locations.

Table 2. Bioconcentration Factor (BCF) for samples from Morogoro.

| Location (vegetables) | BCF for heavy metals | Pb |
|-----------------------|----------------------|----|
|                       | Cu                  | Cr  | Zn  | Cd   |
| Kihonda (Onions)      | 7.41                | 70.4 | 1.23 | *   |
| Mgeta (Tomatoes)      | 0.19                | 6.50 | 3.64 | 0.043 | *   |
| Mgeta (Cabbages)      | 1.75                | 6.75 | 6.28 | *   |
| Mgeta (Cauliflowers)  | 0.43                | *    | 31.94 | *   |
| Mgeta sokoni (Onions) | **                  | **   | **   | **   |
| Mgeta sokoni (Tomatoes)| **                | **   | **   | **   |
| Towelo (Tomatoes)     | 1                   | 10.14 | 61.63 | *   |
| Towelo (Cabbages)     | 24.44               | 2.21 | 31.73 | 1.06 |
| Towelo (Cabbage)      | 1                   | 116  | 23.06 | *   |

* Soil/vegetable ratio was not calculated, one of the results was below detection limit
** Areas where soil was not sampled.
4. Conclusion

The results reported in the present study show that concentration of heavy metals Cd, Cu, Cr, Pb and Zn in the soil from all nine locations were recorded as below the TZS permissible limits for these metals. The results also indicate that levels of these heavy metals in vegetables and tomatoes varied from below detection limits to well above of the FAO/WHO permissible limits in foods [30]. The mean concentrations of these heavy metals (except Cu) in vegetables and tomatoes were also observed to be above the FAO/WHO limits [30] and were therefore concluded as not safe for human consumption. Cu was the only heavy metal detected at levels allowable for human consumption. Zn was present at the highest concentration as compared to the other metals. Although most of the studied area had no industries, it was statistically observed that there was significant difference for Zn content between the studied vegetables and soils. The heavy metals recorded levels above the FAO/WHO permissible limits in vegetables and tomatoes [30] and below the TZS permissible limits in soil [25].

The BCF for Cr, Pb, Cd, Cu and Zn in studied vegetables and tomatoes except for Cd (onions) at Kihonda and Cu (tomatoes and Cauliflowers) at Mgeta, were found to be above 1 indicating high uptake of heavy metals in the vegetables and tomatoes from the soil. Two sites at Towelo for Cu (tomatoes and cabbages) had BCF lower than 1 indicating that Cu was in balanced proportions between the soil and the vegetables. Overall results suggest that vegetables and tomatoes grown within the study area might place the consumer at health risk, therefore they are not safe for consumption in their raw state.

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