

Determination of Cd, Mn and Ni accumulated in fruits, vegetables and soil in the Thohoyandou town area, South Africa

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The accumulation of heavy metals such as Cd, Mn and Ni was investigated in seven different vegetables, fruits and soil samples from Thohoyandou, Limpopo Province, South Africa. Heavy metals were quantified using graphite furnace atomic absorption spectrometry. Concentrations of heavy metals in fruits and vegetables were in the range of 0.23–2.94 mg·kg⁻¹ for Cd, 11.72–50.16 mg·kg⁻¹ for Mn and 5.73–44.11 mg·kg⁻¹ for Ni on a dry weight basis. Analysis of soils from where fruits and vegetables were sampled showed that Cd in the soil was in the range of 0.08–1.07 mg·kg⁻¹, Mn levels were 204.99–249.13 mg·kg⁻¹ and Ni levels were 48.47–88.23 mg·kg⁻¹. Cd was below the instrument detection limit for soils on which onions and bananas were grown. Vegetables showed different accumulation abilities, with leafy vegetables being the highest accumulators of heavy metals. The obtained results showed that concentrations of Cd in fruits, vegetables and soils exceeded the recommended maximum acceptable levels proposed by FAO/WHO and, hence, may pose a health risk to consumers. Ni concentrations in bananas, onion, beetroot, spinach and Chinese cabbage exceeded recommended standards by FAO/WHO.

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INTRODUCTION

Food contamination by heavy metals is commonly due to environmental and industrial contamination from sources such as industrial emissions and irrigation water (Huang et al., 2014). Dust emission from cement production is among the anthropogenic activities that contribute to environmental pollution (Bermudez et al., 2010). Heavy metals, particulates and dioxins are some pollutants that are contained in cement dust, which may pose a health risk to humans (Tajudeen et al., 2011). Soils and plants serve as sinks for atmospheric deposition of heavy metals from industrial emissions (Bermudez et al., 2010; Hao et al., 2009; Hernández-Martínez and Navarro-Blasco, 2012). The accumulation of cement dust in soils and on plants may be a result of wind and seepage waters (Taghipour et al., 2013; Li et al., 2015; Xu et al., 2014). Heavy metals that may be contained in cement dust include As, Cd, Pb, Hg, Tl, Al, Be, Cr, Cu, Mn, Ni, and Zn (Schuhmacher et al., 2002; Engelbrecht et al., 2013; Ogunbileje et al., 2013).

Excessive accumulation of these heavy metals in agricultural soils is a result of phytotoxicity and elevated heavy metal uptake by food crops, hence causing food insecurity (Kabata-Pendias and Mukherjee, 2007; Nagajyoti et al., 2010). Furthermore, the potential of heavy metals to bioaccumulate in the food chain has led to health concerns. Excessive bioaccumulation of toxic heavy metals in vegetables may result in the unavailability of dietary nutrients to humans or cause health problems for both humans and the ecosystem (Ogunkunle et al., 2013; Wuana and Okieimen, 2011; Hu et al., 2013; Yang et al., 2009). Moreover, cement dust deposition on plants can cause stomatal clogging and thereby affect plant growth (Abdel-Rahman and Ibrahim, 2012; Prajapati and Tripathi, 2008).

Non-biodegradability, long biological half-lives and their persistent nature make heavy metals harmful (Arora et al., 2008; Shalini et al., 2017). Consumption of unsafe concentrations of heavy metals continuously through food may lead to chronic accumulation of heavy metals in the human kidney and liver, consequently disrupting numerous biochemical processes and leading to cardiovascular, nerve, kidney and bone diseases (Zhou et al., 2016; Sharma et al., 2009).

Exposure to chronic Cd may trigger acute liver and lung toxicity, induce nephrotoxicity and osteotoxicity, and impair the functions of the immune system (Klaassen et al., 2009; Patrick, 2003). Clinical symptoms such as nausea, vomiting, abdominal discomfort, diarrhoea, visual disturbance, headache, giddiness and cough indicate acute health impacts of high concentrations of Ni (Duda-Chodak and Blaszczyk, 2008).

Vegetables are a rich source of vitamins, minerals and fibre, whereas fruits are a rich source of carbohydrates, proteins, vitamins, minerals and fibre, which are essential for good human health (Cherfi et al., 2014). Zn, Cu, Mn, Ni and Co are essential heavy metals that might be contained in fruits and vegetables. However, they can be toxic when their concentrations exceed the tolerable limit in living organisms. Non-essential heavy metals such as Hg, Pb, As, Cr and Cd are toxic to humans even at low concentrations (Izah et al., 2016).

In this study, the presence of Mn, Cd and Ni was investigated in soil, fruits and vegetables from a small-scale farm in the vicinity of a cement brick manufacturing company in Thohoyandou, South Africa. The close proximity of this farm to the cement brick company might result in the contamination of soil, vegetables and fruits with heavy metals from cement dust. Moreover, this small-scale farm is close to one of the busiest roads in Thohoyandou. There have been few studies that have focused on heavy metal contamination of fruits and vegetables in South Africa (Bvenura and Afolayan, 2012; Kisten et al., 2017). Furthermore, none of the previous studies examined heavy metals in soil, vegetables and fruits simultaneously in Thohoyandou. Monitoring levels of heavy metals can provide useful information for promoting food safety in South African food industries and setting national standard limits since there are presently none.

EXPERIMENTAL

Chemicals

Nitric acid, hydrochloric acid, manganese sulphate, cadmium sulphate and nickel sulphate analytical reagents were purchased from Merck (Johannesburg, South Africa). Polyethylene bags and all the glassware were purchased from Lasec (Johannesburg, South Africa).

Instruments and materials

Heavy metal concentrations were analysed using a graphite furnace atomic absorption spectrometer (GFAAS) technique (Perkin Elmer Model Pinnacle 900T, Perkin Elmer, Germany), fully automated and PC-controlled using Syngistix AA. A Mars 5 microwave assisted digestion system (CEM Corporation, USA) was used for the digestion of fruits and vegetables. A Restch grinder and mesh sieves of 2 mm, 1 mm, 500 µm, 250 µm and 75 µm sizes were purchased from Retsch GmbH (Haan, Germany). The pH and electrical conductivity (EC) of soil samples were measured using a portable multi-probe Boeco pH meter that was purchased from Rochelle (Johannesburg, South Africa).

Study area

Thohoyandou is in the province of Limpopo in South Africa. It is an administrative centre of the Vhembe District Municipality and Thulamela Local Municipality. Daily temperatures in the town vary between 20°C and 40°C in wet seasons and 12°C and 22°C in dry seasons. The average annual rainfall in the town is approximately 800 mm, but ranges between 340 mm and 2 000 mm. In summer and the winter months, the prevailing wind direction is east to southeast. The average wind speed is 11 km·h⁻¹ in summer and 15 km·h⁻¹ in winter (Mzezewa et al., 2010). The town is in an urbanization and development stage, with a modern shopping mall being the most recent large development. There are also houses being built since the town is expanding. There are two brick-making companies which supply building materials and which are situated on the western part of town. The company which is the possible source of heavy metals to the site of interest is about 1 km away from the sampling site in this study. Figure 1 shows the sampling area and the sampling points (indicated by red circles).

Sample collection

A sampling method described by Zhou et al. (2016) and Sharma et al. (2009) was used. Bananas/*Musa acuminata* and 7 vegetable samples of different vegetable species (spinach/*Spinacia oleracea*, Chinese cabbage/*Brassica rapa*, onion/*Allium cepa*, beetroot/*Beta vulgaris*, sweet potatoes/*Ipomoea batatas*, tomatoes/*Lycopersicon esculentum* and cabbage/*Brassica pekinensis*) were collected from a small-scale farm close to a brick-making company in Thohoyandou, using the random sampling method. Tomatoes are a solanaceous vegetable species, onion is an allium vegetable, and sweet potatoes and beetroot are root vegetables, whereas Chinese cabbage, spinach and cabbage are leafy vegetables. All samples were stored at a constant temperature of 4°C in polyethylene bags for transport. Soil samples were collected from the upper soil layer (0–20 cm) in the same location where vegetables were sampled, using a stainless-steel spade. Polyethylene bags were used to store samples for transport.

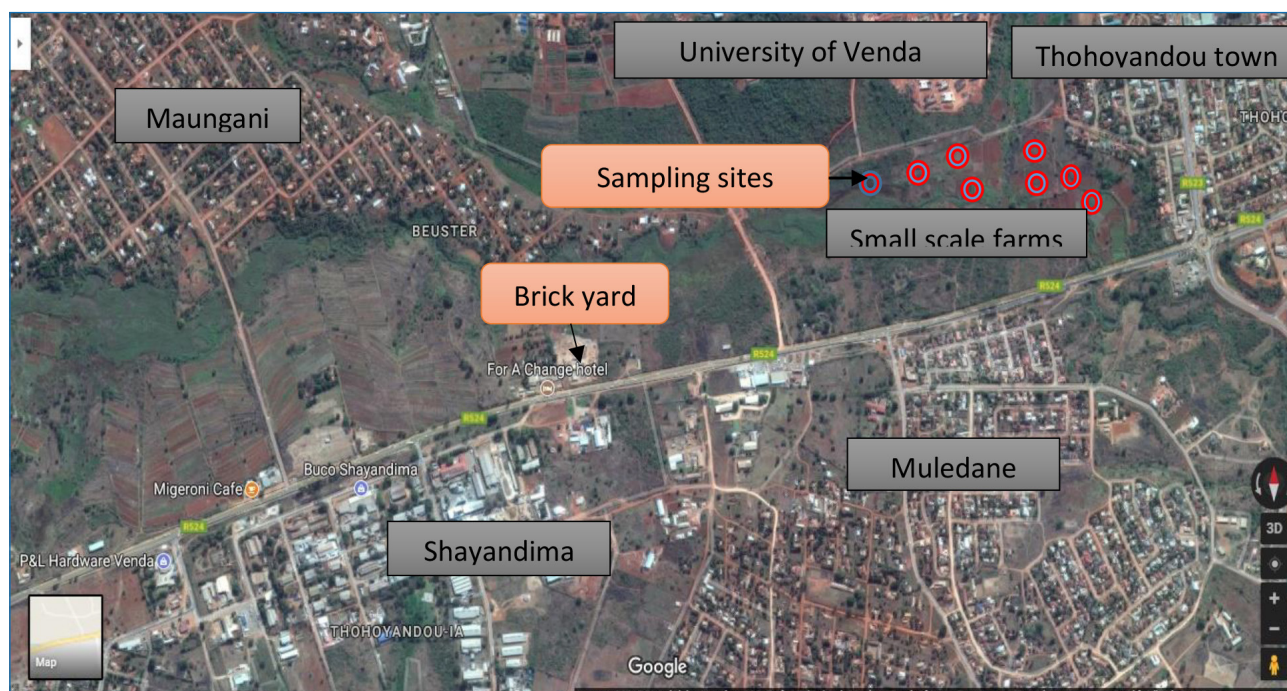


Figure 1. Sampling area showing a brick-making company and the nearby small-scale farms; sampling points shown as red circles

Sample preparation

Fruit and vegetable samples were cleaned with deionised water to remove dust and soil. The edible parts of the vegetables were separated from the plants, chopped into small pieces and air-dried to constant mass. A Restch grinder was used to grind all samples to fine powder; the samples were passed through a series of sieves and the 75 µm fraction was used for analysis. Approximately, 0.2 g of each sample was transferred into a Teflon vessel and digested in 12 mL of HNO₃ using a Mars-5 microwave-assisted digestion system according to the programme shown in Table 1. The resulting solutions were filtered using Whatman No. 42 filter papers into 50 mL volumetric flasks and filled to the mark with deionised water and then analysed for concentrations of Mn, Cd and Ni using a graphite furnace atomic absorption spectrometer. Standard solutions of the three elements under study were prepared. White clover certified reference material (CRM) (BCR-402) was used for quality assurance. Hollow cathode lamps of Mn, Cd and Ni at wavelengths of 193.7, 228.8 and 279.49 nm, respectively, were used to do measurements.

Soil samples were air-dried at room temperature to constant mass and were passed through a 75 µm sieve to eliminate plant roots and other waste materials and stored in sealable plastic bags until analysis. The pH and electrical conductivity (EC) of the soil slurry were measured with a pH multi-meter at 1:5 (w/v) ratio soil to water. For the analysis of the total concentrations of soil metals, approximately, 0.2 g of each sample was weighed into a 250 mL beaker and digested using aqua regia (5 mL HNO₃ and 15 mL HCl). The mixture was heated for 3 h on a hot plate to near-complete evaporation; then 20 mL of 2% (v/v) HNO₃

was added into the beaker. The solution was filtered through Whatman No. 42 filter paper into a 100 mL volumetric flask and filled to the mark using deionised water. The samples were analysed using the GFAAS.

RESULTS AND DISCUSSION

Chemical properties of soil

Table 2 shows some chemical properties of the composite soil sample. The pH of the composite soil sample was 6.73 indicating that the soil was near neutral pH. Soil acidification has the effect of reducing the supply of nutrients and increasing the dissolution of heavy metals such as Mn and Cd and hence increasing their absorption by plants (Dorraj et al., 2010). The pH of the soil is a critical factor in controlling the bioavailability of trace elements, especially for Cd (Adriano, 2001; Kabata-Pendias and Pendias, 2001). The soil electrical conductivity was 95.3 mS·m⁻¹. The soil in this study was clayey, hence it has the ability to store and bind cations. X-ray fluorescence (XRF) results showed that the composite sample contained 87.1 mg·kg⁻¹ of Ni which was above the standard value as stipulated by FAO/WHO. Cd was not detected.

Quality assurance

White clover CRM (BCR - 402) from the Community Bureau of Reference of the Commission of the European Communities was analysed for quality assurance purposes. The obtained results were compared with the certified value (Table 3). Measured values of Ni compared well with a certified value of 8.25 mg kg⁻¹.

Table 1. Digestion programme for the vegetables and fruits with a microwave-assisted acid digestion system

Stage	Power (%)	Ramp time (min)	Pressure (psi)	Temperature (°C)	Hold time (min)
1	100	4	800	180	8
2	100	5	800	180	5

Table 2. Chemical properties of soil

Sample name	pH	EC (mS·m ⁻¹)	Mn (mg·kg ⁻¹)	Ni (mg·kg ⁻¹)	Cd (mg·kg ⁻¹)
Composite	6.73	95.3	–	87.1	0
Standard value in soil*	–	–	–	50	0.3

– not available, *FAO/WHO, 2011

Table 3. Concentrations of heavy metals in edible parts of fruits and vegetables (mg·kg⁻¹ dry weight) from a small-scale farm in Thohoyandou

Sample name	Cd	Mn	Ni
Maximum permissible limit† (mg·kg ⁻¹)	0.05 (fruits) † 0.05 (vegetables) *	NA (fruits) NA (vegetables)	0.80 (fruits) † 10.00 (vegetables)*
Cabbage green outer leaves	0.29	13.68	5.73
Cabbage inner layer leaves	0.54	22.58	6.55
Onion leaves	0.74	30.65	12.51
Onion bulb	0.91	22.65	10.84
Spinach	2.94	50.16	44.12
Chinese cabbage	0.77	31.78	11.38
Beetroot	1.08	29.95	19.07
Sweet potatoes	0.57	23.92	9.34
Tomatoes	0.60	15.75	7.97
Bananas	0.23	11.72	8.54
CRM (White clover)	–	–	8.16

ND – not detected; NA – not available; †FAO/WHO, 2002; *FAO/WHO, 2011

Table 4. Concentrations of heavy metals in soil collected under the mentioned fruit/vegetable (mg·kg⁻¹ dry weight)

Sample name	Cd	Mn	Ni
Maximum permissible limit† (mg·kg ⁻¹)	0.30*	N/A	50.00*
Cabbage	0.20	211.81	48.47
Onion	ND	223.26	74.71
Spinach	1.07	229.13	88.23
Chinese cabbage	0.03	204.99	68.83
Beetroot	0.08	210.14	83.50
Sweet potatoes	0.12	230.27	80.20
Tomatoes	0.32	245.13	74.32
Bananas	ND	249.13	66.52

ND – not detected, *FAO/WHO, 2011

Heavy metal concentrations in edible parts of fruits and vegetables

The concentrations of Cd, Mn and Ni found in the fruits and vegetables are shown in Table 3. The range of concentrations of heavy metals in fruits and vegetables was in the order Mn > Ni > Cd. The same trend was obtained by Kisten et al. (2017) in vegetables. The obtained concentration ranges were 0.23–2.94 mg·kg⁻¹, 11.72–50.16 mg·kg⁻¹ and 5.73–44.11 mg·kg⁻¹ for Cd, Mn and Ni, respectively. The concentration of Cd in fruits and vegetables exceeded the recommended maximum acceptable levels proposed by FAO/WHO (2002 and 2011). Ni concentrations in bananas, onion, beetroot, spinach and Chinese cabbage exceeded recommended standards by FAO/WHO (2002 and 2011). The lowest concentrations of Cd, Mn and Ni were obtained in bananas and cabbage green outer leaves, whereas the highest concentrations were found in the spinach. Generally, vegetable species differ in their ability to take up and accumulate heavy metals (Saumel et al., 2012). The accumulation of Cd in vegetable species decreased in the order of leafy vegetables > root vegetables > allium vegetables > solanaceous vegetables. Onion leaves showed a higher accumulation of Cd compared to the onion bulb. The concentrations of Mn were in descending order, leafy vegetables (spinach, Chinese cabbage, cabbage inner layer leaves) > allium vegetables (onion leaves) > root vegetables (beetroot and sweet potatoes) > solanaceous (tomatoes) > fruit (bananas). This trend was similar to that found by Zhou et al. (2016). However, the concentration of cabbage outer green leaves was lower compared to the rest of the leafy vegetables. The highest concentration of Ni was observed in spinach (leafy vegetable). However, there is no clear trend for the rest of the vegetables according to their species. The interception of heavy metals emitted in the atmosphere by leaves might result in high levels of heavy metals in leafy vegetables. These heavy metals may remain on the leaf surface or enter leaf tissues, even though metal contents in leaf tissues can also be a result of selective metal uptake by roots (Maisto et al., 2013). The exposed surface area of the leaves may influence aerial dust deposition (Prajapati, 2002).

Results of this study are in agreement with the results obtained by Ali and Al-Qahtaini (2012), who reported that Mn and Cd concentrations ranged from 4.16–94.16 mg·kg⁻¹ and 0.92–4.13 mg·kg⁻¹, respectively, in different vegetables. The concentrations of Cd and Mn in cabbage, onion, spinach and tomatoes correspond to the results reported by Bvenura and Afolayan (2012), except that the Cd concentration in spinach samples in this study was higher. In a more recent study by Shaheen et al. (2016), a concentration of 0.05 mg·kg⁻¹ of Cd in tomatoes was reported which was above the recommended standard limit by FAO/WHO. The high concentration levels of heavy metals might be due to atmospheric deposition of contaminated dust on the

leaves. Cement dust from the brick-making company might be responsible for the presence of heavy metals in vegetables and fruits. Cement dust may be carried by wind and deposited on the vegetables and the soil. Automobiles might also be a source of heavy metal contamination. The sampling site is less than 1 km away from one of the busiest roads that connect Thohoyandou and the town of Louis Trichardt.

Heavy metal concentrations in soil

The concentrations of Cd, Mn and Ni in soil samples collected where fruits and vegetables were grown are shown in Table 4. The concentrations of Cd, Mn and Ni ranged from 0.03–1.07 mg·kg⁻¹, 204.99–249.13 mg·kg⁻¹ and 48.47–88.23 mg·kg⁻¹, respectively. The concentrations of heavy metals in soils were in the order Mn > Ni > Cd; the same order applied to the samples of fruits and vegetables. Cd was below instrumental detection limits for soils where onions and bananas were grown. The Cd concentrations of soils where tomatoes and spinach were grown were above FAO/WHO standards. The concentration of Ni was below the FAO/WHO standard only for the soil where cabbage was grown. The concentrations of Cd obtained in this study were similar to the results obtained by Liu et al. (2015) in which Cd concentrations ranged between 0.0541 and 0.8487 mg·kg⁻¹ in vegetable soils. The concentrations of Cd and Mn were comparable to the results obtained by Bvenura and Afolayan (2012), although the concentration of Cd in the soil where spinach was grown was higher. However, Mn concentrations obtained by Bvenura and Afolayan (2012) are slightly higher and ranged between 377.61 mg·kg⁻¹ and 499.68 mg·kg⁻¹.

Phosphate fertilizers are a major source of heavy metals that enter agricultural soils, particularly Cd (Nicholson et al., 2003). Pollution related to traffic is another source of Cd, primarily caused by the aging and wear of automobile tyres, gasoline and car body and brake lining wear (Weckwerth, 2001).

Bioaccumulation factor in fruits and vegetables

The bioaccumulation factor (BF) can be used to estimate the ability of plants to accumulate heavy metals in their edible tissues. The bioaccumulation factor was calculated using the following equation.

$$\text{Bioaccumulation factor (BF)} = \frac{C_{\text{plant}}}{C_{\text{soil}}} \quad (1)$$

where C_{plant} is the heavy metal concentration in edible tissues of a plant and C_{soil} is the heavy metal concentration in the soil.

The bioaccumulation factors for Cd, Mn and Ni were 1.45–25.67, 0.05–0.22 and 0.11–0.50, respectively (Table 5). Cd has the

Table 5. Bioaccumulation factors of fruits and vegetables from a small-scale farm in Thohoyandou

Sample name	Cd	Mn	Ni
Cabbage green outer leaves	1.45	0.06	0.12
Cabbage inner layer leaves	2.70	0.11	0.14
Onion leaves	–	0.14	0.17
Onion bulb	–	0.10	0.15
Spinach	2.75	0.22	0.50
Chinese cabbage	25.67	0.15	0.16
Beetroot	13.33	0.14	0.23
Sweet potatoes	7.58	0.10	0.12
Tomatoes	1.88	0.06	0.11
Bananas	–	0.05	0.13

– not available

highest BF, hence the uptake of Cd by vegetables is higher than for Mn and Ni. The trend of the bioaccumulation factor for Mn and Ni is similar to the trend of the concentrations of these heavy metals in fruits and vegetables, with spinach having the highest BF for both heavy metals, showing that leafy vegetables have a greater ability to bioaccumulate heavy metals. Chinese cabbage (leafy vegetable) had the highest bioaccumulation factor for Cd and this corresponds to the findings by Zhou et al. (2016). The strong ability to accumulate Cd by all the vegetables might be due to acidity of the soil and water used for irrigation. Acidity increases the solubility of heavy metals, hence making them readily available for absorption by plants.

CONCLUSION

The concentration of heavy metals in fruits and vegetables was in the order Mn > Ni > Cd. The obtained results showed that concentrations of Cd in fruits, vegetables and soils exceeded the recommended maximum acceptable levels proposed by FAO/WHO and hence may pose a risk to public health. The concentrations of Ni in bananas, onion, beetroot, spinach and Chinese cabbage exceeded recommended standards by FAO/WHO. Vegetables showed different heavy metal accumulation abilities, with leafy vegetables being the highest accumulators of heavy metals. Heavy metal uptake and accumulation was high for leafy vegetables and low for tomatoes (solanaceous vegetables) and bananas.

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