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Assessment of Heavy Metal Contamination in Vegetables Grown Using Paper Mill Wastewater in Wonji Gefersa, Ethiopia

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Abstract Heavy metals are among the major contaminants of vegetables. A study was conducted at Wonji Gefersa farms where paper wastewater is used for cultivation of vegetable crops. Four vegetable samples, namely Swiss chard, carrot, tomato, green pepper, as well as paper wastewater were examined for heavy metal [Lead (Pb), Zinc (Zn), Cadmium (Cd), Iron (Fe), Copper (Cu), Chromium (Cr) and Cobalt (Co)] contamination using atomic absorption spectroscopy. The levels of Pb, Cd and Cr in paper wastewater were all above the safe limit for FAO standards for wastewater quality for irrigation. The concentration of Pb in Swiss chard and Green peeper was exceeded the permissible limits. The study reveals that Pb metal contamination in the study area which poses health risk with time unless an urgent step is taken by relevant agencies to address this issue.

Keywords Vegetables · Contamination · Concentration · Wastewater · Irrigation · Health risk

Use of wastewater to irrigate agricultural lands is one of the common practices in suburban and industrial areas in many parts around the world (Sharma et al. 2007). Waste water irrigation leads to accumulation of heavy metals into the soil (Singh et al. 2010). Industrial wastewater has been implicated as a potential source of heavy metals such as Copper (Cu), Cadmium (Cd), Zinc (Zn), Lead (Pb), Nickel (Ni) and Iron (Fe) in the edible and non-edible parts of vegetables (Sharma et al. 2006). Food-safety issues and possible health risks make this as one of the most serious environmental concerns.

Wastewater carries appreciable amounts of trace toxic metals which often lead to degradation of soil health and contamination of a food chain, mainly through the vegetable grown on such soils (Rattan et al. 2002). The toxic elements accumulated in the organic matter in soils are taken up by growing plants and lastly exposing humans to this contamination (Khan et al. 2008).

Toxic heavy metals entering the ecosystem may lead to bioaccumulation, particularly by eating fruits and vegetables (Kashif et al. 2009). This may cause an excessive build-up of heavy metals in the body. Some heavy metals that are most often found to be responsible for harmful damage to humans are Pb, Cd, Cr, Co and Ni (Gupta et al. 2008). Some heavy metals such as copper, iron, zinc and manganese, are necessary to the body but in case of overexposure, they can lead to heavy metal toxicity symptoms. Heavy metal concentrations vary among different vegetables, which may be attributed to a differential absorption capacity of vegetables for different heavy metals (Singh et al. 2010).

Heavy metals are among the major contaminants of vegetables. They are not biodegradable, have been long biological half-lives and have the potential for accumulation in the different body organs leading to unwanted effects (Nabulo et al. 2011; Singh et al. 2010).

In view of persistent nature and cumulative behavior as well as the consumption of vegetables and fruits, there is a need to test and analyze food items to ensure that the levels of these contaminants meet the agreed international requirements. Regular survey and monitoring programmes of the concentration of heavy metals in food products have

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been carried out for decades in most developed countries (Sobukola et al. 2010). However, in developing countries like Ethiopia, limited data are available on heavy metals in food products. Some data have been reported for leafy vegetables (Itanna 2002).

In Ethiopia, the quality of water is increasingly threatened due to human population growth and expansion of different industries. Studies indicated that all types of domestic wastewater and more than 90% of the industries in the country discharge their effluents without any form of treatments into the surrounding agricultural farms and streams (AAEPA 2007). These practices cause both environmental, health and economic burden in the country.

Wonji paper mill industry mainly uses different dyes for paper color printing and different chemicals for processing a paper. The wastewater after processing directly drains to the nearby agricultural lands without any treatment. The paper wastewater was being used for the last 6 years to grow different vegetables. The wastewater contains heavy metals and organics, which have a chance of accumulation on vegetables that growing in the vicinity of the industry. Most of these vegetables cultivated in this site are supplied to the wholesale vegetable market in Addis Ababa, Adama and the rest enter to the nearby community with cost effective.

Although there was a study related to vegetable contamination by heavy metal using Awash River as irrigation in the region (Benti 2014), there is limited research on vegetable contamination by heavy metal using paper wastewater as irrigation. The aim of this study was therefore, to evaluate the contamination level of heavy metals in different vegetable grown using paper wastewater.

Materials and Methods

The study area, Wonji Gefersa town, is located within Ethiopia, in the Eastern Shoa zone of Oromia region, South of Adama city. It is located at 8° 27′ North and 39° 17′ East with an elevation of 1588 m above sea level. The mean annual rainfall is 831 mm, with mean annual maximum and minimum temperatures of 27 and 15°C, respectively.

Triplicate samples of paper wastewater and composite samples of four vegetables [Swiss chard (*Beta Vulgaris L. var. cicla*), Carrot (*Daucus carota L.*), Tomato (*Lycopersicon esculentum*), and Green pepper (*Capsicum annum*)] were collected in triplicate from the farm during the months from May to July 2014. Moreover, control water samples were collected in triplicate before the river water entered to the paper industry and composite control vegetable samples were also collected in triplicate from nearby agricultural farm, which irrigated with Awash River. A total of 24 vegetable samples and 6 water samples were collected monthly during the study period.

Wastewater samples were collected in 500 mL plastic bottle where the wastewater diverted to vegetable farms. 5 mL of HNO_3 was added to plastic bottle prior to sample collection to prevent microbial degradation of heavy metals. 50 mL of wastewater samples were digested with 10 mL concentrated HNO_3 at 80°C (APHA 1985). The freshly harvested vegetables were brought to the laboratory and washed primarily with running tap water, followed by three consecutive washings with distilled water to remove soil particles. Samples were cut to small pieces using clean knife and dried in an oven at 70°C for 48 h. The dried samples were grounded using mortar and pestle and 0.5 g of each powdered sample was weighed using the electronic balance. Samples underwent pressurized digestion with HNO_3/H_2O_2 in a high performance microwave digestion system. 0.5 g of ground plant sample was digested with 10 mL of HNO_3 and 5 mL of H_2O_2. The digestion temperature was about 180°C (Itanna 2002). The digested samples carefully transferred into 100 mL volumetric flask, rinsed and diluted with 50 mL distilled water and shaken.

Finally Cd, Pb, Zn, Cr, Cu, Fe and Co concentrations in wastewater and vegetables were analyzed by Graphite Atomic Absorption Spectrometer (nova, Model 400P, analytikjena, Germany). All reagents used were Merck, analytical grade (AR) including Standard Stock Solutions of known concentrations of different heavy metals. The analytical detection limits for Cd, Pb, Zn, Cr, Cu, Fe and Co were 0.016, 0.025, 0.01, 0.02, 0.01, 0.05 and 0.01 mg L^{-1}, respectively. Statistical analysis were performed by SPSS version 16.0 to calculate average mean, standard deviation and Pearson's correlation (r) value to show the degree of metal association in vegetables.

Results and Discussion

The concentration of heavy metal content of paper wastewater and river water used for irrigation purposes of a Wonji Gefersa irrigation scheme is shown in Table 1. The concentrations (µg/L) of heavy metals in paper wastewater ranged from 622 to 625 for Pb, 978 to 982 for Zn, 80 to 81 for Cd, 1620 to 1621.2 for Fe 115 to 116.9 for Cu and 520 to 523 for Cr. In control river water, heavy metal concentrations (µg/L) ranged from 126.9 to 128.7 for Pb, 220.5 to 221 for Zn, 8.8 to 9.6 for Cd, 430 to 431 for Fe, 101 to 102.4 for Cu and 261 to 262.1 for Cr. The concentration of heavy metal in paper wastewater was in the following order of decreasing magnitude Fe > Zn > Pb > Cr > Cu > Cd.

In comparison with the standard guideline of irrigation water (Pescod 1992) it was found that mean Pb, Cd and Cr concentrations of paper wastewater were above the safe limit while the levels of Zn, Fe, and Cu were within the recommended limit of FAO for wastewater quality for

Table 1 Heavy metal concentrations (μg/L) in paper wastewater and river water used for irrigation in Wonji Gefersa, Ethiopia

Parameter	Paper wastewater			River water (control)			Safe limit*
	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	
Pb	623.3 ± 1.5	622	625	127.6 ± 0.9	126.9	128.7	500
Zn	980 ± 2.0	978	982	220.8 ± 0.2	220.5	221	2000
Cd	80.5 ± 0.5	80	81	9.2 ± 0.4	8.8	9.6	10
Fe	1620.6 ± 0.6	1620	1621.2	430.5 ± 0.5	430	431	2000
Cu	116.1 ± 0.9	115	116.9	101.8 ± 0.7	101	102.4	200
Cr	521.5 ± 1.5	520	523	261.6 ± 0.5	261	262.1	100

n number of samples

*Source: Pescod (1992)

irrigation (Table 1). The level of chromium in the control water sample was above the safe limit of FAO standards. The reason might be at the upstream area, there is large tannery industries which Awash River receives wastewater from these industries.

Of all the heavy metals examined, concentration of Fe was highest in both paper wastewater and river water used for irrigation in the study area. The concentration of Pb, Cu and Cr in paper wastewater of the study area was higher than the levels of Pb (0.125 mg/L), Cu (0.064 mg/L), and Cr (0.05 mg/L) in paper wastewater reported in Muktsar, India (Bishnoi et al. 2006). Similarly, the concentration of Fe in the study area was higher than the level of Fe in paper wastewater reported in Lahore, Pakistan (0.156 ppm) (Chaudhry et al. 2013).

The concentration of heavy metals in the vegetables is given in Table 2. The maximum uptake of Fe was in Green pepper (569.9 μg/kg) followed by Swiss chard (368.8 μg/kg), Carrot (341.8 μg/kg) and Tomato (222.2 μg/kg), where the levels of Fe in all the vegetables were below the prescribed safe limit of FAO/WHO. The average concentrations of Fe (222.2–569.9 μg/kg) in vegetables in the present study were significantly lower than those reported in Loumbila, Burkina Faso (0.204–28.98 mg/kg) (Bambara et al. 2015).

Higher Cu concentration (179.2 μg/kg) was found in Green peppers whereas the mean value was (124.1, 88.2, 98.6 μg/kg) for Swiss chard, Carrot and Tomato respectively. The concentration of Cu in all the vegetables was below the recommended limit. The present study revealed that the mean Cu level (88.2–179.2 μg/kg) measured in vegetables from Wonji Gefersa was lower than the vegetables from Tahtay Wukro, Tigray, Ethiopia (1.93–4.10 mg/kg) (Gebrekidan et al. 2013), and Addis Ababa, Ethiopia (0.28–8.22 mg/kg) (Weldegebriel et al. 2012), but higher than the Cu concentrations (0.02–0.172 mg/kg) in vegetables from Nagodi, Ghana (Boamponsem et al. 2012).

Higher concentration of Cd was shown by Swiss chard (138.5 μg/kg) followed by Green pepper (136.7 μg/kg), Carrot (73.5 μg/kg) and Tomato (54.7 μg/kg). The study revealed that the Cd metal content was within the acceptable limit of FAO/WHO. The average concentration of Cd (54.7–138.5 μg/kg) in vegetables in this study were higher than those reported in Addis Ababa, Ethiopia (10–130 μg/kg) (Itanna 1998), and Burayu farm, Addis Ababa, Ethiopia (20–90 μg/kg) (Alemayehu et al. 2011) but lower than the average Cd content in vegetables (30–260 μg/kg) from Gondar vegetable market, Ethiopia (Rahlenbeck et al. 1999).

The concentration of Zn in Green pepper, Swiss chard, Carrot and Tomato was (121, 96, 212.2 and 259.3 μg/kg respectively). The Zn levels in all vegetable samples were within the acceptable limit of FAO/WHO. The mean concentration of Zn (96–259.3 μg/kg) in the present study was substantially lower than the Zn concentration in vegetables (5.06–10.61 mg/kg) from Accra, Ghana (Lente et al. 2014).

Maximum Pb concentration (574.7 μg/kg) was found in Swiss chard whereas the mean value was 376.5, 211.5 and 182.1 μg/kg for green pepper, tomato and carrot, respectively. The lead concentration in Swiss chard and Green peeper exceeded the permissible limit of 300 μg/kg (FAO/WHO). The present study showed that the mean Pb level (182.1–574.7 μg/kg) measured in different vegetables were higher than the vegetables from wastewater irrigated areas of Wonji Gefersa, Ethiopia (0.3–0.4 mg/kg) (Benti 2014), but it was substantially lower than the Pb content (0.21–1.79 mg/kg) of vegetables from Addis Ababa, Ethiopia (Itanna 1998). In the present study, the accumulation of elevated concentration of Pb in Swiss chard and Green pepper might be attributed to the leakage of ink effluent from paper industry to the farm. The other possible reason for the accumulation is the gas emission from the traffic that transport raw and end product paper since the vegetable growing on the roadside which traps the metal Pb.

The highest mean concentration of Cr was found in Green pepper (433.3 μg/kg) followed by Swiss chard (123.7 μg/kg), Carrot (80.9 μg/kg) and Tomato (77.4 μg/kg). The chromium level in all vegetable samples was within the recommended level of FAO/WHO. The mean concentration of Cr (77.4–433.3 μg/kg) in vegetables recorded during

Table 2 Concentration of heavy metals (μg/kg) in vegetables grown using paper wastewater in Wonji Gefersa, Ethiopia

Vegetables	Statistics	Fe	Cu	Cd	Zn	Pb	Cr	Co
Green pepper	Mean ± SD	569.9 ± 2.1	179.2 ± 1.2	136.7 ± 0.8	121 ± 3.1	376.5 ± 4.5	433.3 ± 1.5	184.9 ± 2.1
	Min–max	568.3–572.3	178.2–180.6	135.8–137.4	118.4–124.5	372.2–381.2	432.3–435	182.6–186.7
Green pepper (control)	Mean ± SD	381.1 ± 0.9	93.9 ± 0.2	51.1 ± 0.4	83.1 ± 0.3	102.8 ± 0.8	210.5 ± 0.7	122.4 ± 0.9
	Min–max	380–381.8	93.7–94.1	50.7–51.5	82.8–83.4	102–103.6	209.7–211.2	121.8–123.4
Swiss chard	Mean ± SD	368.8 ± 3.6	124.1 ± 2.7	138.5 ± 6.3	96 ± 5.2	574.7 ± 5.8	123.7 ± 1.6	219.1 ± 2.1
	Min–max	364.8–371.9	121.8–127.1	132.7–145.2	90.3–100.6	570.7–581.3	122.6–125.5	217.2–221.3
Swiss chard (control)	Mean ± SD	220.3 ± 0.5	73.2 ± 1.0	38.5 ± 0.8	61.4 ± 0.4	120.9 ± 0.4	102.2 ± 0.5	127 ± 0.2
	Min–max	219.7–220.8	72–74	37.6–39.1	61–61.8	120.4–121.3	101.7–102.6	126.8–127.2
Carrot	Mean ± SD	341.8 ± 1.7	88.2 ± 1.1	73.5 ± 0.8	212.2 ± 0.5	182.1 ± 3.1	80.9 ± 0.8	26 ± 3.0
	Min–max	340.1–343.6	87.2–89.4	72.8–74.4	211.8–212.7	179.2–185.3	80.4–81.8	22.6–28.1
Carrot (control)	Mean ± SD	236.3 ± 0.4	61 ± 0.4	26.2 ± 0.8	197.3 ± 0.6	97.8 ± 0.8	115.9 ± 0.2	ND
	Min–max	235.9–236.7	60.6–61.4	25.6–27.1	196.7–198	96.9–98.6	115.7–116.1	ND
Tomato	Mean ± SD	222.2 ± 1.5	98.6 ± 0.5	54.7 ± 2.7	259.3 ± 0.6	211.5 ± 3.1	77.4 ± 0.7	38 ± 0.5
	Min–max	220.5–223.3	98.1–99.1	52.4–57.6	258.7–259.9	208.9–215	76.6–78.1	37.5–38.4
Tomato (control)	Mean ± SD	196 ± 0.1	52.4 ± 0.9	18.5 ± 0.6	230.3 ± 1.1	126.6 ± 0.5	111.4 ± 0.6	ND
	Min–max	195.9–196.2	51.7–53.5	17.9–19.1	229.4–231.6	126–127	110.8–112.1	ND
Safe limit*		425,500	40,000	200	60,000	300	2300	50,000

n number of samples

*Source: FAO/WHO (2001)

the present study was lower than those reported in Koka, Ethiopia (0.56–1.51 mg/kg) (Fite and Leta 2015), and Addis Ababa, Ethiopia (0.05–1.65 mg/kg) (Weldegebriel et al. 2012).

The maximum uptake of Co was in Swiss chard (219.1 µg/kg) followed by Green pepper (184.9 µg/kg), Tomato (38 µg/kg), Carrot (26 µg/kg). All the vegetables had cobalt concentration below the recommended level of FAO/WHO. The mean concentration of Co (26–219.1 µg/kg) in vegetables of the present study was found very similar to the values (0.04–0.21 mg/kg) reported by Gebrekidan et al. (2013), but lower than the average concentration of Co (0.06–0.76 mg/kg) in vegetables from Kera's farm, Addis Ababa, Ethiopia (Itanna 1998).

The concentrations of heavy metals in vegetable samples were quite variable. Tomato was generally the least accumulator of Cd and Cr while carrot had lowest concentration of Cu and Pb (Fig. 1). Green pepper had generally the highest concentrations of Fe, Cu, and Cr; while Swiss chard contained the highest concentrations of Cd, Pb and Co. For vegetable samples of Tomato and Carrot, the trend was Pb > Cu > Cr > Cd.

The result from the finding indicated that green pepper bio-accumulated high amounts of Fe, Cr and Pb whereas Swiss chard bio-accumulated excessive amount of Pb and Fe (Fig. 1). This could be attributed due to the distinct nature of the vegetable species that accumulate different metals depending on their environmental conditions, metal species, plant available and forms of heavy metals. A study conducted by Gebrekidan et al. (2013) in Wukro town, Ethiopia also showed that Swiss chard accumulated high concentrations of heavy metals of Fe, Mn, Cr, Cd, Ni and Co.

Pb concentration in Green pepper and Swiss chard was above safe permissible levels recommended by WHO/FAO. Lead is a toxic element that can be harmful to plants, although plants usually show ability to accumulate large amounts of lead without visible changes in their appearance or yield.

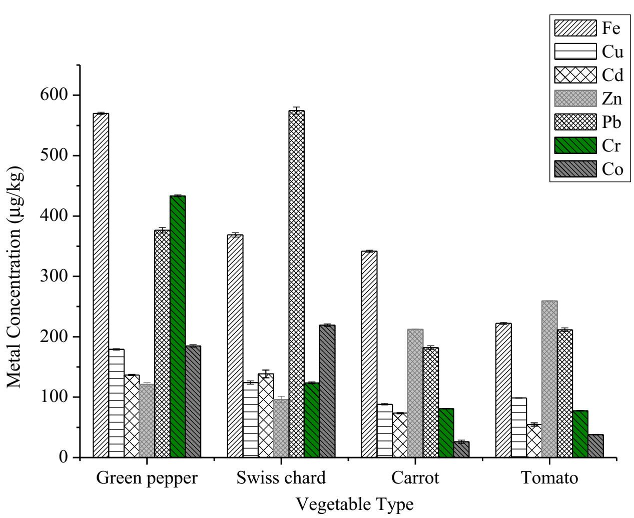

Fig. 1 Mean Concentrations of heavy metals in Vegetables of Wonji Gefersa, Ethiopia

In many plants, Pb accumulation can exceed several hundred times the threshold of maximum level permissible for human (Wierzbicka 1995). The introduction of Pb into the food chain may affect human health and thus, studies concerning Pb accumulation in vegetables have been increasing importance (Coutate 1992). Lead can be deposited in the soft tissues of the body and can cause musculoskeletal, renal, ocular, immunological, neurological, reproductive, and developmental effects (ATSDR 1999). Generally, Green pepper and Swiss chard in the study area were contaminated by lead, and they were toxic to consumer.

Person's Correlation analysis shown in Table 3 was used to determine the degree of metal association. The result indicated a positive correlation of most of the metals. Fe was positively correlated with Cu, Cd and Cr. Cu also positively correlated with Fe, Cd and Cr. Cd correlated with all the metals except to Zn. Zinc is the only metal which negatively correlated with Fe, Cu, Cd, Pb and Cr. The positive correlation probably indicated that the metals came from the same sources and that their geographic distributions were also similar. Cr was not correlated to Pb, and Fe also was not correlated with Pb indicating that these two groups of the metals were thus believed to be contributed by diverse sources. Yousufazi et al. (2001) showed that there was a strong association between Fe/Cu (r = 0.841), Fe/Cd (r = 0.985) in vegetables grown using a mixture of industrial effluent and sewage. A study conducted by Abbasi et al. (2013) reported that Fe was not correlated with Pb (r = 0.109).

The strong association between most of the metal indicated that their common sources might be from ink wastewater that discharged from the paper industry. The weak correlation between Cr and Pb; Fe and Pb indicated that either of the metal might have come from the upper stream like Mojo which different industries discharge their

Table 3 Correlation coefficient (r) matrix of heavy metals in vegetables grown using paper wastewater in Wonji Gefersa, Ethiopia

	Fe	Cu	Cd	Zn	Pb	Cr
Fe	1					
Cu	0.899	1				
Cd	0.788	0.785	1			
Zn	−0.728	−0.693	−0.990[b]	1		
Pb	0.391	0.496	0.873	−0.898	1	
Cr	0.928	0.965[a]	0.651	−0.549	0.268	1

[a]Correlation is significant at the 0.05 level (2-tailed)
[b]Correlation is significant at the 0.01 level (2-tailed)

wastewater to Awash River. The other possible reason might be the gas emission from the traffic deposited these metals, particularly Pb to the vegetable.

The concentration of heavy metal in paper wastewater were in the following order of decreasing magnitude Fe > Zn > Pb > Cr > Cu > Cd. The result showed that the concentration of Pb, Cd and Cr in paper wastewater were all above the safe limit for FAO standards for wastewater quality for irrigation.

Among the vegetables investigated, Tomato was the least accumulator of Cd and Cr while carrot had lowest concentration of Cu and Pb. Green pepper had the highest concentrations of Fe, Cu, and Cr whereas Swiss chard contained the highest concentrations of Cd, Pb and Co. For vegetable samples of Tomato and Carrot the trend was Pb > Cu > Cr > Cd. The result from the finding indicated that green pepper bio-accumulated high amounts of Fe, Cr and Pb whereas Swiss chard bio-accumulated excessive amount of Pb and Fe. The result indicated that level of Pb metal in Green pepper and Swiss chard was above the recommended level set by FAO/WHO. So that human consumption of these two vegetables leads to health impact.

Correlation analysis of heavy metals among vegetables showed that there is a significant positive correlation most of the metals. Generally, the result of the present study revealed Pb metal contamination in vegetables in varying magnitude among vegetables in the study area. Hence it poses an important public health risk. So monitoring heavy metals in plant tissues is essential in order to prevent the excessive buildup of these metals in the human food chain. To avoid entrance of metals into the food chain, a green treatment technique, such as a constructed wetland, should be used as a method to reduce heavy metal concentrations from different types of wastewater.

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