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Heavy Metal (Cu, Zn, Pb) Contamination of Vegetables in Urban City: A Case Study in Lagos

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Abstract: Five leafy vegetable species (*Talinum triangulare*, *Celosia argentea*, *Amaranthus viridis*, *Cucurbita maxima* and *Corchorus olitorius*) sold in Lagos markets were investigated for Cu, Pb and Zn content by atomic absorption spectrophotometry. Both unwashed and washed leaves showed the highest amounts of heavy metals in the urban area. Unwashed leaves showed greater difference between urban and remote areas and among the urban sites than washed leaves for heavy metal concentrations. Water-washing resulted in a significant ($p<0.001$) decrease in vegetable concentrations of Cu, Zn and Pb. However, the levels of Cu and Zn did not indicate excessive contamination that could be considered a serious health hazard to the consumers.

Key words: Unwashed and washed vegetables, copper, lead, zinc

INTRODUCTION

Vegetables constitute an important part of the human diet since they contain carbohydrates, proteins, vitamins, minerals as well as trace elements. The contamination of vegetables with heavy metals due to soil and atmospheric contamination poses a threat to its quality and safety. Dietary intake of heavy metals also poses risk to animals and human health. High concentrations of heavy metals (Cu, Cd and Pb) in fruits and vegetables were related to high prevalence of upper gastrointestinal cancer (Turkdogan *et al*., 2002). Many anthropogenic sources such as waste incineration, industrial processes and most importantly, vehicular traffic emit heavy metals into the atmosphere. Regulations have been set up in many countries and for different industrial set up to control the emission of heavy metals.

The uptake and bioaccumulation of heavy metals in vegetables are influenced by many factors such as climate, atmosphere depositions, the concentrations of heavy metals in soil, the nature of soil and the degree of maturity of the plants at harvest (Scott *et al*., 1996; Voutsa *et al*., 1996). Air pollution may pose a threat to post-harvest vegetables during transportation and marketing causing elevated levels of heavy metals in vegetables (Sharma *et al*., 2008). Elevated levels of heavy metals in vegetables are reported which such as long term uses of treated or untreated wastewater (Sharma *et al*., 2006, 2007; Adeniyi, 1996; Sinha *et al*., 2005). Other anthropogenic sources of heavy metals include the addition of manures, sewage sludge, fertilizers and pesticides which may affect the update of heavy metals by modifying the physico-chemical properties of the soil such as pH, organic matter, bioavailability of heavy metals in the soil (Yusuf and Osibanjo, 2006). Whatmuff (2002) and McBride (2003) found that increasing concentrations of heavy metals in soil increased the crop uptake. Cultivation areas near highways are also exposed to atmospheric pollution in the form of metal containing aerosols. These aerosols can be deposited on soil and absorbed by vegetables, or alternatively deposited on leaves and fruits and then absorbed. High accumulation of Pb, Cr and Cd in leafy vegetables due to atmospheric depositions has been reported by Voutsa *et al*. (1996) and De Nicola *et al*. (2008). The levels of heavy metals (Zn, Mn, Cu and Pb) in vegetable

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(*Talinum triangulare*) collected from dumpsites of Lagos, Nigeria were found to be high due to vehicular traffic emission (Adeniyi, 1996). The magnitude of heavy metal deposition of vegetable surfaces varied with morpho-physiological nature of the vegetables (Singh and Kumar, 2006). Jassir *et al*. (2005) have shown that unwashed leafy vegetables sold in road side of Riyadh city, Saudi Arabia had higher levels of heavy metals as compared to washed leafy vegetables. Demirezen and Aksoy (2006) have reported higher concentrations of Pb, Cd and Cu in *Abelmoschus esculentus* collected from urban areas of Kayseri, Turkey as compared to those from rural areas. The partitioning of heavy metals is well known, with accumulation of greater concentrations in the edible portions of leafy or root crops than the storage organs or fruits (Jinadasa *et al*., 1997; Lehoczky *et al*., 1998; Sharma and Agrawal, 2006).

Urban activities may significantly contribute to elevated heavy metal loads in atmospheric deposits and consequently in the edible portion of the vegetables. Thus the present study was conducted with the objectives to investigate the accumulation of heavy metals (Cu, Zn, Pb) on the (*Talinum triangulare*, *Celosia argentea*, *Amaranthus viridis*, *Cucurbita maxima* and *Corchorus olitorius*) leaf surfaces through the concurrent analysis of unwashed and washed leaves from urban and remote areas. Water-washing by shaking mechanically removes much of the particulate found on the leaf surfaces, allowing the fractions of particle-bound heavy metals to be estimated. This study therefore, aimed to distinguish the water-removable fraction of heavy metals and the fraction present in leaves, potentially supplying useful information on the pollutant fraction damaging to leaf physiological processes.

MATERIALS AND METHODS

The present study was carried out between August and October 2004 in the urban areas of Lagos, located in the South Western Nigeria. The climate of the region is tropical with two distinct seasons i.e dry and rainy. The dry season (November to April) is associated with high temperature during the day ranging from a minimum of 30°C to a maximum of 36°C. Rainy season starts in May and continue till end of October. During the rainy season, the temperature varies from a minimum of 24°C to a maximum of 32°C.

Lagos is one of the most densely populated cities of Nigeria. There are several industrial estate located at the periphery of the city. Heavy traffic on narrow roads leading to frequent traffic congestion is of common occurrence within the city.

Vegetable Markets

Five multipurpose retail markets were sampled. The magnitude of urban activities such as industrial, residential, commercial and vehicular density varied between the sampling locations.

The characteristics and relative position of sampling locations are shown in Table 1.

Leafy Vegetables

Common vegetables sold to the local markets exclusively are Soko (*Celosia argentea*), green (*Amaranthus viridis*) Ugwu (*Cucurbita maxima*), ewedu (*Corchorus olitorius*) and water leaf (*Talinum triangulare*). These vegetables were purchased ostensibly for consumption. They were stored at 4°C till processed.

Table 1: Characteristics of market sampling sites located in the urban area of Lagos

Study sites	Types of markets	Land uses
Mile 12	Wholesale/Retailer	DPRA, HT, CA
Oshodi	Retailer	DPRA, HT, NIA
Iyana Iba	Retailer	DPRA, HT, CA
Alaba	Retailer	DPRA, LT, CA
Yaba	Retailer	DPRA, HT, CA

DPRA: Densely Populated Residential Area, NIA: Near Industrial Area, HT: Heavy Traffic, CA: Commercial Area, LT: Low Traffic

Table 2: Wavelength and detection limits of each heavy metal measured by atomic absorption spectrophotometer (Mode 1 buck 200A)

Elements	Wavelength (mm)	Detection limits (μg mL^{-1})
Copper	324.8	0.0010
Zinc	213.9	0.0008
Lead	213.3	0.0100

Sampling and Pre-Treatment

Samples of edible portion of vegetables were collected from different outlets in each market locations. The samples were collected once in every two weeks between August and October 2004 for all the five vegetables. Samples were kept in pre-distilled water rinsed polyethylene bags, labelled and brought back to the laboratory. In the laboratory, the vegetable samples were chopped into small pieces divided into two groups.

The first group was oven dried at 65°C without washing, while the second group was washed in distilled water to remove airborne particulate before oven drying at 65°C until the constant weight was achieved. The dried samples were pulverized, sieved and kept at room temperature for further analysis. The water-removable heavy metal were calculated as the difference between unwashed and washed leaf concentrations.

Analytical Procedure for Heavy Metal Analysis

Aqua-regia mixture, 25 mL (70% high purity HNO_3 and HCl ratio 3:1) and 5 mL 30% H_2O_2 were added to an empty 100 mL beaker and heated at 80°C till the solution became cleared (Rodrigues-Flores and Rodriguez-Castellon, 1982). The resulting solution was cooled and filtered. The filtrate was made up to 50 mL with de-ionised water and kept at room temperature for further analysis of heavy metals. For the heavy metal analysis of dry vegetable, 1 g sample was taken into a 100 mL acid washed beaker, 25 mL aqua-regia and 5 mL 30% H_2O_2 were added (Rodrigues-Flores and Rodriguez-Castellon, 1982). The mixture was digested at 80°C until a clear solution was obtained. After cooling, the digested samples were filtered and diluted to 50 mL with de-ionised water.

Determination of the heavy metals such as Cu, Zn and Pb in the filtrate of vegetables and blanks was achieved by atomic absorption, spectrophotometer (model: Buck 200A). The instrument was calibrated using manually prepared standard solution of respective heavy metals. An analytical grade of a nitrate salt of Pb and granules of Cu and Zn were used in the preparations of solutions used in the spiking of samples for Pb, Cu and Zn.

Acetylene gas was used as the fuel and air as the support. An oxidizing flame was used in all cases. Table 2 shows different parameters at which the instrument was set for each metal.

Quality Assurance

Blanks and source of quality control standards were measured at every five samples to detect contamination and drift. The elemental concentrations of procedural blanks were generally <5% of the mean analyte concentrations for all the metals. Precision and accuracy of analysis were ensured through replicate analysis of the samples, acceptable percentage recoveries (86.5%±0.005 to 96.4±0.003) were obtained from spiked digested vegetable samples.

Statistical Analysis

The data were processed using statistical package, SPSS 11.0. In order to compare for each pollutant the concentrations in unwashed and washed leaves for both urban and remote sites, a paired t-test was performed. All the correlation analysis were performed by Pearson or Spearnan test.

RESULTS

The concentrations of Cu, Zn and Pb of *Celosia argentea*, *Amaranthus viridis*, *Cucurbita maxima*, *Corchorus olitorius* and *Talinum triangulare* are shown in Table 3-7, respectively.

Table 3: Mean metal concentrations ($\mu g\ g^{-1}$ dry weight) in washed and unwashed *Celosia argentea* at the urban and remote sites

	Urban			Remote			
Elements	Unwashed	Washed	PR[a]	Unwashed	Washed	PR	Safe[b] limit
Cu	12.06±0.26	8.10±0.33	32.8	2.90±0.19	2.83±0.17	2.41	40.0
Zn	34.20±1.80	23.82±0.62	30.4	22.60±2.71	21.90±1.88	3.10	60.0
Pb	3.25±0.02	2.15±0.03	34.0	0.33±0.04	0.31±0.02	6.06	0.3

[a] Percent reduction, [b] Source: FAO/WHO-Codex Alimentarius Commission (2001)

Table 4: Mean metal concentrations ($\mu g\ g^{-1}$ dry weight) in washed and unwashed *Amaranthus viridis* at the urban and remote sites

	Urban			Remote			
Elements	Unwashed	Washed	PR[a]	Unwashed	Washed	PR	Safe[b] limit
Cu	10.86±0.54	6.50±0.29	40.1	1.73±0.61	1.70±0.94	1.73	40.0
Zn	36.76±2.30	23.26±0.96	36.7	20.73±1.34	20.05±1.05	3.50	60.0
Pb	10.57±0.04	5.75±0.03	45.6	2.96±0.06	2.84±0.04	4.05	0.3

[a] Percent reduction, [b] Source: FAO/WHO-Codex Alimentarius Commission (2001)

Table 5: Mean metal concentrations ($\mu g\ g^{-1}$ dry weight) in washed and unwashed *Cucurbita maxima* at the urban and remote sites

	Urban			Remote			
Elements	Unwashed	Washed	PR[a]	Unwashed	Washed	PR	Safe[b] limit
Cu	17.38±1.30	11.04±0.83	36.5	3.76±1.23	3.71±0.45	1.33	40.0
Zn	33.28±2.76	22.23±1.42	33.0	22.10±3.60	21.80±2.91	1.36	60.0
Pb	4.30±0.07	2.56±0.02	40.5	1.57±0.62	1.45±0.26	7.60	0.3

[a] Percent reduction, [b] Source: FAO/WHO-Codex Alimentarius Commission (2001)

Table 6: Mean metal concentrations ($\mu g\ g^{-1}$ dry weight) in washed and unwashed *Corchorus olitorius* at the urban and remote sites

	Urban			Remote			
Elements	Unwashed	Washed	PR[a]	Unwashed	Washed	PR	Safe[b] limit
Cu	11.83±1.58	6.83±0.65	42.3	2.24±0.73	2.21±0.55	1.34	40.0
Zn	31.02±1.46	18.50±1.12	40.0	17.50±2.65	17.21±1.79	1.70	60.0
Pb	13.10±0.05	6.51±0.03	50.3	4.12±0.03	3.84±0.03	1.00	0.3

[a] Percent reduction, [b] Source: FAO/WHO-Codex Alimentarius Commission (2001)

Table 7: Mean metal concentrations ($\mu g\ g^{-1}$ dry weight) in washed and unwashed *Talinum triangulare* at the urban and remote sites

	Urban			Remote			
Elements	Unwashed	Washed	PR[a]	Unwashed	Washed	PR	Safe[b] limit
Cu	18.28±2.03	11.12±1.38	39.2	3.92±0.14	3.89±0.09	0.76	60.0
Zn	24.48±1.21	15.65±1.07	36.0	14.02±1.12	13.91±1.25	0.78	40.0
Pb	3.85±0.08	1.92±0.03	50.0	0.55±0.01	0.52±0.02	5.45	0.3

[a] Percent reduction, [b] Source: FAO/WHO-Codex Alimentarius Commission (2001)

The mean concentrations of heavy metals were highest for Zn followed by Cu and Pb. Among the vegetables, mean concentration of Zn (36.76 $\mu g\ g^{-1}$) was highest in *Amaranthus viridis*, but mean Cu concentration (18.28 $\mu g\ g^{-1}$) was highest in *Talinum triangulare*, while mean Pb concentration (13.0 $\mu g\ g^{-1}$) was highest in *Corchorus olitorius*.

Both unwashed and washed leaves showed lower metal concentrations at the remote than the urban sites (Table 3-7), with significant differences (at least $p<0.05$) in all the investigated heavy metals. However, for washed vegetables the differences in concentrations for each heavy metal between urban and remote sites were narrower than those shown for unwashed vegetables. The greatest differences between urban and remote sites were observed for Pb, both for unwashed and washed vegetables.

The unwashed leaves showed higher metal concentrations than washed leaves (Table 3-7). At the urban sites, the differences were significant ($p<0.001$), moreover, the concentrations of metals in unwashed leaves were correlated (at least $p<0.01$) to those in washed leaves.

Over the investigated period, mean water-removable heavy metal concentrations (calculated as the percentage ratio of the difference between unwashed and washed leaf metal concentration to unwashed leaf metal concentrations) decreased in the order of Pb>Zn>Cu at the remote sites and Pb>Cu>Zn at the urban sites (Table 3-7). Percentages of Zn, Cu (*Amanranthus viridis* and *Talinum triangulare*) and Pb (*Corchorus olitorius* and *Talinum triangulare*) water removable concentrations were similar at the urban sites whereas water removable concentrations of Cu, Zn and Pb were higher at the urban sites.

DISCUSSION

Urban area of developing countries often receive atmospheric depositions of heavy metals resulted in contamination of fresh vegetables at the time of transportation and marketing. Consumption of contaminated vegetables may pose risk to human health. Heavy metals determined in different vegetables showed that the concentrations of Pb exceeded the safe limits (FAO/WHO (Codex Alimentarius Commission, 2001) standard Cu and Zn concentrations, however, did not exceed the safe limit of the test vegetable. Exceedence of safe limits was highest for *Amaranthus viridis*, where, 60% of the sampling locations showed Pb safe limit exceedence of FAO/WHO standards.

The variations in the concentrations of the heavy metals in vegetables may be ascribed to the heavy metals concentrations of soil, air and irrigation water of their production sites and also the absorption of heavy metals from aerial depositions during transportation and marketing.

The water-removable proportion of heavy metals provides information about both the prevailing metal source (natural or anthropogenic) and the leaf absorption amount for each metal. The results of speciation studies on air particulate suggest that metals from anthropogenic sources are mainly in water-soluble forms (Fernandez Espinosa *et al*., 2002). The water removable proportions for Cu, Pb and Zn were more noticeable at the urban than remote sites showing greater atmospheric depositions on vegetables during marketing are a major source of heavy metal contamination in urban sites. The observed trend of the proportion of water removable heavy metals at the urban sites (Pb>Cu>Zn) is confirmed by other researchers for *Quercus ilex* leaves in Italy (De Nicola *et al*., 2008), for *Petunia* leaves in Madrid (Caselles *et al*., 2002) and *Holm oak* leaves in fome (Gratani *et al*., 2000). Cu and Zn are present in airborne particulate as water-soluble sulphate salts (Huggins *et al*., 2000; Heal *et al*., 2005; Karthikeyan *et al*., 2006) and it is generally reported that the leaching of each metal is related to the kinds of liaisons between metals and the solid matrix (Chester *et al*., 1993; De Nicola *et al*., 2008).

Foliage analysis has often been used in heavy metal biomonitoring studies and concurrent analysis of unwashed and washed vegetables allow external composition to be distinguished from internal (Dasch, 1987; De Nicola *et al*., 2008). The differences between unwashed and washed leaves with regard to the heavy metal concentrations suggest that heavy metals reach leaves by aerial deposition and adhere to the leaf surface. The water-washing procedure mechanically removes particles deposited on the leaf surface, hence a large amount of the heavy metals bound to particles and water-soluble aerosols (Kos *et al*., 1996; De Nicola *et al*., 2008; Sharma *et al*., 2008).

In the present study, the mean concentrations of Cu, Zn and Pb (18.28, 24.48 and 3.85 $\mu g\ g^{-1}$, respectively) in *Talinum triangulare* were found higher than the concentrations reported in *Talinum triangualare* (2.76, 22.11 and 0.44 $\mu g\ g^{-1}$, respectively for Cu, Zn and Pb) collected from dump sites at Lagos, Nigeria (Adeniyi, 1996).

CONCLUSION

From this study, it can be concluded that urban activities in Lagos have elevated the levels of heavy metals in urban atmospheric deposits, which consequently increased the concentrations of heavy metal in test vegetables during marketing. Variations in the magnitude of reductions in heavy metal contamination due to washings of vegetables also depicted or showed the variations in heavy metal depositions at various market sites. Vegetables sold in local markets of the city still looking healthy and fresh, may pose a risk to human health particularly due to Pb contamination. Based on safe limit data *Corchorus olitorius* is maximally contaminated with Pb followed by *Amaranthus viridis*. The study suggests that washing technique can be used as a tool to assess the heavy metals load in vegetables through atmospheric depositions. The present study further suggested that to reduce the health risk, vegetables should be washed properly before consumption as washing can remove a significant amount of aerial contamination from the vegetable surface.

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