HEAVY METALS IN SELECTED EDIBLE VEGETABLES AND ESTIMATION OF THEIR DAILY INTAKE IN SANANDAJ, IRAN

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Abstract. The levels of four different heavy metals [cadmium (Cd), lead (Pb), chromium (Cr) and copper (Cu)] were determined in various vegetables [leek (*Allium ampeloprasum*), sweet basil (*Ocimum basilicum*), parsley (*Petroselinum crispum*), garden cress *(Lepidium sativum)* and tarragon (*Artemisia dracunculus*)] cultivated around Sanandaj City. The contributions of the vegetables to the daily intake of heavy metals from vegetables were investigated. One hundred samples (20 samples per month) were collected for five months. Atomic absorption spectrometry was used to determine the concentrations of these metals in the vegetables. The average concentrations of each heavy metal regardless of the kind of vegetable for Pb, Cu, Cr and Cd were 13.60 ± 2.27, 11.50 ± 2.16, 7.90 ± 1.05 and 0.31 ± 0.17 mg/kg, respectively. Based on the above concentrations and the information of National Nutrition and Food Research Institute of Iran, the dietary intake of Pb, Cu, Cr and Cd through vegetable consumption was estimated at 2.96, 2.50, 1.72 and 0.07 mg/day, respectively. It is concluded that the vegetables grown in this region are a health hazard for human consumption.

INTRODUCTION

Food safety is a major public concern worldwide. During the last decades, the increasing demand for food safety has stimulated research regarding the risk associated with consumption of foodstuffs contaminated by pesticides, heavy metals and/or toxins (D’Mello, 2003). Vegetables constitute essential components of the diet, by contributing protein, vitamins, iron, calcium and other nutrients which are usually in short supply (Thompson and Kelly, 1990). Vegetables also act as buffering agents for acidic substances obtained during the digestion process. However, these plants may contain both essential and toxic elements, such as heavy metals, at a wide range of concentrations (Bahemuka and Mubofu, 1999). Metals, such as lead, chromium, cadmium and copper are cumulative poisons. These metals cause environmental hazards and are reported to be exceptionally toxic (Ellen *et al*, 1990).

Contamination of vegetables with havey metal may be due to irrigation with contaminated water, the addition of fertilizers and metal-based pesticides, industrial emissions, transportation, the harvesting process, storage and/or at the point of sale. Human beings are encouraged to consume more vegetables and fruits, which are a good source of vitamins, minerals, fiber and are beneficial for health. However, these plants contain both essential and toxic metals over a wide range of concentrations. It is well known that plants take up metals by absorbing them from contaminated soil as well as from deposits on parts of the plants exposed to the air from polluted environments (Khairiah *et al*, 2004; Chojnacka *et al*, 2005). Publicity regarding the

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high level of heavy metals in the environment has created apprehension and fear in the public as to the presence of heavy metal residues in their daily food. The public is confused and alarmed about their food safety. Keeping in mind the potential toxicity and persistent nature of heavy metals, and the frequent consumption of vegetables and fruits, it is necessary to analyze these food items to ensure the levels of these contaminants meet agreed international requirements (Radwan and Salama, 2006).

Lead and cadmium are among the most abundant heavy metals and are particularly toxic (Radwan and Salama, 2006). Excessive content of these metals in food is associated with a number of diseases, especially of the cardiovascular, renal, nervous and skeletal systmes (WHO, 1992, 1995; Steenland and Boffetta, 2000; Jarup, 2003). These heavy metals are also implicated in carcinogenesis, mutagenesis and teratogenesis (Radwan and Salama, 2006). Other metals, such as copper and zinc are essential for important biochemical and physiological functions and necessary for maintaining health throughout life (Prentice, 1993; ATSDR, 1994; Linder and Azam, 1996).

The aim of this study was to determine the concentrations of heavy metals in selected edible vegetables grown in the main farmlands (along the banks of the Gheshlagh River) around Sanandaj City, and to estimate their contribution to the daily intake of the metals.

MATERIALS AND METHODS

Samples of the edible vegetables were randomly collected from the main farmlands around the city. The samples were collected from these growing areas over a period of five months during the cultivation season (May-September) during the year 2005. A total of 100 samples of five vegetables [leek (*Allium ampeloprasum*), sweet basil (*Ocimum basilicum*), and parsley (*Petroselinum crispum*), garden cress *(Lepidium sativum)* and tarragon (*Artemisia dracunculus*)] were collected (four composite samples of each kind of vegetable during each month). All samples were collected and stored in polythene bags according to their type and brought to the laboratory for preparation and treatment.

For lead, cadmium, chromium and copper analyses, vegetable samples were washed with distilled water to eliminate suspended particles. The leafy stalks were removed from all samples and these were sliced and dried on a sheet of paper to eliminate excess moisture. Once dried, each sample was weighed and oven-dried at 60ºC to a constant weight. Each oven-dried sample was ground in a mortar until it could pass through a 60 mesh sieve. The samples were stored in clean, dry, high density polyethylene bottles of 100 ml capacity with screw caps. Bottles were prewashed with nitric acid, rinsed with de-ionized water, dried and tested for contamination by leaching with 5% nitric acid. The bottles contained no metal liners that could contaminate the samples. Samples were precisely weighed (2 grams each) and ground in a mortar followed by wet digestion with HNO_3:$HClO_4$ (2:1) in a conical flask for 2-3 hours on a sand bath. Some 10 ml of HCl was added. Digested samples were filtered with 0.45 µm pore size cellulose nitrate membrane filter paper (Millipore) and the volume was increased to 100 ml with distilled water and bottles were stored until flame atomic absorption spectrophotometry was performed. The samples were analyzed by an atomic absorption spectrophotometer (ALPHA 4, Chem Tech Analytical, England) using a nitrous oxide-acetylene flame for As and an air-acetylene flame for the four heavy metals- Pb, Cd, Cr and Cu, using at least two standard solutions for each metal. A certified standard reference material (Alpha-Line, Chem Tech Analytical, England) was used to ensure accuracy, and the analytical values were within the range of certified values. All recoveries of the metals studied were greater than 95%. The mean (±SD) concentrations for each heavy metal regardless of the kind of vegetable (the means of twenty replicates for each element in mg/kg by dry weight) were calculated and a comparison of this data in the selected vegetables was studied, and compared with the permissible levels set by the FAO and WHO. Based on the average concentration and the average consumption of edible vegetables, estimates of the amount of each heavy metal consumed were calculated.

Analysis using the Kruskal-Wallis test was carried out to examine the statistical significance of differences in the mean concentration of metals between groups of vegetables using SPSS, version 11. A probability level of $p<0.05$ was considered statistically significant.

RESULTS

The mean concentrations of heavy metals found in the vegetables samples are summarized in Table 1. The results show high levels of Pb, Cd and Cr in all vegetables studied. The levels of Pb in all samples were between 11.23 ± 0.006 mg/kg in parsley and 16.99 ± 0.46 mg/kg in sweet basil. Cd content varied from non-detectable amounts in leeks to 0.65 ± 0.02 mg/kg in garden cress. The highest concentration of Cr was detected in tarragon (8.81 ± 0.25 mg/kg) while sweet basil had the lowest concentration (6.30 ± 0.89 mg/kg). Within the selected vegetables, the highest concentration of Pb was found in sweet basil. The highest concentrations of Cu and Cr were observed in leeks and tarragon. Table 2 shows the mean (± SD) levels of Pb, Cr, Cu and Cd consumed.

DISCUSSION

The results in Table 1 show a high degree of contamination in vegetables, when compared with the permissible levels given by the FAO and WHO. Good agreement of this

Table 1
Concentrations of heavy metals in vegetables cultivated in farmland areas of Sanandaj, in terms of mg/kg dry weight.

Element	Parsley	Leek	Tarragon	Garden cress	Sweet basil	Recom max l for vegetables[b]
Cd	0.20 ± 0.006	ND [a]	0.40 ± 0.05	0.65 ± 0.02	0.32 ± 0.05	0.2
Cr	8.73 ± 0.33	8.18 ± 0.15	8.81 ± 0.25	7.50 ± 0.38	6.3 ± 0.89	2.3
Pb	11.23 ± 0.79	14.88 ± 0.13	11.33 ± 0.23	13.11 ± 0.34	16.99 ± 0.46	0.3
Cu	11.68 ± 0.42	15.06 ± 0.28	10.72 ± 0.21	8.71 ± 1.01	11.35 ± 0.51	40.0

[a] ND = Not detected. Levels were below the detection limit; [b]Recom max l = Recommended maximum limit; [b]Source: FAO/WHO (2001), Joint Codex Alimentarius Commission

Table 2
Heavy metal content in selected vegetables and estimated daily intake through consumption of vegetables in Sanandaj.

	Pb	Cu	Cr	Cd
Average concentration (mg/kg dry wt)	13.60± 2.27	11.50± 2.16	7.90± 1.05	0.31± 0.17
Estimated daily intake (mg/day)	2.96	2.5	1.72	0.07

data was observed when the levels of Pb and Cd were compared with previously reported data. For instance, lead concentration (1.36 mg/100 g) was within the range of 0.05 to 6.70 mg/100 g, and 0.13 to 2.27 mg/100 g, reported from Ireland and New York, respectively. For vegetables from Ireland, the Cd concentration was in a range of 0.005 to 0.06 mg/100 g, whereas that from New York was in a range of 0.004 to 0.061 mg/100 gm (Bahemuka and Mubofu, 1999), which are in agreement with the result of this study. However, the concentration of Cd in this study was below the range of 0.09 to 0.26 mg/100 g reported for vegetables grown in Metropolitan Boston and Washington DC (Hibber *et al*, 1984). Furthermore, the Cd and Pb levels reported in this study were lower than those reported for vegetables in Nigeria (Ndiokwere, 1984) and Pakistan (Parveen *et al*, 2003) and higher than those reported for vegetables in Tanzania (Bahemuka and Mubofu, 1999) and Egypt (Radwan and Salama, 2006). The high contamination levels found in some vegetables may be related to pollutants in irrigation water, farm soil or pollution from highway traffic (Igwegbe *et al*, 1992). All vegetables had lower levels of Cu than the permissible values for food (4 mg/100 g) recommended by the FAO/WHO (2001). Generally, plants contained an amount of Cu, inadequate for normal growth. Application of micronutrient fertilizers and Cu based fungicides may sometimes increase this to alarming levels. In the present study, the concentration of Cu ranged from 8.71 to 15.06 mg/kg. The highest amount was found in leeks and the lowest in garden cress. From the results, it can be noted that the levels of Cu obtained in this study were comparatively higher than those reported by Bahemuka and Mubofu (1999) and Radwan and Salama (2006) for similar green vegetables.

As shown in Table 2, a comparison between the average concentration of each element in all five types of vegetable (Table 2) and the permissible levels recommended by the FAO and WHO (Table 1) clearly indicates that the concentrations of Pb, Cr and Cd were 45, 3.50 and 1.55 times above the permissible levels, respectively. This finding is similar to that reported in previous studied of Pb (Kursad *et al*, 2002) and Cd (Alam *et al*, 2003). However, Cu was within acceptable levels. Variations in metal contents for these vegetables depends on the physical and chemical nature of the soil and the absorption capacity for each metal by each plant, which is altered by numerous environmental and human factors as well as the nature of the plant (Zurera-Cosano *et al*, 1989; Sharma *et al*, 2007). The results show there are significant variations in the levels of these metals among vegetables ($p<0.001$, Kruskal-Wallis test).

Exposure of consumers to health risks are usually expressed as provisional tolerable daily intake (PTDI), a reference value established by the FAO/WHO (1999). Table 2 shows an estimate for each heavy metal consumed in the studied vegetables. The National Nutrition and Food Research Institute of Iran estimated the average consumption of edible vegetables is 218 g/person/day. The average concentration of Pb, Cu, Cr and Cd were 13.60 ± 2.27, 11.50 ± 2.16, 7.90 ± 1.05 and 0.31 ± 0.17 mg/kg, respectively. Based on the above concentrations, if a person consumes 218 g of vegetables/day, he/she will then ingest 2.96 g of Pb, 2.5 g of Cu, 1.72 g of Cr and 0.07 g of Cd/day. Studies from other countries have reported that the dietary intake for lead in adults is between 30 and 427 µg/day (Dabeka *et al*, 1987; Bahemuka and Mubofu, 1999; Radwan and Salama, 2006). For cadmium and copper, the estimated daily intake is from 4.60 µg to 30 mg/day and 0.45 to 20 mg/day, respectively (Bahemuka and Mubofu, 1999; Radwan and Salama, 2006). It is concluded that the estimated daily intake of lead in the present study is above that reported for other countries, whereas the estimated consumption of cadmium and copper are within the normal range. The estimated daily intake of Pb and Cd in this study are above that recommended by the FAO/WHO which has set a limit for heavy metal intake based on body weight for an average adult (60 kg body weight). The PTDI for Pb, Cd and Cu are 214 µg, 60 µg and 3 mg, respectively (Joint FAO/WHO Expert Committee on Food Additives, 1999). Thus, we concludes that a large daily intake of these vegetables is likely to be a health hazard to the consumer.

In conclusion, the present study provides data on heavy metal pollution in Sanandaj. The levels of heavy metals in the studied vegetables, and the permissible levels required for safe food were compared. High Pb content was found in sweet basil. A high level of Cd was found in garden cress. These amounts may be hazardous if the vegetables are taken in large quantities. However, the results of this study indicate that the daily intake of Cu through edible vegetables from farmland areas may not constitute a health hazard for consumers because the values were below the recommended daily intake of this metal. Nonetheless, all these metals have toxic potential, but the detrimental impact becomes apparent only after decades of exposure. It is therefore suggested that regular monitoring of heavy metals in plant tissues is essential in order to prevent excessive build-up of these metals in the human food chain.

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