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Concentration levels of heavy metals in irrigation water and vegetables grown in peri-urban areas of Sanandaj, Iran

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Original Article

Abstract

Concentration and daily intake (DI) of heavy metals [lead (Pb), chromium (Cr), cadmium (Cd) and copper (Cu)] were investigated in four common edible vegetables including coriander, dill, radish root and radish leaf grown at peri-urban sites in Sanandaj, Iran. A total of 120 composite samples of vegetables were taken from ten vegetable farms during six months from May to October 2012. Inductively coupled plasma optical emission spectroscopy (ICP-OES) was used to estimate the levels of heavy metals. The results showed that Pb and Cr concentrations exceeded the safety limits given by Food and Agriculture Organization (FAO) or the World Health Organization (WHO) for human consumption with the exception of copper and cadmium that were lower than the permissible leveling in all of the samples. Furthermore, the results showed that there was a significant variation in the levels of these metals among the examined vegetables ($P < 0.001$). DI values for Pb, Cu, Cr and Cd could be 0.1, 1.5, 0.94 and 0.004 mg per day, respectively. As respect, DI values for Pb and Cd were also below the international guideline bases. Although Pb level was higher than the permissible standard, it seems that daily intake of these vegetables may not have detrimental health hazards to consumers.

KEYWORDS: Vegetables, Heavy Metals, Daily Intake

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Introduction

In recent years, vegetables consumption was increasing in the world and constituted an important part of the human diet and nutrition. This was due to vegetables contain essential diet components of vitamin, protein, minerals, trace elements and other nutrients. Buffering agent is another and important function of vegetables for acidic and some toxic substances produced during the digestion process. In spite of this fact, nutritional value and consumer acceptance must be taken into consideration when vegetables are being considered as foodstuffs; because vegetables can contain both essential and toxic elements over a wide range of concentrations.[1,2] Therefore, Food safety issues and potential health risk are a major public concern worldwide and make it as one of the most serious environmental concerns.[3]

It is well known that heavy metals are among the major contaminants of foodstuffs and can considered as one of the most

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important problems for human health because they have long biological half-lives, non biodegradable and accumulation potential in different organic bodies.[3] Among the organics, vegetables may become contaminated with heavy metals if they grow on soils contaminated by natural or anthropogenic sources such as industrial and agricultural activities.[4] Therefore, intake of heavy metal-contaminated vegetables may pose a direct threat to the human health and considered as one of the most important aspects of food quality assurance.[2,3,5] International and national regulations on food quality have lowered the maximum permissible levels of toxic metals in food items due to an increased awareness of the risk these metals pose to food chain contamination.[3,5]

Unorganized urbanization and rapid industrial and agricultural developments resulted in an increase of heavy metal metals in the environment.[5] Contamination of soil with heavy metals is common and it can be a major source of metals to crops and finally may be a primary path of human exposure to these potentially toxic metals.[6,7] Several authors have shown heavy metals as important contaminants of the vegetables.[5,8,9] However, bioaccumulation of elements in vegetables is more complicated because their uptake by plants depends upon the climate, soil properties, atmospheric depositions, irrigating water quality and plant physiologic factors.[8]

Kurdistan province is one of the major agriculture areas in the west part of Iran. Sanandaj, the capital of Kurdistan province currently covers more than 100 km^2 peri-urban areas for agricultural activities specially vegetables production. Agricultural activities in this area causes significant alterations in the water resources and increases the toxic metals resulting from fertilizers and metal-based pesticides, industrial emissions, transportation and harvesting process. Since crop irrigation is mostly depend upon groundwater, a concern in Sanandaj agricultural products from urban and peri-urban sites is the transfer of toxic metals from vegetables through the food chain to humans. For example, it has been estimated that vegetables consumption contributes up to 70% of the dietary intake of Cd.[6] Adekunle et al. have reported that Pb in vegetables exceeded the recommended values for three cities in Nigeria.[10] Demirezen and Aksoy have shown high concentrations of [lead (Pb), cadmium (Cd) and copper (Cu)] in Abelmoschus esculentus collected from urban areas of Kayseri, Turkey.[11] Similarly, Maleki and Zarasvand observed high concentrations of Pb, Cd and Cr in sweet basil, parsley, leek and garden cress collected from peri-urban areas of Sanandaj, Iran.[12] Zhuang et al. have reported heavy metal concentration exceed food standards for vegetables grown close to metal smelters.[13] Thus, monitoring and assessment of heavy metals concentrations in the vegetables from the peri-urban sites in developing countries are necessary. However, limited published data are available on heavy metals concentrations in the vegetables and human exposure to contaminants from the peri-urban sites of Iran. Therefore, the present study aimed to determine the concentrations of heavy metals in selected edible vegetables grown at peri-urban sites in Sanandaj and to estimate their contribution to the daily intake of the metals.

Materials and Methods

The present study was carried out since May to October 2012 in the peri-urban areas of Sanandaj, located in the west part of Iran. Six major vegetable farms (from two major agricultural area namely Naysar region and Gharezeh region) were selected to study the level of trace metal contamination in vegetables and irrigation waters. Four common vegetables including coriander (Coriandrum sativum), dill (Aniethum graveolens), radish (Raphanus sativus) and radish leaf were selected for the evaluation. Samples of vegetables were randomly and simultaneously collected monthly from the selected farms, prepared and preserved in the laboratory until analyzed. A total of 120 samples of vegetables were taken as described below.

The vegetable samples were randomly collected in the form of sub-samples from each site at various distances (10 m, 30 m, 50 m, 70 m, 120 m and 140 m) away from the first sub-sample. Then they were thoroughly mixed to give a composite sample as a representative fraction of the vegetables. The vegetable samples were washed with distilled water to remove soil particles. Then they were air-dried for two weeks to remove excess moisture until stable weights were obtained. Samples were then grounded in a porcelain mortar with a pestle, passed through a 10 mesh sieve. For vegetable digestion, 1 g of each sample was taken into a Pyrex beaker and 10 ml of tri-acid mixture [Nitric acid (HNO_3), Perchloric acid ($HClO_4$) and Sulfuric acid (H_2SO_4) in 1:1:1 ratio)] was added. It was kept for 24 hours and then heated on the hot plate at 95° C until its volume was reached to 10 ml. Then 10 ml of tri-acid mixture was added again and heated again until the solution was reduced to 4 ml.[14] Thereafter, it was diluted with 50 ml deionized water. The digest was filtered and the final volume of 100 ml was made with double deionized water. The selected farms were irrigated with ground water. Water samples were collected in a 500 ml washed polypropylene bottle and 1 ml of concentrated acid nitric was added to the samples to avoid microbial activity.

Determination of the heavy metals was achieved by inductively coupled plasma optical emission spectroscopy (ICP-OES) (Model Spectro arcos., SPECTRO Inc., Germany). The main operation parameters including torch type, detector type, nebulizer type, nebulizer flow, plasma power, coolant flow and pump rate were flared end EOP Torch 2.5 mm, CCD, cross flow, 0.85 l/min, 1400 W, 14.5 l/min and 30 RPM, respectively. All used chemical reagents were of analytical grade including standard stock solutions of different heavy metals.

The daily intake (DI) of heavy metals from vegetables consumption was calculated using the following formulas:[14]

$$DI = C_{metal\ in\ vegetable} \times M_{vegetable\ ingested}$$

Analysis of Kruskal-Wallis test were employed to examine the statistical significant of differences in the mean concentration of metals between group of vegetable families using SPSS for Windows (version 16.0, SPSS Inc., Chicago, IL, USA). A probability level of $P < 0.05$ was considered statistically significant.

Results and Discussion

Levels of Heavy Metals in Irrigation Waters

The chemical properties and metal concentrations of water from the six water irrigation sources are given in tables 1-3. As it is illustrated, the pH of the irrigation waters ranged from 7.6 to 8.1, the total dissolved solid (TDS) from 0.479 to 0.732 mg l^{-1}, and the electrical conductivity (EC) measured at 25° C from 0.479 to 0.732 dS/m (Table 1). These values are within the acceptable range recommended for irrigation waters.[15] The concentration of cations in water used for irrigation was highest for Mg, followed by Ca, Na, and K (Table 2). The cations and anions concentrations of the water except for Mg and K were found to be lower than the acceptable range recommended for irrigation waters.[15] According to table 3, the concentrations of heavy metals in the all irrigation water sources were lower than the acceptable range recommended for irrigation waters.[15] The presence of low amounts of these metals in irrigation waters may decrease their levels in soils, which in turn may weaken metal uptake by plants ultimately leading to down concentrations in the vegetables.

Levels of Heavy Metals in Vegetables

The mean concentration and range of heavy metals found in vegetables samples from cultivated sites are summarized in table 4. The heavy metal concentrations were determined based on vegetable dry weight. The results

Table 1. Some properties of irrigation waters of vegetable farms

Farm	pH	EC (dS/m)	TDS (mg l^{-1})	Total hardness (mg l^{-1} $CaCO_3$)	Turbidity (NTU)	Total alkalinity (mgl^{-1} $CaCO_3$)
1	8.1	0.585	379	280	5.0	126
2	7.9	0.479	312	208	3.1	130
3	7.6	0.610	396	312	2.5	140
4	7.6	0.600	388	300	2.4	137
5	7.6	0.732	477	440	1.2	166
6	7.7	0.715	465	432	1.0	170
Recommended Max. Concentration[15]	6.5-8.4	0-3	0-2000	-	-	-

EC: Electrical conductivity; TDS: Total dissolved solid

Table 2. Cations and anions concentrations in the irrigation waters of vegetable farms in terms of mg l^{-1}

Farm	Ca^{+2}	Mg^{+2}	Na^{+}	K^{+}	Cl^{-}	Br^{-}	SO_4^{-2}	NO_3^{-}	SAR
1	8.00	9.88	2.44	0.070	1.56	0.003	1.75	1.03	0.81
2	9.00	10.53	2.39	0.100	1.74	0.003	2.20	1.13	0.76
3	9.60	9.87	2.39	0.102	1.60	0.003	1.93	0.85	0.76
4	10.00	9.00	2.60	0.092	1.46	0.003	1.85	0.80	0.84
5	15.60	10.30	1.39	0.023	1.32	0.003	0.63	2.37	0.38
6	14.45	9.71	1.52	0.025	1.40	0.003	0.73	2.33	0.43
Recommended Max. Concentration[15]	0-20	0-5	0-40	0-0.05	0-30	-	0-20	0-10	0-15

Table 3. Heavy metal concentrations in the irrigation waters of vegetable farms

Element (ppb)	Farm						Recommended Max. Concentration[15]
	1	2	3	4	5	6	
Cd	0.10	0.10	0.10	0.10	0.10	0.10	10
Pb	1.00	1.00	2.00	2.00	1.00	1.00	5000
Cu	0.30	0.30	0.30	0.30	0.30	0.30	200
Cr	5.00	4.00	4.00	5.00	5.00	5.00	100
Zn	0.10	0.10	0.10	0.10	5.00	4.00	2000
Co	2.00	1.00	2.00	2.00	2.00	3.00	50
Fe	8.00	9.00	9.00	5.00	84.00	70.00	5000
Al	0.10	0.10	0.10	0.10	0.10	0.10	5000
Hg	0.10	0.30	0.50	0.30	0.20	0.40	-
As	0.10	0.10	0.10	0.10	0.10	0.10	100
Ni	0.20	0.20	0.20	0.20	0.20	0.20	200
Mo	0.20	0.20	0.20	0.20	0.20	0.20	10
Sn	12.00	9.00	8.00	9.00	8.00	8.00	-
Mn	0.10	0.10	0.10	0.10	0.10	0.10	200
Ba	0.00	0.00	0.00	0.00	0.00	0.00	-
B	33.00	30.00	30.00	29.00	18.00	19.00	700-3000
Li	70.00	80.00	90.00	70.00	90.00	80.00	2500
F	290.00	310.00	350.00	330.00	370.00	390.00	1000
Sr	157.00	140.00	169.00	178.00	172.00	167.00	-
Mg	5157.00	3889.00	4797.00	4710.00	5415.00	5400.00	10000
Be	10.00	9.00	10.00	9.00	10.00	10.00	100
P	17.00	19.00	16.00	17.00	10.00	11.00	-
Sb	0.40	0.40	0.40	0.40	0.40	0.40	-
Se	10.00	10.00	11.00	9.00	8.00	10.00	20
V	0.10	0.10	0.10	0.10	0.10	0.10	100

Table 4. Concentration of heavy metals in vegetables cultivated in main farmland areas of Sanandaj, Iran, in terms of mg Kg^{-1} dry weight

Element	Dill	Coriander	Radish root	Radish leaf	Recommended Max. level for vegetables[16]
Cd	0.006 (0.00-0.01)*	0.006 (0.00-0.01)*	0.011 (0.01-0.015)*	0.04 (0.00-0.10)*	0.20
Cr	3.93 (3.70-4.10)*	4.25 (4.00-4.50)*	4.00 (3.50-4.60)*	5.00 (4.20-5.80)*	2.30
Pb	0.48 (0.40-0.60)*	0.47 (0.40-1.00)*	0.42 (0.30-0.50)*	0.40 (0.20-0.60)*	0.30
Cu	10.70 (8.70-13.30)*	9.10 (7.90-11.00)*	3.15 (2.50-3.80)*	4.46 (4.10-5.00)*	40.00

* Values in the parenthesizes are minimum and maximum concentration of each element respectively.

showed a high level of Pb and Cr in all the vegetables from the cultivated area according to Food and Agriculture Organization (FAO) or the World Health Organization (WHO) guidelines.[16] The results showed that the levels of Pb in all the commodities were between 0.42 mg kg^{-1} in radish root and 0.48 mg kg^{-1} in dill with range of 0.3-0.5 and 0.4-0.6, respectively. Cd contents varied from 0.006 mg kg^{-1} in coriander and dill to 0.04 mg kg^{-1} (0.00-0.1) in radish leaf. In this study, maximum quantity of Cr was detected in radish leaf (5 mg Kg^{-1}) while dill had the lowest concentration (3.93 mg Kg^{-1}). Within the selected vegetables, the highest concentrations of Pb were noticed in dill. Furthermore, the highest concentration of Cu was observed in dill. Good agreement of these data was observed when the levels of Pb and Cd were compared to previously reported data by Fergusson.[17] For instance our values for lead were within the range of 0.05 to 6.7 mg/100 g, and 0.13 to 2.27 mg/100 g, reported for vegetables from Ireland and New York, respectively. For Cd, vegetables from Ireland showed a range of 0.005 to 0.06 mg/100 g, and those from New York had a range of 0.004 to 0.061 mg/100 g which was somewhat higher than our results. Besides, the concentrations of Cd in this study were below the range 0.09 to 0.26 mg/100 g reported for vegetables grown in Metropolitan Boston and Washington DC.[18] Furthermore, according to table 5, the Cd and Pb levels reported in this study were lower than those reported for vegetables in China[1] and India[19] and somewhat higher than those reported for vegetables in Uganda[6] for Cd and China for Pb.[20] The high contamination found in some vegetables might be closely associated to the pollutants in irrigation water, farm soil or due to pollution from the highways traffic.[21] All vegetables had lower levels of Cu than the permissible value (4 mg/100 g) in food provided by FAO/WHO.[16] Apart from its function as a biocatalyst, Cu is necessary for body pigmentation in addition to iron, the maintenance of a healthy central nervous system, prevention of anemia, and is interrelated with the function of zing and iron in the body.[22] Generally, plants contain the amount of Cu, which is inadequate for normal growth of plants. Application of micronutrient fertilizers and Cu-based fungicides may sometimes increase it to the alarming levels. In the present study, the mean levels of Cu were ranged from 3.15 to 10.70 mg kg^{-1}. The highest amount was found in dill and the lowest in radish root. From the results, it can be noted that the levels of Cu obtained in this study were comparatively lower than those reported by Begdeli and Seilsepour.[2]

As shown in table 4, the mean levels of Pb and Cr (0.44 and 4.3mg Kg^{-1}) were 1.46 and 1.87 times over the permissible levels set by FAO and WHO for human consumption. This trend was similar to those reported in previous study for Pb.[23] However, Cu and Cd amounts were within the acceptable levels. This trend was dissimilar to those reported in previous studies for Cd by Gupta et al.[19] and Ahmad and Goni[24] The

Table 5. Concentration ranges and safe limitations of heavy metals in vegetables from Iran and other countries (mg Kg^{-1} dry weight).

	Cu	Cr	Pb	Cd	Reference
Ranges					
Our study	5.8-8.28	3.85-4.75	0.33-0.68	0.003-0.03	
China	13.1-34.8	ND-1.07	6.07-15.6	0.14-4.47	1
China	8.65-317	0.08-15.4	0.18-7.75	0.036-0.18	20
India	8.63-27.94	3.70-9.03	11.97-22.09	2.05-2.91	24
India	15.66-34.49	34.83-96.30	21.59-57.63	10.37-17.79	19
India	10.95-28.58	5.37-27.83	3.09-15.74	0.5-4.36	25
Uganda	8.65-317	0.08-15.4	0.18-7.75	0.036-0.18	6
Iran	4.54-39.99	-	0.74-3.83	0.00-1.86	2
Greece	4.25-258	0.28-43.00	0.19-10.86	0.04-2.71	26
Iran	1.26-33.0	0.19-70.5	0.16-5.69	0.01-1.48	12
Safe limitations					
FAO/WHO (1984)	40	5	5	0.3	27
FAO/WHO (2001)	40	2.3	0.3	0.2	16
China EPA	20	0.5	9	0.2	28

FAO: Food and Agriculture Organization; WHO: World Health Organization; EPA: Environmental Protection Agency

variations of metal contents in these vegetables depend on the physical and chemical nature of soil and absorption capacity of each metal by plant, which is altered by innumerable environmental and human factors and nature of the plant.[25] The results showed that there was significant variations in the levels of these metals among the examined vegetables by Kruskal-Wallis test ($P < 0.001$).

Daily Intake Estimate of Heavy Metals

Exposure of consumers and related health risks are usually expressed as provisional tolerable daily intake (PTDI), a reference value established by Joint FAO/WHO.[27] Table 6 represents the estimation of each heavy metal intake through consumption of the studied foodstuffs.

The National Nutrition and Food Research Institute of Iran have estimated that the average consumption of edible vegetables is 218 g per day for every individual.[12] Results showed that the mean levels of Pb, Cu, Cr and Cd were 0.44, 6.85, 4.3 and 0.016 mg Kg^{-1}, respectively. Therefore, the DI of Pb, Cu, Cr and Cd could be 0.1, 1.5, 0.94 and 0.004 mg per day, respectively. Other studies from various countries have reported that the dietary intake for lead in adult was between 30 µg per day[4] and 427 µg per day.[29] For cadmium and copper, the estimated daily intake was from 4.6 µg to 30 mg per day, and 0.45 to 20 mg per day, respectively.[4,29] It could be concluded that estimation of daily intake for Cr and Cu in the present study was above those reported from other countries whereas the estimation for Cd and Pb are lower than the range.[1] Moreover, the estimated daily intake for the Pb and Cd in this study was below that those reported by FAO/WHO who have set a limitation for heavy metal intake based on body weight for an average adult (60 kg body weight). PTDI for Pb, Cd and Cu are 214 µg, 60 µg and 3 mg, respectively.[16] Thus, the authors believe that daily intake of these vegetables is not to cause a detrimental health hazard to the consumer.

Table 6. Mean heavy metal contents in vegetables and estimated of daily intake through consumption of vegetables in Sanandaj, Iran

	Pb	Cu	Cr	Cd
Mean concentration (mg Kg^{-1} dry weight)	0.44	6.85	4.30	0.016
Estimated daily intake (mg day^{-1})	0.10	1.50	0.94	0.004

Conclusion

The present study provided additional data on heavy metals pollution and also can help in risk assessment of consumer exposure to the expected heavy metal levels. A comparison of the levels of heavy metals in the studied vegetables was done with the permissible levels required for safe food. The results clearly showed a divergence from the permissible levels by FAO and WHO. High Pb contents were found in dill. High levels of Cd were found in radish leaf, and these amounts could be toxic and hazardous if taken in large quantities. However, the results of this study indicated that the daily intake of Cu and Cd through edible vegetables from farmland areas may not constitute a health hazard for consumers because the values are 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. Although the concentration of some metals were not excess than permissible levels, the authors strongly recommended that people should not take large quantities of those vegetables so as to avoid large accumulation of the heavy metals in the body. 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. Appropriate measures should be put in place by the companies to always treat their waste effluents before discharging them into the immediate environment, since this will control the levels of heavy metals in the soil.

Conflict of Interests

Authors have no conflict of interests.

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