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Heavy Metals Concentration in Vegetables Irrigated With Contaminated and Fresh Water and Estimation of Their Daily Intakes in Suburb Areas of Hamadan, Iran

Abdolmotaleb Seid-mohammadi (PhD)^a, Ghodrattollah Roshanaei (PhD)^b and Ghorban Asgari (PhD)^{c*}

^a Research Center of Social Determinants of Health, Department of Environmental Health Engineering, School of Public Health, Hamadan University of Medical Sciences, Hamadan, Iran

^b Research Center Modeling of Noncommunicable Diseases, Department of Biostatistics and Epidemiology, School of Public Health, Hamadan University of Medical Sciences, Hamadan, Iran

^c Research Center for Health Sciences, Department of Environmental Health Engineering, School of Public Health, Hamadan University of Medical Sciences, Hamadan, Iran

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* Correspondence

Ghorban Asgari (PhD)

Tel: +98 811 8380398

Fax: +98 811 8380509

E-mail: asgari@umsha.ac.ir

ABSTRACT

Background: This study was conducted to estimate the level of heavy metals accumulate in vegetables irrigated with contaminated water compared with those irrigated with fresh water in Hamadan, west of Iran in 2012.

Methods: Sixty samples of different vegetables i.e., parsley, tarragon, sweat basil and leek irrigated with contaminated water and thirty six samples from three different adjacent areas irrigated with fresh water as control were analyzed to determine heavy metals. The concentration of heavy metals i.e., lead, cadmium and chromium were achieved using atomic adsorption spectrophotometer.

Results: The mean concentration of lead, chromium and cadmium regardless of the kind of vegetables irrigated with contaminated water was 6.24, 1.57 and 0.15 mg/kg, respectively. Moreover, metals uptake differences by the vegetables were recognized to vegetable differences in tolerance to heavy metals. Based on the above concentrations the dietary intakes of metals through vegetables consumption were 0.004, 0.0008 and 6E-05 mg/day in infants for lead, chromium and cadmium, respectively.

Conclusions: The high concentration of these heavy metals in some vegetables might be attributed due to the use of untreated sanitary and industrial wastewater by farmers for the irrigation of vegetable lands. Therefore, treating of these wastewater and bioremediation of excess metals from polluted vegetation land could be considered.

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Introduction

Vegetables contain carbohydrates, proteins, as well as vitamins, minerals, fibers and trace elements since they constitute an important part of the human diet¹. In recent years their consumption is gradually increasing due to increased consciousness on the nutritional values of vegetables. But it also may contain different types of toxic elements at a wide range of concentrations².

Contamination of vegetables with heavy metals may be due to irrigation with water contaminated by industrial and domestic wastes, the addition of fertilizers and metal-based pesticides, as well as from deposits on different parts of the vegetables exposed to the air from polluted environments and application of sludge in vegetable lands^{1,3-5}. Industrial and municipal wastewater is mostly used for the irrigation of crops due to its easy availability, disposal problems and shortage of fresh water. In general, the application of wastewater led to changes in the physicochemical characteristics of soil and consequently heavy metal uptake by vege-

tables^{6,7}. These toxic elements due to their non-biodegradable properties and long biological half-lives readily accumulate in food chain and upon arrival in a critical concentration can cause metabolic and physiological effects on human kind^{2,4,5}. The consumption of heavy metals contaminated food can seriously deplete some essential nutrients in the body which are further responsible for decreasing immunological defenses and disabilities associated with malnutrition and high prevalence of upper gastrointestinal cancer rates⁸.

In recent years, research on heavy metals concentration in vegetables has attracted more attention in the world wide^{1,3, 5-12}. These results indicated that some vegetable species have been contaminated with heavy metals such as lead (Pb), copper (Cu), mercury (Hg), chromium (Cr) and cadmium (Cd) particularly in the areas irrigated with untreated municipal and industrial wastewater. Besides, investigation of heavy metal concentrations in edible vegetables in the

country has indicated that the amount of heavy metals in samples collected from investigated areas in the providence of Tehran, Hamadan, Kurdistan, Gorgan, Mazandaran and Isfahan was higher than the permissible limits for human consumption^{2,13-19}.

Hamadan City in the west of Iran is one of the most important areas for vegetables production. In this vegetable land areas approximately 40 tons of different kind of vegetables per day has produced seasonally. Abbasabad and Moradbaig rivers are two important water bodies elongated from south-west to north-east of Hamadan. These rivers are main part of the city drainage systems that drain approximately 75 million liters untreated sewage per day due to absence of sanitation waste water treatment facilities as well as some other industrial effluents²⁰. The farmers in peri urban areas of Hamadan use water from these rivers for irrigation of plants especially different vegetable spices. Such irrigation practices attain very good crop yields as it contains large amount of organic material and some inorganic elements essential for plant growth. But contaminations of soils and crops with these metals may have adverse effects on soil, crops and human beings. Thus, it is necessary to estimate the amount of toxic heavy metals concentration in plants grown in the suburb of Hamadan in order to assess the health risks.

The present study was conducted to investigate the levels of three toxic heavy metals i.e., Pb, Cr and Cd in the commonly grown vegetables i.e., parsley (*Petroselinum crispum*), tarragon (*Artemisia dracunculus*), leek (*Allium ampeloprasum*) and sweet basil (*Ocimum basilicum*) cultivation in suburb of Hamadan, Iran. The effect of irrigation with fresh and contaminated water was also studied to observe the concentration of accumulated metals to which human being are exposed. Another aim was to illustrate the difference of heavy metal concentrations in vegetables plantation in various regions and different seasons.

Methods

Area study

The field study was in the northeast suburb areas of Hamadan City, located in the west of Iran. An investigation survey was conducted in suburb areas of Hamadan to identify the locations where wastewater from domestic sewage is currently used for irrigation of vegetable crops. Interviews with government authorities and farmers helped to identify areas where wastewater irrigation has become common practice for at least 10 years (polluted sites), and where the irrigation water can be clearly sourced to fresh water (control sites) (Figure 1). A total of sixty samples of some commonly grown vegetables, i.e., parsley, tarragon, leek and sweet basil were taken from five different adjacent sites in that they irrigated with contaminated water. Similarly, thirty six samples from three different agricultural fields irrigated with fresh water were taken as control. The vegetable samples were collected randomly across the field during spring (June, 2012), summer (August, 2012) and autumn (September, 2012).

Plant sampling, preparation and analysis

Samples of edible portions of selected vegetables (2 kg each) were collected from the farmland in each appointed locations and they were taken in plastic bags to laboratory

within 2 h of harvesting. These were properly washed using cleaned tap water and rinsed with deionized water to remove any soil particles attached to the plant surfaces. Samples were then separately dried in an oven at 70-80 °C for about one day to determine their dry weight. The dry samples were crushed in a micro grinding mill and the resulting powder was passed through 2 mm sieve. The sieved samples were kept at ambient temperature in clean and dry high density polyethylene bottles before preparation for analysis. For each vegetable, samples (2 gr each) were accurately weighted and prepared well with adding 15 mL of (HNO₃: H₂SO₄: HClO₄; 5:1:1) into an acid washed beaker and digested at 80 °C until a transparent solutions were obtained by Akan et al.⁷. The resulting solution was cooled and filtered through a Whatman No. 541 filter paper into a volumetric flask and finally maintained to 50 mL with deionized water. Determination of heavy metals such as Pb, Cd and Cr was achieved using an atomic adsorption spectrophotometer (ALPHA4, Chem Tech Analytical, England) according to standard methods for the examination for water and wastewater²¹. Mean, standard deviation and range were calculated by SPSS 16.0 software. Linear regression analysis was used to determine the relationship between the concentration of heavy metals in vegetable samples with different locations, seasons and irrigated water. A probability level of $P < 0.05$ was considered statistically significant.

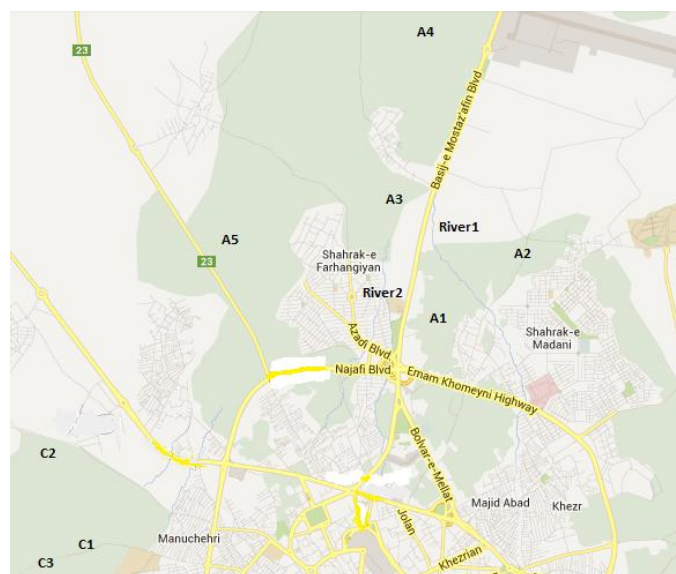


Figure 1: Map of experimental sites (A=Contaminated sites, C=Control sites) in suburban areas of Hamadan, Iran

Calculation of daily intake of metals (DIM) through vegetables

The DIM through vegetables (mg/day) was calculated by multiplying daily vegetable consumption (kg/person. day) with vegetable heavy metal concentration (mg/kg) were calculated as the following equation:

$$DIM = \frac{M \times F \times D}{A}$$

Where M, F, D and A are the heavy metal concentration (mg/kg), conversion factor, daily intake of vegetables and average body weight, respectively. The conversion factor 0.085 was used to convert fresh green vegetable weight to dry weight, as described by Arora, 2008⁵ and Harmanescu, 2011²². The average daily vegetable intakes for adults and

infants were 0.345 and 0.235 Kg/person/day, respectively²³. The average adult and infant body weights were 55.9 and 32.7 kg, respectively, as used in previous studies^{5,23}.

Results

Data for the three heavy metals concentration in different vegetables irrigated with water contaminated with wastewater and vegetables irrigated with fresh water are presented in Figure 2. It can be clearly observed that the concentration of Pb were in the ranged from 4.5 to 7.55 mg/kg in the vegetables irrigated with contaminated water, and were significantly higher ($P<0.001$) than the vegetables irrigated with fresh water, which were in the ranged from 0.6 to 2 mg/kg (Figure 2(a)). The Cd concentration varied between 0.07 to 0.33 mg/kg in the vegetables irrigated with contaminated water and were significantly higher ($P<0.001$) than the vegetables irrigated with fresh water (Figure 2 (b)). The concentration of Cd in fresh water irrigated vegetables were in the ranged from 0 to 0.01mg/kg. The results shown in Figure 2 (a and b) indicated that in all vegetables which irrigated with contaminated water, concentrations of Pb and Cd were exceeded the maximum permissible value (FAO/WHO)²⁴. Moreover, a similar trend was observed for Cr, i.e., maximum accumulation was obtained in contaminated water irrigated vegetables and minimum in fresh water irrigated samples ($P<0.001$). The comparison of mean concentration of Cr reported in the vegetables during the present study with the maximum permissible limits (FAO/WHO)²⁴ showed that it concentration were higher in both parsley and sweat basil irrigated with contaminated water, however were below the maximum permissible limits in tarragon and leek. As shown the concentration of Cr in different vegetables were from 0.7 to 2.9 mg/kg and from 0 to 0.41 mg/kg in contaminated and fresh water irrigation vegetables, respectively. The mean concentrations of Pb, Cr and Cd were 6.24, 1.55, 0.15 mg/kg and 1.11, 0.1, 0.01 mg/kg in vegetables irrigated with contaminated and fresh water resources, respectively (Figure 2).

The relationships between the individual heavy metals and different vegetables grown in the suburb areas of Hamadan shown in Figure 2 indicated that the mean concentrations of Pb in parsley, tarragon, leek and sweet basil were 6.42 ± 1.56 mg/kg, 5.54 ± 0.48 mg/kg, 6.6 ± 0.51 mg/kg and 6.4 ± 0.47 mg/kg, respectively. The trends of Pb concentrations in different vegetables were in the order of leek>parsley>sweet basil>tarragon. From Figure 2 (b), the mean concentrations of Cd in parsley, tarragon, leek and sweet basil were 1.27 ± 0.08 mg/kg, 0.22 ± 0.01 mg/kg, 0.11 ± 0.06 mg/kg and 0.16 ± 0.01 mg/kg, respectively. Besides, Cd accumulation in plants irrigated with contaminated water in the following order of ranking tarragon>sweet basil>parsley>leek. For Cr, the amount of accumulation in different vegetables was as follows: parsley>sweet basil>leek>tarragon. The mean concentrations of Cr were 2.08 ± 0.54 mg/kg for tarragon, 1.7 ± 0.34 mg/kg for sweet basil, 1.37 ± 0.3 mg/kg for leek and 1.24 ± 0.35 mg/kg for tarragon (Figure 2 (c)).

The daily intakes metals results of heavy metals estimates according to the average vegetable consumption for both adults and infants are shown in Tables 1 and 2, respectively. As shown, the DIM values for different metals were high when based on the consumption of vegetables irrigated

with contaminated water than those irrigated with fresh water. The highest intakes of Pb, Cr and Cd were form the consumption of leek, parsley and tarragon, respectively irrigated with contaminated water. Likewise, the DMI of Pb, Cr and Cd ranged from 0.003 to 0.0004, 0.001 to 3.1E-05 and 0.0001 to 1E-06, respectively for adults, while ranged from 0.003 to 0.0007, 0.001 to 3.6E-05 and 0.0001 to 1E-06, respectively for infants.

The highest Pb, Cr and Cd concentrations in different vegetables irrigated with contaminated water were observed at zones 1, 1 and 4, respectively (data not shown). However, the field date showed that there were not any significant variation between sits and heavy metals concentration in different vegetables ($P>0.05$). Mean concentration of Pb, Cd and Cr in vegetables were higher than the permissible limits²³ at all the areas irrigated by contaminated water respectively (data not shown).

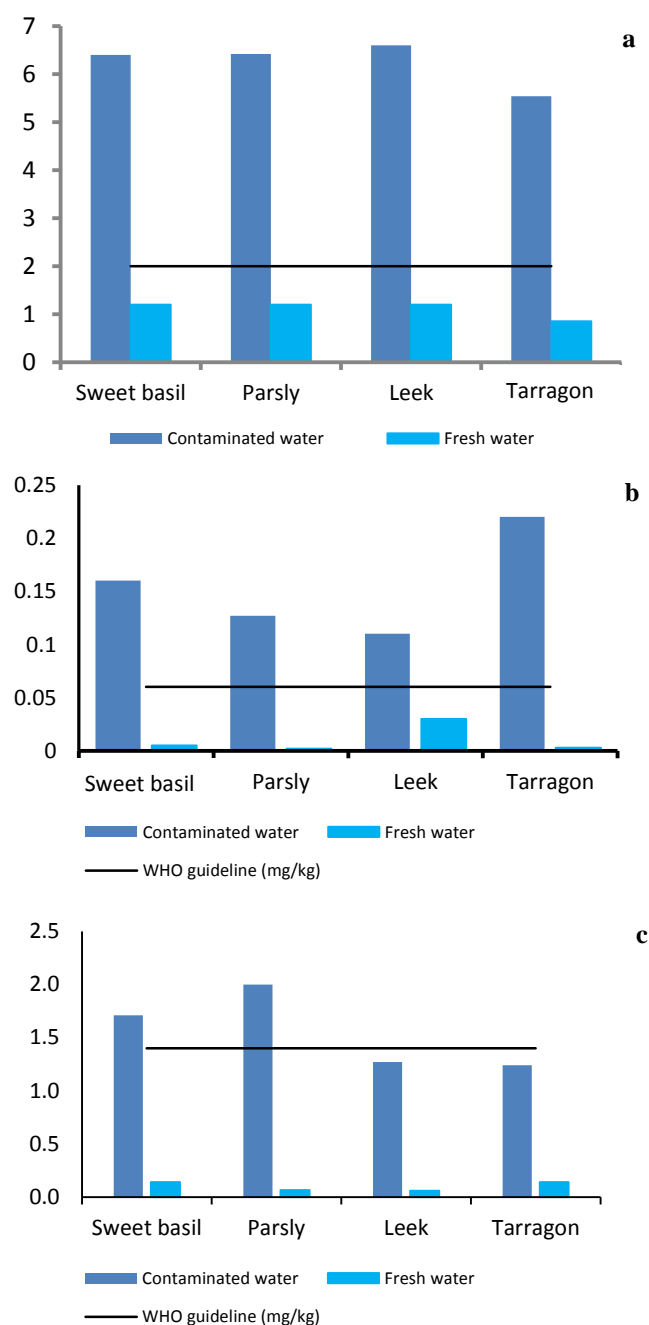


Figure 2: Heavy metal concentrations in different vegetables irrigated with contaminated water and fresh water: (a): pb, (b): Cd and (c):Cr

Discussion

The heavy metal concentrations in different vegetables irrigated with contaminated water were compared with those irrigated with fresh water in suburb areas of Hamadan. The results shown in Figure 2 clearly indicated that the concen-

tration of all the heavy metals were several folds higher in the contaminated water irrigated vegetables as compared to those irrigated with fresh water. The mean concentrations of Pb and Cr recorded during the present study were considerably higher than the earlier reports of Samarghandi et al.¹⁶ in some vegetables from the same areas.

Table 1: Daily intake of metals (mg) for individual heavy metals in different vegetables irrigated with contaminated water

Vegetables	Lead		Chromium		Cadmium	
	Adults	Infants	Adults	Infants	Adults	Infants
Parsley	0.0030	0.0040	0.0010	0.0012	6.7E-05	7.6E-05
Sweet bail	0.0030	0.0048	0.0008	0.0010	8.4E-05	9.6E-05
Leek	0.0030	0.0040	0.0007	0.0008	5.7E-05	6.6E-05
Tarragon	0.0030	0.0030	0.0006	0.0007	0.0001	0.0001

Table 2: DIM (mg) for individual heavy metals in different vegetables irrigated with fresh water

Vegetables	Lead		Chromium		Cadmium	
	Adults	Infants	Adults	Infants	Adults	Infants
Parsley	0.0006	0.0007	3.3E-05	3.8E-05	1.0E-06	1.0E-06
Sweet bail	0.0007	0.0007	7.3E-05	8.4E-05	3.0E-06	3.0E-06
Leek	0.0006	0.0007	3.1E-05	3.6E-05	1.0E-05	1.0E-05
Tarragon	0.0004	0.0005	7.3E-05	8.4E-05	1.0E-06	1.8E-06

The concentration of Pb and Cr were increased by 30 and 56 percent during sixteen years from 1996 to 2012¹⁶. Our results show agreement with previous studies showing eminent concentrations of heavy metals in edible parts of vegetables with the application of wastewater for irrigation²⁵⁻²⁷. Moreover, results from present and previous studies demonstrate that the vegetables grown on wastewater irrigated soils are generally contaminated with heavy metals, which pose a major health concern^{22,27}. However, the comparison of mean heavy metal concentrations in vegetables irrigated with contaminated water from the data of countries indicated that the values of the present study were manifold lower than the levels observed at Sannandaj¹⁹, but higher than those recorded at Gorgan¹⁴ and Shahrood¹⁸. Through regular and systematic investigation, the reason for heavy metals accumulation in vegetable land soils is mainly from the natural and man-made sources^{2,10,13,28}.

In the case of contaminated water irrigated areas of Hamadan City, all urban sanitary wastewater and much of industrial wastewater, which are not completely treated, are directly discharged into vegetable lands, incurring obvious contamination of metals in vegetable samples. In these vegetable lands, it has a history of more than 25 years using vegetables contaminated water irrigation. When sewage and industrial wastewater that are not suitable compared with the field irrigation water quality standard is used for irrigation during a long time, the heavy metal concentrations in plants and crops obviously exceeded the maximum permissible limits. Similar to our results, many other studies indicated that the application of contaminated water generally led to changes in the physicochemical properties of soil and consequently heavy metals uptake by crops, fruits and vegetables^{5,7,10,29}. For instance, the concentration of Pb, Cd and Cr in the vegetable land soils with long term application of untreated wastewater was 4, 10 and 120 times higher than the soil background values, respectively. Besides, other studies shown that heavy metal concentrations in vegetables grown in Tehran², Kurdistan¹⁹ and Isfahan¹⁵ provinces are increasing gradually related to unreasonable wastewater irrigation.

There were considerable variations in heavy metals uptake by different types of vegetables (Figure 1). The trend of metal concentrations in the vegetables would be a function of trace metals concentration in the soil and their absorption in plant tissues^{5,22,30}. According to the highest concentration of Pb, Cr and Cd were shown in leek, parsley and tarragon, respectively. Similar studies carried out by Khan et al.⁸ and Cheraghi et al.¹³ shows that heavy metal concentration varied among different vegetables species, which may be attributed to differential absorption capacity of vegetables for different heavy metals. However, the field data shows that there were not any significant variation between vegetation lands and heavy metals concentration in different vegetables. Mean concentrations of Pb, and Cd in vegetables were higher than the permissible limits²⁴ at all the areas irrigated by contaminated water. Very few studies shown that there were positive correlation between vegetables heavy metal concentrations and seasonal variations due to increasing of soil pH during summer which maintained higher concentrations of heavy metals in the topsoil layer and higher frequency of irrigation during this season compared to other seasons^{6,16}.

In this study, ANOVA tests showed that variations in concentration of heavy metal samples in vegetables were not significant ($P>0.05$) due to different seasons which agreed with the observation of Dadban et al.¹⁴ and Singh et al.³¹ for various heavy metals in vegetables sampling in different seasons. However, the results reported by Sharma et al.²⁶ and Samarghandi et al.¹⁶ suggested that there were significant variations in the concentrations of heavy metals in vegetable samples between areas and seasons. Akan et al.⁷ reported high level of heavy metals in all the vegetable organs in the dry seasons than the rainy seasons due the use of untreated effluents from tanneries and textile by farmers in the dry seasons.

In order to observe the health risk of any pollutant, it is very important to estimate the level of exposure, by detecting the routes of exposure to the target organism. Among several possible pathways of exposures to humans, the DIM was estimated according to the average vegetables consump-

tion for both adults and infants. The results of this study regarding DIM suggested that the consumption of vegetables irrigated with contaminated water is high, but is nearly free of risks, as the dietary limits of Pb, Cr and Cd in adults can range from 0.02 to 0.04 mg, 0.2 to 0.3 mg and 0.01-0.03 mg³², respectively.

Conclusions

Irrigation by water contaminated with untreated sewage has increased the heavy metal concentrations in different vegetables in the study areas. The mean concentrations of Pb and Cd in all vegetables irrigated by contaminated water were higher than maximum permissible limit set by FAO/WHO²² for vegetables, while Cr concentration was higher than maximum permissible limits only in parsley and sweat basil irrigated by contaminated water. However, the daily intakes of studied metals were much less than the dietary limits at present, indicating that there is a relative absence of health risks associated with the ingestion of contaminated vegetables. But the authors strongly suggested that people living in these areas should not eat large quantities of vegetables, so as to avoid excess accumulation of heavy metals in the body. To avoid accumulation of metals into the vegetables not only sanitary and industrial wastewater should not be drained into the rivers without complete treatment but also it is imperative to utilize alternative methods for extraction of excess metals from polluted areas.

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Conflict of interest statement

The authors declare that they have no conflicts of interest.

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