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Pollution of Heavy Metals in Some Farms of Torbat-E Jam, Khorasan Razavi Province, Iran

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ABSTRACT: Agricultural intensification is associated with the use of great amounts of agrochemicals that may result in the accumulation of metals in soils, and –subsequently— agricultural products and food chain. Nowadays, this is a major concern for many Iranian agricultural products, necessitating scientific researches on the issue. Therefore, the present study has been conducted to evaluate the level of metal contamination in some agricultural products of Torbat-e Jam, Iran, with the following purposes: (i) to determine concentrations of cadmium (Cd), nickel (Ni), and lead (Pb) in melon (*Cucumis melo* var. inodorus), sugar beet (*Beta vulgaris*), and maize (*Zea mays*) as well as water and soils of some farms in Torbat-e Jam, Iran; and (ii) to examine chemical fertilizers as a possible source of heavy metals' contamination. To do so it has taken some samples from soils, irrigation waters, chemical fertilizers, and crops, measuring their heavy metals contents by means of atomic absorption spectrometry. Results show that heavy metals' concentrations in groundwater and soil were lower than the adopted global standards. Among fertilizers, only Cd content of triple-superphosphate was higher than the standards, leading to a substantial buildup of Cd in the soil, compared to Pb. The greater use of potassium fertilizer has increased Pb concentration in the soil samples of maize farms during the growing season. Among all elements, Pb had the greatest transfer coefficient. It seems that current farm management practices as well as excessive use of chemical fertilizers may further the contamination and loss of soil quality in agricultural systems of the region.

Keywords: cadmium, concentration factor, fertilizer, melon, transfer coefficient

INTRODUCTION

Nowadays, accumulation of heavy metals and contamination of agricultural soils, both of which are chiefly caused by human activities, are among the most important environmental issues, worldwide. Concurrent with development and expansion of industry, mining activities, and overuse of chemical fertilizers, heavy-metal pollution of agricultural soils has raised a major environmental concern in human societies, especially in developing countries (STAP, 2012). Unfortunately, the inherent soil fertility in most of these areas is already too low, reinforcing the farmers' tendency to use fertilizers frequently and excessively (Pacheco et al., 2001). Apart from enhancing soil fertility, fertilizers cause environmental pollution (Kelly & Tate, 1998), because in addition to creating problems such as

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leaching and eutrophication of surface waters, they are regarded as a source of heavy metals, considered harmful pollutants for human health as well as the environment (Rui et al., 2008).

Worrying heavy metals in fertilizers include arsenic (As), lead (Pb), cadmium (Cd), and to a lesser extent, zinc (Zn) and nickel (Ni) (McCauley et al., 2009). Exposure to such heavy metals is a critical threat to human health (Jarup, 2003). Although the concentration of these elements in fertilizers may seem negligible at first, it should be noted that their accumulation and deposition in the soil, especially in agricultural fields, take place gradually. Thus, after several years of continued use of chemical fertilizers and plant cultivation in soil, contaminated with heavy metals, their concentrations may reach levels that threaten human food security, as these elements could enter the food chain through plant uptake (Frossard, 1993). Among all heavy metals, Cd is of particular importance due to its considerable presence in phosphorus fertilizers as well as its high mobility, because it is present in contaminated soil as free ions or ions that are easy to get dissolved in soil water and thus is readily available for plants (Hardyman & Jakoby, 1984; Smolders & Mertens, 2013).In fact, applying excessive amounts of chemical fertilizers may increase the possibility of heavy metals' building up in farm soils and plant toxicity (Ju et al., 2007), likewise the case of durum wheat (Atafar et al., 2010). Among all, phosphate fertilizers can be regarded as an important source of heavy metals' contamination in soils and water resources (Nicholson et al., 2003; Boudaghi et al., 2012). As such, there is growing concern about their long-term use in agricultural soils (Cheraghi et al., 2012). Therefore, some countries have tried to set regulations on fertilizer quality by setting some standards for heavy metal limits in fertilizers (Tables 1 and 2). There is also a significant difference among plant species in terms of their ability to absorb heavy metals. For example, leafy vegetables absorb Cd more than grasses, and about 12-18% of total Cd absorbed by cereals is imported to grains (Williams & David, 1973).

Table 1. Comparison of the standards, adopted for heavy metals contents in fertilizers in China, Canada, Japan, and Iran (adopted from Wallis-Lage et al., 2012)

Heavy metal	China	Canada		Japan	Iran
	mg/kg fertilizer	Maximum acceptable cumulative metal additions to soil over 45 years (kg metal/ha)	Maximum acceptable limits of heavy metals (mg/kg dry matter)	mg/kg byproduct of phosphate fertilizers	mg/kg fertilizers
Cadmium (Cd)	8	4	20	8	25
Nickel (Ni)	-	36	180	-	-
Lead (Pb)	100	100	500	100	25
Chromium (Cr)	500	210	-	500	-

Table 2. The certified value of some trace elements in multi-nutrient fertilizers, based on SRM 695 (Mackey et al., 2007)

Trace element	Certified concentration (mg/kg)
Arsenic (As)	200
Cadmium (Cd)	16.9
Cobalt (Co)	65.3
Chromium (Cr)	244
Molybdenum (Mo)	20
Nickel (Ni)	135
Lead (Pb)	273

Recently, there has been much attention on soil contamination of agroecosystems to heavy metals in different parts of Iran. Various causes have been identified for this contamination, such as geological sources (Mohammadpour et al., 2016), mining activity (Karbassi et al., 2014), oil exploitation and extraction (Karbasi et al., 2015), irrigation with sewage (Alizadeh et al., 2009), and agrochemical inputs, especially fertilizers (Atafar et al., 2010; Boudaghi et al., 2012; Cheraghi et al., 2012; Nouri et al., 2008; Nazemi and Khosravi, 2011). Many of studies in Iran have been performed mainly in central and western parts of Iran. A wide area of agricultural fields in Eastern parts of Iran, in particular Torbat-e Jam County, are cultivated for melon (*Cucumis melo* L.), sugar beet (*Betal vulgaris* L.), and maize (*Zea mays* L.). Typically, large amounts of different chemical fertilizers like urea, potassium sulfate, and triple superphosphate are used in this area to maximize crop production. These chemical fertilizers may contain high levels of cadmium (Boudaghi 2012), along with other heavy metals, which can act as an important source of heavy metal contamination in the soils and plants of the area. Therefore, the main purpose of this study has been to determine the concentration of heavy metals in soil, water, and important crops of Torbat-e Jam and find their possible correlation with the amount and type of the commonly-used fertilizers.

MATERIALS AND METHODS

In order to investigate the contamination of melons, maize, and sugar beet fields with heavy metals, namely cadmium, nickel, and lead, this study has been conducted in 2012 in Torbat-e Jam county, northeast of Khorasan Razavi Province, Iran. The area is 8184 square kilometers, from 60° 15´E to 60° 30´E, and 34° 35´N to 35° 47´N (Figure 1). The average elevation is 950 m above sea level and the climate, cold and dry, with an average annual precipitation of about 100.6 mm. The average cultivation area in Torbat-e Jam is about 66900 ha, with 98% of the total cultivated lands being under irrigation, producing a wide range of crops, the main examples of which include wheat, melons, sugar beet, tomato, and canola (Agriculture Organization of Khorasan Razavi, 2014).

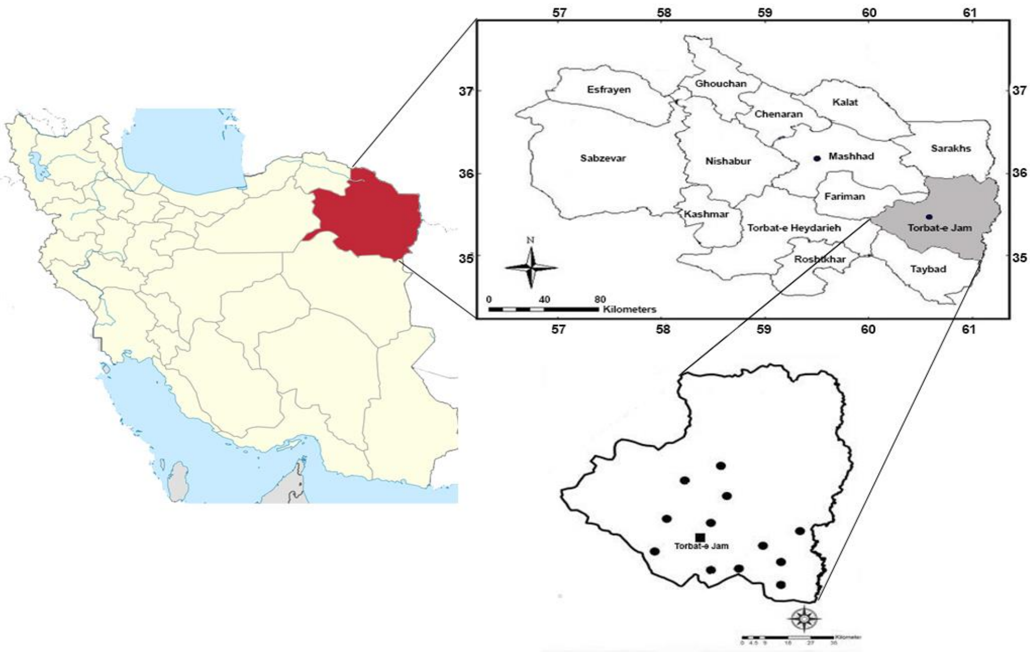

Fig. 1. Geographical location of Torbat-e Jam county, showing the distribution of the sampled points (⬤) as well as the city of Torbat-e jam (■).

In the entire county, 12 locations, and in each location, one field for each of the crops, were selected in random (Figure 1). In each field, a combined soil sample was extracted from a depth of 0-30 cm, both before planting and after harvest. Soil samples were air-dried, ground, and passed through a 2-mm sieve. In each field, some samples were taken from three commonly used fertilizers (triple super phosphate, urea, and potassium sulfate). For water samples, the pH of water was reduced to less than 2 with concentrated nitric acid after sampling, and all samples were kept cool until examination. The concentrations of heavy metals (cadmium, nickel, and lead) in the soil, manure, leaves, and fruit samples, as well as samples of water (filtered with Whitman filter paper) were measured via atomic absorption spectrometry (graphite furnace method) (Gupta 2000). Other soil properties such as pH (pH of saturated soil paste using pH meter), and the electrical conductivity (EC) (1:1 extract EC at a 25 °C with EC-meters) (Tandon 2005) were measured in Soil Laboratory of University of Birjand.

In order to determine the transfer rate of heavy metals (cadmium, nickel, and lead) from the soil to the plant, Plant Concentration Factor (PCF) was calculated based on Equation (1) below (Khan et al. 2008; Xue et al. 2012):

$$PCF = \frac{C_{Plant}}{C_{soil}} \quad (1)$$

In which, C_{plant} and C_{soil} (mg kg^{-1} dry weight) are concentrations of heavy metals in above ground parts of plants and soil, respectively. Transfer coefficient of the plants was determined, according to Equation (2) (Xue et al. 2012):

$$\mathrm{TF} = \frac{\mathrm{C_{Shoot}}}{\mathrm{C_{Root}}} \quad (2)$$

where C_{shoot} and C_{root} are the concentration of heavy metal in the extracts from above and below the ground on dry weight (DW) basis, respectively. All other required data such as type, rate, and method of applying fertilizers were gathered by means of questionnaires.

All data was analyzed, using SPSS 16.0 statistical package, and were subject to normality test of Kolmogorov-Smirnov. Differences at $P<0.05$ level were considered significant. The figures were creted, using the Microsoft Excel (2010).

RESULTS AND DISCUSSION

According to the questionnaires, there was not any difference among the studied farms in terms of the type of used fertilizers (urea, triple superphosphate, and potassium sulfate); however, there was a significant difference ($p<0.05$) between consumption rate of urea and other fertilizers among the fields, with greater usage of fertilizers in maize fields (Tables 3 and 5).

As nickel contents in the soil of all farms did not vary significantly before planting and after fertilization, it seems that chemical fertilizers are the source for plants' nickel absorption; however, we did not measure it due to its negligible amounts in the plants. The mean concentration of nickel in the soil of studied area was much lower than the value, reported by Mohammadpour et al. (2016) for Hamedan farms, which can be attributed to different soil parental materials. According to fertilizers analysis (Table 4), the concentrations of Cd and Pb in these three widely used fertilizers were in the following order: triple superphosphate > potassium sulfate > urea. Considering the concentration of Cd and Pb in these fertilizers (Table 4) as well as the average amount of fertilizers' application (Table 5), if both metals are only adsorbed by the soil, it is expected that the average values of Cd and Pb, added to the soil in maize fields, are more than those of sugar beet and melon fields.

Only triple superphosphate fertilizer had a Cd concentration higher than standard concentrations of CDFA (4 mg/kg), with the concentration of Cd in other fertilizers being insignificant and less than most valid standards of the world. Thus, the use of these fertilizers should not actually cause any increase or decrease in the amount of Cd in the soil, water, and plants system. Cd concentration in commonly used triple super phosphate and urea fertilizers in this region was lower than another report for western part of Iran (0.03 and 12.2 mg/kg in urea and triple super phosphate, respectively, Nouri et al. 2008).

In the whole region, pH of water ranged between 8.5 and 9.1. The maximum concentration of Ni in the water, used for irrigation in the study area, was 0.03 mg/l (Figure 2), which was lower than FAO limits (Ayers & Westcot 1994). The Cd concentration in water used in melons and maize farms were almost similar, again below FAO limits (0.01 mg/l, Ayers & Westcot 1994), while the Cd concentration in the water used for irrigation of sugar beet farms was up to 0.02 mg/l, which was greater than FAO limits and higher than the amount, reported for northern parts of Iran (Boudaghi et al. 2012). The maximum lead concentration in irrigation water used in the studying area was 0.15 mg/l, which was below FAO limits (5 mg/l; Ayers & Westcot 1994) as well as lead levels in irrigation water for vegetable farms of Shahroud, Iran (7.55 mg/l; Nazemi & Khosravi 2011).

Table 3. Results of one-way analysis of variance for fertilizer consumption among different maize, melon, and sugar beet fields in Torbat-e Jam, Iran.

Fertilizer	S.O.V	df	Means of Squares	F value	Significant level
Urea	Between groups	2	15244.44	17.81	0.003
	Within groups	6	855.55		
	Total	8			
Triple super-phosphate	Between groups	2	477.77	1.13	0.38
	Within groups	6	311.11		
	Total	8			
Potassium sulfate	Between groups	2	2233.33	2.83	0.13
	Within groups	6	788.88		
	Total	8			

Table 4. The concentration of Cd and Pb (mg/kg, ±SEM) in fertilizers, widely used in the studied region fields

Fertilizer	Cd	Pb
	mg/kg	
Urea	0.01± 0.005 b	0.01 ± 0.003 c
Triple super phosphate	5.48 ± 0.01 a	2.98 ± 0.06 a
Potassium sulfate	0.031 ± 0.005 b	1.047 ± 0.008 b

In each column, the means with similar letters did not differ significantly at 0.05 probability level.

Table 5. Possible amounts of Cd and Pb (mg/ha/ year, ±SEM), added to the soil system of different crop fields in Torbat-e Jam, Iran, as a result of fertilization

Crop	The average used fertilizer (kg/ha)			Heavy metals added to soil	
	Urea	Triple super phosphate	Potassium solfate	Cd_{total}	Pb_{total}
Melon	120±25	106.66±10	66.67±30	587.75	388.84
Sugar beet	220±10	120±10	66.67±30	661.86	429.6
Maize	220±30	120±25	106.67±10	663.1	471.47

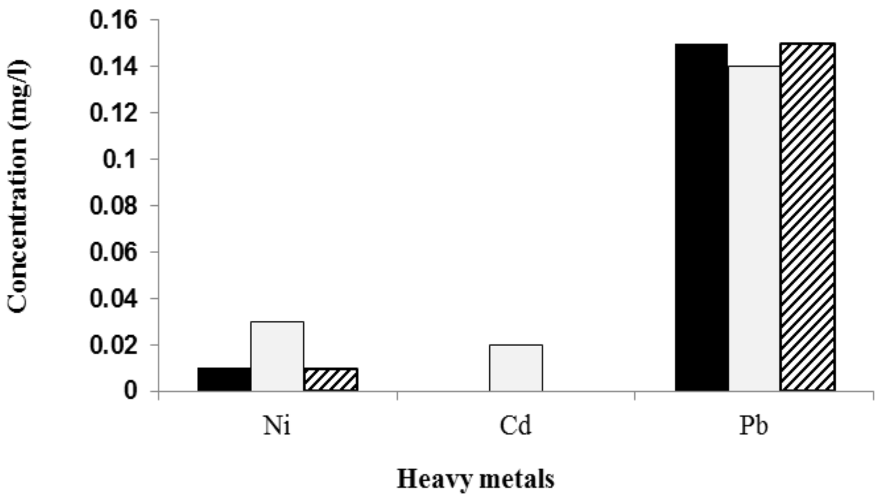

Fig. 2. Average concentration of heavy metals in water, used to irrigate melons (dark columns), sugar beet (light gray columns), and maize (dashed columns) fields of Torbat-e Jam, Iran

Overall, results showed that the concentration of heavy metals (Ni, Cd, and Pb) in all water resources, at least in these areas, was very low. So, irrigating water, at least in the short term, may not play a significant role in increasing the accumulation of these heavy metals in the soil.

According to soil analysis in the region, the average soil Ec was 4.91 dS/m (3.8-6.59) and the average soil pH, 7.83 (7.7-7.9). The Ni_{total} in the soil had no significant variation before and after fertilization, whereas soil Cd and Pb concentrations were differed considerably between these periods (Table 6), indicating that the amount of Ni in used fertilizers was very low, hence not a contributing factor in raising Ni level in the soil. Ni, as a heavy metal, had mainly more mobility in water and soil at low soil pHs; however, this element does not seem more toxic than heavy metals such as Cd and Pb, and there are fewer reports of contamination of chemical fertilizer to this element. In turn, Ni acts as an essential micronutrient for healthy plant growth and even some soils may have deficiency in the Ni content (Wuana1 & Okieimen 2011). In contrast, consumption of fertilizers contributed to the increasing concentrations of Cd and Pb in soils under cultivation of all three crops. The increases in the soil Cd at the end of growing season were 87%, 92%, and 90.9% for melon, sugar beet, and maize, respectively, while the corresponding amounts for soil Pb corresponded to 19.4%, 16.7%, and 35.4% (Table 6). The higher Pb level in soil of maize fields is probably due to using greater amounts of potassium sulfate in this crop than in melons and sugar beet fields.

The amounts of total heavy metals added to the soils under cultivation of melon, sugar beet, and maize through fertilization were 0.87, 0.69, and 1 mg/kg for Cd; 0.7, 0.5, and 0.5 mg/kg for Ni; and 2.74, 2.7, and 6.73 mg/kg for Pb, respectively (Table 6). Their contents in the shoots of melon, sugar beet, and maize were also 0.17, 0.26, and 0.2 mg/kg for Cd; 0.53, 1.21, and 2.1 for Ni; and 3.53, 6.2, and 11.06 mg/kg for Pb, respectively, which seemingly reflects their absorption by plants from their previous reserves in the soil. In comparison, maximal permissible addition (MPA) of Pb, Cd, and Ni, according to the data of Dutch ecologists (Crommentuijn et al. 1997), are 55, 0.76 and 2.6 mg/kg, respectively.

Table 6. The average concentrations of Ni, Cd, and Pb (mg/kg) in soils of melon, sugar beet, and maize fields, before (B.P) and after (A.P.) fertilizer application, in Torbat-e Jam region in 2012

Crop	Heavy metal (total amount)	Sampling time	Mean	Standard deviation	P-Value
Melon	Ni	B.P.	6.4	0.73	0.21 ns
		A.P.	7.1	0.65	
	Cd	B.P.	0.13	0.15	0.00 *
		A.P.	1.0	0.10	
	Pb	B.P.	11.36	0.79	0.02 *
		A.P.	14.1	0.13	
Sugar beet	Ni	B.P.	4.3	0.29	0.16 ns
		A.P.	4.8	0.21	
	Cd	B.P.	0.06	0.06	0.003 *
		A.P.	0.75	0.10	
	Pb	B.P.	13.4	0.97	0.007 *
		A.P.	16.1	0.9	
Maize	Ni	B.P.	5.3	0.63	ns 0.27
		A.P.	5.8	0.59	
	Cd	B.P.	0.1	0.02	* 0.001
		A.P.	1.1	0.32	
	Pb	B.P.	12.25	0.7	* 0.04
		A.P.	18.98	2.8	

ns and * means non-significant and significant at 0.05 probability level, respectively

The nickel, cadmium, and lead contents in roots and shoots of maize were higher than those of melon and sugar beet (Figure 3). In another study in Hamedan province, Iran, Mohammadpour et al. (2016) also found that Ni concentration in corn was higher than other crops (wheat, barley, alfalfa, and potatoes). Averaging on all crops, the concentration of these elements in the roots was greater than the shoot (Figure 3). In case of lead, it is reported that the plants roots can absorb high amounts of Pb, restricting its translocation to the aerial parts (Kumar et al. 1995). Hossain et al. (2007) in a survey of soil cadmium transfer to vegetables found that the accumulation of cadmium in the roots was higher than the shoots, too. Essential elements for plant growth are usually mobile in the plant system, whereas toxic and heavy elements are relatively immobile within a plant, getting accumulated mainly in the roots (Mireles et al. 2004)

The Ni content in melon fruit (Figure 3b) was below than the permissible limit (1 mg/kg; Misra & Mani 1991). The same story went for the amounts of cadmium in melon fruit and maize shoots (0.1 – 2.4 mg/kg; Misra & Mani 1991), while higher than the cadmium level reported in strawberry fruit in the western part of Iran (0.01 mg/kg; Cheraghi et al. 2012). In case of lead, its content in melon fruit was less than the permissible limit (1-13 mg/kg; Misra & Mani 1991) but higher than the reported value for strawberry (3.57 mg/kg; Cheraghi et al. 2012).

Sugar beet's tubers and maize shoots had Ni contents less than the permissible limit (5 mg/kg; Misra & Mani 1991). The concentration of Ni in maize plants in our study (2.1 mg/kg) was similar with the ones, reported by Mohammadpour et al. (2016) in Hamedan. Cd content in sugar beet tuber was also below the limits (1-0.1 mg/kg). In case of lead, its values in the tubers of sugar beet was less than the specified limits, in contrast to the maize shoots which stood higher (5-10 mg/kg; Alizadeh et al. 2009). Pb may enter life cycle from various sources and may contaminate the environment, leading to various disorders in organisms. A critical level (the maximum permissible amount) of 9 mg/kg (on a dry basis) of Pb concentration in plants has been reported (SEPA 2005). Given the average yields of melons, sugar beet and maize forage in Torbat-e Jam, which were 14702, 41000, and 44000 kg/ha, respectively (Agriculture Organization of Khorasan Razavi, 2014), it can be concluded that the total harvested amounts of nickel, cadmium and lead from farms soils were 7.7, 2.4, and 151.4 gr/ha for melon fruits; 49.6, 10.6, and 254.2 gr/ha for sugar beet tubers; and 92.4, 8.8, and 486.6 gr/ha for forages of maize crop, respectively.

Based on the results from means comparison of Plant Concentration Factor (PCF) of nickel and lead, but not nickel alone, transferring from the soil to the plant roots, there were significant differences ($p<0.05$) among the three crops**.** Transfer Coefficients (TC) from root to shoot in three crops showed significant differences ($p<0.05$) only for nickel, without any considerable variation in case of cadmium and lead (Table 7).

Transfer rate of heavy metals from soil to plants is related to their absorption by the root system. The trend of transfer rate of heavy metals from soil to melon roots was in the following order: Cd > Pb > Ni. For maize and sugar beet, this trend was similar: Pb > Ni > Cd (Table 8). Also, the order of metals, according to their transfer from root to consumable aerial parts of melon (fruits) and maize (forage) was Pb > Cd > Ni, and Pb > Ni > Cd. In potatoes, this trend has been reported as the following: Ar > Ni > Cd, Cu, Pb > Cr, Zn (Cheraghi et al. 2012). For some different vegetables in China, the trends of PCF for heavy metals were reported as Cd > Ni > Cu > Zn > Cr > Pb (Khan et al. 2008). The difference in metal concentration of various plants is due to their varying abilities in uptake and accumulation of heavy metals. Moreover, the difference among species in terms of growth period, growth rate, and physical and chemical properties of soils may affect heavy metal uptake (Verloo & Eeckhout 1990; Moseholm et al. 1992)

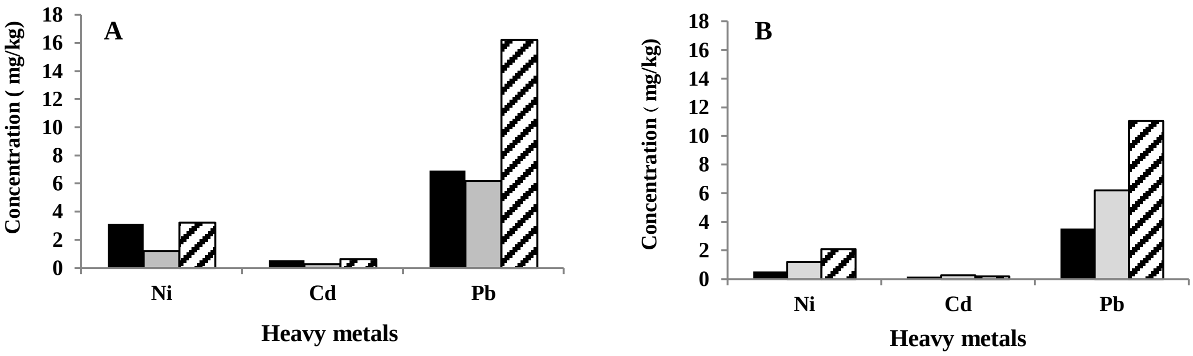

Fig. 3. Average concentrations of heavy metals in the roots (a) and edible parts (b) of melons (dark columns), sugar beets (light gray columns), and maize (dashed columns), harvested from the farms of Torbat-e Jam, Iran

Table 7. Analysis of variance of the transfer of nickel, cadmium, and lead from the soil to roots of melon, sugar beet, and maize, and from roots to consumable parts of melon (fruit) and maize (forage).

Heavy metal	Transfer path	S.O.V	df	M.S	F	Significance level
Ni	Soil to root	Treatment	2	0.07	13.9	0.006
		Error	6	0.005		
		Total	8			
	Root to consumable organ	Treatment	1	0.21	36.2	0.004
		Error	4	0.006		
		Total	5			
Cd	Soil to root	Treatment	2	0.03	0.81	0.48
		Error	6	0.04		
		Total	8			
	Root to consumable organ	Treatment	1	0.01	0.503	0.51
		Error	4	0.03		
		Total	5			
Pb	Soil to root	Treatment	2	0.18	30.34	0.001
		Error	6	0.006		
		Total	8			
	Root to consumable organ	Treatment	1	0.03	1.2	0.31
		Error	4	0.02		
		Total	5			

Table 8. The Plant Concentration Factors (PCF) and Transfer Coefficients (TF) of nickel, cadmium, and lead for melon, maize, and sugar beet

Crop	Parameter	Nickel		Cadmium		Lead	
		Mean	Standard deviation	Mean	Standard deviation	Mean	Standard deviation
Melon	PCF	0.44	0.04	0.52	0.18	0.5	0.13
	TF	0.18	0.08	0.25	0.25	0.51	0.005
Maize	PCF	0.55	0.06	0.5	0.23	0.85	0.02
	TF	0.65	0.15	0.36	0.15	0.65	0.22
Sugarbeet	PCF	0.36	0.09	0.32	0.27	0.38	0.016

CONCLUSION

In general, based on the analysis of cadmium content in three widely consumed fertilizers in the region, the highest level of cadmium was found in triple superphosphate fertilizer, having negligible amounts in the other two (namely, urea and potassium sulfate). Given the cadmium content and application rates, it seems that phosphorous fertilizers enjoy the lion's share in terms of the cadmium amount, added to agricultural soils. Since there is no widespread use of animal manure as well as a shortage of any other source of cadmium contamination, such as industrial and mining activities, in the region, one may suspect that the extensive use of chemical fertilizers has been a major contributor to an increased level of cadmium in the agricultural soils.

Finally, according to the results from this study, it can be concluded that the concentration of the studied metals in edible parts of studied crops were lower than the limit, capable of causing health risks from crop consumption. Although the concentration of heavy metals in water and soil before planting crops (and applying fertilizers) in this region was less or at the permissible limits, the frequent and heavy use of fertilizers in conventional agricultural systems may lead to a substantial buildup of heavy metals as well as contamination of agricultural soils and food crops in the long run. Thus, among all good management practices to mitigate the negative effects of fertilizers on the environment, issues such as mandatory testing of agricultural soils to determine their actual need for fertilizers, continuous monitoring to ensure distribution of authorized and standard chemical fertilizers, setting national and provincial standards, encouraging farmers to use environmental-friendly fertilizers along with relevant supporting programs, and monitoring operation of land owners and training of farmers are recommended.

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