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Quantitative measurement of toxic metals and assessment of health risk in agricultural products food from Markazi Province of Iran

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Abstract

The current study aims to investigate the levels of the toxic metal in agricultural products (legumes, wheat, and potato) collected in Markazi province, Iran, and human health risk by using inductively coupled plasma - optical emission spectrometry (ICP-OES). The levels of arsenic (As) and cadmium (Cd) in all samples were lower than the limit of detection (LOD), while the level of Cd in potato samples was lower than the maximum permissible level (MPL) of the European commission (EC). The non-carcinogenic and carcinogenic risk assessment by direct ingestion of agricultural products was calculated using the United States Environmental Protection Agency (USEPA) method. The highest mean of toxic metals was observed for lead (Pb) in legume samples (562.17 μg kg^{-1}). Mercury (Hg) and Pb levels in all samples were higher than LOD, while Pb level in wheat samples were lower than of EC. The rank order of Hg and Pb levels in all samples based on target hazard quotient (THQ) value was wheat> potato>legume. The THQ index of Hg and Pb by the deterministic method in wheat was 1.37 and 0.454; in potato 0.139 and 0.104; in legume 0.092 and 0.41, respectively. The carcinogenic risk index was at an acceptable range. The high hazard index values were estimated and the THQ index for Hg in wheat suggests a non-negligible health risk.

Keywords: Agricultural products, Health risk assessment, Toxic metals, ICP-OES

Introduction

Agricultural products are an important part of the human daily diet that can be contaminated with toxic metals such as Hg, Cd, Pb, and As, therefore agricultural products are considered a serious concern for human health (Stefanović et al. 2008; Dodangeh et al. 2018). The contamination of agricultural products with toxic metals can be related to either environmental resources or human activities such as industrial processing, mining, waste disposal, application of wastewater, sewage sludge to irrigation of cultivated agricultural products,

and even application of fertilizers (especially phosphate type) and pesticides (Tadesse et al. 2015; Marrugo-Negrete et al. 2015; Shariatifar et al. 2020; Shariatifar et al. 2017). It is worthy to note that the atmospheric deposition could be accounted as the primary sources of soil, and water contaminations and consequently further contaminations in the food chain (Fakhri et al. 2017). In this regard, agricultural products can absorb toxic metals from the soil, water, and air, furthermore concentrate them in different parts such as roots, leaves, and grains (Qu et al. 2012).

Toxic metals as the non-essential compounds for the human body could cause several adverse health effects, even in trace amounts (Yılmaz et al. 2010). Due to their non-degradable nature, they could biologically

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accumulate in different tissues (kidney, liver, bone, and brain) after their intake through diet as well as inhalation pathways (Bjørklund et al. 2017; Raj and Maiti 2019). It has been noted that developmental disorders in infants and children such as lowering intelligence quotient (IQ) can be correlated with Hg and Pb (Marrugo-Negrete et al. 2015; Raj and Maiti 2019). In this context, the International Agency for Research on Cancer (IARC) categorized Pb as (group 2B) and Cd as (group 1) a human carcinogenic compound (Cancer IAfRo 1993). As may cause cancer in humans in various organs including the lung, skin, blood, and skeleton. (Sarkar and Paul 2016). Also, excessive entry of heavy metals and synthetic chemicals into the studied soils may lead to deterioration of the soil biology, thereby altering the physicochemical properties of the soil and causing other environmental problems (Qu et al. 2012; Salama and Radwan 2005; Islam et al. 2020; Proshad et al. 2018; Solgi et al. 2014; Solgi et al. 2012; Ghasemidehkordi et al. 2018a; Ghiyasi et al. 2010a; Hani and Karimineja 2010a). Pruvot et al. (2006) showed that the soil near the smelter is more contaminated than the reference soil. Also, higher levels of toxic metals in industrial and mining areas are due to lack of environmental protection and pollution (Pang et al. 2016; Fang et al. 2014). Agrawal et al. (2003) noted that agricultural lands near industrial centers and highways should also be polluted due to air pollution around them (Agrawal et al. 2003). Therefore, atmospheric sedimentation in agricultural lands located in industrial areas might play a critical role in the uptake of higher levels of toxic metals by plants (Rahman et al. 2013). Moreover, industrialization influences the toxic metals concentration in soil, water, and air leads to an increase in the concentration of these elements in agricultural products. Lei et al. (2015) mentioned that human activities, like mineral exploration, refining, and manipulation, as well as sewage sludge, are the most important sources of toxic metals pollution (Lei et al. 2015).

In Markazi province, the presence of heavy metals in the soil may be from various sources such as industrial activities, lead and zinc mines, and fuel combustion, greenhouse gas emissions, and municipal waste disposal. To the best of our knowledge, few researches have been done to investigate the concentration of toxic metals in agricultural products, obtained from the industrial areas of Markazi province as one of the most industrialized regions of Iran. Also, this is the first attempt to measure and compare toxic metal content in agricultural products in the industrial areas of Markazi province using an ICP-OES. Therefore, the present study is devoted to evaluate the toxic metal concentrations and the potential health effects in selected agricultural products (legumes, potatoes, and wheat) in Markazi province of Iran.

Method and material

Chemical reagents
All chemicals (nitric acid 65%, perchloric acid, sulfuric acid, and hydrogen peroxide 70%) and standard stock solutions of toxic metals with analytical grade (purity > 99%) were purchased from Merck (Darmstadt, Germany). Double-distilled water was used in all dilutions.

Sample collection
The sample size was based on the agricultural fields. In this regard, the samples were collected in January 2019 from agricultural products produced in the Markazi Province. A total of 120 samples were collected (60 samples of legume (chickpea ($n = 20$), lentil (n = 20) and bean (n = 20), 30 samples of potato, and 30 samples of wheat were collected). All samples were labeled after collection and stored in clean polythene bags according to their type and then transfer to the laboratory for analyses.

Preparation and treatment of samples
The collected samples were washed with distilled water to eliminate possible surface contamination. Then the potato samples were cut into small pieces with a knife and then all the samples were dried in an oven at 100 °C. Finally, the samples were grinded into a fine powder using a commercial blender and stored in polyethylene bags, until analysis.

Sample preparation
To minimize effects of organic matrix and prevent the possibility of sample contamination and loss of analyte, in this study acid decomposition of samples was performed in the closed vessel microwave oven system (Milestone Ethos D, maximum power of 1400 W and maximum pressure in Teflon vessels of 100 bar). The agricultural product samples were washed with distilled water and dried at 105 °C for 48 h. The dried samples were ground, then homogenized using an agate pestle, and sorted in glass bottles until analysis. All of the glass containers used were cleaned and soaking, overnight in a 10% nitric acid solution, and then rinsed with deionized water (Türkmen and Dura 2016; Mendil et al. 2004). Triplicate agricultural product samples (0.25 g) were digested with 9 mL of nitric acid (65%) and 1 mL of hydrogen peroxide (70%) in the microwave digestion system for 30 min at a maximum temperature of 300 °C. The residue was then diluted to 10 mL with deionized water in a 10 mL volumetric flask. A blank digest was conducted in the same way (Türkmen and Dura 2016; Torres-Escribano et al. 2010).

ICP-OES measurement

All prepared samples were analyzed in triplicates using an ICP-OES (Spectro Arcos, SPECTRO, Germany) with Torch type of flared end EOP Torch 2.5 mm. The optimum functioning parameters were radio-frequency (RF) generator (1400 W); argon gas grade 6 was used for plasma, nebulizer, and auxiliary gas. The gas flows of plasma, auxiliary, and nebulizer were 14.50, 0.90, and 0.85(L min – 1), respectively. Then, initial stabilization time, rinse time, and sample uptake time were 240 s total and 45 s for preflush. Also, the time between replicate analysis and delay time was zero. The analysis was a three-time replicate, and the frequency (resonance frequency) of the generator of RF was 27.12 MHz. The types of solid state, detector, and spray chamber were cyclonic,, Charge Coupled Device (CCD), and modified Lichte, respectively. The type of pump of sample delivery was four-channel, software-controlled; peristaltic pump enables exact sample flows. The speed of prewash pump was 60 rpm (for15 s), 30 rpm (for 30 s), and the time of prewash was 45 s, and finally, the speed of the pump of sample injection was 30 rpm.

Validation of the analytical method

The validation of the analytical procedure for quantitative analysis of elements in agricultural products and its aqueous extracts was performed by evaluating selectivity, working and linear ranges, LOD, quantification limit (LOQ), re-peatability, and reproducibility (precision). Matrix effects were studied using standard addition method, by adding 200 μL of mixed standard solutions to the original samples (Mix standard certified reference material (CRM): 92091 Supelco LOT BCCB9855, Trace CERT®, 33 elements, 10 mgL^{-1} in nitric acid, Hg standard CRM:28941 Supelco, LOT BCCB8927, 1000 mgL^{-1}, Hg in nitric acid). The recovery values were in the 94–105% range.

Health risk assessment

The Chronic Daily Intake (CDI) of detection of toxic metals due to ingestion of toxic metals via consumption of food products was calculated using the following Equation (Heshmati et al. 2018; Jahanbakhsh et al. 2019):

$$CDI = \frac{C \times IR\,i \times EDi \times EFi}{BW \times AT} \qquad (1)$$

In this study, C is the concentration of the toxic metal (mg.kg^{-1}); IRi ingestion rate was set as (legume 19 g day^{-1}, wheat 320 g day^{-1} and potato 58 g day^{-1}) (Abdollahi et al. 2014; Kalantari et al. 2005); EDi is the exposure duration of toxic metals ingestion (24 years); EFi, is

the exposure frequency (350 days year^{-}) (Yousefi et al. 2018); BWI, bodyweight average (70 Kg) (Yousefi et al. 2018; Nazaroff and Alvarez-Cohen 2001) and AT is the mean exposure years (365 days year^{-1}× number of exposure years) (EPA U 2015). The THQ was used for the non-carcinogenic risk assessment of toxic metals according to Eq. 2(Dadar et al. 2017; Madani-Tonekaboni et al. 2019; Roudbari et al. 2021):

$$THQ = \frac{CDI}{RfD} \qquad (2)$$

Where, CDI is chronic daily intake (mg kg^{-1} per day); RfDo is the oral reference dose that for Hg, Pb and Cd are 0.0001, 0.0035, and 0.0005 mg kg^{-1} per day (EPA U 2015; Dadar et al. 2017; Madani-Tonekaboni et al. 2019; Rezaei et al. 2013).

The carcinogenic risks (CR) for exposure to Pb to the population in were obtained using the Eq. 4 (Yousefi et al. 2018; Fakhri et al. 2018; Fathabad et al. 2018):

$$Cancer\ risk\ (CR) = CDI \times CSF \qquad (4)$$

Where, CSF index is the cancer slope factor of Pb via agricultural products ingestion pathway (0.00085 mg/kg/day), CSF index for Hg via oral ingestion was not provided. In this investigation the calculated CR values, was compared with the upper limit allowed by the USEPA standards which is≤10–6 (USEPA 2016).
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Statistical analysis

The statistical analysis was conducted with SPSS v.24 using analysis of variance (ANOVA) and Chi-square tests. The toxic metal contents represent the mean of three independent experiments (triplicate). Data are presented as mean ± SEM value. The considered significant level was $p < 0.05$.

Results

The graphs of calibration for the toxic metals were prepared from the standard solutions at six points, ranged from 0.50–16 μgL^{-1}. All the toxic metals revealed linear relationships of the instrumental response and the solutions containing the metals with insignificant intercepts and correlation coefficients for all samples ranged from 0.9891 to 0.9922.Perceived LOD and LOQ are ranged 0. 05–2 and 0.16–6.60 μg/kg, respectively. The recoveries gained were found to vary from 94 to 105% that is a suitable range for all toxic metals. The wavelengths applied for the determination of the elements concentration, based on baseline signals and their interferences at selected lines observed experimentally during the measurements (Table 1). Table 2 shows toxic metals content in legume, wheat, and potato samples in Markazi

Table 1 Wavelength and method validation parameters obtained by ICP-OES analysis in agricultural products samples

Element	Wavelength (nm)	R^2 Value	Calibration range	Recovery (%)	LOD (µg/kg)	LOQ (µg/kg)
As	189.042	0.9922	0.3–1200	94	1	3.30
Cd	214.438	0.9891	0.3–1200	98	0.05	0.16
Hg	184.950	0.9899	0.3–1200	101	0.35	1.17
Pb	283.305	0.9918	1.2–1200	105	2	6.60

province as well as permissible levels for toxic metals. Table 3 shows the estimated THQ for toxic metals (Hg, Cd, and Pb) exposure through food products (legume, potato, and wheat) consumption by deterministic method in Markazi province.

Based on deterministic method, the CR index in the legume, wheat, and potatoes due to Pb was 1.24×10^{-7}, 1.39×10^{-6}, and 3.17×10^{-7}, respectively.

Discussion

Toxic metals concentration

The concentration of As and Cd in agricultural products samples (except for potato samples in the case of Cd) was lower than the LOD (1 and 0.05 µg kg^{-1}, respectively). For instance, the mean concentration reported for Cd in wheat and legume samples were lower than LOD, while in potato samples it was 27.46 µg kg^{-1}, which were lower than the MPL proposed by EC (30 µg kg^{-1}). In some of the previous studies also As, Cd concentrations were lower than other elements (Hg and Pb) which probably due to the low absorption of As and Cd through the plant root compared to Hg and Pb (Cheng et al. 2017). In this study, among all food samples, the highest mean concentration of Pb was observed in legume samples (562.17 µg kg^{-1}). which were lower than the MPL proposed by National Standard Organization of Iran (300 µg kg^{-1}) (ISIRI 2000). The highest mean concentration of Hg was observed in wheat samples (39.54 µg kg^{-1}), which were lower than the MPL proposed by Food and Drug Administration (500 µg kg^{-1}) (Food and Drug Administration (FDA) 2018). Similar studies have shown that Pb concentration in vegetables and crops is higher than other toxic metals, due to their high ability to absorb even small amounts of Pb from air, soil, and water pollution (Liu et al. 2013; Sharafi et al. 2019). However, the main source of mercury in agricultural products is through air pollution, while the uptake of mercury through soil is partial (Ghasemidehkordi et al. 2018a).

The rank of Hg concentration of agricultural products can be summarized as wheat˃ legume˃ potato. The rank of Pb concentration of agricultural products was legume˃ potato˃ wheat. Although agricultural products can absorb a variety of toxic metals, there is no uniform relationship between toxic metal concentrations and food type (Fang et al. 2014). Moreover, the concentration of the toxic metal in legume samples was higher than potato samples due to the different susceptibility of vegetables to pollution. Pruvot et al. (2006) reported that the leafy vegetables had higher concentrations of Pb and Cd than potatoes (roots or tuberous vegetables) (Pruvot et al. 2006). Liu et al. (2013), reported the differences in toxic metals concentration between different plants owing to their diverse accumulation capacities and different soil characteristics (Liu et al. 2013). Momen et al. (2006) reported that the concentrations of Pb and Cd were lower than LOQ in all legume samples (except for Cd concentration in the white bean, faba bean, and lentil samples were 900, 800, and 500 µg kg^{-1}, respectively) (Momen et al. 2006).

According to study of Tadesse et al. (2015), Pb concentration in potatoes of Ethiopia was in the range of 2000–17,400 µg kg^{-1} (higher than MPL of EC), while in the present study, the Pb concentration was below the LOD (< 100 µg kg^{-1}). A similar study have been shown that the Cd and Pb concentrations in some of the vegetables were 200 and 3950 µg kg^{-1}, respectively (Bahemuka and Mubofu 1999). Ghasemidehkordi et al.

Table 2 Comparison of the heavy metals content in agricultural products samples with Permissible limit of Iran/EC/FDA (µg/kg)

Type of food	samples	Mean ± SD				Maximum permissible level of EC			Maximum permissible level of Iran			Maximum permissible level of FDA			*p*- value		
		As	Cd	Hg	Pb	Cd	Hg	Pb	Cd	Hg	Pb	Cd	Hg	Pb	Hg	Pb	Cd
Legume	(n = 60)	ND	ND	35.45 ± 6.36	562.17 ± 34.39	–	20	300	–	–	–	–	500	6000	0.040	0.042	–
Wheat	(n = 30)	ND	ND	39.54 ± 7.26	372.69 ± 26.35	–	30	500	–	–	100	–	500	6000	0.025	0.030	–
Potato	(n = 30)	ND	27.46 ± 2.33	17.47 ± 1.67	470.52 ± 12.61	30	5	100	–	–	20	100	500	1000	0.04	0.043	0.043

ND not detect

Table 3 Results of THQ indexes due to consumption of Hg, Cd and Pb content in agricultural products samples by deterministic method

Type of food		deterministic method
Legume	Hg	0.092
	Pb	0.041
Wheat	Hg	1.733
	Pb	0.454
Potato	Hg	0.139
	Pb	0.104
	Cd	0.043

(2018a, b) determined the concentrations of Pb and Hg in 10 kinds of agricultural products from agricultural lands near industrial areas of Iran. The highest amount of Pb and Hg reported 56.147 $\mu g\,kg^{-1}$ and 1733.62 $\mu g\,kg^{-1}$, respectively, which were higher than authenticated regulations of THQ World Health Organization & Food and Agriculture Organization (WHO/FAO) (Ghasemidehkordi et al. 2018a,b).

Wheat considers as an important cereal in the diet and provides a 50–90% protein requirement and total caloric content in many countries especially in Iran (Khaniki et al. 2005). The mean Pb concentration of Pb in different traditional flatbreads in Iran by Jahed Khaniki et al. (2005), was determined between 270 and 520 $\mu g\,kg^{-1}$, while the mean Cd concentration was determined as 120–650 $\mu g\,kg^{-1}$(Khaniki et al. 2005). In general, it has been shown that metal contamination in grain bran is more endosperm. Therefore, the amount of toxic metals in cereal-based products like bread is associated with the extraction rate of flour. Furthermore, the supply of wheat from industrial areas can cause further pollution (Khaniki et al. 2005; Tajdar-oranj et al. 2018).

Sadeghi et al. (2020) determined mean concentrations of As, Cr, Hg, and Ni in wheat were 0.186, 0.9, 0.021, and 05 $mg\,kg^{-1}$, respectively. In this study among the metals, As and Hg were the highest, which may be due to wastewater released from human activity in the area (Sadeghi et al. 2020).

Leblebici et al. (2017) determined lowest and highest levels of Cd, Cr, Ni and Pb in potatoes were 0.01–1.90, 0.08–0.98, 0.02–5.35 and 0.52–13.86 $\mu g g^{-1}$.dw, respectively. These toxic metals can be transferred to the atmosphere, water and soil from the activities of industries, mines, agricultural fertilizers, combustion of fossil fuels and traffic. (Leblebici et al. 2017).

Toxic metal contamination of agricultural products is generally observed in most parts of the world; toxic metals are a serious threat to human health. In general, the main reason for the presence of toxic metals due to the rapid growth of urbanization is the change of agricultural land use to industrial, especially in developing countries (Rai et al. 2019).

Non-carcinogenic risk assessment of toxic metals by intake of agricultural products

The results revealed that the rank order of agricultural products based on THQ was wheat>potato>legume (Table 3). The concentration of As and Cd in agricultural products was lower than the limit of detection (LOD), except Cd in some samples of potato which were lower than the MPLof EC (Commission E. 2006). Therefore, there was no need to estimate their non-carcinogenic risk in the population.

The non-carcinogenic risk of wheat was found higher than potatoes and legumes due to the high consumption rate of wheat (320 g/day). The THQ index of Hg and Pb in wheat was 1.37 and 0.454; in potato 0.139 and 0.104; in legume 0.092 and 0.41, respectively (Table 3). In Markazi province is a considerable non-carcinogenic risk (THQ > 1) due to ingestion of wheat. Because of the high daily consumption of wheat and the persistent nature of toxic metals during food processing and bioaccumulation of them in the human (Stefanović et al. 2008), the associated safety considering the concentration of the toxic metal is highly concern.

Carcinogenic risk assessment of toxic metals by intake of agricultural products

The characteristic of the cancer risk (CR) can be qualitatively described as follows; safe limit CR < 10^{-6}; acceptable limit 10^{-4} > CR > 10^{-6}; threshold risk limit CR > 10^{-4}; considerable risk limit CR > 10^{-3}. The findings carcinogenic risk assessment of Pb in agricultural product samples showed that the CR index was the acceptable limit (> 10–4). In various studies, agricultural soil is generally considered to be one of the main sources and receptors for toxic metals. However, in Markazi Province, the presence of toxic metals in the soil may be from various sources such as industrial activities, Pb and Zn mines, and fuel combustion, greenhouse gas emissions, and municipal waste disposal.

The excessive entry of heavy metals and synthetic chemicals into the studied soils may lead to deterioration of the soil biology, thereby altering the physicochemical properties of the soil and causing other environmental problems. Increasing air, water, and soil pollution caused by traffic jams, mines, various manufacturing industries, and industrial waste is a serious problem that negatively affects public health (Islam et al. 2020; Proshad et al. 2018; Hani and Karmineja 2010b; Ghiyasi et al. 2010b).

Although the urban structure of this province seems to be advanced, despite the advanced urban facilities, the main problems caused by air, water, and soil pollution of the area may cause pollution in agricultural products of the Markazi Province. Agricultural products are the main food used in many countries of the world, and pollution has been reported in most parts of the world due

to soil, groundwater, and air. Concerning the increase, the level of heavy metals in agricultural soils and their uptake in agricultural products may be a serious health problem has arisen in some parts of this province.

Conclusion

The results showed that the concentrations of As and Cd in all samples (legumes, wheat, potatoes) were lower than LOD, and also the concentration of Cd in potato samples was less than the MPL of EC. Pb had the highest mean of toxic metals in all agriculture product samples. The concentrations of Hg and Pb in all samples (legume, wheat, potatoes) were lower than LOD, and also the concentration of Pb in wheat samples was lower than the permitted limit of EC. Wheat was recognized as the most important source of exposure to toxic metals due to its high consumption compared to potatoes and legumes. The results of deterministic method showed exposed population from Markazi Province were at an acceptable cancer risk range, by ingestion of wheat and potato samples. Further research to identify the potential sources of contamination with toxic metals and possible control or corrective strategies and continuous monitoring of agricultural products are recommended. Also, farmers, craftsmen, and other people in the community should be aware of the dangers of food exposed to toxic metals in the Markazi province, which can reduce the level of toxic metals in these foods by providing guidelines and action.

Authors' contributions
NabiShariatifar: Conceptualization, Supervision, Design of study Writing-Reviewing and Editing. Fereshteh Karimi.: data analysis, Writing- Original draft preparation. Mohammad Rezaei: data analysis, Investigation. MahsaAlikord: Writing- Reviewing and Editing, Software Majid Arabameri: Software, Methodology, Validation. The author(s) read and approved the final manuscript.

Availability of data and materials
The datasets applied and analyzed during this research are available from the corresponding author on request.

Declarations

Competing interests
The authors have no conflicts of interest to declare in this work.

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