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Assessment of Human Health Risk for Heavy Metals in Imported Rice and its Daily Intake in Iran

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ABSTRACT
This study investigated the amount of Pb, Cd, Cr, Ni, Zn and Cu in samples of imported Indian rice (*Oryza sativa*) available in Iranian markets. Twenty brands of Indian rice which widely-consumed were collected from Iranian local markets. Five samples of each brand were collected and analyzed. After the rice grains were digested through acid digestion method, Pb, Cd, Cr, Ni, Zn, Cu values of each brand were determined by atomic absorption spectrometer. The results showed that cadmium content could not be detected in rice samples (Cd<0.015 mg kg^{-1}). The mean values of lead, chromium, nickel, Zinc and Cu were 0.33±0.25, 0.631±0.43, 0.128±0.047, 3.47±2.12 and 2.08±0.33 mg kg^{-1}, respectively. The results revealed that the mean value of lead in the samples was a little higher than recommended value by FAO/WHO guidelines. Moreover, with determining the degree of Provisional Tolerable Weekly Intake (PTWI) of Pb, Cd, Cr, Ni, Zn and Cu for one person receives less of these metals compared with the standards set by FAO/WHO. Of course, it should be noted that with the increasing rate of food sources contaminated with heavy metals and due to their bioaccumulation in body tissues and the subsequent health risks, it is recommended to use utter products.

Key words: Heavy metals, food hygiene, weekly intake, bioaccumulation, soil pollution

INTRODUCTION
The heavy metals are dispersed to the environment from human and natural sources such as modern development and industrialization and ranks among one of today's top environmental troubles. Heavy metals also have been spread from various sources. They can enter to environment through anthropogenic sources such as using superphosphate fertilizers and fossil fuels, industrial sludge sewage discharging and traffic as well as natural ways (Liu *et al*., 2003). Currently, concern to universal pollution by these metals is related to their bioaccumulation over the period of time in food chain and agricultural products, which is causing serious health risks for humans, animals

and crops (Liu *et al*., 2003; Lie *et al*., 1990; Aghamirlou *et al*., 2015). As a result, the main route of exposure to these elements in human population is soil-crop-food pathway (Fu *et al*., 2008). All heavy metals clearly can create adverse effects in the human body. Small amounts of pollutants such as lead, cadmium, nickel and mercury can be dangerous and toxic for human health and can create adverse health problems such as kidney disorders, pulmonary problems and damage to the bones, cancer and mutation (Mahvi *et al*., 2006; Moghaddam *et al*., 2008; Shekoohiyan *et al*., 2012).

Excessive intake of cadmium causes kidney damage and bone fractures. Long-term exposure to lead may cause memory deterioration, prolonged reaction times and reduced ability to learn. Also, accumulation of Cr, Cu and Zn over time in the bodies can cause serious illness hazardous effects on human health (Cao *et al*., 2010).

Paddy rice is accumulating heavy metals and is determined as a major exposure route for the dietary intake (Gilbert *et al*., 2015). Rice is one of the most important widely-consumed grains in the world and is highly used in daily diets of world population especially in Iran and other countries of Asia. According to statistical data of the Food and Agricultural Organization of the United Nations (FAOSTAT, 2004), rice significantly provided 30% of the dietary energy supply and 20% of the dietary protein intake around the world (FAOSTAT, 2004). In order to enrich their rice crops, farmers apply chemical fertilizers and pesticides to the land, which are persistent to degradation and causes bioaccumulation in rice and other products (Khaniki and Zazooli, 2005). Another reason to rice contamination with heavy metals like lead is the distance of the plant location from the road as well as the traffic of the and area polluted industrial situation that are in vicinity of rivers that discharge their waste into river and other water resources, so that those plants that grow in vicinity of the road and industrial areas have higher concentration of heavy metals (Bosque *et al*., 1990).

The numerous studies have been carried out on heavy metal contents of the rice (Al-Saleh and Shinwari, 2001; Lin *et al*., 2004; Zazooli *et al*., 2010; Shimbo *et al*., 2001) but no study has been done on Indian imported rice in Iran. Therefore, this study due to the high consumption level of Indian rice by Iranian people and its health-related issues was carried out by aim of determining the concentrations of lead Pb, Cd, Cr, Ni, Zn and Cu in Indian rice samples in Iran and weekly intake of aforementioned heavy metals via the consumption of Indian rice.

MATERIALS AND METHODS

This cross-sectional study was performed in 2012. At the first, 20 samples of Indian rice which widely consumed were provided from local markets of Iran. Out of each brand, five samples were randomly selected (n = 100). All glassware containers for the analysis were acid-washed (soaked in 15% HNO_3 solution for at least 48 h), rinsed in ultra-pure water and then dried in an oven in order to eliminate any possible contamination. Firstly, a portion of rice grains from each brand was appropriately cleaned, then 2 g of that were weighed and dried at 105°C for 48 h and then were weighed again to determine water content. The samples were digested by a nitric-perchloric acid (70%) based on ASTM standards. A pre-mixed solution of concentrated nitric and perchloric (30 mL of nitric acid (70%), 10 mL of perchloric acid and 5 mL of sulfuric acid (70%) was added to each sample. Then they were kept at laboratory temperature for 30 min. The beaker was covered with an acid-washed watch glass and was placed on a hot plate. The temperature was gradually increased until the mixture started to boil. The boiling was continued until evaporation occurred and perchloric fumes evolved. The heating was stopped when about 3 mL of a clear liquid remained. Thereupon, dionized water was added to bring the digest to 25 mL (ASTM., 1999).

The digested solution was analyzed for Pb, Cd, Cr, Ni, Zn and Cu contents using flame atomic absorption spectrometer (Shimudze-AA-670). Concentrations of heavy metals were expressed in terms of mg kg^{-1} on a dry weight basis. Dionized water was prepared by (Millipore Direct-Q3) water purification system and the acids used in this study were purchased from German Merck Company.

Provisional Tolerable Weekly Intake (PTWI) is determined for many contaminants to show the amount of contaminants and appropriate safe exposure levels consumed over a lifetime without significant risk. In 1993, PTWI of 7 µg kg^{-1} b.wt., is established by Jecfa which illustrates Tolerable Daily Intake (TDI) of 1 µg kg^{-1} b.wt. (WHO., 1993). In addition to, Daily Intake Rate (DIR) depends on metal contamination as well as daily consumption of foods. The DIR is calculated as follows (Zazooli *et al*., 2008):

$$\text{DIR} = \frac{C_M \times \text{DI}}{\text{Bw}}$$

where, CM is heavy metal concentration in rice (µg g^{-1}), DI is daily intake of rice (kg/person) and Bw is average body weight (In this study, body weight was determined as 65 kg).

Data was analyzed by SPSS software program. The ANOVA test has been used for determination of correlation coefficient between parameters.

RESULTS AND DISCUSSION

Rice is now grown in varying degrees in nearly all provinces of Iran (Fig. 1). In Iran, about 4.8% of all farmland were used for rice. Paddy fields amounted to about 31.2 percent of farmland in Gilan, 37.5% in Mazanderan, 10.1% in Golestan, 8.5% in Khouzestan, 5.5% in Fars and less than 6.2% in the rest of the country.

The results of Pb, Cd, Cr, Ni, Zn and Cu values in 100 samples of imported Indian rice in Iran are shown in Table 1.

Comparison concentrations of heavy metals in sample of rice are shown in Fig. 2. And Average and standard deviation also heavy metals in sample of rice are shown in Fig. 3.

These results indicated that the mean value of Pb concentration in rice 0.33±0.25 mg kg^{-1} on dry wt basis and range is 0.09-0.90 mg kg^{-1} dry wt. The results revealed that less than 35% of rice samples had Pb content below 0.2 mg kg^{-1} and also the amount of Pb content in 65% samples were above 0.2 mg kg^{-1} level. ANOVA analysis showed that there was a significant difference in lead contents in rice ($p<0.003$).

Cadmium content could not be detected in rice samples (Cd<0.015 mg kg^{-1}). The food sanitary standard of Cd in rice on FAO/WHO codex was 0.2 mg kg^{-1}. Therefore, the content of Cd imported Indian rice in Iran is less than the maximum permitted level for rice.

The mean values of chromium, nickel, zinc and copper content were found to be 0.631±0.43, 0.128±0.047, 3.47±2.12 and 2.08±0.33 mg kg^{-1}, respectively. The results indicated that the contents of Pb, Cd, Cr, Ni, Zn and Cu in rice were different for the different market brand.

Table 2 shows the contents of lead Pb, Cd, Cr, Ni, Zn and Cu in rice samples from several countries (Lin *et al*., 2004; Zazooli *et al*., 2010; Fu *et al*., 2008).

International Agency for Research on Cancer (IARC) has introduced cadmium as a carcinogenic agent which is a major cause of kidney dysfunction (WHO., 1989). The content of cadmium in this study could not be detected (less than 0.015 mg kg^{-1}) but in a study on Iranian rice by Zazouli *et al*. (2008) cadmium content was found 0.4 mg kg^{-1}. In another study by Lin *et al*. (2004) cadmium value in Taiwanese markets was detected 0.02 mg kg^{-1}. Cadmium content in rice was determined 0.2 mg kg^{-1} by FAO WHO (2004).

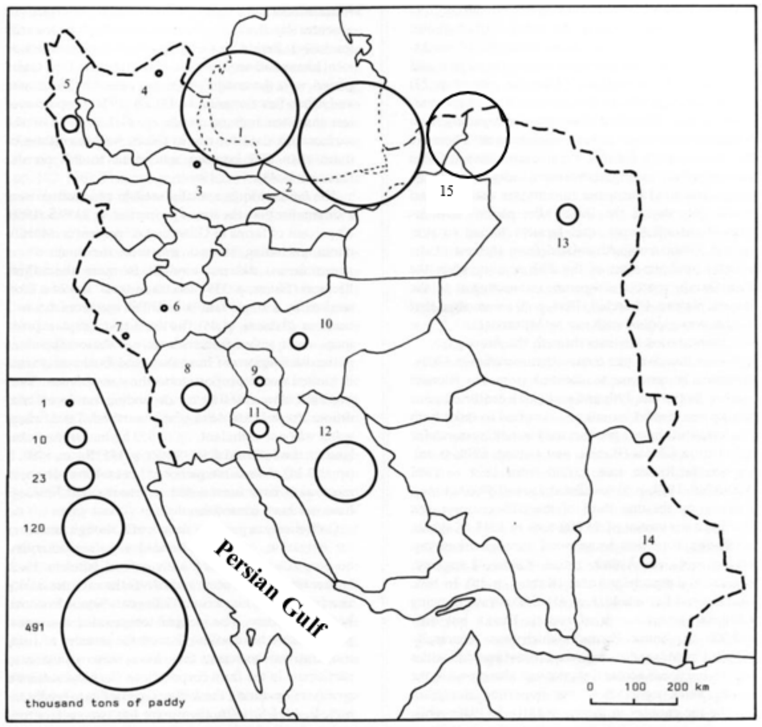

Fig. 1: Paddy production by province; 1: Gilan, 2: Mazanderan, 3: Zanjan, 4: Azarbayejan-e-sharghi, 5: Azarbayejan-e-gharbi, 6: Lorestan, 7: Ilam, 8: Khouzestan, 9: Chahar mahal va bakhtiari, 10: Isfaha, 11: kohgiluye va bouyerahmad, 12: Fars, 13: Khorasan, 14: Sistan va Balouchestan, 15: Golestan

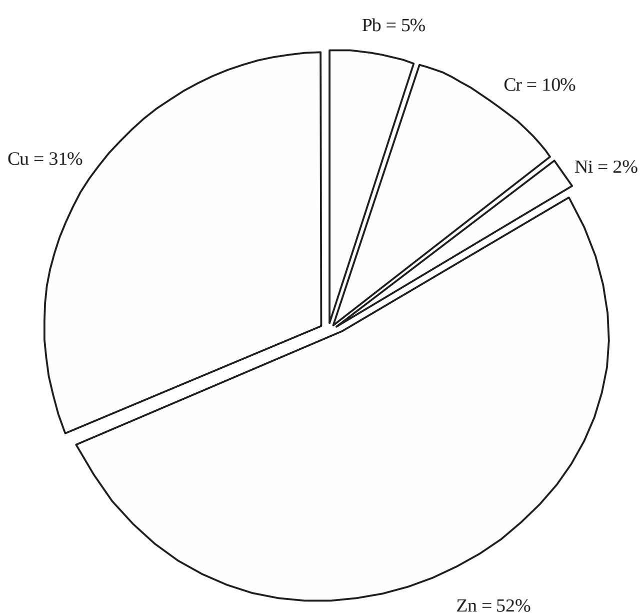

Fig. 2: Comparison concentration of heavy metal in sample of rice

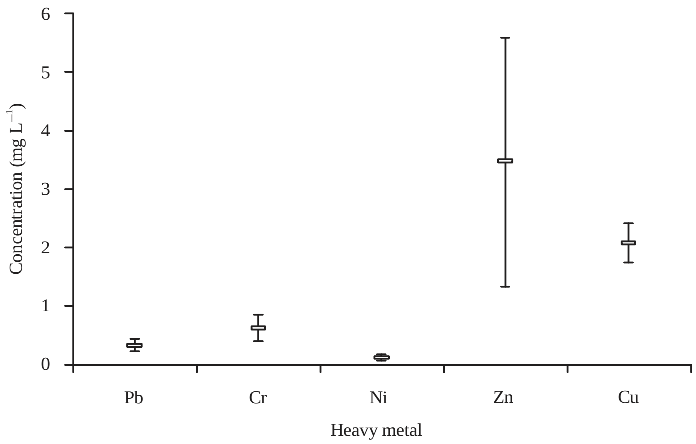

Fig. 3: Average and standard deviation heavy metal in sample of rice

Table 1: Heavy metal content in imported Indian rice in Iran (mg kg^{-1}, b.wt)

Basis of market brand	Pb	Cd	Cr	Ni	Zn	Cu
A[a] (n = 35) [b]	0.137±0.037[c]	BDL	0.790±0.56	0.054±0.01	3.58±2.7	2.04±0.34
B[d] (n = 35)	0.250±0.034	BDL	0.635±0.41	0.168±0.08	2.91±2.5	2.05±0.41
C[e] (n = 10)	0.385±0.035	BDL	0.660±0.25	0.440±0.01	2.85±0.7	2.15±0.07
D[f] (n = 20)	0.785±0.311	BDL	0.282±0.14	0.032±0.01	4.52±2.6	2.17±0.30
Total (n = 100)	0.320±0.230	BDL[g]	0.631±0.43	0.128±0.04	3.47±2.12	2.08±0.33
Standard	0.30	0.2	-	40	-	50
DIR	0.88	-	1.7	0.35	9.54	5.72
PTWI	25.00	7	23.3	35	7000	3500

[a]A: (Lead<0.2 mg kg^{-1}), [b]Number of samples, [c]Data are Means±standard deviation, [d]B: (0.2≤Lead≤0.3 mg kg^{-1}), C[e]: (0.3≤Lead≤0.6 mg kg^{-1}), D[f]: (Lead>0.6 mg kg^{-1}), [g]BDL: Below detectable limit (Cadmium <0.015 mg kg^{-1}), DIR: Daily intake rate, PTWI: Provisional tolerable weekly intake

Table 2: Heavy metal contents in rice from various areas reported in literature mean

Mean area (mg kg^{-1})	Pb	Cd	Cr	Ni	Zn	Cu	Reference
This study	0.320	<0.015	0.630	0.120	3.47	2.08	-
Taiwan	0.020	0.020	0.070	0.260	14.70	2.24	WHO (2004)
China	0.166	0.003	0.062	0.201	-	2.80	Lin *et al.* (2004), Zazooli *et al.* (2010), Fu *et al.* (2008)
Iran	2.230	0.170	-	-	-	-	Gilbert *et al.* (2015)

Lead is an unnecessary metal for human whose varying presence in presents the body's contamination with this metal. It enters to the body dramatically via ingestion and changes according to age. For instance, in adults the average dietary intake of lead is 10% and in children is 40% (Proud *et al*., 1991). According to the results of this study, the mean value of lead concentration (0.32 mg kg^{-1}, p>0.05 and SD = 0.23) was a little more than recommended value by WHO/FAO (0.30 mg kg^{-1}) (WHO., 2004). Therefore, the average content of Pb in Imported Indian Rice to Iran is over the maximum permitted level for rice by WHO/FAO.

In 35% of the samples the lead content was less than recommended value by WHO but in the remaining brands, it was more than the maximum level set by WHO/FAO. Of course, it should be mentioned that, in a study on North Iranian rice found that the lead content was 2.23 mg kg^{-1} which is very higher than WHO/FAO standard. In a similar study on Indian, Egyptian and Thai rice in Arabian market, the mean value of lead was determined 0.052, 0.029 and 0.264 mg kg^{-1}, respectively (Bosque *et al*., 1990) as the lead content in Indian rice was more than Thai and Egyptian rice but it was less than WHO/FAO standard. In another study on rice in Thai market, Lin *et al*. (2004) presented the lead content was 0.01 mg kg^{-1}.

Chromium III is a vital element for the human body and very functional in insulin mechanism, metabolism of fat, carbohydrate and protein but chromium VI is toxic and carcinogenic for human (Rivai *et al*., 1990). In the present study, the mean value of chromium concentration was 0.631 mg kg^{-1}. In a study in Taiwan the chromium value was found 0.10 mg kg^{-1} (Khaniki and Zazooli, 2005).

Provisional Tolerable Weekly Intake (PTWI): The provisional tolerable weekly intake of lead is 25 mg kg^{-1} b.wt. (WHO., 2004). With Assumption 60 kg adult and consumption of 165 g rice per day, the weekly lead intake for one person through rice is 7.001 mg kg^{-1} b.wt., which is less than PTWI set by WHO/FAO (Gulson *et al*., 1995). PTWI for nickel is 35 mg kg b.wt. (Gulson *et al*., 1995). With previous Assumptionss, nickel PTWI for one person is 0.365 mg kg^{-1} b.wt. The PTWI for chromium set by WHO/FAO is 23.3 mg kg^{-1} b.wt. (WHO., 2004). So, any person receives 12.55 mg kg^{-1} b.wt., of nickel weekly, which is less than PTWI set by WHO/FAO.

This study tried to determine lead, nickel, chromium, cadmium, copper and zinc values in Indian rice imported to Iranian markets and found that except 65% of the samples which had lead content more than WHO standard, the remaining values were in the safe range regarding chromium, nickel and cadmium. However, due to bio-accumulation of heavy metals in body tissues, its adverse health effects and the increase of environmental pollutants and food sources, it seems indispensable to need more careful in consuming the products containing these metals and use utter and high-quality products.

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