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# Heavy metal distribution frequency in Iranian and imported rice varieties marketed in central Iran, Yazd, 2012

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## ABSTRACT

**Aims:** This study aimed to determine the concentration of heavy metals including lead (Pb), cadmium (Cd), and arsenic (As) in rice samples in Yazd markets.

**Materials and Methods:** In this study, 108 rice samples were collected from 36 different brands including 26 imported and 10 Iranian rice varieties from July-December 2011. Determination of heavy metals was carried out by wet ashing and digestion methods following graphite furnace atomic absorption spectrometry and hydride generation atomic absorption spectrometry.

**Results:** The average concentrations of lead in both Iranian and imported samples were 328.3 ± 81.44 and 254.55 ± 77.2 μg/kg, respectively, well above the safe limit set by Iranian standard, whereas mean concentrations of cadmium and arsenic were both below their permissible limit. In homemade rice samples, their concentration levels were shown to be 37.25 ± 23.39 and 33.5 ± 18.35 μg/kg, respectively. However, for imported products, their levels were 43.71 ± 13.74 and 87.55 ± 72.99 μg/kg, respectively.

**Conclusion:** Deterministic estimation of exposure to heavy metals from rice showed that there was no health issue concerning exposure to toxic metals through rice intake in Iran when potential risk of each heavy metal is considered individually, cumulative risk assessment must be applied in future studies.

**Key words:** Arsenic, cadmium, daily intake, lead, rice

## INTRODUCTION

Rice is one of the most consumed cereal grain throughout the world. Iran's agricultural ministry issued an official announcement that annual rice consumption is 45 kg per person, rated as the second most consumed grain in Iran.[1-3] In recent years, several reports have been released on the toxic heavy metal contamination of rice in the literature; most of them have been dedicated to the rice varieties from Asia/South Asia, the main producers as well as the main consumers of rice in the world.[4-10]

The hazards relevant to the acute and chronic effects of heavy metals, as well as their impact on human health and environment are well documented.[11-14] Long-term exposure is partly due to food intake, particularly when that type of staple food, and is continuously being affected by environmental contamination of heavy metals.[12,15,16]

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Since heavy metals are long-lasting and accumulating environmental contaminants, the existence of their trace amount in the commercially cultivated products is largely inevitable. However, industrial development and lack of industrial waste management lead to the concentrations exceeding from that of established limits by national and international safety agencies and is considered as a violation.[16] For centuries, in Iran, certain northern, southern, and western areas have been dedicated to the cultivation of rice and upto recent years, most of the public rice consumption was supplied by domestic products inside the country. Lately, to meet the higher demands for rice, farmers' economical problems and failure to compete with the lower price imported ones, rice cultivars from Asian/south Asian countries have being supplied in the market in various brands. A few studies are available regarding the heavy metal concentration in rice in Iran. They mostly considered native cultivars, produced in the particular geographical areas. A report on the heavy metal concentrations in the rice samples from west of Iran, indicated that the average concentration of heavy metals in the studied samples was above the national standard.[17]

Another study performed on the rice planted in Ghaemshahr, in north of Iran, showed that 88% of the samples cultivated in these areas was cadmium contaminated with a higher concentrations than that of the regulatory limit.[18] Lead concentration was also reported to exceed from the standard level in the rice samples produced in the north of Iran.[19] Another study pointed out to an average concentration of cadmium and lead in three types of rice varieties which were cultivated and commonly used in Ahwaz.[20] Moreover, rice planted in Lorestan has been examined for heavy metals in another study, both the latter reports confirmed the safe level of the tested heavy metals in the examined samples.[21]

On the other hand, some focused on the imported rice samples, one of them stated that the average concentrations of two poisonous metals, lead and cadmium were less than the regulatory limits for 20 Indian brands.[22] This was confirmed by another study in which different varieties of rice supplied in Khorasan market were shown to have the average concentration of heavy metals below the national and international standards.[3]

Since the existing rice in the public food basket in an urban community directly depends on various kinds of rice distributed in the market, estimation of heavy metals intake from rice, market studies resulted in a clear view of exposure, particularly, in central Iran where there is no rice field. Furthermore, due to instability of rice outlet, its quality control must be performed on a regular basis. Previous studies in Iran mainly focused on particular rice production areas. Moreover, comparison between domestic and foreign rice verities is rarely disputed in these studies and no similar studies were done in Yazd before.

This study was designed to speculate the frequency distribution of the most toxic heavy metals in different kinds of Iranian and imported rice samples in an urban society of Yazd province of Iran.

## MATERIALS AND METHODS

Totally, 108 rice samples including 30 Iranian and 78 imported rice samples were randomly collected from 33 rice distributors in Yazd city, center of Iran, from July-December 2011. Of the total examined rice types, 10 belonged to domestic products (eight cultivars from north and two from west of Iran). Twenty-six belonged to imported cultivars (21 Indian, four cultivars from Pakistan, and one brand from Uruguay). The rice samples were milled to fine powder using home-used grinder (Moulinex-A2424B-France). All the laboratory glassware and equipment were acid washed with nitric acid solution and all chemical reagents were of analytical grade. Milled samples were oven-dried in two steps. In the first step, the samples were come to a stable weight using 80°C oven (Memmert-C409,0936- Germany) for 2 h, then drying was accomplished using 5 g of each sample at 105°C oven for 6 h.

For lead and cadmium measurement, Association of Analytical Communities (AOAC) 999/11 guideline[23] was followed. Preliminary burning was done on a hotplate (Gerhardt- EV14-1719080019, Germany) and ashing was performed for 10 h in 550°C oven (Barnstead, Barnstead Thermolyne 30400 Muffle Furnace- 1249060781996 -USA) with the initial temperature ramp set at 200°C and 50°C/h until the temperature reached to 550°C. Ash samples were moisturized with distilled water (EC = 0.25 μS/cm) and got dried on water bath (Memmert-L409,0921-Germany). Ten milliliters of 70% nitric acid (Merck-Darmstadt- Germany) were added to each ash sample and the solution was filtered using Whatman filter paper 42 and brought to the volume of 100 ml with double distilled deionized water.

AOAC985/16 guideline[24] based on acid digestion method was followed for arsenic measurement; 30 ml of concentrated nitric acid (Merck- Darmstadt- Germany) was added to each sample, it was stirred and kept at ambient temperature overnight. Then after, the digestion was heated using the hotplate mentioned above. Temperature was gently increased to boil and kept simmered until the solution turned transparent. It was brought to the volume of 3 ml and was filtered using Whatman filter paper 42 and brought to the volume of 100 ml with deionized distilled water. Atomic absorptions spectrophotometer (graphite furnace atomic absorption spectrometry; VARIAN Model AA240 FS) equipped with a graphite furnace (Graphite Tibe Atomizer GTA120) and auto-sampler (Australia) was used for lead and cadmium measurements. In order to measure arsenic in the wet ash samples, the given instrument was equipped with a hydride generation system (hydride generation atomic absorption spectrometry). Furan temperature was set at 1800°C and 2100°C for Pb and Cd, respectively. The main operation parameters including wavelengths of absorption spectra (nm), precision (as relative standard deviation, %), and detection limit (μg/kg) of the three tested elements were: 228.8-2.11 and 0.009 for Cd; 283.3-2.57 and 0.11 for Pb; and 193.7-2.92 and 0.03 for As, respectively.

Calibration standard curves were prepared using standard solutions of 1, 3, 5, and 10 μg/kg from stock standard solution of cadmium ($r^2 = 0.97$) and Arsenic ($r^2 = 0.93$). Lead standard solutions of 15, 30, and 50 μg/kg were used to prepare the calibration curve ($r^2 = 0.99$) for lead measurement.

The experiment quality parameters were assured by the use of triplicate of sample analysis and recovery tests which was shown to be 89-101% for three tested elements.

Descriptive statistics was applied using Statistical Package for the Social Sciences software (SPSS Ins, version 18, IBM colt, USA) for explanation of the results. The data were presented for the total samples (irrespective of their origins) and also individually for either of Iranian/imported products. Moreover, the average concentration of each element was compared with the corresponding permissible limit defined in the Iranian standard, using independent *T*-test with 95% confidence interval. Deterministic exposure assessment was made upon 123 g daily rice consumption and the exposure results were expressed based on the weekly intake of each tested elements (μg) per kg of body weight. Results were compared to provisional weekly intake suggested by International safety authorities.[25]

## RESULTS

Central and spread indicators including mean, quartiles, and standard deviation of lead, cadmium and arsenic are shown in Table 1. The average concentration of lead, cadmium, and arsenic among the total samples, irrespective of their origin, were determined as 275.05, 41.92, and 72.58 μg/kg, respectively.

With respect to lead contamination, most of the samples were placed in the second quartile of lead concentration that ranged from 132.18 to 462.92 μg/kg lead in content, showing a typical normal distribution for lead among the samples. The average concentration of lead in the total samples was significantly above the normal limit ($P < 0.05$) and the large numbers of the tested samples (91%) were detected to be out of the Iranian standard.

Analyzing the results by frequency distribution of lead in two individual groups of Iranian and imported rice samples showed the values of 328.3 and 254.55 μg/kg as the mean lead concentrations in these samples, respectively. Such a concentrations were significantly ($P < 0.05$) higher than permissible safe limit, stated set by the national standard [Table 1]. Nevertheless, lead distribution frequency still followed a normal distribution in the both groups of rice.

The results of cadmium and arsenic measurement in the total samples of rice are presented in Table 1. Concentration of these two elements was mainly ranged in the first quartile of concentration. The average concentrations of both elements were significantly less than the determined permissible limit ($P < 0.05$). However, 13.9% and 8.3%, respectively, of the samples were shown to be unacceptable regarding cadmium and arsenic concentrations.

Evaluating the samples in two individual groups of Iranian and imported products, no change was observed in the results. The concentrations of the latter two elements in both kinds of Iranian and imported rice are shown in Table 1. The average concentration of cadmium and arsenic in the both groups of samples were well below the standard level. Regarding cadmium contamination, most of the Iranian samples were placed in the first quartile of cadmium concentration. However, few samples in the fourth quartile were shown to have considerable amount of cadmium. With respect to the frequency distribution of arsenic, likewise, similar results were observed in different kinds of imported rice verities. Despite the low levels of arsenic in the majority of imported samples, its concentration was found to be quiet worrisome in a few samples. The results of all other reports on the levels of heavy metals in rice, investigated in Iran, were summarized in Table 2.

## DISCUSSION

Heavy metal contamination of rice has become controversial in Iran in recent years. This study, for the first time, considered such a contamination distribution frequency among the rice varieties in Yazd retail market with a comparative perspective to the Iranian and imported rice varieties. According to the obtained results, lead was the most frequent kind of toxic heavy metal to which different kinds of rice available in the market are contaminated. In a way that significant amount of rice supplied in domestic market did not meet the standard in terms of lead. High concentration of lead in different kinds of rice consumed in Iran was reported previously. In 2001, Bakhtiarian *et al*.[17] reported that the concentration of lead in 57 samples of the rice, cultivated around the Korriver in Koorbal region in Fars province, was 2.4 times more than that found in this study in terms of Iranian rice. Malakootian *et al*.[22] studied 20 types of Indian rice, distributed in Iran and found that the mean lead concentration was 364 μg/kg which was almost 1.4 times more than the average concentration of lead in different kinds of imported rice in this study. Besides, lead average concentration of domestic rice varieties cultivated in the north of Iran was reported to be 2230 μg/kg, more than six times higher than what was found in this study. This result was in contradiction with results of three other studies which claimed on the much lower content of lead in Iran rice varieties.[20,26,27]

Among the studies performed in other countries on the Indian rice, the study by Saleh *et al.*[10] in Saudi Arabia showed

**Table 1: Distribution frequency indexes of lead, cadmium and arsenic concentrations in the rice samples collected from Yazd market, corresponded safe limits and the violation percentages**

| Elements | Mean[a,b] (μg/kg) | Safe level[c] (μg/kg) | Violation (%) | Quartiles of heavy metals concentrations | | | |
|---|---|---|---|---|---|---|---|
| | | | | 1 | 2 | 3 | 4 |
| Pb | | 150 | | | | | |
| Total samples | 275.05±77.04 | | 91.7 | 132.18–223.21 | 223.21–268.99 | 268.99–321.5 | 321.5–462.9 |
| Iranian cultivars | 328.77±44.81 | | 100 | 257.03–297.55 | 297.55–323.31 | 323.31–350.0 | 350.04–11.37 |
| Imported rice samples | 254.55±77.2 | | 92.3 | 132.18–210.62 | 210.62–247.35 | 247.35–281.72 | 281.72–62.92 |
| Cd | | 60 | | | | | |
| Total samples | 41.92±17.10 | | 13.9 | 21.87–32.31 | 32.31–37.01 | 37.01–45.44 | 45.44–109 |
| Iranian cultivars | 37.25±23.39 | | 10 | 21.87–27.84 | 27.84–30.21 | 30.21–32.86 | 32.86–109 |
| Imported rice samples | 43.71±13.74 | | 13.8 | 27.71–35.71 | 35.71–37.97 | 37.97–47.63 | 47.63–83.1 |
| As | | 150 | | | | | |
| Total samples | 72.54±67.2 | | 8.3 | 7–34.25 | 34.25–59.5 | 59.5–90.25 | 90.25–421 |
| Iranian cultivars | 33.5±18.35 | | 0 | 7–21.5 | 21.5–27.5 | 27.5–52 | 52–65 |
| Imported rice samples | 87.55±72.99 | | 11.5 | 9–56 | 56–67 | 67–99 | 99–421 |

[a]Data is presented based on dry weigh, [b]Mean concentrations of heavy metals ± Standard deviations, [c]Safe limits of the heavy metal concentrations in rice established in iran's standard, Pb : Lead, Cd : Cadmium, As : Arsenic

**Table 2: Mean concentration of heavy metals under instigation in this study (μg/kg) in domestic and imported rice samples reported in related studies in iran**

| Rice type element | Iranian rice marketed in Yazd | Imported varieties marketed in Yazd | Iranian rice from north of Iran | Rice marketed In Khorasan | Imported varieties marketed in northeast and central Iran | Iranian rice from northeast and central Iran | Iranian rice marketed in Shahrekord | Iranian rice from southwest of Iran | Iranian rice from west of Iran | Iranian rice from north of Iran |
|---|---|---|---|---|---|---|---|---|---|---|
| Pb | 328.3 | 254.55 | - | 26.02 | 369 | 73 | 68 | 87 | 789 | 2230 |
| Cd | 37.25 | 43.71 | 410 | 10.02 | 108 | 45 | 62 | 56 | 65 | 410 |
| As | 33.5 | 87.55 | - | 51.58 | 245 | 70 | - | - | - | - |
| Year of study | 2011 | 2011 | 2007 | 2011 | 2012 | 2012 | | 2010 | 2001 | 2005 |
| Reference | This study | This study | 18 | 3 | 27 | 27 | 26 | 20 | 17 | 19 |

Pb: Lead, Cd: Cadmium, As: Arsenic

that the mean concentration of lead among the Indian rice was 264 μg/kg which is close to the results of this study for imported rice. In Iran, Indian rice varieties examined for heavy metal contamination in two other individual studies and the average lead concentrations were shown to be 364 and 369 μg/kg both far more than the data obtained in this study for imported rice.[22,27]

In opposed to the present result, Janati *et al.* studied almost the same number of rice samples in Khorasan province of Iran. Regardless of samples' origin, they stated that lead concentration in different kinds of rice in the market of that province was less than the standard level.[3] Since their results are similar to the results of this study in terms of the two other elements tested, such difference is unexplainable for us. The only explanation would be a probable difference in the source of rice supplied in center and north-eastern of Iran. The results of cadmium measurement were consistent with the reported results elsewhere in that cadmium was generally less than the permissible local set level.[3,10,22] In Iran, Malakootian *et al.*[22] found similar results by studying Indian rice. However, in contradiction to our results. Studying 60 samples of rice in Ghaemshahr, north of Iran, Maleki *et al.*[18] reported that the cadmium concentration was several times more than the guide value defined in Iranian standard.

Regarding arsenic contamination, there has always been special concern over Asian rice especially those from South East Asia. Various studies indicated that the rice of East Asia, mainly India is polluted with arsenic.[6,7] But, in present research, the average concentration of arsenic detected in the imported samples was less than the safe limit. Note that, in few numbers of samples, arsenic concentration was vigorously high.

Based on the results of this study in the concentrations of three examined elements and considering 165 g daily rice consumption by 60 kg person, it is observed that weekly intake of each of these three elements is quiet lower than respective provisional tolerable weekly intake determined by WHO/FAO [Table 1].

## CONCLUSIONS

The majority of distributed rice samples in the Yazd rice outlet carried unacceptable level of lead, but not cadmium and arsenic. However, it constitutes one-fifth of threshold daily intake of lead, which means that for the time being, such contamination does not pose serious health hazards. Cumulative exposure assessment applying high consumer's data in different age groups of the community should be noted in order to perform more meticulous, realistic studies about potential hazards of heavy metals through rice intake.

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