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Concentration of heavy metals in Iranian market rice and associated population health risk

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RESEARCH ARTICLE

Abstract

Heavy metals contamination, constituting an important public health threat, is considered to be a major problem of environmental systems. In this study, the heavy metal contamination of several famous rice types; both domestic and imported, was investigated. In addition, the risk assessment of heavy metal-contaminated rice was characterised by average estimated daily intakes (EDIs), target hazard quotient (THQs), and hazard index (HI) of the Iranian population. The EDIs for Pb, As and Cd from rice was 0.217, 0.183 and 0.090 μg/kg bw/day, respectively. THQs of individual element for all metals was less than one, indicating that health risk associated with the intake of a single element through consumption of rice was negligible. The HI of heavy metals was 0.331. It shows that for the general people there is no health risk to Pb, As and Cd by the rice intake.

Keywords: food safety, lead, arsenic, cadmium, risk assessment

1. Introduction

Human beings are exposed to heavy metals from many different sources including air, water, soil, food and consumer products. Dietary intake is the main route of exposure for most people (Järup, 2003; Zhuang *et al.*, 2009). Cereals play an important role in the diet and are the main source of many nutrients for both children and adults since they contain protein, vitamins, carbohydrates, fibre, and minerals (McKevith, 2004). Rice is one the most important grain produced mainly for use as human food and provide high percentage of total calories consumed in the world (Juliano, 1993).

Recently, concern has been raised about possible heavy metals contamination in rice. It seems that rice can become contaminated with heavy metals by many ways, such as irrigation, growth in contaminated soil, industrial emissions, the harvesting process, storage and/or at the point of sale (Bennett *et al.*, 2000; Zhuang *et al.*, 2009).

Most of the heavy metals, especially Pb, As and Cd, are toxic because of their non-biodegradable nature, long biological half-lives, solubility in water and their potential to accumulate in different body parts. It is known that serious systemic health problems can develop as a result of exceeding dietary intake of heavy metals such as Cd and Pb. Even low concentrations of heavy metals have harmful effects on human and animals health because their elimination from the body is impaired (Khan *et al.*, 2008; Otitoju *et al.*, 2014). Furthermore, consumption of heavy metal-contaminated food can perilously deplete some essential nutrients in the body causing a decrease in immunological defences, intrauterine growth retardation, impaired psycho-social behaviour, and gastrointestinal cancer (Arora *et al.*, 2008). Therefore, some regulations to restrict the emission of heavy metals and the tolerance limit in food have been set up. It is necessary to decline toxic heavy metal accumulation in cereals used in food production, particularly rice.

Rice is a staple food in Iran, with the quality of cooked rice outweighing all other considerations for Iranian consumers. It is grown in fifteen provinces. However, more than 80% of rice area is distributed in the two northern provinces of Iran (Mazandaran and Gilan). As the demand and supply of rice in Iran is still not evenly balanced, thousands of tons of rice for domestic consumption are imported to country mostly from India and Pakistan (Malakootian *et al.*, 2011; Shobha Rani, 1998). Thought, there are reports regarding heavy metal contents and risk assessment of domestic and imported rice, none of them were comprehensive enough to compare contaminants among a wide range of domestic and imported rice (Morekian *et al.*, 2013; Naseri *et al.,* 2015).

Mashhad is the second most populous city of Iran and every year several million people visit it because of pilgrimage and tourism attractions. The present study was conducted with aim to determine the heavy metals (Pb, Cd and As) concentrations in some of the frequently consumed Iranian (Gilan and Mazandaran) and imported rice (Pakistanis and Indian); to estimate the health risks of Cd, Pb and As via consumption of rice for the subjects based on a probabilistic method. The THQs and HI were calculated to evaluate the non-carcinogenic health effects of single and combined heavy metals due to daily consumption.

2. Materials and methods

Sample collection

One hundred and twenty samples of frequently consumed rice were collected from local agricultural agencies and retail markets from Mashhad, Iran. The collected samples were grouped according to the producing country, Iranian rice (Gilan and Mazandaran) (both n=30) and imported rice (Pakistanis and Indian) (both n=30). Regarding the ethical aspects of research, manufacturers were defined with code and the research was done by an examiner blind to the producers.

Digestion procedures

Dry ashing

After homogenisation of the samples, they were come to a stable weight in oven at 80 °C for 2 h, then drying was accomplished (5 g of each sample) at 105 °C in oven for 6 h. Preliminary burning was done on a hotplate and ashing was performed for 10 h at 550 °C in oven for 10 h with the initial temperature ramp set at 200 °C and 50 °C/h until the temperature reached 550 °C. Ash samples were moisturised with distilled water and evaporated into dryness over a water bath. Then the residue was dissolved with 10 ml HNO_3 and made up to the final volume (100 ml) with deionised water (Morekian *et al.*, 2013).

Wet ashing

For arsenic measurement, 30 ml of concentrated nitric acid was added to each sample. The acidified sample was kept at room temperature for 24 h. After the digestion sample was heated using the hotplate like dry ashing procedure. The temperature was gently increased to boil the solution and kept simmered until turned transparent. It was brought to the volume of 3 ml and was filtered using Whatman filter paper (Whatman, Maidstone, UK) and brought to the volume of 100 ml with deionised distilled water (Morekian *et al.*, 2013).

Apparatus

Atomic absorption spectrophotometer (Varian model AA-1275; Varian Medical Systems, Inc., Palo Alto, CA, USA) (equipped with a graphite furnace and auto-sampler) was used for Pb and Cd measurements. In order to measure As in the wet ash samples, the given instrument was equipped with a hydride generation system. Furan temperature was set at 1,800 and 2,100 °C for Pb and Cd, respectively.

Preparation of the calibration curve

Calibration standard curves were prepared using standard solutions of 5, 10, 15 and 20 mg/kg from the stock standard solution of Cd (R^2=0.998) and Pb (R^2=0.995). As calibration curve is prepared with arsenic standard solutions of 1, 5, 15 and 20 mg/kg (R^2=0.998). The limit of detection was determined and was found to be as follows: Cd = 1 µg/kg; Pb = 7 µg/kg; As = 10 µg/kg.

Quality assurance

All acids used in this study were ultra-pure grade and other reagents were analytical grade. All glass wares were soaked in nitric acid solution for 24 h and rinsed with deionised water. Three replicates were used for the analysis of rice samples. Reagent blanks and a standard plant sample were employed in the analysis to ensure accuracy and precision.

Human risk assessment of heavy metals through rice consumption

Estimated daily intake

The daily intake of metals depends on both the metal concentration in food and the daily food consumption. In addition, the body weight can influence the tolerance of pollutants. The estimated daily intake (EDI) is a concept introduced to take into account these factors. The EDI was calculated with the following equation:

$$EDI = \frac{EF \times ED \times FIR \times C}{W_{AB} \times TA} \quad (1)$$

Where EF is exposure frequency (365 days/year); ED is the exposure duration (70 years), equivalent to the average lifetime; FIR is the food ingestion rate (100 g/person/day); C is the metal concentration in rice (μg/g); W_{AB} is the average body weight (70 kg) and T_A is the averaging exposure time for non-carcinogens (365 days/year × ED) (Qian *et al.*, 2010).

Target hazard quotient

Non-cancer risk assessments were typically conducted to estimate the potential health risks of pollutants using the target hazard quotient (THQ). The THQ for the residents through consumption of contaminated rice can be assessed by comparing with the provisional tolerable weekly intake (PTWI) for each element. The health risks were separately considered since the contact pathway with each exposure medium changes with age. In this respect, the THQ was determined based on the methods modified from Chien *et al.* (2002) by the following equation (Qian *et al.*, 2010):

$$THQ = \frac{EDI \times 7}{PTWI} \quad (2)$$

The applied PTWI for Cd, Pb and As were recommended by the Joint FAO/WHO Expert Committee on Food Additives: 7 μg Cd/kg bw/week; 25 μg Pb/kg bw/week and 15 μg As/kg bw/week (Bilandžić *et al.*, 2011).

Hazard index

To assess the overall potential risk for non-carcinogenic effects posed by two or more pollutants, the hazard index (HI) approach has been developed. The HI is a measure of the potential risk of adverse health effects from a mixture of chemical constituents. Whether or not a particular chemical mixture posed an additive risk depends on the targets (tissue, organ, or organ system) and the mechanisms of action of the individual chemical. The HI values through daily consumption of rice for human beings were calculated with the following equation (Qian *et al.*, 2010):

$$HI = \sum_{n=0}^{i} THQn \quad (3)$$

Statistical analysis

All data were analysed using SPSS 13.0 (SPSS Inc., Chicago, IL, USA). The characteristics of heavy metals concentrations were described in the mean ± standard deviation and examined using descriptive statistics. Mean values of rice heavy metals concentrations were compared among different provinces using ANOVA and the Tukey post-test. The results of statistical tests were considered statistically significant if $P<0.05$.

3. Results and discussion

Heavy metals concentrations in rice samples

Cadmium

Cd is an extremely toxic metal with a natural occurrence in soil, but is also spread in the environment due to human activities. It can be readily taken up by plants and translocated to aerial organs where it can accumulate to high levels. Consequently, it can easily enter the food chain and become detrimental to human and animal health (Alam *et al.*, 2003). Cd causes toxicity in multiple organs in humans and laboratory animals. Chronic Cd exposure through inhalation or by oral ingestion results in renal dysfunction, anaemia, pulmonary tumours, osteoporosis and bone fractures (Godt *et al.*, 2006; Wu *et al.*, 2012).

The Cd concentration in our study is varied greatly, from 0.001 to 0.156 mg/kg and with means 0.064±0.001 mg/kg. As shown in Figure 1, the maximum and minimum levels of cadmium were related to Pakistanis rice (0.082±0.006 mg/kg) and Gilan rice (0.043±0.006 mg/kg), respectively. There is no significant difference in the levels of cadmium between Mazandaran and Gilan rice and the Pakistanis and Indian rice. However, there are significant differences between Iranian and imported rice varieties.

According to the Iranian National Standards Organization (INSO) and FAO\WHO, a maximum allowable concentration of Cd in rice is 0.04 mg/kg and 0.1 mg/kg, respectively. The survey results showed that the mean concentrations of cadmium in Mazandaran (0.052±0.009 mg/kg), Gilan (0.043±0.006 mg/kg), Pakistanis (0.082±0.006 mg/kg) and Indian (0.081±0.009 mg/kg) rice are higher than the maximum allowed set by INSO but under the FAO/WHO (FAO/WHO, 2004; INSO, 2013).

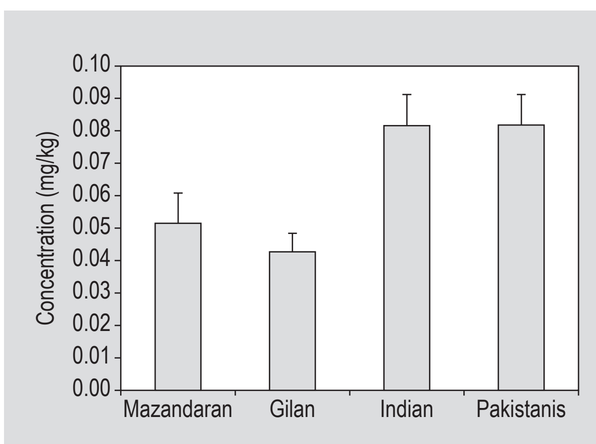

Figure 1. Cadmium concentration in different rice varieties.

More recently, a study by Ghazanfarirad *et al.* (2014) conducted on the amount of Cd in imported rice samples (16 different brands) showed that all samples contained trace amounts of Cd (0.001 to 0.103 mg/kg). Only one type of rice (6.25% of the samples) contained amount of Cd higher than the standard (0.1 mg/kg for Cd) and the rest of the samples contained levels of Cd below the standard threshold (Ghazanfarirad *et al.*, 2014).

Lead

Concern has grown over the adverse effects of exposure to Pb because of its possible detrimental effects on intelligence (Baghurst *et al.*, 1992). The Pb levels ranged from 0.108 to 0.234 mg/kg, with a mean level of 0.153±0.001 mg/kg. Figure 2 shows that the highest Pb levels were found in Indian rice 0.171±0.009 mg/kg and the lowest in Mazandaran rice (0.134±0.008 mg/kg). Statistical analyses showed no significant difference in the levels of Pb between Mazandaran and Gilan and the Pakistanis and Indian rice. However, there are a significant differences between Iranian and imported rice brands. According to the current INSO of 0.12 mg/kg, 96.66% of total samples were not acceptable on Pb contamination level but the average concentration of the Pb in all rice was lower than the maximum allowed set FAO/WHO of 0.2 mg/kg (INSO, 2013).

Morekian *et al.* (2013) investigated the Pb content of 36 different brands including 26 imported ones (anonymous country) and 10 Iranian rice brands by atomic absorption spectrometry in Yazd. They reported that the average concentrations in both Iranian and imported samples (91%) were above the safe limit set by the Iranian standard that are similar to the results of this study. (Morekian *et al.*, 2013). Like the results of this investigation, Naseri *et al.* (2015) had reported the concentrations of Pb in rice samples collected from Shiraz was above the permissible limit of Iran.

Arsenic

Rice is an important route of human exposure to arsenic, especially in populations with rice-based diets. The use of arsenic-containing compounds on soils and arsenic contaminated irrigation water appears to be the driving force for increasing As concentration in rice grains (Williams *et al.*, 2005). Rice contains the highest amount of As in all types of cereals, largely because of the high bioavailability of As under reduced soil conditions (Qian *et al.*, 2010; Zavala *et al.*, 2008).

The As concentrations in all rice samples ranged from 0.039 to 0.378 mg/kg with a mean of 0.129±0.017 mg/kg. As concentrations in different groups (Gilan, Mazandaran, Pakistanis and Indian) are given in Figure 3. The maximum and minimum of As concentration was observed in Indian rice (0.188±0.032 mg/kg and Mazandaran rice 0.093±0.009 mg/kg, respectively. There is no significant difference in the levels of As between Mazandaran and Gilan rice and the Pakistanis and Indian rice. However, there are significant differences between Iranian and imported rice brands. According to INSO and FAO/WHO recommends (0.12 and 0.2 mg/kg) for As, there could be 70 rice samples (58.33%) in this study containing As more than that of reported by Iranian standards and only 18 samples of brands (15%) excess of the above mentioned maximum allowable concentrations set FAO/WHO (INSO, 2013)

Similar results can be found in various studies, which reported that As content of rice samples were upper the food sanitary (Khaniki and Zozali, 2005; Malakootian *et al.*, 2011; Rezaiyan Attar and Javad, 2014).

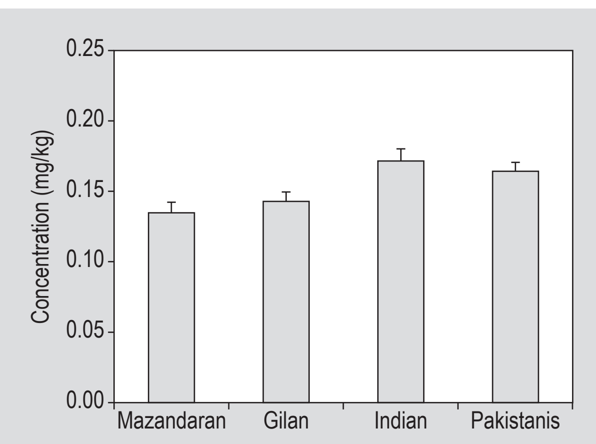

Figure 2. Lead concentration in different rice varieties.

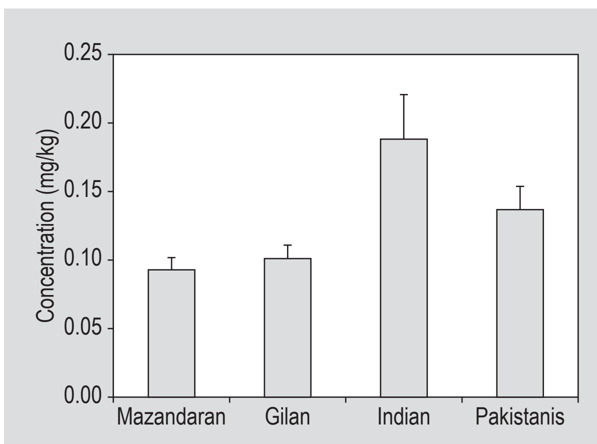

Figure 3. Arsenic concentration in different rice varieties.

Human risk assessment of heavy metals through rice consumption

Estimated daily intake

The EDIs of heavy metals for Iranian people via consumption of different types of rice are presented in Table 1. The trends of EDIs for heavy metals in rice are in the order of Pb > As > Cd.

Target hazard quotient

The THQ is an advantageous parameter for assessment of risks associated with the consumption of food crops contaminated with heavy metals. The THQs of individual heavy metals through rice consumption for Iranian population are given in Table 2. There were no values for an individual element more than one in any types of rice, indicating that Iranian people are not at potential health risk by intake of a single heavy metal of Cd, Pb and As from rice consumption. For all types of rice, the THQs of heavy metals from rice consumption is in decreasing order of Cd > As> Pb. Our results show that Cd intake has the highest potential health risk of adverse effects and Pb ingestion has the minimum risk.

Table 1. Estimated daily intake of heavy metals for Iranian people via consumption of different types of rice.

Type of rice	Pb	Cd	As
Mazandaran	0.191	0.073	0.131
Gilan	0.200	0.060	0.144
Pakistanis	0.242	0.115	0.267
Indian	0.231	0.116	0.194
Mean	0.216	0.091	0.184

Table 2. Target hazard quotient of heavy metals for Iranian via consumption of different types of rice.

Type of rice	Pb	Cd	As
Mazandaran	0.053	0.073	0.060
Gilan	0.056	0.060	0.066
Pakistanis	0.067	0.115	0.122
Indian	0.064	0.116	0.089
Mean	0.060	0.091	0.084

Hazard index

The accepted standard for HI is 1.0 at which there will be no significant health hazard. The probability of experiencing long-term health hazard effects increment with the increasing HI value and according to Lemly (1996); HI=1.1-10 refers to moderate hazard while HI>10 refers to high hazard (Lemly, 1996; Ogunkunle *et al.*, 2013).

HI value through rice consumption for Iranian population is 0.331. This suggests that Iranian population do not experience adverse health effects from the rice intake.

4. Conclusions

Based on the new INSO, most of the imported rice that exists in Iran markets showed Cd, Pb and As contamination. It is noteworthy that the FAO\WHO limits for heavy metals in rice are much higher than that of INSO. However, the residents in Iran tend to eat more rice than those in the other parts of the world. As a result, the potential risk of heavy metals in rice to human beings is not serious. Considerable attention should also be paid to the potential health risks of heavy metals through other exposure pathways.

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