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Arsenic, cadmium, lead and chromium concentrations in irrigated and rain-fed rice and their dietary intake implications

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Abstract

Absorption of heavy metals and contaminants through a rice-based diet may have serious consequences for human health. The present study determined the levels of arsenic (As), cadmium (Cd), lead (Pb) and chromium (Cr) in 71 irrigated and rain-fed rice and assessed dietary (rice) exposure to the heavy metals. The concentration in rice grains were generally higher in the irrigated season which may be related to the use of contaminated irrigation water: As 0.153 ± 0.112 and 0.140 ± 0.080 mg kg^{-1}, Cd 0.073 ± 0.069 and 0.038 ± 0.032 mg kg^{-1}, Pb 0.264 ± 0.125 and 0.147 ± 0.077 mg kg^{-1} and Cr 1.208 ± 0.913 and 0.986 ± 0.796 mg kg^{-1} in irrigated and rain-fed rice, respectively. Elevated concentration of grain As was recorded for rice samples that were collected from the locations with As-contaminated groundwater used for irrigation. For Cd, Pb and Cr, relatively higher concentration was noted for the areas which could be linked to industrial effluent contamination. Daily intake of As and heavy metals from rice is estimated as 18.6-214 µg for As, 2.6-119 µg for Cd, 25.0-241 µg for Pb and 59.0-1846 µg for Cr, based on 400g daily rice consumption for 60 kg Bangladeshi adult people. The rice component of the diet alone may contribute up to 46%, 57%, 50% and 60% of the Maximum Tolerable Daily Intake (MTDI) for As, Cd, Pb and Cr, respectively, making it a more important factor in the dietary intake for these elements than other food stuffs and drinking water. Hence, heavy metals accumulation in rice grains is a big concern in south Asia where people's daily meal largely contains rice or rice based products.

Keywords: Heavy metals, irrigated rice, rain-fed rice, risk exposure.

Introduction

Rice is most important staple food for about 50% of the world's population (Muthayya et al., 2014) and it supplies about 30% of the dietary energy and 20% of the dietary protein in Asia (WHO, 2002). However, rice may contain significant amounts of contaminants such as arsenic (As), cadmium and lead (Meharg et al., 2009, 2013; Watanabe et al., 1996). Intake of As and heavy metals through rice could cause an adverse impact on human health. Serious concerns over heavy metal accumulation in rice grains have been addressed in recent years (Fu et al., 2008; Diyabalanage et al., 2016). Elevated concentration of As (Abedin et al., 2002; Meharg and Rahman, 2003; Das et al., 2004) and Cd in rice grain have been reported (Meharg et al., 2013).

Heavy metal contamination of food is one of the most important assessment parameters of food quality assurance (Wang et al., 2005; Khan et al., 2008). International and national regulations on food quality have lowered the maximum permissible levels of toxic elements in food items due to an increased awareness of the risk that these elements pose to the food chain and tp human health in particular (Radwan and Salama, 2006). Arsenic (As), cadmium (Cd), lead (Pb) and chromium (Cr) are recognized as toxic elements. Absorption of these elements in excess through foods could have serious consequences on human health (Jarup, 2003). Rice is a major food crop in south-east Asia and in Bangladesh it occupies about 75% of the cropped area. Irrigated rice (Boro rice) is grown in the dry season and rain-fed rice (transplant Aman rice) in the wet season. Rice plants are more efficient in assimilating As into their grain than other cereal crops and the bioavailability of As is greatly enhanced in flooded (reducing) soil conditions leading to an excessive As bioaccumulation of wetland rice crops (Islam et al., 2016). Irrigation water often contains elevated concentrations of toxic elements (Ahmad and Goni, 2010; Islam et al., 2015). The As concentration of Boro rice cultivated with As contaminated irrigation water may exceed the acceptable human consumption level of As by as much as 2.65 times (Aziz et al., 2015). Halim et al. (2015) reported higher concentrations of heavy metals in rice grains from contaminated soils. The present study was undertaken to determine the levels of As and heavy metals (Cd, Pb and Cr) in rain-fed and irrigated rice and to assess risk exposure to these elements through typical daily dietary intake levels of Bangladeshi adults.

Results

Variation in arsenic concentration of rice grains

The As concentration of rice greatly varied with growing seasons, varieties and more importantly with locations. Location-wise rain-fed rice As concentration (Table 1) varied more than twofold from Faridpur (mean 0.186 mg kg^{-1}) and Saltha (0.185 mg kg^{-1}) to Gazipur (0.076 mg kg^{-1}). Faridpur

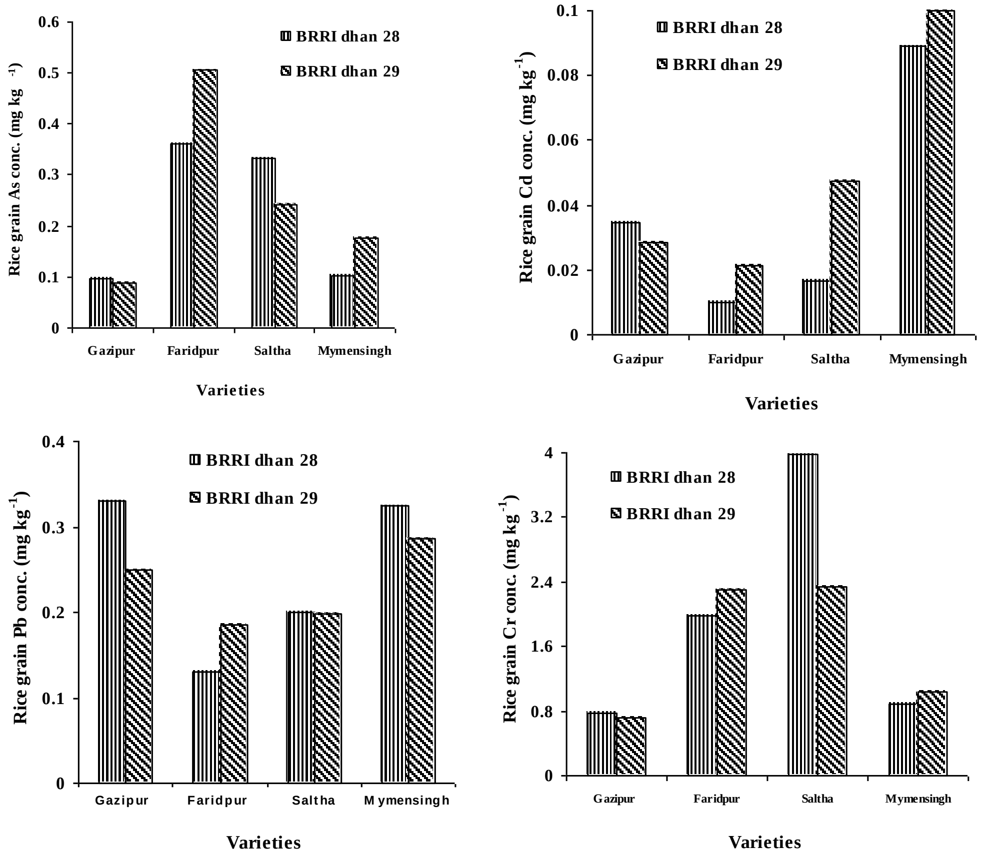

Fig 1. Location variations in grain As, Cd, Pb and Cr concentrations of two rice varieties.

and Saltha are known for elevated concentrations of As in groundwater and consequently in irrigated soils of those areas (Rauf et al., 2011). The As accumulation in rice was considerably higher in irrigated rice than in rain-fed rice. Boro rice is cultivated with irrigation water from shallow tubewells (STW) while the rain-fed rice is cultivated with almost no use of irrigation water.

Looking at the variety comparisons, still the driving force was location and crop season. For this reason, BRRI dhan28 and BRRI dhan29 that grown in dry season demonstrated higher As accumulation in rice, the values being 0.224 and 0.253 mg kg^{-1}, respectively (Fig. 1). On the other hand, BINA dhan7 had an elevated level of rice As (0.371 mg kg^{-1}) since rice samples were collected from As contaminated Faridpur area. Rice varieties of Indian origin (Swarna and Ranjit) had relatively lower content of grain As, showing 0.106 and 0.067 mg kg^{-1}, respectively. Aromatic rice (fine grain rice) varieties viz. Kataribhog, Basmati, Kalajira, Jirashail and Chinisagar that cultivated in wet season (transplant Aman rice) had relatively lower content of grain As, it being on an average 0.127 mg kg^{-1} (data not shown).

Variation in cadmium concentration of rice grains

Irrigated rice samples collected from Mymensingh exhibited the highest Cd concentration (mean 0.080 mg kg^{-1}), which was threefold higher than Saltha (0.024 mg kg^{-1}) and Baghmara (0.024 mg kg^{-1}) (Table 1).

Between the two rice seasons, the difference of Cd concentration was small (Table 1). Unlike rice As, irrigation water may not be a potential source of Cd in rice. The Cd level of rice samples depending on the varieties ranged from 0.007-0.297 mg kg^{-1}, however the majority varieties had Cd concentration below 0.05 mg kg^{-1} (data not shown).

Variation in lead concentration of rice grains

The Pb concentration of rice samples varied significantly with growing seasons, varieties used and locations of cultivation. Comparing location differences, the Pb concentration of rain-fed rice decreased threefold from Bagha (mean 0.232 mg kg^{-1}) and Chapai Nawabganj (0.231 mg kg^{-1}), to Baghmara (0.080 mg kg^{-1}).

Like rice As, the rice Pb level was always higher in irrigated rice than in transplanted rice (rain-fed). The highest difference in rice Pb concentration was observed with Saltha (0.199 mg kg^{-1} for irrigated rice against 0.091 mg kg^{-1} for rainfed rice. Rice from Gazipur (0.306 and 0.201 mg kg^{-1}, respectively) had higher Pb than at Faridpur (0.182 and 0.123 mg kg^{-1} , respectively).

Variation in chromium concentration of rice grains

Variation in rice Cr content between locations and between seasons is displayed in Table 1. The chromium concentration of rain-fed rice collected from 10 sub-districts declined from Gomostapur (mean 1.918 mg kg^{-1}), to Baghmara (0.577 mg kg^{-1}). In season comparisons, Mymensingh and Saltha had higher rice Cr for irrigated rice, but for other locations the reverse was true. The Cr concentration of rice varieties over the locations and seasons was in the range of 0.148- 4.616 mg kg^{-1}. Aromatic rice (fine rice) varieties showed lower rice

Table 1. Concentrations of arsenic, cadmium, lead and chromium in rice grains from 10 sub-districts across Bangladesh.

Location (Sub-district)	Arsenic (mg kg^{-1})		Cadmium (mg kg^{-1})		Lead (mg kg^{-1})		Chromium (mg kg^{-1})	
	Irrigated rice	Rain-fed rice	Irrigated rice	Rain-fed rice	Irrigated rice	Rain-fed rice	Irrigated rice	Rain-fed rice
Faridpur Sadar	0.306 ± 0.159 (n = 4)	0.186 ± 0.165 (n = 7)	0.019 ± 0.006	0.029 ± 0.019	0.182 ± 0.037	0.123 ± 0.046	0.196 ± 0.360	0.767 ± 0.526
Saltha	0.287 ± 0.066 (n = 2)	0.185 ± 0.072 (n = 5)	0.032 ± 0.022	0.024 ± 0.026	0.199 ± 0.001	0.091 ± 0.028	3.17 ± 1.16	0.878 ± 0.509
Gazipur Sadar	0.078 ± 0.021 (n = 6)	0.076 ± 0.021 (n = 2)	0.087 ± 0.072	0.033 ± 0.017	0.306 ± 0.123	0.201 ± 0.106	0.554 ± 0.214	1.49 ± 0.360
Mymensingh Sadar	0.129 ± 0.035 (n = 11)	0.106 ± 0.031 (n = 6)	0.059 ± 0.037 (n = 11)	0.080 ± 0.087 (n = 6)	0.260 ± 1.05 (n = 11)	0.118 ± 0.030 (n = 6)	1.10 ± 0.484 (n = 11)	0.941 ± 0.632 (n = 6)
Bagha		0.123 ± 0.070 (n = 4)		0.053 ± 0.036 (n = 4)		0.232 ± 0.082 (n = 4)		0.629 ± 0.583
C. Nawabganj Sadar		0.153 ± 0.075 (n = 5)		0.062 ± 0.074 (n = 5)		0.231 ± 0.071 (n = 5)		0.773 ± 0.201
Paba		0.109 ± 0.026 (n = 4)		0.048 ± 0.013 (n = 4)		0.124 ± 0.057 (n = 4)		0.762 ± 0.623
Baghmara		0.097 ± 0.036 (n = 3)		0.024 ± 0.011 (n = 3)		0.080 ± 0.008 (n = 3)		0.577 ± 0.327
Gomostapur		0.155 ± 0.046 (n = 7)		0.041 ± 0.024 (n = 7)		0.166 ± 0.097 (n = 7)		1.918 ± 1.464
Charghat		0.114 ± 0.034 (n = 5)		0.032 ± 0.025 (n = 5)		0.115 ± 0.012 (n = 5)		0.629 ± 0.340

T (Transplant) Aman rice is grown in rain-fed condition (wet season) and Boro rice in irrigated condition (dry season)

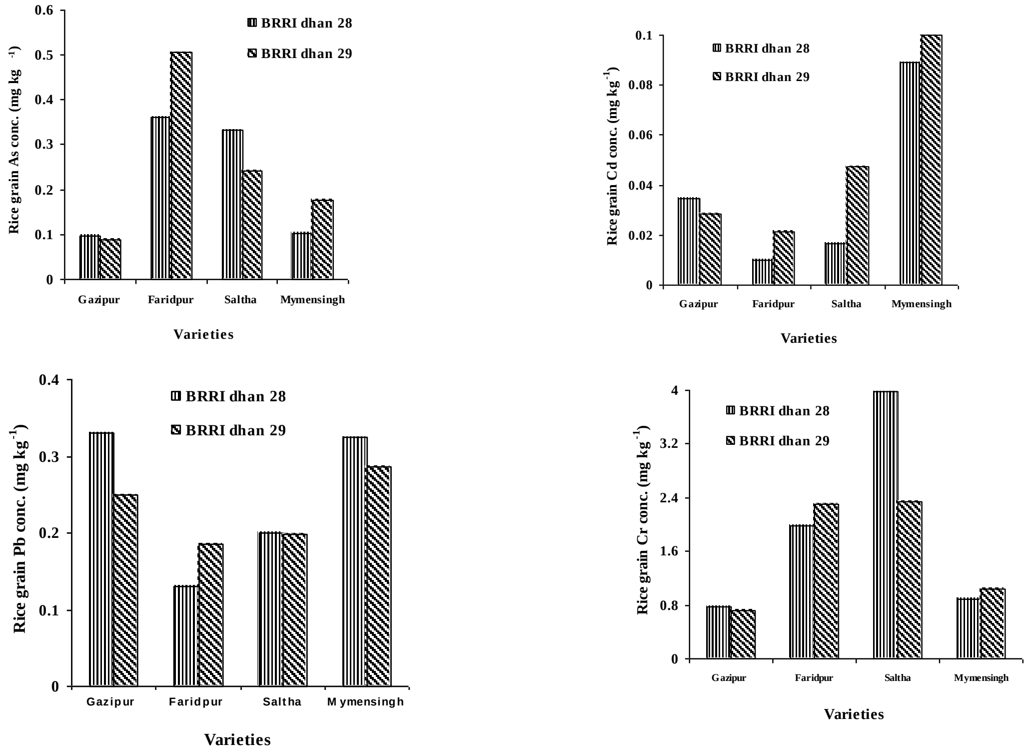

Fig 1. Location variations in grain As, Cd, Pb and Cr concentrations of two rice varieties.

Table 2. Descriptive statistics of arsenic, cadmium, lead and chromium concentrations (mg kg^{-1}) of rain-fed (transplant Aman) and irrigated (Boro) rice.

Element	Rice	Minimum	Maximum	Median	Mean	SD
Arsenic	Boro rice	0.047	0.506	0.110	0.153	0.112
	T. Aman rice	0.047	0.535	0.128	0.140	0.080
	All	0.047	0.535	0.123	0.144	0.091
Cadmium	Boro rice	0.007	0.297	0.062	0.073	0.069
	T. Aman rice	0.007	0.189	0.030	0.038	0.032
	All	0.007	0.297	0.032	0.049	0.050
Lead	Boro rice	0.105	0.602	0.250	0.264	0.125
	T. Aman rice	0.063	0.353	0.120	0.147	0.077
	All	0.063	0.602	0.148	0.185	0.109
Chromium	Boro rice	3.983	0.160	0.894	1.208	0.913
	T. Aman rice	0.148	4.616	0.811	0.986	0.796
	All	0.148	4.616	0.845	1.058	0.836

n = 48 (transplant Aman rice), 23 (Boro rice), 71 (All rice).

Table 3. Statistics of daily intake (µg) of arsenic, cadmium, lead and chromium by an adult male of 60 kg body weight.

Element	Minimum	Maximum	Median	Mean	S.d.
Arsenic (As)	18.6	214	49.0	57.6	36.4
Cadmium (Cd)	2.6	119	12.6	19.7	20.0
Lead (Pb)	25.0	241	59.0	74.1	43.5
Chromium (Cr)	59.0	1846	338	423	334

Table 4. Relationship between heavy metals with respect to grain concentration of rain-fed and irrigated rice.

Elements	Rain-fed samples (n = 23)	Irrigated samples (n = 48)	All samples (n = 71)
As vs Cd	- 0.299*	- 0.422*	- 0.319**
vs Pb	0.179	- 0.286	- 0.018
vs Cr	0.114	0.717***	0.376**
Cd vs Pb	0.114	0.099	0.253*
vs Cr	- 0.126	- 0.524**	- 0.260*
Pb vs Cr	0.325*	- 0.214	0.126

*, P<0.05; **, P<0.01; ***, P<0.001.

Table 5. 'T' statistics of arsenic, cadmium, lead and chromium concentrations (mg kg^{-1}) of rain-fed (transplant Aman) and irrigated (Boro) rice.

Element	Locations/ Soil type	Mean		df	't' value
		Boro rice	T. Aman rice		
Arsenic	All	0.153 (n = 23)	0.140 (n = 48)	33	0.497
	Calcareous	0.300 (n = 6)	0.186 (n = 12)	10	1.78*
	Non-calcareous	0.101 (n = 17)	0.107 (n = 8)	-0.399	-0.399
Cadmium	All	0.073	0.038	27	2.33*
	Calcareous	0.023	0.027	15	-0.434
	Non-calcareous	0.091	0.033	22	2.90**
Lead	All	0.264	0.147	30	4.12**
	Calcareous	0.467	0.151	17	1.58
	Non-calcareous	0.188	0.110	14	4.57**
Chromium	All	1.21	0.986	39	0.997
	Calcareous	2.36	0.814	7	4.10*
	Non-calcareous	0.800	1.229	12	-1.858*

*, P<0.05; **, P<0.01

Cr content (0.340 mg kg^{-1} for BRRI dhan 50 and 0.840 mg kg^{-1} for local aromatic varieties). Variety Miniket rice that grown in dry season had relatively higher Cr level (1.363 mg kg^{-1}).

Dietary intake of heavy metals through rice consumption

The As intake by an adult male was in the range of 18.6-214 µg d^{-1}, with the mean value 57.6 µg d^{-1}and median 49.0 µg d^{-1} (Table 2). The PTWI (Provisional Tolerable Weekly Intake) of Cd for a person is 6.25 µg kg^{-1} body weight, thus for a 60 kg adult the PTWI becomes 375 µg, equivalent to 53.6 µg Cd d^{-1}. Now, in the present study the intake of Cd by the adult male falls in the range of 2.6-119 µg d^{-1}, having the median value 12.8 µg d^{-1} and mean value 19.6 µg d^{-1} which on weekly basis becomes 137.2 µg (Table 2). This value is below the PTWI value. The Pb intake by adult male was found to vary from 25.0-241 µg d^{-1}, with the median 59.0 µg d^{-1} and the mean 74.1 µg d^{-1} (Table 3). The Cr intake was the highest, ranging from 59.0-1846 µg, the mean value being 423 µg which was 13.6, 4.7 and 17.5 times higher than that of As, Cd and Pb, respectively. The MTDI (maximum tolerable daily intake) of chromium (Cr) for an adult person (60 kg body weight) is 33 µg which is equal to 231µg wk^{-1} (WHO, 2002). The present study reveals that the daily intake of Cr by an adult ranged from 59.0-1846 µg, the median and mean values being 338 and 423 µg, respectively (Table 3). Thus, the upper values of Cr exceed the MTDI value showing a high risk of Cr exposure.

Discussion

Variations in As and heavy metals concentrations of rice grains

Concentrations of AS and heavy metals in rice grains were generally higher in irrigated rice (dry season) than in rain-fed rice (wet season). However, within the wet season rice, considerable variation in heavy metal concentrations in rice grains was found for some locations. This result can be explained that contaminated irrigation water is a reason for relatively higher concentration of heavy metals in dry season (Boro) rice and location specific contaminated soil could be a reason for higher concentration of heavy metals in rice e.g. arsenic contaminated Faridpur and Saltha soils (Hossain et al., 2008).

There are a number of reports of elevated rice As in Bangladesh and West Bengal, India where paddy fields are irrigated with As-contaminated groundwater (Meharg and Rahman, 2003; Islam et al., 2004). As reviewed and observed by Williams et al. (2006), the rice As levels in Asia-Pacific regions are 0.06-0.14 mg kg^{-1} for Thailand, 0.00-0.25 mg kg^{-1} for Philippines, 0.02-0.04 mg kg^{-1} for Australia, 0.07-0.19 mg kg^{-1} for China, 0.03-0.07 mg kg^{-1} (Basmati) for India and 0.08-0.36 mg kg^{-1} (rain-fed rice) and 0.09-0.51 mg kg^{-1} (irrigated rice) for Bangladesh. Thus, result of the present study agrees well to the reported values. Meharg et al. (2013) reported Cd levels over 260 rice samples across 12 districts of Bangladesh in the range of <0.005-1.31 mg kg^{-1}, with the mean of 0.099 mg kg^{-1}. The present results are within the range. Fig. 1 shows elevated grain As and Cr concentrations for the rice samples from Faridpur & Saltha upazila (sub-districts) representing calcareous soil ($CaCO_3$ rich), elevated Cd concentration for Mymensingh and Pb for Mymensingh & Gazipur. This result indicates that location is a dominant factor, not the rice varieties.

Relationship between As and heavy metals for rice grain concentrations

Correlation statistics is done to examine the relationship, if any, between the variables (As, Cd, Pb and Cr) in respect of rice grain concentrations, the 'r' values shown in Table 4. The As concentrations of rice grain were negatively correlated with those of Cd (r = -0.319, P<0.01) and positively with Cr (r = 0.376, P<0.01) while Cd in grain was positively correlated with Pb (r = 0.253, P<0.05) and Cr (r = -0.260, P <0.05) across all 71 rice samples. The 'r' values between As and Cd (r = -0.299, P<0.05) and between Pb and Cr (r = 0.325, P<0.01) were significant for rain-fed rice samples only, however other relationships such as As vs Pb or Cr and Cd vs Pb or Cr were not significant. Concerning irrigated rice samples, the 'r' values for As vs Cd (r = -0.422, P<0.05), As vs Cr (r = 0.717, p<0.001) and Cd vs Cr (r = 0.524, P<0.01) were significant. It appears that rice As exhibited negative correlation with rice Cd showing a negative interaction between them.

Comparison between irrigated and rain-fed rice for heavy metals concentrations

Unpaired 'T' statistics was performed to compare mean differences in grain concentrations of heavy metals between irrigated and rain-fed rice (Table 5). This statistics shows that there was a significant difference in grain concentration between irrigated (dry season) and rain-fed (wet season) rice for Cd and Pb, not for As and Cr when all 23 Boro (irrigated) and 48 transplant Aman (rain-fed) rice samples were brought under T-test. But when such comparison was made within data set of calcareous zone (Faridpur & Saltha) and that of non-calcareous zone (Mymensingh & Gazipur), the situation was different. The 't' values were found significant for all elements except Cd for the rice samples from calcareous zone, but for the non-calcareous zone, the 't' values for Cd and Cr showed significant and that for As and Pb was not significant. This result is supportive of higher concentration of heavy metals in irrigated rice than in rain-fed rice.

Human health risk of consuming contaminated rice

The Maximum Tolerable Daily Intake (MTDI) for As is 2.1 µg d^{-1} kg^{-1} body wt^{-1} (WHO, 2001), thus acceptable intake of As is 126 µg d^{-1} for a 60 kg male adult. When As ingestion is modeled on 400 g of daily rice dry weight consumption rate (HIES, 2010) for a 60 kg adult the intake of 57.6 µg equates to 46% of the MTDI. Similarly, intake of Cd, Pb and Cr equates to 57%, 50% and 60% of the MTDI, respectively. Thus, rice diet might alone contribute to 46-60% of the MTDI as far as heavy metals As, Cd, Pb and Cr are concerned. The daily As intake estimates varied substantially among the countries. The average daily intake of As (57.6 µg) by adults in this study through rice exclusively is comparable to estimates reported from Canada (59.2µg; Dabeka et al., 1993), UK (52.6 µg; WHO, 2000) and Australia (73.3 µg; WHO, 2000) and it is much lower than the reported values from Japan (182 µg; WHO, 2000) and India (186 µg; Roychowdhury et al., 2003). Sharma et al. (2006) reported that the estimated daily intake of Cd in different countries ranges from 20-60 µg day^{-1} while the tolerable daily intake of Cd is about 57-72 µg d^{-1}. Thus, the present level of Cd in rice grain is within the safe limit.

The maximum intake for all heavy metals has exceeded the tolerable level (Table 3). To date, contamination of food by arsenic has received much attention in Bangladesh, but not the other metals, although the levels of rice contamination by Cr, Cd and Pb are high in many cases. If we set the upper intake limit, for instance, to be 80% of the tolerable level, the number of samples that would exceed the Maximum Tolerable Daily Intake (MTDI) limit is estimated as 10% for As, 17% for Cd & Pb and 15% for Cr. Arsenic is class 1 carcinogen and hence the WHO (2010) has withdrawn the previous PTWI (Provisional Tolerable Weekly Intake) of 15 µg As, equivalent to 2.1 µg kg^{-1} body wt.d^{-1} (FAO/WHO, 1989). With that consideration, food having any amount of As is unsafe for consumption.

Materials and methods

Collection of rice grain samples

A total of 71 rice grain samples were collected from 10 upazilas (sub-districts) of Bangladesh. More detail on sampling procedure (where were they collected?) and the procedure for avoiding contamination of samples at collection The samples represent different planting seasons (wet and dry seasons), locations (industrial and non-industrial areas) and varieties (local and modern varieties). Proper labeling was done for each of the collected samples. Rain-fed and irrigated rice represent wet and dry seasons, respectively. Rain-fed rice is grown during July to October and irrigated rice during February to May. Elements tested in grain were: arsenic (As), cadmium (Cd), lead (Pb) and chromium (Cr).

Chemical analysis of rice grain samples

The As, Cd, Pb and Cr concentrations of rice grains were chemically analyzed at the Institute of Tropical Agriculture of Kyushu University, Japan. All rice samples were dehusked by grinding with a pestle in a mortar and then finely ground in an electric grinder. The samples were stored for digestion in plastic bags at room temperature ($25 \pm 1\ ^0C$). Samples weighing 0.5 g were digested with ultrapure grade nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) at 120 ^0C for a period until the digest became colorless following the procedure as outlined by The Soil & Plant Analysis Lab, Wisconsin University, USA (2005). Each digest was diluted to 25 ml with deionized water. All the digests were filtered through a 4-µm filter (No. 5B; Toyo Roshi Kaisha, Ltd.) and kept in a refrigerator until analysis. Elemental concentrations in the digested samples were determined by inductively coupled plasma - mass spectrometry (ICP-MS) (Agilent 7500c; Agilent Technologies, Inc

Human health risk assessment of rice consumption

Diets are the main route of exposure to contaminants, so the assessment risks of these elements to humans via dietary intake is important. Daily intake of As, Cd, Pb and Cr through rice grains was estimated using the formula (Kile et al., 2007): $DI = C \times Q$, where DI stands for daily intake, C for element concentration and Q for average daily consumption of rice which is 400 g/day/60 kg adult male (HIES, 2010).

The intake of heavy metals was compared with Maximum Tolerable Daily Intake (MTDI), Provisional Tolerable Weekly Intake (PTWI) or Provisional Tolerable Monthly Intake (PTMI), as recommended by the Joint FAO/WHO Expert Committee on Food Additives (FAO/WHO, 1999, 2011, 2013 and EC, 2003). This value provides an indication of the amount of toxic heavy metals that can be consumed on a daily/weekly/monthly basis without appreciable risk.

Conclusion

Arsenic, cadmium, lead and chromium concentrations of rice grain markedly varied with locations of sample collection and growing seasons. The concentration of elements in rice grains across the locations followed the order of Cr > Pb >As > Cd for both irrigated and rain-fed rice. Concentrations of heavy metals were found higher in irrigated rice than in rain-fed rice. For rain-fed rice, heavy metal concentration generally did not vary significantly by location, which could be due to rain-fed crop culture. In rain-fed culture aerobic condition arises at times which reduces arsenic uptake. Of the 71 rice samples under test, 10% of samples for As, 17% for Cd & Pb and 15% for Cr have exceeded the Maximum Tolerable Daily Intake (MTDI). Further it is estimated that a typical rice diet may alone contribute 46-60% of the MTDI as far as As, Cd, Pb and Cr are concerned. Research is needed to screen out and develop rice varieties for lower uptake of heavy metals by rice grains in order to decrease the risks of As and heavy metals consumption by humans.

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