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Geographical variation and age-related dietary exposure to arsenic in rice from Bangladesh

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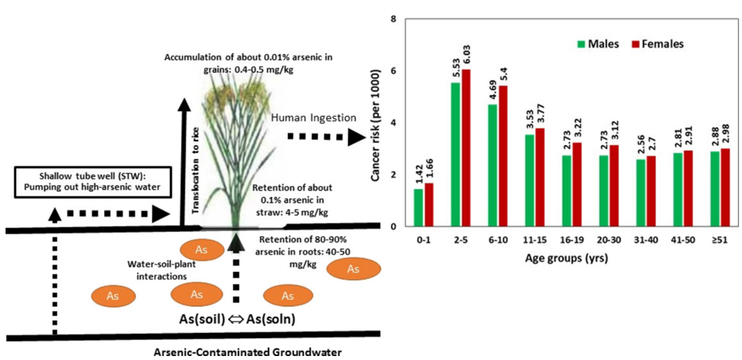
HIGHLIGHTS

- Arsenic levels in rice are dominated by the location and variety.
- Daily intake of inorganic As from rice ranged between 0.38 and 1.92 μg/kg BW.
- The excess cancer risk higher than ranged used by the US EPA as a threshold
- The hazard quotient of iAs suggests very high potential non-carcinogenic risk.
- The female groups are more susceptible than male to carcinogenic risks.

GRAPHICAL ABSTRACT



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ABSTRACT

An extensive number (965) of rice samples collected by household survey from 73 upazilas (i.e. sub-districts) in Bangladesh was analyzed to determine regional variation, distribution and associated health risks from arsenic (As). No previous study had conducted a study examining such a large number of rice samples. The mean and median concentrations of total As were 126 μg/kg and 107 μg/kg, respectively, ranging from between 3 and 680 μg/kg. Importantly, total As levels of aromatic rice were significantly lower (average 58 μg/kg) than non-aromatic rice (average 150 μg/kg) and also varied with rice grain size. The variation in As content was dominated by the location (47% among the upazilas, 71% among districts) and rice variety (14%). Inorganic As content in rice grain ranged between 11 and 502 μg/kg ($n = 162$) with the highest fraction being 98.6%. The daily intake of inorganic As from rice ranged between 0.38 and 1.92 μg/kg BW in different districts. The incremental lifetime cancer risk (ILCR) for individuals due to the consumption of rice varied between 0.57×10^{-3} to 2.88×10^{-3} in different districts, and 0.54×10^{-3} to 2.12×10^{-3} in different varieties, higher than the US EPA threshold. The 2–10 age group experiences higher carcinogenic risks than others and females are more susceptible than males.

1. Introduction

Accumulation of As in rice grain from paddy soils and irrigation water poses a potential health risk to millions of people worldwide (Correll et al., 2006; Nordstrom, 2002; Panaullah et al., 2009; Smedley and Kinniburgh, 2002; Williams et al., 2006). Bangladesh is an example

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of As occurring naturally in alluvial sediments that being mobilized into the groundwater. A substantial number of people rely on this groundwater for drinking as well as irrigation for paddy rice during the dry season and consequently exhibit additional health risks from these types of rice (Meharg et al., 2009; Williams et al., 2006). Rice is grown as a lowland crop in flooded paddy soils under reducing conditions where As availability is higher than under oxidizing conditions (Duxbury et al., 2003). Consequently, the reductive mobilization greatly enhances the bioavailability of As to rice, leading to excessive bioaccumulation in rice grain and straw. In countries like Bangladesh, India, China and the US, rice often predominantly contains even higher levels of inorganic As (10% to 90%), depending on As concentration in irrigation water, soil and the uptake mechanism of rice varieties (Williams et al., 2005). The grain-As concentrations are also impacted by the combined influences of soil characteristics, environmental conditions, and crop management (Khan et al., 2009; Ohno et al., 2007; Panaullah et al., 2009) as well as rice varieties. A number of studies have already reported the concentrations and nature of As in rice from Bangladesh and other countries. Arsenic content in rice varies widely, with most reported concentrations found in the 20 to 900 μg/kg range (Meharg and Zhao, 2012). Recent studies indicated that rice genotypes have w*ide variations* in total grain-As concentrations and As speciation has been reported in rice around the world (Meharg et al., 2009; Rahman et al., 2011; Williams et al., 2007). Total As content in rice grains from Bangladesh varies from 58 to 1835 μg/kg (Meharg and Rahman, 2003). While this initial study shows the As in rice grains can be as high as 1835 μg/kg, it has been shown that As in rice grains was usually below 1000 μg/kg (Rahman et al., 2009b).

Arsenic is classified as a chronic carcinogen as well as an acute toxin at higher concentrations and exposure to it at elevated levels that causes serious illness including different types of cancers (IARC, 2004). Inorganic As in drinking water has been studied and is linked to human carcinogenesis and exposure is associated with various internal cancers as well as other health problems, including skin cancer and diabetes, etc. (Guo et al., 1997). Arsenic in rice is a serious concern for billions of people and a significant number of them may be at risk of developing As-related health problems (Meharg and Zhao, 2012). Arsenic contamination and low levels of micronutrients in rice have been recognized as a major human health issue (Williams et al., 2009). Exposure to arsenic from rice ingestion is a significant risk factor for cancers, especially for people who heavily depend on a rice diet (Mondal and Polya, 2008). Therefore, rice itself plays a potential exposure route to humans but the data on rice's direct impact on humans is very limited. Banerjee et al. (2013) reported that cooked rice with 200 μg/kg As causes genotoxic effects. Since it is the staple food crop in Bangladesh, the dietary exposure to As and associated risks to human health is an important issue. It is thus essential to investigate the levels of inorganic As in rice to assess the human health risks. A number of articles have already been published based on the presence and concentration of As in As-polluted areas of Bangladesh (Duxbury et al., 2003; Meharg and Rahman, 2003; Williams et al., 2006; Williams et al., 2005). A small number of published articles also estimated the intake by adult males and females (Correll et al., 2006; Ohno et al., 2007) but the associated cancer risks could be considerable and currently unclear from the rice samples of Bangladesh.

Here we report the largest household survey of As in Bangladesh rice to date. Nine hundred and sixty-five rice grain samples were collected from household families in different districts. In each case the rice was used for their own consumption. The objectives of this study are to: firstly, provide data of total and inorganic As content rice samples; secondly, assess daily intake and potential carcinogenic and non-carcinogenic health risk to local inhabitants (different male and female age groups) via the consumption of rice; and thirdly, provide data on regional, grain and seasonal variations.

2. Materials and methods

2.1. Sample collection and preparations

In this study, we analyzed 965 polished rice samples collected from a range of locations through household surveys from 73 upazilas in 20 districts of Bangladesh during April 2014. Districts were selected based on the As concentrations (severely, moderately and less affected) in drinking water covering most regions except hilly areas. The sampling sites are shown in Fig. SM1 (Supplementary material, SM). The collected rice samples were from both dry (Boro rice) and wet (Aman rice) seasons, and include most local, high yielding variety (HYV) and hybrid varieties. Raw grain samples were washed with deionized water, and finally dried at 65 °C in an oven for 48 h. The samples were then homogenized by grinding them with mortar and pestle. To avoid cross-contamination the mortar and pestle was wiped clean between samples.

2.2. Sample preparations for total and speciated As

Digestion of the rice samples for total As analysis was carried out employing the method used by Rahman et al. (2009b), i.e. approximately 0.5 g ground rice sample were weighed directly into a 75 mL digestion tube and 5 mL concentrated HNO_3 (trace analytical grade, 70%), obtained from Fisher Chemicals was added to it. The mixture was then allowed to stand overnight under fume hood. In the following day, the digestion tubes was heated using temperature controlled digestion block (A.I. Scientific Block Digestion System AIM 500), programmed to slowly ramp up to 140 °C over 8 h and then to maintain temperature for the digestion of rice samples. Sample digestion was continued until only a small residual liquid remained in each tube. The tubes were removed from the digestion block and allowed to cool to room temperature in the fume cupboard prior to dilution (10 mL). The samples were mixed thoroughly by vortexing and filtered by a 0.45 μm syringe filter directly into plastic containers for storage prior to analysis.

A As speciation study (samples, $n = 162$) was conducted after extracting rice grain samples (approximately 0.25 g) with 2 M trifluoroacetic acid (TFA) as done by Abedin et al. (2002). The digests were diluted to 10 mL and filtered by a 0.45 μm syringe filter and finally, 1 mL aliquot of the digest from each tube was transferred to a 2 mL HPLC glass vial for chromatographic analysis.

2.3. Analytical methods

7500ce series inductively coupled plasma mass spectrometer (ICP-MS) (Agilent Technologies, Tokyo, Japan), coupled with an autosampler (ASX-520, CETAC Technologies) and integrated samples introduction system (ISIS) served to determine the amount of total As. For As speciation a liquid chromatography system (model 1100, Agilent Technologies, Tokyo, Japan) equipped with a guard and Hamilton PRP-×100 separation column, coupled with ICP-MS was used. The details of the operating parameters for ICP-MS and ion chromatography (IC) system were already provided in our earlier publication (Rahman et al., 2009c). The detection limit (DL) and limit of quantification (LOQ) of the total As were 0.01 μg/kg and 0.1 μg/kg, respectively. The detection limit for speciation in As standard solutions [As(III), As(V), DMA and MMA] by the IC-ICP-MS ranged between 0.1 and 0.3 μg/kg (Chen et al., 2008). The limit of quantification (LOQ) for As species in rice were ranged between 2.5 and 3 μg/kg.

2.4. Analysis of standard reference materials (SRM)

During the analysis of total As, a SRM1568b rice flour [National Institute of Standard and Technology (NIST), USA] was used to validate the total As concentration in rice. We found the concentration of total As in rice flour was SRM 270 ± 13 ($n = 65$) μg/kg, which indicates 95%