

# Arsenic Exposure from Rice and Water Sources in the Noakhali District of Bangladesh

Mohammad Mahmudur Rahman · M. Asaduzzaman · Ravi Naidu

Received: 15 August 2010 / Revised: 30 November 2010 / Accepted: 2 December 2010 / Published online: 24 December 2010  
© Springer Science+Business Media B.V. 2010

**Abstract** Drinking water and food are the two major routes of the consumption of arsenic (As) by people in Bangladesh. This study reports exposure of adults in two adjacent rural villages of Bangladesh to As from drinking water and rice. Uncooked and cooked rice, drinking and cooking water, were sampled from 14 families to determine total As concentrations. Arsenic speciation in uncooked rice was also conducted to determine the percentage of inorganic As (arsenite and arsenate). Arsenic intake by people from rice and drinking water was also assessed. The average water intake rates were 3.2 and 2.7 L per day for adult males and females, respectively, whereas no significant gender difference was observed for rice intake. The study revealed that with one exception, all the families examined drank As-contaminated groundwater. They used As-safe pond water for cooking. The mean As concentrations in drinking and cooking water were 328 and 2.5 µg/L, respectively. The average As concentrations in uncooked and cooked rice were 153 and 139 µg/kg (dry weight), respectively. On average, 73% of the As present in uncooked rice was inorganic. The study showed that adult males and females consume 1099 and 933 µg of inorganic As per day, respectively, from rice consumption and drinking water. Rice contributed 49 and 47 µg per day of inorganic As for adult males and females, respectively. The study concluded that the villagers in the

study area consumed considerable amount of As from drinking water and that exposure from rice was also not negligible, despite intensive programs by governmental and non-governmental agencies to reduce villagers' exposure to As consumption.

**Keywords** Arsenic · Drinking water · Rice · Arsenic speciation · Arsenic intake

## Introduction

Chronic exposure of humans to As through drinking water causes a range of dermatological signs and symptoms such as melanosis, leuco-melanosis and keratosis (NRC 2001; WHO 2001). Arsenic exposure may also cause respiratory problems, nervous system disorders, obstetric problems, diabetes, hypertension, cardiovascular diseases, as well as cancers relating to the skin, lung, liver, kidney and bladder (WHO 2001; NRC 2001; IARC 2004). Groundwater extracted by hand tubewells is the major source of potable water for the majority of the rural population of Bangladesh. Geogenic As has been detected in the groundwater of Bangladesh since 1992 (Dhar et al. 1997). The British Geological Survey (BGS) analysed water samples from 3,534 tubewells from 61 of the 64 districts of Bangladesh and reported that 35 million people are exposed to water with As concentrations above 50 µg/L, the country's maximum permissible As concentration for drinking water (BGS 2001). The mean As concentration was 55 µg/L and the maximum As concentration detected in a tubewell was 1670 µg/L (BGS 2001). Based on 52,202 hand tubewell water samples from throughout Bangladesh, Chakraborti et al. (2010) recently found that 42% of the samples had As concentrations above 10 µg/L and 27% were above 50 µg/L.

---

M.M. Rahman · M. Asaduzzaman · R. Naidu  
Centre for Environmental Risk Assessment and Remediation (CERAR), University of South Australia, Mawson Lakes Campus, Mawson Lakes, SA 5095, Australia

M.M. Rahman · M. Asaduzzaman · R. Naidu (✉)  
Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC-CARE), P.O. Box 486, Salisbury South, SA 5106, Australia  
e-mail: ravi.naidu@crccare.com

In addition to supplying drinking water, As-contaminated groundwater is extensively used for the irrigation of crops in Bangladesh and West Bengal, India. As a result, food crops may accumulate elevated levels of As so that it poses potential threat to human health. A huge amount of As is deposited on to the farmlands of Bangladesh due to extensive use of groundwater for irrigation. It is therefore important to assess As exposure from commonly used food crops such as rice and vegetables grown on agricultural lands. Rice is the main food of the Bangladeshi population (Abedin et al. 2002) and the majority (>95%) of the people use groundwater for drinking. For this reason, it is crucial to determine As levels in rice and drinking water. Several published articles have already reported the levels of As in rice and vegetables grown in As-contaminated regions of Bangladesh (Alam et al. 2003; Duxbury et al. 2003; Meharg and Rahman 2003; Das et al. 2004; Williams et al. 2005, 2006; Rahman et al. 2009). A few articles also estimated the intake of As by adult Bangladeshis (males and females) from food and drinking water (Correll et al. 2006; Smith et al. 2006; Ohno et al. 2007; Kile et al. 2007; Rahman et al. 2009). Various studies (Bae et al. 2002; Smith et al. 2006; Ohno et al. 2007; Pal et al. 2009; Mondal et al. 2010) have already demonstrated the level of As in cooked rice from Bangladesh and West Bengal in India. The concentration of As in cooked rice can considerably vary compared to uncooked rice based on the concentration of As in cooking water (Mondal et al. 2010; Pal et al. 2009; Ohno et al. 2007; Smith et al. 2006; Rahman et al. 2006; Bae et al. 2002). Thus, it is essential to determine As levels in cooked rice in contrast to uncooked rice, to determine the contribution of As from cooking water.

The form of As present in rice is also important as the main Bangladeshi consume food items such as rice and vegetables, which predominately contain inorganic As [arsenite, As(III) and arsenate, As(V)] which is more toxic than organic forms [monomethyl arsenic acid, MMA(V) and dimethyl arsenic acid, DMA(V)] of As (Williams et al. 2005). The main As species detected in Bangladeshi rice is As(III) or As(V) with DMA(V) being a minor component (Williams et al. 2005; Smith et al. 2006; Ohno et al. 2007).

In this study, we collected drinking and cooking water sources used by 14 families in two adjacent rural villages (Chiladi and Basantapur) of Senbagh thana in the Noakhali district of Bangladesh to determine the consumption of As by adults from drinking water and cooking water. Noakhali is one of the most highly As-contaminated districts in Bangladesh. It was reported in a recently published article that 95% of 843 hand tubewell water samples from Noakhali district had As concentrations above 50 µg/L, and 11% exceeded 1000 µg/L (Chakraborti et al. 2010). The maximum concentration detected in a hand tubewell was 4,730 µg/L (Chakraborti et al. 2010). Rice samples (uncooked and cooked) were also collected to estimate

As ingestion from the villagers' main foodstuff. The main objective of this study were to assess the quantity of total and inorganic As consumed by the adults of the surveyed families from rice and water sources, including drinking and cooking sources. Mondal et al. (2010) recently estimated the total As exposure from different exposure routes (drinking water, rice and cooking of rice) for three contrasting areas [high (Bhawangola-I block, Murshidabad district), moderate (Chakdha block, Nadia district) and low (Khejuri-I block, Midnapur district) As levels in drinking water] of West Bengal. The data of the present study was compared with the results of Mondal et al. (2010). The concentrations of inorganic As in uncooked rice were also measured to improve understanding of the risks from rice consumption by adults.

## Materials and Methods

### Sample Collection and Preparation

Water sources (both drinking and cooking water) and rice (both uncooked and cooked) were collected from 14 families in two villages (Chiladi and Basantapur) of Noakhali district of Bangladesh in December 2008. Most of the villagers in the study area consume the rice grown in their own paddy fields. Generally As-contaminated groundwater is used for irrigating the agricultural crops, especially rice in dry season in the study area. All surveyed families revealed that the collected rice samples were grown locally and harvested in wet season (Aman rice). We were unable to collect cooked rice samples from one family (No. 9). All surveyed families used pond water for cooking at the time of the survey. All family members drank water from their own tubewells. All groundwater and pond water samples were collected in polyethylene bottles without filtering. Nitric acid was added after collection as a preservative. During the field sampling, the intake rates of water and rice per day of each individual were noted for all surveyed families by interviewing them. Age, gender and knowledge of As contamination or toxicity were also noted during field survey.

Uncooked rice samples were washed with de-ionized water, and after removing the excess water, the samples were dried in an oven at 65°C. Cooked rice samples were dried without washing. The samples were homogenized by grinding.

### Sample Digestion and Analysis

Concentrated nitric acid (Mallinckrodt Chemicals, USA) was used for the digestion of the uncooked and cooked rice samples by the procedure of Rahman et al. (2009). For As speciation, rice samples were extracted with 2 M trifluoroacetic acid (TFA) according to the procedure of Abedin et al. (2002).

An Agilent 7500c (Agilent Technologies, Tokyo, Japan) inductively coupled plasma mass spectrometry (ICPMS) was used for the determination of As in drinking and cooking water, and digested rice samples. An Agilent 1100 liquid chromatography system (Agilent Technologies, Tokyo, Japan) equipped with a guard column and a Hamilton PRP-X100 separation column, coupled with ICPMS was used to determine As species [As(III), DMA(V), MMA(V) and As(V)] in rice samples. The details of the instruments and the procedure for speciation of As have been described earlier (Chen et al. 2008).

#### Quality Control: Analysis of Standard Reference Materials (SRM)

Trace elements in natural water (SRM 1640) and trace elements in water (SRM 1643e), and rice flour (SRM 1568a) from the National Institute of Standard and Technology (NIST), USA, and unpolished rice flour (CRM 10) from the National Institute for Environmental Studies (NIES), Japan were used to verify the analytical results for As. The standard reference materials (SRM) were analysed after employing the same digestion and analysis procedures as the samples. The certified concentration of As in NIST SRM 1640 is  $26.67 \pm 0.41$   $\mu\text{g/L}$  and in NIST SRM 1643 it is  $58.98 \pm 0.7$   $\mu\text{g/L}$ . From the analytical results, the mean As concentration of NIST SRM 1640 was detected as  $26.86 \pm 0.45$   $\mu\text{g/L}$  ( $n = 8$ ) with a recovery of 100.7% and the mean As concentration of NIST SRM 1643 was  $56.42 \pm 1.1$   $\mu\text{g/L}$  ( $n = 8$ ) with a recovery of 95.6%.

The certified concentration of As in NIST SRM rice flour is  $0.290 \pm 0.03$   $\mu\text{g/g}$  and the reference value of As in NIES CRM rice flour is  $0.170$   $\mu\text{g/g}$ . Our results were an average As concentration in NIST SRM rice flour of  $0.263 \pm 0.012$   $\mu\text{g/g}$  ( $n = 3$ ) with a recovery of 91%, and in NIES CRM unpolished rice flour, it was  $0.155 \pm 0.02$   $\mu\text{g/g}$  ( $n = 3$ ), with a recovery of 93%.

There is no certified material currently available for rice grain speciation. However, NIST SRM 1568a was used to validate the results of As speciation in rice samples using the extraction procedures of 2M TFA (Abedin et al. 2002). The detection limit for speciation in As standard solutions by the IC-ICPMS ranged between 0.1 and 0.3  $\mu\text{g/L}$  (Chen et al. 2008). The As speciation result of NIST SRM is given in Table 1. The sum of As species was  $257 \pm 6$   $\mu\text{g/kg}$  ( $n = 4$ ), which was consistent with other studies on As speciation in NIST SRM rice sample. Table 1 also presents the comparison of the results (As speciation of standard reference material rice flour, NIST SRM 1568a) of this work with those published by others. The percentage of DMA(V) and inorganic As were close to other published studies (Table 1). Surprisingly, this study could not detect any MMA(V) in the NIST SRM rice samples whereas most of the studies reported a small amount of MMA(V) except Smith et al. (2006) study.

## Results and Discussion

### Arsenic Content in Drinking Water Sources

The mean, median and range of As in tubewell water samples recorded were 328, 279 and 14–772  $\mu\text{g/L}$ , respectively. Arsenic concentrations in all drinking water samples exceeded the recommended level for drinking water set by the WHO (10  $\mu\text{g/L}$ ). The average As concentration in tubewell water samples was almost 33 times greater than the WHO recommended level. Out of 14 water samples, only a single sample was found to be As-safe according to the Bangladesh standard (50  $\mu\text{g/L}$ ). Therefore all surveyed families except one drank water from tubewells that contained levels of As above those considered safe. In a recent study conducted from three contrasting areas (high, moderate and low As levels in drinking water) of West Bengal, the mean As concentrations was reported as 130, 40 and 1  $\mu\text{g/L}$  for Bhawangola-I, Chakdha and Khejuri I blocks, respectively (Mondal et al. 2010). The mean As concentration in the study area is 2.5 and 8 times greater than the highly (Bhawangola-I) and moderate (Chakdha) As-impacted area of West Bengal as reported by Mondal et al. (2010).

### Arsenic in Cooking Water Sources

All cooking water samples were found to have As concentrations below the WHO guideline value (10  $\mu\text{g/L}$ ). The range for As in all cooking water samples was 0.3–8.6  $\mu\text{g/L}$ , with a mean value of 2.5  $\mu\text{g/L}$ . Based on the recent household survey conducted from three contrasting areas of West Bengal, Mondal et al. (2010) reported that the mean As concentrations in cooking water for Bhawangola-I, Chakdha and Khejuri I blocks were 130, 37 and 1  $\mu\text{g/L}$ , respectively. Mondal et al. (2010) also reported that all surveyed families used As-contaminated tubewell water for cooking except Khejuri I block which was used as control, whereas all surveyed families involved in this study used As-safe pond water for cooking. Figure 1 shows bar diagram of As concentrations in drinking and cooking water sources used by 14 families included in this study.

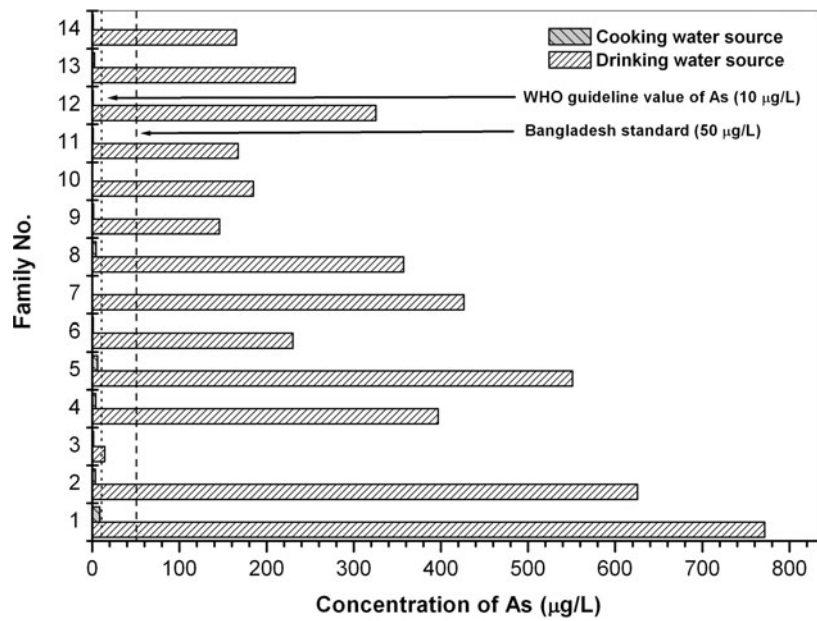
### Arsenic Content in Uncooked and Cooked Rice

The mean As concentration in 14 uncooked Aman rice samples was 153  $\mu\text{g/kg}$  (dry wt.) with a range of 74–302  $\mu\text{g/kg}$ . In a previous study from 25 villages in four districts of Bangladesh (Comilla, Brahmanbaria, Munshiganj and Manikganj), the mean concentration of As in rice grain (polished) was found to be 143  $\mu\text{g/kg}$  (Rahman et al. 2009). Duxbury et al. (2003) reported that the mean As concentration in Aman rice was 117  $\mu\text{g/kg}$ , based on the rice grain samples collected from different districts of Bangladesh.

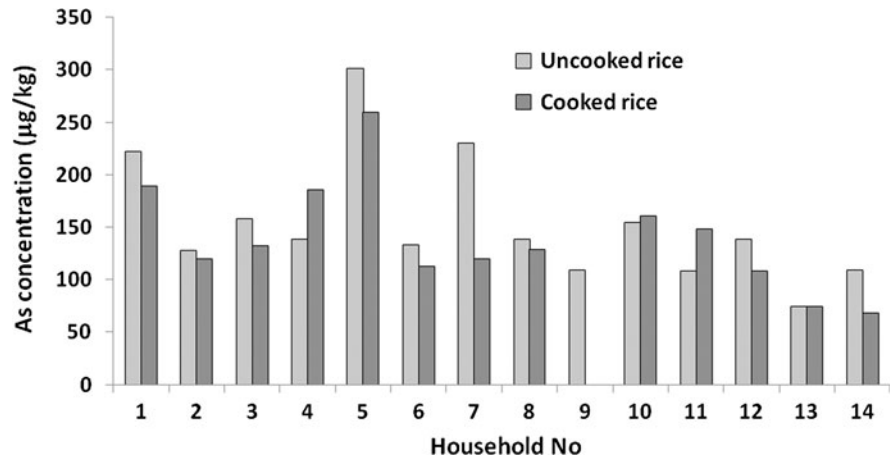
**Table 1** Comparison of the results (As speciation) of standard reference material for rice flour, NIST SRM 1568a from this work with those published by others

Certified values ( $\mu\text{g}/\text{kg}$ )	<i>n</i>	As(III) ( $\mu\text{g}/\text{kg}$ )	As(V) ( $\mu\text{g}/\text{kg}$ )	Total inorganic As ( $\mu\text{g}/\text{kg}$ )	DMA(V) ( $\mu\text{g}/\text{kg}$ )	MMA(V) ( $\mu\text{g}/\text{kg}$ )	Species sum ( $\mu\text{g}/\text{kg}$ )	Extraction efficiency (%)	% of DMA(V)	% of inorganic As	Reference
290 $\pm$ 30	4			91 $\pm$ 11	166 $\pm$ 13		257 $\pm$ 6	89	57	31	This study
	3			92 $\pm$ 4	174 $\pm$ 9	8 $\pm$ 2	274	94	63	34	Heitkemper et al. (2001)
	13	67 $\pm$ 4	39 $\pm$ 3	106 $\pm$ 3	158 $\pm$ 5	13 $\pm$ 2	277	96	54	38	Kohlmeier et al. (2003)
	16	55 $\pm$ 1	54 $\pm$ 3	109	165 $\pm$ 8	15 $\pm$ 2	289	99	57	38	D'Amato et al. (2004)
	6			80 $\pm$ 14	160 $\pm$ 24	2	242 $\pm$ 40	83 $\pm$ 12	54	26	Williams et al. (2005)
	3	77 $\pm$ 2		97 $\pm$ 10	132 $\pm$ 10	1.5	245 $\pm$ 10	84 $\pm$ 3	51 $\pm$ 3	33 $\pm$ 2	Williams et al. (2006)
	3	90 $\pm$ 4	22 $\pm$ 1	112	163 $\pm$ 32		240	83	56	26	Smith et al. (2006)
	3	68 $\pm$ 4	21 $\pm$ 2	89	162 $\pm$ 10	11 $\pm$ 1	285	98	56	38	Ohno et al. (2007)
	3				135 $\pm$ 4	8 $\pm$ 1	232 $\pm$ 5	80	47	31	Sanz et al. (2007)

**Fig. 1** Arsenic levels in drinking and cooking water used by 14 households



**Fig. 2** Bar diagram of As concentrations in uncooked and cooked rice used by 14 households



Williams et al. (2005) previously reported that the mean As concentration in uncooked rice samples ( $n = 15$ , majority of the rice samples was Aman) collected from Bangladesh was  $130 \mu\text{g}/\text{kg}$ . Arsenic concentrations in uncooked rice in this study are almost similar to those of previous studies. Williams et al. (2006) analysed 189 Aman rice samples from 25 districts of Bangladesh and the range of As concentration was  $<40\text{--}920 \mu\text{g}/\text{kg}$ . However, most of the Bangladeshi rice samples analysed in previous studies showed almost similar mean As values (range:  $117\text{--}183 \mu\text{g}/\text{kg}$ ), exceptions are there. Recently Mondal et al. (2010) reported that the mean As concentrations in raw rice samples for Bhawangola-I, Chakdha and Khejuri I blocks of West Bengal were  $120, 160$  and  $120 \mu\text{g}/\text{kg}$ , respectively. It is noteworthy that Bhawangola-I, Chakdha blocks are As-exposed areas and Khejuri I block is unexposed area. But the mean As levels in rice are broadly comparable with other studies conducted from Bangladesh and West Bengal (Mondal et al.

2010). Meharg and Rahman (2003) reported much higher concentrations of As ( $496 \mu\text{g}/\text{kg}$ ) in rice from Bangladesh, yet Al Rmalli et al. (2005) found very low concentrations of As ( $11.3 \mu\text{g}/\text{kg}$ ) in Bangladeshi rice sold in UK markets. These variations are probably due to either soil variations in As or potentially cultivar variations (Lu et al. 2009; Norton et al. 2009). Khan et al. (2009) mentioned that if the rice was obtained from markets or imported to the village from neighbouring localities or from other countries, it hence was potentially lower in As due to the mixing of rice from various geographic locations.

The mean, median and range of 13 cooked rice samples were  $139, 128$  and  $68\text{--}260 \mu\text{g}/\text{kg}$ , respectively. There was no significant difference in the mean As concentrations between uncooked and cooked rice samples in this study. Figure 2 shows a bar diagram of As concentrations in uncooked and cooked rice used by 14 households. Smith et al. (2006) reported that the mean As level in 46 cooked rice



samples was 358  $\mu\text{g}/\text{kg}$  (range: 46–1110  $\mu\text{g}/\text{kg}$ ). Based on their study in Bangladesh, Ohno et al. (2007) reported that the average As contents in uncooked and cooked rice were 340 and 460  $\mu\text{g}/\text{kg}$ , respectively, which were much higher than in our study. Recently Ohno et al. (2009) further reported that the average As contents in uncooked and cooked rice samples collected from Chapai Nawabganj district were 220 and 260  $\mu\text{g}/\text{kg}$ , respectively. Both these studies (Ohno et al. 2007, 2009) were conducted in highly As-contaminated Chapai Nawabganj district of Bangladesh and their studies revealed that cooked rice contained higher As concentrations than uncooked rice. They also suggested that As in cooking water affected the As concentrations of rice after cooking (Ohno et al. 2009). In a recent study conducted from three contrasting areas (high, moderate and low As-affected areas) of West Bengal in India, the mean As concentrations in cooked rice were 310, 220 and 100  $\mu\text{g}/\text{kg}$  for Bhawangola-I, Chakdha and Khejuri I blocks, respectively (Mondal et al. 2010). The study revealed that cooking raw rice with As-contaminated water increased the level of As in cooked rice in Bhawangola-I, Chakdha blocks whereas slightly decrease As concentration in cooked rice in Khejuri block. In our study, As concentrations in cooked rice were less in most of the cases than those of uncooked rice, except two samples. The probable reason was that all surveyed families used pond water low in As ( $<10 \mu\text{g}/\text{L}$ ) for cooking. It is most common practice in Bangladesh and West Bengal for people to wash rice with water till the washing water has become clear (5–6 times), water is discarded and then rice is boiled in excess water till cooked, finally discarding the excess water before consumption (Mondal et al. 2010). This

washing and cooking process with As-safe water, and discarding of the excess water, probably reduces As levels in cooked rice.

#### Arsenic Speciation in Uncooked Rice

The results of As speciation in uncooked rice samples are presented in Table 2. From the analytical results, it appears that inorganic As was the predominant species present in uncooked rice samples in this study. DMA(V) was also present in all uncooked rice samples. This study could not detect any MMA(V) in uncooked rice. The study shows that an average of 72.8% of total As was present as inorganic As in uncooked rice samples, with a range of 68 to 78% and on average 10.5% of total As was DMA(V). Williams et al. (2006) reported a range of 60–83% for inorganic As in Aman and boro rice samples from Bangladesh. Williams et al. (2006) reported that the relative amount of inorganic As in boro rice (mean 82%) was higher than in Aman rice (mean 66%) in Bangladesh. In a recent study conducted from West Bengal, it was reported that 90–100% of total recovered As was present in the inorganic form in Aman rice collected from As-contaminated areas (Pal et al. 2009). In this study, the percentage of inorganic As in uncooked rice (Aman) is comparable with Williams et al.'s (2006) study conducted from Bangladesh but less compared to West Bengal's Aman rice (Pal et al. 2009). Ohno et al. (2007) reported that the percentage of inorganic As was 87–107% for uncooked rice of Bangladesh although the rice grain type was not mentioned in that study.

**Table 2** Total and speciated As in uncooked rice samples

Family No.	Total As in rice ( $\mu\text{g}/\text{kg}$ )	Inorganic As in rice ( $\mu\text{g}/\text{kg}$ )	DMA(V) in rice ( $\mu\text{g}/\text{kg}$ )	Species sum ( $\mu\text{g}/\text{kg}$ )	% of inorganic As in rice <sup>a</sup>	% of recovery <sup>b</sup>
1	222	159	39	197	71	89
2	127	86	21	107	68	84
3	158	126	9	135	80	85
4	139	98	19	117	71	84
5	302	238	14	252	79	83
6	133	94	19	113	71	85
7	231	180	21	201	78	87
8	138	97	20	116	70	84
9	109	77	8	85	71	78
10	154	115	7	122	75	79
11	109	76	18	94	70	86
12	138	97	19	116	70	84
13	74	51	8	59	69	80
14	109	84	7	91	77	83
Average	153	113	16	129	73	84
Range	74–301	51–237	7–39	59–252	68–78	78–89

<sup>a</sup>% of recovery = (inorganic As/total As)

<sup>b</sup>% of recovery = (sum of As species/total As)

**Table 3** Drinking water and rice consumption rate for adults

Group	Gender	<i>n</i>	Average age (year)	Water intake (L)	Intake rate of rice per day (g)
Adults	Males	30	36 (12–80)	3.2 (2.0–4.0)	432 (200–600)
	Females	41	32 (12–70)	2.7 (1.5–3.0)	420 (200–600)

Arsenic speciation in cooked rice was not conducted in this study. Smith et al. (2006) reported an average of 87% inorganic As is present in cooked rice samples whereas Ohno et al. (2007) reported that the percentage of inorganic As was 97–102% in cooked rice, based on the study conducted from Bangladesh. In a recent study, Pal et al. (2009) determined whether the cooking procedures can change the form of As in cooked rice: they cooked two boro rice samples from contaminated areas by using the traditional rice cooking method (Bae et al. 2002; Sengupta et al. 2006) using distilled deionized water. The results showed that 95% of recovered As is present as inorganic form in the cooked boro rice (Pal et al. 2009). Thus the cooking procedures did not change the nature of As in cooked rice.

#### Water and Rice Consumption Rates

The water and rice consumption rates for adult males and females are presented in Table 3. The survey data showed that the average water intake rates were 3.2 and 2.7 L per day for adult males and females, respectively. Recently Mondal et al. (2010) estimated the mean daily water intake was  $3.1 \pm 1.0$  L/day for males and  $2.6 \pm 0.9$  L/day for females from West Bengal. The daily water intake rates for males and females in our study were almost similar to Mondal et al.'s (2010) study.

The study also revealed that the average rice consumption rates for adult males and females were 432 and 420 g (uncooked basis) per day. No significant difference was observed on rice intake between adult males and females. Ohno et al. (2007) reported that adult males and females consume 776 and 553 g of cooked rice daily, based on their study from Bangladesh.

#### Consumption of As from Drinking Water and Uncooked Rice by Adults

Based on average intake rates of drinking water and rice for adult males and females and As concentrations observed in rice and drinking water, we simply estimated the daily intake of As from these sources. The groundwater of Bangladesh contains mainly inorganic As (Samanta et al. 1999), so the intake of inorganic As from rice and drinking was also evaluated. The concentrations of inorganic As in uncooked rice samples along with total As content in uncooked rice are presented in Table 2. The concentrations of As in drinking

water are presented in Table 4. Table 4 also presents the daily total and inorganic As consumption from drinking water and rice for adult males and females. The estimated average As intake from rice was 66 and 64  $\mu\text{g}$  per day for adult males and females, respectively. The calculated average inorganic As intake from rice was 49 and 47  $\mu\text{g}$  per day for adult males and females, respectively. The calculated daily average As intake from drinking water was 1050 and 886  $\mu\text{g}$  for adult males and females, respectively. Taken together, adult males and females consumed 1116 and 950  $\mu\text{g}$ , respectively, of total As; 1099 and 933  $\mu\text{g}$ , respectively, of inorganic As daily from rice and drinking water in the villages studied here. In our previous study (Rahman et al. 2009) in Bangladesh, we observed that adult males and females consumed 888 and 706  $\mu\text{g}$  of total As from rice and drinking water daily, which was lower than found in this study. The daily As intake from rice and drinking water was 1186  $\mu\text{g}$  according to the study conducted by Smith et al. (2006), also in Bangladesh. The As intake rate from rice and drinking water for adults was also higher than that found by Roychowdhury et al. (2002) conducted in Jalangi (males—693  $\mu\text{g}$  and females—560  $\mu\text{g}$ ) and Domkal (males—574  $\mu\text{g}$  and females—474  $\mu\text{g}$ ) in the Murshidabad district in West Bengal of India.

Mondal et al. (2010) reported that the median contributions to total As exposure from drinking water were found to be 70% for Bhawangola-I block, 58% for Chakdha block and just only 2% for the unexposed area (Kejuri-I block). Rice contributes 12% for Bhawangola-I block, 34% for Chakdha block and 113% for uncontaminated area Kejuri-I block (Mondal et al. 2010). Thus, raw rice contributes almost all the total As exposure in the unexposed area (Mondal et al. 2010). Ohno et al. (2007) reported that the average contributions to the total As intake were 13% from drinking water and 56% from cooked rice. Ohno et al. (2007) indicated that many families had changed their drinking water sources to less contaminated ones and thus the average As contributions from drinking water was less compared to the cooked rice. In the present study, the average contributions to the total As intake were 94 and 93% from drinking water; 6 and 7% from uncooked rice, for adult males and females, respectively. The average As contribution from drinking water was significantly high in this study as almost all the surveyed families used highly As-contaminated water for drinking. The present study also estimated that 98% of the total As consumed by adult males and females was inorganic. Thus consuming a high amount of inorganic As

**Table 4** Daily total and inorganic As intake by adult males and females from drinking water and rice

Family No.	Total As in drinking water ( $\mu\text{g/L}$ )		Average water intake rate (L)		Intake of As from drinking water ( $\mu\text{g}$ ), A		Average rice intake rate (g)		Intake of As from rice ( $\mu\text{g}$ ), B		Intake of inorganic As from rice ( $\mu\text{g}$ ), C		Total intake of As from rice and drinking water ( $\mu\text{g}$ ) (A+B)		Total intake of inorganic As from rice and drinking water ( $\mu\text{g}$ ) (A+C)	
	M	F	M	F	M	F	M	F	M	F	M	F	M	F	M	F
1	772	3.2	2469	2083	432	420	96	93	69	67	2565	2176	2538	2150		
2	626	2.7	2002	1689	432	420	55	54	37	36	2057	1742	2039	1725		
3	14		45	38			68	67	55	53	114	105	100	91		
4	397		1269	1071			60	58	42	41	1329	1129	1312	1112		
5	551		1764	1488			130	127	103	100	1894	1615	1866	1588		
6	230		736	621			57	56	41	39	793	677	777	660		
7	426		1364	1151			100	97	78	75	1464	1248	1442	1227		
8	357		1142	964			60	58	42	41	1202	1022	1184	1005		
9	146		467	394			47	46	33	32	514	439	500	426		
10	185		591	498			67	65	50	48	657	563	641	547		
11	167		536	452			47	46	33	32	583	498	569	484		
12	325		1041	879			60	58	42	41	1101	937	1083	920		
13	233		745	628			32	31	22	22	777	659	767	650		
14	165		529	446			47	46	36	35	576	492	565	481		
Average	328		1050	886			66	64	49	47	1116	950	1099	933		
Range	14–772		45–2469	38–2083			32–130	31–127	22–103	22–100	114–2565	105–2176	100–2538	91–2150		

M/F—Adult males/Females



poses a real As-related health risk to the study population in this study.

## Conclusions

The study concludes that all surveyed households except one used As-contaminated water for drinking as set by the Bangladesh guideline value. Most of the family members of the surveyed families are well aware of As contamination in tubewell water and the consequent symptoms. Due to the social conventions, some villagers are reluctant to collect drinking water from neighbouring houses that have access to safe tubewells. Thus the study population in the study area were consuming a significant amount of As from drinking water during the study period. All surveyed households used As-safe pond water for cooking. So, cooking water did not contribute additional As to the daily As consumption. There was no significant difference between mean As concentrations in uncooked and cooked rice samples. Thus cooking water did not increase As levels in cooked rice in this study. The study shows that inorganic As was the main species present in uncooked Aman rice samples and a low amount of DMA(V) was also detected. The water consumption rates for adult males and females were in close agreement with the previous studies conducted from West Bengal (Mondal et al. 2010). Drinking water contributes to the majority of As consumed by adults, in comparison to that consumed in rice. In this study, about 98% of the total consumed As was inorganic which cause As-related health hazards to the study population. The study reveals that rice can also be a most important contributor to the exposure of inorganic As. The study did not consider the As intake from other food sources such as vegetables. It would therefore be useful to evaluate As intake from other food sources to assess their contribution to total intake. Besides As, concentrations of other heavy metals also need to be evaluated to determine the other possible health risk. More effort is required to reduce As consumption from drinking water and crops. Detailed study is required on the actual health risks from consuming As from all sources for this section of population. The government should educate rural people about the risk of long-term As exposure through drinking water and devise means for socially acceptable means of sharing safe tubewells.

**Acknowledgements** The authors thank the authority of CERAR, University of South Australia for the laboratory support. Financial support from CRC-CARE is greatly acknowledged. Md. Asaduzzaman is grateful to the Crawford Fund, Australia for the fellowship during his training in the CERAR. The authors are grateful to the authority of the Dhaka Community Hospital, Dhaka, Bangladesh for the laboratory support for processing the rice samples.

## References

- Abedin MJ, Cresser MS, Meharg AA et al (2002) Arsenic accumulation and metabolism in rice (*Oryza sativa* L.). *Environ Sci Technol* 36:962–968
- Al Rmali SW, Haris PI, Harrington CF, Ayub M (2005) A survey of arsenic in foodstuffs on sale in the United Kingdom and imported from Bangladesh. *Sci Total Environ* 337:23–30
- Alam MGM, Snow ET, Tanaka A (2003) Arsenic and heavy metal contamination of vegetables grown in Samta village, Bangladesh. *Sci Total Environ* 308:83–96
- Bae M, Watenab C, Inaoka P et al (2002) Arsenic in cooked rice in Bangladesh. *Lancet* 360:1839–1840
- BGS (British Geological Survey) (2001) Arsenic contamination of groundwater in Bangladesh. BGS technical report WC/00/19, British Geological Survey, Keyworth, UK
- Chakraborti D, Rahman MM, Das B et al (2010) Status of groundwater arsenic contamination in Bangladesh: A 14-year study report. *Water Res* 44:5789–5802
- Chen Z, Akter KF, Rahman MM et al (2008) The separation of arsenic species in soils and plant tissues by anion-exchange chromatography with inductively coupled mass spectrometry using various mobile phases. *Microchem J* 89:20–28
- Correll R, Huq SM, Smith E et al (2006) Dietary intake of arsenic from crops. In: Naidu R, Owens G, Smith E, Nadebaum P (eds) *Managing arsenic in the environment: from soil to human health*. CSIRO, Melbourne, pp 255–271
- D'Amato M, Forte G, Caroli S (2004) Identification and quantification of major species of arsenic in rice. *J AOAC Int* 87:238–243
- Das HK, Mitra AK, Sengupta PK et al (2004) Arsenic concentrations in rice, vegetables, and fish in Bangladesh: a preliminary study. *Environ Int* 30:383–387
- Dhar RK, Biswas BK, Samanta G et al (1997) Groundwater arsenic calamity in Bangladesh. *Curr Sci* 73:48–59
- Duxbury JM, Mayer AB, Lauren JG et al (2003) Food chain aspects of arsenic contamination in Bangladesh: effects on quality and productivity of rice. *J Environ Sci Health, Part A, Environ Sci Eng* 38:61–69
- Heitkemper DT, Vela NP, Stewart KR et al (2001) Determination of total and speciated arsenic in rice by ion chromatography and inductively coupled plasma mass spectrometry. *J Anal At Spectrom* 16:299–306
- IARC (International Agency for Research on Cancer) (2004) IARC monograph 84: some drinking water disinfectants and contaminants including arsenic. World Health Organization, International Agency for Research on Cancer (IARC), Lyon
- Khan NI, Bruce D, Naidu R et al (2009) Implementation of food frequency questionnaire for the assessment of total dietary arsenic intake in Bangladesh: Part b, preliminary findings. *Environ Geochem Health* 31:221–238
- Kile ML, Houseman EA, Breton CV et al (2007) Dietary arsenic exposure in Bangladesh. *Environ Health Perspect* 115:889–893
- Kohlmeyer U, Jantzen E, Kuballa J et al (2003) Benefits of high-resolution IC-ICP-MS for the routine analysis of inorganic and organic arsenic species in food products of marine and terrestrial origin. *Anal Bioanal Chem* 377:6–13
- Lu Y, Adomako EE, Solaiman ARM et al (2009) Baseline soil variation is a major factor in arsenic accumulation in Bengal delta paddy rice. *Environ Sci Technol* 43:1724–1729
- Meharg AA, Rahman MM (2003) Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption. *Environ Sci Technol* 37:229–234
- Mondal D, Banerjee M, Kundu M et al (2010) Comparison of drinking water, raw rice and cooking of rice as arsenic exposure routes in three contrasting areas of West Bengal, India. *Environ Geochem Health* 32:463–477

- National Research Council (NRC) (2001) Arsenic in drinking water—2001 update. National Academy Press, Washington
- Norton GJ, Islam MR, Deacon CM et al (2009) Identification of low inorganic and total grain arsenic rice cultivars from Bangladesh. *Environ Sci Technol* 43:6070–6075
- Ohno K, Yanase T, Matsuo Y et al (2007) Arsenic intake via water and food by a population living in an arsenic-affected area of Bangladesh. *Sci Total Environ* 381:68–76
- Ohno K, Matsuo Y, Kimura T et al (2009) Effect of rice-cooking water to the daily arsenic intake in Bangladesh: results of field surveys and rice-cooking experiments. *Water Sci Technol* 59:195–201
- Pal A, Chowdhury UK, Mondal D et al (2009) Arsenic burden from cooked rice in the populations of arsenic affected and non-affected areas and Kolkata city in West-Bengal, India. *Environ Sci Technol* 43:3349–3355
- Rahman MA, Hasegawa H, Rahman MA et al (2006) Influence of cooking method on arsenic retention in cooked rice related to dietary exposure. *Sci Total Environ* 370:51–60
- Rahman MM, Owens G, Naidu R (2009) Arsenic levels in rice grain and assessment of daily dietary intake of arsenic from rice in arsenic-contaminated regions of Bangladesh—Implication to groundwater irrigation. *Environ Geochem Health* 31:179–187
- Roychowdhury T, Uchino T, Tokunaga H et al (2002) Survey of arsenic in food composites from an arsenic-affected area of West Bengal, India. *Food Chem Toxicol* 40:1611–1621
- Samanta G, Roychowdhury T, Mandal BK et al (1999) Flow injection hydride generation atomic absorption spectrometry for determination of arsenic in water and biological samples from arsenic-affected districts of West Bengal, India and Bangladesh. *Microchem J* 62:174–191
- Sanz A, Munoz-Olivas R, Camara C et al (2007) Arsenic speciation in rice, straw, soil, hair and nails samples from the arsenic-affected areas of Middle and Lower Ganga plain. *J Environ Sci Health, Part A, Environ Sci Eng* 42:1695–1705
- Sengupta MK, Hossain MA, Mukherjee A et al (2006) Arsenic burden of cooked rice: traditional and modern methods. *Food Chem Toxicol* 44:1823–1829
- Smith NM, Lee R, Heitkemper DT et al (2006) Inorganic arsenic in cooked rice and vegetables from Bangladeshi households. *Sci Total Environ* 370:294–301
- WHO (World Health Organization) (2001) IPCS environmental health criteria 224 Arsenic and arsenic compounds. International Programme on Chemical Safety, World Health Organization, Geneva
- Williams PN, Price AH, Raab A et al (2005) Variation in arsenic speciation and concentration in paddy rice related to dietary exposure. *Environ Sci Technol* 39:5531–5540
- Williams PN, Islam MR, Adomako EE et al (2006) Increase in rice grain arsenic for regions of Bangladesh irrigating paddies with elevated arsenic in groundwaters. *Environ Sci Technol* 40:4903–4908