Article

Arsenic Burden from Cooked Rice in the Populations of Arsenic Affected and Nonaffected Areas and Kolkata City in West-Bengal, India

Arup Pal, Uttam Kumar Chowdhury, Debapriya Mondal, Bhaskar Das, Bishwajit Nayak, Arijit Ghosh, Sumana Maity, and Dipankar Chakraborti

Environ. Sci. Technol., **2009**, 43 (9), 3349-3355• DOI: 10.1021/es803414j • Publication Date (Web): 01 April 2009

Environmental Science & Technology is published by the American Chemical Society. 1155 Sixteenth Street N.W., Washington, DC 20036

Environ. Sci. Technol. **2009,** *43,* 3349–3355

Arsenic Burden from Cooked Rice in the Populations of Arsenic Affected and Nonaffected Areas and Kolkata City in West-Bengal, India

ARUP PAL, UTTAM KUMAR CHOWDHURY, DEBAPRIYA MONDAL, BHASKAR DAS, BISHWAJIT NAYAK, ARIJIT GHOSH, SUMANA MAITY, AND DIPANKAR CHAKRABORTI*

School of Environmental Studies, Jadavpur University, Kolkata 700032, India

Received December 2, 2008. Revised manuscript received February 27, 2009. Accepted March 5, 2009.

Arsenic contamination of rice irrigated with contaminated groundwater contributes to the additional arsenic burden of the population where rice is the staple food. In an arsenic contaminated area, an experimental field-based study done on nine fields elucidated significant positive correlation between arsenic in irrigation water and soil, irrigation water and rice, and also soil and rice both for Boro (groundwater) and Aman (rainwater) rice. Speciation studies showed that for both Boro (cooked) and Aman (raw) rice from contaminated area, 90% of total recovered arsenic was inorganic. In arsenic contaminated, uncontaminated villages, and Kolkata city, daily quantities of arsenic ingested by adult population from cooked rice diet are equivalent to 6.5, 1.8, and 2.3 L, respectively, of drinking water containing WHO guideline value. In contaminated area, daily intake only from cooked Boro rice for 34.6% of the samples exceeded the WHO recommended MTDI value (2 μg In−As day^{-1} kg^{-1} body wt), whereas daily intake from Aman rice was below MTDI value as was rice from uncontaminated areas and Kolkata city. Our study indicated that employing traditional rice cooking method as followed in Bengal delta and using water having arsenic $<3 \mu$g L^{-1} for cooking, actual exposure to arsenic from rice would be much less.

Introduction

Arsenic contamination of groundwater has emerged as a major public health problem in South East Asia, particularly in Bangladesh, all states in the Ganga flood plains in India (*1*), and many parts of China (*2*). In these countries arsenic contaminated groundwater is not only used for drinking but also widely used for irrigation of food crops, particularly paddy rice (*Oryza sativa L*), which is the staple food and provides more than 73% of the caloric intake of the population of the Bengal delta (*3*). Groundwater is extensively used to irrigate the rice crop in West-Bengal, India and Bangladesh, particularly during the dry season. Rice grown on arsenic contaminated soils may possess a high level of arsenic, which may potentially increase arsenic exposure to the population, especially where rice is the principal staple food (*4*).

In the context of the South East Asian groundwater arsenic contamination, it has been speculated and reported that rice could be a major source of arsenic for the exposed population (*3*). Arsenic contamination of rice in Bangladesh (*3*−*14*) and in West-Bengal, India (*15*−*21*) is well reported.

The arsenic content of cooked rice can be significantly different from that of raw rice depending on the arsenic content of the cooking water (*9*−*13, 15, 16, 20, 22*) and cooking method (*10, 16, 20*). Normally villagers of the Bengal delta cook rice following their traditional procedure (*12, 20*), which can lead to an appreciable amount of arsenic being leached out of rice when cooked in water having arsenic concentration $<3 \mu$g L^{-1} (*20*).

In West-Bengal, India 9 of the total 19 districts have groundwater arsenic levels exceeding 50 μg L^{-1}. The area and population of these nine districts are 38 865 sq. km and 50.2 million, respectively, while the area and population of West-Bengal are 88 000 sq. km and 80.2 million (*23*). Using underground water for agricultural irrigation since the 1980s, West-Bengal has become self-sufficient in food production (*24*). In West-Bengal underground water is used for Boro rice, and rainwater is usually used for Aman rice cultivations. We had already reported that many of the irrigation tube-wells contain arsenic in elevated levels (*21*). Therefore, in many agricultural fields in the Ganga-Meghna-Brahamaputra (GMB) plain, a huge quantity of arsenic enters the soil, and crops when arsenic contaminated tube-wells are used for irrigation. We made a thorough study in the North-24-Pargana district (*25*) on arsenic groundwater contamination and its health effects, and found that 20 of its 22 blocks (administrative subdivision after district) have arsenic above 50 μg L^{-1} in the water. Deganga block (our present study area) is one of the 20 known arsenic affected blocks of the North-24-Pargana district, where groundwater arsenic contamination is very high (*25*).

In the present study (a) Boro and Aman rice samples were collected and analyzed for arsenic, along with irrigation water and soil from nine experimental fields, which had been cultivated by villagers from the arsenic contaminated North-24-Pargana district. A similar study was made for samples from seven control fields of the uncontaminated areas of East-Medinipur district. (b) Arsenic exposure and uptake were estimated for households of the contaminated and the control areas using cooked rice prepared from samples obtained in the study and using traditional rice cooking procedures with water having arsenic $<3 \mu$g L^{-1}. (c) The arsenic burden on the population of Kolkata city, where the rice comes from both contaminated and uncontaminated districts of West-Bengal, is estimated and compared with contaminated and uncontaminated areas. Finally (d) the daily intake of arsenic from rice consumption using the traditional cooking procedures using water with arsenic concentration $<3 \mu$g L^{-1} has been estimated for the three different studied areas and compared to the World Health Organization maximum tolerable daily intake (WHO-MTDI) value of 2 μg day^{-1} kg^{-1} body wt.

Materials and Methods

Areas of Sample Collection. The samples from contaminated areas were collected from the Deganga block of North-24-Pargana district of West-Bengal. The arsenic contamination situation in Deganga block is shown in the Supporting Information Figure 1.

Boro and Aman rice samples were collected from nine paddy fields (size of each field approximately 100 × 100 m) of contaminated area from Kolsur village of Deganga block and seven fields of uncontaminated area from Silampur village of Contai-I block, East Medinipur district. We call those fields our experimental fields as owners of those fields

* Corresponding author phone: +91 33 2414 6233; fax: +91 33 2414 6266; e-mail: dcsoesju@vsnl.com.

10.1021/es803414j CCC: $40.75

Published on Web 04/01/2009

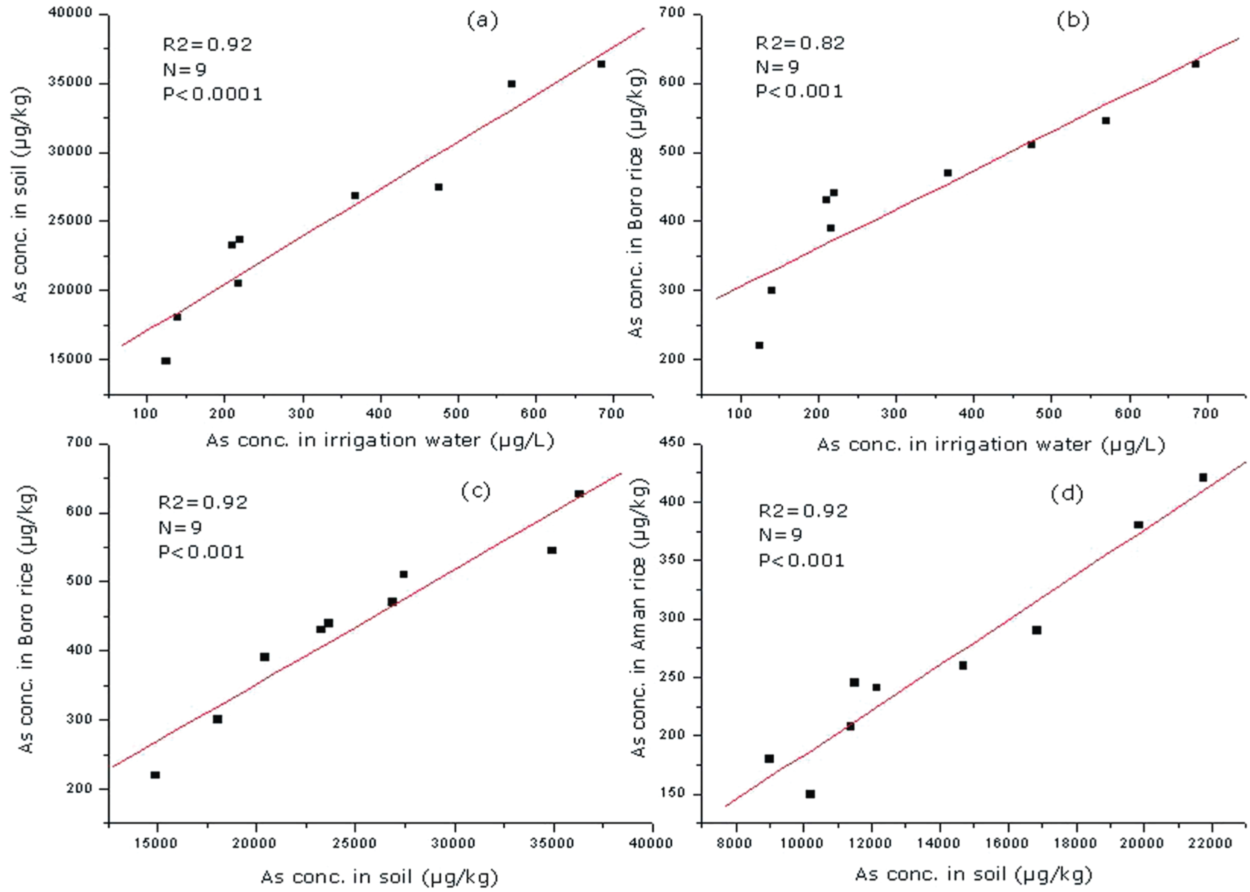

FIGURE 1. (a) Correlation between arsenic concentrations in irrigation water versus soil ($Y = 13639.76 + 34.26X$). (b) Correlation between arsenic concentrations in irrigation water versus Boro rice ($Y = 249.08 + 0.56X$). (c) Correlation between arsenic concentrations in soil versus Boro rice ($Y = 20.71 + 0.01X$). (d) Correlation between arsenic concentrations in soil versus Aman rice ($Y = -8.92 + 0.01X$).

allowed us to collect irrigation water, soil, and Boro and Aman rice from January to December 2003. We also collected some paired raw and cooked rice samples from the owners of those fields. Besides, we collected irrigation water samples from 596 shallow, small and big diameter irrigation tube-wells out of a total of 3200 in use for agricultural irrigation covering Deganga block. We collected 55 raw and cooked (paired) Boro and Aman rice samples along with cooking water ($N = 55$) from 55 different households of this area covering all the 14 Gram Panchayets (GPs) of Deganga block, having an area of 202 sq. km and population 275 350. Raw rice samples were also collected from 201 individual households. From the uncontaminated area we collected 27 raw and cooked (paired) Boro and Aman rice samples along with cooking water ($N = 27$) from 27 different households and 104 raw rice samples from 84 households.

We collected 148 raw and cooked (paired) rice samples and cooking waters ($N = 148$) from 70 out of the total 141 wards in Kolkata city having an area of 185 sq.km and population of 4.5 million. Rice samples were collected from households of Kolkata city in such a way that we could cover an appreciable portion of the city. During our rice collection from Kolkata, we could not obtain the information from hotels and households about whether the rice collected was Boro or Aman or whether the influx of rice had been from arsenic contaminated or uncontaminated districts.

Methods of Samples Collection and Rice Intake Habit Information. Surface soils (0−15 cm) were collected from experimental fields. Six samples were collected from each field and combined to give a composite sample. Samples were air-dried, powdered with quartz pestle and mortar, and 2 mm sieved. Methods of groundwater and rice sample collection and preservation have been discussed in our earlier publications (*1, 20*). During our sample collection we also collected information on the daily rice and water consumption by an adult person based on a questionnaire survey (*21*). From the arsenic contaminated and uncontaminated villages we collected samples; almost 100% of the population drinks water from hand tube-wells. We found that an adult on an average consumed 1.84 kg of cooked rice which is equivalent to 0.7 kg of raw rice and 3.5 L of direct water per day. In uncontaminated areas, the daily average of rice and water consumption rate of an adult person was almost the same as in contaminated areas. The daily cooked rice consumption rate of an adult person in Kolkata averaged 0.84 kg. In rural West-Bengal (in both contaminated and uncontaminated areas) almost all families eat cooked rice three times in a day and still use traditional rice cooking methods. In the traditional procedure raw rice is washed until the washings become clear, washings are discarded and then the rice is boiled in excess water until cooked, finally discarding the remaining water (*12, 20*). In Kolkata city many families, due to urban life style, eat rice only one or two times in a day, and do consume bread and likely use a pressure cooker while cooking rice. Hotels use the traditional cooking procedure.

Instrumentation and Method of Analysis. Flow injection hydride generation atomic absorption spectrometry (FI-HG-AAS) was used for total arsenic analysis. Details of the instrumentation, chemicals used, and procedures for rice and soil digestions have already been described in our earlier publications (*1, 20, 26*). The speciation was done for four raw Aman rice samples and two cooked Boro rice samples from the contaminated area. Rice was cooked using the traditional procedure as described earlier (*12, 20*). Arsenic speciation of the raw rice was done by IC-ICP-MS at the Forensic Chemistry Center, U.S. Food and Drug Administration (US-FDA) based on the protocol of Heitkemper et al. (*27*). Speciation of arsenic in cooked rice was done by LC-ICP-MS using the enzymatic ultrasonic probe for rice treatment at Facultad de Ciencias Químicas, Universidad Complutense de Madrid (UCM), in Ciudad University, Spain. This procedure was described by Sanz et al. (*28*). Using the IC-ICP-MS method As(III) and As(V) were determined together as total inorganic arsenic, whereas for the LC-ICP-MS procedure using the enzymatic ultrasonic probe for rice treatment the inorganic arsenic species were determined separately for cooked rice. The accuracy of our analytical methods was verified by analyzing the Standard Reference Materials (SRM) and by an interlaboratory comparison of rice analysis between our laboratory and that of Ciudad University, Spain.

Results and Discussion

Quality Control Analysis. Results of SRM water, soil, and rice analyses were found to be in good agreement with the certified values [water SRM (quality control sample for trace metal analysis) from the U.S. Environmental Protection Agency's Environmental Monitoring Laboratory, Cincinnati, Ohio (certified value 17.6 ± 2.21 μg L^{-1}, observed value 16 ± 3.5 μg L^{-1} (N = 5); soil SRM No. 2709 from National Institute of Standards and Technology, U.S. (certified value 17 700 ± 800 μg kg^{-1}, observed value 17 350 ± 520 μg kg^{-1}) (N = 5); rice SRM No. 1586a from National Institute of Standards and Technology, U.S. (certified value 290 ± 3 μg kg^{-1}, observed value 273 ± 2 μg kg^{-1}) (N = 5)]. Performing the interlaboratory comparison by comparing our results with results obtained from Ciudad University, Spain, a good agreement (94%) was observed (for the same sample analyzed by Ciudad University, Spain to be 290 ± 10 μg kg^{-1}; our corresponding result was 273 ± 2 μg kg^{-1}).

Experimental Field Samples. The arsenic concentration in irrigation groundwater during Boro rice cultivation in nine experimental fields was found to be quite high (334 ± 201 μg L^{-1}) (Supporting Information Table 1). The arsenic concentration in soil for Boro and Aman rice cultivations was 25 100 ± 7174 μg kg^{-1} and 14 155 ± 4451 μg kg^{-1} (N = 9), respectively, and the arsenic concentration in Boro and Aman rice from these fields was 439 ± 124 μg kg^{-1} and 265 ± 89 μg kg^{-1} (N = 9), respectively (Supporting Information Table 1). High soil arsenic concentration during Aman cultivation could be due to accumulation of arsenic in the same field from previous Boro cultivation using groundwater for irrigation. A similar study has been done by van Geen et al. (*14*) on Bangladeshi Boro rice samples.

It is also evident that the arsenic concentration in the soil in each field decreased by an appreciable amount during Aman rice (rainwater) compared to Boro rice (groundwater irrigated) cultivation (Supporting Information Table 1). We have two explanations for such decrease: (i) The arsenic in the soil was washed away by the rain (rain starts in June). (ii) The other reason could be biomethylation of arsenic from soil to air. The two hypotheses were further supported by our field experiments. In 2000, as a result of the flood, a good portion of North-24-Pargana, including our experimental fields, were submerged under water for about 5 months. The average arsenic in the nine fields before the flood was 26 882 ± 8554 μg kg^{-1}, and the average arsenic in irrigation tube-wells was 352 ± 218 μg L^{-1}. After the flood the arsenic in the soil declined to 13 632 ± 4660 μg kg^{-1} but the arsenic concentration in irrigation tube-wells remained almost the same. We found that arsenic input to the soil during irrigation is counteracted by arsenic leaching out during the monsoons leading to strong temporal changes of soil arsenic contents. Two previous studies supported our findings (*29, 30*). We have already reported that if arsenic rich sludge is mixed with cow dung, 76% of the sludge arsenic is lost (*31*). After harvesting of Boro rice in fields of West-Bengal, many cows graze the fields to eat the left over straw in the fields. This could facilitate biomethylation. To confirm this, methylated species of arsenic have to be identified by further experiments. Our present findings do not follow the prediction that soil arsenic levels increases at a rate of 1000 μg kg^{-1} yr^{-1} when the irrigation water arsenic concentration is 100 μg L^{-1} (*3*). However, to reliably quantify these long-term trends, detailed spatially and temporally, resolved data on the arsenic content in a well-defined area is needed over an extended monitoring period (*29*).

The regression line in Figure 1a indicates that during the Boro rice cultivation season, an increase of 100 μg L^{-1} of arsenic in irrigation water may cause an increase of 3426 μg kg^{-1} arsenic in the soil. We also found that arsenic concentrations in Boro rice increases with increases in irrigation water arsenic concentrations (Figure 1b) and soil arsenic (Figure 1c). It appears that a 100 unit change in soil arsenic concentrations can change arsenic concentrations in Boro rice by one unit. Although rainwater is used during Aman rice cultivation in the nine experimental fields, arsenic in the soil is less compared to Boro. There is a good correlation between arsenic in soil and in Aman rice (Figure 1d).

For the experimental fields (N = 7) of the uncontaminated area, all irrigation groundwater and rainwater samples were found to have arsenic concentrations <3 μg L^{-1}, and the soil arsenic concentration was found to be in the range of 1264 − 2264 μg kg^{-1} (mean 1818 μg kg^{-1}). Arsenic concentrations in Boro (N = 7) and Aman (N = 7) rice were in the range of 18−46 μg kg^{-1} (mean 31 μg kg^{-1}) and 14−33 μg kg^{-1} (mean 25 μg kg^{-1}), respectively.

Arsenic Concentrations in the Samples Collected from the Contaminated, Uncontaminated Area and Kolkata City. The arsenic content of the paired raw and cooked rice samples (N = 55) collected from the households of the contaminated area was found to be in the range of 138−482 μg kg^{-1} (mean 249 μg kg^{-1}) and 33−138 μg kg^{-1} (mean 65 μg kg^{-1}), respectively, for raw and cooked Boro rice samples and between 28 and 163 μg kg^{-1} (mean 82 μg kg^{-1}) and 8 and 57 μg kg^{-1} (mean 23 μg kg^{-1}), respectively, in raw and cooked Aman rice samples (Table 1). All the cooking water samples were found to have arsenic concentration <3 μg L^{-1}. The comparison between cooked Aman and cooked Boro rice showed that a mean arsenic content for cooked Boro rice to be 2.8 times higher than that for cooked Aman rice. For raw Boro and Aman rice samples this ratio was 3 times. This indicates that the arsenic concentration differences between raw Boro and Aman rice and cooked Boro and Aman rice samples are similar.

For the paired raw and cooked rice samples (N = 27) collected from the households of the uncontaminated area it was found arsenic concentration in raw and cooked Boro rice ranged between 31 and 84 μg kg^{-1} (mean 53 μg kg^{-1}) and 7 and 25 μg kg^{-1} (mean 13 μg kg^{-1}), respectively, whereas in raw and cooked Aman rice, the arsenic values ranged between 21 and 49 μg kg^{-1} (mean 36 μg kg^{-1}) and 7 and 17 μg kg^{-1} (mean 12 μg kg^{-1}), respectively, (Table 1). It was also observed that arsenic concentration in cooking water was <3 μg L^{-1}.

For paired raw and cooked rice samples (N = 148) collected from Kolkata city it was found that arsenic concentration in raw and cooked rice ranged between 22 and 395 μg kg^{-1} (mean 137 μg kg^{-1}) and 6 and 96 μg kg^{-1} (mean 34 μg kg^{-1}), respectively, (Table 1).

From the plots of arsenic concentration in paired raw versus cooked Boro rice (Figure 2a) and that in paired Aman rice samples from contaminated area (Figure 2b), it is observed that during cooking 64−84% of arsenic was removed from the raw rice samples. We have conducted

TABLE 1. Distribution of As Concentration (μg kg^{-1}) in Raw and Cooked (Boro and Aman) Rice Collected from Households of Contaminated, Uncontaminated Area and Kolkata City

areas	rice type	no. of samples	min. as conc.	max. as conc.	mean as conc.	median as conc.	distribution of as conc. in rice: ≤3–100	101–200	201–300	301–400	>400
contaminated	Boro (raw)	55	138	482	249	174		29 (52.7%)	11 (20%)	11 (20%)	4 (7.3%)
	Boro (cooked)	55	33	138	65	51	44 (80%)	11 (20%)			
	Aman (raw)	55	28	163	82	83	41 (74.5%)	14 (25.5%)			
	Aman (cooked)	55	8	57	23	18	55 (100%)				
uncontaminated	Boro (raw)	27	31	84	53	51	27 (100%)				
	Boro (cooked)	27	7	25	13	13	27 (100%)				
	Aman (raw)	27	21	49	36	36	27 (100%)				
	Aman (cooked)	27	7	17	12	13	27 (100%)				
Kolkata	raw	148	22	395	137	134	53 (35.8%)	70 (47.3%)	21 (14.2%)	4 (2.7%)	
	cooked	148	6	96	34	34	148 (100%)				

experiments to determine the difference between the weight of raw and cooked rice on a series of paired samples (N = 10) following the traditional procedure and using water arsenic concentration of <3 μg L^{-1}. We found that on an average, the weight of cooked rice was 2.6 ± 0.15 times higher than the weight of raw rice. Thus after mass balance we found that the removal of arsenic was 8–58% for both Boro and Aman rice samples.

A strong correlation between arsenic in raw and cooked rice for both Boro and Aman (Figure 2a and b) indicates that when water with arsenic concentrations <3 μg L^{-1} is used for cooking, the arsenic content of cooked rice depends on the arsenic content of the raw rice. At the same, the variance in arsenic concentration of raw rice compared to cooked rice for Boro rice samples (Figure 2a) indicates that arsenic removal from rice by cooking with arsenic safe water depends largely on the cooking method. We have observed that the percentage removal depends on the rice washing procedure, whether excess water is discarded after cooking, and also the rice variety. This is in agreement with our previous study (*20*) where we reported that up to 57% of the arsenic could be removed from arsenic contaminated rice using traditional rice cooking methods and using water containing arsenic <3 μg L^{-1}.

The arsenic content of the Boro (N = 201) and Aman (N = 77) raw rice samples colleted from individual households (N = 201) in the arsenic contaminated Deganga block was found to be in the range of 138–527 μg kg^{-1} (mean 270 μg kg^{-1}) and 13–163 μg kg^{-1} (mean 80 μg kg^{-1}) respectively (Supporting Information Table 2). The arsenic concentration in raw Boro rice from uncontaminated area was in the range of 18–86 μg kg^{-1} (mean 51 μg kg^{-1}) (N = 56) and for raw Aman rice 14–49 μg kg^{-1} (mean 32 μg kg^{-1}) (N = 48) (Supporting Information Table 2).

The arsenic concentration range in 596 irrigation shallow tube-wells from contaminated area was <10–840 μg L^{-1} with the average arsenic concentration at 71 μg L^{-1} (Supporting Information Table 3). On the basis of total groundwater withdrawal during the Rabi (cereals) crop production and Boro rice cultivation from November to June (2003–2004), we calculated that approximately 6400 kg of arsenic had inundated the agricultural lands of Deganga block alone. On this basis we feel that thousands of tons of arsenic are entering on the irrigation lands of arsenic contaminated areas in the GMB plain.

Comparisons of Rice Arsenic Concentration between Contaminated, Uncontaminated Areas and in Kolkata City. The mean arsenic concentrations in cooked Boro (65 μg kg^{-1})

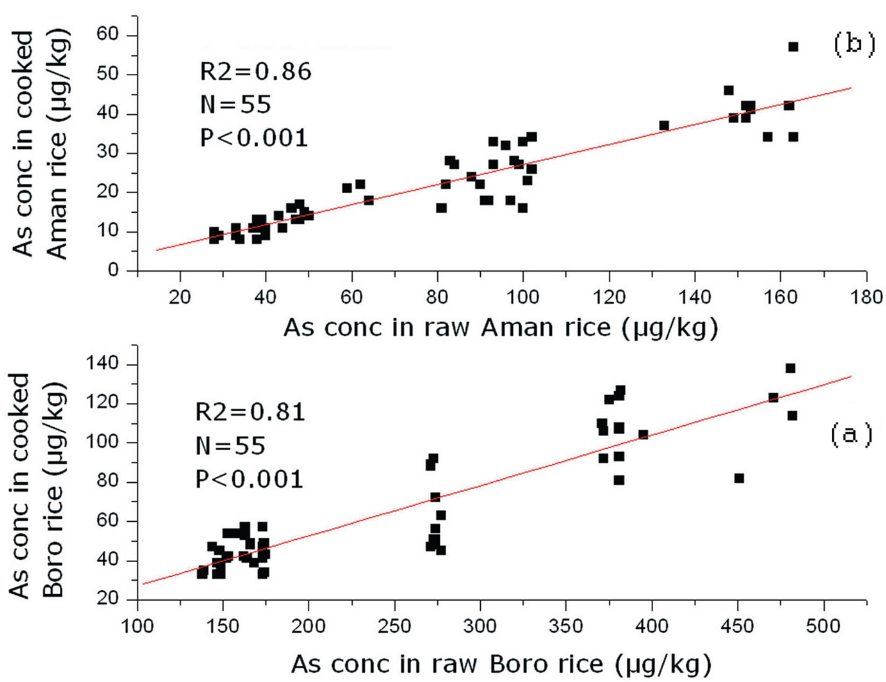

FIGURE 2. (a) Correlation between arsenic concentrations in raw and cooked Boro rice collected from contaminated areas (Y = 1.39 + 0.26X). (b) Correlation between arsenic concentrations in raw and cooked Aman rice collected from contaminated areas (Y = 1.56 + 0.26X).

and Aman (23 μg kg^{-1}) rice collected from households in contaminated areas were 5 and 2 times higher than from Boro (13 μg kg^{-1})and Aman (12 μg kg^{-1}) rice collected from households of uncontaminated areas (Table 1). The same comparison also holds for the raw rice samples (Supporting Information Table 2). For both raw Boro and Aman rice samples collected from contaminated areas, the arsenic concentration was significantly higher ($p < 0.001$) than those in uncontaminated areas and in Kolkata city. The mean arsenic concentration of cooked rice samples collected from Kolkata (34 μg kg^{-1}) was 1.29 times lower than the Boro and Aman combined cooked rice collected from contaminated areas, but 2.8 times higher than cooked rice collected from uncontaminated areas. Similar findings for the corresponding raw rice samples show that rice collected from Kolkata (131 μg kg^{-1}) was 1.6 times lower than contaminated and 3.1 times higher than uncontaminated area. This result indicates that Kolkata probably receives rice from both contaminated and uncontaminated areas.

Comparison of Aman and Boro rice collected from the nine experimental fields of contaminated areas show a mean arsenic content for Boro rice to be 1.66 times higher than that for Aman rice which is in agreement with two previous studies (*5, 8*). In uncontaminated field areas, Boro rice arsenic was 1.2 times higher than that of Aman rice. For the household survey from contaminated areas the mean arsenic content for Boro rice was found to be 3.4 times higher than that of Aman rice, whereas in the uncontaminated area, the ratio was 1.6. The greater difference between Boro and Aman rice for the households in contaminated areas indicates that there is either extensive use of contaminated groundwater for irrigation during Boro rice cultivation or lower arsenic concentration of soil for the Aman rice cultivations with respect to the field soil samples. It could also reflect the significance of rice genotype in accumulating arsenic.

Arsenic Speciation in Raw and Cooked Rice. In the raw rice speciation study with Aman rice from contaminated areas, we found that 90−100% of total recovered arsenic was present in the inorganic form (As^{III} + As^{V}) (Supporting Information Table 4). In Boro raw rice from contaminated area Sanz et al. (*28*) reported 95% of total recovered arsenic was inorganic with DMA, MMA and AsB being the minor components.

The findings of speciation of rice with respect to inorganic arsenic versus total arsenic content for different countries by previous studies (*7, 15, 17, 21, 28, 32*) are tabulated in Supporting Information Table 5. Here we have considered the inorganic arsenic content of rice to be 90% estimated as the average for all those studies having a recovery greater than 70%, though species recovery efficiency ranges between 75 and 106% (Supporting Information Table 6).

To determine whether cooking procedures can change the form of arsenic in cooked rice, we cooked two Boro rice samples from contaminated areas by the traditional rice cooking method (*12, 20*) using distilled deionized water. Sanz et al. (*28*) reported the raw rice speciation study of these two samples in their study. From the results, it appears that for cooked Boro rice 95% of recovered arsenic is present in the inorganic form (As^{III} and As^{V}). Also inorganic arsenic (As^{III} and As^{V}) was the predominant species in the discarded water with DMA, MMA and AsB as the minor components (Supporting Information Table 7). Previously Smith et al. (*13*) reported that in Bangladeshi cooked rice ($N = 46$) 87% of total arsenic was present in inorganic form. However, Mihucz et al. (*33*) supported our finding that As^{III} could be removed most effectively if rice was washed and cooked in abundant water.

Inorganic Arsenic Body Burden from Rice Intake for the Adult Population from the Study Areas Compared to WHO MTDI (Inorganic Arsenic). The WHO recommended MTDI for inorganic arsenic is 2 μg day^{-1} kg^{-1} body wt (*34*). In our present study we found that in contaminated and uncontaminated areas adults had an average body weight of 53 kg. Considering the bioavailability of arsenic in cooked rice to be 90% (*35*) and the inorganic arsenic content of cooked rice to be 90% and based on the arsenic content of the cooked Boro and Aman rice samples collected from contaminated areas (Table 1) we observed that 34.6% of cooked Boro rice samples contributed more than the recommended MTDI value (Table 2). For the Aman rice samples, however, the estimated daily intake was below MTDI value. Considering daily intake of arsenic from raw rice, we found that in raw Boro rice, the daily intake from 47.3% of the samples contributed more than the WHO recommended MTDI value, but for the Aman rice samples, the estimated daily intake was below the MTDI value. In uncontaminated areas as well as in Kolkata city the daily intake from both raw and cooked rice samples contributed less than the MTDI value (Table 2).

Because raw rice is not eaten, recently researchers used cooked rice to calculate the dietary exposure from rice, but in their work cooking water contained arsenic (*11, 13, 15, 16*). To estimate arsenic burdens from rice, we need to consider cooked rice and take into account the fact that the actual arsenic exposure from cooked rice to Bengal delta population would be much less if the traditional cooking method and water with arsenic concentrations <3 μg L^{-1} was used for cooking. This is supported by this study and our previous study (*20*). From Table 2, based on the calculation of the percentage of the population exposed to different MTDI ranges, especially for contaminated areas, it is clear that considering the arsenic concentration of raw rice rather than cooked rice will significantly overestimate the exposure.

Arsenic Exposure through Cooked Rice Compared to That from Water. As the actual rice arsenic exposure comes from cooked rice, using data from Table 1, we found that in contaminated areas, uncontaminated areas, and Kolkata city, an adult consumes 65 μg (Boro 97 μg and Aman 34 μg), 18 μg (Boro 19 μg and Aman 18 μg), and 23 μg inorganic arsenic per day from cooked rice on average. These values are equivalent to 6.5, 1.8, and 2.3 L (WHO guideline value is 10 μg L^{-1} on the basis of 2 L day^{-1}) and 1.3, 0.36, and 0.46 L [according to the interim Indian Standard 50 μg L^{-1} (*36*)] of water per day in contaminated, uncontaminated and Kolkata city respectively.

Arsenic exposure through drinking water in arsenic contaminated villages is considered to be the main source of arsenic exposure. In recent years, due to the wide awarenesss of arsenic contamination in contaminated villages, particularly where patients are suffering from the effects of arsenic toxicity, villagers usually do not use arsenic contaminated water for drinking and cooking and this is supported by the fact that in this study we found that all families were using water containing arsenic <3 μg L^{-1} for drinking and cooking purposes. This is only true for limited arsenic contaminated areas in GMB plain.

It is evident from this study that employing traditional rice cooking method as followed in Bengal delta and using water containing arsenic <3 μg L^{-1} for cooking, actual exposure to arsenic from rice would be much less than previously predicted (*8, 16*). Still, Boro rice could be a major source of arsenic exposure for arsenic contaminated rural areas of West-Bengal.

In the world's groundwater arsenic contamination scenario, Asian countries are worst affected. Rice is the staple food for Asian countries and with the present water scarcity and population increase, groundwater use for agriculture and domestic purposes should increase rather than decrease. Automatic exclusion of these fertile rice growing areas would have disastrous consequences. Solutions to favor a decrease

TABLE 2. Expected Number of Exposed Adult Population at Different MTDI (μg In−As Day^{-1} kg^{-1} Body Wt) Ranges from Raw and Cooked Rice (Both Boro and Aman) Collected from Households of Contaminated, Uncontaminated Area and Kolkata City

area	total adult population[a] (*23*)	rice type	raw/cooked rice	expected number of exposed adult population at different MTDI range from raw and cooked rice (percentage of samples) ≤1.00	1.01−2.00	2.01−3.00	3.01−4.00	4.01−5.00	>5.00
contaminated	165210	Boro (*N* = 55)	raw		87066 (52.7%)	33042 (20%)	9252 (5.6%)	26929 (16.3%)	8921 (5.4%)
			cooked	20982 (12.7%)	87066 (52.7%)	30068 (18.2%)	27094 (16.4%)		
		Aman (*N* = 55)	raw	105074 (63.6%)	60136 (36.4%)				
			cooked	138116 (83.6%)	27094 (16.4%)				
uncontaminated	91024	Boro (*N* = 27)	raw	91024 (100%)					
			cooked	91024 (100%)					
		Aman (*N* = 27)	raw	91024 (100%)					
			cooked	91024 (100%)					
Kolkata (70 wards out of 141 wards)	2760000		raw (*n* = 148)	2312880 (83.8%)	447120 (16.2%)				
			cooked (*n* = 148)	2760000 (100%)					

[a] Considering 60% of the total population is adult i.e. above 18 years age (*37*).

in arsenic contamination of the rice crop include continued education in the need for traditional cooking methods in safe water; proper watershed management to maximize the use of surface and rainwater; drip water irrigation to conserve water; use of rice types requiring minimal water; and ultimately, development of a specific rice genotype that would not accumulate arsenic.

Acknowledgments

Thanks to Hamdard National Foundation, India for the award to Dipankar Chakraborti INR 5 million to continue arsenic work. We thank Dr. Douglas T. Heitkemper, USFDA, Forensic Chemistry Centre, USA and Dr. Riansares Muñoz Olivas, Facultad de Ciencias Quimicus, Universidad Complutense de Madrid (UCM), Ciudad Universitaria, Madrid, Spain for their help in the speciation study. Arup Pal is also thankful to the Indian Council of Medical Research, New Delhi, India, for awarding the ICMR-SRF.

Supporting Information Available

Additional tables and figure. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

(1) Chakraborti, D.; Sengupta, M. K.; Rahman, M. M.; Ahamed, S.; Chowdhury, U. K.; Hossain, M. A.; Mukherjee, S. C.; Pati, S.; Saha, K. C.; Dutta, R. N.; Zaman, Q. Q. Groundwater arsenic contamination and its health effects in the Ganga-Meghna-Brahmaputra plain. *J. Environ. Monitor.* **2004**, *6*, 72N–83N.

(2) Sun, G. Arsenic contamination and arsenicosis in China. *Toxicol. Appl. Pharmacol.* **2004**, *198*, 268–271.

(3) Meharg, A. A.; Rahman, M. Arsenic Contamination of Bangladesh Paddy Field Soils: Implications for Rice Contribution to Arsenic Consumption. *Environ. Sci. Technol.* **2003**, *37*, 229–234.

(4) Abedin, M. J.; Cresser, M. S.; Meharg, A. A.; Feldmann, J.; Cotter-Howells, J. Arsenic accumulation and metabolism in rice (*Oryza sativa* L.). *Environ. Sci. Technol.* **2002**, *36*, 962–968.

(5) Duxbury, J. M.; Mayer, A. B.; Lauren, J. G.; Hassan, N. Food chain aspects of arsenic contamination in Bangladesh: Effects on quality and productivity of rice. *J. Environ. Sci. Health, Part A* **2003**, *38*, 61–69.

(6) Das, H. K.; Mitra, A. K.; Sengupta, P. K.; Hossain, A.; Islam, F.; Rabbani, G. H. Arsenic concentrations in rice, vegetables, and fish in Bangladesh: A preliminary study. *Environ. Int.* **2004**, *30*, 383–387.

(7) Williams, P. N.; Price, A. H.; Raab, A.; Hossain, S. A.; Feldmann, J.; Meharg, A. A. Variation in arsenic speciation and concentration in paddy rice related to dietary exposure. *Environ. Sci. Technol.* **2005**, *39*, 5531–5540.

(8) Williams, P. N.; Islam, M. R.; Adomako, E. E.; Raab, A.; Hossain, S. A.; Zhu, Y. G.; Feldmann, J.; Meharg, A. A. Increase in rice grain arsenic for regions of bangladesh irrigating paddies with elevated arsenic in groundwater. *Environ. Sci. Technol.* **2006**, *40*, 4903–4908.

(9) Huq, S. M. I.; Joardar, J. C.; Parvin, S.; Correll, R.; Naidu, R. Arsenic contamination in food-chain: transfer of arsenic into food materials through groundwater irrigation. *J. Health Popul. Nutr.* **2006**, *24*, 305–316.

(10) Rahman, M. A.; Hasegawa, H.; Rahman, M. A.; Rahman, M. M.; Miah, M. A. M. Influence of cooking method on arsenic retention in cooked rice related to dietary exposure. *Sci. Total Environ.* **2006**, *370* (1), 51–60.

(11) Ohno, K.; Yanase, T.; Matsuo, Y.; Kimura, T.; Rahman, M. H.; Magara, Y.; Matsui, Y. Arsenic intake via water and food by a population living in an arsenic affected areas of Bangladesh. *Sci. Total Environ.* **2007**, *381*, 68–76.

(12) Bae, M.; Watenab, C.; Inaoka, P.; Sekiyama, M.; Sudo, N.; Bokul, M. H.; Ohtsuka, R. Arsenic in cooked rice in Bangladesh. *Lancet* **2002**, *360*, 1839–1840.

(13) Smith, N. M.; Lee, R.; Heitkemper, D. T.; Cafferky, K. D.; Haque, A.; Henderson, A. K. Inorganic arsenic in cooked rice and vegetables from Bangladeshi households. *Sci. Total Environ.* **2006**, *370*, 294–301.

(14) van Geen, A.; Zheng, Y.; Cheng, Z.; He, Y.; Dhar, R. K.; Garnier, J. M.; Rose, J.; Seddique, A.; Hoque, M. A.; Ahmed, K. M. Impact of irrigating rice paddies with groundwater containing arsenic in Bangladesh. *Sci. Total Environ.* **2006**, *367*, 769–777.

(15) Mondal, D.; Poyla, D. A. Rice is a major exposure route for arsenic in Chakdaha block, Nadia district, West Bengal, India: A probabilistic risk assessment. *Appl. Geochem.* **2008**, *23*, 2986–2997.

(16) Singes, A.; Mitra, K.; Burlo, F.; Carbonell-Barrachina, A. A. Contribution of water and cooked rice to an estimation of the dietary intake of inorganic arsenic in a rural village of West Bengal, India. *Food Addit. Contam., Part A.* **2008**, *25* (1), 41–50.

(17) Mandal, B. K.; Suzuki, K. T.; Anzai, K. Impact of arsenic in foodstuff on the people living in the arsenic affected areas of West Bengal, India. *Environ. Sci Health, Part A.* **2007**, *42*, 1741–1752.

(18) Roychowdhury, T.; Uchino, T.; Tokunaga, H.; Ando, M. Survey of food composites from an arsenic-affected area of West Bengal, India. *J. Food Chem. Toxol.* **2002**, *40*, 1611–1621.

(19) Roychowdhury, T.; Tokunaga, H.; Ando, M. Survey of arsenic and other heavy metals in food composites and drinking water and estimation of dietary intake by the villagers from an arsenic affected area of West Bengal, India. *Sci. Total Environ.* **2003**, *308*, 15–35.

(20) Sengupta, M. K.; Hossain, M. A.; Mukherjee, A.; Ahamed, S.; Das, B.; Nayak, B.; Pal, A.; Chakraborti, D. Arsenic burden of cooked rice: traditional and modern methods. *J. Food Chem. Toxol.* **2006**, *44*, 1823–1829.

(21) Chowdhury, U. K.; Rahman, M. M.; Mandal, B. K.; Paul, K.; Lodh, D.; Biswas, B. K.; Basu, G. K.; Chanda, C. R.; Saha, K. C.; Mukherjee, S. C.; Roy, S.; Das, R.; Kaies, I.; Barua, A. K.; Palit, S. K.; Quamruzzaman, Q.; Charaborti, D. Groundwater arsenic contamination and human sufferings in West Bengal, India and Bangladesh. *Environ. Sci.* **2001**, *83*, 93–415.

(22) Torres-Escribano, S.; Leal, M.; Velez, D.; Montoro, R. Total and Inorganic arsenic concentrations in rice sold in spain, effect of cooking, and risk assessment. *Environ. Sci. Technol.* **2008**, *42*, 3867–3872.

(23) *Census of India*; Office of the Director of Census Operation: West Bengal, India, 2001.

(24) *West Bengal Human Development Report*; Development and Planning Department, Government of West Bengal: West Bengal, 2004.

(25) Rahman, M. M.; Mandal, B. K.; Chowdhury, T. R.; Sengupta, M. K.; Chowdhury, U. K.; Lodh, D.; Chanda, C. R.; Basu, G. K.; Mukherjee, S. C.; Saha, K. C.; Chakraborti, D. Arsenic groundwater contamination and sufferings of people in North 24-Parganas, one of the nine arsenic affected districts of West Bengal, India. *J. Environ. Sci. Health, Part A:* **2003**, *28*, 25–59.

(26) Chatterjee, A.; Das, D.; Chakraborti, D. A study of ground water contamination by arsenic in the residential area of Behala, Calcutta due to industrial pollution. *Environ. Pollut.* **1993**, *80*, 57–65.

(27) Heitkemper, D. T.; Vela, N. P.; Stewart, K. R.; Westphal, C. S. Determination of total and speciated arsenic in rice by ion chromatography and inductively coupled plasma mass spectrometry. *J. Anal. Atom. Spectrom.* **2001**, *16*, 299–306.

(28) Sanz, E.; Munoz-Olivas, R.; Camara, C.; Sengupta, M. K.; Ahamed, S. Arsenic speciation in rice, straw, soil, hair and nail samples from the arsenic affected areas of middle and lower Ganga plain. *Environ. Sci. Health, A* **2007**, *42*, 1695–1705.

(29) Dittmar, J.; Voegelin, A.; Roberts, L. C.; Hug, S. J.; Saha, G. C.; Ali, M. A.; Badruzzaman, A. B. M.; Kretzschmar, R. Spatial distribution and temporal variability of arsenic in irrigated rice fields in Bangladesh. 2. Paddy soil. *Environ. Sci. Technol.* **2007**, *41*, 5967–5972.

(30) Saha, G. C.; Ali, M. A. Dynamics of arsenic in agricultural soils irrigating with arsenic contaminated groundwater in Bangladesh. *Sci. Total Environ.* **2007**, *379*, 180–189.

(31) Das, D.; Chatterjee, A.; Samanta, G.; Chowdhury T. R.; Mandal, B. K.; Dhar, R. K.; Chanda, C. R.; Lodh, D.; Chowdhury, P. P.; Basu, G.; Biswas, B. K.; Chowdhury, U. K.; Rahman, M. M.; Paul, K.; Chakraborti, D. A. simple household device to remove arsenic from ground water and two years performance report of arsenic removal plant for treating groundwater with community participation. In *Technologies for Arsenic Removal from Drinking Water*; Ahamed, M. F., Ali, M. A., Adeel, Z., Eds.; BUET, Dhaka and UNU: Tokyo, 2001; pp. 231−250.

(32) Ackerman, A. H.; Creed, P. A.; Parks, A. N.; Fricke, M. W.; Schwegel, C. A.; Creed, J. T.; Heitkemper, D. T.; Vela, N. P. Comparison of a chemical and enzymatic extraction of arsenic from rice and an assessment of the arsenic absorption from contaminated water by cooked rice. *Environ. Sci. Technol.* **2005**, *39*, 5241–5246.

(33) Mihucz, V. G.; Tatar, E.; Virag, I.; Zang, C.; Jao, Y.; Zaray, G. Arsenic removal from rice by washing and cooking with water. *Food Chem.* **2007**, *105*, 1718–1725.

(34) *Guidelines for Drinking-water Quality*, 2nd ed.; World Health Organization: Geneva, 1993.

(35) Laparra, J. M.; Velez, D.; Barbera, R.; Farre, R.; Montoro, R. Bioavailability of inorganic arsenic in cooked rice: practical aspects for human health risk assessments. *J. Agric. Food Chem.* **2005**, *53*, 8829–8833.

(36) Ministry of Rural Development, 2004 Department of Drinking Water Supply, Goverment of India, Letter No. W11036/01/2002 TM II, dated 20/10/2004.

(37) www.smilefoundationindia.org/ourchildren.htm (accessed November 11, 2008).

ES803414J