Arsenic intake via water and food by a population living in an arsenic-affected area of Bangladesh

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Abstract

More and more people in Bangladesh have recently become aware of the risk of drinking arsenic-contaminated groundwater, and have been trying to obtain drinking water from less arsenic-contaminated sources. In this study, arsenic intakes of 18 families living in one block of a rural village in an arsenic-affected district of Bangladesh were evaluated to investigate their actual arsenic intake via food, including from cooking water, and to estimate the contribution of each food category and of drinking water to the total arsenic intake. Water consumption rates were estimated by the self-reporting method. The mean drinking water intake was estimated as about 3 L/d without gender difference. Arsenic intakes from food were evaluated by the duplicate portion sampling method. The duplicated foods from each family were divided into four categories (cooked rice, solid food, cereals for breakfast, and liquid food), and the arsenic concentrations of each food category and of the drinking water were measured. The mean arsenic intake from water and food by male subjects was 0.18 ± 0.13 (n = 12) and that by female subjects was 0.096 ± 0.007 mg/d (n = 6), and the range for all 18 respondents was 0.043–0.49 mg/d. The average contributions to the total arsenic intake were, from drinking water, 13%; liquid food, 4.4%; cooked rice, 56%; solid food, 11%; and cereals, 16%. Arsenic intake via drinking water was not high despite the highly contaminated groundwater in the survey area because many families had changed their drinking water sources to less contaminated ones. Instead, cooked rice contributed most to the daily arsenic intake. Use of contaminated water for cooking by several families was suspected based on comparisons of arsenic concentrations between drinking water and liquid food, and between rice before and after cooking. Detailed investigation suggested that six households used contaminated water for cooking but not drinking, leading to an increase of arsenic intake via arsenic-contaminated cooking water.

Key words: Arsenic contamination; Bangladesh; Cooking water; Duplicate portion; Rice

1. Introduction

Arsenic contamination of groundwater has been reported from many parts of the world (Mandal and Suzuki, 2002; Hossain, 2006). The most severely contaminated areas include West Bengal, India (Das et al., 1996; Chakraborti et al., 2002), and Bangladesh (Nickson et al., 1998; Ohno et al., 2005). The first reported patient with arsenical dermatosis in West Bengal was diagnosed in 1983 (Chakraborty and Saha, 1987). Patients with skin disease caused by arsenic have also been observed in Bangladesh (Karim, 2000). Dhar et al. (1997) estimated that more than 50 million people in Bangladesh were at risk from arsenic, while Hossain (2006) estimated that 85 million were at risk.

To assess the health risks posed by arsenic, the comprehensive oral intake of arsenic must be estimated. However, this estimation presents some challenges with respect to drinking water quality and quantity. Recently, many people try to avoid drinking arsenic-contaminated groundwater because they have become aware of the risk of the arsenic contamination; instead, they obtain drinking water from less contaminated sources. Therefore, the use of arsenic concentrations in contaminated tubewells to indicate drinking water concentrations may lead to overestimation of the recent arsenic intake. Furthermore, although a mean daily water consumption rate of 2 L/d is generally used, it may not be always applicable for all over the world. WHO (2004) has stated, for instance, "In deriving WHO guideline values, it is assumed that the daily per capita consumption of drinking-water is approximately 2 liters for adults, although actual consumption varies according to climate, activity level and diet." Watanabe et al. (2004) estimated the mean daily water intake of people in Bangladesh to be 3 L/d. Nevertheless, there are few studies of water intake in developing countries.

Arsenic is consumed not only in water but also via food. Roychowdhury et al. (2003) estimated the daily arsenic intake via food in West Bengal, India, by the market basket sampling method, but this sampling method cannot take into account the effects of the cooking process and cooking water. To determine the actual arsenic intake via food, duplicate portion sampling is required. Methods of cooking rice, a staple food in Bangladesh, vary in different countries. In Japan, for example, people cook rice with very little water, all of which is absorbed by the rice during cooking, whereas in

Bangladesh, rice is cooked with excess water and water that is not absorbed during cooking is discarded (Bae et al., 2002; Rahman et al., 2006). Thus, the arsenic concentration may differ between uncooked and cooked rice and according to the method of cooking. Furthermore, there is a possibility of using arsenic contaminated-water for cooking, even though they obtain drinking water from less contaminated sources. These effects can be observed by the duplicate portion sampling.

In the present study, we evaluated arsenic intake via water and food by people living in an arsenic-affected district of Bangladesh. Many people in this district have symptoms of chronic arsenic poisoning, and many have recently stopped drinking arsenic-contaminated tubewell water, instead drinking less-contaminated (at least, by arsenic) water from dug wells. The principal aims of this study are therefore to investigate the contribution of food and water to the total arsenic intake after they obtain the less-contaminated drinking water sources, and to investigate whether they use uncontaminated water sources not only for drinking but for cooking. To evaluate arsenic intake via drinking water, we conducted a questionnaire survey to assess the 24-h water consumption rate and analysed arsenic concentrations in drinking water. To evaluate arsenic intake via food, including via cooking water, we used a duplicate portion sampling method. Although we could not directly measure the arsenic intake via cooking water, we evaluated its impact by comparing arsenic concentrations between uncooked and cooked rice. The arsenic concentration in liquid food such as soup was also used to estimate the contribution of cooking water to arsenic intake.

2. Materials and methods

2.1. Sample collection

We collected samples and conducted interviews in June 2005, in the rainy season; the maximum temperature during the survey period was around 38 °C. The study area was one block in Chunakhali village (24°36’N, 88°12’E), Chapai Nawabganj district, Rajshahi division, Bangladesh. This typical poor rural village has a population of about 2500. The block, where about 160 people live, is severely affected by arsenic and many people are afflicted with skin lesions associated with chronic arsenic poisoning. In addition, there are no arsenic treatment utilities or safe water distribution systems in the block. Water from dug wells contains much less arsenic than that from tubewells, but whereas some people have a dug well in or near their home, others have to walk up to a couple of hundred meters to get this less arsenic-contaminated water. After consulting with the local block leader and a dermatologist, we selected 77 people from 18 families in this block as our target subjects. We selected families representative of the block both financially and geographically and that had at least one family member with arsenical skin lesions. All the selected families agreed to participate the survey and many of them reported that they had changed their drinking water source from the arsenic-contaminated tubewells to a less-contaminated source from several months to several years previously.

We collected water from the present drinking water source of each family and also from previous drinking water sources when they were still available, in polyethylene bottles. Ideally, we sampled water directly from the source, but in some cases, we sampled water from a container in a house. In the latter case, we collected the sample after shaking the container vigorously. Water samples were transported to the laboratory by air, and then 1% v/v nitric acid (ultrapure grade; Kanto Chemical Co., Inc., Tokyo, Japan) was added to the samples, which were kept in a dark container at 4 °C until analysed.

We collected food samples by the duplicate portion sampling method (Tsuda et al., 1995; WHO/IPCS, 2000). We selected one adult from each family as the respondent, who submitted his or her duplicate meals (breakfast, lunch, and supper) for one day. Each cooked item was collected in a separate plastic bag and weighed. We estimated food intakes of non-respondent family members in relation to the food intake of the respondent from interview results, and these food intakes were used for the estimation of arsenic intakes via cooking water of the non-respondents. Food samples are typically composited by meal or by day (WHO/IPCS, 2000), but we composited the food separately into the following four categories: cooked rice, cereal, solid food, and liquid food. Cereal was defined in this study as cereals that the people ate for breakfast, and did not include rice or rice gruel. Cereal was mainly of two types: one, called “Kalai rooti” locally, was a variety of chapati made from lentil powder and rice flour, and the other was oat powder mixed with water, sugar, and salt. Solid food was defined as the solid portion of meals such as fried vegetables, excluding staples such as cooked rice and cereal. Liquid food was the liquid portion of soup, but solid ingredients in soups were categorised as solid food. Spices and lentils that could not be separated using a spoon and fork were treated as liquid food. Uncooked rice was also collected for comparison. After collection and arrangement of the food samples, solid food samples were homogenised and then transported to the laboratory by air. In the laboratory, solid food, rice, and cereal samples were freeze-dried for two to three days, and the water contents were estimated by measuring the weight of the samples before and after freeze-drying. The samples were stored in individual plastic bags and kept in desiccators. Liquid food samples were stored at –80 °C until analysed.

2.2. Estimation of direct water consumption rate

To quantify the daily water intake by direct drinking, we used a method similar to the Cup Method (Watanabe et al., 2004), in which direct water intake is estimated by asking the respondent how many cups of water are consumed in a day. At the first visit, the cup used for drinking water was identified and the capacity of the cup was measured. Each subject self-recorded the number of cups drunk (the “water diary” method) (Shimokura et al., 1998; Levallois et al., 1998). Recording sheets were provided to the subjects, who were asked to mark the sheet every time they drank water from their own cup. About 24 h later, the sheets were collected, and the number of cups marked was multiplied by the capacity of that subject's cup to estimate the water consumption rate. Water diary data were obtained for 65 of the 77 subjects (84%). Beverages made with water, such as tea, were not considered in this study because they are not common drinks for the poor village people. Most of the poor village people take tea in a small cup, and the amount of water used is negligible, not more than one cup per day.

2.3. Analytical method for total arsenic concentration

Arsenic in water was quantified by inductively coupled plasma–mass spectrometer (ICP-MS; HP-4500; Agilent Technologies, Inc., Palo Alto, CA, USA). The instrumental parameters were as follows: RF power, 1200 W; RF matching, 1.8 V; sample skimmer cone in Ni; monitoring masses; 75 (As) and 77 ($ArCl^+$); plasma flow rate, 16 L/min; auxiliary flow rate, 1.1 L/min; nebulizer flow rate, 1.2 L/min. Yttrium (Y; m/z = 89) was used as the internal standard.

Dried samples of rice, cereal, and solids were finely ground in a mill (A11 basic; IKA Werke GmbH & Co. KG, Staufen, Germany). Then the ground solid samples and liquid samples were digested using a microwave digestion system (ETHOS TC; Milestone S.r.l., Bergamo, Italy) by the following procedure. A 0.5-g (dry weight) portion (solid sample), or a 1.0-g (wet weight) portion (liquid sample) was weighed into a PTFE vessel, and 3 mL of nitric acid and 2 mL of hydrogen peroxide (ultrapure grade; Kanto Chemical Co., Inc., Tokyo, Japan) were added. The basic program of the microwave digester was as follows: increase the temperature from room temperature to 210 °C over 30 min, hold at that temperature for 15 min, and cool down to room temperature over 10 min; maximum power was 1000 W. Times and temperatures were modified slightly, depending on the sample type. The digested solution was made up to 50 mL with ultrapure water and filtered through a 0.45-µm membrane filter before injection into the ICP-MS instrument.

The validity of the analysis was confirmed with the Standard Reference Materials (SRM) Rice Flour (SRM1568a) and Typical Diet (SRM1548a), purchased from the National Institute of Standards and Technology (NIST), Gaithersburg, MD, USA. The certified arsenic concentration and our observed concentration (mg/kg dry wt) of the Rice Flour were 0.29 ± 0.03 and 0.26 ± 0.01 (n = 3, mean ± standard deviation [SD]), and those of the Typical Diet were 0.20 ± 0.01 and 0.21 ± 0.00 (n = 3), respectively. The certified and the observed values were thus in good agreement.

2.4. Speciation analysis of arsenic

Analyses of arsenic species were carried out for liquid food samples and some rice samples. Liquid food samples of 5 mL were centrifuged for 10 min at 3000 rpm and at 4 °C. The supernatant was filtered through a 0.45-µm membrane filter, and then 1 mL was taken and made up to 25 mL with ultrapure water.

Rice samples of 0.5 g (dry weight) were soaked in a solvent mixture of methanol (5 mL) and water (5 mL) and put in the digester. The program of the digester was as follows: increase the temperature from room temperature to 114 °C over 4 min, hold at that temperature for 21 min, and then cool down to room temperature over 10 min; maximum power was 500 W. The sample was centrifuged for 15 min at 3000 rpm and at 4 °C. Then, the supernatant was filtered through a 0.45-µm membrane filter and made up to 25 mL with ultrapure water. Sep-pak C18 cartridges (Waters Corp., Milford, MA, USA) were used to remove coexisting hydrophobic substances, because organic matter in the raw extracts may interfere with chromatographic separation (Yuan et al., 2005).

Arsenic species were quantified with high-performance liquid chromatography (HPLC; HP-1100, Agilent Technologies, Inc., Palo Alto, CA, USA) coupled with ICP-MS. A cationic column (RSpak NN-614; Showa Denko K.K., Tokyo, Japan) and a mobile phase consisting of a mixture of 6 mM NH_4NO_3, 5 mM HNO_3, and 1.5 mM PDCA were used for speciation of dimethylarsinic acid (DMA) and As(III). An anionic column (Excelpak G1836A; Agilent Technologies) and a mobile phase consisting of a mixture of 2 mM NaH_2PO_4 and 2 mM EDTA-2Na at pH 8.7 were used for speciation of methylarsonic acid (MMA) and As(V). The instrumental parameters were as follows: injection volume, 50 µL; flow rate, 0.8 mL/min; and column temperature, 25 °C. Standard solutions were prepared from MMA and DMA (Tri Chemical Laboratories Inc., Yamanashi, Japan), and diarsenic trioxide (As(III)) and disodium hydrogen arsenate heptahydrate (As(V)) (Wako Pure Chemical Industries, Ltd., Osaka, Japan).

The certified value of total arsenic in the Rice Flour SRM was 0.29 ± 0.03 mg/kg dry wt. Our observed values (n = 3) were 0.09 ± 0.004 for As(III), 0.022 ± 0.001 for As(V), 0.011 ± 0.001 for MMA, and 0.162 ± 0.01 for DMA. The sum of the mean values was 0.285 mg-As/kg dry wt, which is in the range of the certified value.

2.5. Estimation of individual arsenic intake from drinking water and food

The daily arsenic intake of each individual from drinking water and food was estimated as follows. The arsenic intake from drinking water (mg/d) was calculated by multiplying the arsenic concentration in water of the drinking source (mg/L) by the water consumption rate (L/d). The daily arsenic intake from liquid food was calculated by multiplying the arsenic concentration in liquid food (mg/kg wet wt) by the consumption rate of liquid food (kg wet wt/d). The arsenic intake from each food category other than liquid food was estimated by the following equation:

$$\text{Arsenic intake}_{i,j}\ (\text{mg/d}) = (\text{Arsenic concentration}_{i,j}\ [\text{mg/kg dry wt}]) \times (\text{Daily consumption rate}_{i,j}\ [\text{kg wet wt/d}]) \times (1 - \text{Water content}_{i,j})$$

where *i* is the food category (cooked rice, solid food, and cereal) and *j* is the subject.

To estimate the arsenic intake from drinking water, we assumed that the subjects drank water only from the collected drinking water source. This may have introduced error, probably underestimation, in the estimated arsenic intake. Levallois et al. (1998), for example, reported that 25% of water intake was away from home as the case in the Quebec City region in Canada. This type of study is required for better understanding of the actual water consumption habit in Bangladesh.

3. Results and discussion

3.1. Daily water intake via direct drinking and daily food consumption rates

Estimated daily water intakes by gender and age are shown in Table 1. Direct water intake did not differ between adult men (3.1 ± 1.3 L/d) and adult women (2.9 ± 1.0 L/d). Maximum water intake was as much as 5.7 L/d. These results are similar to those of Watanabe et al. (2004), who estimated a mean water intake of around 3 L/d with no gender difference, and a maximum water intake of 6.0 L/d. Even though both studies were conducted during the hot season, which may have caused increased water intakes, these results show that the water consumption rate of 2 L/d generally used for risk assessment may not always be adequate, especially in agricultural countries in the tropics such as Bangladesh.

Daily food consumption rates measured by the duplicate portion sampling method are shown in Table 2. Mean consumption rates of female subjects were 71% (cooked rice), 71% (cereal), 68% (solid food), and 72% (liquid food) of those of male subjects. All families ate cereals, including "Kalai rooti" and oat powder, as the staple food for breakfast, except one family, which ate rice gruel. Almost all of the solid food items were vegetables and fruits. Exceptions were mutton, eaten in two meals, and fish, eaten in four meals. Eleven of the 18 families had liquid food such as lentil soup at least once on the survey day.

3.2. Arsenic concentrations in food and drinking water

Arsenic concentrations in each food category and in the present drinking water sources are shown in Table 3. The mean arsenic concentrations of the food categories on a dry basis were 0.34 ± 0.15 (uncooked rice), 0.46 ± 0.51 (cooked rice), 0.20 ± 0.11 (cereal), and 0.44 ± 0.58 mg/kg dry wt (solid food), and the mean values on a wet basis were 0.34 ± 0.15 (uncooked rice), 0.13 ± 0.13 (cooked rice), 0.13 ± 0.082 (cereal), 0.10 ± 0.15 (solid food), and 0.038 ± 0.046 mg/kg wet wt (liquid food). Considerably large SD and some outliers can be observed in the categories of cooked rice (on a dry basis), solid food, and liquid food. Water was generally used for cooking these foods, and the cooking water quality may affect the arsenic concentrations in cooked foods. Compared with uncooked rice, arsenic concentrations in cooked rice on a dry basis were distributed over a wider range, suggesting that cooking water quality may affect daily arsenic intakes via rice.

Two types of well, tubewells and dug wells, were used as drinking water sources at the time of the survey. The mean arsenic concentration in tubewell water was 0.032 ± 0.026 mg/L (n = 5), and 0.0035 ± 0.0030 mg/L in dug well water (n = 12). The concentrations were lower than the Bangladeshi drinking water standard, 0.05 mg/L, except for water from one tubewell. On the other hand, the mean arsenic concentration in water of previously used tubewells was 0.44 ± 0.32 mg/L (n = 7); water from six of seven contained more than 0.05 mg/L, and water from four contained more than 0.5 mg/L.

3.3. Daily arsenic intakes via drinking water and food

Daily arsenic intakes via drinking water and food estimated for the 18 respondents by the duplicate portion sampling are shown in Fig. 1. The mean arsenic intake was 0.15 ± 0.11 mg/d (n = 18; minimum, 0.043 mg/d; maximum, 0.49 mg/d). The mean arsenic intakes of male and female subjects were 0.18 ± 0.13 (n = 12) and 0.096 ± 0.007 mg/d (n = 6), respectively. Total arsenic intake was not normally distributed (Fig. 1); some households had higher arsenic intakes than would be expected with normal distribution, implying sources of contamination other than drinking water. The contamination source may be cooking water, discussed in the next section.

The mean arsenic intakes from different sources were 0.023 ± 0.027 (drinking water), 0.0056 ± 0.011 (liquid food), 0.090 ± 0.079 (cooked rice), 0.017 ± 0.019 (solid food), and 0.020 ± 0.014 mg/d (cereal). Arsenic intake from drinking water was not high, even though the groundwater in the survey area was highly contaminated, because many families had changed to less-contaminated drinking water sources, as mentioned above. Instead, the contribution of cooked rice to the daily arsenic intake exceeded that of drinking water. The average contribution of each category to the total daily arsenic intake was drinking water, 13.3%; liquid food, 4.4%; cooked rice, 55.9%; solid food, 10.8%; and cereal, 15.6% (Fig. 2). Cooked rice contributed most to the daily arsenic intake. These results are consistent with those of earlier studies conducted by market basket methods in India (Roychowdhury et al., 2003) and Bangladesh (Meharg and Rahman, 2003), in that rice contributed most to the arsenic intake when arsenic concentrations in drinking water were not very high.

Regarding other foodstuffs, high arsenic contents in fish and seafood (Schoof et al., 1999) and seaweed (Hanaoka et al., 2001) have been reported. The interviewed respondents stated that they do not eat seafood or seaweed because the survey area is very far from the ocean, but many of them reported that they ate river fish a few times a week. During the survey period, fish was contained in four meals in three families, but distinct increases in arsenic concentrations were not observed in the solid food samples including fish. Two possible reasons are that fish accounted for a relatively small proportion of the solid food consumed by the respondents, or that the fish did not contain a high arsenic concentration.

3.4. Contribution of cooking water to the total arsenic intake

We compared the arsenic concentrations in liquid food and in drinking water of each family. The arsenic concentrations were similar except for four families (Families #5, 10, 14, and 17; Fig. 1). In these four families, the concentrations in liquid food were considerably higher than those in drinking water. In one of these families (#17), the drinking water was known to come from a different source than the water used for cooking, suggesting that the other exceptions were also due to the use of water from a more contaminated source for cooking. To confirm this, we analysed the concentrations of dissolved inorganic arsenic in liquid food after removing small particles and suspended solids by centrifugation and filtration, and found that 71% to 98% of the total arsenic in liquid food consumed by these families was present in the dissolved inorganic form. Arsenic in natural waters is mostly found in inorganic form (Smedley and Kinniburgh, 2002), and thus this result indicated that most of the arsenic in the liquid foods was derived from water. Therefore, we inferred that more-contaminated water sources were used for cooking liquid foods in these four families.

To investigate the possibility of more contaminated water being used for cooking rice, we compared the difference in arsenic concentration between uncooked and cooked rice (dry basis) and the arsenic concentration in drinking water of each family. When arsenic concentrations (dry basis) in cooked rice were more than those in the uncooked rice, we attributed the increased arsenic to arsenic in the rice-cooking water, because only water is added when rice is cooked. The concentration difference between uncooked and cooked rice thus depends on the arsenic concentration in cooking water. We found that the difference in arsenic concentration between cooked and uncooked rice in four families (Family #3, 8, 10, and 14 in Fig. 1) was high considering that the arsenic concentrations in their drinking water were lower than 0.02 mg/L. An experiment in which Bangladeshi rice was cooked in a Bangladeshi manner, using water in which the arsenic concentration was 0.05 mg/L, showed an increase in the arsenic concentration of the rice before and after cooking of less than 0.1 mg/kg dry wt (unpublished results, K. Ohno and T. Kimura), compared with an increase of more than 0.1 mg/kg dry wt for these four families (Table 4). Therefore, we suspected that these households used arsenic-contaminated water for rice cooking, as was the case with liquid food. To confirm this, we carried out speciation analyses of arsenic in uncooked and cooked rice of the suspected households (Table 4). Almost all of the increases in arsenic concentrations in cooked rice were in arsenic of the inorganic form. Therefore, we inferred that these households used more arsenic-contaminated water for cooking rice.

Although several households apparently used contaminated water for cooking, different families (Families #5, 10, 14, and 17) were suspected to use it for liquid food than used it for cooked rice (Families #3, 8, 10, and 14). Therefore, we checked the validity of these results. Families #3 and #8, who were suspected to use contaminated water for cooked rice but not liquid food, did not consume liquid food during the survey period. Thus, this result is valid. Similarly, Families #5 and #17 were suspected to use contaminated water for liquid food but not for cooked rice. This result is also valid for the following reason. The arsenic concentrations of the more-contaminated water used for cooking by Families #5 and #17 were around 0.05 mg/L, as determined by the inorganic arsenic concentrations in their liquid foods. At this arsenic concentration, a distinct difference in arsenic concentration between uncooked and cooked rice would not necessarily be observed. Thus, we considered six of the 18 households to use more-contaminated water for cooking than for drinking.

3.5. Arsenic intake from water via direct drinking and cooking water

We estimated daily arsenic intakes from water via direct drinking, liquid food, and rice cooking water for all respondents to the survey on water consumption rates. Arsenic intake from drinking water was estimated by multiplying the water consumption rate by its arsenic concentration, and that from liquid food was estimated in the same manner. Arsenic intake from rice cooking water was estimated by multiplying the daily rice consumption rate by its arsenic concentration difference of rice before and after cooking (dry basis). We allocated zero to arsenic intake from rice cooking water instead of allocating negative values when the arsenic concentration in cooked rice is lower than that in uncooked rice. The averages estimated daily arsenic intake from water was 0.14 ± 0.14 mg/d for the families suspected to use contaminated cooking water, and 0.028 ± 0.033 mg/d for those that probably used uncontaminated water for cooking (Fig. 3). The mean estimated intakes of the families likely using contaminated cooking water were 0.061 ± 0.094 mg/d in drinking water, 0.014 ± 0.018 mg/d in liquid food, and 0.069 ± 0.097 mg/d in rice cooking water, whereas the corresponding values for families likely using uncontaminated water were 0.023 ± 0.026, 0.0007 ± 0.0008, and 0.0041 ± 0.0079 mg/d. Relatively high amounts of arsenic were thus ingested via cooking water in the households suspected to use contaminated water for cooking. There are several reasons why families might use arsenic-contaminated water for cooking: They may not be conscious of the importance of arsenic intake via cooking water; their drinking water sources may be too far away from home to carry sufficient water for cooking use; it may be difficult to carry the water in such cases as after dark or in heavy rain; or they might think that arsenic, like pathogenic microbes, can be detoxified by boiling, or that it may evaporate. In any case, the arsenic intake from using contaminated water for cooking can be reduced by making householders aware of the risks of using contaminated water, although it may still be difficult for them to carry sufficient uncontaminated water to their home.

In this study, we found that cooked rice contributed most to the daily arsenic intake after many families had changed their drinking water sources to less contaminated ones. In addition, we estimated that six of 18 households likely used uncontaminated water for drinking but not for cooking, even though all subjects reported using the same water for cooking as for drinking. This discrepancy may be caused by a response bias, especially by the inclination to answer questions in a manner that is viewed favorably by others, and needs to be explored in further work. The sample size in this study was small and the findings need to be confirmed with a larger sample size. In addition, we did not consider the effect of the cooking water used for cereal and solid food preparation on the total arsenic intake in this study, so there are additional opportunities for clarification of the intake pathway.

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Tables and Figures

Table 1 Estimated daily water intakes via direct drinking

Table 2 Food consumption rates measured by the duplicate portion sampling method

Table 3 Arsenic concentrations in food and in water from present drinking water sources

Table 4 Arsenic speciation in the uncooked and cooked rice of the families suspected to use arsenic-contaminated water for cooking

Fig. 1. Arsenic intake from drinking water and food estimated for the respondents to the duplicate portion sampling. “M” and “F” denote male and female subjects, respectively.

Fig. 2. Average contributions of each category to the total daily arsenic intake.

Fig. 3. Averages of estimated daily arsenic intake from water via direct drinking, liquid food, and rice cooking water (n = 65). Six of 18 families were suspected to use more-contaminated water for cooking than for drinking. Details of the estimation method are described in the text.

Table 1 Estimated daily water intakes via direct drinking

	n	Age [a]	Water intake [a] (L/d)
Adults (age >15 years)			
Male	28	34 ± 18 (16–80)	3.1 ± 1.3 (1.0–5.7)
Female	23	40 ± 15 (20–70)	2.9 ± 1.0 (1.2–5.0)
Children			
Male	4	11 ± 3 (8–15)	2.7 ± 0.7 (1.7–3.3)
Female	10	11 ± 3 (6–15)	2.0 ± 0.5 (1.2–2.7)

[a] mean ± SD (min–max)

Table 2 Food consumption rates measured by the duplicate portion sampling method

		Cooked rice	Cereal	Solid food [a]	Liquid food [b]
		(g wet wt/d)			
Male					
	n	12	11	12	6
	mean ± SD	776 ± 183	188 ± 68	273 ± 184	220 ± 124
	(min–max)	(486–1045)	(87–353)	(60–698)	(102–384)
Female					
	n	6	6	5	5
	mean ± SD	553 ± 183	133 ± 49	185 ± 94	159 ± 48
	(min–max)	(292–794)	(82–208)	(86–335)	(114–218)

[a] Consists of the solid portion in meals except for staple foods (rice and cereal). Solid ingredients in soup are also categorised here.
[b] Consists of the liquid portion of soup. Spices and lentils in soup that could not be separated using a spoon and fork are also categorised here.

Table 3 Arsenic concentrations in foods and water from present drinking water sources

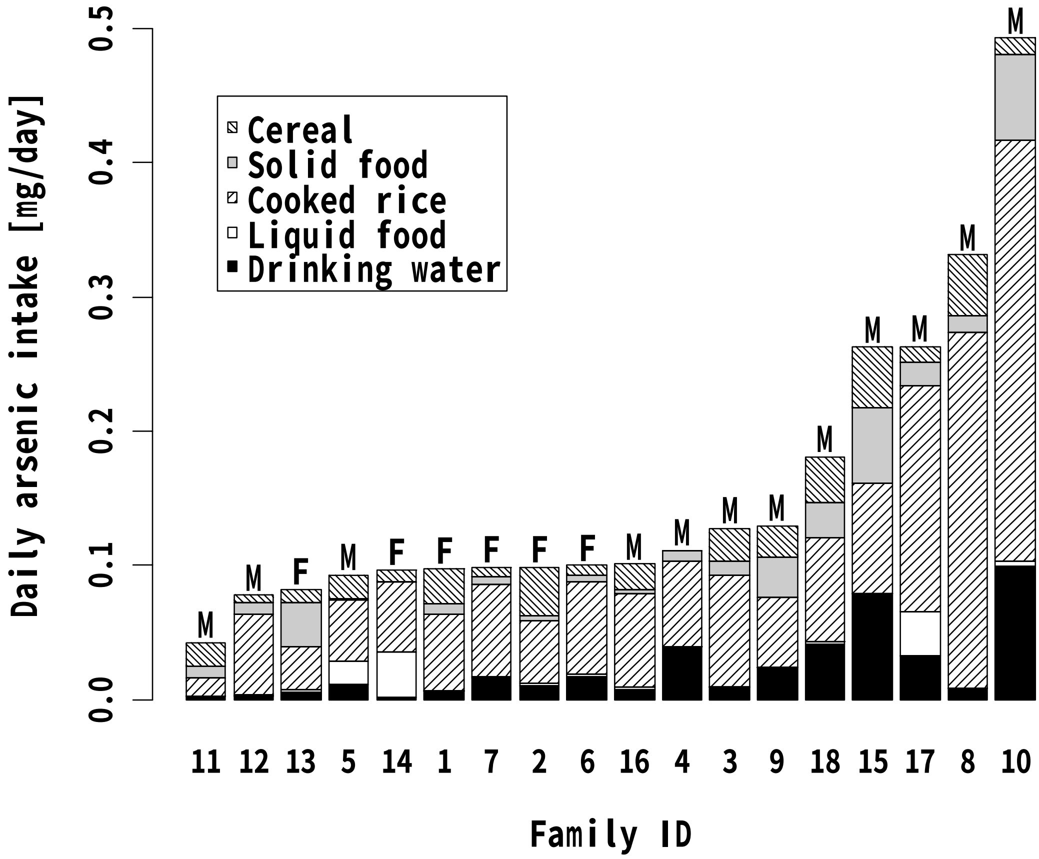
Category	n	Arsenic concentrations					
		Mean ± SD	Range and quartiles				
			min.	lower quartile	median	upper quartile	max.
Food on a dry basis (mg/kg dry wt)							
Uncooked rice	18	0.34 ± 0.15	0.15	0.22	0.30	0.44	0.59
Cooked rice	18	0.46 ± 0.51	0.10	0.23	0.30	0.51	2.37
Cereal	17	0.20 ± 0.11	0.086	0.12	0.16	0.23	0.44
Solid food	17	0.44 ± 0.58	0.074	0.12	0.15	0.40	1.85
Food on a wet basis[a] *(mg/kg wet wt)*							
Uncooked rice	18	0.34 ± 0.15	0.15	0.22	0.29	0.43	0.59
Cooked rice	18	0.13 ± 0.13	0.028	0.064	0.10	0.15	0.60
Cereal	17	0.13 ± 0.082	0.060	0.073	0.090	0.16	0.33
Solid food	17	0.10 ± 0.15	0.018	0.026	0.029	0.075	0.50
Liquid food	11	0.038 ± 0.046	0.0082	0.012	0.014	0.044	0.16
Drinking water sources (mg/L)							
Tubewell water	5	0.032 ± 0.026	0.0065	0.019	0.020	0.045	0.071
Dug-well water	12	0.0035 ± 0.0030	0.0010	0.0016	0.0025	0.0039	0.011

[a] Water contents measured by freeze-drying were 1.7 ± 1.7% (uncooked rice), 70 ± 3% (cooked rice), 35 ± 20% (cereal), and 78 ± 4% (solid food).

Table 4 Arsenic speciation of the uncooked and cooked rice of the families suspected to use arsenic-contaminated water for cooking

Family	Rice type	Arsenic concentration (mg-As/kg dry wt)				Extraction efficiency [c] (%)
		Inorganic arsenic [a]	MMA [a]	DMA [a]	Total arsenic [b]	
#3	Uncooked	0.28	nd	0.001	0.26	106
	Cooked	0.39	nd	0.001	0.40	97
#8	Uncooked	0.58	nd	0.010	0.58	101
	Cooked	0.88	nd	0.008	0.89	100
#10	Uncooked	0.44	nd	0.002	0.42	104
	Cooked	2.42	nd	nd	2.37	102
#14	Uncooked	0.26	nd	0.009	0.30	89
	Cooked	0.62	nd	0.008	0.61	102

nd: not detected, [a] by HPLC-ICP-MS, [b] by ICP-MS,
[c] (sum of arsenic species by HPLC-ICP-MS) / (total arsenic by ICP-MS)



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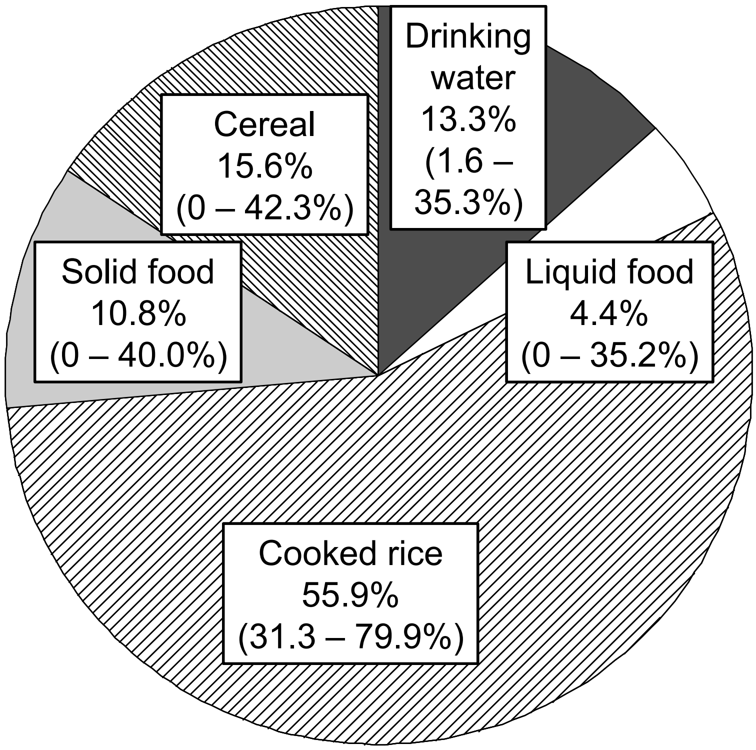
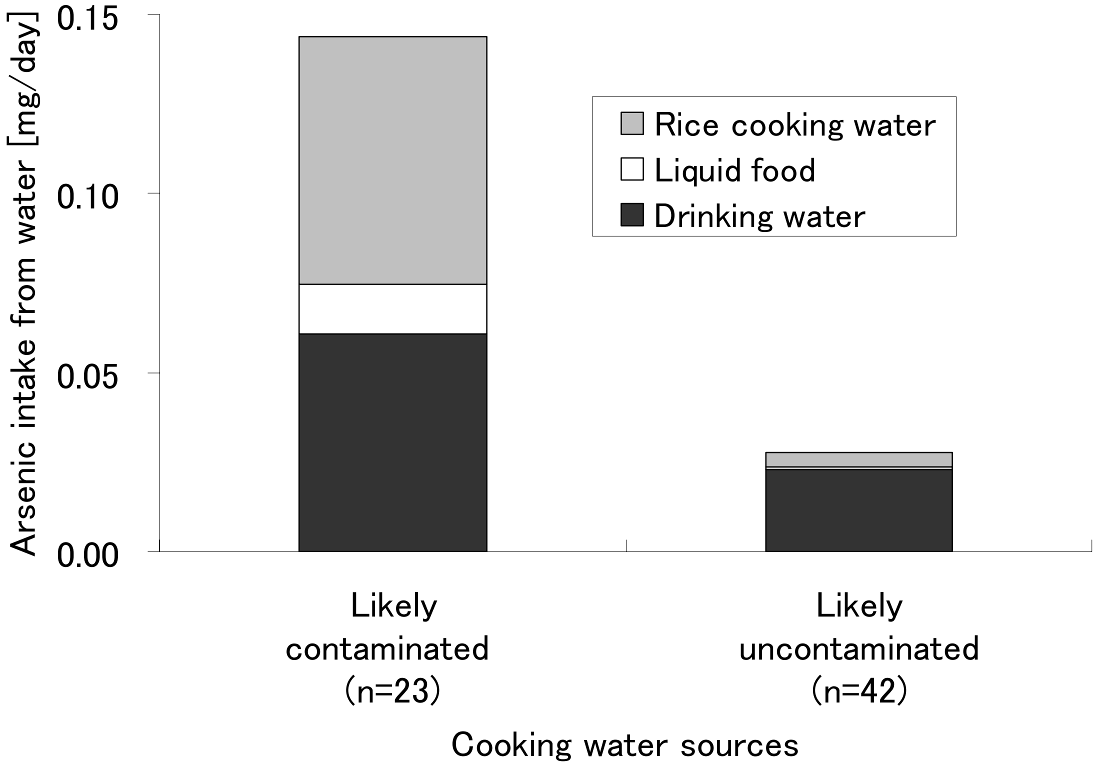


Fig. 2. Average contributions of each category to the total daily arsenic intake.



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