

Risk of Arsenic Exposure from Drinking Water and Dietary Components: Implications for Risk Management in Rural Bengal

Dipti Halder,^{*,†,‡} Subhamoy Bhowmick,^{‡,§} Ashis Biswas,^{†,‡} Debashis Chatterjee,[‡] Jerome Nriagu,^{||} Debendra Nath Guha Mazumder,[⊥] Zdenka Šlejkovec,[#] Gunnar Jacks,[†] and Prosun Bhattacharya[†]

[†]KTH-International Groundwater Arsenic Research Group, Department of Land and Water Resources Engineering, KTH Royal Institute of Technology, Teknikringen 76, SE-100 44 Stockholm, Sweden

[‡]Department of Chemistry, University of Kalyani, Kalyani 741235, West Bengal, India

[§]Department of Chemistry, University of Girona, Campus de Montilivi, Girona 17071, Spain

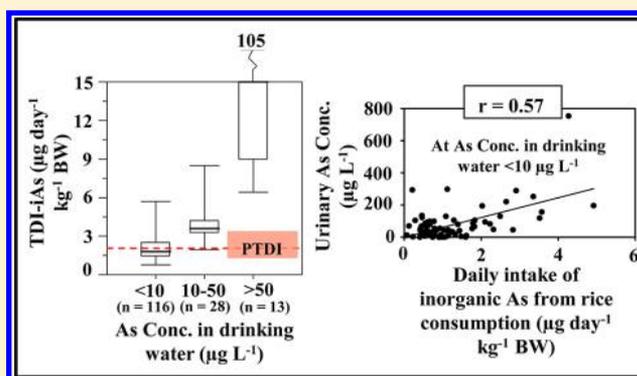
^{||}Department of Environmental Health Sciences, School of Public Health, University of Michigan, 109 Observatory Street, Ann Arbor, Michigan 48109-2029, United States

[⊥]DNGM Research Foundation, 37C Block B, New Alipore, Kolkata 700053, West Bengal, India

[#]Department of Environmental Sciences, Jožef Stefan Institute, Jamova 39, Ljubljana, SI-1000, Slovenia

Supporting Information

ABSTRACT: This study investigates the risk of arsenic (As) exposure to the communities in rural Bengal, even when they have been supplied with As safe drinking water. The estimates of exposure via dietary and drinking water routes show that, when people are consuming water with an As concentration of less than $10 \mu\text{g L}^{-1}$, the total daily intake of inorganic As (TDI-iAs) exceeds the previous provisional tolerable daily intake (PTDI) value of $2.1 \mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$, recommended by the World Health Organization (WHO) in 35% of the cases due to consumption of rice. When the level of As concentration in drinking water is above $10 \mu\text{g L}^{-1}$, the TDI-iAs exceeds the previous PTDI for all the participants. These results imply that, when rice consumption is a significant contributor to the TDI-iAs, supplying water with an As concentration at the current national drinking water standard for India and Bangladesh would place many people above the safety threshold of PTDI. We also found that the consumption of vegetables in rural Bengal does not pose a significant health threat to the population independently. This study suggests that any effort to mitigate the As exposure of the villagers in Bengal must consider the risk of As exposure from rice consumption together with drinking water.



INTRODUCTION

Over the last few decades, the contamination of groundwater by geogenic arsenic (As) has been highlighted as an environmental disaster in many regions of the world.¹ The problem is most acute in South and Southeast Asia, particularly in the eastern to northeastern part of India and adjoining Bangladesh, where as many as 60 million people are at risk of chronic As poisoning.^{2,3} Most of the studies conducted in As affected regions have mainly focused on the characterization of source and processes responsible for As mobilization into groundwater and development of low-cost mitigation alternatives.^{4–11} Although various national and international aid agencies and local governments are trying to decrease the extent of As exposure by supplying safe water to the local communities, the success has been limited.¹² A recent development is the recognition that the local staple diet is an important route of As exposure.^{13–20}

As a consequence of the “Green Revolution,” farmers in India and Bangladesh are now cultivating their land three to four times per year, which has led these countries to become self-reliant for food. These cultivations are largely dependent on groundwater irrigation. In the last few decades, thousands of high capacity, large diameter motorized pumps have been installed to meet this irrigation requirement.^{21,22} These pumps are mostly abstracting groundwater from shallow aquifers, which are heavily contaminated with dissolved As. Consequently, As is increasing on the top soil of the irrigated lands. By monitoring As concentration in the Bangladeshi paddy field soil over a period of three years, Dittmar et al.²³ have estimated that continuation of the current irrigation

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practice would increase the As concentration in the top 40 cm of the paddy field soil by a factor of 1.5–2 by the year 2050. The increased As concentration in the irrigated lands ultimately results in the accumulation of As in the food crops cultivated on these lands.^{24,25} General cultivation practices such as continuous flooding of irrigation land for the cultivation of rice (*Oryza sativa* L.) also increase the mobility of As in the irrigation land.^{25,26} Previous studies have attempted to quantify the As concentration in different food components generally cultivated in these regions with a view to assessing the dietary As exposure among the local population.^{13–20} However, most of these studies were undertaken in the areas where people were being exposed to high levels of As in their drinking water. So far, not much attempt has been made to assess the risk of As exposure when people are supplied with As safe drinking water.

The objective of the present study is to assess the risk of As exposure to the communities of rural Bengal, where people have been supplied with As safe drinking water for the past few years. A questionnaire-based survey is combined with the collection and quantification of As concentration in drinking water and different food components that comprise a standard diet of the local population. Speciation is conducted to evaluate the accumulation of major As species in the food components prior to the assessment of dietary As exposure. An attempt is made to critically compare the extent of As exposure from drinking water and different components of the local diet. Urinary As concentration of the participants is determined to assess the current status of As exposure.

MATERIALS AND METHODS

Questionnaire Survey and Sample Collection. This study includes the same cohort of 157 participants (male: 68; female: 89) reported by Halder et al.²⁷ The participants were selected randomly from the three neighboring villages, namely, Chhoto-Itna, Debogram, and Tehatta of Nadia District, West Bengal, India. The only selection criterion was that they must have lived in their villages at the time of the study for at least 10 years. The participants were interviewed face to face in the local language of Bengali following a structured questionnaire, which included demographic information (e.g., age, height, body weight, level of education, occupation, marital status, smoking habit, ongoing medication, etc.), dietary habit (frequency, amount of consumption, and source of the components), and pattern of drinking water intake (for further details, we refer to Nriagu et al.²⁸). Following the questionnaire survey, rice samples ($n = 157$) were collected from the household of each participant. Since, in the rural households of Bengal, vegetables are not always available in the household's basket like rice, vegetables were collected directly from the sources. The questionnaire survey revealed that, although some participants had a home garden within their premises, a major portion of daily vegetables for all the participants was collected from the local market. Accordingly, the available vegetable samples were collected both from the home gardens ($n = 28$) and vegetable shops in the local market ($n = 52$). Vegetable samples were collected two times over the year (summer and winter) to include most of the available types of seasonal vegetables that people consume. Drinking water samples ($n = 24$) were collected in 15 mL prewashed polyethylene bottles from the sources, which supply water to the participants, and then acidified onsite with 0.15 mL of HNO₃ (14 N, Suprapur, Merck). Urine samples ($n = 80$) were collected from the group of participants, who were drinking Public Health Engineering

Department (PHED)-supplied tap water, presumed to be safe for As ($<10 \mu\text{g L}^{-1}$). In the field after collection, urine samples were preserved in the ice box, and after being returned to the laboratory, the samples of urine and vegetables, water, and rice were preserved at $-20 \text{ }^\circ\text{C}$, $4 \text{ }^\circ\text{C}$, and room temperature, respectively, until they were analyzed.

Sample Analysis. Methods for the characterization and analysis of total As concentration in the rice samples have been described previously by Halder et al.²⁷ In the subsequent section of this paper, we are reporting only the methodology adopted for As speciation. Collected vegetable samples were washed with Millipore water (18 M Ω) and dried in a hot-air oven at $65 \text{ }^\circ\text{C}$ for 48 h. The dried vegetable samples were milled with a mechanical grinder and subsequently digested with HNO₃ followed by H₂O₂ for the quantification of total As. The same digestion procedure of rice (see Halder et al.²⁷ for details) was applied for the vegetables samples. The urine samples were also digested with HNO₃ and H₂O₂ after filtering through a $0.45 \mu\text{m}$ membrane for the quantification of total As (see the Supporting Information for details of the digestion procedure). The concentration of total As in the digested vegetable and urine samples and in drinking water was analyzed by a hydride generation atomic absorption spectrometer (HG-AAS, Varian AA240, detection limit $<1 \mu\text{g/L}$) at the Department of Chemistry, University of Kalyani.²⁹ Since the vegetable samples were digested following the same procedure as the rice, one sample of standard reference material of rice flour (SRM 1568a) from the National Institute of Standards and Technology (NIST) was included in every batch of As analysis of the vegetable samples to ensure instrumental accuracy. For urine and drinking water analyses, SRMs of 2670a and 1643e were used, respectively. For all SRM samples of rice flour, urine, and water, the recovery was $>97\%$. One-fifth of the drinking water, vegetables, and urine samples were reanalyzed to test the precision of the analysis (see the Supporting Information for details of the quality assurance).

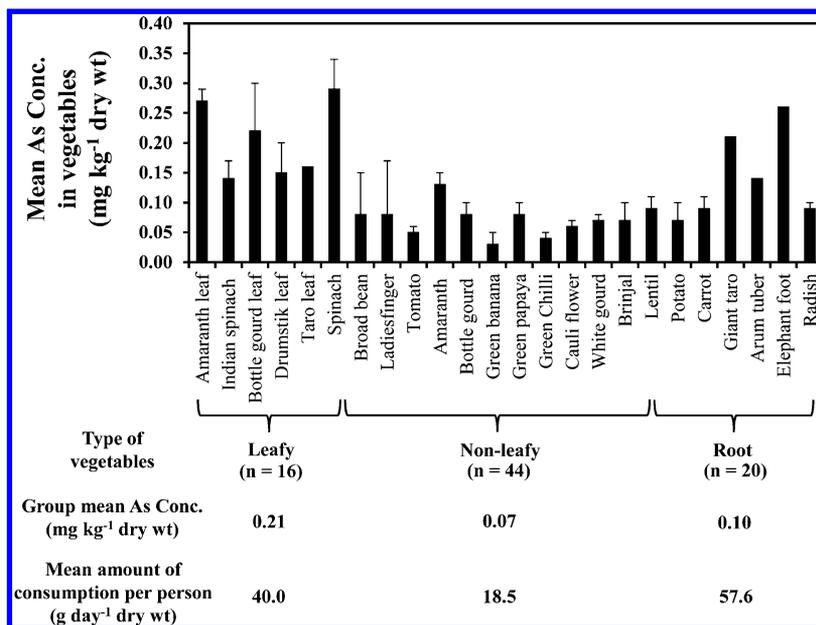
Speciation of As in the food components was conducted in the Department of Environmental Sciences, Jožef Stefan Institute. Different As species (As(III), As(V), MMA, and DMA) in the rice and vegetable samples were extracted with a mixture of water and methanol (1:1). The mild extractant was selected because a stronger extraction condition using a mixture of acids would increase the extraction yield but at the same time might cause some decomposition of the organically bound As and falsely indicate higher inorganic As content.³⁰ The chromatographic separation of the species in the extract was performed with a Hamilton PRP-X100 anion-exchange column and subsequently quantified with a hydride generation atomic fluorescence spectrometer (HG-AFS, PS Analytical, Kent, UK).³⁰ The total As concentration in the samples, in which speciation was performed, was reanalyzed by radiochemical neutron activation analysis (RNAA) to estimate the fraction of As recovery with the extractant (see the Supporting Information for detailed methodology of As speciation and total As determination with RNAA in rice and vegetables samples).

Assessment of As Exposure through Consumption of Drinking Water, Rice, and Vegetables. Daily intake of inorganic As due to consumption of a particular exposure medium (e.g., drinking water, rice, and vegetables) (DI-iAs_i) for a participant was computed using the equation as follows:³¹

$$\text{DI-iAs}_i (\mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}) = (C_i \times X_i \times W_i) / \text{BW} \quad (1)$$

Table 1. Consumption Pattern of Drinking Water and Dietary Components (Rice and Vegetables) among the Participants of Different Age Groups

dietary component	18–30 years (<i>n</i> = 13)			31–50 years (<i>n</i> = 112)			51–65 years (<i>n</i> = 32)		
	min	median	max	min	median	max	min	median	max
rice (g day ⁻¹ dry wt)	200	350	400	100	350	500	150	400	450
vegetables (g day ⁻¹ dry wt)	86.3	118	131	22.7	121	147	78.3	119	137
drinking water (L day ⁻¹)	2.5	3.5	6.0	2.0	3.5	6.0	2.5	3.5	5.0

**Figure 1.** Mean As concentrations in the vegetables generally consumed in the study area. Error bars represent standard deviation.

where C_p , X_p , W_p , and BW represent the concentration of total As in the exposure medium ($\mu\text{g L}^{-1}$ for drinking water and $\mu\text{g kg}^{-1}$ for rice and vegetables), the fraction of inorganic As content in the medium, the amount of daily consumption of the exposure medium by the participant (L day^{-1} for drinking water and kg day^{-1} for rice and vegetables), and the body weight of the participant (kg), respectively. The daily intake of inorganic As due to consumption of rice (DI-iAs-R) was calculated for the participants by Halder et al.²⁷ previously. However, in this study, the fraction of inorganic As content in rice was accounted as 0.81, as reported by Williams et al.³² To make an exact estimation, the present study recalculates the values of DI-iAs-R by quantifying the fraction of inorganic As content in the rice samples consumed in the study area as 0.92. The fraction of inorganic As content in the collected vegetable samples was quantified as 1. No attempt was made to speciate As in drinking water, as it was reported that As in groundwater is present primarily in inorganic form,^{33,34} and total As concentration was used for the subsequent daily intake calculation. By combining DI-iAs-R, daily intake from drinking water (DI-iAs-DW), and daily intake from vegetables (DI-iAs-V), the total daily intake of inorganic As (TDI-iAs) for each participant was calculated. It should be mentioned here that, at the recent 72nd meeting of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), the previous provisional tolerable daily intake (PTDI) value for inorganic As intake ($2.1 \mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$) has been withdrawn, because the value was in the lower range of the $\text{BMDL}_{0.5}$ (bench mark dose level for 0.5% increased prevalence of lung cancer).³⁵ Thus currently, there is no established guideline to assess health

risk due to dietary intake of inorganic As. However, the Codex Committee on Contaminants in Foods (CCCF) has argued that a TDI-iAs value below the $\text{BMDL}_{0.5}$ does not necessarily indicate that there is no risk and cannot be regarded as a safety standard,³⁶ which motivated us to compare TDI-iAs of the participants with the previous PTDI value of $2.1 \mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$ to assess the health risk of inorganic As exposure from drinking water and dietary components.

RESULTS AND DISCUSSION

Dietary Habit in Rural Bengal. Similar to previous studies conducted in other parts of India and Bangladesh,^{13,14,17} the food frequency questionnaire survey adopted in the present study reveals that rice with vegetables is the main staple food in rural Bengal. People mostly consume rice three times per day with vegetables. Daily laborers and farmers even carry homemade food and drinking water to the working places. The consumption of wheat, fruits, and animal protein (in the forms of egg, fish, chicken, mutton, and beef) is very occasional, estimated to be 2–4 times per month; therefore, these routes are not considered for the assessment of dietary As intake in this study. By weight, rice contributes almost 76% of the total diet, while the rest is composed of vegetables. Since the villagers either do not have land for rice cultivation or do not grow enough rice to last a whole year, the market place is the major source of rice to the villagers. The consumption pattern of different dietary components and drinking water shows that amount of rice consumption varies according to the age of the participants (Table 1). The older (51–65 years) participants consume a higher amount of rice compared to the younger

Table 2. Accumulation of Inorganic As in Food Stuffs Collected from the Study Area and Its Comparison to the Other Studies Conducted in Different Parts of West Bengal and Bangladesh

dietary component	type of component	reference	sample location	range of total As conc (mg kg ⁻¹)	% extracted	% of inorganic As
rice	household rice	present study	West Bengal	0.01–0.64	61	91.7 ± 9
	household rice	Mondal and Poly ¹⁶	West Bengal	0.09–0.17	82	74 ± 13
	market rice	Williams et al. ³²	India	0.03–0.08	80 ± 12	81 ± 4
	market rice	Williams et al. ³²	Bangladesh	0.03–0.30	75.5	80 ± 3
	paddy rice	Signes-Pastor et al. ¹⁷	West Bengal	–	98.4	49.8
	atab rice	Signes-Pastor et al. ¹⁷	West Bengal	–	98.9	45.1
	boiled rice	Signes-Pastor et al. ¹⁷	West Bengal	–	100	80.7
	puffed rice	Signes-Pastor et al. ¹⁷	West Bengal	–	100	33.3
	rice	Rahman et al. ⁴⁰	West Bengal	0.12–0.66	–	95
	cooked rice	Smith et al. ⁴¹	Bangladesh	0.04–1.11	–	87
	rice	Roychowdhury ²⁰	West Bengal	0.09–0.24	70.5	89.9
leafy vegetables	spinach	present study	West Bengal	0.24–0.35	14.7	100
	Indian spinach	present study	West Bengal	0.11–0.18	25	100
	vegetables	Roychowdhury ²⁰	West Bengal	0.13–0.29	70	89.2
non-leafy vegetables	green papaya	present study	West Bengal	0.04–0.11	51.4	74.3
	bottle gourds	Williams et al. ¹⁵	Bangladesh	0.32–0.47	90 ± 24	100 ± 0
	green banana	Williams et al. ¹⁵	Bangladesh	0.05–0.50	89 ± 37	100 ± 0
	long yard bean	Williams et al. ¹⁵	Bangladesh	0.33–0.87	87	100 ± 0
	tomato	Signes-Pastor et al. ¹⁷	West Bengal	–	96.4	100
	cauliflower	–	West Bengal	–	85.7	100
	brinjal	Signes-Pastor et al. ¹⁷	West Bengal	–	90.6	100
	potato	present study	West Bengal	0.03–0.12	91.3	100
root vegetables	potato	Williams et al. ¹⁵	Bangladesh	0.05–0.89	128	100 ± 0
	arum stolon	Williams et al. ¹⁵	Bangladesh	0.05–1.93	79 ± 33	100 ± 0
	arum tuber	Williams et al. ¹⁵	Bangladesh	0.09–0.31	100	100 ± 0
	carrot	Signes-Pastor et al. ¹⁷	West Bengal	–	92.5	80.3
	radish	Signes-Pastor et al. ¹⁷	West Bengal	–	92.2	61
	onion	Signes-Pastor et al. ¹⁷	West Bengal	–	102	100
	betel nut	Signes-Pastor et al. ¹⁷	West Bengal	–	76.5	100
	potato	Signes-Pastor et al. ¹⁷	West Bengal	–	101	58
	vegetables	Rahman et al. ⁴⁰	West Bengal	0.01–0.12	–	94
	vegetables	Smith et al. ⁴¹	Bangladesh	0.02–2.33	–	96

(18–30 years) and middle age group (31–50 years) participants. However, the amount of vegetable consumption and drinking water intake is similar for the participants in all age groups (Table 1).

Vegetables that are generally consumed in the villages can be classified into three categories based on the edible part: leafy ($n = 16$), non-leafy ($n = 44$), and root ($n = 20$) vegetables (Figure 1). For all age groups, leafy and root vegetables represent the major portion of vegetable intake in the study area (Figure 1). The types of daily leafy vegetable consumption depend on the seasonal availability, and the amount of individual leafy vegetable intake over the year is roughly the same. Potato is available in the market throughout the year and constitutes a significant portion of the daily intake of vegetables.

Distribution of As in Drinking Water and Vegetables.

The questionnaire data indicates that people in the study area drink mostly groundwater. The drinking water sources include PHED-supplied tap water, government installed deep tube wells, and shallow private tube wells. The concentration of As in drinking water varies largely from below the instrumental detection limit ($<1 \mu\text{g L}^{-1}$) to $875 \mu\text{g L}^{-1}$. Out of the total surveyed drinking water sources ($n = 24$), only 9 (37.5%) sources are safe compared to the WHO provisional drinking water guideline of $10 \mu\text{g L}^{-1}$ for As. In 6 (25%) and 9 (37.5%) tube wells, the levels of As concentration are >10 – 50 and $>50 \mu\text{g L}^{-1}$, respectively. The PHED-supplied tap water and

government installed deep tube wells are safe, while privately owned household shallow tube wells are mostly contaminated with As. The survey data reveals that, though the number of drinking water sources with As concentration less than $10 \mu\text{g L}^{-1}$ is small, the majority of the participants ($n = 116$, 73.9%) collect drinking water from these sources for the last 3–4 years. Only 17.8% ($n = 28$) and 8.28% ($n = 13$) of the participants are consuming drinking water with As concentrations of >10 – 50 and $>50 \mu\text{g L}^{-1}$, respectively. Presently, people in the study area are sharing common low As drinking water sources due to increased social awareness about As exposure from drinking water according to a recent study by Guha Mazumder et al.³⁷

The comparison of mean As concentrations in the different groups of vegetable samples collected from the study area shows higher As accumulation in the leafy vegetables (mean: 0.21 mg kg^{-1}) compared to that in the non-leafy and root vegetables (mean: 0.07 and 0.1 mg kg^{-1} , respectively) (Figure 1). Though the study conducted by Alam et al.¹⁴ in Samta village of Bangladesh did not find a significant difference in As accumulation among the different groups of vegetables, studies by Williams et al.¹⁵ from different parts of Bangladesh and Roychowdhury et al.¹³ from As affected regions of Murshidabad, West Bengal, also reported similar higher As accumulation in leafy vegetables. If As accumulation is compared among the individual vegetables of different groups, higher As concentrations are observed in spinach and amaranth

leaf for leafy vegetables, amaranth stem for non-leafy vegetables, and giant taro, arum tuber, and elephant foot for root vegetables (Figure 1). The high accumulation of As in taro, arum tuber, and elephant foot has also been reported in Bangladesh.^{14,15,38} In Bangladesh, Das et al.³⁹ and Huq and Naidu³⁸ have also reported high concentration of As in potato. However in West Bengal, Roychowdhury et al.¹³ have found high concentration of As in potato skin compared to potato flesh. Since the consumption of potato skin is not very common in our study area, the present study attempted to quantify As concentration in potato flesh only and has found that it shows the lowest As accumulation (mean: 0.07 mg kg⁻¹) among the root vegetables (Figure 1). The comparison between As concentrations in vegetables collected from the households and market places did not show any significant difference.

Major As Species in the Dietary Components.

Speciation of As (organic and inorganic species) accumulated in the foods is necessary to accurately estimate the potential dietary As exposure.¹⁶ Few previous studies (e.g., Rahman et al.,⁴⁰ Williams et al.,^{15,32} Smith et al.,⁴¹ Mondal and Polya,¹⁶ Signes-Pastor et al.,¹⁷ and Roychowdhury²⁰) have attempted to quantify inorganic As accumulation in food components collected from the different As affected regions of West Bengal and Bangladesh. The present study tried to verify these trends by determining the inorganic As content in rice and different groups of vegetables that generally people prefer to consume in the study area (Table 2). Although, a significant amount of As was not extracted by the mixture of methanol and water, the present study further supports the predominant accumulation of inorganic As in the rice and vegetables grown in West Bengal and Bangladesh. This study indicates that 91.7% and 100% of the extractable As is present as inorganic species in rice and most of the studied vegetables, respectively (Table 2). It is further revealed that the percentage of inorganic As content in rice varies to some extent according to location (Table 2), which may be because of a complex interaction between edaphic and environmental factors.^{42–44} However, in most types of the vegetables, As is entirely present (100%) in inorganic form throughout the Bengal region. This prompted us to assume total As concentration for vegetables and 0.92 times of the total As concentration for rice during calculation of DI-iAs.

Human Exposure to As through Consumption of Drinking Water, Rice, and Vegetables. The comparison of DI-iAs-DW, DI-iAs-R, and DI-iAs-V has been shown in Figure 2, which indicates that the consumptions of rice and drinking water are the major sources of inorganic As intake in the study area. If these DI-iAs values for each participant are compared with the previous WHO recommended PTDI value, 17% of the participants are above the threshold for risk due to intake of inorganic As from the consumption of rice and drinking water separately. For none of the participant did the DI-iAs-V exceed the threshold value of PTDI (Figure 2). Although As in most of the vegetables is present exclusively as the inorganic species, consumption of vegetables alone is not a potential health risk to the population. When DI-iAs-V is considered together with DI-iAs-R to estimate the total dietary intake of inorganic As (DI-iAs-DC = DI-iAs-R + DC-iAs-V), the extent of health risk to the population is increased by 7% (Figure 2). It is worthwhile to mention here that recently Gamble et al.⁴⁵ have found increased As methylation capacity that decreased the blood As concentration level in Bangladeshi people with the supplement of a marginal level of folate. Thus, because of high folate

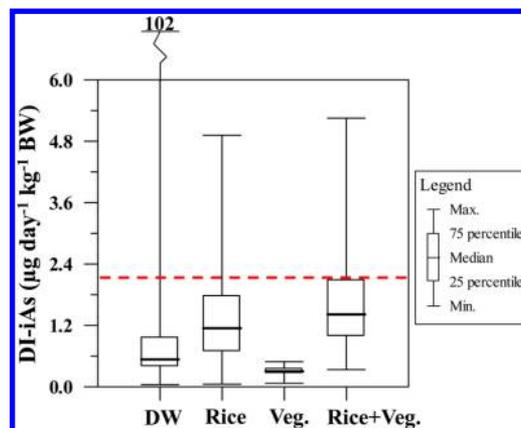


Figure 2. Comparison among daily intakes of inorganic As (DI-iAs) due to consumption of drinking water (DW), rice, and vegetables (Veg.). The red line represents the previous WHO recommended provisional tolerable daily intake (PTDI) value of 2.1 $\mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$.

content in the vegetables, particularly in the leafy type, consumption of vegetables might have a significant effect in decreasing the health burden of As toxicity to the rural population in Bengal. However, the critical comparison between the advantage of a folate supplement and the disadvantage of increased inorganic As intake with the consumption of vegetables is beyond the scope of the present study.

The values of TDI-iAs for each participant were categorized according to the ranges of As concentration in drinking water, namely, <10, 10–50, and >50 $\mu\text{g L}^{-1}$, to examine the contribution of drinking water and dietary components (rice and vegetables) to the TDI-iAs (Figure 3). The comparison of TDI-iAs for each category with the previous PTDI value indicates that, even when the As concentration in drinking water is less than 10 $\mu\text{g L}^{-1}$, for 35% ($n = 41$) of the participants, the TDI-iAs value exceeds the threshold value of

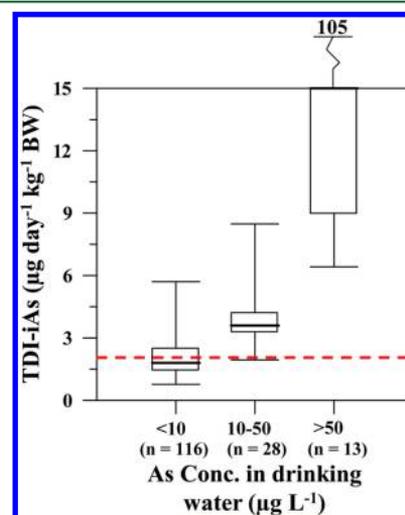


Figure 3. Comparison of total daily intake of inorganic As (TDI-iAs) due to the consumption of drinking water, rice, and vegetables at different concentration ranges of As in drinking water. The red line represents the previous WHO recommended provisional tolerable daily intake (PTDI) value of 2.1 $\mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$. The legend of the box whisker plot is the same as that in Figure 2.

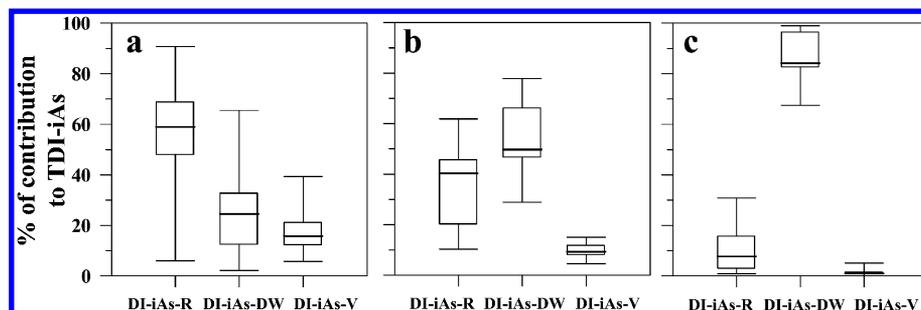


Figure 4. Percentage of contributions of daily intake of inorganic As from rice (DI-iAs-R), drinking water (DI-iAs-DW), and vegetables (DI-iAs-V) to the total daily intake of inorganic As (TDI-iAs) at As concentration in drinking water of (a) <10 , (b) $10\text{--}50$, and (c) $>50 \mu\text{g L}^{-1}$. The legend of the box whisker plot is the same as that in Figure 2.

$2.1 \mu\text{g day}^{-1} \text{kg}^{-1} \text{BW}$ (Figure 3). At this concentration level, the relative contribution of DI-iAs-R to TDI-iAs does significantly predominate over the contribution of DI-iAs-DW (Figure 4a). By assessing the risk of As exposure at the three regions of West Bengal, Mondal et al.¹⁹ have also reported that the risk of As exposure from rice consumption predominates in the area where As concentration in drinking water is low. Furthermore, the determination of urinary As concentration of the participants who were drinking PHED-supplied tap water indicates that, despite low intake of As from drinking water, the concentration of As in urine is considerably high (range: BDL– $753 \mu\text{g L}^{-1}$; median: $42.3 \mu\text{g L}^{-1}$). The urinary As concentration also strongly positively correlates with DI-iAs-R ($r = 0.57$), while this correlation with DI-iAs-V is not significant ($r = 0.08$) (Figure 5). This indicates that, even when people in rural Bengal are supplied with As safe drinking water, they still are at potential risk of As exposure due to consumption of rice. When the level of As concentration in drinking water is $>10\text{--}50 \mu\text{g L}^{-1}$, for all the participants, the TDI-iAs value exceeds the previous PTDI value (Figure 3), and DI-iAs-R and DI-iAs-DW

contribute almost equally to the TDI-iAs of the participants (Figure 4b). This signifies that, when consumption of rice contributes significantly to the TDI-iAs, the current national drinking water standard of India and Bangladesh are no longer protecting the population from As health risk. When the As concentration in drinking water exceeds $50 \mu\text{g L}^{-1}$, the relative contribution of DI-iAs-DW becomes so high that the influence of DI-iAs-R on the TDI-iAs becomes negligible (Figure 4c). At all categories, the contribution of DI-iAs-V to TDI-iAs is very small (Figure 4a–c).

Implications. This study suggests that the supply of As safe ($<10 \mu\text{g L}^{-1}$) drinking water to the population in rural Bengal alone is not enough to reduce the risk of As poisoning. Consumption of rice is another potential pathway of inorganic As exposure that must also be considered. Introduction of policies for sustainable agricultural practices that minimize the transfer of As from groundwater to soils to the human food chain is clearly needed. It should also be mentioned that, when consumption of rice significantly contributes to the TDI-iAs, an attempt should be made to decrease inorganic As intake from the drinking water further, so that the safe level of TDI-iAs can be maintained. It can be concluded that, in rural Bengal, any mitigation of As poisoning needs integrated approaches rather than the traditional fragmented strategies.

■ ASSOCIATED CONTENT

📄 Supporting Information

Details of the digestion procedure of urine for the analysis of total As; and quality assurance and analytical procedures adopted for the determination of As compounds by HPLC-HGAFS and total As by RNAA in rice and vegetable samples. This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: +46 8790 7919; fax: +46 8790 6857; e-mail: dipti@kth.se.

Notes

The protocol used in this study was approved by the institutional ethical committees of University of Kalyani, DNGM Research Foundation, and University of Michigan for the studies on human beings.

The authors declare no competing financial interest.

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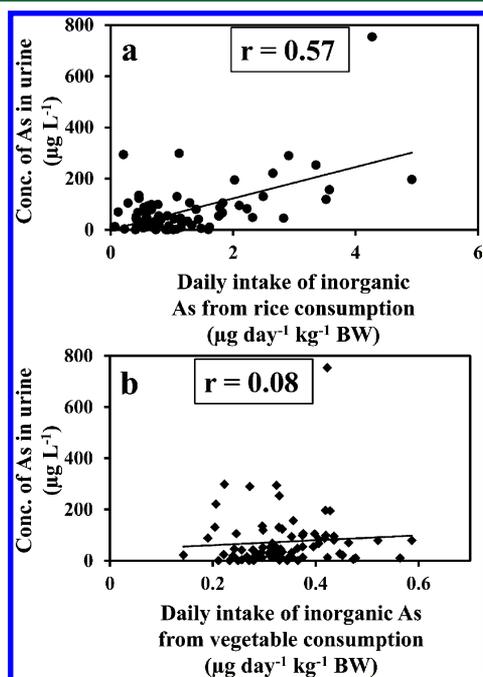


Figure 5. Correlation of urinary As concentration with (a) daily intake of inorganic As from rice consumption (DI-iAs-R) and (b) daily intake of inorganic As from vegetable consumption (DI-iAs-V).

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