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# Arsenic Speciation in Food and Estimation of the Dietary Intake of Inorganic Arsenic in a Rural Village of West Bengal, India

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Arsenic (As) species were quantified by HPLC-HG-AFS in water and vegetables from a rural area of West Bengal (India). Inorganic species predominated in vegetables (including rice) and drinking water; in fact, inorganic arsenic (i-As) represented more than 80% of the total arsenic (t-As) content. To evaluate i-As intake in an arsenic affected rural village, a food survey was carried out on 129 people (69 men and 60 women). The data from the survey showed that the basic diet, of this rural population, was mainly rice and vegetables, representing more than 50% of their total daily food intake. During the periods when nonvegetarian foods (fish and meat) were scarce, the importance of rice increased, and rice alone represented more than 70% of the total daily food intake. The food analysis and the food questionnaires administrated led us to establish a daily intake of i-As of about 170  $\mu$ g i-As day<sup>-1</sup>, which was above the tolerable daily intake of 150  $\mu$ g i-As day<sup>-1</sup>, generally admitted. Our results clearly demonstrated that food is a very important source of i-As and that this source should never be forgotten in populations depending heavily on vegetables (mainly rice) for their diet.

KEYWORDS: Inorganic arsenic; total arsenic; West Bengal; rice; water

# INTRODUCTION

Contamination of groundwater with arsenic (As) and the impact of this contamination on humans have been reported in 23 countries, but the magnitude of this problem is especially severe in Bangladesh and West Bengal (India). The origin of As in groundwater in the Bengal delta is geological, and there are three main pathways for its release from soil: (1) oxidation of As bearing pyrites, (2) reduction of As rich oxy-hydroxides, and (3) increased use of phosphorus fertilizers (1). West Bengal is composed of 18 districts, and in half of them (South 24-Parganas, North 24-Parganas, Howrah, Hooghly, Nadia, Bardhaman, Murshidabad, Malda, and South Calcutta), groundwater has a concentration above 0.05 mg L<sup>-1</sup>. Approximately 42.7 million people live in these 9 districts. This does not mean that all are drinking arsenic-contaminated water and will suffer from arsenic toxicity (2); however, there is no doubt that they are at risk because the maximum permissible limit of As established by the World Health Organization in 2006 is 0.01 mg  $L^{-1}$  (3). There is strong evidence of elevated As levels in rice grain and vegetables in regions of West Bengal and Bangladesh, where fields have been irrigated with As-contaminated water. Signes et al. (4) reported that the most popular types of commercial rice from West Bengal (paddy, atab, and boiled) have As concentrations as high as  $550 \pm 13 \ \mu g \ kg^{-1}$ ,  $339 \pm 14 \ \mu g \ kg^{-1}$ , and  $507 \pm 52 \ \mu g \ kg^{-1}$ , respectively. Farid et al. (5) reported As contents in different vegetables irrigated with As-contaminated water: amaranth,  $572 \ \mu g \ kg^{-1}$ ; saishim (China shak),  $539 \ \mu g \ kg^{-1}$ ; red amaranth,  $321 \ \mu g \ kg^{-1}$ ; stem amaranth (katua data),  $284 \ \mu g \ kg^{-1}$ ; Indian spinach,  $189 \ \mu g \ kg^{-1}$ ; chilli,  $112 \ \mu g \ kg^{-1}$ ; and potatoes,  $103 \ \mu g \ kg^{-1}$ .

These high As contents in rice and vegetables are indicators of the level of As contamination in the environment. In order to asses the risk posed by As in the diet, however, As speciation must be ascertained because methylated organic As (o-As; monomethyl arsenic acid, MA; dimethyl arsenic acid, DMA) is much less toxic than inorganic As (i-As: arsenate and arsenite) (6), which has been classified by the International Agency for Research on Cancer (IARC) as carcinogenic to humans (7). In addition, to realistically asses the risk posed by As in the diet it is important to remember that the As concentration in food tends to rise when it is cooked (8-10). In As-endemic areas, the high As content in water used for cooking purposes

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column guard column mobile phase 1 injection volume flow rate	HPLC Hamilton PRP-X100 Hamilton PRP-X100 10 mM K <sub>2</sub> HPO <sub>4</sub> /KH <sub>2</sub> PO <sub>4</sub> adjusted to pH 6.0 (isocratic) 50 μL 0.8 mL min <sup>-1</sup>
reducing agent flow rate of reducing agent HCI HCI flow rate carrier gas carrier gas flow rate hydrogen flow rate resonance wavelength	HG-AFS 1.4% (w/v) NaBH <sub>4</sub> in 0.4% (w/v) NaOH 1.0 mL min <sup>-1</sup> 1.5 M 1.5 mL min <sup>-1</sup> argon 200 mL min <sup>-1</sup> 60 mL min <sup>-1</sup> 193.7 nm

is a further source of contamination, mainly as i-As. This is indicated by the high total As (t-As) contents found in the studies on t-As contents in cooked foods from As-contaminated areas (11-14).

As rice is cultivated in As-contaminated soils under anaerobic conditions (at which As is highly available for plant uptake), the As concentration in rice is high compared with that of other crops (15). This agrees with Signes et al. (1) who reported that in West Bengal soils maximum soluble As and iron concentrations were found under anaerobic conditions at the interval [-50, 150] mV.

There are several studies on the evaluation of As daily intake of the population of West Bengal, and in all of them, rice is an important source of As; rice is the background of the diet of the West Bengal population and represents about 70% of the total daily calories (6). Roychowdhury et al. (2) reported that rice contributes over 90% of the total intake of As from food composites (rice, vegetables, and spices) by population of Jalangi and Damkal blocks in West Bengal.

This study is included within a project of the European Union Asia Pro Eco Programme called Development of a low cost technology for in situ treatment of ground water for potable and irrigation purposes (*16*). Its aim was to determine the t-As and As speciation (arsenite, arsenate, MA, and DMA) contents in rice and vegetables coming from As alert zones in Eastern India and evaluate the daily intake of t-As and i-As of the inhabitants of an As-affected rural village of West Bengal.

#### MATERIAL AND METHODS

Study Area. The study was conducted in Kasimpur. This village is located around 5 km north of Barasat in North 24-Parganas district, approximately 25 km from Calcutta (India). The total area of the village is 5.0 km<sup>2</sup>, with 22270 people living in the residential area of the village, with the remaining area being cultivated land. The average annual income of the villagers is 350 US\$ annum<sup>-1</sup>. The main source of water for drinking and irrigation purposes in the village is 100 wells and tube wells, mainly shallow. The village was chosen as the model village in our study because 70% of its tube wells, which are the main source of water for drinking and irrigation, had As concentrations above 50  $\mu$ g L<sup>-1</sup>. It is important to mention that the WHO limit for As in drinking water was established at 10  $\mu$ g L<sup>-1</sup> (3); after a risk assessment conducted in 2003, this guideline value was designated as provisional in view of the scientific uncertainties.

**Food Questionnaire.** A mixture of weekly purchase, observations, and 24-h recall food questionnaire was administered to several homes in the studied village. The design of the questionnaire was carried out by Leiden University (Netherlands) and approved by Ramakrisma

Vivekananda Mission (West Bengal), a registered society (NGO) with expertise in working with villagers in the studied area. The questionnaire was administered by professionals from this society. The number of interviewees was set at 129 (69 men and 60 women). The people interviewed were between 12 and 60 years of age, and their marital status was as follows: 27% single, 68% married, and 5% widowed. The selected interviewees were mainly farmers and housewives who normally eat at home, buy or obtain their food from local markets and/ or farms, cook themselves, and take drinking water from the surrounding tube wells. People working at nearby cities were not included in the survey because they are not representative of the endemic As area (they frequently eat in the city).

Instrumentation. For As speciation analyses, a high performance liquid chromatography (HPLC) system consisting of a Varian 9012 ternary pump (Varian, San Fernando, CA, USA), a Rheodyne 7125 injector, and a 50 mL loop for sample introduction was used. Separations of As species were performed on a Hamilton PRP X-100 anion-exchange column (10  $\mu$ m, 250 mm  $\times$  4.1 mm i.d; Hamiton, Reno, NV, USA). A guard column packed with the same material (12-20  $\mu$ m; 25 mm  $\times$  2.3 mm i.d) preceded the analytical column. Hydride generation of volatile arsines before the detection was performed by way of adding online solutions of HCL and NaBH4 using a Gilson Minipuls 3 peristaltic pump. The quantification of As was performed on a hydride generation system (PSA 10.044, PS Analytical, Kent, UK) using an atomic fluorescence spectrometer system (AFS) (PSA 10.044 Excalibur, PS Analytical) equipped with a boosted-discharge hollow cathode lamp (Photron Pty. Ltd., Victoria, Australia). The analog signal output was connected to a computer equipped with chromatographic software (PS Analytical).

Determination of t-As was performed with a Unicam Model Solaar 969 atomic absorption spectrometer equipped with a continuous hydride generator Unicam Solaar VP90 (AAS-HG).

Other equipment used included a hot air oven (Selecta, Barcelona, Spain) with a maximum temperature of 250 °C, a grinder (Molinex, Valencia, Spain), a mechanical shaker Vibromatic (J.P Selecta S.A., Barcelona, Spain), a centrifuge (Heraeus BioFuge, Heraeus Instruments, Hanau, Germany), and a lyophilizer (B. Biotech International, Christ Alpha 2-4, Osterode, Germany).

**Reagents.** Deionized water (18 M $\Omega$  cm) was used for the preparation of the reagents and standards. All glassware were treated with 10% (v/v) of HNO<sub>3</sub> for 24 h and then rinsed three times with deionized water before use. All chemicals were at least of proanalysis quality. Commercial standards of NaAsO<sub>2</sub> (sodium meta-arsenite) and Na<sub>2</sub>HAsO<sub>4</sub> 7H<sub>2</sub>O (sodium hydrogen arsenate) were obtained from Panreac (Barcelona, Spain), while CH<sub>4</sub>AsNaO<sub>3</sub> 1.5H<sub>2</sub>O (monosodium methylarsinate sesquihydrate, MA) and (CH<sub>3</sub>)<sub>2</sub>AsO(ONa) 3H<sub>2</sub>O (monosodium dimethylarsinate trihydrate, DMA) were from Supelco (Bellefonte, PA, USA) and Fluka (Buchs, Germany), respectively. Finally, anhydrous trifluoroacetic acid (TFA) was from Sigma (St Louis, MO, USA).

**Food Samples.** Raw vegetables (carrot, kidney bean, radish, tomato, onion, betel nut, cauliflower, brinjal, and potato), commercial rice [paddy, atab (dehusked), boiled (parboiled), and puffed (4)] and spices (coriander seeds, turmeric powder, cumin seeds, mustard, ginger mango, and fenugreek seeds) were collected in farms and local markets surrounding the previously cited rural village. Samples were sun-dried in India until constant weight was obtained and sent to Spain for analyses. All samples were analyzed for t-As and As-species.

**Quantification of Total Arsenic.** A 0.5 g portion of the dried food sample was weighed and digested using the ashing method previously described by Muñoz et al. (17). Calibration standards were prepared using the same HCl concentration of the samples and certified materials. The instrumental conditions used for As determination by HG-AAS were as follows: reducing agent, 1.4% (m/v) NaBH<sub>4</sub> in 0.4% NaOH, 5 mL min<sup>-1</sup>; HCl solution, 10% (v/v) and 10 mL min<sup>-1</sup>; and carrier gas, argon, 250 mL min<sup>-1</sup> flow rate. For atomic absorption spectrometry, wavelength, 193.7 nm; spectral band-pass, 0.5 nm; hollow cathode lamp current setting, 8 mA; and air/acetylene flame with a fuel flow rate of 0.8 L min<sup>-1</sup>.

The certified reference materials (rice flour = NIST SRM 1568a; and bush, branches and leaves = GBW07603) that were used for testing

this analytical method were provided by CYMIT Química, S.L. (Barcelona, Spain) and produced by the US National Institute of Standards and Technology and the Institute of Geophysical and Geochemical Exploration of China, respectively.

Quantification of Arsenic Species. The method described by Heitkemper et al. (18) was used for the extraction of the As species. A dried and milled food sample (0.5 g) was treated with 3 mL of 2 M TFA. The mixture was allowed to stand for 6 h at 100 °C in a 60 mL capped HDPE centrifuge tube. The mixture was then centrifuged and the supernatant collected and diluted to volume with deionized water. The TFA extracts were filtered through a 0.45 mm nylon syringe filter before analysis by HPLC-HG-AFS. The As species (arsenite, arsenate, MA, and DMA) were determined in the water extract using HPLC-HG-AFS. Separation of the As compounds was carried out in about 15 min in the anion-exchange column using 25 mM phosphate buffer (pH 6.0) as the mobile phase at a 1.1 mL min<sup>-1</sup> flow rate. The elution order was arsenite, DMA, MA, and arsenate. A total of 50  $\mu$ L of the sample was injected in the HPLC system following the instrumental and analytical conditions described in Table 1. Under these conditions, the retention times were 3.15, 4.24, 5.07, and 8.75 min for arsenite, DMA, MA, and arsenate, respectively. Figure 1 shows the separation obtained in (1) a 50  $\mu$ g L<sup>-1</sup> standard of arsenite, DMA, MA, and arsenate; (2) ginger mango; (3) radish root; and (4) kidney bean. External calibration was accomplished using standards concentrations of 1, 10, 20, 30, 40, and 50  $\mu g L^{-1}$  of each of the four As species studied (arsenite, DMA, MA, and arsenate). Information on the certified NIST SRM 1568a (rice flour) and GBW07603 (bush, branches and leaves) was used to assess the precision and accuracy of this analytical method.

### **RESULTS AND DISCUSSION**

**Analytical Quality Assurance.** *Total As.* The analytical characteristics of the t-As methodology were as follows: detection limit,  $7 \mu g \text{ kg}^{-1}$ ; precision 2%; accuracy for rice flour (NIST SRM 1568a), found value =  $0.29 \pm 0.04 \text{ mg kg}^{-1}$  (certified value =  $0.29 \pm 0.03 \text{ mg kg}^{-1}$ ); and accuracy for bush branches and leaves (GBW07603), found value =  $1.20 \pm 0.03 \text{ mg kg}^{-1}$ .

As Speciation. An estimate of the instrumental detection limit (IDL) for each of the four As species was calculated on the basis of three times the standard deviation of peak area measurement for replicate 50 µL injections of an As standard containing 2.0  $\mu$ g L<sup>-1</sup> of arsenite, arsenate, MA, and DMA. The IDL estimates were 1.7, 1.5, 1.1, and 1.4  $\mu$ g L<sup>-1</sup>, respectively. Finally, estimates of the method detection limits (MDLs) were calculated using the IDLs multiplied by a dilution factor of 10 (0.5 g rice diluted to a final volume of 5 mL). Experimental MDLs were 17, 15, 11, and 14  $\mu$ g As kg<sup>-1</sup> fresh matter for arsenite, DMA, MA, and arsenate, respectively. The average fortification recoveries (for a 0.75  $\mu$ g spike on 10 mL of a 20  $\mu$ g L<sup>-1</sup> standard) through the method were 94, 89, 90, and 92% for arsenite, DMA, MA, and arsenate, respectively. No significant reduction/oxidation reactions were observed in these fortification studies.

**Total Arsenic. Table 2** shows Indian food samples from the North 24-Parganas district, where As-contaminated water is regularly used for watering crops. The analyzed foods were classified into three different groups: vegetables, grains (including rice), and spices. Significant As concentrations were found in all samples analyzed, and in most cases, these concentrations were high. The food with the highest As concentration was rice, ranging from  $120 \pm 10$  and  $496 \pm 17 \ \mu g \ kg^{-1}$  and presenting a mean value of  $339 \ \mu g \ kg^{-1}$ . Samples of bulbs and roots, such as carrot, radish, potato, and ginger mango showed high As concentrations as well. These experimental data agree with previous studies in which it was established that as rice is cultivated in As-contaminated soils under anaerobic conditions



**Figure 1.** Examples of experimental chromatograms of As speciation: (A) standard of 50  $\mu$ g L<sup>-1</sup> of arsenite, DMA, MA, and arsenate (in order of elution); (B) ginger mango; (C) radish; (D) kidney beans.

(at which As is highly available for plant uptake), the As concentration in rice is higher compared with that in other crops (1, 15, 19). The mean values for the vegetable and spice groups were 75 and 91  $\mu$ g kg<sup>-1</sup>, respectively.

Carbonell et al. (20) and Lario et al. (21) showed that the largest quantities of As residues in plants were found in roots, with above-ground vegetable parts (leaves and stems) presenting intermediate values and the lowest contents being found in fruits and seeds. The data obtained in the present study agree with this statement because roots and bulbs contained the highest As concentrations detected; the only exception to it was rice because of its special growing conditions (flooding and low soil redox potentials).

Arsenic Speciation. Table 3 shows the concentrations of the four As species under study in Indian vegetables, rice, and spices, all of them from local markets and farms. These analyses demonstrated that As was mainly present as inorganic forms, basically arsenite because arsenate was not detected in any sample. Only a few samples had a low percentage of i-As, for

 Table 2. Total Arsenic Concentration in Vegetables, Cereals, and Spices from Kasimpur (West Bengal) Analyzed by AAS-HG

samples	total arsenic ( $\mu { m g~kg^{-1}}$ ) mean $\pm$ SE
Vegeta	bles (dw) <sup>a</sup>
carrot	121 ± 2
kidney beans	$41 \pm 2$
radish	$167\pm5$
tomato	$56\pm4$
onion	$55\pm5$
betel nut	$34\pm4$
cauliflower	$70 \pm 12$
brinjal	$53\pm11$
potatoes	$80\pm7$
Cere	als (dw)
paddy rice	496 ± 17
atab rice	$271\pm20$
boiled rice	$469\pm 6$
puffed rice	$120 \pm 10$
Spic	ces (dw)
coriander seeds	89 ± 2
turmeric powder	78 ± 7
cumin seeds	$84\pm2$
mustard	$50\pm5$
ginger mango	$214\pm8$
fenugreek seeds	$29\pm7$

<sup>a</sup> Concentrations are based on dry weights.

Table 3.	Arsenic S	Speciation in	n Vegetabl	es, Cereals,	and Spices	Samples
from Kasi	impur (We	est Bengal)	Analyzed	by HPLC-H	G-AFS	

	arsenic speciation ( $\mu$ g kg <sup>-1</sup> )								
	inorganic	arsenic	organic	arsenic					
samples	arsenite	arsenate	MA	DMA	total As				
	Vegetables (dw) <sup>a</sup>								
carrot	$90\pm7$	nd <sup>b</sup>	$22 \pm 11$	nd	$112\pm10$				
kidney beans	$42 \pm 1$	nd	n.d	nd	$42\pm3$				
radish	$94\pm12$	nd	$60\pm7$	nd	$154\pm11$				
tomato	$54\pm5$	nd	n.d	nd	$54\pm5$				
onion	$56\pm 6$	nd	n.d	nd	$56\pm 6$				
betel nut	$26 \pm 5$	nd	n.d	nd	$26\pm5$				
cauliflower	$60\pm9$	nd	n.d	nd	$60\pm9$				
brinjal	$48 \pm 2$	nd	n.d	nd	$48 \pm 2$				
potatoes	$47 \pm 2$	nd	$34 \pm 7$	nd	$81\pm5$				
		Cereals (dv	N)						
paddy rice	$243\pm10$	nd	nd	$242\pm12$	$488 \pm 11$				
atab rice	$121 \pm 10$	nd	nd	$147\pm22$	$268\pm15$				
boiled rice	$380\pm11$	nd	nd	$91\pm9$	$471\pm10$				
puffed rice	$40\pm2$	nd	nd	$80\pm4$	$120\pm3$				
		Spices (dv	v)						
coriander seeds	$41\pm9$	nd	$35 \pm 11$	nd	$76\pm10$				
turmeric powder	$24\pm7$	nd	$47\pm3$	nd	$71\pm5$				
cumin seeds	$76\pm8$	nd	n.d	nd	$76\pm8$				
mustard	$48\pm5$	nd	n.d	nd	$48\pm5$				
ginger mango	$148\pm5$	nd	$60\pm7$	nd	$208\pm5$				
fenugreek seeds	$5\pm1$	nd	$23\pm3$	nd	$28\pm2$				

<sup>a</sup> Dry weight. <sup>b</sup> nd, not detected.

instance, fenugreek seeds 18%, paddy rice 50%, atab rice 44%, and puffed rice 33%. All the other samples presented i-As percentages above 50%. Puffed rice had the lowest percentage of i-As of the types of rice studied, perhaps due to its manufacturing process, where heating could have induced transformation of the initial arsenite into DMA (however, more research is needed to support this statement). Paddy and atab rice had similar percentages of i-As because As is distributed homogenously between the husk and grain (4). Boiled rice, however, had a high percentage of i-As because the water used to cook it usually has high concentrations of i-As.

 
 Table 4. Average Daily Intake of Food (on a Fresh Basis) According to the Food Questionnaire Administrated in Kasimpur (West Bengal)

	mean food intake (mL or g day $^{-1}$ )						
food group	men ( <i>n</i> = 69)	women ( <i>n</i> = 60)					
water	1761	1765					
rice	385	362					
vegetables	205	182					
fruits	10	13					
poultry meat	28	27					
fish	20	22					
milk	66	60					
eggs	9	9					
bakery products	25	23					
spices	3	2					

From a toxicological point of view, this experiment shows that the worst possible scenario is happening in rural villages of West Bengal because the majority of As present in the studied foods was present as the most toxic species (arsenite), which is about 60 times more toxic than arsenate, while o-As is nontoxic for humans (22).

**Table 4** shows the estimated average of the daily intake of food of the rural population of West Bengal. These data demonstrated that the most important food for the rural population of West Bengal was rice, which represented more than 51% of the total daily food intake and more than 70% of the total daily caloric intake (6). After rice, vegetables were the most important food, representing about 21% of the total daily food intake. However, none of the other food items included in **Table 4** represents more than 10% of the total daily food intake.

**Figure 2** demonstrates that the importance of rice for the rural populations of West Bengal increases when the daily food intake is minimal. Under circumstances of food shortage, rice becomes the most important food and represents about 70% of the daily food intake, and at the same time, it becomes the largest source of both t-As and i-As. During times when vegetables, milk, and bakery products are more abundant, the importance of rice decreases and represents about 35% of daily food intake.

Estimation of Inorganic As Intake and Evaluation of Risks. The intakes of t-As and i-As were calculated and expressed as  $\mu g$  As day<sup>-1</sup> (**Table 5**) from the data provided by the food questionnaire administered to inhabitants of Kasimpur regarding the quantity of water (mL) and food (g) consumptions and also from the t-As and i-As concentrations from Tables 1 and 2. Data included in Table 5 were calculated on a fresh weight basis. The contribution of rice to the total intake of As from foods, other than drinking water, ranged between 46 to 60%. The highest percentage was found when food consumption was low, and the lowest percentage was found when food consumption was high. From data in Table 5, it seems evident that vegetables play a minor role in t-As and i-As daily intakes of this rural population, with values being close to 0-1%. The low importance of vegetables in As daily intake was mainly due to the fact that vegetables have a high water content (approximately 90%). However, the low water content of rice and its special growing conditions leading to high As accumulation in grains makes rice a significant source of As in the daily intake of villagers.

There is overwhelming evidence from epidemiological studies that consumption of elevated levels of arsenic through drinking water is causally related to the development of cancer at several sites, particularly the skin, bladder, and lung (3). Inorganic As compounds are classified by IARC in group 1 (carcinogenic to



Figure 2. Total daily intake of foods (%) by population of Kasimpur (West Bengal) under circumstances of food shortage (gray columns) and food abundance (black columns).

Table 5.	Dailv	Dietarv	Intake c	of Arsenic	(from	Water.	Rice.	and	Vegetables)	) bv	/ the	Population	of	Kasimpu	r (West	t Bena	al) <sup>a</sup>
					\ -	,	,										

	arsenic (µg kg	<sup>-1</sup> or $\mu$ g L <sup>-1</sup> )	AslWR&V ( $\mu$ g As day <sup>-1</sup> ) <sup>b</sup>		DAsI (µg As	s day <sup>-1</sup> ) <sup>b</sup>			
type of food	As(III) + As(V)	MA + DMA	$\overline{As(III) + As(V)}$	MA + DMA	As(III) + As(V)	MA + DMA	TDAsI ( $\mu$ g As day $^{-1}$ ) $^{b}$		
water <sup>†</sup>	50		36						
rice <sup>†</sup>	167	119	33	24					
vegetables <sup>†</sup>	6	4	0	0					
total <sup>†</sup>	223	123	69	24	69	24	93		
water <sup>‡</sup>	50		88						
rice <sup>‡</sup>	167	119	62	44					
vegetables <sup>‡</sup>	6	4	1	1					
total‡	223	123	151	45	151	45	196		
water§	50		167						
rice§	167	119	83	60					
vegetables§	6	4	1	1					
total§	223	123	251	61	251	61	312		

<sup>a</sup> This estimation was done for three different scenarios, when daily food intake was at (a) its minimum (<sup>†</sup>), (b) its mean (<sup>‡</sup>), and (c) its maximum (<sup>§</sup>). <sup>b</sup> AsIWR&V, Arsenic intake through water, rice, and vegetables; DAsI, daily arsenic intake; TDAsI, total daily arsenic intake; AsIDW, arsenic intake through drinking water.

humans) on the basis of sufficient evidence for carcinogenicity in humans and limited evidence for carcinogenicity in animals (3). The parameter most commonly used for the evaluation of As risk assessment is the Provisional Tolerable Weekly Intake (PTWI) established by FAO/WHO: 15  $\mu$ g i-As week<sup>-1</sup> kg<sup>-1</sup> body weight (23); it is necessary to stress that there is a narrow margin between this PTWI and intakes reported to have toxic effects in epidemiological studies. The provisional status of the maximum weekly intake was meant to convey the desirability of seeking ways of reducing the As intake of populations exposed to naturally elevated levels of i-As in drinking-water (23). The PTWI value can be transformed into the Tolerable Daily Intake (TDI) of i-As as follows: TDI = PTWI/7 days = 124  $\mu$ g i-As day<sup>-1</sup> for adults, if we assumed that adults in West Bengal have a mean body weight of approximately 58 kg.

In the present study, the daily As intake obtained from just water, rice, and vegetables (no fish, meat, milk, or eggs were included) was 196  $\mu$ g As day<sup>-1</sup>, from which 151  $\mu$ g As day<sup>-1</sup> was i-As. This daily i-As intake was higher than the recommended TDI (124  $\mu$ g i-As day<sup>-1</sup>). Data in **Table 5** shows that under circumstances of minimal food consumption, i-As intake was lower than the TDI (93  $\mu$ g As day<sup>-1</sup>), while when food is abundant, i-As intake was 312  $\mu$ g As day<sup>-1</sup>, approximately 2.5 times higher than the TDI. In addition, for an overall estimation of i-As intake, it is important to keep in mind that all data obtained in this study came from raw food samples, and it is probable that their final As content will increase after cooking, especially if As-contaminated water is used (*11–14*).

Conclusions. Our results demonstrated that rice was the food stuff with the highest As concentration (between  $120 \pm 10$  and  $496 \pm 17 \ \mu g \ kg^{-1}$ ), although bulbs and/or root samples, such as carrot, radish, potato, and ginger mango showed high As concentrations as well (between  $47 \pm 2$  and  $148 \pm 5 \,\mu g \, kg^{-1}$ ). Arsenic was mainly present in inorganic forms, basically all arsenite, which is the worst possible scenario because arsenite is the most toxic As species for humans. The most important food for the rural West Bengal population was rice, which represented more than 51% of total daily food intake and could reach up to 70% under food shortage circumstances. Since rice had the highest arsenite concentration, and it is the most abundant/popular food, it represented a high percentage (54%) of the total daily arsenite intake from water, rice, and vegetables. Daily arsenite intake from water, rice, and vegetables will be lower than the tolerable daily intake (TDI) when there is food scarcity, but it will be much higher, approximately 2.5 times, than the TDI when food is abundant. Finally, it is important to mention that only raw materials were analyzed and that the described As concentrations will increase during the cooking process if As-contaminated water is used.

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