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Arsenic exposure from food exceeds that from drinking water in endemic area of Bihar, India

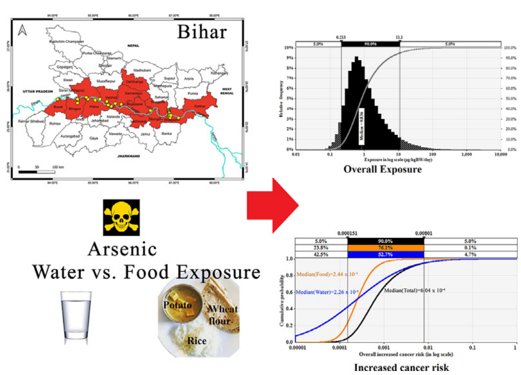
Debapriya Mondal [a,*], Mohammad Mahmudur Rahman [b], Sidharth Suman [a,c,d], Pushpa Sharma [c,d], Abu Bakkar Siddique [b], Md. Aminur Rahman [b], A.S.M. Fazle Bari [b], Ranjit Kumar [c], Nupur Bose [e], Shatrunjay Kumar Singh [d], Ashok Ghosh [c], David A. Polya [f]

[a] *School of Science, Engineering & Environment, University of Salford, Salford M5 4WT, UK*
[b] *Global Centre for Environmental Remediation (GCER), Faculty of Science, The University of Newcastle, Callaghan, NSW 2308, Australia*
[c] *Mahavir Cancer Institute and Research Center, Patna, India*
[d] *Department of Environment and Water Management, A.N. College, Patna, India*
[e] *Department of Geography, A.N. College, Patna, India*
[f] *Department of Earth and Environmental Sciences, Williamson Research Centre for Molecular Environmental Science, University of Manchester, Manchester M13 9PL, UK*

HIGHLIGHTS

- Food contributes equally as drinking water towards total As exposure in Bihar, India.
- Cooked rice was the most significant As contributor to food exposure.
- With increase in As concentrations in drinking water As exposure from food increased.
- Median excess lifetime cancer risk of 2 per 10,000, from food As exposure in Bihar.

GRAPHICAL ABSTRACT



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ABSTRACT

Extensive evidence of elevated arsenic (As) in the food-chain, mainly rice, wheat and vegetables exists. Nevertheless, the importance of exposure from food towards total As exposure and associated health risks in areas with natural occurring As in drinking water is still often neglected, and accordingly mitigations are largely focused on drinking water only. In this study, the contribution of food over drinking water to overall As exposure was estimated for As exposed populations in Bihar, India. Increased lifetime cancer risk was predicted using probabilistic methods with input parameters based on detailed dietary assessment and estimation of As in drinking water, cooked rice, wheat flour and potato collected from 91 households covering 19 villages. Median total exposure was 0.83 μg/kgBW/day (5th and 95th percentiles were 0.21 and 11.1 μg/kgBW/day) and contribution of food (median = 49%) to overall exposure was almost equal to that from drinking water (median = 51%). More importantly and contrary to previous studies, food was found to contribute more than drinking water to As exposure, even when drinking water As was above the WHO provisional guide value of 10 μg/L. Median and 95th percentile excess lifetime cancer risks from food intake were 1.89×10^{-4} and 7.32×10^{-4} respectively when drinking water As was below 10 μg/L and 4.00×10^{-4} and 1.83×10^{-3} respectively when drinking water As was above 10 μg/L. Our results emphasise the importance of food related exposure in As-endemic areas, and, perhaps surprisingly,

* Corresponding author.
E-mail address: d.mondal@salford.ac.uk (D. Mondal).

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particularly in areas with high As concentrations in drinking water – this being partly ascribed to increases in food As due to cooking in high As water. These findings are timely to stress the importance of removing As from the food chain and not just drinking water in endemic areas.

1. Introduction

The severity of arsenic (As) contamination in the state of Bihar, located in the eastern region of India, next to West Bengal is well acknowledged (Chakraborti et al., 2003; Chakraborti et al., 2016a; Chakraborti et al., 2016b; Chakraborti et al., 2017; Kumar et al., 2016c; Richards et al., 2020; Saha, 2009). Out of 38 districts (the highest administrative division in a state), 22 were reported to have As in drinking water above the WHO provisional guide value of 10 μg/L (Chakraborti et al., 2018). More than 9 million people were estimated to be drinking water with arsenic above 10 μg/L and 33% of the tested hand tube wells samples ($n = 19{,}961$), normally used for drinking water had As above the WHO provisional guide value (Chakraborti et al., 2017). In a previous study by Nickson et al. (2007) covering 11 districts of Bihar, 29% ($n = 66{,}623$) of drinking water samples, were reported to have As concentration higher than 10 μg/L. Many of these studies focussed either exclusively or to a large extent on areas of Bihar close to the Ganga River. In a recent study, systematically covering all 38 districts of Bihar, just 16% of the samples ($n = 273$), were found to have As above the concentration of 10 μg/L (Richards et al., 2020). These differences likely reflect differences in sampling frame, successful mitigations and public education/awareness interventions by various agencies, including the Government of Bihar, over the last decade. Nevertheless, all these studies indicate that a majority of sampled sources might have As lower than 10 μg/L.

Consumption of As contaminated food can be a relatively important route of exposure (Mondal et al., 2010; Mondal et al., 2019; Mwale et al., 2018). However, estimates of overall As exposure from food in Bihar exposed populations is sparse: only three studies to date in international journals have reported As concentrations in food items from rural Bihar: (a) Kumar et al. (2016b) reported mean total As concentrations of 51 ± 41 μg/kg ($n = 15$); 27 ± 24 μg/kg ($n = 35$); 13 ± 8.4 μg/kg ($n = 31$); 23 ± 15 μg/kg ($n = 6$) and 452 ± 712 μg/kg ($n = 34$) in raw rice, wheat, maize, green gram and vegetable samples, respectively, collected from households in the Samastipur district of Bihar, India; (b) we have reported total As in wheat grain (44 ± 48 μg/kg, $n = 72$) and wheat flour (50 ± 74 μg/kg, $n = 58$) samples collected from As exposed districts of Bihar (Suman et al., 2020) and (c) Singh and Ghosh (2011) reported highest concentrations of total As in wheat grains (24 μg/kg), followed by rice grains (19 μg/kg) and lentils (15 μg/kg) in a single food sample ($n = 1$) collected from farmers of Rampur Diara in Maner block of Patna district. In a laboratory-based study, Mandal et al. (2019) estimated risks associated with consumption of wheat and maize grown experimentally under different conditions in Bihar soils. These studies are either focussed on a particular aspect of exposure or localised and limited by small sample sizes.

To more fully understand the As exposure from food in As exposed populations of Bihar, India, in this study, we conducted a detailed dietary assessment of adult participants, male and female from each household using 24-h recall and collected food samples from 91 households covering 19 villages widely distributed in eight known As affected districts of Bihar. We focused on rice, wheat and potato since cereals, followed by vegetables and milk, constitute a major share in the diet of the rural populations of India (Gupta and Kumar, 2015). In rural Bihar, consumption of wheat (68.7 kg/capita/year in 2011–12) is second only to rice (75.4 kg/capita/year in 2011–12) (Kumar et al., 2016a).

The notion that As exposure from food is equally important as that from drinking water in areas, such as in India and Bangladesh, with elevated geogenic As in drinking water is sparsely recognised (Huq et al., 2006; Kile et al., 2007; Kumar et al., 2016b; Mondal and Polya, 2008; Mondal et al., 2010; Suman et al., 2020) in spite of numerous studies showing high total As and more specifically inorganic As (iAs) in the food-chain (Santra et al., 2013; Zhao et al., 2010). This is illustrated by the fact that As mitigations are by far focused towards lowering exposure from drinking water rather than from food (Huq et al., 2006; Mondal et al., 2014). In Bangladesh, elevated childhood exposure to As is found despite reduced drinking water concentrations (Kippler et al., 2016). It is therefore timely to consider the relative importance of As exposure from food over drinking water in an area, such as Bihar, India, where exposure from drinking water is still widespread. Hence, we determined the relative contribution of food over drinking water towards total iAs exposure and excess lifetime cancer risk. We focused on (a) cooked rice rather than raw rice since cooking can either increase or decrease the As concentration of rice depending on cooking method, rice variety and As concentration of cooking water (Mwale et al., 2018; Mondal and Polya (2008), Raab et al., 2009); (b) wheat flour (which is used to make homemade bread-*chapati*) rather than wheat grain as they were the major forms in which wheat is normally consumed in rural populations of India, particularly in Bihar; and also determined As exposure from (c) potato, the main vegetable consumed throughout the year.

To best of our knowledge, this is the first study where a detailed dietary assessment was conducted on As exposed populations of Bihar, India to estimate the As exposure from the three major staple foods, cooked rice, wheat flour and potato. Furthermore, modelled As-attributable health risks from both drinking water and food and their contributions towards total As exposure have been estimated using probabilistic methods.

2. Materials and methods

2.1. Survey and study area

Drinking and cooking water and food samples including cooked and raw rice, wheat grain and flour, and potato were collected from 91 households in Bihar across eight known As affected districts (Fig. 1) based on Government of Bihar data from 2009 (Phed.bih. Nic.in., 2020) as a part of the project- "Nature and nurture in arsenic induced toxicity of Bihar, India". We visited 19 villages in 2017–19 and in every village 3–6 households were randomly selected for this study (Fig. 1). While the selection of the villages was done based on access, contact and, more importantly, covering a wide range of As contaminations as per Government of Bihar data, the selection of the households was done based on socioeconomic status covering both low and high ones (often the type of house, concrete, mixed and mud was used as the guiding factor). After informed consent was obtained, 24-h recalls along with a detailed food frequency questionnaire (FFQ) was administered with one adult male and one adult female participant from each household along with information on demographic and socioeconomic conditions. The study was conducted in accordance with national and international guidelines for the protection of human subjects and was approved by both the University of Salford Ethics Committee (STR1718–10) and Mahavir Cancer Sansthan Institutional Ethics Committee.

A close-ended FFQ comprised of 25 food items ideally consumed by rural Bihar population was used to collect the data on frequency of food

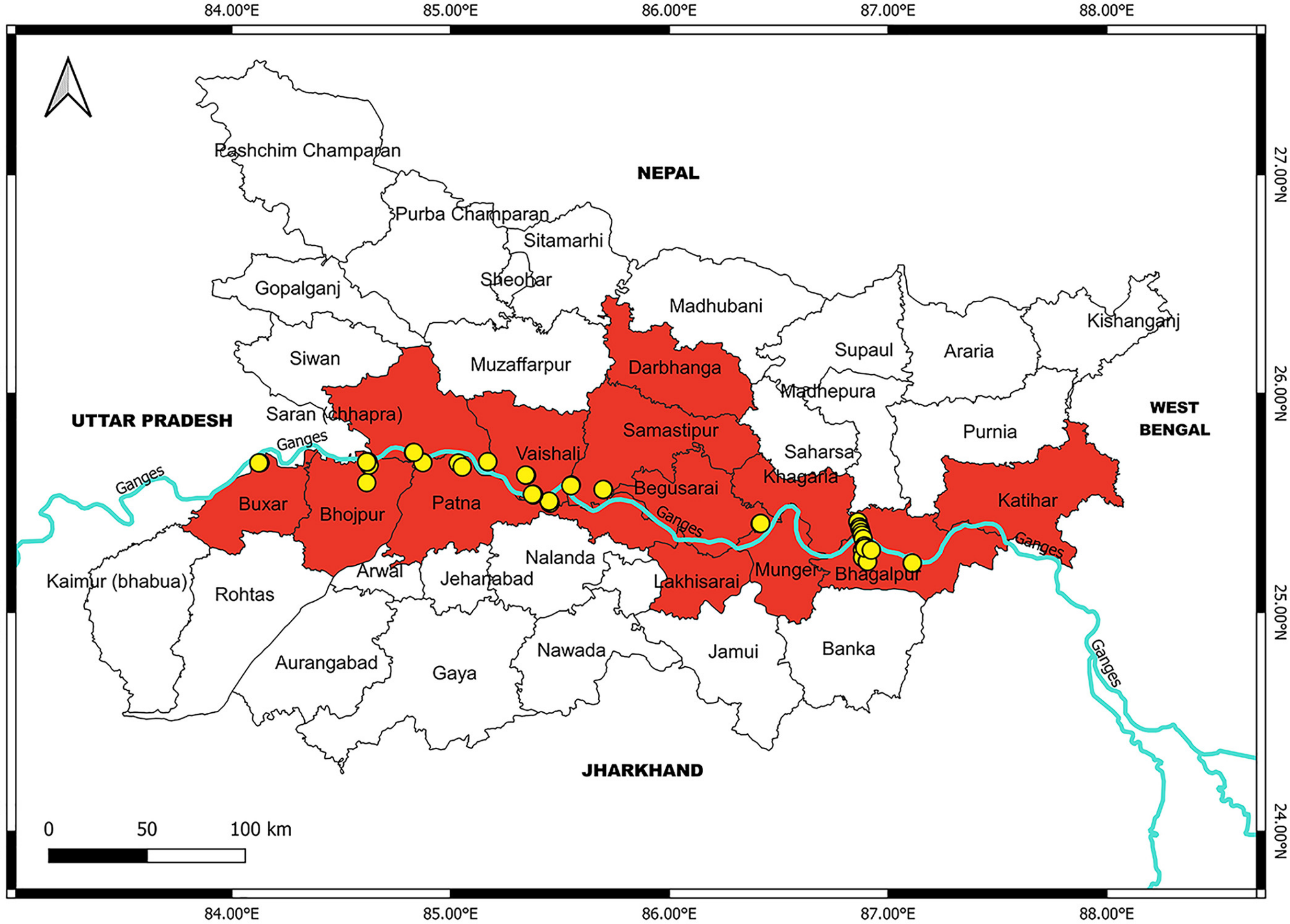

Fig. 1. Sampling locations are shown by yellow dots in this map of Bihar, India, spread over 19 villages in 8 out of 13 As effected districts shown in red as defined by Phed.bih.nic.in. (2020). See Richards et al. (2020) for a more recent coverage of the whole state.

items consumed. The responses were recorded as daily, weekly, occasionally or rarely to determine the food consumption pattern. Generally, in a rural Indian population such as in rural Bihar, food consumption does not vary greatly every day. Therefore the 24-h recall method could provide a reliable indication of actual food intake while FFQ provides the usual food habit (Shim et al., 2014). Those respondents whose food habit was totally different from their actual food intake (due to a special function or occasion) over the last 24h, were asked to give the quantitative details of their usual food intake (dietary recall). Out of 182 respondents, 14 (7.2%) reported of such special occasions and among those nine female respondents who were fasting couldn't provide the dietary recall and hence were excluded. An open-ended 24-h recall was used to collect quantitative data on food consumed in the preceding 24 h starting with the first thing eaten by the participant in the morning until the last food item consumed before the participant went to bed. Using a household item such as a bowl, glass and spoon, from each of the respective households surveyed, the amount of food consumed over the last 24h was recorded, alongside the timing of each meal. The amount of wheat, rice and potato consumed was then estimated for both male ($n = 91$) and female ($n = 82$) participants. Due to the limited number of participants ($n = 35$) being surveyed to determine the intake rate of water, we have included data from published studies in Bihar (Kumar et al., 2016b; Singh et al., 2014; Singh and Ghosh, 2012; Yasmin et al., 2013) and combined with our results to determine the overall water intake rate. Since the published studies had reported the water intake data as a combined value for male and female rather than by gender, we have used a single distribution for both males and females.

2.2. Sample collection and analysis

Drinking ($n = 90$) and cooking water samples, if different to drinking water ($n = 22$) were collected from each household in pre-cleaned polypropylene bottles and transported to the University of Manchester where it was analysed for total As following the protocol as detailed in Richards et al. (2020). Briefly, samples were acidified using 1% analytical grade nitric acid and then filtered through 0.45 μm nylon filters (Fisher) and analysed using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500cx).

All coupled food samples collected from each household: raw rice ($n = 89$); cooked rice ($n = 70$); wheat grain ($n = 82$); wheat flour ($n = 72$); potato ($n = 86$) were stored in plastic zip-lock bags and transported to the laboratory in the Mahavir Cancer Research Centre in Patna. Cooked rice samples were dried in an oven at 80–100 °C and all the remaining food samples were first cleaned in tap water (three times) then with deionised water before drying in an oven at 80–100 °C. Potato samples were chopped into small pieces before drying. All dried samples were ground to powder and were shipped to the University of Newcastle, Australia for total As analysis. To analyse total As in food samples, they were digested using an established protocol (Rahman et al., 2009) and analysed using ICP-MS (Agilent 7900). A portion of dried cooked rice ($n = 23$) and wheat flour ($n = 30$) samples were sent to University of Manchester for iAs estimation. Around 0.5 g of powdered samples were extracted with 5 mL of 0.28 M analytical grade nitric acid using microwave assisted extraction (Mars 5, CEM corporation). The estimation was carried out using ICP-MS (Agilent 7500cx) coupled with an HPLC system (Agilent 1200 series) utilizing a Hamilton PRP-X100 anion exchange column (250 × 4.6 mm) with detection limit of 0.2 to 0.5 μg/L. For quality assurance and quality control, certified reference materials (CRMs), duplicates, blanks and continuing calibration verification (CCV, for total As in food samples) were included in each batch of analysis.

2.3. Probabilistic risk assessment

Probabilistic iAs exposure from food and water (Eq. 1), and the lifetime increased cancer risk (Eq. 2) for the studied population in Bihar, India was estimated based on the USEPA one hit model (USEPA, 1989):

$$Exposure_i = C_i \times Cing_i \times \left(\frac{IR_i}{BW}\right) \quad (1)$$

$$TR = CPS_o \sum_{i=1}^{i=N} C_i \times Cing_i \times \left(\frac{IR_i}{BW}\right) \times BCF_i \times \left(\frac{ED}{LT}\right) \quad (2)$$

where, TR is the excess lifetime cancer risk; CPSo is the oral cancer potency slope factor for iAs; i is potential exposure medium (drinking water, cooked rice, wheat flour or potato); C is the total As concentration; Cing is the proportion of iAs in the subscripted medium; IR is the ingestion rate of the subscripted medium; BW is the body weight of the exposed person; BCF is the bioconcentration factor; ED is the exposure duration; LT is the life expectancy of the exposed person.

2.4. Data analysis

For all variables, descriptive statistics and point estimates: mean ± standard deviation, median, range (minimum and maximum) and interquartile range (IQR) represented by 25th and 75th percentiles were determined. Percentage change in total As concentration of cooked rice over raw rice due to cooking was determined for all paired samples and to visualise the trend in the data, scatter plot was used. One-way analysis of variance (ANOVA) was used, to determine the differences in food intake between male and female participants and the nonparametric Kruskal-Wallis H test was used to determine any significant differences in As concentrations in food items associated with drinking water with less than and more than 10 μg/L of As.

Probability distribution of input variables was characterized by fitting different distributions to each variable using the complete range of data. Intake rates and age distribution were stratified by gender. The goodness of fit was assessed using Akaike's information criterion (AIC) values. Monte Carlo simulations were run and after 100,000 iterations, the output was rendered as cumulative probability and/or relative frequency. Finally, overall excess lifetime cancer risk was estimated taking into account for the gender distribution of Bihar and respective life expectancies.

Data was analysed using Stata 11.2 and Microsoft Excel for Windows for descriptive analysis and for comparisons. Probability distributions for input variables were determined and simulations were run with the software @Risk (version 7.6, Palisade Corp., USA) in combination with Microsoft Excel for probabilistic exposure and risk estimation.

3. Results and discussion

3.1. Analytical quality control data

Percentage recoveries of CRMs used during analysis of different batch of samples are shown in Table 1.

Table 1
Percentage recovery of As in different CRMs and percentage variation in replicates analysed.

CRM **analysis**		
Sample type	N	mean % recovery ± SD
% Recovery of total As (analysis of food samples)[a]		
Spinach Leaf (1570a)	15	104 ± 14
Rice flour (NIST 1568b)	16	95 ± 6
% Recovery of inorganic As (analysis of food samples)[b]		
Rice flour (NIST 1568b)[c]	12	79 ± 12
% Recovery of total As (analysis of water samples)[b]		
Surface water - Trace metals (SPS-SWS1)	2	105
Hard drinking water - Metals (ERM-CA011)	2	105
Trace Elements in Natural Water (NIST-1640a)	2	100
Variation in duplicates		
Sample type	N	Mean % Variation[d]
Food samples [a]	34	12 ± 12
Water samples [b]	6	32 ± 27

[a] Analysis performed at University of Newcastle, Australia.
[b] Analysis performed at University of Manchester, UK.
[c] The recovery is against the certified value of total iAs of 92 μg/kg.
[d] This is the average of the %variation between each paired sample.

3.2. As concentration in water and food

Total As concentration in food and water samples is summarised in Table 2. In 76% of surveyed households, the cooking and drinking water was the same and 37% of households were using treated/filtered water for drinking. While a wide range of As concentration in drinking water was noted, 77% of the households had As concentration in drinking water below the WHO provisional guide value of 10 μg/L, 14% had As concentration greater than or equal to 10 μg/L but less than 50 μg/L, the Indian permissible limit in absence of an alternative source (Cgwb.gov.in., 2020) and 9% had As concentration more than or equal to 50 μg/L. The observed distribution of As concentrations in water samples was similar to that reported by Richards et al. (2020). They reported 16% ($n = 273$) of their samples exceeding As concentrations of 10 μg/L compared to 23% in this study and 4% exceeding 50 μg/L compared to 9% in this study. Their sampling was not restricted to known As impacted districts and water samples were randomly collected from existing private and government wells over households.

We found higher concentration of total As in cooked rice compared to raw rice, similar to the findings of the Kumar et al. (2016b) study of Samastipur, Bihar. But, in this study (Table 2), the concentrations found in cooked rice was much higher than in the previous study of Kumar et al. (2016b). They have reported mean, median and range of total As concentration in cooked rice samples ($n = 15$) of 119, 77 and 10–728 μg/kg compared to 190, 97 and 16–1128 μg/kg, respectively observed in this study. The raw rice samples in Kumar et al. (2016b) study also had lower total As concentrations (mean = 51, median = 58 and range = 2–132 μg/kg) compared to this study (mean = 95, median = 94 and range = 15–231 μg/kg.

Table 2
Concentration of total As in drinking water and food components collected from households ($n = 91$) of As-affected areas in Bihar, India.

Parameters	Sample size	Mean ± SD	Median	Range
Drinking water (μg/L)	90	35 ± 127	3.6	0.01–732
Cooking water (μg/L)	90	78 ± 240	3.6	0.01–1542
Raw rice (μg/kg)	89	95 ± 33	94	15–231
Cooked rice (μg/kg)	70	190 ± 227	97	16–1128
Wheat grain (μg/kg)	82	41 ± 46	23	0.96–234
Wheat flour (μg/kg)	72	48 ± 68	25	3.6–448
Potato (μg/kg)	86	42 ± 32	31	5.6–176

There was no significant correlation between paired raw and cooked rice samples, but a correlation between As concentration in cooking water and total As in cooked rice samples (Spearman's Rho = 0.4838, $P < 0.05$) was observed. Fig. 2 shows the percentage change in As concentration in rice due to cooking. When cooking water As was less than 10 μg/L ($n = 54$) the median percentage change was −3% (IQR: −29% - 54%). Besides, a decrease in total As content of rice due to cooking was observed in 46% of the rice samples ($n = 31$) and median As concentration in the cooking water of those households was 1.41 μg/L (IQR: 0.39–5.35 μg/L). Similar findings were noted by Chowdhury et al. (2020) and authors reported a decrease of 34–89% and 23–84% in sunned and parboiled cooked rice samples when cooked in water with As concentrations less than 3 μg/L. This was also noted in a previous household-based survey in Khejuri-I block, Midnapur district, West Bengal, India where cooked rice samples had lower As than the raw grains when cooking water was less than 1 μg/L (Mondal et al., 2010). Conversely, when cooking water As was greater than or equal to 10 μg/L ($n = 13$) the median percentage increase in total As in cooked rice was 231% (IQR: 125% - 384%); and when cooking water As was more than or equal to 50 μg/L ($n = 7$) the median percentage increase was 265% (IQR: 80% - 692%). Chowdhury et al. (2020) also found an increase of 24–337% in sunned and 114% in parboiled rice due to cooking with water having As concentrations greater than 50 μg/L. While increase in total As concentration in cooked rice when cooking water As was either greater than 10 μg/L (Chowdhury et al., 2020; Mondal and Polya, 2008; Ohno et al., 2009) or greater than 50 μg/L (Bae et al., 2002; Chowdhury et al., 2020; Roychowdhury, 2008; Sengupta et al., 2006) were noted in previous studies, some very high increase of total As in rice due to cooking even at relatively low As in cooking water, as observed in this study, needs further investigation.

Total As concentrations in wheat grains and flour was similar to those observed in our previous publication (grain: 44 ± 48 μg/kg, $n = 72$; flour: 50 ± 74 μg/kg, $n = 58$) which had a lower sample size (Suman et al., 2020). Total As concentration in potato was ten times lower than concentrations reported in vegetables (mean ± SD: 452 ± 712 μg/kg: median: 145 μg/kg ($n = 34$)) but not potato, in a previous study (Kumar et al., 2016b).

3.3. Model input variables

Due to the low sample size in this study, to determine the distribution function for water intake we combined all existing results from Bihar, India (Table 3) giving a mean water intake of 4.6 ± 1.2 L/day. It is that the water intake reported by Singh and Ghosh (2012) and

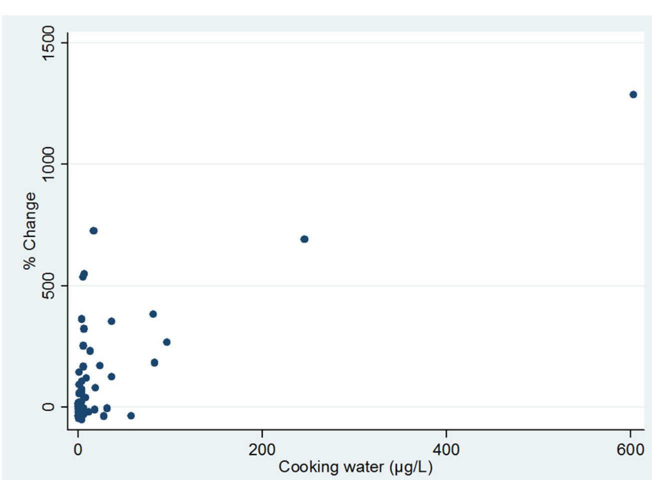

Fig. 2. Percentage change in total As concentration due to cooking of rice with change in As concentration of cooking water.

Table 3
Drinking water intake rates in Bihar, India.

Districts	Study Area	Sample Size	Age (Years)	Water Intake (L/day)	References
Patna	Rampur Diara	264	> 20	5.95	(Singh and Ghosh, 2012)
	Haldichapra	222	> 20	6.11	
Gaya	Bodh Gaya	60	Adults	4.00	(Yasmin et al., 2013)
Vaishali	Chaukia	281	> 20	5.70	(Singh et al., 2014)
	Terahrasiya	283	> 20	5.35	
Bhagalpur	Mamalkha	159	> 20	6.03	
	Masharu	149	> 20	5.30	
Samastipur	Mohiuddinagar	23	20–60	3.50	(Kumar et al., 2016b)
	Mohanpur	23	20–60	3.50	
Samastipur	Dharampur	10	> 20	3.30	This study
Patna	Maner	7	> 20	3.86	
Bhojpur	Sinha	8	> 20	3.19	
Begusarai	Gyantoli	10	> 20	3.50	

Singh et al. (2014) were almost double that of other studies, including ours, from Bihar. This could be attributed to seasonal variation (month of study was not reported except by Kumar et al. (2016b) whose study took place in May - the survey in the current study was done in August), sampling method and sample size. All available results from Bihar were combined to ensure that the likelihood and consequence of very high or low intake is probabilistic and represented by an appropriate function (Arunraj et al., 2013).

We found a significant difference in rice (one-way ANOVA (F = 4.58, $P = 0.03$) and wheat (one-way ANOVA, $P < 0.001$) intake between male and female respondents, with males having a higher intake (Table 4); while potato intake was not significantly different (one-way ANOVA (F = 0.01, $P = 0.91$)). Overall adult average rice intake (82 ± 74 g/day) estimated in this study was much lower than that of Kumar et al. (2016b) study (average 378 g/day; range 250–500 g/day). This could be attributed to one specific district (Samastipur) being sampled in their study. That said, the reported wheat intake in this study (average 226 ± 106 g/day) was similar to that of Kumar et al. (2016b) study (average 259 g/day; range: 100–400 g/day). Previously, Singh and Ghosh (2012) reported rice consumptions of 159 g/day and 169 g/day from two different areas of Patna district in Bihar. Though their findings were higher than the average intake observed in this study they were much lower than those of the Kumar et al. (2016b) study. In our survey we found many respondents (around 25%) did not consume rice and preferred homemade bread (*chapati*) over rice due to the perceived risk of conditions such as obesity and diabetes. Moreover, a decreasing trend in rice consumption has also been noted previously, for example consumption in 1993–94 was reported to be 86.5 kg/capita/year compared to 75.4 kg/capita/year in 2011–12 (Kumar et al., 2016a).

We considered iAs concentrations in cooked rice and wheat flour for exposure assessment rather than the concentrations in raw rice and wheat grain, respectively (Table 4). As detailed in Suman et al. (2020), wheat flour was often found to have slightly higher total As concentrations compared to the grain: this is attributed to indigenous processing at households, sometime using As contaminated water.

3.4. Overall iAs exposure

Using probabilistic estimation, overall median iAs exposure was 0.84 μg/kg BW/day, with a wide range: 5th and 95th percentile of 0.21 and 11.1 μg/kg BW/day (Fig. 3A). The 85th percentile was 3.49 μg/kg BW/day for this surveyed population hence at least 15% of the population had As exposure above 3 μg/kg BW/day, the recommended upper limit for iAs exposure by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) using the benchmark dose lower confidence limit for a 0.5% (BMDL0.5) increased incidence of lung cancer

Table 4
Input parameters used in calculation of exposure distribution and As attributable lifetime cancer risks.

Input variable	Point estimate (mean ± SD)	Input parameter/ distribution	Data source
Total As in drinking water (μg/L)	35.41 ± 127.01	Lognormal (σ = 28.38, μ = 269.03)	This study
Total As in cooked rice (μg/kg)	189.85 ± 227.14	Pearson5 (α = 2.21, β = 242.28)[a]	This study
Total As in wheat flour (μg/kg)	47.73 ± 68.37	Pearson5 (α = 1.76, β = 40.53)[a]	This study
Total As in potato (μg/kg)	42.10 ± 31.99	Pearson5 (α = 3.80, β = 130.39)[a]	This study
iAs in cooked rice (%)	87.78 ± 7.13	Kumaraswamy (α1 = 0.15, α2 = 0.25, a = 74.81, b = 100.26)[b]	This study
iAs in wheat flour (%)	99.38 ± 0.60	Extreme Value (a = 99.60, b = 0.33)	This study
iAs in potato (%)	84.2 ± 2.2	Constant	(Jia et al., 2019)
Bioconcentration factor of rice	90%	Constant	(Mondal et al., 2010)
Bioconcentration factor of wheat	80%	Constant	(Althobiti and Beauchemin, 2018)
Bioconcentration factor of potato	100%	Constant	Assumed (Upadhyay et al., 2019)
Water Intake (L/day)	4.56 ± 1.18	Uniform (a = 2.94, b = 6.35)	Combined studies (Table 3)
Rice intake (g/day)	Male: 93.30 ± 86.58 Female: 69.27 ± 56.10	Exponential (λ = 93.29)[c] Exponential (λ = 69.26)[c]	This study
Wheat intake (g/day)	Male: 273.01 ± 105.35 Female: 173.77 ± 80.34	Kumaraswamy (α1 = 1.37, α2 = 2.39, a = 85.69,b = 566.7)[b] Normal (σ = 173.76, μ = 80.33)	This study
Potato intake (g/day)	Male: 154.94 ± 145.50 Female: 152.95 ± 100.27	Exponential (λ = 154.95)[c] Triangular (m = 0, a = 0, b = 441.42)[d]	This study
Body weight (kg)	Male: 61.26 ± 13.94 Female: 52.38 ± 10.78	Extreme value (a = 54.83, b = 11.14) Weibull (α = 2.15, β = 24.847)[e]	This study
Exposure duration (years)	40 if age > 40 or equal to age (Mondal et al., 2010)	Male = Triangular (m = 19.25, a = 40, b = 40); Female = Triangular (m = 18.62,a = 40, b = 40)[d]	Age based on this study
Life expectancy (years)	Male: 67.8 Female: 68.4	Constant	(Niti.gov.in, 2020)
Gender distribution (%) Based on 2011 data	Male: 52.2% Female: 47.8%	Constant	(Censusindia.gov.in, 2020)
Cancer potency slope factor $((mg/kg)/d)^{-1}$	1.5	Constant	(USEPA, 2020)

[a] Pearson 5 distribution has the form $f(x) = \frac{e^{-\beta/x}}{\beta\Gamma(\alpha)(x/\beta)^{\alpha+1}}$ where α = continuous shape parameter and β = continuous scale parameter;
[b] Kumaraswamy distribution has the form $f(x) = \frac{\alpha_1\alpha_2 z^{\alpha_1-1}(1-z^{\alpha_1})^{\alpha_2-1}}{(b-a)}$ where α_1 and α_2 = continuous shape parameter and *a*, *b* = continuous boundary parameters;
[c] Exponential has the form $f(x)=\lambda e^{(-\lambda x)}$ where λ = continuous inverse scale parameter;
[d] Triangular distribution has the form $f(x) = \begin{cases} \frac{2(x-a)}{(m-a)(b-a)} \\ \frac{2(b-x)}{(b-m)(b-a)} \end{cases}$ where m = continuous mode parameter and *a*, *b* = continuous boundary parameters;
[e] Weibull distribution has the form $f(x) = \frac{\alpha}{\beta}\left(\frac{x}{\beta}\right)^{\alpha-1} e^{-\left(\frac{x}{\beta}\right)^{\alpha}}$ where α = continuous shape parameter and β = continuous scale parameter.

(Cubadda et al., 2017). That said, the probabilistic mean iAs exposure was 3.51 μg/kg BW/day, which was higher than this recommended upper limit.

Median iAs exposure from drinking water (0.31 μg/kg BW/day) was similar to iAs exposure from food (0.34 μg/kg BW/day, Fig. 3B). Contribution of food to overall exposure was 36% (median) when drinking water exposure was below WHO guideline value of 10 μg/L but 53% (median) when concentrations in drinking water was above 10 μg/L (Fig. 4A). This is in contrary to previous studies (Cubadda et al., 2017; Mondal et al., 2010; Rasheed et al., 2018) where food was found to be a major contributor to iAs intake only at As concentrations in drinking water below 10 μg/L. This is largely due to very high total As concentration in cooked rice samples in those households with As in drinking water greater than 10 μg/L (Table 5). Median percentage change in the As concentration of cooked rice compared to that in raw rice due to cooking of rice was −3% (IQR: −29% - 54%; $n = 54$) when drinking water was less than 10 μg/L while it was 231% (IQR: 125% - 384%; $n = 13$) when drinking water As was more than 10 μg/L (Kruskal-Wallis H test showed that there was a statistically significant difference in percentage increase on cooking between the two groups, $\chi^2 = 13.88$, $P < 0.001$). This suggests that when As concentration was high in drinking water, there was higher possibility of increased As concentrations in cooked rice. The influence of high increase in total As concentration in cooked rice on overall exposure at As in drinking water greater than 10 μg/L was also reflected by its highest contribution of 45% towards total food exposure (Fig. 4B). There was no significant difference in As concentration in wheat flour at below or above 10 μg/L (Table 5), and the contribution of exposure from wheat flour (median = 35%) was almost equal to exposure from cooked rice (median = 38%) where As is drinking water was less than 10 μg/L (Fig. 4B). The other potential explanation could be the contribution of drinking water at concentration above 10 μg/L was not as high as could be expected because the median was 36 μg/L (IQR: 17–96 μg/L) and out of 21 households with drinking water As above 10 μg/L only five had concentrations above 100 μg/L.

3.5. Excess lifetime cancer risk

Median age and gender adjusted overall excess lifetime cancer risk for this studied population (6×10^{-4}) and 95th percentile (8.01×10^{-3}) were higher than the commonly inferred USEPA regulatory threshold target cancer risk levels of 10^{-4}–10^{-6} (Fig. 5A). Median risk from food intake (2.44×10^{-4}) was marginally higher than risk from water intake (2.26×10^{-4}) demonstrating food-related risks were of serious concern. Similar observation was made previously in our study from Chakdaha in West Bengal, India where median risk from cooked rice was found to be slightly higher than from drinking water (Mondal and Polya, 2008) but contrary to previous studies (Cubadda et al., 2017; Mondal et al., 2010; Rasheed et al., 2018) we found risk from food intake (median = 1.89×10^{-4}; 95th percentile = 7.32×10^{-4}) was slightly lower than risk from water intake (median = 3.34×10^{-4}; 95th percentile = 5.96×10^{-4}) when drinking water As was less than 10 μg/L (Fig. 5B) and risk from food intake (median = 4.00×10^{-4}; 95th percentile = 1.83×10^{-3}) was higher than risk from water intake (median = 3.57×10^{-4}; 95th percentile = 6.09×10^{-4}) when drinking water As was greater than 10 μg/L (Fig. 5C).

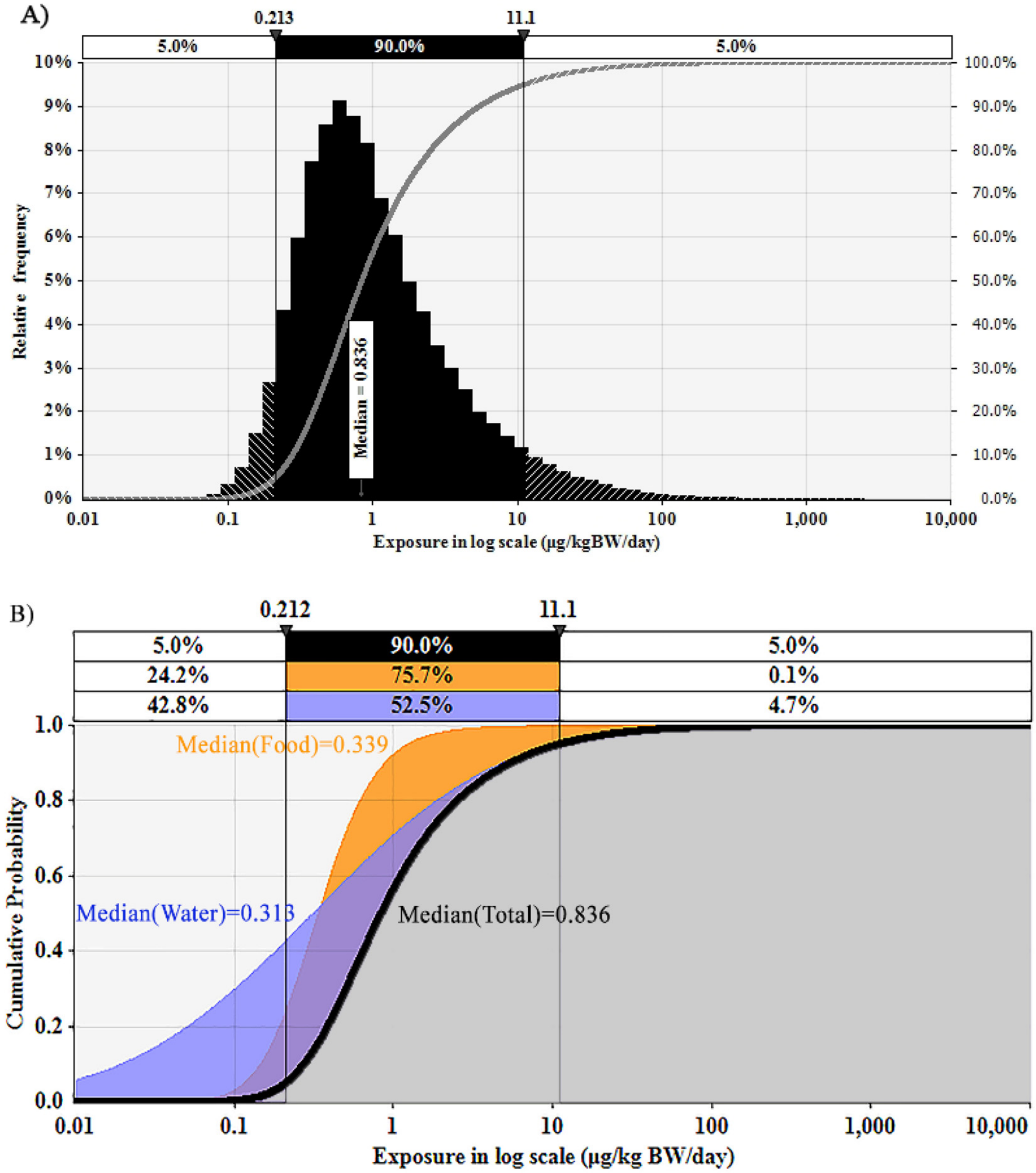

Fig. 3. Probabilistic exposure assessment in Bihar, India A)Frequency distribution of Total inorganic As exposure, B) Cumulative frequency distribution of inorganic arsenic exposure from food and from drinking water.

While the probabilistic risk assessment is dependent on the accuracy and representativeness of the input data, in contrast to deterministic risk assessment, probabilistic risk assessment attempts to characterize uncertainty and variability based on the complete range of data for the input parameters developing the most appropriate distribution and hence are less likely to under-or over-estimate the risks when compared with deterministic assessment (Peng et al., 2016). For example, in this study both for the exposure assessment, 5th (0.21 μg/kg/day) and 95th (11.1 μg/kg/day) percentiles (Fig. 3) and for risk assessment, 5th (1.51×10^{-4}) and 95th (8.01×10^{-3}) percentiles (Fig. 5), provided the range below and above which the chances of both exposure and risk was less likely.

Compared to the recent study covering whole of Bihar ($n = 273$) where 16% of tube-well water samples were above WHO guideline value of 10 μg/L (Richards et al., 2020) we found 23% of the samples above the guideline value but this study was conducted in As exposed areas over whole of Bihar. Both these results show that human health risk estimates based upon tube well water As concentrations in some previous studies may be somewhat overestimated, for example, Chakraborti et al. (2017) estimated that 32.7% of population had As exposure of greater than 10 μg/L in drinking water. Of course, the datasets are not directly comparable and subject to temporal and spatial variations along with changes in exposure due to mitigation. If the estimate of Chakraborti et al. (2017) stating 3.1 million people in Bihar consuming As greater than 10 μg/L in drinking water was accurate then based on the overall risk estimate of 7.89×10^{-4} at drinking water greater than 10 μg/L (Fig. 5C) an estimated 2445 people could be at excess risk of cancer due to As exposure from food and water. But a much higher number of excess cancer risk with around 5.54 in 10,000 will be expected due to As exposure from food and water at As in drinking water below 10 μg/L. Hence exposure to As from drinking water and food with As concentrations below the WHO provisional guide value of 10 μg/L is of equal if not of more importance due to much higer proportion of populations being exposed to As at concentrations lower than 10 μg/L. Besides, other than cancers significant detrimental health effects could be prevalent at exposures less than 10 μg/L. For example,

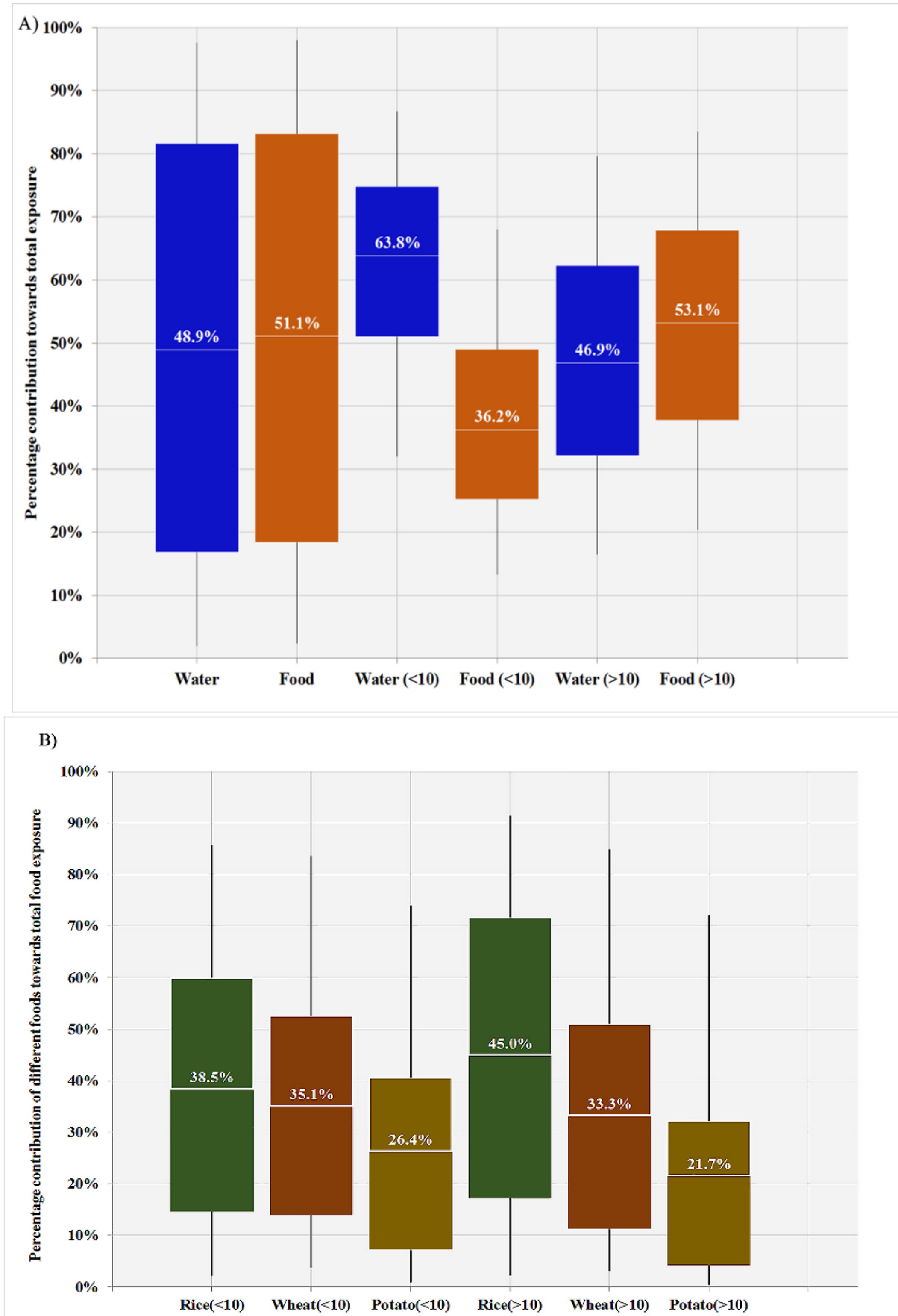

Fig. 4. Probabilistic assessment of contribution of different exposure media to overall iAs exposure grouped by below and above 10 μg/L in drinking water: A) water vs. food; B) cooked rice vs. wheat vs. potato towards total food exposure.

increased risks of cardiovascular diseases (CVD) like increased risks of coronary heart disease (CHD) mortality and CVD mortality as well as combined fatal and non-fatal CHD, CVD, carotid atherosclerosis disease and hypertension have been determined in those exposed to drinking water below or at 10 μg/L compared to 1 μg/L (Xu et al., 2020). Hence, further studies in Bihar As exposed areas are warranted to determine the true disease burden from As exposure.

3.6. Limitations

There are inevitable constraints on efforts to assess exposure and health risks using probabilistic methods, some of these are discussed below and could form a basis for improvement for future risk assessments. While we have determined the As exposure from drinking water based on samples collected from the households, exposure from

Table 5
Median (interquartile range, sample size) total As concentration in exposure media stratified by As in drinking water of <10 μg/L, the WHO provisional guide value.

Exposure media	Drinking water <10 μg/L	Drinking water >10 μg/L	Kruskal-Wallis H test
Drinking water (μg/L)	1.4 (0.66–3.7; n = 69)	36 (17–96; n = 21)	$\chi^2 = 47.77$, P < 0.001
Cooking water (μg/L)	1.8 (0.50–3.7; n = 69)	37 (17–172; n = 21)	$\chi^2 = 36.64$, P < 0.001
Cooked rice(μg/kg)	90 (70–145; n = 55)	269 (116–594; n = 14)	$\chi^2 = 10.58$, $P = 0.001$
Wheat flour (μg/kg)	23 (15–42; n = 52)	45 (22–57; n = 19)	$\chi^2 = 3.84$, $P = 0.049$
Potato (μg/kg)	29 (21–42; n = 66)	34 (26–68; n = 19)	$\chi^2 = 1.38$, $P = 0.239$

other secondary sources could take place. Due to potential misclassification bias in self-reported estimates of time spent outside the house and locating the appropriate secondary sources, this study was restricted to As concentrations in drinking water of the primary sources. In terms of limitations with respect to food exposure, from vegetables other than potato, have not been considered even though high concentartions of mean As in vegetables of 452 ± 712 μg/kg have been noted in a previous study (Kumar et al., 2016b). Therefore, total As exposure from food ingestion might have been somewhat underestimated, however the frequency of consumption of the vegetables like gourd, cucumber, brinjal, luffa and ladyfinger studied by Kumar et al. (2016b) was not regular. Use of wheat flour instead of homemade bread (*chapati*) and raw potato over cooked ones might have introduced some error towards the absolute exposure and risk estimates but our results may have only been somewhat underestimated if the As in cooking water was contributing to an overall increase of As concentration in the cooked food. While the cooked food is most preferable for exposure and risk assessment, procuring the samples in a cross-sectional survey was difficult as in rural Bihar most villagers do not store cooked food and leftovers are often discarded after the meal.

Although we covered a wide range of As contamination in drinking water, this study was limited to 19 villages from eight districts rather than the whole of Bihar. Furthermore the selection of villages did depend on access and contact as was therefore somewhat opportunistic. Besides, even though all our food samples were collected from households and most likely to be cultivated locally, that said, if the produce was not enough villagers often procured rice and wheat from the Public Distribution System (PDS). The PDS is where the produce of Bihar is centrally procured by Government of Bihar after harvesting and distributed to the households at subsidised rate to ensure affordability (Dfpd.gov.in., 2020) resulting in mixing of grains between As endemic and non-endemic areas. Hence, future studies should include systematic sampling covering the whole state of Bihar rather than only endemic areas.

Though 24-h recall is a preferred method for dietary intake assessment it is limited by possible recall bias, interviewer bias and any potential change to the diet (Shim et al., 2014). As a part of another ongoing study, a subgroup ($n = 55$) of this studied population was surveyed after a few months and the 24-h recall was repeated. Though the individual food intake rates varied between the two surveys, the observed average wheat, rice and potato intakes in the first (wheat: 250 ± 118 rice: 65 ± 64; potato: 166 ± 132 g/day, respectively) and second (wheat: 261 ± 99; rice: 77 ± 76; potato: 143 ± 105 g/day, respectively) surveys were not significantly different. We performed a sensitivity test by excluding those respondents who reported a special occasion during the 24-h recall (n = 5) and compared the average intake with that of the overall surveyed population but there was no noticeable difference in the average food intakes.

The limited database on exposure duration has resulted in their general point estimates being entered in the model limiting the variabilities and uncertainties to be ascertained. We estimated the cancer risks by multiplying the resulting lifetime average daily iAs dose by the current USEPA Integrated Risk Information System iAs cancer slope factor of 1.5 $(mg/kg)/d)^{-1}$ which is based on the risk of skin cancer (USEPA, 2020). In 2010, the USEPA IRIS calculated a new cancer slope factor of 25.7 $(mg/kg)/d)^{-1}$ for combined lung and bladder cancers (USEPA, 2010). If the proposed slope factor of 25.7 $(mg/kg)/d)^{-1}$ was used, the median age and gender adjusted overall excess lifetime cancer risk for this studied population will be 1.03×10^{-2} (95th percentile = 1.38×10^{-1}) suggesting 1 per 100 people could be at risk in this studied As exposed populations of Bihar, India.

4. Conclusions

In this studied As exposed population of Bihar, India, overall iAs exposure from food, determined based on intake of rice, wheat and potato was almost equal to that from drinking water. 77% of the households had drinking water As concentration below the WHO guideline value of 10 μg/L and 37% were using some form of improved water for drinking indicating that a significant proportion of drinking waters in this studied population might have As lower than 10 μg/L. Median contribution of food to overall iAs exposure was 36% when drinking water exposure was below 10 μg/L, but contrary to previous studies, food was found to contribute more than drinking water (by 6%) when arsenic concentrations in drinking water was above 10 μg/L. This suggested that the presence of high As concentrations in water may result in higher total As concentrations in the food which could be due to both, increased As on cooking and naturally high total As in the grains and potato.

The median and 95th percentile excess lifetime cancer risk was estimated to be 6 per 10,000 and 80 per 10,000 respectively in the studied As exposed population in Bihar, which is higher than the 10^{-4} -10^{-6} range typically used by the USEPA as a threshold to guide determination of regulatory values. Median and 95th percentile excess lifetime cancer risks from food intake were 1.89×10^{-4} and 7.32×10^{-4} respectively when drinking water As was below 10 μg/L and 4.00×10^{-4} and 1.83×10^{-3} respectively when drinking water As was above 10 μg/L. Hence exposure to iAs from food is of equal if not of more importance to that from drinking water for many of the studied population. The highest contribution to overall iAs exposure from food was from cooked rice, largely due to the relatively high As content in cooked rice. While provision of low As irrigation water for growing the crops and vegetables in As-endemic areas is an immediate requirement, food safety regulations must be emphasised in India. At household level, improving cooking practices, like cooking in low arsenic water, cooking rice in excess water and kneading the wheat flour in low arsenic water may lead to reduction in dietary As exposure.

CRediT authorship contribution statement

The study was conceptualised by DM, Data was acquired by MMR, SS, PS, ABS, MAR, AFB, RK, NB and DAP; Formal sample analysis was done by MMR, SS, PS, ABS, MAR, AFB; Funding acquisition was by DM, SKS, AG; Investigation was done by ALL authors; Methodology was developed by DM, MMR, AG, NB and DAP; Project administration was by DM, SKS, AG and DAP; Resources provided by DM, MMR, AG, SKS and DAP; Software was run by DM; Supervision by DM, MMR, SKS, AG and DAP; Writing – DM; Writing - review & editing-DM, MMR and DAP.

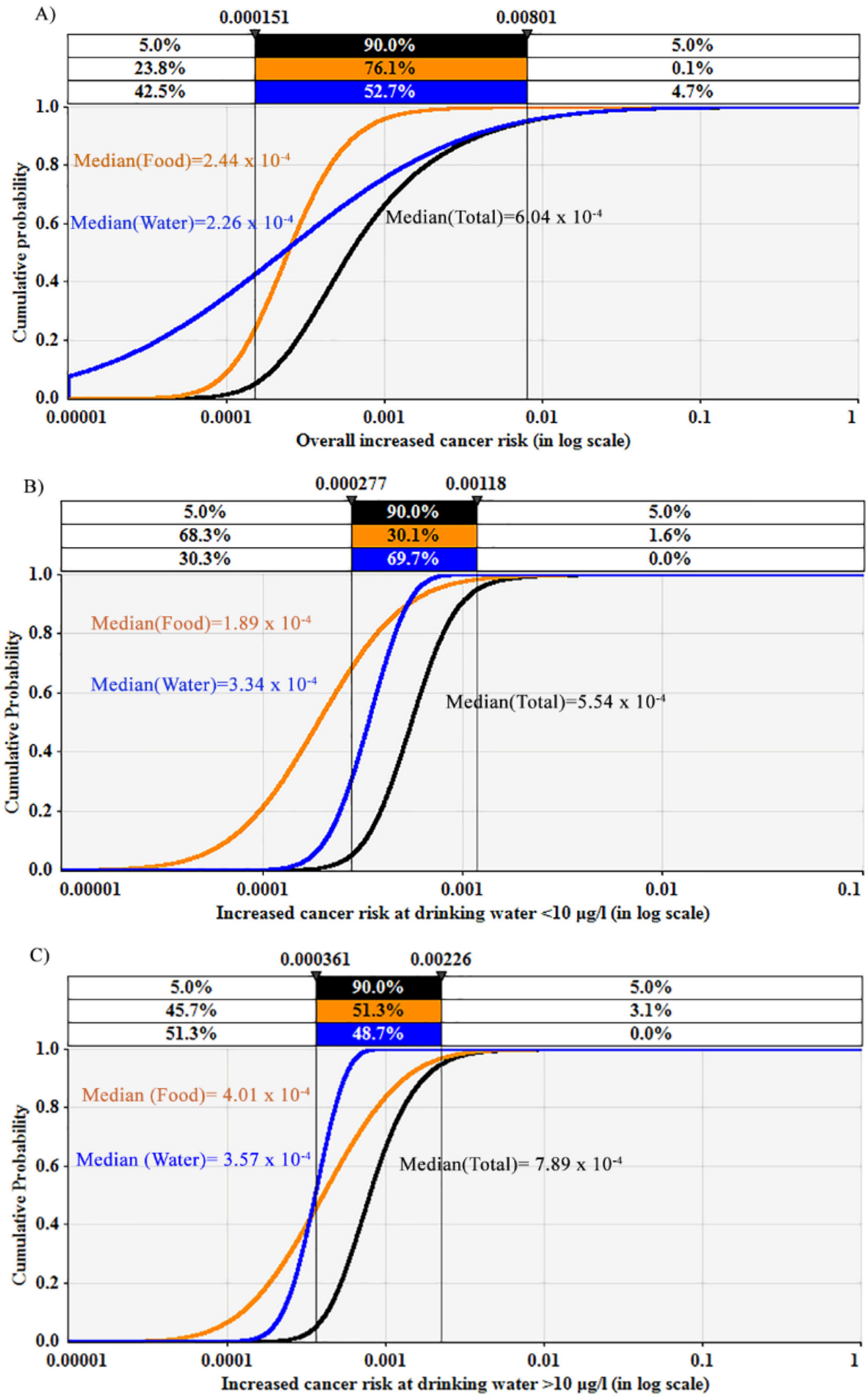

Fig. 5. Cumulative probability distributions of age and gender adjusted excess lifetime cancer risk from food (rice, wheat and potato) intake, water intake and both (total) for the studied population of Bihar, India: A) Overall; B) when As in drinking water is less than 10 µg/L; C) when As in drinking water is more than 10 µg/L.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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