Applied Geochemistry 23 (2008) 2987–2998

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Rice is a major exposure route for arsenic in Chakdaha block, Nadia district, West Bengal, India: A probabilistic risk assessment

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ARTICLE INFO

Article history:
Available online 15 July 2008

ABSTRACT

The importance or otherwise of rice as an exposure pathway for As ingestion by people living in Bengal and other areas impacted by hazardous As-bearing groundwaters is currently a matter of some debate. Here this issue is addressed by determining the overall increased cancer risk due to ingestion of rice in an As-impacted district of West Bengal. Human target cancer health risks have been estimated through the intake of As-bearing rice by using combined field, laboratory and computational methods. Monte Carlo simulations were run following fitting of model probability curves to measured distributions of (i) As concentration in rice and drinking water and (ii) inorganic As content of rice and fitting distributions to published data on (i) ingestion rates and (ii) body weight and point estimates on bioconcentration factors, exposure duration and other input variables. The distribution of As in drinking water was found to be substantially lower than that reported by previous authors for As in tube wells in the same area, indicating that the use of tube well water as a proxy for drinking water is likely to result in human health risks being somewhat overestimated. The calculated median increased lifetime cancer risk due to cooked rice intake was 7.62×10^{-4}, higher than the 10^{-4}–10^{-6} range typically used by the USEPA as a threshold to guide determination of regulatory values and similar to the equivalent risk from water intake. The median total risk from combined rice and water intake was 1.48×10^{-3}. The contributions to this median risk from drinking water, rice and cooking of rice were found to be 48%, 44% and 8%, respectively. Thus, rice is a major potential source of As exposure in the As-affected study areas in West Bengal and the most important exposure pathway for groups exposed to low or no As in drinking water.

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1. Introduction

Arsenic concentrations in shallow, reducing groundwater in West Bengal constitute a major health hazard to millions of people using these waters for drinking, cooking or irrigation (Charlet and Polya, 2006). An environmental tragedy is developing in West Bengal and an alarming number of toxicity cases associated with ingestion of As-enriched drinking water have been reported (Mazumder et al., 2000; Mukherjee et al., 2005; von Ehrenstein et al., 2005).

Of the 3 major possible routes of As exposure – inhalation (Pal et al., 2007), ingestion (Huq and Naidu, 2003) and dermal contact (Watts and Halliwell, 1996) – ingestion is potentially the greatest contributor to exposure and hence for healthy humans who are not occupationally exposed the most significant route of exposure to As is through the oral intake of food and beverages (WHO, 2001b). Among the many possible pathways of As ingestion illustrated in Fig. 1, drinking water is considered as the most significant as epidemiological data that has been accumulating during last couple of years has mainly relied on the concentration of As in the drinking water as the proxy for exposure (Cantor and Lubin, 2007; Chen et al., 1988; Smith et al., 2000, 2006a). But the chronic As toxicity symptoms

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doi:10.1016/j.apgeochem.2008.06.025

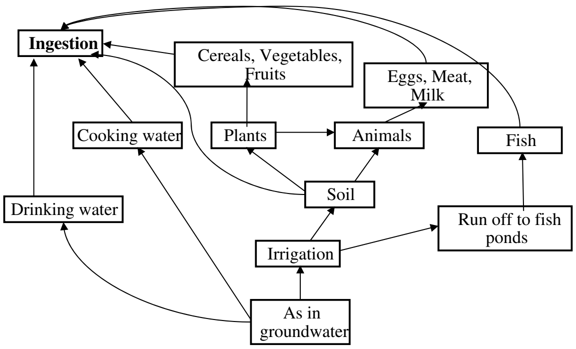

Fig. 1. Possible pathways of As ingestion.

recorded in Bangladesh and West Bengal may reflect pathways other than the consumption of water (Huq and Naidu, 2003). Soil-crop-food transfer, as well as cooking in As-enriched water has been suggested to be among the major exposure pathways (Alam et al., 2003).

A number of studies have reported the presence of As in rice, ranging in concentration from 0.03 to 1.83 mg As/kg (Abedin et al., 2002a,b; D'Amto et al., 2004; Heitkemper et al., 2001; Kohlmeyer et al., 2003; Meharg, 2004; Schoof et al., 1998; Williams et al., 2005) and have implicated As-enriched groundwater used extensively for crop irrigation, particularly in the dry season to be the cause. But there still exist different views, based on different objectives and study designs in addressing the importance of rice as a further potential exposure route in the Bengal delta. Williams et al. (2006) based on a major food market-basket survey from Bangladesh addressing the inorganic and total As content of rice, predicted for all the surveyed districts except one that the consumption of rice would be equivalent to drinking water with an As content of 10 μg/L and suggested that rice is already a major exposure route. However van Geen et al. (2006), studying the soil and soil–water As profiles for Bangladeshi paddy fields reported no evidence of proportional transfer of As to rice and suggested that exposure of the Bangladeshi population to As contained in rice is less of an immediate concern than the continued use of groundwater containing elevated levels of As for drinking or cooking.

Notwithstanding the differences between Williams et al. (2006) and van Geen et al. (2006) regarding the relative importance of rice as an exposure route, various studies suggest that rice is likely to become an even more important exposure route in the future. Roberts et al. (2007) suggest that the As content of soils in rice-growing areas irrigated by As-bearing groundwaters may be undergoing a secular increase, suggesting that As exposure from rice might become even more significant in the future. Lastly, there has been extensive intervention in West Bengal to reduce human exposure to As in ground waters utilised as drinking water and this will also lead to an increase in the importance of rice as an exposure route relative to that of water.

In an average Bengali home, the main meal would consist of boiled rice served with some vegetables (Alam et al., 2003). In As endemic areas with subsistence rice diets, as in Bengal where 73% of the calorific intake is from rice (Ninno and Dorosh, 2001), the contribution of inorganic As from rice should be considered in assessments of risk from As exposure. Huq et al. (2006) noted high-As in some vegetables including arum, gourd leaf, *Amaranthus* (*shak*, both *data shak* and *lal shak*) and *Ipo–mea* (*kalmi*), with concentrations ranging from 0.8 mg As/kg in gourd to 1.58 mg As/kg dry weight in arum. But Williams et al. (2006) reported vegetables, pulses and spices contribute less to As body burden than rice due to the lower As concentrations in them and lower rates of consumption (Williams et al., 2006).

Variation in As content of rice depends on the rice variety (different genotypes) (Meharg and Rahman, 2003) and cooking methods (Bae et al., 2002; Rahman et al., 2006; Sengupta et al., 2006). Cooking of rice with As-enriched water leads to an increase in the As content of rice (Ackerman et al., 2005; Bae et al., 2002; Laparra et al., 2005; Ohno et al., 2007; Signes et al., 2008) whereas cooking with low-As water (Sengupta et al., 2006; Signes et al., 2008) and a large excess of water discarding the gruel (Rahman et al., 2006) leads to a decrease. Thus risk assessments should ideally consider exposure from cooked rice rather than from raw rice (Bae et al., 2002).

Inorganic As and DMA(V) are the predominant species present in rice (Cullen and Reimer, 1989; Schoof et al., 1999; Signes et al., 2008; Williams et al., 2005) though there are large genotypic differences in the levels of DMA and inorganic As (Liu et al., 2006).

Arsenic is associated with both cancer and non-cancer effects. Notwithstanding differences in reported standardised mortality ratios (Mondal et al., 2008), there is sufficient evidence to conclude that As causes internal cancers and the NRC (2001) summarise large epidemiological studies on dose–response relationships. There are, however, large uncertainties in the dose–response relationship for non-malignant conditions, such as ischemic heart disease and diabetes mellitus, from As exposure (Adamson and Polya, 2007) making the magnitude of non-malignant conditions in humans arising from As exposure difficult to quantify (Adamson and Polya, 2007).

The traditional approach to risk assessment is to deal with variability and uncertainty by successively adding safety factors or selecting parameter estimates that will almost certainly overestimate the risks involved (Paustenbach, 2000). This deterministic approach gives a distinct estimate of maximum exposure, which can subsequently be compared with reference values for health and environmental effects. It is not possible however to obtain an indication of the uncertainty in this value or the margin of safety (NRC, 1994).

Probabilistic risk assessment, on the other hand, provides a method to deal with these problems. Variability and uncertainty in input parameters are described by probability distributions, and output is likewise presented as a probability distribution (USEPA, 2001). Probabilistic analysis addresses the main deficiencies of point estimates because it imparts more information and uses all of the available data (Beck and Cohen, 1997). The purpose of probabilistic methods is to find a rational and scientifically justifiable method of dealing with uncertainty and variability (Oberg and Bergback, 2005). The selection of probability distributions for input variables is the single most important factor determining the outcomes of a probabilistic risk assessment (Hope, 1999; USEPA, 1997). Standard probability distributions are used for factors that do not vary greatly between different sites (Finley and Paustenbach, 1994).

Here the issue of the relative importance of rice and water as exposure pathways is quantitatively addressed by determining the overall increased cancer risk due to ingestion of As-bearing rice by using combined field, laboratory and probabilistic methods for typical As impacted districts of West Bengal. Chakdaha block of Nadia district was selected because it is one of the 4 highly As-affected areas of West Bengal (Chakraborti et al., 2004) among the 9 arsenic effected districts. To estimate the cancer risk, the risk model will have many input variables that vary substantially between individuals, areas and over time. Because risk assessment should acknowledge and quantify the uncertainty in risk predictions, to facilitate use by policy makers, probabilistic methods have been used. In this study only cancer-related health risks have been considered because of the large uncertainties in dose–response relationships for non-malignant conditions. Lastly, the importance of cooking on the rice risk is also quantified by analysing cooked rice collected from the households within the survey area and comparing calculated risks from exposure to cooked rice compared to raw rice.

2. Materials and methods

2.1. Sample collection

Two surveys, one in October-2006 and other in April–May-2007, were carried out in Chakdaha block, Nadia district, West Bengal. Stratified clustered random sampling weighted by population was the sampling strategy. Clusters were selected from both urban (1 cluster) and non-urban areas (9 clusters) the ratio reflecting that only 12% of the population of Chakdaha is located in urban areas. Logistical difficulties prevented sampling at one of the randomly selected clusters in eastern Chakdaha and this was substituted by another randomly selected cluster from the same non-urban stratum. Fig. 2 shows the location of the 10 clusters. Five households were selected from each cluster by targeting every fifth household following a pre-determined pattern. Although not strictly random, the pre-determination of the sampling lines meant that there was no observer-related sampling bias introduced.

For each household the observation units were drinking water, cooking water, raw rice and cooked rice. A number of households shared a common water source, mostly from the community tube wells or government supplied pipe water, resulting in a smaller number of drinking water samples ($n = 41$) than the number of surveyed households. Use of different cooking water rather than using the same source as the drinking water was observed only in 18% of the surveyed households ($n = 9$). Raw rice samples were collected from every household surveyed resulting in 100% sample collection ($n = 50$) while it was only possible to obtain the equivalent cooked rice sample from just 75% of the surveyed households ($n = 39$) because cooked rice was only obtainable during the mid day of the survey, when people in those areas cook their food as there is hardly any food preservation. A subset of 17 samples (30% of the total raw rice samples) was analysed for As species to determine the inorganic As content of rice: this subset was selected to ensure that it included samples from each of the 10 clusters.

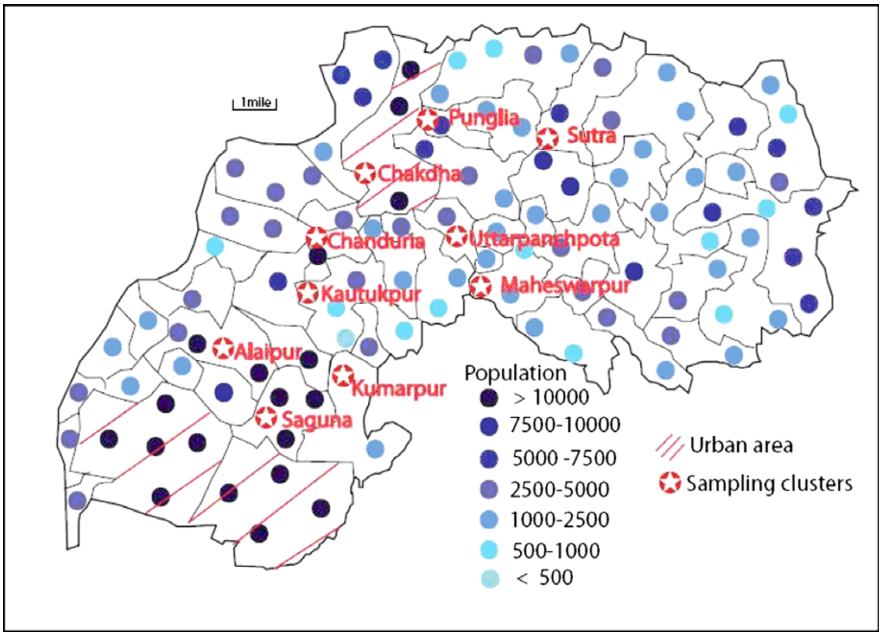

Fig. 2. Sampling locations of 10 clusters within the Chakdaha block.

The unfiltered drinking and cooking water samples collected after pumping the tubewell for a few minutes were transferred to acid cleaned translucent HDPE bottles followed by acidification to 1% v/v HNO_3 (Aristar grade) and were heated to 55 °C for 1 h to deactivate any polio wild virus, which is still endemic in parts of India, and finally stored at room temperature until transported to the University of Manchester. Although sampling of filtered water samples is desirable for the purpose of geochemical equilibrium and mass-transfer modelling, the authors chose not to filter the water samples in this study as the purpose of the sampling was different: to determine human exposure to As whatever its form: dissolved, colloidal or particulate. After collection, the rice samples were placed individually in polythene bags. Raw rice samples were stored at room temperature while cooked rice samples were stored at 4 °C before shipping to the University of Manchester for analysis.

2.2. Sample preparation

Raw rice samples were washed with deionised water (18 MΩ). Cooked rice samples were not. All the rice samples were dried at 65 °C for 48 h and then ground either manually using a pestle and mortar or using an agate ball mill (Fritsch).

2.2.1. Preparation for total arsenic analysis

Ground sub-samples (0.1–0.2 g) of rice were weighed into Pyrex glass digestion tubes, steeped in 2 mL of concentrated subdistilled HNO_3 (Aristar grade) and pre-digested overnight at room temperature. Samples were then heated to 120 °C on a heating block, until clear, and then evaporated to dryness. The residue was spiked with an internal standard (50 μg/L Ge) and dissolved in deionised water (18 MΩ) to a weight of 10 g.

2.2.2. Arsenic extraction for speciation

Following the protocol of Williams et al. (2005, 2006) ground sub-samples (0.25 g) were weighed into Pyrex glass digestion tubes and 2 mL of 2 M trifluoroacetic acid was added. The mixture was allowed to stand overnight for pre-digestion. The tubes were then placed on a heating block at 100 °C for 6 h. The digest was evaporated to dryness and made up to a weight of 5 g with deionised water. It was then centrifuged and the supernatant filtered through 0.45 μm nylon membrane syringe filters (VWR).

2.3. Sample analysis

2.3.1. Total arsenic

Water samples and rice samples were analysed for total As by ICP-MS (PlasmaQuad II (Fisons)). Operating conditions for the instrument were the same as described by Gault et al. (2005). Arsenic was detected at $m/z = 75$ with mass interference from $^{40}Ar^{35}Cl^{+}$ corrected through monitoring of $m/z = 77$ and 82 using TJA Solutions' Plasma Lab software. All the samples were run in triplicate and the standards were run after every set of 10 samples. Concentrations were determined using a 5 point calibration curve. External calibration was accomplished using standards with concentrations of 5, 10, 50, 100 and 200 μg/L for the water analysis and 1, 3, 5, 10 and 50 μg/L for rice analysis. Calibration standards were prepared immediately prior to analysis by dilution of concentrated multielement stock solutions (Alfa Aesar, UK) with 2% subdistilled HNO_3 in 18 MΩ deionised water.

2.3.2. Arsenic speciation

Raw rice samples were analysed for As species: As(III), As(V), DMA and MMA by IC-ICP-MS. The chromatographic system consisted of a 790 Personal IC chromatograph (Metrohm, Switzerland), a liquid chromatography solvent delivery pump, fitted with a 100 μL sample loop and an anion exchange column Hamilton PRP-X 100 (250 × 4.1 mm i.d.) with a guard column (4 × 3 mm i.d.), Polymerx RP-1 (Phenomenex, USA). Operating conditions for the instrument were same as described by Gault et al. (2005). The mobile phase for rice speciation consisted of 6.66 mM $NH_4H_2PO_4$ and 6.66 mM NH_4NO_3 adjusted to pH 6.2 and spiked with an internal standard of 50 μg/L Ge and 50 μg/L Rb. The mobile phase flow rate was maintained at 1 mL/min and the chromatography system was coupled to the ICP-MS. Signals at m/z 75, 77, 51, 53, 72, 78, 82, 83, 85 were monitored. External calibration was accomplished using standards of concentration 5, 10 and 20 μg/L for each of the 4 As species studied. Calibration standards were prepared immediately prior to analysis by dilution of concentrated stock solutions in 18 MΩ deionised water.

Concentrations corrected for drift, blank and sensitivity, with errors calculated explicitly from the measurements of individual samples and of the goodness of fit of the calibration curve were determined using an in house Turbo Pascal program DBSCORR Version P (Polya, 1998a), after peak area calculation by the in house Turbo Pascal programme TRPEAK Version 13 (Polya, 1998b, 2006).

2.4. Risk assessment model

The excess lifetime cancer risks were based on the one the hit model of the USEPA (1989) (see Eq. (1))

$$\mathrm{TR} = \mathrm{CPSo} \times 10^{-3} \sum_{i=1}^{i=N} C_i \times \mathrm{Cing}_i \times \left(\frac{\mathrm{IR}_i}{\mathrm{BW}}\right) \times \mathrm{BCF}_i \times \left(\frac{\mathrm{ED}_i}{\mathrm{LT}}\right) \tag{1}$$

where

TR is the excess lifetime cancer risk.
CPSo is the oral cancer potency slope factor for As.
i = refers to the different potential ingestion medium, i.e. water or cooked rice
N is the total number of exposure media, in this study $N = 2$.
C_i is the total As concentration in the subscripted medium.
Cing_i is the proportion of inorganic As in the subscripted medium.
IR_i is the ingestion rate for the subscripted medium.
BW is body weight of exposed person.
BCF_i is the bioconcentration factor for the subscripted medium.

ED_i is the exposure duration for the ingestion pathway.
LT is the life expectancy of the exposed person.

This equation is only valid for low risk levels ($<10^{-2}$) and is based on the assumption that the dose–response relationship is linear in the low dose portion of the multistage model (USEPA, 1988). Under this assumption the slope factor is constant and the risk is directly related to intake (USEPA, 1988). It is noted that a number of the input variables are reported as a function of age and gender and this dependence is also incorporated in the risk model.

Of the input variables in Eq. (1), C_i, and $Cing_i$ were determined in this study, the remaining parameters were estimated from analogous studies. Based upon the lack of change of As speciation upon cooking of rice (Signes et al., 2008) it has been assumed that the Cing for cooked rice is the same as that for raw rice. In order to avoid any possible bias resulting from differences of As concentrations in cooking water of households from which cooked rice samples were not collected, compared to those from which sampling was possible, a regression equation between As concentration in cooked rice and raw rice was used to estimate the As concentrations of cooked rice for households where samples were not collected.

2.5. Published input variables

All the published input variables used in the risk model are summarized in Table 1.

An extensive study by Watanabe et al. (2004) on water intake by an As-affected adult population of Bangladesh reported the mean intake to be 3.1 L/day with no significant difference between the genders. Ohno et al. (2007) also reported similar results with both the studies being conducted during the hot season and based on similar sampling strategy to quantify the water intake while Kile et al. (2007) observed no seasonal or daily difference in drinking water intake rate. Chowdhury et al. (2000) reported water intake by the children of West Bengal to be 2 L/day. These publications were considered best representative for the study population and run through the model even though they were point estimates.

Constant rice ingestion rates normalised to bodyweight were used in the risk model. Using rice ingestion rate and body weight data for the population of West Bengal from NNMB (2002), it was determined that there was a strong correlation between these two variables and normalised rice ingestion rates were obtained from a linear regression analysis (Fig. 3). The use of such normalised data minimises scattering of calculated risk values that would result from the use of largely co-variant variables with large variability amongst the target population.

The age and gender dependent body weight distribution used in the model were based on the NNMB (2002) database. Age distributions were assumed to be the same as documented for India by the WHO (2001b) (See Table 2).

The percentage of inorganic As in water is typically indistinguishable from 100% (Gault et al., 2005; Kile et al., 2007) and the BCF_w is therefore also considered 100% (Laparra et al., 2005).

The study on bioavailability of As in cooked rice is limited, the only data generated by Juhasz et al. (2006) stated that 90% of the As is bioavailable for rice varieties with high inorganic As content as applicable to this study.

The exposure duration is considered equal to the age under consideration for less than 40 years and equal to 40 years for all ages greater than that because in West Bengal the tube wells were first dug in 1950 s and 1960 s (Bagla and Kaiser, 1996) to provide clean water and thereby reduce deaths attributable to diarrhoeal diseases and it is therefore considered unlikely that any persons, whatever their age would have experienced substantially greater exposure than 40 years.

To determine the dose of As relating to cancer in an exposed population, the USEPA, (1998) has used a linearized multistage model which assumes that extrapolation from high doses to low doses is possible with a straight line and at low doses the slope of the dose–response curve is

Table 1
The published input variables used in the risk model

Input variable	Symbol	Fitted distribution	Parameter	Data source
Water intake rate (L/day)	IR_w	Constant	<18 years–2 >18 years–3.1	Watanabe et al. (2004) Chowdhury et al. (2000)
Rice intake rate normalised to body weight (gm/kg day)	IR_r	Constant	Male – 11.604 Female – 11.273	NNMB (2002)
Body weight (kg)	BW	Lognormal	Table 2	Burmaster and Crouch (1997) and NNMB (2002)
Bioconcentration factor	BCF_w BCF_r	Constant	Water 100% Rice 90%	Gault et al. (2005) Juhasz et al. (2006)
Exposure duration (years)	ED_w ED_r	Constant	Equal to the age (for age < 40) and equal to 40 (for age > 40)	Assumed based on the information by Bagla and Kaiser (1996)
Oral cancer potency slope factor $((mg/kg)/d)^{-1}$	CPSo	Constant	1.5	USEPA, IRIS (1998)
Life expectancy (years)	LT	Constant	Male – 61 Female – 63	WHO (2006)
Age		Empirical	Table 2	WHO (2001a)
Gender		Empirical	Male – 51.89% Female – 48.10%	WHO (2006)

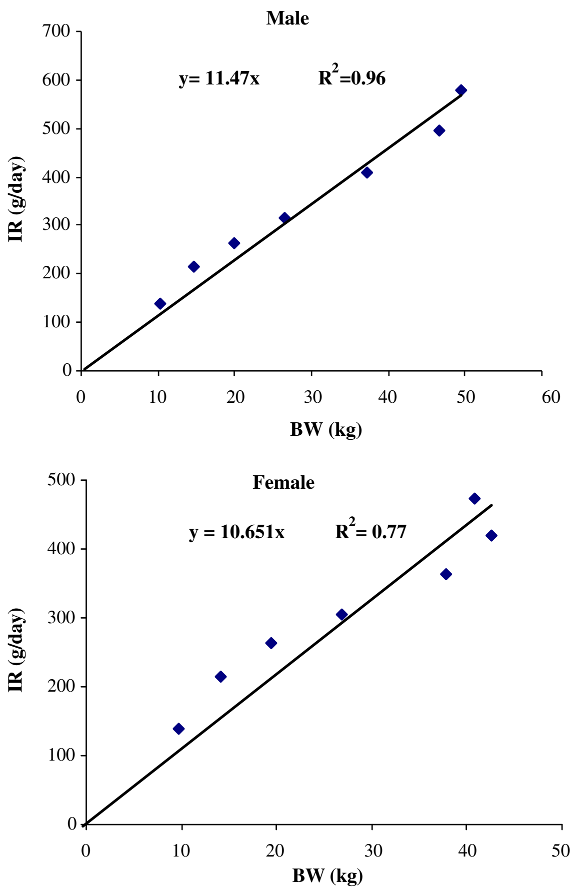

Fig. 3. Correlation between the rice ingestion rate and body weight for the population of West Bengal. Data from NNMB (2002).

Table 2
The age and gender dependent body weight distribution based on the NNMB (2002) database and age distributions as documented for India by the WHO (2001a)

Age	Proportion of population	Male		Female	
		Wt (kg)	SD	Wt (kg)	SD
<1	0.02	6.40	1.60	5.80	1.60
1–3	0.07	10.33	1.40	9.67	1.40
4–6	0.05	14.63	1.80	14.10	1.80
7–9	0.08	19.93	2.93	19.43	2.93
10–12	0.05	26.43	5.00	26.80	5.00
13–15	0.08	37.17	6.67	37.83	6.67
16–17	0.05	46.70	5.50	42.60	5.50
18–24	0.11	50.80	7.50	42.50	7.50
25–34	0.16	51.30	7.60	42.90	7.60
35–44	0.13	50.80	8.40	44.30	8.40
45–54	0.09	50.20	7.60	42.50	7.60
55–64	0.06	48.50	8.80	41.10	8.80
65–74	0.04	46.30	7.60	39.30	7.60
>75	0.02	48.90	9.10	33.60	9.10

represented by a slope factor (Davis and Cornwell, 1998). The USEPA toxicological database, IRIS, suggests an As oral cancer potency slope factor of 1.5 per (mg/kg)/day (USEPA, 1998). The reported and utilized factor is not age or gender dependent.

2.6. *Parameterising probability distributions for input variables*

Fitted distributions of the input variables for As concentration in water and rice, and inorganic As content of rice were established by statistical (χ^2 test) methods. Statistically the goodness of fit for the fitted distribution was determined by fitting different probability distributions (at least 10) to the data set and finally ranking them by χ^2 test value. Inspection of P–P graphs and Q–Q graphs was carried out to determine if there was any systematic variation in the magnitude of residuals. For the best fitted distribution the distribution parameters were estimated by maximum likelihood estimators. The software @Risk (Version 4.5, Students Edition, Palisade Corp., USA) was used in Microsoft Excel for this analysis.

2.7. *Monte Carlo simulation*

Monte Carlo simulation is a statistical model in which input variables to an equation are varied simultaneously (Paustenbach, 2000). To run the risk model using Monte Carlo simulation, the input values were chosen from variable distributions, with the frequency of a particular value being equal to the relative frequency of the variable in the distribution. The following 3 steps based on the methods by Paustenbach (2000) were carried out:

1. Probability distribution for each input variable was characterized and the distribution was specified for the Monte Carlo simulation.
2. For each iteration, one value was randomly selected from each input variable distribution, and Eq. (1) was run. 100000 iterations were performed in Microsoft Excel.
3. The output risks were rendered as cumulative probability plots.

Finally, the sensitivity of the cumulative probability plots for the input variables were determined by repeating the Monte Carlo simulation with each variable in term modified by the standard error of the measurement.

2.8. *Statistics*

Microsoft Excel 2003 and SPSS v.13 for windows were used for data analysis and statistical applications.

3. Results and discussion

3.1. *Analytical quality control data*

Arsenic concentration in the standard water reference material (SRM 1640) was measured as 28 ± 4 μg/L ($n = 3$) in agreement with the certified value of 26.6 ± 0.4 μg/L. Accuracy of total rice digestion and analysis was confirmed by a mean As total recovery of 99 ± 11% ($n = 9$) from a NIST1568a rice flour reference material. The mean deviation between duplicate sample analyses ($n = 8$) was 4.9% (0.0–14.8%). The As speciation of NIST 1568a rice flour reference material was determined to validate the method. Although not certified for As species, its speciation has been characterised prior to this study. Total inorganic As of 0.11 ± 0.06 mg/kg, DMA(V) of 0.13 ± 0.04 mg/kg, MMA(V) of 0.01 ± 0.02 mg/kg and total recovery ((sum of species recovered from chromatographic separation obtained from the TFA extraction/total[As] from HNO_3 extraction) × 100) of 82% observed were in agreement with previously published data (D'Amto et al., 2004; Heitkemper et al., 2001; Kohlmeyer et al., 2003; Williams et al., 2005).

3.2. As concentration in drinking water and rice

The mean concentration of As in drinking water for the survey area was around 17 μg/L (Table 3). This is lower than might have been expected for an area that has been previously been identified as an As-affected area (e.g., Chatterjee et al., 1995), in part reflecting differences between drinking water and well water compositions (see discussion) and in part because of government interventions to provide As-free drinking water: some of the water supplies were from recently installed Public Health Engineering Department (PHED) and Rural Water Supply Schemes (RWSS) which utilise low-As waters from greater than 100 m depth (Rahman et al., 2003).

The rice samples, which were either cultivated ($n = 22$) or purchased from local market ($n = 28$), had a mean concentration of 0.13 mg As/kg (Table 3). This is comparable to the studies of, Das et al. (2004) (0.136 ± 0.08 ($n = 10$) for a Bangladesh field survey) and Roychowdhury et al. (2002) (0.21 ($n = 6$) for a West Bengal household survey) but less than that reported by Williams et al. (2006) (0.08 to 0.51 mean As for different areas of Bangladesh ($n = 330$) for a field and market based study) and Ohno et al. (2007) (0.34 ± 0.15 ($n = 18$) for a household survey from a highly As-affected area of Bangladesh. It is noted that several authors, including Duxbury et al. (2003), have recorded a significant difference in the As content of boro (0.18, $n = 78$) and aman (0.11, $n = 72$) rice ($n = 150$) from Bangladesh field surveys. It was not possible to obtain comprehensive information from the households from which samples were collected to be able to determine the growing season of the rice sampled. It is also noted that variations in As content of rice may be related to a number of other factors including rice type (Signes et al., 2008; Williams et al., 2005).

No correlation was observed between the As concentration in raw rice and drinking water As from the 10 clusters ($r^2 = 0.05$). Indeed, As concentration in drinking water was significantly different among the clusters ($P < 0.001$) whilst no significant difference was observed for raw rice As concentrations between the clusters ($P > 0.05$) when one way analysis of variance was performed after normalizing the data. The lack of correlation of As concentration in rice and drinking water As concentrations is thought to reflect the greater variety of sources of rice and particularly from sources beyond the immediate area.

Speciation results confirmed that inorganic As was the predominant form (74 ± 13%) of As in all rice varieties in agreement with the studies by Williams et al. (2005) (80% inorganic As for Bangladeshi rice and 81% for Indian rice) and Sanz et al. (2005) (80% for Indian rice).

3.3. Effect of cooking of rice

The mean As concentration of 0.17 mg/kg observed in cooked rice was less than that reported by Smith et al. (2006b) (0.35, $n = 46$) for a Bangladesh household survey), Bae et al. (2002) (0.27, $n = 5$, for a Bangladesh on site survey), Rahman et al. (2006) (0.32, $n = 4$) for a Bangladesh field survey) and Roychowdhury et al. (2002) (0.37, $n = 9$) for a West Bengal household survey). The comparatively low concentrations of As in cooked rice in this study may be in part because of low-As in cooking water in

Table 3
Summary statistics of analyses of As in water and rice for the 10 clusters of the surveyed area of Chakdaha block

Clusters	Statistics	As in drinking water (μg/L)	As in cooking water (μg/L)	As in raw rice (mg/kg)	As in cooked rice (mg/kg)
Alaipur	Mean	11.60	11.60	0.10	0.12
	SD	8.11	8.11	0.02	0.05
Chakdaha	Mean	46.00	46.00	0.12	0.26
	SD	9.00	9.00	0.05	0.03
Chanduria	Mean	2.40	2.40	0.12	0.11
	SD	0.89	0.89	0.05	0.02
Kautukpur	Mean	1.47	1.47	0.09	0.07
	SD	0.92	0.92	0.03	0.03
Kumarpur	Mean	1.10	3.92	0.11	0.09
	SD	0.53	6.22	0.05	0.05
Maheserpur	Mean	42.60	44.20	0.15	0.25
	SD	26.50	25.95	0.03	0.05
Punglia	Mean	2.10	33.52	0.17	0.25
	SD	0.22	70.70	0.11	0.11
Saguna	Mean	7.90	8.57	0.14	0.12
	SD	12.04	8.68	0.09	0.13
Sutra	Mean	27.28	33.38	0.17	0.34
	SD	30.57	26.68	0.06	0.15
Uttarpanchpota	Mean	28.00	17.80	0.13	0.14
	SD	19.34	24.90	0.02	0.06
Total	Mean	17.30	20.57	0.13	0.17
	SD	21.67	29.44	0.06	0.11

households in the survey area. Mean cooking water As concentrations were not found to be significantly different from mean drinking water As concentrations (Table 3).

For rice samples cooked in water with As concentration less than 10 μg/L, the As content is proportional to the As concentration in the equivalent raw rice (Fig. 4A(I)) but when cooked in water with As concentration greater than 10 μg/L, a poor correlation was observed between As in cooked and raw rice (Fig. 4A(II)) – this supports the findings that the As concentration of cooked rice depends on As content of cooking water (Bae et al., 2002; Laparra et al., 2005; Sengupta et al., 2006; Signes et al., 2008). An increase in the As content of rice cooked in high-As water was also reported by Signes et al. (2008), but for As concentrations in water greater than 50 μg/L. In the present study such a relationship was only observed with As concentrations in cooking water greater than 10 μg/L (Fig. 4A(II) and 4B). The difference may relate to rice variety (Juhasz et al., 2006) and/or to the cooking method (Bae et al., 2002; Rahman et al., 2006; Sengupta et al., 2006).

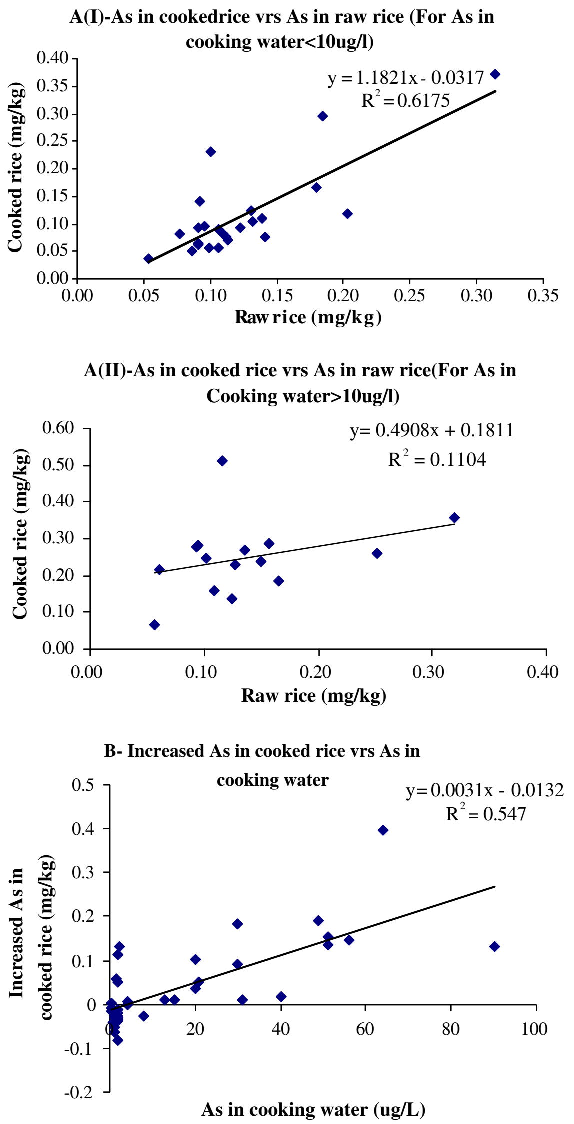

Fig. 4. Correlation between (A) As content of raw and cooked rice; and (B) increased As content of cooked rice with cooking water.

The mean cooking water As concentration in households from which cooked rice samples were obtained was 16 μg/L whereas that from households from which such samples were not obtainable was much higher – 36 μg/L. Although not significantly different, in part because of the large inherent variability of this parameter, this difference suggests that there may nevertheless be some systematic difference in the water quality of the two groups of households. It is speculated that households in which a rice meal was not being cooked at the time of survey may be of lower socio-economic status and may consequently have access to poorer quality drinking and cooking water. The regression equation for the relationship between As concentration in cooked rice and that in raw rice is shown in Fig. 4B and was used to model the As concentration in cooked rice samples from households ($n = 11$) in which such samples were not obtained.

3.4. Model input variables based on this study

The optimal fitted distributions, established by χ^2 tests, of the input variables for As concentration in water and rice, and the inorganic As content of rice are illustrated in Fig. 5.

The As concentration in water was best characterised by a BetaGeneral distribution of the form

$$f(x) = \frac{(x - \min)^{\alpha-1}(\max - x)^{\beta-1}}{B(\alpha,\beta)(\max - \min)^{\alpha+\beta-1}} \tag{2}$$

characterised by a set of parameters $(\alpha, \beta, \min, \max)$ where α and β are shape parameters greater than zero, min and max refer to the minimum and maximum observed values and $B(\alpha,\beta)$ is the Beta function.

The As concentration in raw rice was best fitted by LogLogistic distribution of the form

$$f(x) = \frac{\alpha \times t^{\alpha-1}}{\beta(1 + t^{\alpha})^2} \tag{3}$$

$$\text{where} \quad t = (x - \gamma)/\beta \tag{4}$$

and characterised by a set of parameters (γ, β, α) where γ is the location parameter and α and β are shape parameters greater than zero.

The As content in cooked rice was best fitted by LogNormal distribution of the form

$$f(In(x)) = \frac{e^{-\frac{(In(x)-\mu)^2}{2\sigma^2}}}{\sigma\sqrt{2\pi}} \tag{5}$$

characterized by set of parameters (μ, σ) where μ is the mean and σ is the standard deviation.

The inorganic As content was characterised by an ExtremeValue distribution function of the form

$$f(x) = \frac{1}{b}\left(\frac{1}{e^{z+\exp(-z)}}\right) \tag{6}$$

$$\text{where} \quad z = (x - a)/b \tag{7}$$

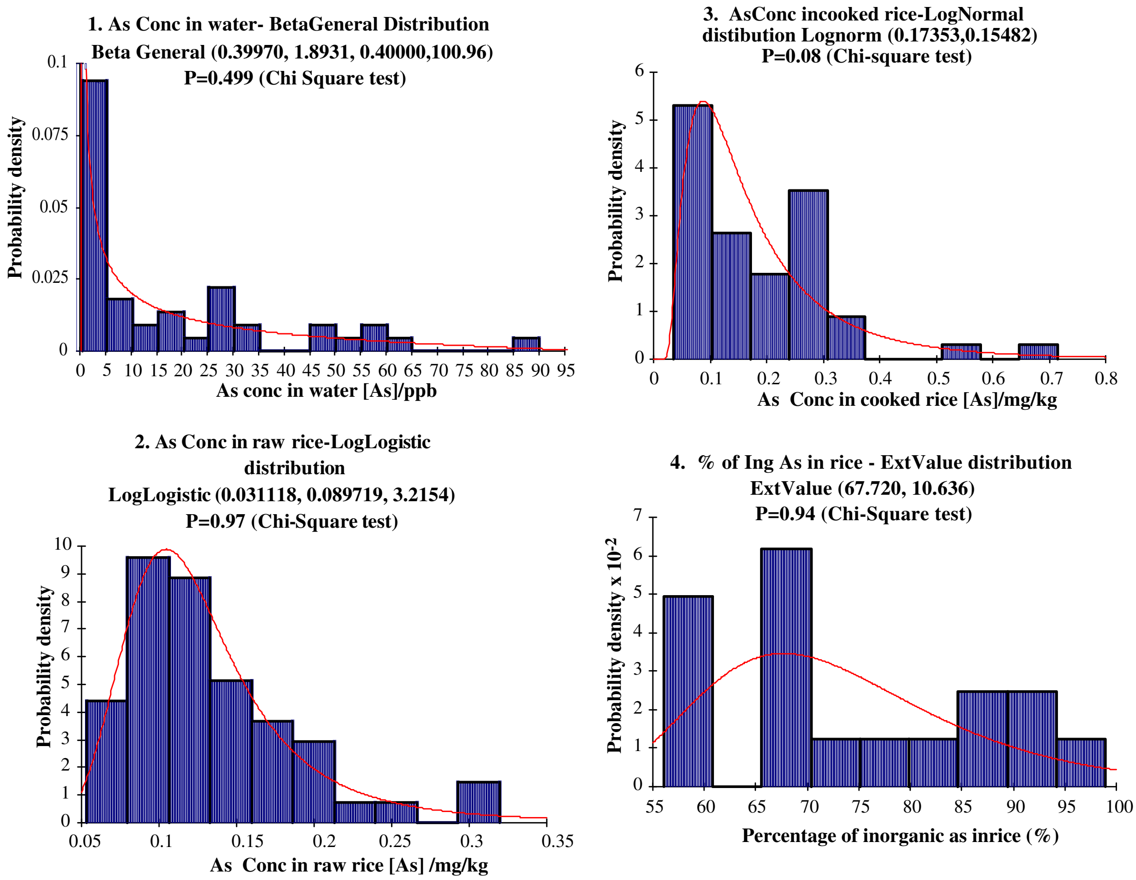

Fig. 5. Distributions of the input variables: (1) As concentration in water, (2) As concentration in raw rice, (3) As concentration in cooked rice and (4) inorganic As content of rice, established by statistical (χ^2 test) and graphical (probability plots) methods. The input data is represented by bars and the fitted distribution by the line in the plots.

and characterised by a set of parameters (a, b) where a is the location parameter and b is a scale parameter.

3.5. Risk plots

Risk plots derived from Monte Carlo simulations of Eq. (1) are presented in Fig. 6. Age and gender adjusted excess lifetime cancer risk from cooked rice intake had a median of 7.62×10^{-4} and was fractionally higher than the equivalent risk from water intake. The total excess lifetime median cancer risk of 1.48×10^{-3} was higher than the USEPA regulatory threshold target cancer risk level of 10^{-4}–10^{-6}. Risk from cooked rice intake was marginally higher than risk from water intake for this study area demonstrating that rice-related risks are of serious concern.

3.6. Sensitivity analysis

Changing the mean (baseline) input values for As concentration in water and rice, and the inorganic As content of rice by the standard error on the mean for these samples analysed led to a change by 9%, 6% and 7%, respectively, for the median output risk. Among the input variables from

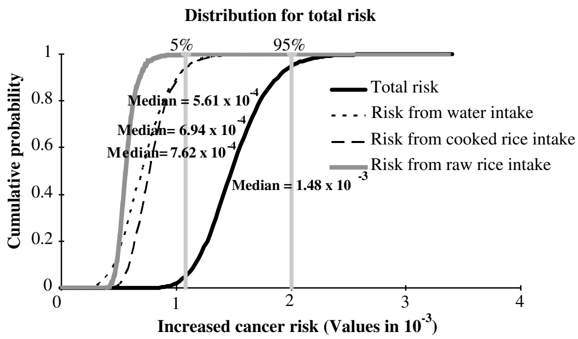

Fig. 6. Cumulative probability distributions of age and gender adjusted excess lifetime cancer risk from rice intake, water intake and both (total risk) per year for the studied population of Chakdaha. The 5th and 95th percentiles for the total excess lifetime cancer are also indicated. Note that the 10^{-4}–10^{-6} range typically used by the USEPA as a threshold to guide determination of regulatory values would plot at values from 0.001×10^{-3} to 0.1×10^{-3}.

the published data the maximum change in the output risk estimate was for a baseline 10% change for exposure duration and CPSo.

3.7. Benefits of probabilistic assessment

The probabilistic approach to risk assessment has proved its value in taking into account the variabilities and parameter uncertainties (Oberg and Bergback, 2005). Whereas a deterministic approach will provide only a single point estimate of risk, using the probabilistic risk assessment approach, it has been possible to determine the 5th percentile (1.09×10^{-3}) and 95th percentile (2.01×10^{-3}) values for total cancer-related risks due to combined water and rice exposure (Fig. 6) – this will ultimately have particular relevance to determining high risk sub-groups of the population.

3.8. Limitations

There are necessarily various limitations to the calculations that have been carried out in this study. Some of these are discussed below and are used as a basis for highlighting fruitful areas of further research. This study was limited to a small sample size for the study area but is nevertheless considered to be broadly representative of the study population, as discussed in Section 3.2. Limitations due to sample size might be overcome in future studies by substantially increasing the sample size and also by using regression-kriging techniques (Rodriguez-Lado et al., 2008) to interpolate As-in-groundwater concentration data between the sample points.

Arsenic concentrations in drinking water analysed in this study were found to exceed the WHO provisional guide value of 10 μg/L in 44% of cases and to exceed the Indian permissible limit of 50 μg/L in 11% of cases. However, in the larger database summarised by SOES, 2007 a much greater proportion of samples from tube wells were found to exceed these limits, viz. 55% of samples with >10 μg/L and 22% with >50 μg/L. Of course, the two sets of data are not directly comparable. In this study drinking waters have been analysed from households, whereas the SOES, 2007 dataset describes tube well water, which may or may not be used as drinking water. Thus, the difference in the two sets of data are ascribed to intelligent transfer by users of drinking water supplies to lower As concentration supplies, including those supplied by the Government of West Bengal through its various schemes. This highlights that human health risk estimates based upon tube well water As concentrations (e.g. Samadder and Subbara, 2007) rather than more representative drinking water As concentrations may be somewhat overestimated.

Exposure from ingestion pathways other than rice and water have not been considered even though some of the vegetables consumed in this area like arum can contain as much as 1.5 mg/kg As (Huq et al., 2006). Therefore, total As exposure from food ingestion may have been somewhat underestimated, however the frequency of consumption of these vegetables is not regular (Huq et al., 2006) and it is difficult to quantify what bias may have been introduced by this limitation to the survey.

The variable, water intake rate, is thought to be rather inaccurate as information on the daily water intake has been lacking for West Bengal and other developing countries. Both of the studies by Watanabe et al. (2004) and Chowdhury et al. (2000) from which the data has been derived for the model are limited to point estimates. Age and gender distributions for water intake in the USA (Hope, 1999) indicate significant variation as a function of age and gender.

Only point data for rice and water exposures has been obtained both of which are likely to exhibit seasonal and secular variations. Fletcher et al. (2006), for example, noted significant differences between current As concentrations in drinking water (median 1.4 μg/L; minimum 0.1 μg/L; maximum 94.8 μg/L) and lifetime average concentrations in drinking water (median 2.1 μg/L; min 0.1 μg/L; maximum 181 μg/L) in a study of As exposure in eastern Europe. These differences reflect improvements in drinking water quality either due to changes in supply or changes in water procurement behaviour over a period of years. Secular changes in As exposure from drinking water are likely to be even more substantive in West Bengal, where the recent installation of tube wells has resulted in massive changes in As exposure, and where ongoing mitigation efforts by the Government of West Bengal and other agencies are aimed to substantially reducing such exposure in the future.

The limited database on exposure duration, the bioconcentration factor for rice and dose–response relationship as discussed in Section 2.5, has resulted in their general point estimates being entered in the model limiting the variabilities and uncertainties to be ascertained.

4. Conclusions

Arsenic-in-rice-related human health risks are of serious concern in West Bengal, a median lifetime cancer risk from cooked rice of 7.62×10^{-4} being calculated for the population in Chakdaha block considered in this study, is higher than the 10^{-4}–10^{-6} range typically used by the USEPA as a threshold to guide determination of regulatory values. The median total risk from combined rice and water intake was 1.48×10^{-3}. The contributions to this median risk from drinking water, rice and cooking of rice were found to be 48%, 44% and 8%, respectively. Thus, rice is a major potential source of As exposure in the As-affected study areas in West Bengal and the most important exposure pathway for groups exposed to low or no As in drinking water.

The influence of the cooking method and As concentration in cooking water on the As concentration of cooked rice confirms that modification of rice cooking methods may be a cheap way of reducing As exposure, although measures addressing As concentrations in drinking water and irrigation waters have the potential to be ultimately quantitatively more significant.

The probabilistic risk assessment model presented here is very much limited by the accuracy and representativeness of the input data, but constitutes a framework upon which improved data can readily be added to establish a tool to inform policy makers considering the relative merits of various As mitigation schemes. In particular, the future provision of suitably spatially distributed age, gender and other socio-economic data will enable probabilistic risk assessment models to better identify at-risk areas and populations within West Bengal and also elsewhere in the world where similar groundwater As hazards exist.

Acknowledgements

We acknowledge and thank Professor Dipankar Chakraborti (School of Environmental Studies at Jadavpur University) and Professor Debashis Chatterjee (Chemistry Department, University of Kalyani) and their groups for their kind help during the field surveys, Paul Lythgoe and Dr. Andrew Gault for their help in developing the analytical methods and Mike Lawson for his help during the field work. DM acknowledges the receipt of a NERC funded Dorothy Hodgkins postgraduate award. Aspects of this study were presented at the launch of the UKIERI/British Council funded PRAMA project at IICB, Kolkata in January 2008: we thank Ashok Giri, Bhaswati Ganguli, D.N. Guha Mazumder and Ross Nickson for discussions at and subsequent to this meeting. We particularly thank Tony Fletcher and two anonymous referees for detailed review comments that enabled us to considerably improve the clarity of the manuscript.

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