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ARTICLE

Arsenic in irrigated water, soil, and rice: perspective of the cropping seasons

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Abstract Arsenic contamination of shallow groundwater and related health problems are threats for the millions in endemic regions of West Bengal. Contamination of rice grain creates the food chain pathway of mineral arsenic besides drinking water contamination. Present study concentrated on association of arsenic concentration in irrigated water, paddy field soil and rice with the cropping seasons. Irrigated ground water arsenic concentration decreased significantly ($p = 0.007$) from summer (median 0.42 mg l^{-1}) to winter (median 0.35 mg l^{-1}). Carried over effect created significant decrease ($p = 0.03$) of paddy field soil arsenic concentration from summer (median 8.35 mg kg^{-1}) to winter (median 6.17 mg kg^{-1}). Seasonal variation was observed in rice straw ($p = 0.03$) but not in husk ($p = 0.91$). Arsenic concentration decreased significantly ($p = 0.05$) in the rice grains collected in winter season (median 0.23 mg kg^{-1}) than the samples collected in the summer season (median 0.30 mg kg^{-1}). In conclusion, seasonal effects need to be considered in case of human health risk assessment from arsenic consumption.

Keywords Arsenic · Ground water · Rice grain · Cropping season

Introduction

Elevated levels of arsenic in groundwater is confirmed in some states, like West Bengal, Bihar, Uttar Pradesh, Assam, Jharkhand, Chhattisgarh, and Madhya Pradesh (Chakraborti et al. 2002; Acharyya and Shah 2006) in India. Arsenic is considered as human carcinogen (Ellington et al. 1991; NRC 2001). Long time arsenic exposure may cause melanosis, keratosis, non-pitting edema, gangrene, leucomelanosis, apart from increasing cancer risk of skin, bladder, and lungs (Guha Mazumder et al. 2000; Kile et al. 2007; Mondal et al. 2010). Especially boro rice is the major route of arsenic exposure in arsenic affected parts of West Bengal (Mondal and Polya 2008) due to the excessive requirement of ground water irrigation, extracted from shallow aquifers contaminated with arsenic (Alam and Sattar 2000; van Geen et al. 2006; Meharg and Rahman 2003). Harvested vegetables are also source of arsenic (Roychowdhury et al. 2003; Samal et al. 2011; Biswas et al. 2012). Elevated arsenic concentration in rice straw and grain is phytotoxic also (Abedin et al. 2002a). Transfer of arsenic from soil to rice straw into grain and finally human consumption through rice is an additional arsenic consumption pathway for human (Roychowdhury et al. 2002, 2003, 2008; Samal et al. 2011) although arsenic intake through drinking water is sometimes limited (below maximum permissible limit). Several field studies conducted in arsenic endemic parts of Bangladesh and Nadia district, West Bengal concluded that irrigation with arsenic enriched groundwater causes elevated arsenic concentrations in the upper field soil layers (Rahman et al. 2008; Bhattacharya et al. 2010; Khan et al. 2010). Investigation on the effect of season on paddy fields soil in Bangladesh concluded that the fields those are not deeply flooded during monsoon season and nearly all arsenic added through irrigation, retained in the soil (Dittmar et al. 2007; Norra et al. 2005; Panaullah et al. 2009). Arsenic concentration in topsoil of paddy fields are controlled by depth of pedon and mineralogy (Ghosh et al.

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2006) as well as by pronounced monsoon flooding showing significant decrease during the monsoon (Khan et al. 2009; Roberts et al. 2010). Field studies in Bangladesh concluded that irrigation with arsenic rich groundwater resulted in a continuing accumulation of arsenic in paddy field soils (Hossain et al. 2008; Panaullah et al. 2009), which may be severe in regions those were not intensely flooded during monsoon season (Roberts et al. 2010). Thus, some variations of arsenic concentration especially in rice field soil exist as there are temporal variations of irrigation water arsenic concentration (Roberts et al. 2010).

Information regarding variability of arsenic concentration on l seasonal scale particularly in the Gangetic delta part of Nadia district, West Bengal is insufficient till date. Study was carried out to find any significant difference in arsenic concentration in irrigated water, soil to rice plant parts.

Material methodology

Sampling sites selection

Sampling sites were selected in the 5 km east of the river Ganges in Nadia district. Since early 1980s, there is a practice of *Boro* rice cultivation depending on ground water irrigation. During the rice growing period from December to June (RKMP 2011), the fields are irrigated with ground water from shallow (depth 30–60 m) aquifers (summer rice). Alternatively, another rice cultivation period is winter or monsoon rice, known as *Aman*, grown from June to December (RKMP 2011). This typification is on the basis of crop harvesting periods (RKMP 2011). Four blocks: Chakdaha, Haringhata, Shantipur, and Krishnanagar (sampling sites in onwards texts) were selected for samples collection (Table 1). The occurrence of arsenic is well documented in the irrigated ground water (Chakraborti et al. 2002; Roychowdhury et al. 2003; Das et al. 2004), in the paddy field soil (Norra et al. 2005; Biswas and Santra 2011) and harvested rice (Roychowdhury et al. 2002, 2003; Mondal and Polya 2008; Biswas and Santra 2011) and vegetables (Das et al. 2004; Biswas et al. 2012) in these blocks.

Samples collection

Irrigated water samples were collected (50 ml) in polyethylene bottle during the uniform rate of discharge from the irrigation sources as per availability or else directly from the field, and were acidified with one drop of concentrated nitric acid (HNO_3; Merck, Germany). Composite soil samples (following standard soil sampling procedure) were collected from field surface (0–15 cm depth).Composite sampling of straw, husk, and rice grain was done directly from the fields. Rice plants were harvested by cutting at 4 cm above the soil.

Total arsenic estimation

Water samples were filtered through 0.45 μm filter and were kept in 4 °C till further analysis. Soil samples were immediately sun dried followed by hot air oven drying at 60 °C until constant weight. Spikelet containing rice grains were separated from the panicles by hand. Rice grains were separated from husks using pestle and mortar followed by grinding. Rice straw samples were washed with tap water followed by de-ionized water and dried in hot air oven at 60 °C for 72 h. Oven dried soil and plant parts samples were made fine homogenized powder by grinding.

Soil, rice straw, husk, and rice grain were digested in acid on the heating block (Rahman et al. 2007; Biswas

Table 1 Arsenic concentration range in irrigated water (mg l^{-1}), soil and rice plant parts (mg kg^{-1} dry weight) samples collected from fields in different season; sampling sitewise distribution

	Chakdaha		Haringhata		Shantipur		Krishnanagar	
	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter
Irrigated water	0.48–0.67	0.34–0.49	0.40–0.70	0.15–0.55	0.41–0.65	0.18–0.54	0.44–0.61	0.09–0.54
	($n = 23$)	($n = 20$)	($n = 24$)	($n = 21$)	($n = 24$)	($n = 18$)	($n = 23$)	($n = 19$)
Soil	2.73–9.84	1.64–6.16	2.45–14.09	2.41–5.24	6.39–13.48	5.18–8.75	3.42–9.51	3.09–7.28
	($n = 23$)	($n = 23$)	($n = 24$)	($n = 24$)	($n = 24$)	($n = 24$)	($n = 23$)	($n = 23$)
Straw	0.81–1.35	0.57–0.89	0.75–1.51	0.81–1.24	1.03–1.70	1.20–1.78	0.72–1.44	0.75–0.83
	($n = 30$)	($n = 30$)	($n = 36$)	($n = 36$)	($n = 23$)	($n = 23$)	($n = 35$)	($n = 35$)
Husk	0.18–0.98	0.28–0.70	0.62–0.92	0.35–0.91	0.77–1.08	0.42–0.98	0.43–0.91	0.87–1.14
	($n = 30$)	($n = 30$)	($n = 36$)	($n = 36$)	($n = 23$)	($n = 23$)	($n = 35$)	($n = 35$)
Rice grain	0.18–0.32	0.09–0.26	0.21–0.36	0.19–0.35	0.20–0.58	0.09–0.31	0.57–0.92	0.43–0.73
	($n = 30$)	($n = 30$)	($n = 36$)	($n = 36$)	($n = 23$)	($n = 23$)	($n = 35$)	($n = 35$)

et al. 2012). The digested samples were cooled down to room temperature and filtered with Whatman No. 42 filter paper; filtrate was used for analysis against internal laboratory working arsenic standards using flow injection hydride generation atomic absorption spectrophotometer (FI-HG-AAS; Perkin Elmer A Analyst 400). Lower detection limit was 0.03.

Quality control of total arsenic estimation

Instrument was calibrated using matrix-matched standards. In each analysis batch, reagent blanks, internationally certified reference material and five analytical duplicates were included in the acid digests to assess precision and accuracy for chemical analysis. Precision and accuracy level of analysis was evaluated by analyzing standard reference rice flour sample (SRM-1568a, NIST; USA) following same treatment procedure ($n = 9$; three replicas, three sample batch) as the samples. We could record 96 % recovery of the total arsenic in SRM.

Statistical analysis

All the data were kept and analyzed using the statistical package MINITAB 14. The aims and objective hypothesis was tested in 95 % confidence level over the normally distributed data.

Result and discussion

Arsenic in irrigated water and soil

In the present study, we focused on the seasonal variation of arsenic concentration in irrigated water and soil in the endemic regions. Median arsenic concentration in the irrigated ground water samples collected in the summer and winter seasons were 0.56 and 0.53 mg l^{-1} ($p = 0.07$); 0.38 and 0.34 mg l^{-1} ($p = 0.05$); 0.41 and 0.33 mg l^{-1} ($p < 0.05$); 0.42 and 0.37 mg l^{-1} ($p < 0.05$), respectively, for sampling sites Chakdaha, Haringhata, Shantipur, and Krishnanagar (Fig. 1). Sampling sites wise range of arsenic concentration (mg kg^{-1}) in irrigated water samples collected for both the sampling seasons have been reported in Table 1. On the whole, the median arsenic concentration in irrigation water was 0.42 mg l^{-1} (range 0.40–0.58 mg l^{-1}) in summer season. Whereas arsenic concentration ranged from 0.34 to 0.53 mg l^{-1} (median 0.35) in winter season samples (Table 2). The significant ($p = 0.007$) decrease of arsenic concentration in the irrigated ground water used in the winter rice than the summer rice is attributed to the fact of influence of rain water and resultant dilution of the dissolved arsenic as also described in the case of arsenic concentrations in some rivers in France (Masson et al. 2007). In another report on the seasonal effect on arsenic concentration in tube well waters in West Bengal, Savarimuthu et al. (2006) could not explain the possible cause of the difference except the effect of rain water. In the present study also, we assume the same cause behind the seasonal variation of irrigated water arsenic concentration as the samples sizes and sampling design are not sufficient to conclude on any other relative possibilities. Irrespective of sampling seasons, the water arsenic concentration was higher than allowable concentration in irrigation water for crops (0.1 mg l^{-1}) and allowable concentration in drinking water used for livestock (0.2 mg l^{-1}) given by USEPA (Ellington et al. 1991). The consequences were field soil with over load of mineral arsenic due to arsenic rich irrigated water.

Sampling sites wise arsenic concentration (mg kg^{-1} dry weight) range in soil samples collected for both the sampling seasons have been reported in Table 1. Median arsenic concentration in the irrigated paddy field soil samples collected in the summer and winter seasons were 4.38 and 3.14 mg kg^{-1} ($p = 0.08$); 7.09 and 4.02 mg kg^{-1} ($p < 0.05$); 9.81 and 7.13 mg kg^{-1} ($p < 0.05$); 5.72 and 5.09 mg kg^{-1} ($p = 0.09$), respectively, for sampling sites Chakdaha, Haringhata, Shantipur, and Krishnanagar

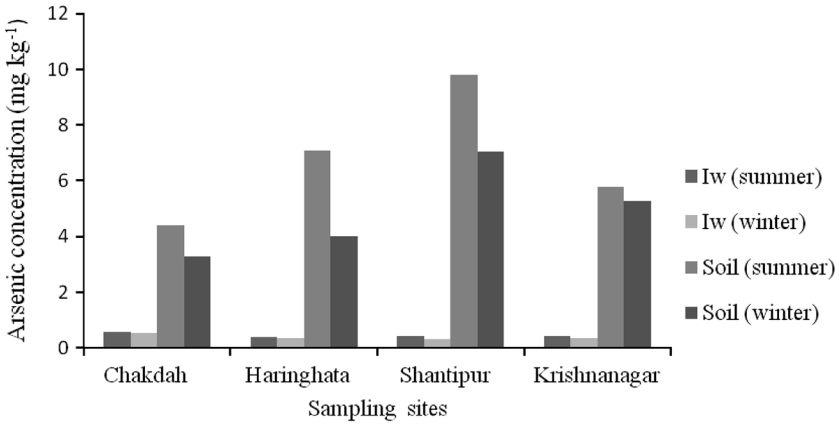

Fig. 1 Arsenic concentration in irrigated water (Iw) and soil in samples from different sampling sites

Table 2 Arsenic concentration in irrigated water (mg l^{-1}), soil, and rice plant parts (mg kg^{-1} dry weight) samples collected from fields in different seasons

Samples	Median arsenic concentration summer	Median arsenic concentration winter	t	p value
Irrigated water	0.42 ($n = 94$)	0.35 ($n = 78$)	6.73	0.007
Soil	8.35 ($n = 94$)	6.17 ($n = 94$)	3.96	0.03
Straw	0.96 ($n = 124$)	0.85 ($n = 124$)	2.89	0.03
Husk	0.75 ($n = 124$)	0.72 ($n = 124$)	0.12	0.91
Rice grain	0.33 ($n = 124$)	0.23 ($n = 124$)	3.08	0.05

(Fig. 1). Arsenic concentration in soil ranged from 5.82 to 9.72 mg kg^{-1} (median 8.35 mg kg^{-1}) in the samples of summer season, whereas in the soil samples of winter season arsenic concentration range was 5.01–7.34 mg kg^{-1} (median 6.17 mg kg^{-1}) (Table 2). Although there is no limit set for arsenic in soil, in both the seasons arsenic concentration in soil exceeded the upper limit of 5.0 mg kg^{-1} for the USEPA Toxicity Characteristic Leaching Procedure (TCLP) (Ellington et al. 1991). We assume the cause behind the variation of soil arsenic concentration over season is the difference of irrigated water arsenic concentration.

Arsenic in rice plant parts

In the previous section, we ended with the seasonal variation of arsenic concentration in irrigated water and resulted variation in rice field soils. In the same way, rice plant parts samples were accounted to evaluate any possible variation of arsenic concentration in rice plant parts depending on cultivation seasons.

Median arsenic concentration (mg kg^{-1} dry weight) in rice straw samples for the summer and winter season were 0.90 and 0.78 mg kg^{-1} ($p = 0.38$); 1.03 and 0.91 mg kg^{-1} ($p = 0.13$); 1.32 and 1.21 mg kg^{-1} ($p = 0.08$); 0.81 and 0.78 mg kg^{-1} ($p = 0.25$) respectively for sampling sites Chakdaha, Haringhata, Shantipur, and Krishnanagar (Fig. 2). Median total arsenic concentration in rice straw ranged from 0.72 to 1.20 mg kg^{-1} (median 0.96) in summer, whereas the range was between 0.86 and 1.70 mg kg^{-1} (median 0.85) in the samples collected during winter season (Table 1). The significant variation ($p = 0.01$), when considered as a whole, depicted the influence of season on rice straw arsenic concentration. The wide range of rice straw arsenic concentrations could be the cause of not considering the rice cultivar effects, because the presence of arsenic in rice straw is directly related to rice plant physiology (Abedin et al. 2002b). Irrespective of seasons, the rice straw arsenic concentrations are high enough to be problems for the cattle (though there are not any satisfactory safe levels of arsenic concentration in straw) as it is widely used as fodder in the study areas.

Median arsenic concentration (mg kg^{-1} dry weight) in the rice husk was 0.75 mg kg^{-1} (range: 0.67–0.79 mg kg^{-1}) and 0.72 mg kg^{-1} (range 0.40–0.87 mg kg^{-1}), respectively, for the summer and winter with no significant difference ($p = 0.91$) over season (Table 2). Summer and winter season arsenic concentration median values in the rice husks were 0.62 and 0.41 mg kg^{-1}; 0.79 and 0.69 mg kg^{-1}; 0.78 and 0.81 mg kg^{-1}; 0.71 and 0.82 mg kg^{-1}, respectively, for sampling sites Chakdaha, Haringhata, Shantipur, and Krishnanagar (Fig. 2). Sampling sites wise arsenic concentration range over the season was reported in Table 1.

Most interesting part of our study was significant differences ($p = 0.05$) of arsenic concentration (mg kg^{-1} dry weight) in raw rice samples. Arsenic concentration in rice grain ranged from 0.23 to 0.61 mg kg^{-1} (median 0.33 mg kg^{-1}) in the summer. In the winter season arsenic

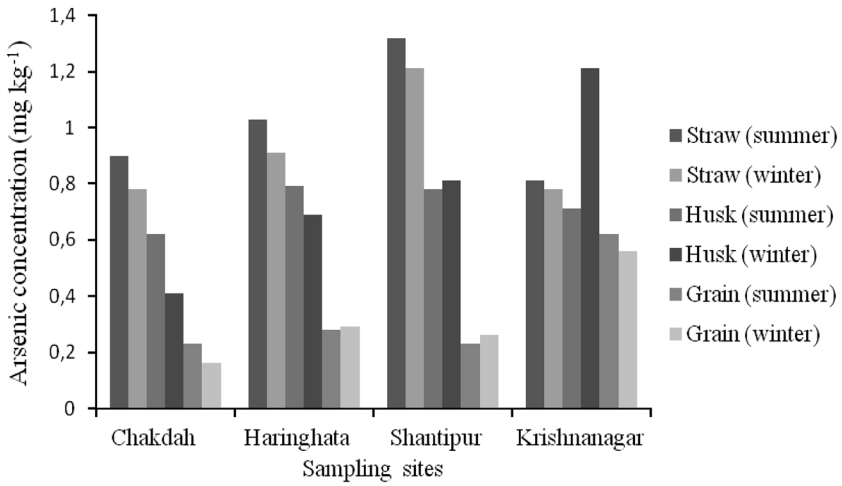

Fig. 2 Arsenic concentration in rice plant part samples from different sampling sites

concentration in rice grain ranged from 0.16 to 0.38 mg kg^{-1} (median 0.23 mg kg^{-1}) (Table 2). Median arsenic concentration in the rice grain samples collected in the summer and winter seasons were 0.23 and 0.16 mg kg^{-1} ($p < 0.05$); 0.29 and 0.26 mg kg^{-1} ($p = 0.05$); 0.20 and 0.26 mg kg^{-1} ($p < 0.05$); 0.62 and 0.56 mg kg^{-1} ($p < 0.05$), respectively, for sampling sites Chakdaha, Haringhata, Shantipur, and Krishnanagar (Fig. 1). Sampling sites wise arsenic concentration range over the season was reported in Table 1. Besides the role of rice plant physiology (Abedin et al. 2002a), we can assume, the availability or supply of mineral arsenic through irrigated water can play role in the mechanism for decrease of arsenic concentration in rice grains from the summer season samples to the winter season samples. Although there was seasonal variation of arsenic occurrence in rice grains, samples from neither of the seasons exceeded the WHO permissible limit of arsenic in rice (1.0 mg kg^{-1}). But obviously, it will finally be hazardous for human depending on cooking (Rahman et al. 2006) and amount of serving a day (Biswas et al. 2013).

Conclusion

Arsenic in food chain and related human health risk is of serious concern in the arsenic endemic parts of West Bengal. Arsenic rich ground water irrigation creates the food chain pathways for human and other dependents. Rice is susceptible to arsenic accumulation. Study concentrated on seasonal pattern of arsenic occurrence starting from irrigated ground water to rice grain. We got significant effect of season on the arsenic concentration of irrigated ground water, paddy field soil and rice grains. The total risk assessment from arsenic is also needed an insight of seasonal variation.

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