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Transfer of Arsenic from Groundwater and Paddy Soil toRice Plant (*Oryza sativa* L.): A Micro Level Study in West Bengal, India

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Abstract: Rice (*Oryza sativa* L.) is one of the major food crops in many countries. As the cultivation of rice requires huge volume of water, long term use of arsenic contaminated groundwater for irrigation may result in the increase of arsenic concentration in the agricultural soil and eventually accumulation in rice plants. Thus a micro level study was conducted to investigate the transfer of arsenic from irrigation water and soil to rice plants in the arsenic affected 17 villages of Chakdaha block, Nadia district, West Bengal, India. Results showed that the level of arsenic in irrigation water (0.11±0.012 and 0.76±0.014 mg l^{-1}) was very much above the WHO permissible limit of 0.01 mg l^{-1} for drinking water and was also above the FAO permissible limit of 0.10 mg l^{-1} for irrigation water. The paddy soil gets contaminated by the irrigation water and thus enhances more probable condition for bioaccumulation of arsenic in rice plants. The total soil arsenic concentrations ranged from 1.38±0.108 to 12.27±0.094 mg kg^{-1} dry weight of arsenic, which was below the maximum acceptable limit for agricultural soil of 20.0 mg kg^{-1} as recommended by the European Community. In the rice plant, the highest accumulation of arsenic was noticed in the root (7.19±0.166 to 18.63±0.155 mg kg^{-1}) and the lowest in the grain (0.25±0.014 to 0.73±0.009 mg kg^{-1}). Regardless of the sampling locations the arsenic accumulation follows the order of root > straw > husk > grain. Consumption of rice straw containing considerable amount of arsenic (1.17±0.014 to 4.15±0.033) by cattle could potentially lead to increased arsenic levels in meat or milk. As because the total amount of arsenic in raw rice is not taken in human body due to its distribution in root, straw, husk and grain parts and also because in any rice sample from the study area concentration of arsenic in the grain part did not exceed 1.0 mg kg^{-1} dry weight of arsenic (the permissible limit of arsenic in rice according to WHO recommendation), thus atleast for now rice has remained harmless for consumption in the study area. But the results clearly showed that the arsenic content in rice plant is correlated to the degree of arsenic contamination of irrigation water and soil.

Key words: Arsenic, Bioaccumulation, Rice (*Oryza sativa* L.), Irrigation water, West bengal

INTRODUCTION

Groundwater arsenic contamination in the Bengal Delta Plain has been termed as the largest mass poisoning in history of human kind [1]. Groundwater of nine out of total eighteen districts of West Bengal has been contaminated with arsenic [2, 3, 4]. Among them, the groundwater arsenic concentration in Nadia district is of deep concern in terms of level of arsenic and area coverage [4]. Anthropogenic activities, such as pesticide and herbicide applications, mining and irrigation with contaminated groundwater have significantly enhanced arsenic levels in agricultural soil in many parts of the world [5]. There are various reports about the elevation of arsenic concentration in agricultural fields of West Bengal [6,7]. So, there is a possibility of transfer of arsenic from contaminated irrigation water and soil to rice plant (*Oryza sativa* L).

Arsenic accumulation in rice is viewed as a newly recognized disaster for South-East Asia, where rice is a staple food [8]. Rahman *et al.* [7] reported that regardless of rice varieties, accumulation of arsenic was 28 and 75 folds higher in root than that of shoot and raw rice grain, respectively. This trend of higher arsenic accumulation in root was found to be concurrent with the reports by Samal [4], Abedin *et al.* [9] and Liu *et al.* [10]. Abedin *et al.* [9] reported accumulation of arsenic (upto 92.0 mg kg^{-1}) in rice straw through a green house pot experiment. The presence of high levels of arsenic in the rice straw is a potential threat to cattle that consume

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the contaminated straw and thus indirectly to human health, via presumably contaminated bovine meat and milk [9, 11]. Rice grain has been reported to accumulate arsenic upto 2.0 mg kg^{-1} by Meharg *et al.* [12], Islam *et al.* [13] and Delowar *et al.* [14], which is much above the permissible limit in rice (1.0 mg kg^{-1}) according to WHO recommendation. Ohno *et al.* [15] concluded that the average contribution to total arsenic intake from drinking water was 13%, whereas from cooked rice it was 56%, thus making it clear that rice contributed most to the daily arsenic intake.

Most of the published reports focused mainly on the uptake of arsenic in the rice plant irrigated with arsenic contaminated water and soil through a green house pot experiment [9,11,14]. However, the field level investigation is quite inadequate on this aspect. Thus, the objective of the present study was to determine the concentration of arsenic in irrigation water, paddy field soil and in the rice plant cultivated with the arsenic contaminated irrigation water and soil in the Chakdaha block of Nadia district (West Bengal, India). This study would help to evaluate the transfer of arsenic from irrigation water and paddy soil to the rice plant.

MATERIALS AND METHODS

Study Area: Nadia district of West Bengal, India is highly contaminated with arsenic [4]. Chakdaha block of Nadia district has been chosen for the present study where the level of arsenic in groundwater is frequently exceeding WHO permissible limit (0.01 mg l^{-1}) for drinking water [16] and FAO permissible limit for irrigation water (0.10 mg l^{-1}) [17]. Samples were collected from all the 17 villages of the Chakdaha block namely Tatla 1, Tatla 2, Ghetugachi, Dubra, Dewli, Hingnara, Shilinda-1, Shilinda-2, Chanduria-1, Chanduria-2, Madanpur-1, Madanpur-2, Sarati, Saguna, Simurali, Routari and Kanchrapara.

Sample Collection: Irrigation water samples have been collected from the shallow tube well pumps which are used for irrigation in the study area. Prior to sample collection, the pumps were kept running for about 10-15 minutes in order to get an uniform rate of discharging water. Then the water samples were collected in polyethylene bottles and preserved with concentrated HNO^3. Soil and rice samples were collected by composite sampling from the fields irrigated with the arsenic contaminated water and transferred to airtight polyethylene bags. Soil samples were collected from 10-15 cm depth in a 2 m^2 area and rice plant samples were collected from a selected plot of 2 m^2 areas during harvesting time.

Sample Treatment: The irrigation water samples were filtered using 0.45 Millipore filter paper and were kept in polyethylene bottles at 4°C for analysis. The soil samples after collection were immediately sun dried and were later dried in the Hot Air Oven at 60°C for 72 hours. The dried soil samples were then grinded and passed through 2.0 mm pore sized sieve to get homogenized representative powder sample. Finally the samples were stored in airtight polyethylene bags at room temperature. The rice plant samples were washed thoroughly with tap water to remove soil and other contaminants and finally rinsed with de-ionized water with continuous shaking for several minutes. The samples were then dried in the Hot Air Oven at 60°C for 72 hours and were stored in airtight polyethylene bags at room temperature. Proper care was taken at each step to minimize any sort of contamination.

Sample Digestion: Soil samples, root, straw, husk and grain portions of the rice samples were digested separately following heating block digestion procedure [7]. About 0.5 g of the sample was taken into clean dry digestion tubes and 5 ml of concentrated HNO_3 was added to it. The mixture was allowed to stand over night under fume hood. In the following day, the digestion tubes were placed on a heating block and heated at 60°C for 2 hours. The tubes were then allowed to cool at room temperature. About 2 ml of concentrated $HClO_4$ was added to the plant samples. For the soil samples 3 ml of concentrated H_2SO_4 was added in addition to 2 ml of concentrated $HClO_4$. Then the tubes were heated at 160°C for about 4-5 hours. The heating was stopped when the dense white fume of $HClO_4$ was emitted. The content was then cooled, diluted to 25 ml with de-ionized water and filtered through Whatman No. 42 filter papers for soil samples and Whatman No. 41 filter papers for plant samples and finally stored in polyethylene bottles. Prior to sample digestion all glass goods were washed with 2% HNO_3 followed by rinsing with de-ionized water and drying.

Sample Analysis: Total arsenic of the digested soil, rice plant (root, straw, husk and grain) and irrigation water samples were analyzed by flow injection hydride generation atomic absorption spectrophotometer (FI-HG-AAS, Perkin Elmer Aanalyst 400) using external calibration through arsenate as standard [18]. The optimum Hcl concentration was 10% v/v and 0.4%

$NaBH_4$ produced the maximum sensitivity. For each sample three replicates were taken and the mean values were obtained on the basis of calculation of those three replicates. Standard Reference Materials (SRM) (Soil: Item number 2709 and Rice Flour: Item number 1568A, from National Institute of Standards and Technology, USA) were analyzed in the same procedure at the start, during and at the end of the measurements to ensure continued accuracy. The certified arsenic concentration and our observed concentration (mg kg^{-1} dry weight) of the Rice Flour was 0.29±0.03 and 0.25±0.01 (n=3), respectively. The certified and the observed values were thus in good agreement.

Statistical Data Analysis: Pearson's correlation coefficient (r) was carried out to find out the correlation among arsenic concentrations in irrigation water, irrigated field soil and in different parts (root, straw, husk and grain) of the rice plant by SPSS software, version 14.0 for windows (SPSS Inc., Chicago, USA).

RESULTS AND DISCUSSION

Cultivation of rice is mostly depended on groundwater in West Bengal, particularly in the dry season, as the surface water sources (river, dam, pond, etc) become dry throughout the season in this region. Rice is widely cultivated in West Bengal to ensure food security and it requires huge volume of groundwater for its irrigation. But groundwater of West Bengal has been highly contaminated with arsenic. Thus there is a possibility of induction of arsenic in rice, cultivated with contaminated irrigation water and soil. In the present study, samples of irrigation water, agricultural field soil and rice plant were collected from the arsenic effected 17 villages of Chakdaha block and analyzed. The selected physicochemical properties of the soil and the concentrations of arsenic in irrigation water, soil and in the various parts (root, straw, husk and grain) of the rice plant cultivated in the Chakdaha block are presented in Table 1 and Table 2, respectively. The results show that the average arsenic concentration in the irrigation water lies between 0.11±0.012 and 0.76±0.014 mg l^{-1} and that in the agricultural field soil has been found to be between 1.38±0.108 and 12.27±0.094 mg kg^{-1} dry weight of arsenic. This clearly indicates that the agricultural soil of the study area has become highly contaminated with arsenic due to

Table 1: Soil characteristics of the arsenic contaminated rice fields of Chakdaha block

Soil parameters	Values
Clay (%)	70.6-81.1
Sand (%)	5.7-6.8
Silt (%)	12.1-24.3
Texture	Clay loam
pH	7.66±0.03
Organic carbon (%)	0.72±0.09

Table 2: Concentrations of arsenic in irrigation water, soil and in different parts of rice plant cultivated in the Chakdaha block

	Concentrations of arsenic (mean±SD)					
	(mg l^{-1})	(mg kg^{-1})				
Villages	Irrigation water	Soil	Rice root	Rice straw	Rice husk	Rice grain
Tatla-1 (n=3)	0.76±0.014	7.73±0.101	8.32±0.141	3.58±0.021	0.96±0.014	0.52±0.001
Tatla-2 (n=3)	0.69±0.008	8.61±0.114	10.48±0.098	1.90±0.019	0.94±0.009	0.59±0.004
Ghetugachi (n=4)	0.71±0.002	1.38±0.108	7.74±0.147	1.88±0.014	0.74±0.016	0.41±0.014
Dubra (n=3)	0.19±0.004	4.36±0.145	10.42±0.125	2.96±0.010	1.35±0.019	0.73±0.009
Dewli (n=4)	0.11±0.012	3.50±0.094	9.02±0.128	1.17±0.014	0.67±0.025	0.25±0.014
Hingnara (n=4)	0.64±0.007	10.87±0.123	8.93±0.239	3.05±0.026	0.56±0.018	0.47±0.004
Shilinda-I (n=4)	0.71±0.009	12.27±0.094	12.22±0.175	1.94±0.031	0.71±0.026	0.58±0.011
Shilinda-2 (n=3)	0.57±0.014	6.18±0.114	7.19±0.166	2.62±0.035	0.79±0.030	0.30±0.003
Chanduria-I (n=4)	0.40±0.005	7.10±0.175	10.09±0.194	1.47±0.009	0.89±0.014	0.72±0.011
Chanduria-2 (n=3)	0.53±0.001	5.52±0.099	18.63±0.155	1.41±0.024	1.34±0.017	0.60±0.003
Madanpur-I (n=3)	0.19±0.013	3.03±0.089	10.89±0.145	1.17±0.018	0.99±0.015	0.66±0.017
Madanpur-2 (n=3)	0.35±0.007	9.88±0.150	16.83±0.140	3.09±0.013	1.28±0.019	0.69±0.021
Sarati (n=4)	0.30±0.011	5.26±0.137	11.85±0.125	3.25±0.026	0.91±0.024	0.42±0.006
Saguna (n=4)	0.40±0.002	4.60±0.130	9.51±0.205	1.79±0.037	0.74±0.012	0.34±0.001
Simurali (n=3)	0.73±0.004	9.22±0.154	17.06±0.194	4.15±0.033	0.74±0.014	0.48±0.014
Routari (n=3)	0.16±0.014	4.94±0.141	14.09±0.056	1.17±0.022	1.05±0.036	0.54±0.002
Kanchrapara (n=3)	0.44±0.003	5.82±0.123	9.33±0.144	2.13±0.009	0.78±0.014	0.65±0.015

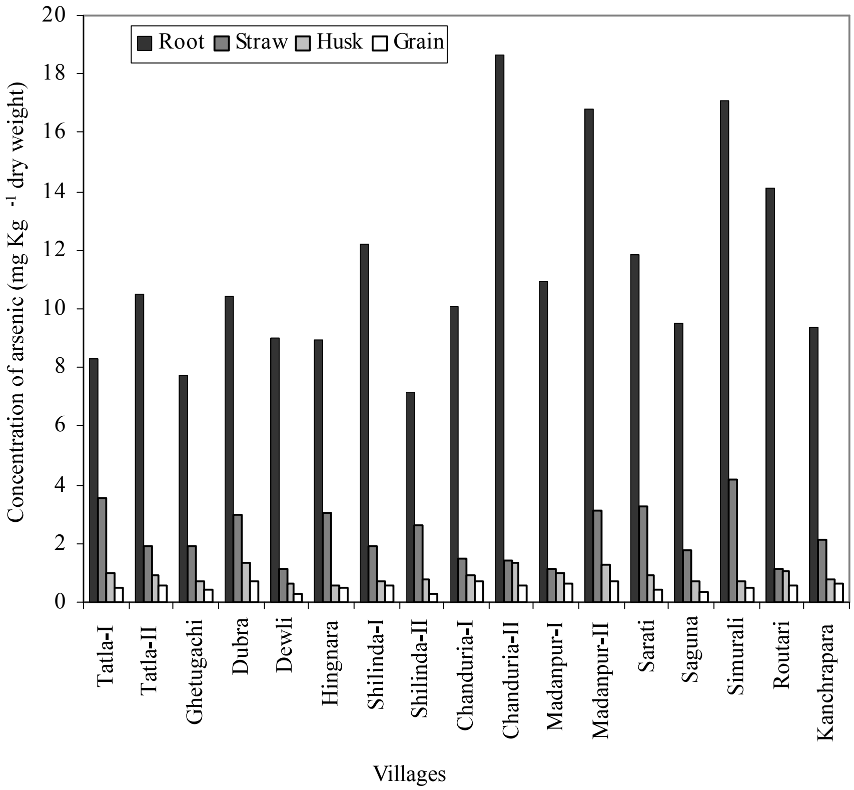

Fig. 1: Distribution of arsenic in different parts of the rice plant in the 17 villages of Chakdaha block

the excessive use of arsenic rich groundwater for irrigation. It can be seen that for all the studied samples the soil arsenic concentration was below the maximum acceptable limit for agricultural soil of 20.0 mg kg^{-1} as recommended by the European Community [7,19,20], but the arsenic concentration in groundwater was found to be much higher than the recommended World Health Organization permissible limit of 0.01 mgl^{-1} for drinking water [7,19,20] and Food and Agriculture Organization permissible limit for irrigation water (0.10 mg l^{-1}) [17].

Long term use of arsenic contaminated groundwater for irrigation may result in the further increase of arsenic concentration in the agricultural soil and eventually hyper-accumulation in crops including rice plants. The accumulation of arsenic (mg kg^{-1} dry weight) in the various parts of rice plant was found to have the following ranges: 7.19±0.166 to 18.63±0.155 in root, 1.17±0.014 to 4.15±0.033 in straw, 0.56±0.018 to 1.35±0.019 in husk and 0.25±0.014 to 0.73±0.009 in grain (Table 2). Graphical representation of the distribution of arsenic in different parts of rice plant cultivated in the Chakdaha block is shown in Figure 1. The results showed that the arsenic accumulated mostly in the root of rice plant (Table 2 and Fig. 1). Previously Rahman *et al.* [7] have reported that regardless rice varieties, accumulation of arsenic was 28 and 75 folds higher in root than that of shoot and raw rice grain, respectively. Although the actual mechanism of higher accumulation of arsenic in the rice root is still not well understood, Liu *et al.* [21] reported that iron oxides (iron plaques) formed around the rice root, bind the arsenic and reduce its translocation to the above ground tissues of the plant. The higher accumulation of arsenic in root of the rice plant is found to be followed by the accumulation (mg kg^{-1} dry weight) in straw (1.17±0.014 to 4.15±0.033), husk (0.56±0.018 to 1.35±0.019) and the lowest is observed in grain (0.25±0.014 to 0.73±0.009) of the rice plant. This is in good agreement with the previous published reports [4,7,9,10]. It can be seen that the rice plant has a relatively good accumulation of arsenic in its straw portion. Thus severe health hazard to the large cattle population of the study area consuming the straw part of the rice plant has become a concern. The Muslim populations of the area are used to eat beef as a readily available and cheap source of meat. The cow milk has served the protein requirement of people of all the religions. Thus there is a further risk of entering arsenic to human bodies [12].

Table 3: Correlation coefficients (r) among arsenic concentrations in irrigation water, soil and in different parts (root, straw, husk and grain) of the rice plant cultivated in the Chakdaha block

	Irrigation water	Soil	Root	Straw	Husk	Grain
Irrigation water	1.000					
Soil	0.514*	1.000				
Root	-0.059	0.285	1.000			
Straw	0.428*	0.451*	0.102	1.000		
Husk	-0.344	-0.142	0.538*	-0.021	1.000	
Grain	-0.091	0.239	0.363	-0.022	0.613**	1.000

*Correlation is significant at the 0.05 level (1-Tailed)

**Correlation is significant at the 0.01 level (1-Tailed)

The results reveal that arsenic supplied with irrigation water thankfully accumulates much less in rice grains (Table 2 and Fig. 1). The concentration of arsenic in the grain of all the studied rice samples was found to be between 0.25±0.014 to 0.73±0.009 mg kg^{-1} dry weight of arsenic, which did not exceed the permissible limit in rice (1.0 mg kg^{-1}) according to WHO recommendation [7,9, 22]. The remarkable shielding of rice grains from the contaminated irrigation water and from the soil arsenic is consistent with several previous studies [7,9,10]. The outer fraction of rice (husk) might act as a translocation barrier for which arsenic could not mobilize easily into the rice grains. The children spend a considerable amount of time playing in the fields surrounding their village. Wasserman *et al*. [23] have shown that the mental development of the children is impaired by the exposure to arsenic. Thus the increase exposure of arsenic to the children is of particular concern.

The correlation coefficients (r) among arsenic concentrations in irrigation water, soil and in different parts (root, straw, husk and grain) of the rice plant is shown in Table 3. It is interesting to note the fact that the arsenic content of the soil is significantly correlated with the arsenic content of irrigation water (r=0.514). This clearly indicates that the usage of arsenic rich irrigation water in the study area have contaminated the agricultural soil with arsenic. High significant correlation is obtained between arsenic concentrations in rice husk and grain portions (r=0.613). Significant correlation of the arsenic content in straw with the arsenic contents in irrigation water (r=0.428) and soil (r=0.514) and that between root and husk (r=0.538) can also be seen. Strong negative correlation among arsenic concentrations in irrigation water with rice root, husk and grain parts are observed. Similar trend of negative correlations of arsenic concentration can also be seen among straw and husk (-0.021) and straw and grain (-0.022) parts. Thus from the results it can be concluded that the arsenic contaminated irrigation water and the agricultural field soil are highly responsible for the transfer and uptake of arsenic in rice plant.

CONCLUSION

Our investigation show that the level of arsenic in irrigation water in the study area is very much above the WHO permissible limit of 0.01 mg l^{-1} for drinking water and FAO permissible limit of 0.10 mg l^{-1} for irrigation water. Soil gets contaminated with arsenic due to the irrigation with the arsenic contaminated water. Although it is got clear that the arsenic is transferred from irrigation water and paddy soil to different parts (root, straw, husk and grain) of the rice plant with different pattern of distribution, the results suggest that irrigation with groundwater enriched in arsenic affects the rice grain quite limitedly. In none of the studied samples the concentration of arsenic in rice grain exceeds the permissible limit of 1.0 mg kg^{-1} dry weight (WHO recommendation). Thus the exposure of the people living in the study area to arsenic contained in rice is less of an immediate concern than the continued use of highly arsenic contaminated groundwater for drinking and cooking purposes. But other indirect pathways of human exposure to arsenic can not be ruled out. Consumption of rice straw by cattle could potentially lead to increased arsenic levels in meat or milk by which there is a further risk of arsenic entrance to human bodies.

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