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Arsenic Fate in the Ground Water and its Effect on Soil-Crop Systems

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ABSTRACT

Environmental pollution is a major global concern. When sources of water pollution are enumerated, agriculture is, with increasing frequency, listed as a major contributor. One of the major factors determining uptake and toxicity to plants is the form of arsenic (As). The present study deals with the source of arsenic contamination in groundwater, accumulation of the toxin in soils and crops in the affected belt of West Bengal irrigated with contaminated groundwater and in animal tissues and products and demonstrates the pathways, other than drinking water, through which arsenic may have access to human, animal and crop systems. This article can be established well with the findings of a rice pot-culture experiment with different crop plants raised in a crop cafeteria experiment exhibited varying tendencies to accumulate arsenic in different plant parts in the following sequence: root>stem>leaf>economic produce.

Key words: Groundwater, contamination, arsenic and soil-crop system

INTRODUCTION

The presence of naturally elevated levels of As in groundwater was confirmed in seven Indian states, namely West Bengal, Bihar, Uttar Pradesh, Assam, Jharkland, Chattisgarh and Madhya Pradesh. Except for West Bengal, the extent of the problem is not fully known and the number of people at risk is impossible to estimate with any degree of confidence. In West Bengal, investigations suggest that eight districts show As content in well-water to be above 0.050 mg L⁻¹ with, according to United Nations Children's Fund (UNICEF), over 13.8 million people at risk (Nickson *et al.*, 1998; Chowdhury *et al.*, 2000).

Arsenic contamination of groundwater in the Gangetic alluvial zones of West Bengal has assumed the proportion of a drinking water-related disaster in recent years with reports of arsenic related health hazards for millions of people. As many as 75 blocks in 9 districts covering an area of 38865 km² are reported to be severely affected (Chowdhury *et al.*, 2000). Groundwater is also used extensively for crop irrigation in the arsenic belt of West Bengal and elevated arsenic levels in groundwater-irrigated soils are now well documented. Arsenic uptake by crop plants grown in soils contaminated with high concentrations of arsenic and irrigated with arsenic-contaminated groundwater has also been reported (ICAR., 2001; Abedin *et al.*, 2002). Soil acts as a major sink of

arsenic inflow to agro-ecosystems (Carey *et al.*, 1996; Ghosh *et al.*, 2002; Livesey and Huang, 1981; Majumdar and Sanyal, 2003; Mukhopadhyay, 2002), thereby reducing the availability of the toxicant to the cropped species.

The sedimentary rocks generally have higher As content than igneous and metamorphic rocks, while suspended and bottom sediments in most aquatic systems contain more arsenic than most natural water (Welch *et al.*, 1988). The capacity to retain arsenic is primarily governed by the sediment grain-size and the presence of surface coating composed of clays, clay-sized iron, manganese oxides and organic matter. Arsenic held by solid phases within the sediments, especially iron oxides, organic matter and sulphides may constitute the primary arsenic sources in groundwater under conditions conducive to arsenic release from these solid phases. These include abiotic reactions (oxidation/reduction, ion exchange, chemical transformations) and biotic reactions (microbial methylation) (Mok and Wai, 1994).

Arsenic in groundwater is generally present as dissolved, deprotonated/protonated oxyanions, namely arsenites ($\text{As}^{\text{III}}\text{O}_3^{3-}$; $\text{H}_n \text{As}^{\text{III}}\text{O}_3^{(3-n)-}$, with $n = 1, 2$) or arsenate ($\text{As}^{\text{V}}\text{O}_4^{3-}$, $\text{H}_n\text{As}^{\text{V}}\text{O}_4^{(3-n)-}$ with $n = 1, 2$), or both, besides the organic forms. The toxicity of As compounds in groundwater/soil environment depends largely on its oxidation state and hence on redox status and pH, as well as whether As is present in organic combinations. The toxicity follows the order: arsine (valence state of As: -3) > organo-arsine compounds > arsenites (+3) and oxides (+3) > arsenates (+5) > arsonium metals (+1) > native arsenic (0). The arsenites are much more soluble, mobile and toxic than arsenates in aquatic and soil environments. At pH 6-8, in most aquatic systems, both $\text{H}_2\text{As}^{\text{V}}\text{O}_4^-$ and $\text{HAs}^{\text{V}}\text{O}_4^{2-}$ ions occur in considerable proportions in an oxidized environment ($E_h = 0.2-0.5$ V), while $\text{H}_3\text{As}^{\text{III}}\text{O}_3$ is the predominant species under reduced conditions ($E_h = 0-0.1$ V) (Sadiq, 1997). Reduction of As (V) to As (III) would be accompanied by mobilization of As in aquatic system.

High arsenic concentration in groundwater is generally associated with the geothermal environments of volcanic deposits, geothermal systems and basin-fill deposits of alluvial lacustrine origin (Welch *et al.*, 1988). As regards the widespread As contamination in groundwater in parts of West Bengal, India and Bangladesh, confined within the delta bound by the rivers Bhagirathi and Ganga-Padma, two major hypotheses, both of geogenic origin, have been proposed. According to the earlier one (Mandal *et al.*, 1996), iron-containing minerals in the alluvial sediments, or formed *in situ*, combine with sulphur to form iron pyrites which have associated arsenic. The latter finds its way into groundwater through oxidation of arsenopyrite in aquifer sediments as atmospheric oxygen invades the aquifer in response to lowering of groundwater level by its large-scale abstraction for agricultural irrigation, especially for cultivation of summer (boro) paddy during the lean period of January to April when the groundwater recharge is at its minimum (Sanyal, 1999). This process would lead to the formation of iron sulphates and sulphuric acid.

This assumption or hypothesis is not consistent with the slightly alkaline status of groundwater in the affected delta, nor with its low (trace) concentration of sulphate, or high concentrations of bicarbonate, iron (II), arsenite, calcium and magnesium (Sanyal, 1999). Bhattacharaya *et al.* (1997) and Nickson *et al.* (1998) put forward the alternative hypothesis that the burial of the sediments, rich in organic matter, has led to strongly reducing conditions in groundwater aquifer, which is facilitated by high water table, fine-grained surface layers and widely practiced wetland paddy cultivation, as well as microbial oxidation of sedimentary organic matter, depleting thereby the dissolved oxygen in groundwater. Arsenic is released when arsenic-rich iron oxyhydroxides, which are efficient arsenic-scavengers, are reduced in anoxic groundwater. Such reduction is driven by

concentrations of sedimentary organic matter. Notwithstanding these hypotheses, the exact sequence of geochemical reactions, leading to As release in groundwater from the aquifer sediments, is still debated.

In particular, currently 75 blocks, located primarily in five districts on the Eastern bank of the river Bhagirathi, are affected with 5-6 million population reportedly drinking groundwater in West Bengal, laden to various degrees with As contamination. A large number of people have actually shown the symptoms of As poisoning, culminating into several deaths. Since, a greater part of the above stated delta has fallen in Bangladesh, the districts of Bangladesh, contiguous to the contaminated zones of West Bengal, are more widely affected by arsenic contamination of groundwater. The inclusion of pulses/other legumes/green manure crops in cropping sequences, coupled with organic manuring, was found to moderate arsenic build-up in soil and plant parts. Among the microorganisms, two genera of blue-green algae (*Anabaena* sp. and *Nostoc* sp.) and four different types of bacteria showed promise of As-decontamination ability. However, arsenic concentration bio-magnified as one passed from the groundwater to crop plants *via* soil.

The present study overviewed the complex problem of arsenic toxicity in agro-ecosystems receiving contaminated groundwater for irrigation purpose with emphasis on soil as an efficient sink.

MATERIALS AND METHODS

Sample collection and analysis: Water, soil and plant samples were collected from the two gram-panchayets (g.p), out of a total of 10, from Hariharpara block and from two gram-panchayets, out of a total of 9, from Raninagar-II block in the Murshidabad district where severely arsenic-affected locations for collection of samples where people were suffering with arsenical skin lesions. The present study was examined on selective plants/microbial species which are used for irrigation at the particular area (Chatterjee *et al.*, 1993; Roychowdhury *et al.*, 2005).

Water samples were collected from the shallow, large-diameter tube-wells, used for agricultural irrigation. The water samples were not filtered during collection or prior to analysis, stored in polyethylene bottles, which were pre-washed with concentrated nitric acid (1:1) and nitric acid (0.1% v/v) was added as preservative (Chatterjee *et al.*, 1993). The analyses of water samples represented the total load of arsenic (dissolved plus colloid-bound) in the water samples.

Agricultural land soils (20-25 g) were collected from the (a) Upper surface of the land (with a Teflon knife after scraping away the top 0.5 cm and up to a depth of maximum 5 cm from the upper surface), (b) Root after pick off the plants, cultivated on the agricultural lands and (c) Below ground level (up to a depth of 30 cm and at every 5 cm interval from the upper surface). The soil samples were mainly clay in nature.

Soil samples were placed in individual polyethylene bags and transported to our laboratory by air. The samples were dried in open air under diffused sunlight followed by drying in an oven at 50°C for 24 h, manually ground to a fine powder with a mortar and passed through a 30-mesh sieve (Chatterjee *et al.*, 1993; Roychowdhury *et al.*, 2005).

The plants (each sample consisted of three sub-samples having a distance of approximately 300 cm between each other) were approximately 30-35 days old. The plants from all the four agricultural lands were sampled by hands protected with vinyl gloves, carefully placed in individual polyethylene bags, stored in a cold box at 4°C and transported to our laboratory by air. All the plants were kept at 4°C until further treatment. After keeping at room temperature for several hours, the root, stem and leaf parts of each plant were separated with a quartz knife. At first, all

the parts (special attention was made on root to remove the soil) were washed manually by tap water and finally by deionized water (Queirolo *et al.*, 2000) in an ultrasonic cleaner (50/60 Hz, Model No. B-220; Branson, USA). All the parts were dried in open air under diffused sunlight for 24 h, followed by drying in an oven at 50°C for complete dryness, manually ground to a fine powder (homogenized) with a mortar and passed through a 30-mesh sieve (Chatterjee *et al.*, 1993; Roychowdhury *et al.*, 2005).

RESULTS AND DISCUSSIONS

Arsenic accruing plants/microbial species: The described hyper accumulation of arsenic from the contaminated soils by the brake-fern, *Pteris vittata* and its succeeding translocation into the above-ground biomass (Ma *et al.*, 2001) advises that the plant-accumulated arsenic was present almost entirely in the toxic inorganic forms, with the proportion of highly toxic As (III) being, in fact, much greater in the plant body than that of the less toxic As (V) form, as compared to the distribution of these two forms in the contaminated soil in which the fern grows (Ma *et al.*, 2001). Conversion of the plant-accumulated inorganic forms of the toxin to non (or less)-toxic organometallic forms by plant metabolism would certainly aid the detoxification process. Such detoxification within the plant body assumes importance, particularly in view of the report that arsenic in plant residues may be mobilized by a unicellular alga, namely *Polyphysa peniculus* (Cullen *et al.*, 1994), resident in normal (moist) agricultural soils.

The previous study and literature reveals a number of plant/microbial species, known for arsenic accumulation/bioindicator, which can effectively remove arsenic (and other heavy metals) from the aquatic system, for instance, to the tune of 170 and 340 $\mu\text{g As g}^{-1}$ dry weight of water hyacinth in its stem and leaves, respectively, when grown in a pond containing 10 mg As dm^{-3} (Chigbo *et al.*, 1982). However, such accumulated arsenic in water hyacinth (*Eichhornia crassipes*) is also liable to leaching out in the water body, particularly so on decomposition of such aquatic weed. Consequently, appropriate precaution has to be exercised while interpreting the arsenic status of aquatic environment by water hyacinth accumulation. Hydrilla (*Hydrilla verticillata* Casp.) is another such aquatic plant (Lee *et al.*, 1991). Pointed gourd, a vegetable creeper plant (*Trichosanthes dioica*), has also been found to accumulate arsenic when cultivated in the contaminated soils of West Bengal (Panda and Das, 2001a).

Some other crop plant species (rice, elephant-foot yam, green gram, etc.) are also reported to accumulate As in substantial quantities (ICAR., 2001). Abedin *et al.* (2002) have reported accumulation of As and transformation of As species in rice plant. However, information on the transformation of As species within plants is limited. The toxicity of As species in its plant body is reported to follow the order $\text{AsH}_3 > \text{As(III)} > \text{As(V)} > \text{MMA}$ (monomethylarsonic acid) $> \text{DMMA}$ (dimethylarsinic acid) (NRCC., 1978).

Arsenate tolerance by the grassy weeds, namely *Agrostis castellana* and *A. delicatula*, has been discussed in terms of the comparison of the corresponding reduction of Maximum Root Growth (MRG) with that in the sensitive populations upon exposure to arsenic (De Koe and Jaques, 1993). These findings tend to suggest that most, if not all, cropped plant species irrigated with As contaminated water, or such as those cultivated on soils, or in water bodies containing high levels of the toxic metalloid, tend to accumulate Arsenic.

A group of microbial species (e.g., the bacterial species, namely *Proteus* sp., *Escherichia coli*, *Flavobacterium* sp., *Corynebacterium* sp. and *Pseudomonas* sp.; the fungus, namely *Candida humicola*; the freshwater algae, namely *Chlorella ovalis*, *Phaeodactylum tricornutum* and *Oscillatoria rubescens*) have been reported to possess varying degrees of arsenic accumulating

abilities. However, the extent of transformation of species and detoxification of such accumulated arsenic is varied and also to an extent, uncertain like arsenic speciation, redox potential in soil, pH of the soil, organic matter in soil, soil texture, as bound to Fe-Mn oxides, etc. soil (McLaren *et al.*, 1998; Mukhopadhyay, 2002; Livesey and Huang, 1981; Carey *et al.*, 1996; Majumdar and Sanyal, 2003).

Accumulation of arsenic in soil-plant system: Soil contamination with arsenic may have toxic effect on vegetation and the animals feeding on the same. Arsenic may not be readily trans located from root to shoot and different crop plants exhibit different tendencies to accumulate and tolerate As (Table 1). Lowland paddy seems to be especially susceptible to As toxicity, due possibly to prevailing reducing conditions in flooded paddy soils.

Some important findings have emerged from the studies on impacts of As contamination in groundwater *vis-a-vis* management of agricultural systems. More important highlights of the study are given below.

The As concentration tends to build up from the contaminated groundwater, *via* the soil, to the crop, irrigated with such water. Among the plant parts, As concentration was higher in roots, followed by stem and leaves, in that order, while the economic or edible parts recorded the lowest concentration of the toxin (Adak and Mandal, 2000). However, potato tuber, despite being an underground part (a modified stem), contained relatively lower amount of As (Adak and Mandal, 1999).

A crop cafeteria experiment was conducted in the farmers' fields in the affected area in West Bengal, involving several farmer-attractive/remunerative cropping sequences, designed to take off partially the pressure on the contaminated groundwater resource for irrigation, especially during the lean period of groundwater recharge. A number of these sequences proved profitable and worth

Table 1: Relative sensitivity of various crop plants to arsenic

Tolerant	Moderately tolerant	Low tolerant
Fruit crops		
Apples	Cherries	Peaches
Grapes	Strawberries	Apricots
Raspberries		
Dewberries		
Vegetables and fruit crops		
Rye	Beets	Peas
Mint	Corn	Onion
Asparagus	Squash	Cucumber
Cabbage	Turnips	Snap beans
Carrots	Radish	Lima beans
Parsnips		Soybeans
Tomato		Rice
Potato		Spinach
Swiss chard		
Wheat		
Oats		
Cotton		
Peanuts		
Tobacco		
Forage crops		
Sudan grass	Crested wheat grass	Alfalfa
Bluegrass	Timothy	Bromegrass
Italian ryegrass		Clover
Kentucky bluegrass		Vetch
Meadow fescue		
Red top		

further examination. In particular, inclusion of pulses/other legumes/green manure crops in the cropping sequences, coupled with organic manure incorporation, was found helpful in moderating As build-up in soil and plant parts.

The field study was examined with lowland rice (boro paddy) and revealed that the extractable As in soil and As build-up in plants was drastically reduced by zinc application. The grain yield of rice in this experiment was also found not to vary significantly between the treatments of continuous ponding and judicious intermittent ponding (Garai *et al.*, 2000); however, the latter saved the irrigation water, thereby bringing less toxin to the soil/crop system.

Laboratory studies on coupled transport of aqueous arsenic across and down the plugs/columns of soils from the affected zone revealed that these soils did act as effective sink for As through its accumulation and relative hold-up from the contaminated irrigation water, in agreement with findings from the corresponding breakthrough curves (Mukhopadhyay *et al.*, 2002). These laboratory findings were supported by the field observations that the extractable As content of soils decreased sharply in subsurface and sub-soils of the profiles in the contaminated zone (Ghosh *et al.*, 2002) and that the leachate water, collected from auger holes in the As-affected command area of deep tubewell (discharging contaminated water), showed low As concentrations (Das and Panda, 2000).

Seasonal fluctuations in As content in groundwater and pond water in the affected zone was noted (Panda and Das, 2001b). Suspended solids in pond water were observed to contribute to the total As content of unfiltered pond water. These findings suggest that storing of groundwater in ponds would aid in reducing As content in water, while sedimentation of the suspended solids in ponds would further lower such concentration. The process of dearsenification of As-contaminated groundwater on storing in ponds would be an effective method involving peoples' participation at the rural level (Das and Panda, 2000; Panda and Das, 2001b).

The findings from an incubation study tended to demonstrate the dependence of As release in the soil solution of the As-contaminated soil samples (from the affected zone of West Bengal) on the applied phosphate and FYM with FYM being able to bind As in the soil matrix (Mukhopadhyay *et al.*, 2002). This is illustrated by the step-wise multiple regression equations given in Table 2 (Mukhopadhyay *et al.*, 2002). This also reflects itself in the findings obtained from a supporting pot-culture experiment with rice crop, where application of FYM helped to moderate the As accumulation in both the soil/plant as illustrated in Table 3-5 (Mukhopadhyay and Sanyal, 2000). The latter tended to derive support from much higher yield of the crop observed in the As-treated soils in presence of the said organic manure (Table 4). Organo-arsenic complexation with humic/fulvic colloids of the native soil and the incorporated organic manures, which would be expected to moderate hazards of As toxicity, was also demonstrated in the present study (Mukhopadhyay, 2002), thereby adding confidence to the trends of the findings as reported in Table 2-5.

Interestingly, the surface water bodies, located in the affected belt, have remained largely free of As. This tends to suggest that the soil, which receives As-contaminated water, acts as an effective sink to contain the toxin (as stated earlier), thereby preventing the surface run-off to carry it to the adjoining water systems (Sanyal, 1999).

The As-decontaminating ability of the microorganisms, prevalent in the contaminated soils, was monitored. This led to the identification of two genera of Blue-Green Algae (BGA) (namely, *Anabaena* sp. and *Nostoc* sp.) and four different types of bacteria showing promise of As-decontamination ability.

Table 2: Stepwise multiple regression equations showing effect of different treatments on NaHCO₃ (pH 8.5)-extractable arsenic content (mg kg⁻¹) in soils

Soil and regression equations	R ²
Site-1	0.618
Y = 0.7951+0.4326 X ₁	0.757
Y = 0.5665+0.4326 X ₁ +0.0069 X ₂	0.899
Y = 0.7713+0.4287 X ₁ +0.0070 X ₂ -0.4162 X ₃	0.943
Y = 0.6087+0.4287 X ₁ +0.0070 X ₂ -0.4199 X ₃ +0.0097 X ₄	
Site-2	
Y = 0.7723+0.3848 X ₁	0.638
Y = 0.965+0.3813 X ₁ -0.3828 X ₃	0.788
Y = 0.7717+0.3812 X ₁ +0.0059 X ₂ -0.3902 X ₃	0.931
Y = 0.7007+0.3812 X ₁ +0.0059 X ₂ -0.3918 X ₃ +0.0040 X ₄	0.940
Site-3	
Y = 0.9837+0.0467 X ₄	0.530
Y = 0.5476+0.4361 X ₁ +0.0467 X ₄	0.872
Y = 0.4195+0.4361 X ₁ +0.0038 X ₂ +0.0467 X ₄	0.895
Y = 0.4861+0.4348 X ₁ +0.0039 X ₂ -0.1387 X ₃ +0.0468 X ₄	0.901
Site-4	
Y = 0.5077+0.3041 X ₁	0.384
Y = 0.1738+0.3041 X ₁ +0.0196 X ₄	0.596
Y = 0.0541+0.3041 X ₁ +0.0068 X ₂ +0.0196 X ₄	0.779
Y = 0.0851+0.3014 X ₁ +0.0069 X ₂ -0.2894 X ₃ +0.1980 X ₄	0.866

Y: Extractable arsenic content in soil, X₁: Arsenic addition, X₂: Phosphorus addition, X₃: FYM incorporation, X₄: Incubation period

Table 3: Arsenic content (mg kg⁻¹) in straw and grain of rice at harvest under pot-culture study

Treatments	Straw			Grain		
	O ₀	O ₁	O ₂	O ₀	O ₁	O ₂
As ₀	5.11	1.91	1.47	0.07	0.09	0.05
As ₁	9.17	8.53	7.36	2.11	2.11	2.29
As ₂	19.3	11.2	11.1	3.24	2.47	2.45
C.D. (p = 0.05)	As = 0.87 O = 0.87 As×O = 1.52			As = 0.39 O = 0.39 As×O = NS		

O₀: No FYM incorporation, O₁ and O₂: FYM incorporation at 0.5 and 1%, respectively, by weight of soil, As₀: No arsenic addition, As₁ and As₂: Arsenic addition at 50 mg kg⁻¹ and 100 mg kg⁻¹ soil, respectively, Weight of soil in each pot = 5 kg

Table 4: Effect of FYM and arsenic on yield attributes and yield (g/pot) of rice (IET-4786) at harvest in pot experiment

Treatments	Panicle number/plant	Grain/panicle	Filled grain (%)	1000 grain weight g ⁻¹	Yield (g pot ⁻¹)
As ₀ O ₀	14	76	75	23	18.40
As ₀ O ₁	16	81	77	26	25.90
As ₀ O ₂	19	86	82	28	37.50
As ₁ O ₀	11	69	66	19	9.52
As ₁ O ₁	14	73	72	21	15.50
As ₁ O ₂	16	83	76	23	23.20
As ₂ O ₀	10	62	66	19	7.77
As ₂ O ₁	15	73	71	21	16.30
As ₂ O ₂	15	73	75	24	19.70
C.D. (p = 0.05)	As = 1.19 O = 1.19 As×O = 2.06	As = 1.38 O = 1.38 As×O = 2.38	As = 1.63 O = 1.63 As×O = 2.82	As = 1.71 O = 1.71 As×O = 2.97	As = 3.68 O = 3.68 As×O = 6.36

Table 5: Residual arsenic (mg kg⁻¹) in soil at harvest of rice crop under pot-culture study

Treatments	O ₀	O ₁	O ₂
As ₀	0.39	0.20	0.21
As ₁	8.88	7.54	5.17
As ₂	24.60	15.10	12.10
C.D. (p = 0.05)		As = 0.66 O = 0.66 As×O = 1.14	

CONCLUSION

The present study has established that, equal if not greater, attention is necessary for understanding the complex aspects of accumulation of arsenic in the food web *vis-a-vis* in drinking

water alone and its ultimate passage to the human populations. This study also highlights the distinct possibility of exposure to toxicity of human and livestock populations consuming arsenic laden agricultural produce at locations away from arsenic affected areas of West Bengal and elsewhere. Further studies are necessary to confirm and extend such initial findings and document the mobilization and transformation of species of As in food web.

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