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Impact of irrigating rice paddies with groundwater containing arsenic in Bangladesh

A. van Geen [a,*], Y. Zheng [a,b], Z. Cheng [a], Y. He [b,1], R.K. Dhar [b], J.M. Garnier [c], J. Rose [c], A. Seddique [d], M.A. Hoque [d], K.M. Ahmed [d]

[a] *Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY 10964, USA*
[b] *Queens College, City University of New York, Flushing, New York 11367, USA*
[c] *Centre Européen de Recherche et d'Enseignement de Géosciences de l'Environnement UMR 6635 CNRS, University Paul Cézanne, IFR PSME 112, Europôle Méditerranéen de l'Arbois, 13545 Aix-en-Provence, France*
[d] *Deparment of Geology, University of Dhaka, Bangladesh*

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Abstract

Soil and soil–water As profiles were obtained from 4 rice paddies in Bangladesh during the wet growing season (May–November), when surface water with little arsenic is used for irrigation, or during the dry season (January–May), when groundwater elevated in arsenic is used instead. In the upper 5 cm of paddy soil, accumulation of 13±12 mg/kg acid-leachable As (n=11) was observed in soil from 3 sites irrigated with groundwater containing 80–180 μg/L As, whereas only 3±2 mg/kg acid-leachable As (n=8) was measured at a control site. Dissolved As concentrations averaged 370±340 μg/L (n=7) in the upper 5 cm of the soil at the 3 sites irrigated with groundwater containing 80–180 μg/L As, contrasting with soil water As concentrations of only 18±7 μg/L (n=4) over the same depth interval at the control site. Despite the accumulation of As in soil and in soil water attributable to irrigation with groundwater containing elevated As levels, there is no evidence of a proportional transfer to rice grains collected from the same sites. Digestion and analysis of individual grains of boro winter rice from the 2 sites irrigated with groundwater containing 150 and 180 μg/L As yielded concentrations of 0.28±0.13 mg/kg (n=12) and 0.44±0.25 mg/kg (n=12), respectively. The As content of winter rice from the control site was not significantly different though less variable (0.30±0.07; n=12). The observations suggest that exposure of the Bangladesh population to As contained in rice is less of an immediate concern than the continued use of groundwater containing elevated As levels for drinking or cooking, or other potential consequences of As accumulation in soil and soil–water.

Keywords: Arsenic; Bangladesh; Rice; Irrigation; Paddy soil

1. Introduction

A pioneering study of five households over a period of 2 years in West Bengal, India, demonstrated that the substitution of well water used for drinking and cooking containing 100–230 μg/L As with water containing 3 μg/L As led to a several-fold reduction in the As

* Corresponding author.
E-mail address: avangeen@ldeo.columbia.edu (A. van Geen).
[1] Now at: John Jay College, City University of New York, New York, NY 10019, USA.

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content of urine, nails, and hair (Mandal et al., 1998). This was a clear demonstration that the As content of well water is a major factor controlling As exposure and therefore the risk of contracting various As-induced diseases in the many areas of South Asia where the rural population relies heavily on groundwater (Dhar et al., 1997; BGS and DPHE, 2001; Berg et al., 2001). Significantly, the West Bengal study also documented large temporal fluctuations in urinary As and a failure to reach the background level indicated by urinary As concentrations for an unexposed control population. These observations were tentatively attributed by Mandal et al. (1998) to the intake of As through food, possibly herbs grown on groundwater elevated in As, rice cooked in groundwater, and occasional consumption of well water containing elevated levels of As. There have since then been a number of conflicting reports regarding the possibility of significant exposure to As through food, particularly the large quantities of rice grown throughout South Asia on paddies irrigated with groundwater.

The As content of uncooked rice reported for the affected study area of Mandal et al. (1998) in West Bengal averaged 0.07 ± 0.01 mg/kg ($n=5$). A more recent set of analyses for rice from West Bengal reported by the same group (Chakraborti et al., 2004) yielded an average of 0.32 mg/kg ($n=21$). Arsenic concentrations generally within this range have been reported for samples of raw and cooked rice grains from different parts of West Bengal and Bangladesh, with concentrations only occasionally exceeding 1 mg/kg (Roychowdhury et al., 2002; Meharg and Rahman, 2003; Das et al., 2004). In what may be the most comprehensive study to date, the As content of 150 rice samples from various locations in Bangladesh averaged 0.18 and 0.12 mg/kg for boro (winter) and amon (summer) rice, respectively (Duxbury et al., 2003). A simple calculation based on these observations indicates that significant As exposure from eating rice cannot be dismissed out of hand. A daily dose of 0.1 mg As could equally plausibly be attained by eating a daily diet of 500 g of rice containing 0.2 mg/kg As or the consumption of 2 L of groundwater containing 50 μg/L As, the Bangladesh standard for As in drinking water. Such a comparison fails to take into account the speciation of As in water and food which could significantly affect its toxicity, however (Schoof et al., 1999; Chakraborti et al., 2004).

This study does not challenge the indisputable fact that consumption of rice results in some exposure to As, and not only in Bangladesh (Williams et al., 2005). The question addressed here, instead, is whether the As content of rice grains grown in Bangladesh is very sensitive to the As content of groundwater used for irrigation. Two recent studies that included a set of control samples suggest that this might not be the case. The first reported 2–3 times higher As concentrations in rice grains produced in a region of Bangladesh known to be irrigated with groundwater elevated in As compared to a region that is not, while also pointing out that the average and range of As concentrations in the combined Bangladesh sample of rice grains ($n=62$) was not significantly different from the outcome of analyses of rice grown in Japan ($n=30$) on water that was presumably not elevated in As (Hironaka and Ahmad, 2003). The second study reported that although the As content of paddy soil and various parts of the rice plant other than rice grains was significantly higher in two areas of Bangladesh impacted by As compared to a control area, the average As content of rice grains from the same three areas (~0.5 mg/kg) was comparable (Ali et al., 2003). More recently, no difference in As content was observed for rice grains grown in fields irrigated with high and low-As irrigation water in West Bengal, India (Norra et al., 2005). Conflicting results were reported by the same team of researchers that conducted two greenhouse studies using irrigation water with extremely high As concentrations (up to 8 mg/L). In one study, the As content of amon rice grains was observed to be much less sensitive to the As content of irrigation water compared to other parts of the rice plant (Abedin et al., 2002a). In the other, seemingly very similar study, the As content of rice grains instead showed a modest response, an increase from 0.3 to 0.5 mg/kg As, to irrigation water containing 1 mg/L As (Abedin et al., 2002b).

Less controversy surrounds the question of As accumulation in paddy soil irrigated with groundwater. Meharg and Rahman (2003) addressed the issue at the regional scale by pointing out that concentrations of As in soils could reach ~30 mg/kg (compared to a crustal content of ~2 mg/kg) in areas of Bangladesh where groundwater is generally elevated in As and has been used for irrigation for over a decade. In two subsequent studies where samples of soil and irrigation water were carefully matched, significant enrichments in soil As extending to 10–20 cm depth were attributed to the As content of irrigation water (Ali et al., 2003; Huq et al., 2003). The latter studies also documented considerable variability in the level of soil As enrichments from one season to the other and for different paddies irrigated with the same groundwater.

This study seeks to contribute to the debate surrounding the potential impact of irrigation with groundwater in Bangladesh by comparing the As content of soil and rice grains measured for a suite of paddies in an area that is limited in extent but where the As content of irrigation water spans nearly two orders of magnitude. We report, to our knowledge for the first time, the concentration of As in soil water extracted from paddy soil in Bangladesh. The motivation for this particular measurement was that As contained in the dissolved phase could conceivably be more directly linked to the uptake of As by rice plants than the As content of the solid phase. The investigation was conducted within a well-studied 25 km^2 area where a long-term program conducted with partner institutions in Bangladesh by Columbia University has examined the health effects and geochemistry of As, while also evaluating the effectiveness of various mitigation options (van Geen et al., 2003a; Wasserman et al., 2004).

2. Methods

2.1. Study sites

The main area selected for this study encompasses ~4 km^2 of rice paddies and 29 motorized irrigation wells pumping groundwater from 6 to 20 m depth. Groundwater sampled from these irrigation wells contains 50–200 μg/L As and is used only during the dry season, from January/February to April/May, to grow boro rice (Bangladesh Rice Research Institute). The control site is a separate rice paddy located 3 km to the northwest that is irrigated with groundwater from a deeper aquifer containing only 3 μg/L As (Fig. 1). During the wet season, the same paddies are either flooded or irrigated with surface water low in As from the surrounding streams and ponds to grow amon rice (Agrahani) from May to November. Each season, the fields are plowed and irrigated before young rice plants are transplanted by hand.

2.2. Sample collection and processing

In October 2003 when the paddies in the area were flooded with surface water, two 5-cm sections of PVC pipe (~4-cm ID) held together with electrical tape

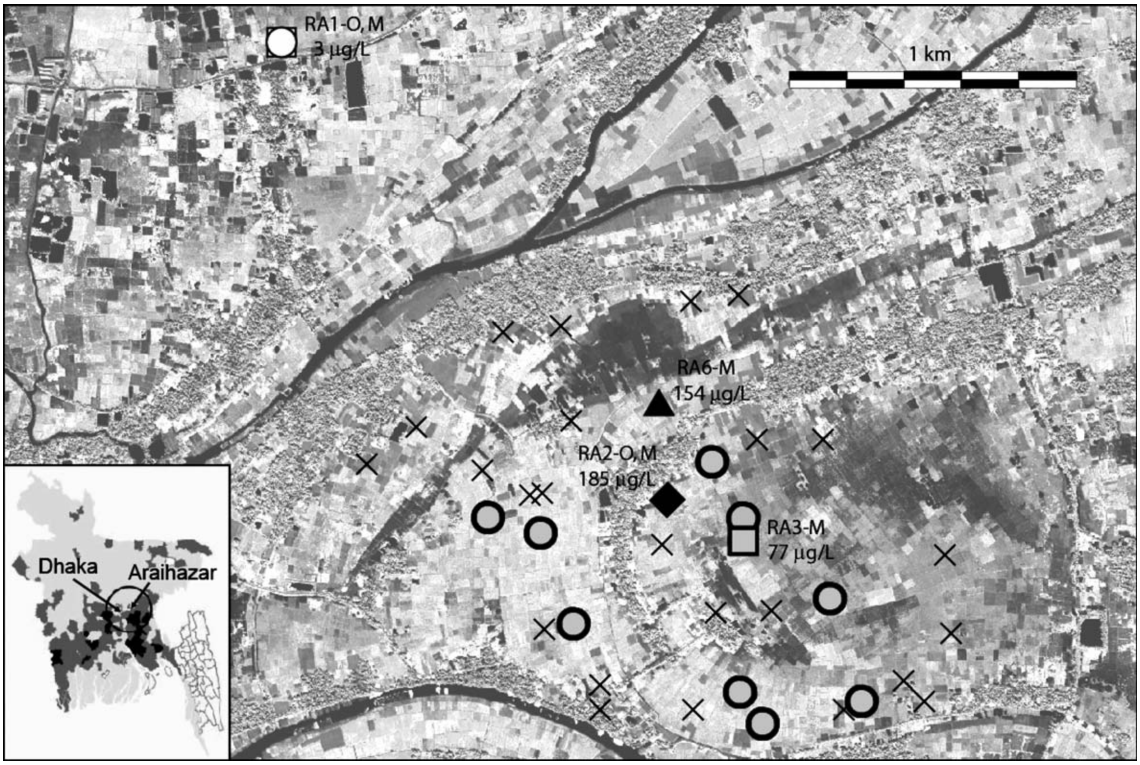

Fig. 1. Map of Araihazar study area showing the location of irrigation wells and sites where soil, soil water, and rice grain samples were collected. Inset shows the location of the study area relative to Dhaka, as well as the proportion of unsafe wells in various parts of the country determined by DPHE/UNICEF workers with a field kit (van Geen et al., 2003b). Shading corresponds to <20% unsafe wells (light grey), 20–80% (dark grey), and >80% (black). The four sites where detailed soil profiles were obtained in October 2003 ("O") and/or March 2004 ("M") are labeled; the As content of the irrigation water at these sites is also indicated. Grey circles indicate the locations of 9 additional sites where samples of soil and soil water integrating the 0 to 5 cm depth range were collected in March 2004. Crosses indicate the location of additional irrigation wells from which groundwater could not be sampled in March 2004 because they were not pumping during the day. Data summary provided in Table 1; all data included as Supplementary Material.

were used to collect two soil and soil–water samples between 0–5 and 5–10 cm depth, respectively. The stacked sections of PVC pipe were gently pushed between rows of rice plants to just below the surface of the soil at the control site (RA1-O) and at a site that is irrigated during the dry season with groundwater containing 185 μg/L As (RA2-O). A single 5-cm section of PVC pipe was used to collect soil and soil water from 0 to 5 cm depth at an additional 9 sites during the dry season in March 2004 (Fig. 1). At this time, soil profiles were also obtained at higher resolution with 2-cm wide (~12-cm ID) stainless steel rings stacked together with electrical tape at an additional 2 sites (RA3-M, RA6-M), as well as the 2 previously sampled sites (RA1-M, RA2-M). Conditions were not representative of the growing season at RA2-M, however, because the irrigation pump had stopped functioning for several days and surface soil was nearly dry. In all cases, cores of soil and soil water were obtained within a radius of ~10 m of the irrigation pump and the inlet of the irrigation channel to the field.

The stacks of PVC pipe and stainless steel rings were tightly capped and taped at the time of collection and processed within a few hours, without taking additional precautions to limit contact with atmospheric oxygen. Between 2 and 10 mL of soil water was extracted from each slice of paddy soil with a stainless-steel sediment squeezer placed in a large C-clamp (Manheim, 1966). After discarding the first few drops and filtering the extracted soil water through a 0.4 μm syringe filter into acid-leached scintillation vials with a Poly-Seal cap, the samples were acidified to 1% high purity HCl (Optima). Approximately 1 g of each soil sample was also placed in separate scintillation vials and covered with 10 mL of 10% reagent grade HCl. The soil slurries were placed in a hot bath (~80 °C) for 30 min to leach the As and Fe content of the soil as a measure of the fraction that is relatively easy to mobilize (Horneman et al., 2004).

In March 2004, samples of irrigation water were collected from the subset of irrigation wells that were operating at the time. These samples were collected with an all polypropylene–polyethylene syringe, transferred through a 0.4 μm syringe filter to acid-leached scintillation vials, and acidified to 1% high purity HCl (Optima).

Grains of amon and boro rice grown during 2002–2003 were obtained directly from the owners of the control site (RA1). Boro rice grown in 2003 and 2004 was also collected from the owners of two paddies irrigated with groundwater elevated in As (RA2 and RA6). Individual grains were randomly selected for analysis from the material provided to us in a container by the owners. We were assured of the provenance of these rice grain samples from a particular field, but could not determine from where within the field because the product of the harvest had been mixed.

2.3. Analyses

In March 2004, a subset of soil-water samples was analyzed on the day of collection for dissolved As(III) by differential pulse cathodic stripping voltammetry (He et al., 2004). Profiles of the redox state of the rice-paddy soil were also determined on the day of collection by measuring with ferrozine the proportion of Fe(II) in the total Fe fraction that can be leached from the soil in 10% hot HCl (Horneman et al., 2004).

Soil–water and irrigation-water samples were analyzed in the laboratory by high resolution inductively coupled plasma mass spectrometry (HR ICP-MS) for dissolved As and Fe following 1 : 5 dilution in a 2% HNO_3 solution (Cheng et al., 2004). The detection limit of the method for dissolved As is ~0.1 μg/L and the precision on the order of 2% for both As and Fe. The soil leachates and rice digests were also analyzed for As by HR ICP-MS following 1 : 50 dilution in 2% HNO_3. Procedural blanks for As in soil leachates were negligible.

Single whole rice grains without their husk were dried overnight in an oven at ~50 °C in open 1.5 or 0.5 mL centrifuge tubes (Eppendorf). After the weight of individual grains was recorded (15–20 mg), 100 μL of concentrated Optima HNO_3 was added to each tube as well as a set of blank tubes. The rice grains were digested by placing the open centrifuge tubes in a hot bath for 10–20 min. High-purity water was then added to the tubes (1.4 or 0.4 mL depending on the tube size), the tubes were capped and shaken vigorously, and a minor amount of translucent matter that remained after the digestion was separated by centrifugation. Procedural blanks averaged 0.7 ± 1 ng As for analyses distributed over the 3 batches of rice grains that were analyzed, which is equivalent to 0.04 ± 0.07 mg/kg As for a typical 18 mg rice grain. The same procedure was followed to measure the As content of Standard Reference Material 1568a from the National Institute of Standards and Technology (NIST), which is rice flour made from 100% Arkansas long-grain. The As concentrations measured by analyzing 15–20 mg batches of SRM 1568a using the above procedure (0.31 ± 0.03 mg/kg; $n = 6$) were comparable to the certified value of 0.29 ± 0.03 mg/kg.

3. Results

Concentrations of As in soil water (30–1500 μg/L) spanned almost two orders of magnitude in the 0–5 cm depth interval of the 13 paddies that were sampled, and this for samples that always contained >1 mg/L dissolved Fe and were therefore unmistakably anoxic (Fig. 2a). Anoxia within 1 cm of the rice paddy surface was confirmed by oxygen microelectrode profiles obtained in March 2004 (Y. Z., unpublished data). Voltammetric concentration and speciation measurements conducted in Bangladesh indicate that As in the soil water was essentially all in the form of As(III) (see Supplementary Material). The lowest soil–water As concentrations were measured on both occasions at the control site: 28 μg/L (n=1) and 15±2 μg/L (n=3) at RA1-O and RA1-M, respectively (Table 1). Soil-water measurements from all sites irrigated with high-As groundwater (with the exception of RA2-M, which is discussed separately) indicate significantly higher As levels, although there is no simple relationship with the concentration of As in irrigation water (Fig. 2a). Significantly, soil–water As concentrations during the wet season, when groundwater is not used for irrigation, were also high at a site where irrigation water during the dry season is high in As (RA2-O). This suggests that the impact of irrigation with groundwater elevated in As on rice paddies may not be limited to the dry season.

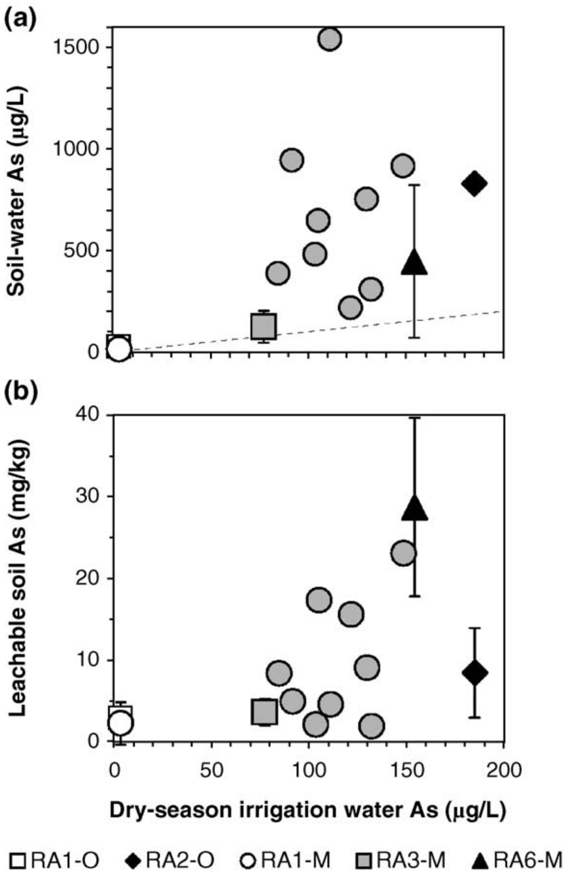

Fig. 2. Comparison of the As content of dry-season irrigation water with that of soil water (a) and soil (b). The symbols are the same as in Fig. 1. Dashed line in (a) corresponds to equal As concentrations in irrigation water and soil water. The standard deviation of soil and soil–water As concentrations is indicated for those locations where multiple samples were collected within the 0–5 cm interval (see Table 1 and Supplementary Material).

Accumulation of As in paddy soil offers a plausible explanation for transferring the impact of irrigation across the seasons because an As enrichment in soil water of 1000 μg/L requires the transfer of only 0.5 mg/kg from the solid phase, assuming a water content of 20% and a dry solid density of 2.5 g/cm^3. In our study area, the As content of rice paddy in the 0–5 cm depth that is leached in hot HCl ranged from 1 to 41 mg/kg. Despite considerable variability, the range of acid-leachable As concentrations in surficial soils generally appears to increase with the As content of irrigation water (Fig. 2b). For 4 out of 13 sites that were sampled, the leachable As concentration in surface soil exceeds 10 mg/kg, the regulatory limit established in the U.K. for domestic gardens (Huq et al., 2003).

Soil–water As concentrations at the site irrigated with groundwater containing 77 μg/L As (RA3-M) are intermediate between soil–water concentrations at the control site and sites irrigated with groundwater containing 185 and 154 μg/L As (RA2-O and RA6-M, Table 1). Detailed profiles obtained at RA3-M and RA6-M indicate higher soil–water As concentrations in the upper 5 cm compared to deeper intervals, whereas site RA2-O indicates elevated As concentrations extending beyond a depth of 5 cm (RA2-O, Fig. 3a). The profiles show no systematic link between the level of As enrichment in corresponding soil and soil–water horizons, although the number of data points is limited (Fig. 3a, b). Leachable As concentrations in the soil are relatively constant to a depth of 5 cm for 3 out of 5 cores (RA1-O, RA3-M, RA6-M); the leachable As content decreases and increases with depth at RA1-M and RA2-O, respectively.

The general association of elevated As and Fe concentrations in soil water, for sites other than the control, suggests that the mobilization of As is related to redox processes that affect the transfer of Fe in rice paddies between the solid and the dissolved phase. This is supported by the unusual conditions encountered at the high-As site resampled in March 2004 (RA2-M),

Table 1
Arsenic content of irrigation water, surface soil, and soil water in 0–5 cm depth interval at 4 sites

Field ID	Dry-season irrigation water As (μg/L)	Soil collection date	Leachable soil As (mg/kg)	Pore water	
				As (μg/L)	Fe (mg/L)
RA1-O	3	Oct ’03	2.9±0.3 (n=5)	28 (n=1)	18 (n=1)
RA2-O	185	Oct ’03	8.4±5.5 (n=5)	830 (n=1)	26 (n=1)
RA1-M	3	Mar ’04	2.2±2.6 (n=3)	15±2 (n=3)	4.7±4.6 (n=3)
RA2-M	–	Mar ’04	2.6±1.1 (n=3)	9±4 (n=3)	0.2±0.1(n=3)
RA3-M	77	Mar ’04	3.6±0.9 (n=3)	150±80 (n=3)	3.5±2.6 (n=3)
RA6-M	154	Mar ’04	29±11 (n=3)	450±370 (n=3)	2.7±2.2 (n=3)

when soil–water As and Fe concentrations were both very low (Fig. 3a, c). We tentatively attribute the low soil–water concentrations at RA2-M to Fe oxidation and co-precipitation of As to oxygen diffusion into the soil in the absence of standing water. A potential relation between soil–water As or Fe concentrations and the redox state of the sediment is more difficult to evaluate because leachable Fe(II)/Fe measurements did not reach deep enough at some of the sites. It is worth noting that the leachable Fe(II)/Fe ratio of ~0.5 was somewhat higher in the upper 5 cm than at depth (~0.3) at the 2 sites where extended profiles were obtained (RA3-M, RA6-M; Fig. 3d). At site RA2-M, where the irrigation pump was not functioning, leachable Fe(II)/Fe ratios evidently did not respond as markedly as the As and Fe content of soil–water to the presumed diffusion of atmospheric oxygen.

In contrast to the composition of soil and soil water, there appears to be no strong impact of the As content of irrigation water on the As content of rice grains at RA2 and RA6 (Table 2). The average As content of a dozen grains of winter boro rice from the site with the highest soil enrichment (29 mg/kg at RA6) is essentially identical to that of boro rice obtained from the control site. The As content of grains of boro rice from the site irrigated with groundwater with the highest level of As (185 μg/L at RA2) is slightly higher and more variable (0.44±0.25 mg/kg). The As content of summer amon rice grown at the control site (0.18±0.05 mg/kg) was about a factor of two lower than for boro rice from all sites.

4. Discussion

4.1. Impact of irrigation on soil and soil–water As

Soil–water As concentrations measured at the control site were lower than at the high-As sites sampled under

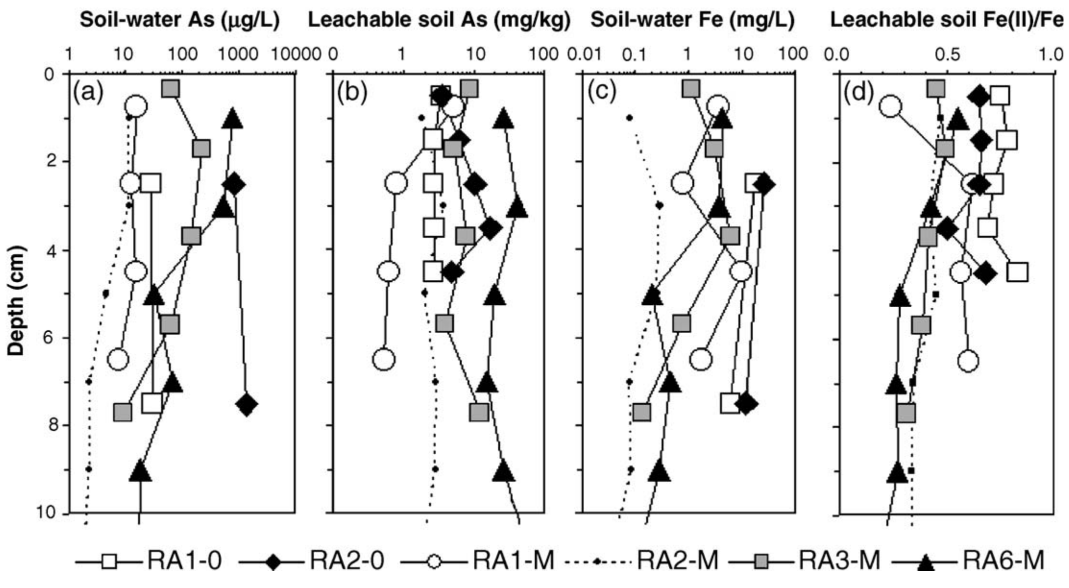

Fig. 3. Profiles of the dissolved As and Fe content of soil water measured at four sites in October 2003 and/or March 2004. Also shown are acid-leachable As concentrations and Fe(II)/Fe ratios in the soils. Symbols are the same as in Figs. 1 and 2. Dashed lines show profiles at RA-2 in March 2004, when the paddy field went dry because the irrigation pump had stopped functioning.

Table 2
Arsenic content of irrigation water and rice grains at 3 sites

Field ID	Dry-season irrigation water As (μg/L)	Winter boro rice As (mg/kg)	Summer amon rice As (mg/kg)
RA1	3	0.30±0.07 (n=12)	0.18±0.05 mg/kg (n=12)
RA2	185	0.44±0.25 (n=12)	
RA6	154	0.28±0.13 (n=12)	

flooded conditions (Fig. 2a), but still a factor of 10 higher than the highest soil–water concentration measured at an uncontaminated experimental rice paddy in Japan that was carefully monitored over several years (Takahashi et al., 2004). The distributions of As and Fe in soil water observed in Araihazar suggest that the connection between reducing conditions and the mobilization of As documented in the Japanese study (albeit at lower As levels) also applies to Bangladesh. The drastic effect of the break-down of the irrigation pump at RA2-M on soil water As and Fe concentrations indicates that at least some of the As supplied by irrigation water is temporarily trapped in the soil as relatively oxic conditions return to rice paddies between growing seasons.

In contrast to soil-water As concentrations, the leachable As content in soil at some of the sites irrigated with groundwater elevated in As was comparable to that of the control site (Fig. 2b). Ali et al. (2003) recently reported a similarly wide range of As concentrations in paddy soil for two regions of Bangladesh where As concentrations in irrigation water are elevated, as well as soil As levels that were systematically at the low end of the range in a control area. A simple mass balance calculation confirms that As supplied by irrigation could lead to substantial enrichments in soils. Rice cultivation requires on the order of 1 m of water over the cultivated area per growth season (Meharg and Rahman, 2003; Huq et al., 2003). To produce this amount of water, 29 irrigation wells distributed over the ~4 km^2 main study area (Fig. 1), pumping at an average rate of 30 L/s (unpublished flow-rate data, A. Horneman), would have to operate 50% of the time over the 4 month growing season. This is consistent with our own anecdotal observations. Irrigation with groundwater containing 100 μg/L As will therefore annually supply on the order of 100 mg of As/m^2 of paddy. The corresponding As enrichment in paddy soil would be ~0.5 mg/kg per year, assuming that the flux is retained within the upper 10 cm of soil, a 20% water content, and a soil density of 2.5 g/cm^3. If there is no major remobilization of As from rice paddies during the wet season, a decade or two of irrigation could therefore lead to significant soil enrichments, as observed in the Araihazar study area and elsewhere in Bangladesh (Meharg and Rahman, 2003; Ali et al., 2003; Huq et al., 2003).

The wide ranges of soil and soil–water As levels observed in Araihazar for a given As concentration in irrigation water suggest several confounding factors that were not controlled for in this preliminary study. These include spatial variability in the supply of As due to co-precipitation of As with Fe oxyhydroxides in irrigation canals and close to the irrigation well (Ali et al., 2003). Some of the variability could also reflect partial remobilization during the wet season of As accumulated in the soil during the dry season (Ali et al., 2003). Such remobilization is suggested by the low leachable As concentration in soils measured at RA2-M compared to RA2-O (Table 1), although binding of As to an oxidized Fe phase that was less sensitive to the acid leach cannot be excluded. As an alternative explanation for the high degree of spatial variability of As enrichments in soils, it is worth pointing out that the paddy with the highest As concentration of 29 mg/kg in our study area happens to be the only site where in March 2004 the irrigation pump was operating essentially around the clock, as opposed to several hours per day at the other sites. Auger cores combined with a geophysical survey of the area indicate that the same paddy field is underlain by coarser sand deposits than surrounding fields (Beth Weiman, Steve Goodbred, and Zahid Aziz, unpublished data). The owner confirmed that his paddy required more irrigation than nearby fields because of water loss, attributed to percolation through local sands. While a greater requirement of irrigation water for certain fields could lead to more build-up of As in the soil, such flushing might also limit soil–water As concentrations. We speculate that this may be why soil–water As concentrations at RA6-M were not particularly high, even if As levels in the soil at the same site clearly were (Fig. 2a, b).

4.2. Limited impact of irrigation on As content of rice

Comparison of the As content of individual grains of boro rice obtained from two fields with documented enrichments of As in soil and soil–water with boro rice from the control site suggest a modest if any impact of the As content of irrigation water (Table 1). Can this observation, from admittedly a limited number of sites, be generalized? Given the importance of the issue for the health and livelihood of millions of people, it would be unwise to do so outside the context of other studies.

In a review of the literature, Meharg and Rahman (2003) lists a range of 0.1–0.5 mg/kg for the As content of rice grains grown in North America and Taiwan on soils that are not contaminated with As. The average As content of all rice grains collected in Araihazar, the control site as well as the high-As sites, fall squarely within that range (Table 2). So does the average As content of rice grains from Bangladesh and West Bengal reported by Duxbury et al. (2003) and Chakraborti et al. (2004), respectively, which presumably include a significant fraction of samples from paddies that are irrigated with groundwater containing elevated As concentrations. The available data therefore suggest that if irrigation with groundwater enriched in As affects the composition of rice grains, the effect is quite limited. The rare reports of rice-grain samples containing >1 mg/kg As (e.g. Meharg and Rahman, 2003) should certainly be investigated further. Perhaps certain geological settings or varieties of rice indeed do not shield the grain as effectively from As contained in soil and soil water as others. Overall, however, alarmist views that rice paddies should not be irrigated with groundwater containing As in Bangladesh do not appear to be justified. If anything, such a recommendation could lead to the exploitation of deeper aquifers presently low in As for irrigation. This would be a serious mistake because withdrawals for irrigation that are very large compared to the small volumes pumped with hand-tube wells could potentially entrain shallower groundwater enriched in As into deeper aquifers, possibly leading to their contamination (Zheng et al., 2005).

4.3. Other potential consequences of irrigation

If rice grains do not appear to be strongly enriched in As by irrigation with groundwater containing As, other indirect paths of human exposure to As cannot be ruled out. It has been pointed out that consumption of rice straw by cattle could potentially lead to increased As levels in meat or milk (Abedin et al., 2002a). Other crops grown in fields irrigated with groundwater can contain significantly higher levels of As than rice (e.g. Mandal et al., 1998; Roychowdhury et al., 2002; Das et al., 2004), but no properly controlled study has to our knowledge been conducted to directly link these elevated levels to the As content of irrigation water. Two potentially significant consequences of continued build-up of As in fields irrigated with groundwater are reduced crop yields (Abedin et al., 2002b; Duxbury et al., 2003) and exposure of As through ingestion of soil elevated in As.

Increased exposure to As of children is of particular concern since they spend a considerable amount of time playing in fields surrounding their village during the dry season and, a recent study from Araihazar has shown, their mental development is impaired by exposure to As from groundwater (Wasserman et al., 2004). A typical figure for soil ingestion by children that is used for risk estimates is 100 mg/day of soil, although one order of magnitude higher ingestions have been reported (Hemond and Solo-Gabriele, 2004). Even for a worst-case scenario under which 1 g/day of soil containing 30 mg/kg As that is entirely bioavailable is ingested, the corresponding dose of 30 μg As per day would be less than a third of the dose for a person drinking 2 L of water containing 50 μg/L As. All the consequences of continued As build-up in soils need to be carefully considered in future studies, but providing safe water to the rural population of Bangladesh and other South Asian countries afflicted with elevated As concentrations in groundwater should clearly remain the priority.

5. Conclusion

No one doubts that the tens of millions of households in several South Asian countries that drink untreated groundwater should stop doing so if their well water is elevated in As. The potential health effects of eating rice irrigated with groundwater containing As, on the other hand, are still debated. By comparing several rice paddies from Bangladesh, including a control site, we have shown here that As supplied with irrigation water accumulates in soil and in soil water but, thankfully, much less so if at all in rice grains. The remarkable shielding of rice grains from the build-up of As in soil and soil water of paddies irrigated with groundwater is consistent with several previous studies. The health risks due to ingestion of As contained in rice therefore appears to be dwarfed in countries such as Bangladesh where people are exposed to much higher As levels by drinking groundwater. High priority should nevertheless be given to continued study of all the potential impacts of continued irrigation with groundwater that contains elevated As concentrations.

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Appendix A. Supplemantary Data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.scitotenv.2006.01.030.

References

Abedin MdJ, Cresser MS, Meharg AA, Feldmann J, Cotter-Howells J. Arsenic accumulation and metabolism in rice (*Oryza sativa* L.). Environ Sci Technol 2002a;36:962–8.

Abedin MdJ, Cotter-Howells J, Meharg AA. Arsenic uptake and accumulation in rice (*Oryza sativa* L.) irrigated with contaminated water. Plant Soil 2002b;240:311–9.

Ali MA, Badruzamman ABM, Jalil MA, Hossain MD, Ahmed MF, Masud AA, et al. Arsenic in plant-soil environment in Bangladesh. In: Ahmed MF, Ali MA, Adeel Z, editors. Fate of Arsenic in the Environment. Tokyo: Bangladesh University of Engineering and Technology, Dhaka/The United Nations University; 2003. p. 85–112. (http://www.unu.edu/env/Arsenic/BUETSymposiumProc.htm).

Berg M, Tran HC, Nguyen TC, Pham HV, Schertenleib R, Giger W. Arsenic contamination of groundwater and drinking water in Vietnam: a human health threat. Environ Sci Technol 2001;35:2621–6.

BGS, DPHE. Arsenic contamination of groundwater in Bangladesh. In: Kinniburgh DG, Smedley PL, editors. Final report, BGS technical report WC/00/19. Keyworth, UK: British Geological Survey; 2001.

Chakraborti D, Sengupta MK, Rahman MM, Ahamed S, Chowdhurry UK, Hossain MA, et al. Groundwater arsenic contamination and its health effects in the Ganga–Meghna–Brahmaputra plain. J Environ Monit 2004;6:74N–83N.

Cheng Z, Zheng Y, Mortlock R, van Geen A. Rapid multi-element analysis of groundwater by high-resolution inductively coupled plasma mass spectrometry. Anal Bioanal Chem 2004;379:513–8.

Das HK, Mitra AK, Sengupta PK, Hossain A, Islam F, Rabbani GH. Arsenic concentrations in rice, vegetables, and fish in Bangladesh: a preliminary study. Environ Int 2004;30:383–7.

Dhar RK, Biswas BK, Samanta G, Mandal BK, Chakraborti D, Roy S, et al. Groundwater arsenic calamity in Bangladesh. Curr Sci 1997;73:48–59.

Duxbury JM, Mayer AB, Lauren JG, Hassan N. Food chain aspects of arsenic contamination in Bangladesh: effects on quality and productivity of rice. J Environ Sci Health 2003;A38(1):61–9.

He Y, Zheng Y, Ramnaraine M, Locke DC. Differential pulse cathodic stripping voltammetric speciation of trace level inorganic compounds in natural water samples. Anal Chim Acta 2004;511:55–61.

Hemond HF, Solo-Gabriele HM. Children's exposure to arsenic from CCA-treated wooden decks and playground structures. Risk Anal 2004;24:51–64.

Hironaka, H., Ahmad, SA. Arsenic concentration of rice in Bangladesh. In: Ahmed, MF, Ali, MA, Adeel, Z, editors. Fate of Arsenic in the Environment. Tokyo: Bangladesh University of Engineering and Technology, Dhaka/The United Nations University; 2003. p. 123–130. (http://www.unu.edu/env/Arsenic/BUETSymposiumProc.htm).

Horneman A, van Geen A, Kent DV, Mathe PE, Zheng Y, Dhar RK, et al. Decoupling of As and Fe release to Bangladesh groundwater under reducing conditions. Part I: evidence from sediment profiles. Geochim Cosmochim Acta 2004;68:3459–73.

Huq SM, Rahman A, Sultana N, Naidu R. Extent and severity of arsenic contamination in soils of Bangladesh. In: Ahmed MF, Ali MA, Adeel Z, editors. Fate of Arsenic in the Environment. Tokyo: Bangladesh University of Engineering and Technology, Dhaka/The United Nations University; 2003. p. 69–84. (http://www.unu.edu/env/Arsenic/BUETSymposiumProc.htm).

Mandal BK, Chowdhury TR, Samanta G, Mukherjee DP, Chanda CR, Saha KC, et al. Impact of safe water for drinking and cooking on five arsenic-affected families for 2 years in West Bengal, India. Sci Total Environ 1998;218:185–201.

Manheim FT. A hydraulic squeezer for obtaining interstitial waters from consolidated and unconsolidated sediments. US Geol Surv Prof Pap 1966;500-C:256–61.

Meharg AA, Rahman MdM. Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption. Environ Sci Technol 2003;37:229–34.

Norra S, Berner ZA, Agarwala P, Wagner F, Chandrasekharam D, Stüben D. Impact of irrigation with As rich groundwater on soil and crops: a geochemical case study in West Bengal Delta Plain, India. Appl Geochem 2005;20:1890–906.

Roychowdhury T, Uchino T, Tokunaga H, Ando M. Survey of arsenic in food composites from an arsenic-affected area of West Bengal, India. Food Chem Toxicol 2002;40:1611–21.

Schoof RA, Yost LJ, Eickhoff J, Crecelius EA, Cragin DW, Meacher DM, et al. A market basket survey of inorganic arsenic in food. Food Chem Toxicol 1999;37(8):839–46.

Takahashi Y, Minakawa R, Hattori KH, Kurushima K, Kihou N, Yuita K. Arsenic behavior in paddy fields during the cycle of flooded and non-flooded periods. Environ Sci Technol 2004;38:1038–44.

van Geen A, Zheng Y, Versteeg R, Stute M, Horneman A, Dhar R, et al. Spatial variability of arsenic in 6000 tube wells in a 25 km^2 area of Bangladesh. Water Resour Res 2003a;39(5):1140. doi:10.1029/2002WR001617.

van Geen A, Ahmed KM, Seddique AA, Shamsudduha M. Community wells to mitigate the current arsenic crisis in Bangladesh. Bull World Health Organ 2003b;82:632–8.

Wasserman GA, Liu X, Parvez F, Ahsan H, Factor-Litvak P, van Geen A, et al. Water arsenic exposure and children's intellectual function in Araihazar, Bangladesh. Environ Health Perspect 2004;112:1329–33.

Williams PN, Price AH, Raab A, Hossain SA, Feldmann J, Meharg AA. Variation in arsenic speciation and concentration in paddy rice related to dietary exposure. Environ Sci Technol 2005;39:5531–40.

Zheng Y, van Geen A, Stute M, Dhar R, Mo Z, Cheng Z, et al. Geochemical and hydrogeological contrasts between shallow and deeper aquifers in two villages of Araihazar, Bangladesh: implications for deeper aquifers as drinking water sources. Geochim Cosmochim Acta 2005;22:5203–18.