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Arsenic Contamination of Paddy Fields through Groundwater Irrigation in Bangladesh: Risks for Rice Production and Mitigation Perspectives

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Abstract—In a three-year field study in Munshiganj District, Bangladesh, we investigated the biogeochemical fate of arsenic (As) in groundwater-irrigated and seasonally-flooded paddy fields. We found that most irrigation-derived As was retained in the top few centimetres of paddy soil, and that As input to soil decreased with increasing distance from the irrigation inlet within each field. Based on concentration profiles measured in soil porewater and floodwater, it was shown that monsoon flooding leads to reductive mobilization of As from paddy soil. Combining this understanding with a three-year data set of soil As contents determined before and after flooding, we estimated that 24–48% of the As annually added to the study field was removed from the soil during the monsoon season. These results indicate that monsoon flooding strongly attenuates, but does not prevent the accumulation of As in paddy soils. Our study further showed that intermittent irrigation, as practiced by farmers at our study site, spatially constrains and temporally limits As release into porewater during rice growth. These findings suggest that intermittent irrigation, currently promoted in Bangladesh for water-saving purposes, may be a promising means of reducing As input to paddy soils and rice plant exposure to As.

Keywords: arsenic, Bangladesh, paddy fields, intermittent irrigation, monsoon flooding, long-term accumulation

INTRODUCTION

Geogenic contaminants in groundwater pose a health risk to hundreds of millions of people worldwide. In Bangladesh, arsenic (As) contamination of groundwater

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is threatening both drinking water supply and food production. National rice production has more than doubled over the past three decades (MoA, 2007), mainly due to increasing cultivation of dry-season rice (*boro*), which requires intense irrigation. Since the irrigation demand is predominantly met by groundwater from shallow aquifers, often containing high As concentrations, an estimated average of ~0.4 kg ha^{-1} As is added to paddy soils each year (Ali *et al.*, 2003; Dittmar *et al.*, 2010b). Arsenic is a non-threshold class 1 carcinogen, present in the environment as mainly inorganic arsenate (As^{V}) under oxic conditions and mainly arsenite (As^{III}) under reducing conditions (Smedley and Kinniburgh, 2002). As^{III} is more toxic than As^{V} (Basu *et al.*, 2001). At circumneutral pH, As^{III} is present as an uncharged species, and is therefore considered more mobile than the negatively charged As^{V} species (Smedley and Kinniburgh, 2002). Though groundwater irrigation has greatly improved food security in Bangladesh, the associated input of As to paddy soils may jeopardize sustainable rice production in the long-term. The adverse impacts of As input to paddy fields therefore urgently need to be understood and appropriate mitigation strategies need to be devised.

The aims of our study were (i) to assess the spatial distribution of As in irrigated rice fields, (ii) to elucidate the geochemical behaviour of As in paddy fields and estimate As gains and losses from paddy soil during irrigation and monsoon flooding, (iii) to investigate the effect of intermittent irrigation, as practiced by local farmers on As dynamics in soil porewater during rice growth and (iv) to assess possible long-term effects of continued As input on rice plant As uptake.

MATERIALS AND METHODS

Field site

The study was conducted in farmers' fields in Munshiganj district, 30 km south of Dhaka and 5 km north of the river Ganges. The fields are subject to intense monsoon flooding between mid-June and late October (Harvey *et al.*, 2006) and have been used for single crop *boro* cultivation during the dry season (January–May) since the early 1990s. The fields are irrigated with ~1.1 m a^{-1} groundwater from a single irrigation well delivering water containing 400 μg L^{-1} As (84% As^{III}), 11 mg L^{-1} Fe, 2 mg L^{-1} P and 20 mgL^{-1} Si (Roberts *et al.*, 2007). Irrigation water is distributed by shallow channels and enters each field through a breach (inlet) in the field bund. Irrigation events last 2–4 hours at a time and add 3–10 cm of water to the fields. Between irrigation events, water level on the fields is allowed to decrease until the moist soil surface is exposed (local farming practice).

Sample collection and analysis

Irrigation water was sampled from the well, along irrigation channels, and from two paddy fields situated at different distances from the irrigation well for selected irrigation events during the *boro* seasons 2005–2007. Within fields, water samples were collected at different distances from the irrigation inlet and at different times after irrigation. In addition, samplers yielding vertical porewater profiles of 1.5 cm resolution were deployed during vegetative crop growth in February 2006, and during the grain-filling stage in April 2005 and 2007. Water samples were collected into pre-acidified vials (i) unfiltered, (ii) after 0.2 μm-filtration and (iii) after filtration through a 0.2-μm filter combined with a modified As speciation cartridge (Roberts *et al.*, 2007), thus allowing for differentiation between dissolved and total element contents, and between dissolved As^{III} and As^{V}. Arsenic was determined by hydride generation atomic fluorescence spectrometry (HG-AFS; PS Analytical Ltd., U.K.); Fe, P, Si, Ca, Mg, Na, K and S by inductively coupled plasma optical emission spectrometry (ICP-OES; Spectro Ciros CCD).

Over three consecutive years (2005–2007), soil cores (0–40 cm) were collected on a grid of 38 sampling points in a single paddy field at the end of the irrigation season and after monsoon flooding, respectively. Cores were divided into 0–10, 10–25 and 25–40 cm depth segments and analyzed by X-ray fluorescence spectrometry (XRF) for total concentrations of As and other elements. In the same years rice plants were sampled along a transect across the same paddy field, corresponding to a soil As concentration gradient. Complete tillers were collected, divided into subsamples of dehusked grains and straw, and analyzed for total As by HG-AFS after milling and microwave digestion with nitric acid and H_2O_2.

During the monsoon seasons 2006 and 2007, floodwater samples were collected in the water column overlying the paddy fields and in a channel connecting the field area to the local river, to assess concentration depth profiles of As and other elements at different time points after the onset of flooding. In addition, soil porewater profiles were obtained from a paddy field at different time points after the onset of flooding in 2006 and 2007. Floodwater and monsoon porewater samples were preserved and analysed as described above for water samples collected during the irrigation season.

RESULTS AND DISCUSSION

The analyses of irrigation water samples showed that Fe^{II} was oxidized during water flow along channels and across fields resulting in Fe(hydr)oxide colloid formation and sorption of As and PO_4^{3-} to these colloids (Roberts *et al.*, 2007). Due to fast water flow and short residence times, aggregation and settling of As-bearing colloids was not significant within irrigation channels. Total As concentrations therefore remained unchanged during channel flow and rice paddies received the same total amount of As, independently of the distance between irrigation well and field inlet (Roberts *et al.*, 2007). By contrast, the slow spread of irrigation water across paddy fields favoured both colloid settling and As adsorption to soil minerals, and thereby led to pronounced As gradients in water sampled across paddy fields at the close of irrigation events (Fig. 1a) (Roberts *et al.*, 2007). Consistent with this, clear gradients in soil As content were found in paddy field top soil, with soil As contents decreasing with increasing

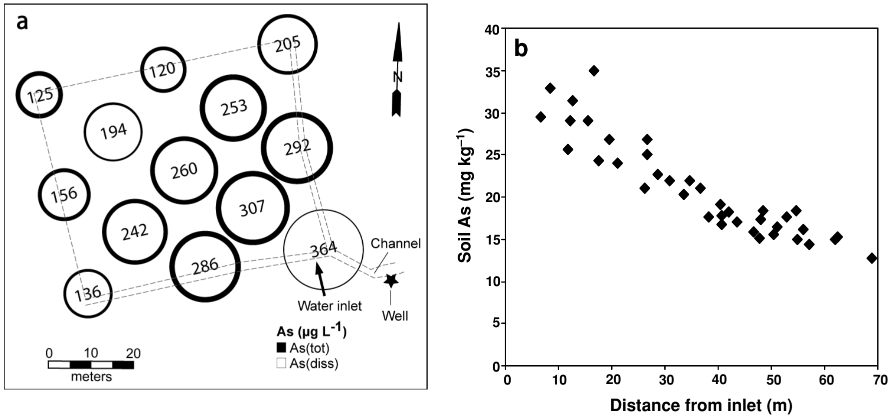

Fig. 1. Spatial distribution of arsenic in a paddy field in Munshiganj District, Bangladesh. (a) Total (As(tot)) and dissolved (As(diss)) arsenic concentrations (μg L^{-1}) in irrigation water shortly after an irrigation event in February 2006. (b) Topsoil (0–10 cm) As content (mg kg^{-1}) at the end of the irrigation season in May 2005 (data from Roberts *et al.*, 2007 (a) and Dittmar *et al.*, 2007 (b)).

distance from the irrigation water inlet (Fig. 1b) (Dittmar *et al.*, 2007). Depth profiles of As soil concentrations between 0 and 60 cm depth revealed that soil As accumulation mainly affected the top 20 cm of soil mixed once a year by ploughing, and that seasonal As accumulation was especially pronounced in the top few centimetres of soil (Dittmar *et al.*, 2007). The three-year data set on soil As concentrations collected across a paddy field showed that increases in soil As contents after the irrigation season were followed by decreases in soil As content after monsoon flooding (Dittmar *et al.*, 2010b).

We investigated mechanisms of As release from paddy soil during monsoon flooding based on soil porewater and floodwater data obtained in 2006 and 2007. Porewater concentration profiles showed concomitant release of Fe, Mn, P and As in the top 20 cm of soil corresponding to the layer above the plough pan, suggesting mobilization of P and As by reductive dissolution of Fe(hydr)oxides (Roberts *et al.*, 2010). Arsenic concentrations in porewater increased over time of flooding (Fig. 2a) and were dominated by As^{III}, indicating that As mobilization was further promoted by reduction of As^{V} to the less competitively sorbing As^{III} (Roberts *et al.*, 2004). Floodwater samples showed pronounced concentration gradients of As and other elements (with concentrations decreasing towards shallower water depths) on some days (Fig. 2b), and vertically well-mixed water columns on other days (Roberts *et al.*, 2010). Both situations were observed at early as well as late stages of monsoon flooding, indicating that As release occurred throughout the flooded period and that released As was distributed throughout the water column by vertical mixing. Increasing As concentrations in a channel connecting the field area to the local river on days of receding

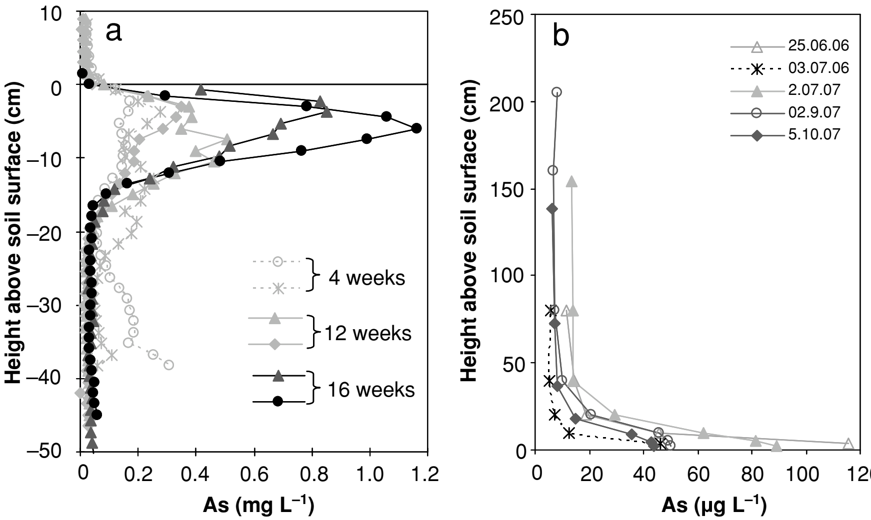

Fig. 2. Release of arsenic from paddy soil during monsoon flooding. (a) Soil porewater profiles of total As (mg L^{-1}) in a paddy field after 4, 12 and 16 weeks of flooding in 2007. (b) Floodwater profiles of total As (μg L^{-1}) on selected days in 2006 and 2007 when the floodwater column was characterized by distinct concentration gradients (data from Roberts *et al.*, 2010).

floodwater level exemplarily revealed As export from the field area to the river (Roberts *et al.*, 2010).

Based on vertical porewater As gradients in topsoil porewater and on changes in floodwater As concentrations over delimited time periods we derived estimates of seasonal As flux from soil into floodwater. It was estimated that between 51 and 250 mg m^{-2} of soil As was released into floodwater over the course of one monsoon season, corresponding to a loss of 13–62% of the As added to soil through irrigation each year. Mass balance calculations based on the three-year data set of soil As concentrations before and after flooding further constrained this release estimate to between 105 and 210 mg m^{-2} a^{-1} (Dittmar *et al.*, 2010b). In addition, the mass balance showed that not all As added during irrigation was retained in the top 40 cm of soil, thereby constraining the estimate of net annual As accumulation in this depth range to 240 ± 40 mg m^{-2} a^{-1}. Predictive scenarios assuming As loss from paddy soil to be constant or proportional to soil As content, suggest an increase in soil As content by a factor of 1.5 to 2 by 2050 (Dittmar *et al.*, 2010b). Analysis of rice plants collected between 2005 and 2007 showed straw and grain As content to be more elevated close to the irrigation inlet that in more distant parts of the field where soil As content was lower (Dittmar *et al.*, 2010a). This suggests that the expected future increase in soil As content will likely result in further increases in straw and grain As content. Our conclusion is corroborated by data from a pot study using As-spiked soil from our field site, which shows As in plant tissue to increase beyond levels currently found at our field site (Dittmar *et al.*, 2010a).

Soil porewater profiles obtained during the *boro* season when the fields were intermittently irrigated showed As concentration and speciation in porewater to vary significantly over space and time. Porewater As concentrations measured close to the irrigation inlet were higher than at the far field corner, thus reflecting the As gradient in the topsoil of the study field (Roberts *et al.*, 2011). During early rice growth, As porewater concentrations near the inlet reached up to 500 μg L^{-1} and were dominated by As^{III}, but As release was constrained to the lower portion of the soil above the plough pan (Roberts *et al.*, 2011). In the later part of the season, soil conditions were oxic throughout the depth range relevant to rice roots and porewater concentrations only intermittently increased to ~150 ug L^{-1} As^{V} following irrigation events. These results suggest that rice plant exposure to As may be lower under intermittently irrigated conditions than under continuous water ponding, where As porewater concentrations generally increase or reach a plateau until the soil is drained for harvest (Bogdan and Schenk, 2008; Xu *et al.*, 2008; Li *et al.*, 2009; Panaullah *et al.*, 2009). This suggests that intermittent irrigation, which is currently advocated in Bangladesh for water-saving purposes, may be a promising means of reducing As input to paddy soils and rice plant exposure to As.

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