



## Arsenic transport in irrigation water across rice-field soils in Bangladesh



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### ABSTRACT

Experiments were conducted to analyze processes impacting arsenic transport in irrigation water flowing over bare rice-field soils in Bangladesh. Dissolved concentrations of As, Fe, P, and Si varied over space and time, according to whether irrigation water was flowing or static. Initially, under flowing conditions, arsenic concentrations in irrigation water were below well-water levels and showed little spatial variability across fields. As flowing-water levels rose, arsenic concentrations were elevated at field inlets and decreased with distance across fields, but under subsequent static conditions, concentrations dropped and were less variable. Laboratory experiments revealed that over half of the initial well-water arsenic was removed from solution by oxidative interaction with other water-column components. Introduction of small quantities of soil further decreased arsenic concentrations in solution. At higher soil-solution ratios, however, soil contributed arsenic to solution via abiotic and biotic desorption. Collectively, these results suggest careful design is required for land-based arsenic-removal schemes.

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### 1. Introduction

Arsenic transport via surface and overland flow is a potentially significant pathway for increasing arsenic contamination in the environment. Arsenic is believed to be relatively immobile in surface water, due to its low solubility under oxic conditions (Smedley and Kinniburgh, 2002). Yet it is frequently found as a contaminant in variety of flowing surface-water settings. Across the world, above-ground transport of arsenic is threatening water quality and plant health by redistributing arsenic from sources such as irrigation water, animal wastes, pesticides, mine wastes, and geothermal waters (Brown et al., 2007; Church et al., 2010; Pichler et al., 2008; Roberts et al., 2007; Wilkie and Hering, 1998).

In Bangladesh, dry-season irrigation with groundwater has enabled the expansion of rice production, greatly improving food security and economic opportunity for farm households (Hossain et al., 2003). Extensive use of arsenic-contaminated groundwater

for irrigation during the dry season threatens these benefits. Following years of irrigation with groundwater, soil arsenic concentrations have risen, and arsenic is now transferring into rice at concentrations sufficient to decrease yields and create dangerous levels of arsenic in rice grains (Abedin et al., 2002; Brammer and Ravenscroft, 2009; Meharg and Rahman, 2003; Meharg et al., 2009; Stroud et al., 2011a; Williams et al., 2006). Due to the large volumes of irrigation water required, as well as the cost of highly technical treatment options, there are currently no practical methods for large-scale removal of arsenic from these systems (Brammer, 2009; Brammer and Ravenscroft, 2009). Whereas numerous studies have investigated technology-based arsenic removal from drinking water (e.g., Garelick et al., 2005) and streams (e.g., Vaclavikova et al., 2008), there has been less work investigating options that maximize arsenic removal from water flowing across soils – a task needed to prevent current and impending environmental and food security risks (Brammer, 2009; Duxbury et al., 2003; Stone, 2008).

A necessary first step for better managing arsenic loading to fields is to understand the processes impacting arsenic transport in and removal from flowing irrigation water. In Bangladesh and other areas of Southern Asia, once well water is brought to the surface for irrigation, it is commonly distributed through a network of channels and then used to fill rice fields (Hossain et al., 2008; Roberts

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et al., 2007). With a complete understanding of arsenic behavior within this context, it may be possible to design land-based arsenic removal schemes to reduce arsenic loading to field soils.

Broadly, arsenic retention and transport depend on pH, arsenic speciation, redox conditions, mineralogy, and aqueous chemistry (Smedley and Kinniburgh, 2002). Arsenic concentration distributions in flowing water are determined by the extent of oxidation, adsorption, and precipitation reactions, relative to flow rates, particle settling capacities, and (re-)release of arsenic from soils (e.g., Cadwalader et al., 2011; Ciardelli et al., 2008; Dixit and Hering, 2003; Langner et al., 2001; Saha et al., 2006; Smedley and Kinniburgh, 2002). Redox cycling of iron largely impacts arsenic solubility through reductive dissolution of Fe(III)(hydr)oxides, which may concomitantly release arsenic to solution, and oxidative precipitation of dissolved Fe(II), which forms Fe(III)(hydr)oxides that may scavenge arsenic from solution (Smedley and Kinniburgh, 2002). Phosphate (Darland and Inskeep, 1997), silicic acid (Waltham and Eick, 2002), carbonate (Appelo et al., 2002), and natural organic matter (Redman et al., 2002) may each compete with arsenic for sorption and/or co-precipitation sites, limiting arsenic removal from flowing water and enhancing arsenic transport.

As a result of these (and other) processes, the distribution of arsenic species along surface flow systems may be heterogeneous (Frau and Arda, 2003; Wilkie and Hering, 1998). In Southern Asia, dissolved arsenic concentrations in irrigation water flowing through distribution channels generally decrease with distance from wells (Hossain et al., 2008; Roberts et al., 2007), but arsenic may be further transported in suspended colloids (Roberts et al., 2007). Across fields, where irrigation water flows much more slowly than in channels, the extent of arsenic transport has been inferred by decreases in arsenic concentrations in rice field soil (Dittmar et al., 2007; Hossain et al., 2008; Norra et al., 2005; Stroud et al., 2011a) and standing irrigation water (Roberts et al., 2007; Stroud et al., 2011a). Overall, rice fields tend to be a sink for arsenic (Neumann et al., 2011), although arsenic may potentially be lost from fields due to irrigation-water infiltration along field boundaries (Neumann et al., 2009), porewater cycling (Roberts et al., 2011), desorption from soils (Saha et al., 2006), monsoonal floodwater retreat (Roberts et al., 2010; Saha and Ali, 2007), and uptake into rice (Stroud et al., 2011b).

To date, there has been only limited investigation of spatio-temporal distributions of arsenic in flowing irrigation water in Bangladesh. Here, we describe field and laboratory experiments analyzing physicochemical processes impacting arsenic concentrations and transport in contaminated irrigation water flowing across bare rice-field soils. Our specific objectives were to 1) examine arsenic transport in flowing irrigation water; 2) compare spatiotemporal distributions of arsenic with distributions of representative elements (Fe, P, and Si) expected to impact arsenic concentrations during flowing- and static-water conditions; and 3) define abiotic and biotic processes impacting arsenic removal from solution.

## 2. Methods

### 2.1. Field site

Our field studies were conducted in Munshiganj, Bangladesh at a well-characterized site that suffers from arsenic-contaminated groundwater (Harvey et al., 2002; Hug et al., 2011; Neumann et al., 2010; Polizzotto et al., 2005, 2006), where farmers are highly dependent on groundwater-irrigated rice agriculture (Neumann et al., 2009), and where arsenic removal from irrigation water has been previously demonstrated by variable spatial distributions of arsenic in rice-field soil and standing water (Dittmar et al., 2007; Roberts et al., 2007). Elevated arsenic concentrations (i.e.  $>10$   $\mu\text{g/L}$ , the World Health Organization drinking-water guideline (WHO, 2011)) in groundwater are persistent throughout the shallow

Holocene aquifer at the site; concentrations peak to  $\sim 400$   $\mu\text{g/L}$  at  $\sim 30$  m depth, the approximate depth of irrigation wells in the area (Harvey et al., 2002). Well water at the site is anoxic, with redox conditions at the potential of methanogenesis (Polizzotto et al., 2005), elevated concentrations of dissolved iron (10.9 mg/L), and dissolved oxygen  $<1$  mg/L (Roberts et al., 2007). During the dry season, well water is fed through a network of distribution channels to irrigate rice fields (Fig. 1).

Field experiments and soil sampling were performed in December, 2011, following monsoon floodwater recession and prior to boro rice planting. Fields were bare and plowed, which left an irregular soil surface and turned over the portion of soil previously exposed to irrigation and monsoon water. Soil samples for laboratory experimentation were collected from a field between the two experimental plots prior to the first seasonal application of irrigation water (Fig. 1). The surface layer of soil ( $\sim 0.5$  cm) was scraped away and samples were collected from the next 2–3 cm. Soils were stored in plastic bags at  $4^\circ\text{C}$ .

Experiments were conducted in two fields (Fig. 1). The first field was rectangular ( $22\text{ m} \times 41\text{ m}$ ), and the inlet for irrigation water was linked to the irrigation well by a  $\sim 94$  m long irrigation channel. In the second field, to restrict water loss through the unplowed field perimeter, two circular plots (diameters of 30.5 m and 11.9 m) were constructed with walls of mounded soil over plowed areas of the field (Neumann et al., 2009; Patil et al., 2011), resulting in a field layout that is atypical to that commonly observed in Bangladesh. The circles were connected with each other and the inlet was located approximately 40 m from the irrigation well.

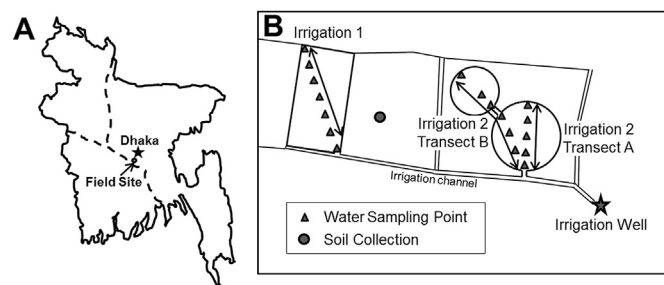
### 2.2. Field flowing irrigation water experiments

Fields were irrigated to  $\sim 10$  cm water depth, a typical depth for rice production in the area. The first field took 1 h and 3 min to fill, and the second field took 1 h and 35 min to fill. Pumping rates were  $\sim 18$ – $20$  L/s and remained constant through the experiments. Flowing and static irrigation water were sampled along transects that spanned the fields (Fig. 1). Samples were collected in a time series. Initial samples (“wetting front”) were collected as a thin, turbid layer of irrigation water flowed preferentially and first arrived at each sampling point (Supplementary Fig. S1). Next, “sheet flow” samples were collected as the irrigation water progressively covered the field by  $\sim 2$ – $3$  cm of water. “Full flow” samples were collected when field irrigation was completing and water depths were  $\sim 10$  cm. After the fields were filled, “static” (i.e. standing, infiltrating and evaporating) water samples were collected  $\sim 4$ ,  $\sim 24$ , and  $\sim 48$  h after irrigation was initiated, mimicking previous studies (Roberts et al., 2007; Stroud et al., 2011a).

Water samples were collected from approximately the middle of the water column into field-rinsed 60 mL syringes, and filtered through  $0.2$   $\mu\text{m}$  nylon filters (VWR). During flowing water conditions, water was allowed to flow directly into the open top of the syringe. During sampling, pH, dissolved oxygen, and conductivity were measured using a Hanna HI 9828 multiparameter meter; to make measurements, irrigation water was collected into a sampling cup that was screwed directly onto the meter, and values were recorded following parameter stabilization. Samples were acidified with concentrated trace-metal-grade nitric acid (1 drop acid/10 mL sample) and stored at  $4^\circ\text{C}$  until analysis.

### 2.3. Adsorption and desorption studies

To examine limits on arsenic removal from solution and retention by rice-field soils, microcosm arsenic adsorption/desorption studies were conducted. Prior to each experiment, soils were homogenized in the laboratory in HDPE beakers. Sub-



**Fig. 1.** A. Map of Bangladesh indicating location of field site. Dotted lines represent major rivers. B. Field area in Munshiganj, Bangladesh. Experiments were conducted in two fields. The first field, Irrigation 1, was  $22\text{ m} \times 41\text{ m}$ , and the inlet for irrigation water was linked to the irrigation well by a  $94\text{ m}$  long irrigation channel. A diagonal transect across the field was sampled at regular increments. In the second field, Irrigation 2, two interconnected circular plots with diameters of  $30.5\text{ m}$  and  $11.9\text{ m}$  were constructed with walls of mounded soil over plowed areas of the field. The inlet was located approximately  $40\text{ m}$  from the irrigation well, and two transects, A and B, were sampled. Soil samples for laboratory experimentation were obtained in an open field between the Irrigation 1 and 2 fields.

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