



## Evidence of decoupling between arsenic and phosphate in shallow groundwater of Bangladesh and potential implications



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### ABSTRACT

Reductive dissolution of iron oxyhydroxides and reduction of arsenic are often invoked as leading causes of high dissolved As levels in shallow groundwater of Bangladesh. The second of these assumptions is questioned here by comparing the behavior As and phosphate (P), a structural analog for As (V) which also adsorbs strongly to Fe oxyhydroxides but is not subject to reduction. The first line of evidence is provided by a detailed groundwater time-series spanning two years for three wells in the 6–9 m depth range showing removal of As(III) from shallow groundwater during the monsoon without loss of P. The data indicate a loss of ~90% of the dissolved As from groundwater in the intermediate well relative to a level of 3  $\mu\text{mol/L}$  As predicted by conservative mixing between groundwater sampled from the shallower and the deeper well. In contrast, P concentrations of ~30  $\mu\text{mol/L}$  in the intermediate well closely match the prediction from conservative mixing. Reduction therefore appears to inhibit the release of As to groundwater at this site relative to P instead of enhancing it. A re-analysis of existing groundwater As and P data from across the country provides a broader context for this finding and confirms that, without reduction, elevated concentrations of As would probably be even more widespread in shallow aquifers of Bangladesh. Without providing definite proof, X-ray absorption spectroscopy of sediment from the time-series site and elsewhere suggests that the loss of As from groundwater may be coupled to precipitation of As sulfide. Further study is needed to assess the implications of these observations for shallow aquifers that have been subjected to increased withdrawals for irrigation in recent decades.

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### 1. Introduction

The presence of arsenopyrite in shallow sediment of the Bengal basin was initially interpreted as an indication that elevated groundwater As concentrations, known by then to be widespread in the region, were attributable at least in part to oxidation caused by enhanced recharge due to irrigation pumping (Chowdhury et al., 1999). Subsequent studies showed that this was unlikely because elevated As levels were associated with low sulfate levels instead of the other way around (BGS/DPHE, 2001; Harvey et al., 2002). Over the past decade, much of the attention has focused instead on reductive dissolution of iron oxyhydroxides and an enhanced

supply of reactive organic carbon that might promote the release of As to groundwater (e.g. Fendorf et al., 2010).

In comparison, less attention has been paid to the still potentially important role of As-sulfur interactions. This may be in part because such interactions could plausibly affect groundwater As in opposite ways and would therefore be difficult to disentangle. On one hand, the reduction of sulfur supplied by recent recharge or organic matter in the sediment could lead to the formation of insoluble As sulfides or the sequestration of As in iron sulfide phases (O'Day et al., 2004; Kirk et al., 2004; Lowers et al., 2007; Hery et al., 2010; Kocar et al., 2010; Xie et al., 2015). Without direct evidence from sediment analysis but on the basis of an extensive set of groundwater data, Buschmann and Berg (2009) made a convincing case that sulfate reduction had an inhibiting effect on the release of As to groundwater in South and Southeast Asia. In a field experiment in shallow aquifer of Bangladesh, the

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injection of molasses and sulfate resulted in a temporary reduction of As concentrations in groundwater (Saunders et al., 2008). On the other hand, the presence of sulfide has also been shown to enhance the release of As from sediments through the formation of As-sulfide complexes in solution, although primarily in laboratory studies (Keimowitz et al., 2007; Kocar et al., 2010; Burton et al., 2013).

In order to shed some light on the roles of Fe and S, as well as reduction to As(III), in controlling groundwater As concentrations in reducing aquifers, we follow a somewhat different approach in the present study by tracking the behavior of As and phosphate P(V), its structural analog in the oxidized state. This is done first on the basis of a detailed time series of these and other groundwater constituents across a redox interface in a village of Bangladesh and, subsequently, through the re-analysis of a previous landmark survey of the entire country. Synchrotron spectroscopy was also applied to sediment from the time-series site in attempt to identify As-sulfide phases but the outcome is ambiguous. The paper concludes by pointing out why further study is needed in order to predict the future evolution of As concentrations in reducing aquifers whose hydrology is being modified by pumping.

## 2. Methods

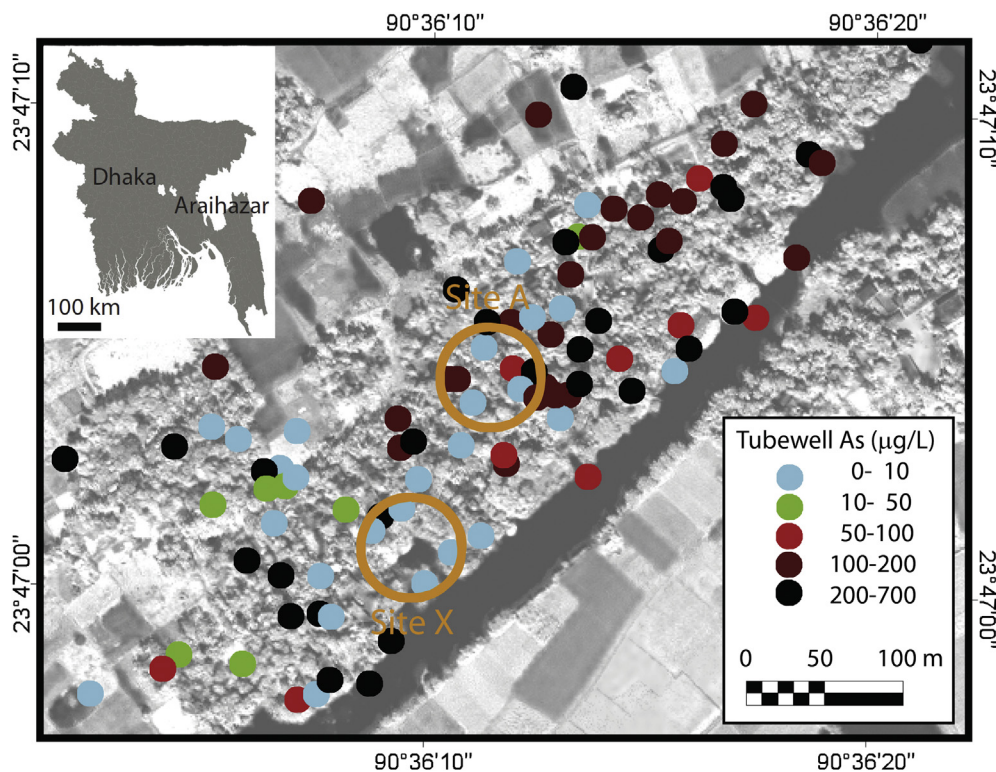
### 2.1. Study site

Time-series data were obtained from three monitoring wells installed in the village of Dari Satyabandi in central Bangladesh (Fig. 1). The vertical distribution of As in the shallow aquifer of Dari Satyabandi is representative of the initial rise in concentrations with depth observed throughout the impacted portions of the

Bengal Basin (BGS/DPHE, 2001; Harvey et al., 2002; van Geen et al., 2003). Sixty percent of shallow wells (<10 m) sampled in the village do not meet the WHO guideline for As in drinking water of  $10 \mu\text{g L}^{-1}$  ( $0.13 \mu\text{mol L}^{-1}$ ), and 90% exceed this value at >20 m depth (Fig. 2). Repeated sampling in Dari Satyabandi has shown that concentrations of As in groundwater at any given location increase consistently with depth and typically do not vary much over time (Cheng et al., 2005; Dhar et al., 2008). The vertical distribution of As in groundwater in the village is consistent with the presence of a sizeable clay layer that inhibits local recharge (Aziz et al., 2008). One documented exception is shallow household well #4115 where seasonal fluctuations in As concentrations ranged from 0.13 to  $1.6 \mu\text{mol L}^{-1}$  over a 3-year period (Cheng et al., 2005). Three new ~4-cm diameter PVC monitoring wells were installed within 2 m of this household well in January 2005 using the local “hand-flapper” method. The 0.9-m (3 ft) wells screen made of finely slotted PVC were centered on depths of 6, 7.5, and 9 m, respectively. The surficial clay layer extends to less than 3 m depth at this particular site. It is also located within 50 m of a local stream and may be therefore be more sensitive to surface forcing than the northern portion of the same village (Fig. 1).

### 2.2. Water sampling and field measurements

From January 2005 through December 2006, groundwater was sampled from the new monitoring wells with a submersible pump every two weeks after the well volume was flushed approximately 10 times using a submersible pump at a flow rate of 15 L/min. At the time of each sampling, groundwater levels were measured manually using a Solinst (Georgetown, Ontario, Canada) Model 102 m. Groundwater conductivity, oxidation–reduction potential (ORP),



**Fig. 1.** Map of As concentrations in shallow tubewells of Dari Satyabandi. IKONOS satellite image of the study area showing in dark grey the local stream passing through the area and the location of wells <20 m deep along its northern bank. The color-coded circles show the As content and location of 99 wells within the village in the 9–20 m depth range. Shown in orange are the location of the three monitoring wells (Site X) and that of additional vertical profiles shown in Fig. 2. The inset shows the location of the site relative to Dhaka. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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