

Solid-phases and desorption processes of arsenic within Bangladesh sediments

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Abstract

Arsenic is a contaminant in the groundwater of Bangladesh and West Bengal, India, where an estimated 57 million people may be drinking water with unsafe arsenic levels. The source of arsenic appears to be natural, solid-phase arsenic in the sediments, and various theories have been put forth regarding the modes of arsenic release, ranging from the oxidative or reductive degradation of arsenic-bearing solids to competitive ligand displacement by phosphate. Currently, however, reductive dissolution of Fe(III) (hydr)oxides and concomitant arsenic release is the most widely accepted explanation of high arsenic groundwater concentrations. Using micro-X-ray fluorescence elemental mapping and micro-X-ray absorption near-edge structure spectroscopy, we detect arsenic-bearing sulfides in the aquifer sediments from our field site in Munshiganj, Bangladesh. Reduction of Fe and As in surface soil layers is apparent, but Fe(III) (hydr)oxides are not detected in the Holocene aquifer materials. Rapid abiotic desorption of arsenic from sediments is observed in batch experiments, and positive controls with ferrihydrite dismiss the role of ferric (hydr)oxides in arsenic retention within the aquifer sediments. Based on our laboratory results, we do not see evidence for ferric (hydr)oxide reductive dissolution at well-depths in the Holocene aquifer. In contrast, our data suggest that arsenic is only released via redox cycling in surface soils/sediments and thus must then be transported to well-depth through the sandy aquifer.

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1. Introduction

Arsenic is a contaminant in the groundwater of Bangladesh and West Bengal, India, and an estimated 57 million people may be drinking water with arsenic concentrations above the World Health Organization standard of 10 µg/L (Yu et al., 2003). In an effort to

reduce the number of diseases due to surface water consumption, the Bangladesh government initiated a widespread project to obtain drinking water through groundwater retrieval in the late 1960s. Water-borne diseases have since decreased, but recent incidences of arsenicosis and cancer have resulted due to arsenic poisoning (Yu et al., 2003).

Bangladesh is a country sitting on kilometers of eroded Himalayan sediments, transported and deposited by the Ganges, Brahmaputra, and Meghna Rivers

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(Acharyya et al., 2000; Anwar et al., 2002), and it is believed that arsenic concentrations in groundwater are the consequence of the release of natural arsenic from these sediments (Acharyya et al., 1999; Chowdhury et al., 1999; Nickson et al., 2000; McArthur et al., 2001; Anwar et al., 2002; Harvey et al., 2002). Though regional variations are present, Bangladesh sediments typically consist of a thick Pleistocene orange sand aquifer, overlain by a clay aquitard, a thick dark-gray sandy Holocene aquifer, and a 0–5 m clay layer cap of overbank deposits (Ahmed et al., 2004). Throughout the country, groundwater arsenic concentrations decrease with depth (Yu et al., 2003), and at some intensive study sites the concentration profile has been found to peak between depths of 28 and 45 m in the Holocene aquifer. Concentrations are below detection limits in the Pleistocene aquifer (McArthur et al., 2001). Solid phase arsenic concentrations are relatively uniform with depth and are typically below 3 $\mu\text{g/g}$ in the aquifer sands (Swartz et al., 2004), but average ~ 20 $\mu\text{g/g}$ (Meharg and Rahman, 2003) and can be as high as 800 $\mu\text{g/g}$ (Breit et al., 2004) in the surficial clay layer. Well depths vary but tubewells in central Bangladesh generally tap groundwater in the Holocene aquifer at depths of ~ 30 m.

Currently, the mechanism of arsenic release remains unclear. Initial theories suggested that ion displacement by fertilizer-derived phosphate (Acharyya et al., 1999) or oxidation of arsenic-bearing sulfides (Chowdhury et al., 1999) has resulted in the partitioning of arsenic in the aqueous phase. However, these hypotheses have been discounted as viable mechanisms within the aquifer. Phosphate concentrations decrease with depth and do not correspond with arsenic levels (McArthur et al., 2001). Following the oxidative dissolution of sulfide minerals, resulting ferric (hydr)oxides would rapidly scavenge any concomitantly released arsenic (Mok and Wai, 1994). Furthermore, it has been proposed that primary sulfide grains are completely oxidized prior to deposition, and, therefore, sulfides detected within the aquifer materials are authigenic, formed following microbial sulfogenesis (McArthur et al., 2001).

The most widely accepted theory to date is that arsenic is released from aquifer sediments during the microbial reductive dissolution of ferric (hydr)oxides (Nickson et al., 1998, 2000; McArthur et al., 2001; Dowling et al., 2002; Harvey et al., 2002). Though yet to be detected in sediments from the Holocene aquifer at depths where groundwater arsenic concentrations are highest, Fe(III) (hydr)oxides are presumed to be derived from the weathering of micas, iron sulfides, and other primary Fe-bearing minerals. Support for reductive

dissolution of iron (hydr)oxides has been gleaned from the low redox potential (<90 mV) of the aquifer, high dissolved Fe content (Nickson et al., 2000; McArthur et al., 2001; Dowling et al., 2002; Harvey et al., 2002; Swartz et al., 2004), injection-withdrawal experiments (Harvey et al., 2002), and correlations of arsenic with methane, ammonium, and dissolved inorganic carbon (DIC)—indicators of biological activity (Dowling et al., 2002; Harvey et al., 2002).

Despite the support for the reductive dissolution theory, the organic carbon source fueling this process remains contentious. McArthur and colleagues (2001) argue that dissolved organic carbon is from sedimentary peat deposits within the aquifer. Alternatively, Harvey et al. (2002) have shown that groundwater dissolved organic carbon (DOC) ages are 3000–5000 years while dissolved inorganic carbon is substantially younger. Microbial processes in the aquifer are thought to be driven by young, labile organic carbon drawn down from surface waters. Tritium concentrations suggest that groundwater flow is rapid in the upper 30 m of the Holocene aquifer and since the onset of extensive dry-season irrigation with groundwater, surface to groundwater travel times have decreased from ~ 80 years or more to less than 40 years today (Harvey et al., this issue).

Conclusions concerning arsenic release have largely been drawn from observations of solution-phase chemistry. Recent studies have used X-ray diffraction (XRD), electron probe X-ray microanalysis (EPMA), transmission electron microscopy (TEM) (Akai et al., 2004), scanning electron microscopy (SEM) (Nickson et al., 2000; Akai et al., 2004), extractions (Swartz et al., 2004; Horneman et al., 2004), color (van Geen et al., 2003) and reflectance and magnetic properties (Horneman et al., 2004) to rectify this limitation; however, in general, solid-phases remain incompletely characterized or even identified. Microcosm incubation experiments with aquifer materials have been conducted, and while reductive dissolution has been observed from sediments from what appear to be the Pleistocene aquifer and oxidized portions of the Holocene aquifer (Akai et al., 2004; Islam et al., 2004; van Geen et al., 2004), it has not been shown for the reducing Holocene aquifer sediments where groundwater arsenic concentrations are typically highest (i.e. >50 $\mu\text{g/L}$ and often ≥ 600 $\mu\text{g/L}$).

Despite the broad consensus that reductive dissolution of ferric (hydr)oxides is the dominant mechanism of arsenic release within Bangladesh aquifers, a number of unexplained observations linger. Arsenic concentrations are spatially variant; wells separated by less than 100 m may provide water with drastically different levels (van

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