Arsenic in Shallow, Reducing Groundwaters in Southern Asia: An Environmental Health Disaster

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Arsenic concentrations in shallow, reducing groundwaters in Bengal, Southeast Asia, and elsewhere constitute a major hazard to the health of people using these waters for drinking, cooking, or irrigation. A comparison of occurrences in the Ganges–Brahmaputra, Mekong, and Red River basins shows that common geological characteristics include (1) river drainage from the rapidly weathering Himalayas, (2) rapidly buried organic-bearing and relatively young (ca. Holocene) sediments, and (3) very low, basin-wide hydraulic gradients. Anaerobic microbial respiration, utilizing either sedimentary or surface-derived organic carbon, is one important process contributing to the mobilization of arsenic from host minerals, notably hydrous iron oxides. In spite of the paucity of data from before the extensive development of groundwater pumping in these areas, there is sufficient evidence to make a *prima facie* case that human activity might exacerbate arsenic release into these groundwaters. The difficulties in implementing comprehensive groundwater remediation suggest serious attention should be given to developing treatment technologies for alternative surface-water supplies.

KEYWORDS: arsenic, groundwater, hazard, health, hydrogeology

INTRODUCTION

The use of hazardous, arsenic-bearing groundwater for drinking, cooking, and irrigation has led in Bangladesh to what has been described by a leading epidemiologist as the worst mass poisoning of a human population in history (Smith et al. 2000). While surface waters usually have a very low dissolved arsenic content, the widespread occurrence of hazardous pathogens in rivers and ponds prompted national and international agencies to turn to groundwater resources for both irrigation and drinking water. While mega-cities, such as Kolkata, can handle the high arsenic content in groundwater because of centralized adduction systems and treatment plants, small villages, where as many as 10 million new domestic wells may have been drilled in the last 40 years, face an enormous problem because the efficiency of water treatment is hard to control for such a large number of wells, and also because arsenic hotspots are extremely patchy. As a result of this usage, over 100,000 people in Bangladesh have already developed skin lesions. Millions of people are at risk of developing debilitating illnesses, including cancers, and excess deaths of the order of several thousand per year might be anticipated in the absence of effective remediation strategies (Yu et al. 2003). There is an extensive literature on the distribution and origin of arsenic in shallow groundwaters in Bangladesh (BGS, DPHE 2001; Chakraborti 2001; Harvey et al. 2005), on arsenic uptake by rice (Williams et al. 2005) and other foods (Al Rmalli et al. 2005), on human health impacts and regulatory issues (Smith and Smith 2004), and on remediation strategies (van Geen et al. 2003; Kanel et al. 2005).

Despite this, there remains considerable debate about the key controls on arsenic concentrations in these groundwaters, notably (1) the controls on arsenic transfer from sediments to groundwater by sediment mineralogy and chemistry (Gault et al. 2005) and microbiology (Islam et al. 2004), (2) the controls on arsenic release and flushing rates by the nature of the permeability structure in the host sediments (Harvey et al. 2005), (3) the relative importance of microbiological and inorganic release processes, and (4) the impact of surface-derived organic carbon on arsenic transfer from the host sediments to the groundwaters and hence the impact of anthropogenic processes on arsenic release rates. Here we provide a brief perspective on the latter two issues. We utilize data predominately from West Bengal and Bangladesh, which are among the worst-affected areas known in the world, and also from the Mekong and Red River basins in Cambodia and Vietnam, which are among the many countries where similar arsenic-bearing shallow, reducing groundwaters constitute a significant hazard to human health (see Smedley and Kinniburgh 2002 for an extensive list of affected areas).

ARSENIC IN GROUNDWATERS OF THE GANGES, MEKONG, AND RED RIVER BASINS

The Ganges–Meghna–Brahmaputra delta (BGS, DPHE 2001), the Mekong basin near Phnom Penh (Polya et al. 2003, 2005), and the Red River basin near Hanoi (Berg et al. 2001) have a variety of common features, including river drainage from the rapidly weathering Himalayas (*sensu lato*) (FIG. 1). Although groundwater contamination has also been reported upstream, for example, in Nepal and in Bihar, India, the major concerns are the delta areas of these river systems because of the high populations they support and will continue to support in the near future. In these delta areas, the three systems are also characterized by rapidly buried organic matter–bearing, relatively young (ca. Holocene) sediments and very low, basin-wide hydraulic gradients.

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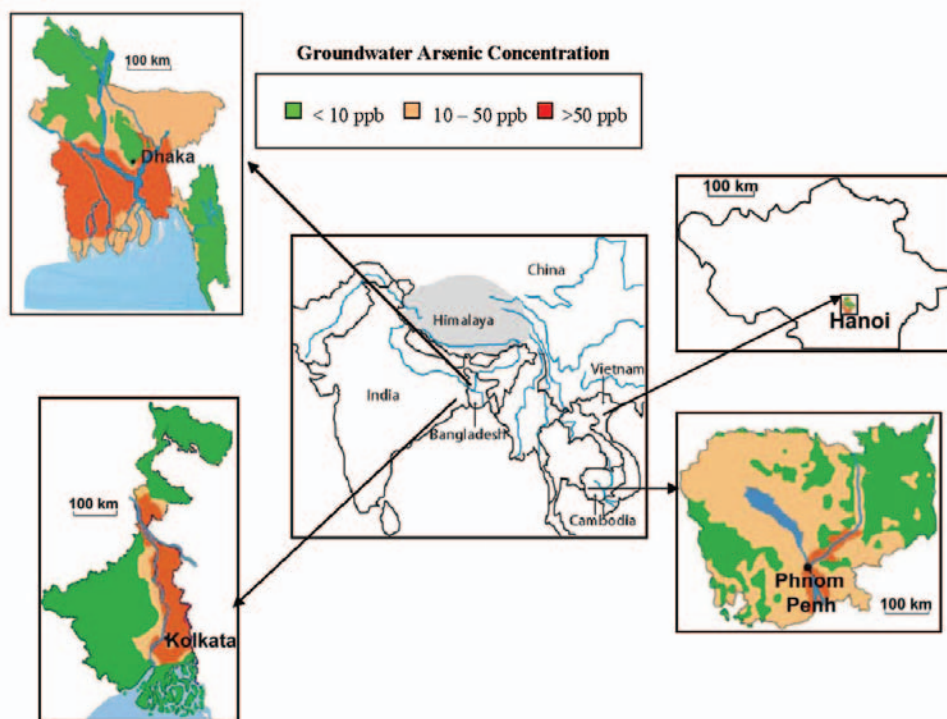*   Corresponding author: david.polya@manchester.ac.uk

tions are much more prevalent in West Bengal and Bangladesh than in Cambodia. Is this related to the relative magnitude and length of time of groundwater abstraction in the Bengal delta plain or to geological differences between the two regions?

In all three systems, the spatial heterogeneity of arsenic content occurs on very small scales, with high concentrations (>50 ppb) commonly being found within tens of metres of low concentrations. This may reflect the complex lithological structures of the aquifers, which include highly permeable channel fills as well as very low-permeability scoured channels filled with organic-rich overbank deposits. The interconnectivity of highly permeable channel sand units may be critical to determining groundwater arsenic distribution because of its control on groundwater flow patterns (Harvey et al. 2005) and on the rate of flushing of arsenic from the system. The British Geological Survey (BGS, DPHE 2001) and Smedley and Kinniburgh (2002) note that regional-scale half-lives for groundwater in the Bengal delta plain are of the order of tens of thousands of years or more. The interconnectivity of high-permeability channels depends upon the rate and frequency of both gradual and sudden channel migrations, which, in turn, depend upon alluvial sediment fluxes and topographic gradients during sedimentation. These further factors depend upon the climate-dependent (monsoon strengths, global temperature distribution) physical and chemical weathering rates of the Himalayas, eustatic sea level changes, and tectonic controls on the rates of basin subsidence.

On a local scale, complex groundwater flow patterns are further affected by a high abstraction-to-recharge excess. This over-abstraction leads not only to salt water intrusion along the coast (Dowling et al. 2003), but also to water table drawdown below villages (Harvey et al. 2002; Charlet et al. 2005), with typical annual fluctuations of groundwater levels of around 5 m in a land only a few metres above mean sea level. The combined impact of pumping and evapotranspiration is evidenced by diurnal drops in water level, particularly once monsoon-related floodings recede in December (Harvey et al. 2005). The local and seasonal increase in hydraulic gradients may lead to mixing between high- and low-arsenic waters. In the fields, intensive agri-

culture and high population density may further alter water quality through the input of organic matter (from irrigated fields or latrines), fertilizer (phosphate), or arsenic-based pesticides. Groundwater recharge is ensured by river and rainwater infiltration, as well as by the many ponds dug in villages through the surface clay layer. The recharge is especially rapid during the monsoon season, when previous drawn-down water levels rise rapidly. The balance of groundwater recharge from meteoric, oxygen-rich water and oxygen-poor, organic-rich water from ponds and irrigated fields has recently been evaluated (Metral et al. submitted).

A common feature of all these arsenic-contaminated waters is their anoxia. This anoxia may be inherited from the conditions prevailing at the time of sediment deposition, as discussed below, or may result from the surface input of organic material. As Eh decreases, the classical cascade of electron acceptors involved in the oxidation of this organic material leads to the successive appearance of Mn^{2+}, Fe^{2+}, NH_4^+, AVS (acid volatile sulfides), and methane. Mn^{2+} is seldom reported, except in the pore water of surface soil horizons (Stüben et al. 2003). Ammonium is found in Vietnam below the city of Hanoï and in Bangladesh. Close to Dhaka, a well-investigated site shows the coincidence, at a depth of 30 m, of arsenic, ammonium and methane maxima, a sulfate minimum, and high $\delta^{34}S$ values (Harvey et al. 2002; Stüben et al. 2003; Dowling et al. 2003). This coincidence is not expected from a consideration of the stepwise decrease in energy yield gained by microorganisms through the respiration of the different corresponding electron acceptors. Instead, it may be due to local groundwater flow dynamics, with a convergence of groundwater flow at a depth of 30 m, the approximate depth of abstraction from many wells in the area (Harvey et al. 2005; Klump et al. 2006).

The most common feature of all these groundwaters is their high concentration of Fe^{2+}—often close to the level required for siderite saturation—in many of the Bengali,

FIGURE 2 Tube wells in Kandal Province, Cambodia.

(A) Orange and brown coloration around the wellhead is due to the precipitation of hydrous ferric oxides, the result of rapid oxidation of dissolved Fe^{2+} upon exposure of anoxic groundwaters to the atmosphere. Such well waters have a bad taste and sometimes smell of rotten eggs (cf. hydrogen sulfide); the local population frequently declines to use such water.

(B) Alternatively, where used, similar well waters are commonly stored overnight in large pottery jars. There, a similar process results in substantive, though by no means complete, removal of groundwater arsenic, which readily sorbs onto high specific surface area, poorly crystalline hydrous iron oxides.

Cambodian, and Vietnamese shallow aquifers (Nickson et al. 2000; Berg et al. 2001; Polya et al. 2003). Since ferruginous waters have a bad taste, iron removal is often carried out, and since hydrous iron oxides are excellent scavengers of both trivalent and pentavalent arsenic (Dixit and Hering 2003), this fortuitously leads to a partial reduction of the arsenic hazard (FIG. 2). Although such arsenic removal may be done in situ by air or Fe nanoparticle injection (Kanel et al. 2005), it is most often done elsewhere, commonly overnight using household sand filters. Both Fe^{2+} and As^{3+} can also be adsorbed and oxidized by Mn oxides (Tournassat et al. 2002), or photochemically oxidized in plastic bottles in the presence of lemon juice (citric acid) and light (Hug et al. 2001). Such methods may also have application in the treatment of hazardous, arsenic-bearing surface waters (mine-contaminated lakes) and groundwaters in other areas of the world, including the People's Republic of China. Here, several hundred million people are expected to migrate from the countryside to urban and peri-urban areas sited largely on river deltas similar to those in Bengal and Southeast Asia.

ARSENIC IN SEDIMENTS OF THE GANGES, MEKONG, AND RED RIVER BASINS

Most researchers now agree that the origin of the arsenic in the groundwaters of these three deltas is geological. Furthermore, although the arsenic content of the host sediments is mostly unremarkable, there are numerous lines of evidence that point to Fe-bearing phases as the dominant hosts of mobilizable arsenic in these sediments. The higher arsenic groundwaters are typically found in shallow, young (ca. Holocene) aquifers, while much lower arsenic concentrations are generally found in deeper aquifers, e.g. below 50–100 m (BGS, DPHE 2001). The sandier parts of both sets of sediments are dominated mineralogically by quartz, feldspar, and mica. The deeper sediments were typically deposited in rivers with a high hydraulic gradient under oxic conditions during the last glacial event, when sea level was lower than now. As sea level rose throughout the Holocene, the shallower sediments were deposited in more-reducing, stagnant waters, such as are found nowadays in the coastal mangrove forest. The deeper aquifers are often characterized by orange to brown, coated sand particles, whereas the shallower aquifers tend to be grey. These readily seen color changes have been documented objectively using reflectance spectroscopy by Horneman et al. (2004), who report Fe^{2+}/Fe_{TOTAL} ratios of around 0.2 for the deeper sediments and from 0.3 to 0.9 for the shallower sediments.

Notwithstanding this, the exact mineralogical and sedimentological origin of groundwater arsenic in these systems is still a topic of intense debate. The data of Horneman et al. (2004) demonstrate that ferrihydrite, often taken as a model for As-source material, is not an adequate model for the Fe-rich coatings in these systems. Recent studies by Foster and co-workers have shown coatings on micas to be a mixed Fe-phase, and other studies have shown arsenic to be reduced at the surface of micas, either directly (by biotite) or indirectly, via co-adsorption of As^{5+} and Fe^{2+} and surface redox reaction (Charlet et al. 2002, 2005; Chakraborty et al. submitted) (FIG. 3). Iron sulfides, including "amorphous" acid volatile sulfides and framboidal pyrite (rather than arsenopyrite), are also often found, particularly in Bangladesh (Nickson et al. 2000). The arsenic associated with HCl extracts suggests the additional presence of arsenic sorbed onto mackinawite (cf. Farquhar et al. 2002; Wolthers et al. 2005) or As^{3+} substituting for carbonate in calcite (Roman-Ross submitted). The occurrence of carbonates and sulfides acting as solid-phase hosts for arsenic is supported by the sequential extraction data of Akai et al. (2004). These workers also suggest that organic matter is an important arsenic host, although it is not clear whether or not the arsenic released due to the hydrogen peroxide extraction step is associated with mineral colloidal inclusions within the reacted organic matter. Lastly, the highly reducing character of the "grey" sediments has led some workers (cf. Harvey et al. 2005) to suggest that it is not the host sediments but the overlying sediments in the vadose zone that are the immediate sedimentary source of groundwater arsenic.

ARSENIC MOBILIZATION: BIOLOGICAL VERSUS INORGANIC CONTROLS

There is a consensus that arsenic in these contaminated groundwaters is ultimately sourced from the host sediments. Various hypotheses have been invoked regarding the release mechanism. First, the oxidation of arsenic-rich pyrite was assumed to result from oxygen intrusion following water table drawdown. However, this drawdown is typically limited to 3 to 5 m, whereas maximum groundwater arsenic concentrations are found much deeper, often at a depth of 20 to 30 m. Second, the input of phosphate from irrigated fields and its competitive adsorption has been presumed to have led to the desorption of arsenic. But here, too, the mass of sorbent material present between the paddy field and the productive aquifer, as well as the rather constant phosphate concentration in the groundwaters,
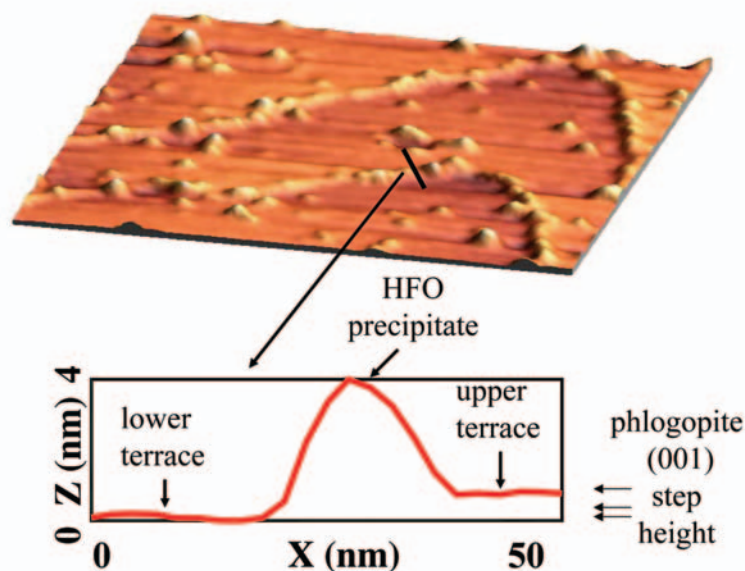
FIGURE 3 Atomic force microscope image of inferred hydrous ferric oxide (HFO) reaction products on a phlogopite (001) surface (Charlet et al. 2002). Area shown is 380 nm × 380 nm. Profile shows the oxide preferentially located at a step on the (001) surface. Iron-rich phases are thought to host the bulk of readily mobilizable arsenic in shallow, reducing aquifers in Bengal and Southeast Asia.

undermine this hypothesis. The third postulated mechanism is the microbiologically mediated reductive dissolution of arsenic-rich hydrous iron oxides (McArthur et al. 2001, 2004; Akai et al, 2004), with production of bicarbonate, which may further exacerbate the arsenic release (Appelo et al. 2002). Direct DNA-based evidence for this phenomenon has been presented for sediments from Bengal (Islam et al. 2004) and Cambodia (Rowland et al. 2004). However, there is no consensus as to the relative importance of biological and inorganic processes in controlling arsenic concentrations in these systems, or to the source of organic carbon driving these microbial processes. Both Islam et al. (2004) and Horneman et al. (2004) noted a decoupling of iron mineral dissolution and arsenic mobilization in Bengali sediments. This decoupling may be related to inorganic sorption processes, which in turn may be controlled by step decreases in redox potential or changes in the nature and specific surface area of secondary reduced iron phases (or both) (BGS, DPHE 2001; Smedley and Kinniburgh 2002).

Microbial reduction of hydrous iron oxides requires a source of degradable organic carbon. This has variously been proposed to be peat layers within the sediments (McArthur et al. 2001), other organic carbon within the sediments (McArthur et al. 2004; Gault et al. 2005), or surface-derived organic carbon (Harvey et al. 2002). Harvey et al. (2002) suggest that massive groundwater irrigation may lead to surface-derived organic matter being drawn down into the aquifer systems, thereby accelerating arsenic release. Tritium data (Aggarwal et al. 2003; Dowling et al. 2003) may be interpreted as suggesting that surface waters reach aquifer depths of 30–50 m on a time scale of decades, and Harvey et al. (2002) cite ^{14}C evidence for surface-derived organic carbon at these depths. If this model is correct, then the policy implications are profound. It suggests that, while well switching (van Geen et al. 2003) may be a practical short-term remediation option (FIG. 4), in the longer term, continued irrigation might simply exacerbate the arsenic contamination problem. There is considerable debate over whether or not this model is correct (van Geen et al. 2003). Long-term secular changes in groundwater arsenic concentration would provide some insight into the validity of this model.

TEMPORAL CHANGES IN GROUNDWATER ARSENIC CONCENTRATIONS

Groundwater abstraction has been taking place in Bangladesh since the 1940s, and there has been a massive increase in the number and usage of wells since the 1970s. However, there are no published studies of secular changes in groundwater arsenic from a single well over a period of tens of years, the time scale that ^{14}C and tritium data suggest may be required for surface-derived organic carbon to reach typical shallow aquifer depths of around 30 m (Dowling et al. 2003; Aggarwal et al. 2003; Klump et al. 2006). Therefore, our understanding of such changes relies on correlations of groundwater arsenic with age of the wells and on shorter-term monitoring studies. The results are highly equivocal.

Analysis of the BGS, DPHE (2001) dataset by McArthur et al. (2004) suggests that older wells within the shallow aquifer in Bangladesh have significantly higher arsenic concentrations than more recent wells. This lends support to the idea that arsenic concentrations are higher where pumping has been going on for a longer period of time. However, it is argued by others (Cheng et al. 2003) that there is a systematic bias inherent in the dataset because wells drilled after knowledge of the arsenic problem became widespread are more likely to be drilled into deeper, less-contaminated groundwaters. Furthermore, the much larger volumes pumped for irrigation purposes mean that the age of a well used for drinking is unlikely to be a good proxy for the time-integrated volume of groundwater abstraction in a particular area. Indeed, monitoring over a 2–3-year time period (Cheng et al. 2005) suggests, as expected for such a short time period, the opposite to be the case, with little evidence of secular increases in groundwater arsenic concentrations. Chakraborti's group at Jadavpur University, on the other hand, has found substantive secular increases in groundwater arsenic in several villages in West Bengal (Chakraborti et al. 2001). This indicates that there is a *prima facie* case for groundwater pumping being linked to enhanced groundwater arsenic levels.

Of course, much of the argument over the impact of pumping is complicated in Bangladesh because there are no data concerning groundwater arsenic concentrations prior to the development of extensive groundwater-based irrigation. For this reason, consideration of groundwater arsenic in Cambodia is particularly pertinent. Extensive groundwater development did not take place in Cambodia prior to the mid-1990s, so groundwater arsenic concentrations measured over the period between 2000 and 2004 (Polya et al. 2003, 2005) might be considered as representative of the values that were present in Bangladesh 40 years ago. Although arsenic concentrations of hundreds of ppb are not uncommon in Cambodia, the magnitude of the problem is considerably less than in present-day Bangladesh. Also, the most significant arsenic hotspots found in both Cambodia (Polya et al. 2003, 2005) and northern Vietnam (Berg et al. 2001) are immediately downstream from major cities (Phnom Penh and Hanoi), in densely populated areas. This provides tantalizingly incomplete evidence that human activity may, indeed, play a significant role in accelerating arsenic release.

CONCLUSION

It is now over 20 years since hazardous, arsenic-bearing groundwaters were discovered in West Bengal (Chakraborty and Saha 1987). This hazard has since been found to exist over much wider areas of Asia, including major river deltas in Cambodia and Vietnam. Despite impacting the lives of millions of people, no fully comprehensive solution to the problem has been implemented. Furthermore, consensus on the controls of arsenic release into these groundwaters remains to be achieved. Mineralogists and geochemists have an important role to play in answering this question. In particular, the impact of human activities on arsenic release rates is an issue that remains hotly debated and worthy of further research.

Whatever the outcome of this debate, we should recognize the temptation to over-emphasize subject-centered solutions to the problem. This is perhaps best exemplified by the attitude, in Tagore Rabindranath's novel *The Waterfall*, of Bibhuti the engineer: "My object was to make Man triumphant over the sands and water and stones… I had not time to trouble my mind about what would happen to… some wretched cultivator in some place or other." With this in mind, particularly given the difficulties in implementing comprehensive remediation of groundwaters and the recently highlighted risk of arsenic transfer from paddy fields to rice corn, further consideration might be given to the practicalities of making safe and utilizing the abundant surface-water supplies (Hossain et al. 2005).

FIGURE 4 Needle sampler and narrow-gauge casing being used in West Bengal to sample groundwater down to depths reaching several tens of metres These needle samplers (van Geen et al. 2004) obviate the need for a full-size well to be drilled; they allow sampling of both water and particulates under anoxic conditions at different depths, and thus readily enable on-site testing for groundwater arsenic at potential well sites. These samplers will be of particular use in implementing well switching as a short-term remediation strategy.

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