

A transect of groundwater and sediment properties in Araihaazar, Bangladesh: Further evidence of decoupling between As and Fe mobilization

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Abstract

A new sampling device was used to obtain 8 detailed profiles of groundwater and associated sediment properties to ~30 m depth in a 4 km² area of Bangladesh that is characterized by high spatial variability in groundwater As. Concentrations of dissolved As, Fe, and Mn ranged from <0.1 to 600 µg/L, <0.1 to 18 mg/L, and <0.1 to 4 mg/L, respectively. Voltammetric measurements conducted in the field indicate that >95% of the dissolved arsenic was As(III). The proportion of Fe(II) in the hot-leachable iron fraction of the sediment ranged from 0.2 to 0.9. The concentration of phosphate-extractable As in aquifer sands ranged from <0.1 to 1.1 mg/kg. Although the highest dissolved As concentration was measured at the site where the highest groundwater Fe and Mn concentrations were also observed, the dissolved As maximum was located several meters below the dissolved Fe and Mn maxima. Fe(II)/Fe ratios and P-extractable As concentrations in the solid phase were particularly elevated at the same site, though again the profiles of these properties did not consistently match that of groundwater As concentrations. A 500-m section composed of 5 vertical profiles confirms that whereas low As concentrations are broadly associated with low dissolved Fe and Mn concentrations, as well as low Fe(II)/Fe ratios in the solid phase, the spatial distributions of these properties in the subsurface do not follow a simple relationship. In combination with other observations from the area, the results suggest that elevated local recharge in areas where the permeability of surface soils is high prevents As from accumulating in groundwater. Conversely, dissolved As concentrations tend to be high in areas where local recharge is restricted by a surface cover of low permeability. The data demonstrate that further investigation of the mechanisms underlying As mobilization is likely to require sampling at lateral scales <1 km.

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1. Introduction

The landmark survey of groundwater pumped from thousands of tube wells in Bangladesh conducted by

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DPHE/MMD/BGS (1999) and BGS/DPHE (2001) has demonstrated that the concentrations of As in aquifers throughout Bangladesh is spatially highly variable on scales of 1–100 km. The spatial variability of As in Bangladesh groundwater has led to considerable confusion about the extent of the arsenic problem, the underlying mechanisms, and the very real need for targeting aquifers that are low in arsenic as a source of drinking water and avoiding those that are not. Even after depth and broad-scale patterns associated with the geology of the country are taken into account, the average arsenic content of wells in a given village, let alone individual wells, is difficult to predict (BGS/DPHE, 2001; Yu et al., 2003; van Geen et al., 2003a; McArthur et al., 2004; Horneman et al., 2004). This observation, and the well-known spatial variability of deltaic and floodplain deposits suggest that understanding As mobilization will require consideration of vertical (Harvey et al., 2002; Swartz et al., 2004) as well as lateral processes. The goal of this study is to show that tractable changes in aquifer properties are observed if they are mapped at spatial scales < 1 km. The implication is that processes that control As concentrations in Bangladesh aquifers should probably be studied at a comparable spatial resolution.

When spatial considerations are set aside, geochemical associations between dissolved As concentrations and other properties of either the groundwater or the sediment have not been straightforward to interpret either. There is considerable scatter in the relationship between dissolved As and Fe in Bangladesh groundwater, for instance, even for samples collected within a limited region (BGS/DPHE, 2001; Zheng et al., 2004; McArthur et al., 2004; Horneman et al., 2004). Microbial reduction of As-containing iron oxyhydroxides is most widely accepted today as a key step in the mobilization of As in groundwater, but a critical question such as the origin of the electron donors that lead to iron reduction remains a matter of conjecture (Nickson et al., 1998, 2000; BGS/DPHE, 2001; Harvey et al., 2002; van Geen et al., 2003b; McArthur et al., 2004; Islam et al., 2004). It is also not clear to what extent reduction of As(V) is a requirement for mobilization or merely a by-product of a generally reducing environment (Inskeep et al., 2002; van Geen et al., 2004a). On the basis of paired tritium–helium ages and dissolved As measurements, Stute et al. (submitted for publication) recently even proposed that the recharge rate of shallow aquifers, i.e. hydrology rather than biogeochemistry, may be the primary factor controlling groundwater As concentrations in shallow Bangladesh aquifers.

Potential relationships between As concentrations and the recharge rate of groundwater are not the focus of the present study. Instead, the issue of spatial variability at the village scale is revisited by documenting subsurface patterns with a new tool, the needle-sampler, which efficiently collects paired groundwater and sediment samples through a modification of the local drilling method (van Geen et al., 2004b). Section 2 of this paper describes the geology of the area where the needle-sampler profiles were obtained. The methods used to collect and analyze groundwater and sediment samples are described in Section 3. The main results are presented in Section 4, with an emphasis on the relationship, in reality the lack thereof, between groundwater As concentrations and other properties of the groundwater and the sediment obtained with the needle-sampler. The discussion in Section 5 starts by comparing the As data collected with the needle-sampler with the distribution of As in neighboring wells to then focus on a high-resolution transect in the context of complementary studies conducted in the same area. The section concludes with an interpretation of the spatial patterns that brings into play differences in hydrology between different parts of the study area, specifically differences in local recharge that appear to be dictated by differences in the permeability of surface soils in the region.

2. Geological setting

The profiles of groundwater and sediment properties were collected within a 25 km² region of Araihaazur upazila where the distribution of groundwater As has previously been documented by sampling and analyzing groundwater from over 6500 wells (van Geen et al., 2003a, 2005). Columbia University scientists and Bangladeshi partners have since 2000 been investigating the health effects of elevated As on a cohort of 12,000 recruited from the 70,000 inhabitants of the area (e.g. Wasserman et al., 2004). The team has also been investigating the origin of elevated groundwater As concentrations in the area and conducting an extensive mitigation program by encouraging the sharing of safe wells and installing over 50 community wells that continue to be monitored (van Geen et al., 2002, 2003c; Opar et al., in press).

Araihaazur upazila straddles the present Meghna River floodplain to the southeast and much older deposits of the uplifted Madhupur tract to the northwest (BGS/DPHE, 2001; Goodbred et al., 2003; Zheng et al., 2005). Consequently, shallow wells in the northwestern portion of the previously surveyed area reach the

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