The role of aquifer flushing on groundwater arsenic across a 35-km transect in the upper Brahmaputra River in Assam, India

R. Choudhury¹, C. Mahanta¹, M.R. Khan², B. Nath³, T. Ellis³ & A. van Geen³

¹Department of Civil Engineering, Indian Institute of Technology, Guwahati, Assam, India

²Department of Geology, University of Dhaka, Dhaka, Bangladesh

³Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY, USA

ABSTRACT: In contrast to high concentrations observed near the Brahmaputra River in Bangladesh, well testing in an upstream portion of floodplain in the Indian state of Assam has shown groundwater arsenic (As) concentrations increasing with distance from the river. To study the cause of this pattern, over 900 wells <60 m deep were tested for As and 9 sites were drilled manually over the same depth range along a 35-km transect perpendicular to the river. No relation was observed between groundwater As concentrations ranging from <0.04 to 660 $\mu\text{g L}^{-1}$ along the transect and the As <1–5 mg kg^{-1} content of recovered sand cuttings. Drilling showed a marked increase in the thickness of a clay layer capping the aquifer starting from <5 m closest to the river to >60 m at the most distant site near the base of the Naga foothills. Radiocarbon ages of 18–46 kyr indicate pre-Holocene deposition of the underlying sands across the entire transect. With the exception of two drill sites closest to the river, the cuttings were consistently grey and indication of reduced Fe oxides. Radiocarbon ages of DIC of 0.2, 4.7, and 17.8 kyr were measured in groundwater from 3 monitoring wells installed to 30–60 m depth at distances of 10, 20, and 40 km from the river, respectively. A conceptual groundwater flow model consistent with monitored heads and groundwater ages suggests that thick clay layers capping the aquifer inhibited the flushing of As towards the foothills and, as a result, maintained higher levels of As in the groundwater.

1 INTRODUCTION

Elevated arsenic (As) concentrations in well water is a major public health concern for over 100 million people across South and Southeast Asia (Smedley & Kinniburgh, 2002). Reductive dissolution of Fe(III) oxyhydroxides minerals has emerged as the leading mechanism for As release to groundwater (Bhattacharya *et al.*, 1997). Yet, several key factors including the relative importance of sedimentary and advected sources of reduced carbon for dissolution of Fe(III) oxides remain poorly understood (Datta *et al.*, 2011). This is one reason why As heterogeneity across different regions has been difficult to explain (Winkel *et al.*, 2011).

This study aims to understand the underlying processes contributing to spatial As variability along a 35 km transect in the upper Brahmaputra floodplains in India, where observed patterns demonstrates groundwater As concentrations increasing with distance from the river.

2 METHODS AND EXPERIMENTAL

2.1 Groundwater sampling and analysis

private and public wells <60 m deep straddling the districts of Golaghat and Jorhat in the upper Brahmaputra Valley bounded by the Brahmaputra River to the north, and Naga Patkai Hill ranges in the south were analyzed using a field kit between June and

August 2015, and an additional 136 wells in November 2015.

2.2 Sediment sampling and analysis

Sediment cuttings were recovered from 9 boreholes drilled along a 35-km transect perpendicular to the Brahmaputra river. At 3 of the drill sites located 10, 20, and 40 km from the river, respectively, monitoring wells screened at 45, 32, and 57 m depth were installed. Field analysis included reflectance measurement using a CM700d diffuse reflectance spectrophotometer and bulk solid phase estimation using an InnovX Delta Premium X-ray fluorescence analyzer. Field data were complemented by radiocarbon dates for organic carbon in a set of clay cuttings and dissolved inorganic carbon (DIC) in groundwater samples collected from 3 monitoring wells installed along the same transect. The various observations are then linked with a simplified 2-D cross sectional groundwater flow model that extends from the Naga foothills to the center of the Brahmaputra river.

3 RESULTS AND DISCUSSION

3.1 Groundwater and sediment data

Field kit data for a total 913 wells <60 m deep indicate that 33% of wells in the area met the WHO guideline of 10 $\mu\text{g L}^{-1}$ for As in drinking water and are mostly concentrated within 10 km of the main course

of the Brahmaputra. Another 21% contained >10 to $50 \mu\text{g L}^{-1}$, and 46% samples exceeded $50 \mu\text{g L}^{-1}$ and concentrated within 10 km of the Naga foothills. Comparison of field and laboratory data for a subset of 288 wells confirms that the kit results are by and large consistent with the ICP-MS measurements, with 94% of wells containing $<10 \mu\text{g L}^{-1}$ As and 83% of the wells with $>50 \mu\text{g L}^{-1}$ correctly classified by the kit.

Borehole lithology indicates a marked increase in thickness of a surface clay layer starting from none to <5 m at the 3 sites closest to the river to 60 m of almost continuous clay at the most distant site near the base of the Naga foothills. The reflectance data indicate particularly reduced Fe oxides ($\Delta R < 0.1\%$, Horneman *et al.*, 2004) prevail in the middle portion of the transect when compared to both the less reduced sands above the layer of brown sand at the site closest to the river and the thinner sand layers at two sites closer to the Naga Hills.

Bulk As concentrations measured by X-ray fluorescence in as many as 121 of total 147 analyzed sand cuttings were below the limit of detection of $1.4\text{--}1.8 \text{ mg kg}^{-1}$ calculated by the manufacturer's software. Concentrations of As in the remaining samples range from 1.5 to 4.6 mg kg^{-1} , without any clear pattern distinguishing drill sites close to the Brahmaputra River and the Naga Hills.

Cuttings were also analyzed for exchangeable As with the field kit. Readings were limited to 0 and $10 \mu\text{g L}^{-1}$ As for all 49 samples of sand cuttings from the 5 sites closest to the river. The corresponding range in leachable As concentrations is $0\text{--}1 \text{ mg kg}^{-1}$ without adjustment in reference chart. Out of the 9 samples of sand cuttings analyzed from the 4 sites closest to the Naga Hills instead, 3 samples gave readings of 25 and $50 \mu\text{g L}^{-1}$ corresponding to $2.5\text{--}5 \text{ mg kg}^{-1}$ leachable As.

With the exception of one shallow sample at 0.3 m bgl, uncorrected radiocarbon age of organic matter in all clay cuttings ranged from 18 to 46 kyr, indicating pre-Holocene (>12 kyr) deposition along most of the transect. Uncorrected DIC ages for groundwater span a wide range from 0.2 kyr at site closest to river to 4.7 kyr near the middle of the transect and 17.8 kyr closest the Naga Hills.

Measured groundwater levels range from 2 to 10 m below the local land surface across the transect. When referred to a constant datum, shallow groundwater elevations decrease 95 m near the hills to 80 m at the river, corresponding to a lateral head gradient of 0.3×10^{-3} .

Observed spatial pattern along the 35 km transect can only partially be explained by difference in extent of reduced Fe oxide in aquifer sediment. Redox of the sediment alone cannot account for the trend in groundwater because the most reduced sands prevail in the middle portion of the transect instead of the region closest to the Naga Hills.

3.2 Groundwater flow modeling

Among the different groundwater flow models that were tested, the best fit model that replicated field

observations was the one where a constant head boundary near the hill was assumed. The implication is that water recharges far from the transect and flows a considerable distance before reaching it. It is therefore already old when it reaches the hillside boundary of the transect and ages further while flowing towards the river. The model indicates that thickness of this flow-system depends on the horizontal hydraulic conductivity as well as the recharge rate.

4 CONCLUSIONS

Observed increase in As concentrations in shallow aquifers, with distance away from the Brahmaputra River documented in this study can be attributed to flushing of exchangeable As playing a dominant role. High surface permeability and therefore recharge and flushing of coarse sands closest to the Brahmaputra River has flushed the aquifers of their exchangeable As. In contrast, such flushing was inhibited by the thick clay capping aquifers near the Naga foothills.

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