Journal of Hydrology 518 (2014) 342–353

Contents lists available at ScienceDirect

Journal of Hydrology

journal homepage: www.elsevier.com/locate/jhydrol

Hydrologic control of temporal variability in groundwater arsenic on the Ganges floodplain of Nepal

T.H. Brikowski [a,*], A. Neku [a], S.D. Shrestha [b], L.S. Smith [c]

[a] Geosciences Dept., The University of Texas at Dallas, United States
[b] Tribhuvan University, Kathmandu, Nepal
[c] Filters for Families, Kathmandu, Nepal

ARTICLE INFO

Article history:
Available online 30 September 2013

Keywords:
Arsenic
Nepal
South Asia
Hydrology

ABSTRACT

Elevated arsenic in groundwater affects some 100 million people in South Asia, yet mitigation efforts are hindered by persistent uncertainty about the proximal source of arsenic and mechanisms for its mobilization. At the core of this uncertainty are the relative roles of surficial organic clays vs. deeper aquifer matrix iron oxyhydroxides. Temporal variations in groundwater chemistry can serve to distinguish the contributions of these two sources, and such variation is especially pronounced in headwater areas of the Ganges floodplain immediately adjacent to the Himalayan foothills (e.g. the Terai of Nepal). Tubewells down to 50 m in the Terai commonly exhibit cyclical, temporally-correlated variation in dissolved arsenic, iron and other species. In Nawalparasi, the most arsenic-affected district, these wells tap thin (2 m) gray sand aquifers embedded in a thick (>50 m) sequence of organic clays. Monsoon recharge refreshes these aquifers, temporarily minimizing arsenic concentrations. Post-monsoon, average groundwater compositions exhibit increasing trends in water–rock interaction (higher TDS, with cation exchange to form increasingly Na–HCO_3 waters), arsenic and iron. This cycle can be repeated during dry-season precipitation events as well, revealing direct correlation between trends in degree of clay interaction (sodium fraction of major cations) and arsenic concentrations. During the year, reversals in vertical head gradient yield reversals in arsenic temporal trend, and downward gradients in the dry season correlate with increases in arsenic. Collectively these observations strongly support a model of reductive mobilization of arsenic from adjacent clays into aquifers, tempered by repeated flushing during periods of appreciable rainfall.

1. Introduction

Mobilization of natural arsenic from shallow aquifers impacts the health of some 100 million people in South Asia, yet after 25 years of study, the responsible geochemical mechanisms remain controversial (e.g. Reich, 2011). A relatively recent issue is the role of surficial fine-grained sediments, which contain significantly greater matrix arsenic than intercalated coarse sediments (Dowling et al., 2002; Gurung et al., 2005), and play a crucial role as the proximal source of arsenic in some areas (Nakaya et al., 2011). Also controversial is the prevalence and significance of temporal variation in groundwater arsenic concentrations, which has been observed in settings heavily influenced by surficial clays (Polizzotto et al., 2008). This study addresses both issues simultaneously through field work in Nepal, motivated by persistent reports from the Terai (northern Ganges floodplain) of large temporal variations in groundwater arsenic from its clay-dominated surficial sediments (Suenaga et al., 2004).

The headwaters setting of the Terai offers a unique perspective on the Asian Arsenic Crisis, since the bulk of affected population, and therefore much of the scientific focus, has been in the delta regions of the Ganges River. Surficial sediments in the Terai exhibit extreme heterogeneity, and the thin aquifers can be traced directly back to their source materials in the Himalayan foothills less than 10 km away. Highly organic clays predominate in the shallow hydrologic system (the upper 50–100 m of surficial sediments contain >70% clay), and aquifer hydraulic conductivities are two orders of magnitude lower than in the delta. In the delta, high levels of labile organic carbon in fines have been shown to promote reductive mobilization of arsenic Neumann et al., 2010; Postma et al., 2007; Berg et al., 2008; Harvey et al., 2006, although not in all cases (Datta et al., 2011). Low hydraulic conductivity of surficial fines limits infiltration, which likely enhances reducing conditions and

---

[*] Corresponding author. Tel.: +1 972 883 6242; fax: +1 972 883 2537.
E-mail address: brikowi@utdallas.edu (T.H. Brikowski).

http://dx.doi.org/10.1016/j.jhydrol.2013.09.021

mobilization of arsenic (Aziz et al., 2008). In the Terai these factors combine to yield highly heterogeneous groundwater arsenic concentrations both in space and time, providing a valuable setting for exploring the arsenic mobilization process.

2. Background

The spatial and temporal distribution of elevated groundwater arsenic in Nepal are unique in South Asia. Downstream on the Ganges, arsenic-affected areas predominate in the upper delta plain (Fig. 1), while farther upstream along the central Ganges, arsenic appears to be concentrated in near-channel areas (Chakraborti et al., 2003; Kumar et al., 2010). Distributed areas of elevated arsenic are again found in headwater regions of the Ganges floodplain (i.e. the Terai), but almost exclusively between major cross-cutting (antecedent) rivers that penetrate the core of the Himalaya (Fig. 1). In the Terai districts, elevated arsenic is found exclusively in the foreland basin south of the Main Frontal Thrust (MFT), on the undisturbed floodplain (compare points on either side of the Churia Hills, Fig. 2). Surficial aquifers here are formed from material eroded from the thrust wedge (immediately north of the MFT), which is composed of earlier floodplain and later debris fan sediments exhumed by thrusting. These units comprise the Siwalik Formation (Nakayama and Ulak, 1999; Suresh et al., 2004), forming the Churia Hills, rising up to a kilometer above the Terai. Arsenic occurrences are further limited to the areas immediately downslope from exposures of the fine-grained Lower Siwalik Formation (Fig. 2 As, and Smith et al., 2004), comprised of meandering stream deposits laid down during the initial uplift of the Himalaya.

Beyond the regional heterogeneity discussed above, South Asian arsenic localities also exhibit prominent local heterogeneity over scales of hundreds of meters (McArthur et al., 2001; van Geen et al., 2003; Eiche et al., 2008). Much of this can be attributed to the effects of hydraulic heterogeneity (Aziz et al., 2008; Stute et al., 2007). In the broadest terms, shallow wells in low hydraulic conductivity clay-rich surficial deposits (Holocene in Bangladesh, especially topographic lows such as abandoned river channels, Hoque et al., 2012) tend to have elevated groundwater arsenic, while higher conductivity sands beneath these clays or where sands are present at the surface have low arsenic (DHPE, 1999; van Geen et al., 2003; van Geen et al., 2006; Nath et al., 2005). In particular, surficial hydraulic conductivity and organic content appears to affect aquifer redox state, where higher conductivity sands are flushed and oxygenated by distant or monsoonal recharge (e.g. van Geen et al., 2008). Correlation of elevated arsenic with elevated iron, phosphorous and dissolved organic carbon (DOC) and decreased sulfur is consistent with this hypothesis, where microbially-mediated redox reactions are by far the most likely mechanism for mobilization (e.g. Smedley and Kinniburgh, 2002; Mukherjee et al., 2008; Fendorf et al., 2010). These hydrologic controls can be extremely localized, as indicated by geophysical correlation of arsenic with clay abundance at the surface (Bangladesh, Versteeg et al., 2002), and more significantly at the sampling depth (Nepal, Brikowski et al., 2005), as well as sensitivity of dissolved arsenic concentrations to the degree of hydraulic connection between clay and sand layers (Eiche et al., 2008). Evidence from the Mekong Delta indicates arsenic can be leached from overlying clays into sandy aquifers below (Polizzotto et al., 2008). Kinetic barriers to arsenic release may also limit arsenic concentrations in higher-permeability settings (making flushing easier), as suggested by an observed linear relationship between arsenic content and groundwater age in Bangladesh (i.e. the degree of water–rock interaction may be a key variable, Stute et al., 2007). Observed chemical changes during groundwater irrigation also support the concept that the balance between competing influences of atmospheric oxygen, reaction with soil (clay) and time (biological processes) drive transient changes in arsenic (Polizzotto et al., 2013).

All of these observations suggest that temporal variation in groundwater arsenic should be prominent given the intensely seasonal precipitation characteristic of the monsoon climate in this region. Instead, strong temporal variation has been reported in only a few locations, associated primarily with seasonal shifts in

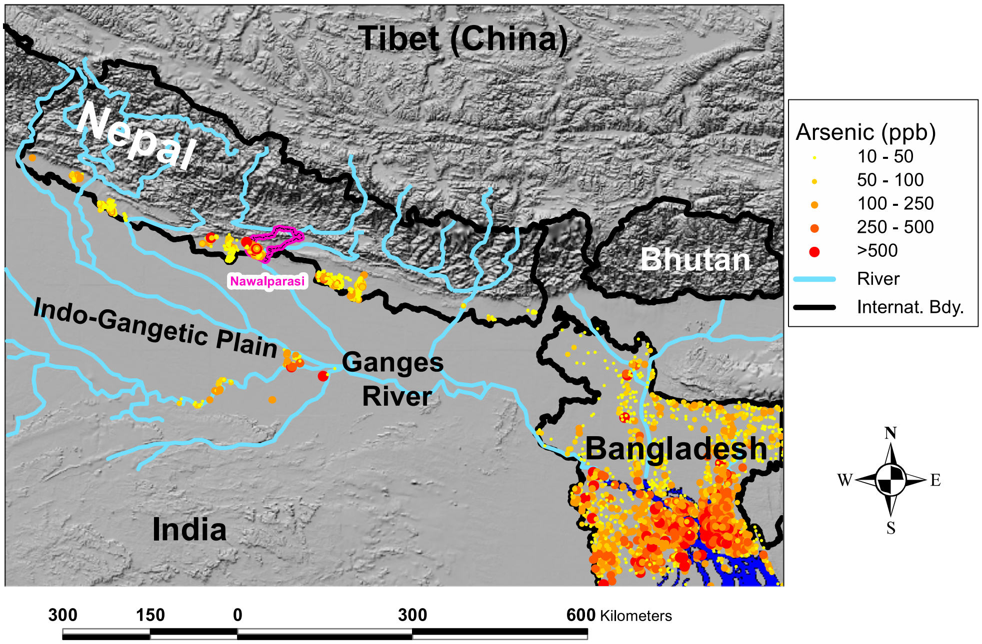

Fig. 1. Distribution of arsenic measured in tubewells, Terai, Nepal (Shrestha et al., 2004a), Bangladesh (Kinniburgh and Smedley, 2001) and India (omitting West Bengal, Shah, 2010). Values less than 10 ppb (below WHO limit) not shown. Note arsenic occurrences in Nepal cluster between transverse (antecedent) rivers. Topography shown as shaded relief, study area is in western Nawalparasi district (magenta outline). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)