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High resolution profile of inorganic aqueous geochemistry and key redox zones in an arsenic bearing aquifer in Cambodia

Laura A. Richards^{a,*}, Daniel Magnone^a, Chansopheaktra Sovann^b, Chivuth Kong^c, Sebastian Uhlemann^{d,e}, Oliver Kuras^d, Bart E. van Dongen^a, Christopher J. Ballentine^f, David A. Polya^a

^a School of Earth and Environmental Sciences and Williamson Research Centre for Molecular Environmental Science, The University of Manchester, Williamson Building, Oxford Road, Manchester M13 9PL, UK

^b Department of Environmental Science, Royal University of Phnom Penh, Phnom Penh, Cambodia

^c Faculty of Agricultural Economics and Rural Development, Royal University of Agriculture, Phnom Penh, Cambodia

^d British Geological Survey, Environmental Science Centre, Keyworth, Nottingham NG12 5GG, UK

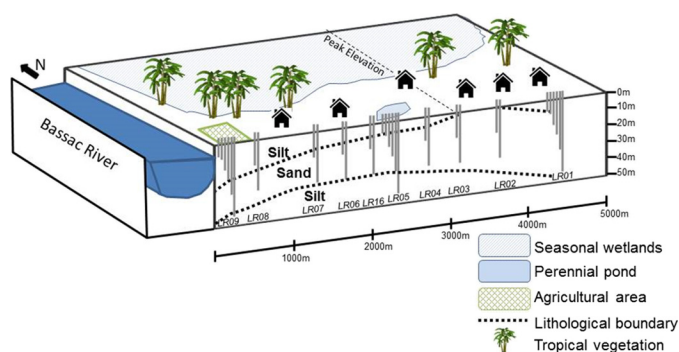
^e ETH Zurich, Institute of Geophysics, Sonneggstrasse 5, 8092 Zurich, Switzerland

^f Department of Earth Sciences, University of Oxford, South Parks Road, Oxford OX1 3AN, UK

HIGHLIGHTS

- Key changes in inorganic aqueous geochemistry monitored at high spatial resolution.
- Groundwater arsenic is highly heterogeneous and often exceeds health guidelines.
- Arsenic associated with iron, sulfate and dissolved oxygen along natural flowpaths.
- Geochemical models used to determine model redox equilibrium and mineral saturation.
- Key characterized aquifer redox zones can vary with depth, lithology, site & season.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic contamination of groundwaters in South and Southeast Asia is a major threat to public health. In order to better understand the geochemical controls on the mobility of arsenic in a heavily arsenic-affected aquifer in northern Kandal Province, Cambodia, key changes in inorganic aqueous geochemistry have been monitored at high vertical and lateral resolution along dominant groundwater flow paths along two distinct transects. The two transects are characterized by differing geochemical, hydrological and lithological conditions. Arsenic concentrations in groundwater are highly heterogeneous, and are broadly positively associated with iron and negatively associated with sulfate and dissolved oxygen. The observed correlations are generally consistent with arsenic mobilization by reductive-dissolution of iron (hydr)oxides. Key redox zones, as identified using groupings of the PHREEQC model equilibrium electron activity of major redox couples (notably ammonium/nitrite; ammonium/nitrate; nitrite/nitrate; dissolved oxygen/water) have been identified and vary with depth, site and season. Mineral saturation is also characterized. Seasonal changes in groundwater chemistry were observed in areas which were (i) sandy and of high permeability; (ii) in close proximity to rivers; and/or (iii) in close proximity to ponds. Such changes are attributed to monsoonal-driven surface-groundwater interactions and are

* Corresponding author.

E-mail address: laura.richards@manchester.ac.uk (L.A. Richards).

consistent with the separate provenance of recharge sources as identified using stable isotope mixing models.

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1. Introduction

Millions of people in South and Southeast Asia are chronically exposed to dangerous concentrations of geogenic arsenic in groundwater, at levels which exceed the World Health Organization drinking water provisional guideline of 0.13 μM ($10 \mu\text{g}\cdot\text{L}^{-1}$) (Smedley and Kinniburgh, 2002; Charlet and Polya, 2006; World Health Organization, 2011; Ravenscroft et al., 2009). The arsenic in these shallow aquifers is widely thought to be released into groundwaters following the reductive dissolution of arsenic-bearing Fe(III) minerals (Islam et al., 2004), a process which is driven by metal reducing bacteria and fuelled by organic matter providing an electron donor (Charlet and Polya, 2006; Islam et al., 2004; Postma et al., 2007; Bhattacharya et al., 1997; Rowland et al., 2009). The sub-surface location where arsenic mobilization takes place (*e.g.* near surface *versus* in-aquifer), and the subsequent controls on arsenic mobility, remains vigorously debated (Harvey et al., 2002; McArthur et al., 2011; Neumann et al., 2011; Datta et al., 2011; Lawson et al., 2013), not in the least because of the spatial heterogeneity of arsenic observed in affected aquifers, but also because of questions regarding the nature of the organic matter implicated in arsenic release (Rowland et al., 2009; Harvey et al., 2002; Lawson et al., 2013; Nickson et al., 1998; McArthur et al., 2004; Neumann et al., 2009; van Dongen et al., 2008; Rowland et al., 2007; Al Lawati et al., 2013; Al Lawati et al., 2012); the high sensitivity of arsenic mobilization to redox environments (Lee et al., 2008; Mukherjee et al., 2008; Sharif et al., 2008; Ying et al., 2015; Gulens et al., 1978; Smedley and Edmunds, 2002); and the potential impact of large-scale groundwater abstraction on future arsenic hazard (Harvey et al., 2002; Polya and Charlet, 2009).

Aqueous and sediment redox environments crucially influence the mobility and spatial variability of arsenic in groundwater (Lee et al., 2008; Mukherjee et al., 2008; Sharif et al., 2008; Ying et al., 2015; Gulens et al., 1978; Smedley and Edmunds, 2002; Cherry et al., 1979; Kocar et al., 2008; Polizzotto et al., 2008; Tufano et al., 2008). In the Mekong Delta of Cambodia, anoxic sediment conditions created by extensive monsoonal flooding cause favourable anaerobic processes such as arsenate respiration (a microbial process significantly contributing to arsenic mobilization *via* reductive dissolution of arsenic-bearing iron minerals) to occur (Charlet and Polya, 2006; Islam et al., 2004; Postma et al., 2007; Bhattacharya et al., 1997; Rowland et al., 2009; Ying et al., 2015). Cyclic seasonally induced oxidation and reduction of near-surface sediments has been proposed to lead to eventual arsenic draw-down to deeper parts of the aquifer in circum-Himalayan groundwaters (Ying et al., 2015; Kocar et al., 2008; Polizzotto et al., 2008). In-aquifer geochemistry also has a significant and complex impact on the mobility of arsenic, and delineating redox boundaries can assist in understanding arsenic speciation and mobility (Lee et al., 2008). A number of common groundwater redox classifications exist (Lee et al., 2008; Bjerg et al., 1995; Christensen et al., 2000; Chen and Liu, 2003), however these classifications are often based on a single parameter or limited set of parameters, such as dissolved oxygen (DO) and/or nitrate. These may not be applicable to other locations with different groundwater characteristics, be able to encompass seasonal changes, or be sufficient to describe the complex nature of arsenic mobilization and/or speciation. Furthermore, chemometrically defined redox zones, as defined by trends in various redox-sensitive solutes such as arsenic, methane, iron, manganese, sulfate, nitrate and ammonium, can overlap as result of overprinting of different redox conditions, leading to partial equilibrium conditions and complex interactions among electron acceptors (Mukherjee et al., 2008), as well as affecting kinetic limitations on redox processes and/

or metastable species (Balzer, 1982; O'Day et al., 2004; Borch et al., 2010).

Thus, detailed characterization of the groundwater geochemistry and interacting redox processes is essential to understanding the key controls on arsenic transport in these aquifers. The aim of this paper is to characterize the inorganic aqueous environment in a heavily arsenic-contaminated aquifer in northern Kandal Province, Cambodia, at a high spatial resolution along two distinct transects ("T-Sand" which is sand-dominated and "T-Clay" which is clay-dominated) in relatively close proximity which have differing geochemical and hydrological characteristics. The specific objectives are to: (i) develop high resolution profiles of groundwater geochemistry along groundwater flowpaths across the distinct transects T-Sand and T-Clay; (ii) identify areas of elevated arsenic occurrence and the corresponding relationship to other inorganic groundwater parameters (such as iron, nitrogen-containing compounds, DO and sulfate); (iii) identify and characterize key site-specific redox zones affecting groundwater geochemistry; and (iv) quantify the seasonal/monsoonal influence on these key redox zones.

2. Methods and materials

2.1. Field site description

The field sites are located along two distinct transects (Table 1), T-Sand and T-Clay, in the Kien Svay district of northern Kandal Province, Cambodia, south east of Phnom Penh, in an area generally well-known to be heavily affected by arsenic in its aquifers (Lawson et al., 2013;

Table 1
Description of field sites in northern Kandal Province, Cambodia along two distinct transects (T-Sand and T-Clay).

Transect	T-Sand	T-Clay
Number of sites	10	5
Wells installed (N)	32	17
Depth range (m)	6–45	6–30
Length of transect (m)	~4500	~2500
Wells sampled pre-monsoon (N)	24	11
Wells sampled post-monsoon (N)	26	12
Surface water proximity & description	Perpendicular to Bassac River, large pond near one site, heavily affected in parts by seasonal wetlands	Perpendicular to Mekong River, large pond near one site, heavily affected in parts by seasonal wetlands
Lithological description	Sand/silt dominated, near-surface sand windows in some locations	Clay cap (of variable thickness) over sandy aquifer
Previous characterization	Limited (Richards et al., 2015; Richards et al., 2017)	Well-characterized (Lawson et al., 2013; van Dongen et al., 2008; Polya and Charlet, 2009; Kocar et al., 2008; Polizzotto et al., 2008; Polya et al., 2003; Polya et al., 2005; Charlet and Polya, 2006; Tamura et al., 2007; Appelo and Postma, 1993; Benner et al., 2008; Rowland et al., 2008; Lawson et al., 2016)
Site names	LR01–LR09, LR16	LR10–LR14

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