



Tracing organic matter composition and distribution and its role on arsenic release in shallow Cambodian groundwaters

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Abstract

Biogeochemical processes that utilize dissolved organic carbon are widely thought to be responsible for the liberation of arsenic from sediments to shallow groundwater in south and southeast Asia. The accumulation of this known carcinogen to hazardously high concentrations has occurred in the primary source of drinking water in large parts of densely populated countries in this region. Both surface and sedimentary sources of organic matter have been suggested to contribute dissolved organic carbon in these aquifers. However, identification of the source of organic carbon responsible for driving arsenic release remains enigmatic and even controversial. Here, we provide the most extensive interrogation to date of the isotopic signature of ground and surface waters at a known arsenic hotspot in Cambodia. We present tritium and radiocarbon data that demonstrates that recharge through ponds and/or clay windows can transport young, surface derived organic matter into groundwater to depths of 44 m under natural flow conditions. Young organic matter dominates the dissolved organic carbon pool in groundwater that is in close proximity to these surface water sources and we suggest this is likely a regional relationship. In locations distal to surface water contact, dissolved organic carbon represents a mixture of both young surface and older sedimentary derived organic matter. Ground–surface water interaction therefore strongly influences the average dissolved organic carbon age and how this is distributed spatially across the field site. Arsenic mobilization rates appear to be controlled by the age of dissolved organic matter present in these groundwaters. Arsenic concentrations in shallow groundwaters (<20 m) increase by 1 µg/l for every year increase in dissolved organic carbon age compared to only 0.25 µg/l for every year increase in dissolved organic carbon age in deeper (>20 m) groundwaters. We suggest that, while the rate of arsenic release is greatest in shallow aquifer sediments, arsenic release also occurs in deeper aquifer sediments and as such remains an important process in controlling the spatial distribution of arsenic in the groundwaters of SE Asia. Our findings suggest that any anthropogenic activities that alter the source of groundwater recharge or the timescales over which recharge takes place may also drive changes in the natural composition of dissolved organic carbon in these groundwaters. Such changes have the potential to influence both the spatial and temporal evolution of the current groundwater arsenic hazard in this region.

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1. INTRODUCTION

The extensive contamination of groundwaters by geogenic arsenic (As) utilized by millions of people as a primary source of drinking water in South and South East Asia has long been recognised as a public health catastrophe (Chakraborty and Saha, 1987; Balga and Kaiser, 1996; Dhar et al., 1997; Smith et al., 2000; Nickson et al., 1998a; Winkel et al., 2008; Ravenscroft et al., 2009). The consumption of groundwater containing As above 10 $\mu\text{g/l}$ has already resulted in deleterious health effects in millions of people (Yu et al., 2003), with as many as 20% of deaths in arsenic impacted areas of Bangladesh attributable to arsenic (Argos et al., 2010). However, despite more than a decade of active research, several fundamental gaps remain in our understanding of the As release process that limit our ability to better predict locations and anthropogenic activities that risk As contamination. Such information is crucial for governments and those responsible for mitigating the effects of the current crisis and for predicting the potential for secular changes in the As hazard in the future.

The primary source of As in the groundwaters of this region is the rapid weathering of As-bearing rocks in upper Himalayan catchments. This As is then transported by the large rivers that drain these catchments, and deposited in the low-lying young alluvial floodplains of the region. Arsenic is initially sorbed to secondary iron (oxy)hydroxide mineral phases that coat sedimentary grains under aerobic conditions, but is released to solution following the reductive dissolution of these As-bearing phases (Bhattacharya et al., 1997; Nickson et al., 1998b; McArthur et al., 2001; Dowling et al., 2002; Stuben et al., 2003; Horneman et al., 2004; Zheng et al., 2004; Charlet and Polya, 2006). Iron-reducing bacteria catalyse this release of As to solution, with organic matter (OM) critically implicated as an electron donor in this process (Islam et al., 2004; van Geen et al., 2004). However, whilst the biogeochemical processes that are responsible for liberating As to groundwater are now widely understood, identification of the source of OM that drives these processes remains unresolved, elusive and even controversial (Fendorf et al., 2010; Lawson et al., 2013; Neumann et al., 2014).

Several studies have suggested that a sedimentary origin of OM is responsible for driving microbially mediated As release processes; with buried peat (McArthur et al., 2001), aerated root zones and animal burrows (Meharg et al., 2006), or thermally mature hydrocarbons that have migrated from depth (Rowland et al., 2006, 2007, 2008; van Dongen et al., 2008; Al Lawati et al., 2012) amongst suggested potential sources. In contrast, surface derived OM may be utilized in the biogeochemical processes that liberate As, with recharge from ponds, wetlands and through geomorphic features such as abandoned floodplains, meander channel deposits, and scroll bars introducing young dissolved organic carbon (DOC) to shallow groundwaters (Kocar et al., 2008; Papacostas et al., 2008; Polizzotto et al., 2008; Neumann et al., 2010; Lawson et al., 2013). The introduction of this surface derived OM has also been suggested to be exacerbated by the extensive pumping of groundwaters in this region, resulting in the

draw down of increased volumes of organic rich recharge waters (Harvey et al., 2002). This particular model is intensely debated (Aggarwal et al., 2003; Harvey et al., 2003; van Geen et al., 2003; Lawson et al., 2008, 2013; Sengupta et al., 2008; McArthur et al., 2011; Neumann et al., 2010; Mailloux et al., 2013), not least because of the implications it holds for future agricultural production in these densely populated countries reliant on rice as a staple food source.

Given the considerable debate surrounding the origin of OM that is responsible for driving As release, the identification and quantification of the different sources of OM present within these aquifer systems is of great importance. This is of particular significance when one considers that it is not necessarily the amount, but the type and reactivity of OM that are the key controls on As release to groundwater (Gault et al., 2005; Rowland et al., 2007; van Dongen et al., 2008; Mladenov et al., 2010). Indeed, it has been suggested that utilisation of different sources of OM will give rise to different rates of As release to groundwaters (Fendorf et al., 2010). Older sources of OM, such as particulate organic carbon (POC) present in buried peat deposits, are likely to have experienced a long history of involvement and consumption in metabolic processes that began immediately following deposition. It is assumed that such metabolic processing may give rise to a less reactive residual OM that may support a slow rate of As release into the groundwater. In contrast, young surface-derived OM that is introduced into the subsurface has had less time to be utilized in metabolic processes, and as such may provide a higher reactivity source of OM that may support a more rapid rate of As release. Distinguishing between the sources of OM present in any groundwater system, and assessing the vulnerability of the system to changes with respect to its dominant OM source, is therefore of critical importance in predicting future spatial and temporal changes in the current As hazard.

In this study we interrogate the isotopic signature of ground and surface waters at a known As hotspot in Cambodia to (i) determine the residence times of groundwater prior to the onset of extensive groundwater pumping, and (ii) characterize and quantify contributions from different sources of dissolved organic carbon present in this aquifer. The chemical composition of the groundwaters at this site has been well characterized in previous studies (Polizzotto et al., 2008; Lawson et al., 2013) and as such will not be discussed in detail here. This study focuses on the radiogenic and stable isotopes of hydrogen (tritium) and carbon ($\delta^{13}\text{C}$, ^{14}C) to determine groundwater residence times and provenance OM present in the aqueous phase. In addition, the stable isotopes of oxygen ($\delta^{18}\text{O}$) and hydrogen (δD) provide insights into the mode and extent of interaction between ground and surface waters at different locations within the field area. This characterization of pre-development conditions allow for a robust evaluation of the relative contributions of in-aquifer and surface derived OM where minimal groundwater-pumping-driven draw-down of surface waters prevail. We consider bulk DOC age as a proxy for the residual reactivity of organic matter because it provides an opportunity to account for the time-integrated impact of the reaction history and consumption

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