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Size-fractionation of groundwater arsenic in alluvial aquifers of West Bengal, India: The role of organic and inorganic colloids



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HIGHLIGHTS

• We study the role of colloidal particles on groundwater arsenic in West Bengal (India).

- · Arsenic concentration decreases with sequential micro- and ultra-filtration steps.
- Majority of As_{T} remain in the 'truly dissolved' phase (i.e., pass through 0.05 μm).
- Larger Fe-rich inorganic colloids (>0.05 μm) scavenge As(III).
- Smaller organic/organometallic colloids (<0.05 µm) scavenge As(V).

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ABSTRACT

Dissolved organic carbon (DOC) and Fe mineral phases are known to influence the mobility of arsenic (As) in groundwater. Arsenic can be associated with colloidal particles containing organic matter and Fe. Currently, no data is available on the dissolved phase/colloidal association of As in groundwater of alluvial aquifers in West Bengal, India. This study investigated the fractional distribution of As (and other metals/metalloids) among the particulate, colloidal and dissolved phases in groundwater to decipher controlling behavior of organic and inorganic colloids on As mobility. The result shows that 83-94% of As remained in the 'truly dissolved' phases (i.e., <0.05 µm size). Strong positive correlation between Fe and As (r² between 0.65 and 0.94) is mainly observed in the larger (i.e., >0.05 µm size) colloidal particles, which indicates the close association of As with larger Ferich inorganic colloids. In smaller (i.e., <0.05 µm size) colloidal particles strong positive correlation is observed between As and DOC ($r^2 = 0.85$), which highlights the close association of As with smaller organic colloids. As(III) is mainly associated with larger inorganic colloids, whereas, As(V) is associated with smaller organic/ organometallic colloids. Scanning Electron Microscopy and Energy Dispersive X-ray spectroscopy confirm the association of As with DOC and Fe mineral phases suggesting the formation of dissolved organo-Fe complexes and colloidal organo-Fe oxide phases. Attenuated total reflectance-Fourier transform infrared spectroscopy further confirms the formation of As-Fe-NOM organometallic colloids, however, a detailed study of these types of colloids in natural waters is necessary to underpin their controlling behavior.

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1. Introduction

Naturally occurring carcinogenic arsenic (As) is ubiquitous in the environment and commonly observed in high concentrations in the alluvial aquifers of south-east Asia (Bhattacharya et al., 1997; Nickson et al., 1998; Chatterjee et al., 2003; Charlet and Polya, 2006; Mukherjee et al., 2008; Nath et al., 2011). In south-east Asia, the use of As-

0048-9697/\$ - see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.scitotenv.2013.08.087 enriched groundwater as the primary source of drinking and other household activities has led to an unprecedented human health problem, creating a major concern for the environmental and public health experts (e.g., Smedley and Kinniburgh, 2002). The human health problem is particularly alarming in the rural Bengal Delta Plain (BDP), i.e., in West Bengal (India) and Bangladesh, yet there are no sustainable alternatives of drinking water sources available (Mukherjee et al., 2011).

The role of dissolved organic carbon and/or Fe-oxyhydroxide solids on As mobility in natural groundwater was described in detail (e.g., Charlet and Polya, 2006). For example, dissolved organic carbon (DOC) or natural organic matter (NOM) can compete with As for

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sorption sites on mineral surfaces and influence As redox processes (Redman et al., 2002; Dixit and Hering, 2003; Bauer and Blodau, 2006; Buschmann et al., 2006). Arsenic can sorbed onto organic matter (especially humic components) through covalent bonding (Buschmann et al., 2006). On the other hand Fe-oxyhydroxide solids bind As more efficiently in the presence of organic matter (Ritter et al., 2006). However, the inorganic As species [i.e., As(III) and As(V)] react differently in the presence of organic matter depending on As/DOC ratios (Buschmann et al., 2006).

The competition between As and NOM for the sorption sites can notably influence As mobility in groundwater (Redman et al., 2002; Warwick et al., 2005; Bauer and Blodau, 2006; Ritter et al., 2006). The formation of As-NOM complexes was reported to change the course of As redox reactions in aqueous phase (Thanabalasingam and Pickering, 1986; Lin et al., 2004; Wang and Mulligan, 2006; Buschmann et al., 2006). These complexes mostly belong to the colloidal size fraction ranging between 2 and 300 nm or higher (Mueller, 1996; Sharma et al., 2010). Sharma et al. (2010) demonstrated that 94% of As is associated with colloidal Fe-NOM fractions through a laboratory investigation. Astrom and Corin (2000) have reported that As can be scavenged by colloidal Fe and dissolved organic matter (DOM) in river waters through a field-based study. In addition to that both inorganic and organic colloids can control the fate of trace metals in aqueous medium due to adsorption (Pokrovsky and Schott, 2002; Pourret et al., 2007).

The widely practiced field method for groundwater sampling is the use of 0.45 μ m filters followed by preservation with a few drop of HNO₃ (e.g., Gault et al., 2005). This procedure is normally followed to avoid interference from particles in the groundwater during laboratory determination of As using sophisticated spectroscopic instruments (e.g., Gault et al., 2005). Therefore, the uncertainty in determination of total As concentration in groundwater remains as the particulate As is not considered. However, some reports are available in the literature which considered the determination of As concentrations in particulate, colloidal and dissolved phases in groundwater (Hinkle and Polette, 1999; Guo et al., 2011). The rural people do not use any filters normally (e.g., 0.45 μ m) and drink the water directly from their household or nearby wells. Thus the ritual of not considering the particulate phase As, during field sampling/monitoring and subsequent determination, may not give a proper picture while proposing the mitigation options.

There is a very little information available in the literature on the possible formation of As-bearing colloids in groundwater (Bauer and Blodau, 2009; Sharma et al., 2010). Guo et al. (2011) recently reported the role of dissolved or colloidal DOC and Fe phases on As mobility in groundwater of Hetao Basin in Inner Mongolia. Furthermore, there is no data available on the colloidal distribution of As in different size fractions in the alluvial aquifers of West Bengal, the major drinking water source to millions of inhabitants. Therefore, size-fractionation of groundwater As will provide some useful information on the overall pattern of particulate, colloidal and dissolved phase presence of As and to ascertain its safe use in rural areas of BDP in India and Bangladesh as well as in similar geological settings worldwide.

This study aims to elucidate the controls of dissolved and colloidal (organic and inorganic) particles on groundwater As mobility in an As-contaminated alluvial aquifer in West Bengal, India. The ground-water samples collected from the field were characterized to investigate the fractional distribution of As (and other metals/metalloids) in different size fractions (i.e., particulate, colloidal and dissolved phases) and subsequently assessed the factors responsible for As mobility.

2. Materials and methods

2.1. Study area

The study area (latitude 23°00′20″–23°05′20″N; longitude 88°31′40″– 88°49′00″E) is located within the active flood plains of river Hooghly (a distributory of river Ganges) and lies within the BDP in Chakdaha block, Nadia district, West Bengal, India (Fig. 1). The BDP forms an integral part of the Ganga–Brahmaputra–Meghna river system, the world's largest deltaic alluvial deposit (Charlet et al., 2007). Mukherjee et al. (2009) have provided a good account of the local geology and geomorphology. This includes large flood plains (both newer and older alluvium), spreading over extensive areas, and formed by successions of a fining upward sequence containing finer and coarser grained sandy and clayey alluvial deposits. The flood plains are extensively used for agricultural purposes throughout the year, which include numerous large and small natural and/or manmade ponds, characterized geomorphologically as oxbow lakes, abandoned channels and meander scars (Nath et al., 2005; Charlet et al., 2007; Bhowmick et al., 2012).



Fig. 1. Map of the study area highlighting the sampling points as yellow open rectangle with As concentrations (µg/L, unfiltered samples) indicated inside the symbol (inset C). Inset a) shows map of India where state West Bengal is marked, b) shows study area within state West Bengal with an arrow head. Satellite imagery adapted from Google Earth show fluvial geomorphology of the studied region.

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