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Influence of monsoonal recharge on arsenic and dissolved organic matter in the Holocene and Pleistocene aquifers of the Bengal Basin

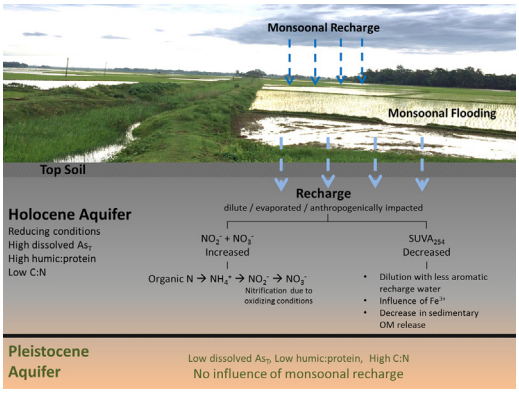
Harshad V. Kulkarni [a,*], Natalie Mladenov [b], Saugata Datta [c], Debashis Chatterjee [d]

[a] 2118 Fiedler Hall, Department of Civil Engineering, Kansas State University, Manhattan, KS, USA
[b] Department of Civil, Construction and Environmental Engineering, San Diego State University, San Diego, CA, USA
[c] Department of Geology, Kansas State University, Manhattan, KS, USA
[d] Department of Chemistry, University of Kalyani, Kalyani, Nadia, West Bengal, India

HIGHLIGHTS

- Shallow groundwater (<40 m) showed high As_T and humic:protein and low C:N.
- Monsoonal recharge influenced groundwater chemistry at shallow (<40 m) aquifer.
- Increases in NO_2^- and NO_3^- occurred in shallow groundwater due to monsoonal recharge.
- Monsoonal recharge resulted in a two-fold decrease in $SUVA_{254}$ in shallow groundwater.
- Humic-like DOM character coincided with isolated As hotspots in the deep groundwater.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As) mobilization in the Bengal Basin aquifers has been studied for several decades due to the complex redox bio-geochemistry, dynamic hydrogeology and complex nature of dissolved organic matter (DOM). Earlier studies have examined the changes in groundwater As in the dry season before monsoon and during the wet season after monsoonal recharge. To investigate the more immediate influence of recharge during the active monsoon period on As mobilization and DOM character, groundwater samples were analyzed in the pre-monsoon and during the active monsoon period. Groundwater samples were collected from shallow (<40 m) and deep (>40 m) tube-wells in West Bengal, India. Dissolved As_T in shallow groundwater ranged from 50 to 315 μg/L exceeding the WHO guideline of 10 μg/L. Shallow groundwater also showed high total dissolved nitrogen, carbon to nitrogen (C:N) <1, and humic-like DOM with a humic:protein ratio >1. By contrast, deep groundwaters contained As_T between 0.5 and 11 μg/L with carbonaceous and protein-like DOM, C:N >1, and humic:protein <1. Stable isotopes of $\delta^{18}O$ and $\delta^{2}H$ and Cl/Br results indicated three recharge scenarios in the shallow aquifer including direct recharge of dilute rainwater, evaporated surface water, and anthropogenically impacted surface water. Monsoonal recharge did not cause notable changes in As_T in deep or shallow groundwater, including two As hotspots in the Pleistocene aquifer. However, the monsoon did result in a two-fold decrease in $SUVA_{254}$, increase in nitrite and nitrate in the shallow groundwater. The DOM in the deep groundwater at the two As hotspots (with As_T 132 and 715 μg/L) had optical properties with much greater humic-like DOM than the surrounding groundwater, which had low As_T and highly protein-like DOM. Overall, these results support that protein-like DOM associated

* Corresponding author.
E-mail address: harshad.env@gmail.com (H.V. Kulkarni).

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with low groundwater As concentrations and suggest that the monsoonal influence on nitrate and nitrite is limited to shallow aquifers.

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1. Introduction

Monsoonal precipitation and flooding occurs in South Asia from July to October, bringing over 1000 mm of rainfall to the area. For the Bengal Basin, where 90% of the total annual precipitation (1200 to 3945 mm) occurs during the four months (July–October) of wet season (Nath et al., 2008) and 15% of the total precipitation contributes towards the aquifer recharge (Mukherjee et al., 2007a), monsoonal precipitation is a major source of groundwater recharge.

In this hydrologically dynamic landscape, groundwater is the main drinking water source for millions of people. Naturally elevated dissolved arsenic (As) concentrations in groundwater in reducing aquifers of the Bengal Basin have put millions of people at severe health risk. Nearly 20 million people in Bangladesh and over 4 million in West Bengal, India have been estimated to rely on As contaminated (>10 μg/L) groundwater as a major source of drinking water (Chakraborti et al., 2009; Fendorf et al., 2010; Flanagan et al., 2012). Geochemical evidence has indicated that shallow (<40 m) groundwater in the Holocene aquifer composed of younger sediments deposited ~7000 years before present (y BP) has higher dissolved As concentrations (Acharyya et al., 2000; Mukherjee et al., 2007a; Neidhardt et al., 2013). In contrast, the Pleistocene aquifer (40–150 m deep), which is composed of older sediments deposited 12,300–48,600 y BP, contains groundwater with low dissolved As (Acharyya et al., 2000; Moran et al., 2000; Eiche et al., 2008; Burgess et al., 2010; Fendorf et al., 2010; Datta et al., 2011; Hoque et al., 2011; Knappett et al., 2016). The shallow Holocene aquifer in the Bengal Basin is recharged by various sources including precipitation, irrigation return flow and seepage from surface water bodies or losing streams (Ravenscroft et al., 2005).

Rapid monsoonal recharge during the wet season (July–October) in the Bengal Basin (Bangladesh) can elevate the groundwater levels in the shallow aquifer (20–30 m) by up to 5 m more than in the dry season (November–June) when the monsoon ceases and irrigation pumping resumes (Harvey et al., 2005). Several studies have investigated the influence of monsoonal recharge on dissolved As concentrations in shallow aquifers in the Bengal Basin. A three-year time series study in Bangladesh found no significant seasonal variation in total dissolved arsenic (As_T) concentrations in the shallow aquifer; however a few isolated wells showed a systematic long-term decline in dissolved As_T concentrations (Dhar et al., 2008). Farooq et al. (2010) reported that only 14% of the sampled wells in Murshidabad (West Bengal) showed decrease in post-monsoon dissolved As_T concentrations and attributed this variation to the dilution by As free rain or surface water. Sankar et al. (2014) reported no significant change in dissolved As_T concentrations between pre- and post-monsoon (December–January 2010, 2011 and 2012) samples from Murshidabad (West Bengal). Planer-Friedrich et al. (2012) observed that over 12-months of sampling from the Holocene aquifer in Bangladesh, the dissolved As_T concentrations increased 1.5 times from February (dry period) to August (wet period) and coincided with an increase in the water table. In another study, wet season influences on dissolved As in a shallow aquifer of the Yangtze River Basin in China were reported to have a more complex temporal pattern, showing a temporary decrease in dissolved As_T concentration at the beginning of the precipitation period (May) due to vertical mixing followed by an increase in dissolved As_T during the rest of the wet season (Schaefer et al., 2016). Previous studies collected groundwater samples in the pre-monsoon (dry) and post-monsoon (wet) seasons, but much remains to be understood about the changes underway during the active monsoon period. In the geological setting of the Bengal Basin, it has been shown that the monsoonal precipitation rapidly recharges the shallow aquifer (Mukherjee et al., 2007b). Therefore, the insights into the effects of recharge on As mobilization during the transition from dry to wet season may advance our understanding of hydrological processes on As mobilization.

Arsenic mobilization in younger Holocene aquifers in the Bengal Basin is known to be driven mainly by microbial reductive dissolution of ferric oxy(hydroxide) rich sediments (McArthur et al., 2001; Dowling, 2002; Hasan et al., 2007). Labile organic matter originating from aquifer sediments (McArthur et al., 2004; Sengupta et al., 2008; Datta et al., 2011; Neumann et al., 2014) or drawn to the depth from surface water sources (Harvey et al., 2006; Neumann et al., 2010; Lawson et al., 2016) serves as the energy source for microbial reductive dissolution. Additionally, biologically recalcitrant dissolved organic matter (DOM), such as fulvic acid, has been shown to enhance As mobilization by serving as an intermediate electron shuttle (Kulkarni et al., 2018), forming aqueous complexes with Fe and As (Sharma et al., 2010; Liu et al., 2011), and competing with As for sorption sites (Bauer and Blodau, 2006). Recent studies have documented the predominantly humic-like character of DOM in shallow high As groundwater in Bangladesh (Mladenov et al., 2010, 2015) and West Bengal, India (Kulkarni et al., 2017; Vega et al., 2017). However, less is known about the influence of aquifer recharge during monsoon on the quality of groundwater DOM as it relates to As mobilization.

Previous efforts to track temporal changes in dissolved organic carbon (DOC) concentration in groundwater have also shown contrasting effects of monsoonal recharge. Majumder et al. (2016) observed that the DOC concentration increased during the post-monsoon season in Nadia district in West Bengal (India) and was related to increased reduction of As (V) to As (III). In contrast, Planer-Friedrich et al. (2012) reported an inverse seasonal trend between DOC and dissolved As_T, as the lowest DOC concentration coincided with the highest As_T concentration. However, such seasonal changes in the DOM properties have not been previously investigated, especially during the active monsoon period.

In terms of spatial variability in DOM chemical quality, the deeper groundwater in Pleistocene aquifers that is generally considered to be free from As contains substantially low DOC concentrations (Sutton et al., 2009; Datta et al., 2011; Sankar et al., 2014) that limit the release of As from these sediments (Harvey et al., 2002; Postma et al., 2007; Berg et al., 2008; Polizzotto et al., 2008; Fendorf et al., 2010). Heterogeneous occurrences of elevated As levels in deeper groundwater in the Pleistocene aquifer have been reported, although not very commonly (Burgess et al., 2010; Erban et al., 2013) and often are attributed to drawdown of As rich shallow groundwater to greater depths (Neidhardt et al., 2013). It has been suggested that the presence of discontinuous clay lenses with higher organic carbon content within the deep aquifer providing labile DOM for microbial metabolism (Mukherjee et al., 2007a, 2007b; Schaefer et al., 2016) may be responsible for such heterogeneous elevated As levels. However, the chemical quality of DOM associated with As hotspots in the Pleistocene aquifer still has not been investigated.

In this study, we collected and analyzed shallow (high As, Holocene aquifer, Nadia district) and deep (low As, Pleistocene aquifer, Hooghly district) groundwater samples from drinking water tube wells. Each tube well was sampled once during the dry season (May–June 2015) and once during the monsoon (July–August 2015). Analyses of groundwater chemistry and optical properties of DOM were conducted in shallow and deep groundwater samples to investigate the influence of monsoonal recharge on As biogeochemistry in the Bengal Basin.

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