



Reductive dissolution of iron-oxyhydroxides directs groundwater arsenic mobilization in the upstream of Ganges River basin, Nepal



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ABSTRACT

Mobilization of arsenic (As) in groundwater has become a focus of attention in recent years. A few years back, groundwater As is found as a geogenic contaminant in the upper stream Ganges River basin immediately adjoining to the Himalayan foothills (the Terai Region of Nepal). The source is said to be leached due to weathering of As-bearing minerals. In this study, geologic, hydrogeologic, and geochemical data were used to characterize the As source and the geochemical process controlling As-mobilization in the groundwater aquifers of Terai Region. The findings suggest that the reductive dissolution of iron-oxyhydroxide is the main mechanism of As-mobilization in Terai aquifers. This mechanism is supported by a number of evidences including, 1) presence of arsenolite and scorodite As-bearing minerals; 2) presence of high concentration of As, Fe and low concentration of SO_4^{2-} and NO_3^- ; 3) presence of strong linkage between As and HCO_3^- and negative correlation between As and oxidation reduction potential; 4) presence of abundant percentage of Fe and Mn-oxides in the sediment samples; 5) presence of similar depth profile trend of As with those of Fe and Mn-oxides; 6) presence of Fe, O and Mn confirmed by energy dispersive X-ray spectroscopy analysis; and 6) presence of predominant form of As adsorbed on organic matter and Fe-oxyhydroxide phases of sequential leaching.

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1. Introduction

Groundwater is a significant source of drinking water in many parts of the world (Jousma and Roelofsen, 2004). Historically, groundwater supplies are thought to be safer in terms of pathogenic microbes than water from open dug wells and surface water bodies like rivers, streams, lakes, and ponds (World Bank, 2005). This is because of the natural filtering ability of the subsurface water and the distance a microbe would have to travel in order to reach the groundwater source. However, groundwater is more prone to chemical and other such contaminants, derived either from natural sources or anthropogenic activities (EPA, 1993). Arsenic (As) contamination in groundwater is one of several chemical contaminants. The most important route of exposure of As in human is due to drinking of As-contaminated groundwater. Today, majority of the world's chronic As-related health problems are because of consumption of As-contaminated groundwater (WHO, 2009).

Groundwater extracted from majority of As-affected region showed that high concentrations of As are associated with young alluvial sediments. Elevated level of As is also characterized by high dissolved iron (Fe) together with low nitrate (NO_3^-) and sulfate (SO_4^{2-}) under reducing conditions of groundwater (Nordstrom, 2002; Smedley et al.,

2003). Water–rock interactions and favorable physical and geochemical conditions (by mobilization and accumulation of As bearing inorganic compounds) in groundwater are the principal reason behind substantial concentration of As. As is quite mobile at pH values of 6.5–8.5, mostly found in groundwater under both oxidizing (aerobic) and reducing (anaerobic) conditions.

Naturally, As content in soil generally ranges below 10 ppm; however, it can cause major chaos once it gets into groundwater. Historically, it was believed that As is released in the soil as a result of weathering of arsenopyrite or other primary sulfide minerals (Ahuja, 2008). However, at present, it is well-known that As-contamination of groundwater is a by-product of several biogeochemical process such as oxidative dissolution of As-rich pyrite minerals (Chowdhury et al., 1999; Das et al., 1996), reductive dissolution of Fe(III) oxide/oxyhydroxides and the consequent release of sorbed and/or co-precipitated As (Chauhan et al., 2009; Dowling et al., 2002; Guo et al., 2008, 2011; Harvey et al., 2002; Larsen et al., 2007; McArthur et al., 2001; Mladenov et al., 2010; Pedersen et al., 2006; Reza et al., 2010a,b; Smedley and Kinniburgh, 2002; van Geen et al., 2006), displacement of adsorbed As by phosphate by the agricultural application of fertilizers (Acharyya et al., 1999), by carbonate (Appelo, 2002) produced through microbial metabolism (Harvey et al., 2002), or by changes in the sorptive capacity of ferric oxyhydroxides (Smedley and Kinniburgh, 2002), and anthropogenic activities and combustion of fossil fuels via precipitation from the atmosphere (Ferguson and Gavis, 1972) that metabolize and release As in

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groundwater. Because of complication in understanding of these biogeochemical processes, the identification of mechanisms responsible for a particular case of As contamination is a difficult task. For example, there has been considerable controversy over the cause of As-contamination of groundwater in Bangladesh. Some researchers have proposed sulfide oxidation as the predominant mechanism for As-release (Bagla and Kaiser, 1996; Dipankar et al., 1994; Mandal et al., 1996) while others contend that the reduction of Fe-oxyhydroxides is the cause of high dissolved As-concentrations in groundwater (Bhattacharya et al., 1997; McArthur et al., 2001; Mladenov et al., 2010; Pedersen et al., 2006; Reza et al., 2010a, b; Reza et al., 2011; van Geen et al., 2006).

The Terai region is placed in the bottoms of the Himalayas. It is the source of sediment for several Indian River located in North India. Terai Region inhabits about 47% of the country's population (Rahman et al., 2006). The unconfined quaternary aquifers, which are on the order of 300m thick (Khadka, 1993), are tapped by around 400000 (Panthi et al., 2006) to 800000 wells (Smedley, 2005). They supply water to 11 million people, or about 90% of the residents of Terai (Rahman et al., 2006). Elevated As level was first discovered in 1999 in the Jhapa, Morang, and Sunsari districts of eastern Terai region of Nepal, which border the Indian state of West Bengal. About 8% of the 268 groundwater samples had As > 10 ppb with two samples exceeding 50 ppb (Panthi et al., 2006). Subsequent surveys of groundwater quality in Terai have revealed the presence of As in some tube wells with depths < 50 m. However, most of the samples had < 10 ppb of As. It is likely to note that Nawalparasi district is the most As-hit among 20 As-contaminated Terai districts of Nepal (Gurung et al., 2005; Thakur et al., 2011; Yadav et al., 2011, 2012). Nearly, 50% of the country's rural populations inhabiting in Terai region rely on groundwater for its drinking water supply and are prone to As exposure (NASC/UNICEF, 2008; Yadav et al., 2012, 2014). The biogeochemical processes mainly responsible for As-release and its mobilization are not well understood in Terai aquifers of Nepal. Therefore, the present study aims to identify the different geochemical processes responsible for As release in groundwater of upstream Ganges river basin.

1.1. Geological setting of Terai Region

Tectonically, Nepal can be classified into four different zones from north to south followed by Terai Plain bordering them on the south: a) Tibetan (Tethys) Himalayas, b) Higher Himalayas, c) Lesser Himalayas, d) Sub-Himalayas or Siwalik group and, e) Terai Plain.

The Tethysian zone which lies in the northern part of western central Nepal consists of high mountain ranges crowned by cretaceous fossiliferous sediments. The higher Himalayan zone consists of crystalline rocks (schist and gneiss). Main control thrust (MCT) marks the contact of crystalline zone to that of lesser Himalayan sedimentary pile. The contact of crystalline with overlying Tethys sediments is gradational. To the south of higher Himalayas, the lesser Himalaya zones lie which consists of meta-sediments with different grades of metamorphism. The sub-Himalayan zone, which lies to south of the lesser Himalayas and separated by main boundary thrust (MBT), consists of sedimentary rocks containing silt, sand, shale and pebble and boulder beds. The pebbles and boulders are quartzite, sandstone, phyllites etc. These formations form the low lying hill ranges that rise from the Indo-Gangetic plain on the south. These hills are called Churia hills and are the Nepalese designation of Siwalik. The Terai plain lies in the southern part of Nepal and is separated from the Churia formations by the Himalayan Frontal thrust. It is the continuation of Indo-Gangetic trough. It gently slopes towards south and covered by recent and older alluvium having the thickness up to 1500 m. The recent alluvium is deposited by the rivers coming from the mountains and hills and hence the different Himalayan ranges lying north of the Terai plain are its provenance.

Geomorphologically, Terai plain can also be divided into two parallel zones i.e. Bhabhar zone and Gangetic alluvial plain (GAP) (Sharma, 1990). The Bhabhar zone lies along the foot of Churia hills. It is composed of boulders, cobbles, pebbles, gravels and sand which are extremely poorly sorted. GAP lies to south of the Bhabhar zone. The Gangetic alluvium underlay, interfinger with and boarder the Bhabhar zone deposits. The sediments in this part consist of silt, clay, sand, gravel and pebble beds intercalating and sometimes intermixing with each other. The sediments are generally finer towards South.

1.2. Study area

Nawalparasi district, a part of Lumbini zone (one of the 75 districts of Nepal), is located in the western Terai region of Nepal. It lies 147 km. west of the capital city Kathmandu, and surrounded by Chitwan district in the east, Tanahun district in north, Palpa and Rupandehi districts in the west, and the Indian state of Uttar Pradesh in the south. The present study was conducted in three villages (ThuloKunwar, Kasia and Panchgawa) in the eastern Parasi Bazar of Nawalparasi district (Fig. 1) where several cases of As-poisoning in groundwater have been documented (Gurung et al., 2005; Thakur et al., 2011; Yadav et al., 2011; 2012; 2014).

2. Materials and methods

Geological, hydrogeological, and geochemical data are utilized to identify the source of As-contamination and the principal geochemical processes responsible for As-mobilization in the upstream of Ganges river basin.

2.1. Sample collection and analysis

2.1.1. Water samples

A total of 24 groundwater samples were collected from selected tubewells (depth 8–50 m) in 2010. Groundwater samples (250 ml) were collected in meticulously cleaned polyethylene bottle (TARSON, India) for analysis. Tubewells were flushed well before the collection of sample to remove the stagnant water. Two sets of samples were collected from each location. One set of sample was collected with 1 ml of conc. HNO₃ for cation and heavy metal analysis including As. The other set of sample was collected without any preservatives for the analysis of anions and other parameters. The water samples were not filtered because the residents of the areas use it for drinking unfiltered. The samples were preserved in ice box and brought to the laboratory and stored at 4 °C until analysis.

The pH and oxidation reduction potential (ORP) were measured onsite using portable kits (Hanna, HI 98121 waterproof pH/ORP/Temp) and the values are reported as the electrical potential of water sample relative to the reference electrode. Water quality parameters that were analyzed in the laboratory included major anions (SO₄²⁻, PO₄³⁻, HCO₃⁻, and NO₃⁻) and major cations (Ca, Mg, Na and K) along with Fe, Mn, Ni, Pb, Zn and As. SO₄²⁻, PO₄³⁻ and NO₃⁻ were analyzed spectrophotometrically (Systronics Visiscan 167) while HCO₃⁻ titrimetrically. Na, K and Ca were analyzed by Flame photometer (Systronics 128; Compressor 126). Water samples were digested with conc. HNO₃ following standard methods (APHA, 1998) prior to heavy metal analysis. Fe, Mg, Mn, Ni, Pb, and Zn contents in the groundwater samples were quantified by Flame Atomic Absorption Spectrometer (AAAnalyst 800; Perkin Elmer). Total As was analyzed using Atomic Absorption Spectrometer coupled with Hydride Generator (AAS-HG). The detail about AAS-HG has been described elsewhere (Yadav et al., 2014).

2.1.2. Sediment samples

Core sediment sample was collected using manual drilling percussion method, locally known as 'Dhikuli'. The details about drilling

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