Abstract

The present study delineates the groundwater solute chemistry, hydrogeochemical evolution, aquifer sediments provenance and geochemical processes that influences the fate of groundwater arsenic (As) in aquifers of three district tectono-morphic regions of Brahmaputra river basin (BRB). These regions are located in the north-western (NW) and the northern (N) parts of BRB, in the northern bank of the Brahmaputra river and along the western and eastern parts of Eastern Himalayas, and the southern (S) study area of BRB in southern bank of the river, situated near Indo–Burmese Range and Naga hills in the BRB. Stable isotopic composition (δ^2H and δ^{18}O) in groundwater suggest that some evaporation may have taken place through recharging water in the study areas. The major-ion composition shows that groundwater composition of the NW and N parts lies between Ca–HCO_3 and Ca–Na–HCO_3 hydrochemical facies, while Na–Ca–HCO_3 facies dominates the S-region region. X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyses of BRB aquifer sediments indicate the presence of feldspar (K-feldspar and plagioclase), mica (biotite and muscovite), ferromagnesian minerals, i.e., amphibole, highly altered pyroxene along with some heavy minerals, i.e., monazite, and rutile in NW and N aquifer sediments. Accordingly, the aquifers sediments in NW and N regions are derived from the Himalayan orogenic belt (Siwalik Group, medium to highly metamorphosed gneisses and schists rocks of Lesser and Higher Himalayas and igneous and metamorphic rocks of Eastern Syntaxis). The mineral compositions of S-aquifers differ from northern aquifers, by the presence of the higher proportion of Fe/Mg aluminosilicates, phyllosilicates or clay minerals, and heavy minerals i.e. titanite and ilmenite, derived from weathering of mafic igneous rock and associated pegmatites of Naga Hills. These mafic minerals, aluminosilicates, and clay minerals might offer the available reactive surface for As-adsorption and co-precipitation with amorphous Fe. These associated adsorbed and co-precipitated As might be released due to reductive dissolution of Fe-oxide and oxyhydroxides in groundwater. These minerals are assumed to be possible sources of As in groundwater. Reaction-path models suggest intense chemical weathering of S-aquifer matrix, suggested by the higher equilibrium of secondary mineral phases, as compared to NW and N regions. The chemical index of alteration (CIA) analysis supports the outcome of reaction-path modelling. Dissolved As distribution is highly variable among the different alluvial aquifers. The groundwater of S-region is much enriched in groundwater As (bdl to 5.53 μM or 415 μg/L, mean 1.77 μM) compared to NW and N regions (bdl to 1.8 μM or 134 μg/L, mean 0.28 μM; bdl to 2.45 μM or 184 μg/L, mean 0.68 μM, respectively). Almost more than 92% of groundwater samples in the S-region are enriched with As, which draws a distinct difference from the NW and N regions of

BRB aquifers. Surface complexation modeling suggests that competitive adsorption/desorption reactions in aquifer sediments also can have an influence on As mobilization mechanism. Reductive dissolution of minerals, i.e., Fe/Mn oxides/hydroxides, possibly due to microbially mediated redox reaction is the primary mechanism for As mobilization, followed by competitive adsorption/desorption reactions in aquifer sediments. Therefore, high variation in dissolved As concentrations among various tectono-morphic regions might be explained in term of geology and rock type of sediment provenance. The occurrence of As-enriched groundwater in Himalayan foreland basin in the BRB is probably a result of crustal evolution through which As is subsequently mobilized from aquifer matrix to groundwater by water–sediment reaction under favorable biogeochemical conditions. The present study proposes geological control (i.e. change in lithofacies, tectonic set-up) on groundwater chemistry and distribution of redox-sensitive solutes such as As in BRB

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