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Arsenic (As) contamination of groundwater in sedimentary aquifers is a major global public health crisis. Natural occurrences of high As groundwater have been reported in several geologically young aquifers around the world including the Datong basin, China. It is generally accepted that microbial processes play a key role in the mobilization and enrichment of arsenic (As) in groundwater. However, the detailed mechanism of the metabolic processes remain poorly understand. We apply isotopic measurements of iron (δ^{56} Fe vs. IRMM-14), sulfur ($\delta^{34}S_{SO4}$ vs. V-CDT) and carbon ($\delta^{13}C_{DIC}$ vs. V-PDB) to an experimental field plot in the Datong Basin, northern China. An array of monitoring wells was installed in a ≈ 1700 -m² plot in which high concentrations of As, ranging from 4.76 to 469.5 µg/L, were detected in the groundwater. The measured range of $\delta^{34}S_{SO4}$ values from 10.0 to 24.7 % indicates the prevalence of microbial sulfate reduction within aquifers. The range of δ^{56} Fe values measured in the groundwater suggests microbial Fe(III) reduction and the occurrence of isotopic exchange between Fe(II)(aq) and FeS precipitation. The low $\delta^{13}C_{DIC}$ values (up to -33.6‰) measured in groundwater are evidence for the microbial oxidation of organic matter, which is interpreted as the light carbon pool within the aquifer sediments. The high As (As>10 µg/L) groundwater, which has higher $\delta^{34}S_{so4}$ and $\delta^{56}Fe$ values and lower $\delta^{13}C$ values, indicates the following: (1) microbial reduction of sulfate causes the mobilization of As through HS⁻ abiotic reduction of Fe(III) minerals; and (2) direct microbial reduction of Fe(III) oxides, hydroxides and oxyhydroxides cannot increase As concentrations to greater than 50 μ g/L. The results provide new insight into the mechanism of As enrichment in groundwater.