Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.37744

Sources, occurrence and mobilization of arsenic related to iron oxide concretions in former redbed sandstones, Colorado Plateau

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Naturally occurring arsenic in drinking water remains a worldwide public health concern with arsenic often associated with iron oxide minerals. In the Colorado Plateau, the Jurassic Navajo Sandstone aquifer system commonly contains elevated dissolved As concentrations and is an important source of domestic water supply for rural populations, yet the source and mechanisms for arsenic mobilization are relatively unknown. Previous studies show arsenic in the bulk Navajo Sandstone is generally low (mean concentration of 10 ppm), while siltier layers within the Navajo Sandstone and underlying Kayenta and Chinle formations contain relatively high As (up to 500 ppm). Iron oxide cements, along joints in bleached portions of the Navajo Sandstone, near the Escalante Anticline, collected as part of this study, are also enriched in As (>7000 ppm), consistent with other types of iron oxide concretions elsewhere in the Colorado Plateau. Combining new rock and water chemistry data with reactive transport modeling (RTM) using PFLOTRAN, this study aims to constrain paleo fluid flow and fluid-rock reactions that led to arsenic accumulation on iron oxides and eventual release to groundwater dependent on source, flow paths, and hydrogeochemical conditions.

Preliminary RTM results show that arsenic can be adsorbed on iron oxide cements along joints in the bleached Navajo Sandstone in discharge areas from upward migration of As-bearing reducing fluids from underlying formations. Alternatively, arsenic may have accumulated from As-bearing lateral regional groundwater flow in the geologic past, although the original source of arsenic is unclear. Today, Holocene to Pleistocene age groundwater in the Navajo Aquifer is relatively oxic with near-neutral pH indicating that arsenate ($HAsO_4^{2-}$) is the dominant arsenic species. There is also no relationship between dissolved As and Fe, suggesting As is not coming from reductive dissolution of As-bearing iron minerals (e.g., arsenopyrite) in the study area. Rather, elevated As in well water is associated with lower $Na/(Ca)^{0.5}$ molar ratios and longer residence times, suggesting cation exchange and desorption reactions with iron oxide concretions and/or other clay-rich units may release arsenic to groundwater, as seen in other semi-arid oxic aquifer systems.