

# Isotope tracing of iron cycling in a crustal deep fracture-hosted hydrogeosphere

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Deep fracture fluids hosted in crystalline basement rocks may represent 30% of the groundwater inventory (1) and feature diverse geochemistry and microbial ecosystems. These include fluids with geochemical characteristics of prolonged water-rock interaction and noble gas components with multiple-billion-year ages (1–3). These ancient and highly saline fluids host chemolithoautotrophic microbial ecosystems that derive energy primarily from coupling sulfate reduction to oxidation of hydrogen produced by serpentinization and/or radiolysis, potentially in isolation from Earth surface process (4–6). Little is known, however, about associated metal cycling within these systems, specifically, how dissolved metal loads may reflect diverse host rock lithologies and respond to *in situ* biogeochemical processes.

One such system is encountered in Kidd Creek mine, Ontario, a Cu-Zn sulfide mine hosted within a volcanogenic massive sulfide (VMS) deposit in the Neoarchean Abitibi greenstone belt (1, 2), where pyrite accounts for 70% of the VMS. Here fracture fluids contain a small standing sulfate pool maintained by indirect radiolytic oxidation of pyrite (IROP), and consumed by *in situ* microbial sulfate reduction (4). We analyzed iron isotope ratios ( $\delta^{56}\text{Fe}$ ) for fracture fluids, and the surrounding VMS and other host rocks from this locality, to constrain likely lithological sources of high dissolved  $\text{Fe}^{2+}$  to these fluids and to determine whether iron is, like sulfur, actively cycled within the fracture fluid environment (4). Fracture fluid  $\delta^{56}\text{Fe}$  and Fe/Mn suggest that the dissolved  $\text{Fe}^{2+}$  pool is impacted by secondary sulfide mineral formation, likely coupled to microbial reduction of IROP-derived sulfate. This study identifies these key processes that may control the Fe and S cycle in deep crustal settings such as Kidd Creek, and thereby sustain long-term chemolithoautotrophic ecosystems in the deep subsurface.

## References

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