

## Sulfur cycling in ancient terrestrial fracture waters transports Archean signatures

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Subsurface microbial communities inhabiting saline fracture waters in the deep terrestrial subsurface such as those in South Africa [1], may rely on sulfate (SO<sub>4</sub>) reduction coupled to hydrogen (H<sub>2</sub>) oxidation [2] as a key metabolic strategy. The cycling of sulfur in anoxic fracture waters in Canada with average crustal residence times >1 Ga [3] is important to understand SO<sub>4</sub> sources for potential subsurface life in other deep Earth environments. The production of SO<sub>4</sub> and associated redox gradients in subsurface aqueous environments also has implications for habitability on other planets (e.g., Mars). In order to constrain the origins of SO<sub>4</sub> in these systems, we carried out multiple sulfur (S) isotope analyses on dissolved SO<sub>4</sub> and sulfide in fracture fluids from five mines in the Precambrian Canadian Shield, including one entirely hosted in Archean rocks (Kidd Creek, ON), and four located at the boundaries between Archean and Paleoproterozoic terranes (Thompson and Birchtree, MN; two mines in the Sudbury basin, ON).

Characteristic Archean mass independent fractionations in S isotopes (S-MIF) were observed in SO<sub>4</sub> and sulfide in fluids from all locations. To our knowledge this is the first evidence of S-MIF signals in fluids freely discharging from fractures at rates of > 1 L/min. Negative  $\Delta^{33}\text{S}$  values in fracture water SO<sub>4</sub> are found at 2.4 km and 2.9 km depth at Kidd Creek while positive  $\Delta^{33}\text{S}$  values are observed at Thompson and Birchtree mines and at both mines in the Sudbury basin. The similarity in S-MIF signatures between the SO<sub>4</sub> and the host rock sulfides indicate that the Archean S is mobilized into the fluids through water-rock reaction over long time- and space-scales. This finding connects the ancient S cycle and S cycling in modern subsurface fluids.

- [1] Lin et al. (2006) *Science* **314**, 479-482; [2] Sherwood Lollar et al. (2014) *Nature* **516**, 379-382; [3] Holland et al. (2013) *Nature* **497**, 357-360.