Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.17137

Preferential partitioning of sulfate species into slab fluids: insights from the DEW model and implications for sulfur isotope cycling

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The release of aqueous fluids from subducting slabs to the mantle wedge drives arc magmatism and elemental cycles over geological timescales. However, there are widely differing views about the nature of these fluids, their speciation, and how they impact and modify the composition and oxidation state of the mantle wedge. Slab-derived sulfur may play a key role, with potential for oxidizing the mantle wedge and producing S-rich arc magmas. This is supported by enriched isotopic signatures (i.e. $\delta^{34}S > 0‰$) documented in a growing number of melt inclusions and volcanic gases (Fig. 1). However, there is conflicting evidence from arc crustal sections and exhumed metamorphic rocks, hence the source, speciation and transport of sulfur in the mantle wedge remains speculative. Here we use the Deep Earth Water model [1,2] to calculate the speciation of fluids in equilibrium with mafic and ultramafic lithologies, from 2 to 3 GPa and 450 to 650°C at varying oxygen fugacities from FMQ-2 to FMQ+4. We show that oxidized fluids can carry orders of magnitude more sulfur, as sulfate species, than reduced fluids (Fig. 2). This effect is enhanced with increasing pressure and decreasing temperature. We also predict a significant role for aqueous $CaHSO_4^+$, required to match the high-pressure experimental solubility of anhydrite [3]. This is consistent with complementary evidence that metal-sulfate complexes contribute to mass transfer in subduction zones [4,5]. Furthermore, at mildly oxidizing conditions (~FMQ+2) sulfate-rich fluids are in equilibrium with pyrite, which could cause large S isotope fractionation with the fluid being enriched up to 10‰ relative to the slab. Therefore, such fluids could impart enriched S isotope signatures to the mantle wedge and ultimately be transferred to arc magmas.

[1] Sverjensky et al. 2014, *Geochimica et Cosmochimica Acta;* [2] Huang & Sverjensky 2019; *Geochimica et Cosmochimica Acta;* [3] Newton & Manning 2005, *Journal of Petrology;* [4] Pons et al. 2016, *Nature Communications;* [5] Debret et al. 2020, *Geochemical Perspectives Letters*

Figure 1: Compilation of sulfur isotope compositions in the subduction zone cycle
Modified after Rezeau et al. (2023), *Chemical Geology*

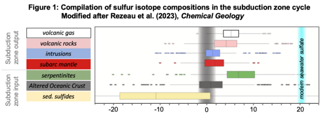

Figure 2: DEW model prediction of sulfur speciation in fluids at equilibrium with eclogite and a S-bearing mineral