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Multiple sulfur isotopes evidence deep intra-slab transport of sulfate-rich fluids

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Sub-arc mantle oxidation is often attributed to sulfates from seawater transported in aqueous fluids derived from slab dehydration, but sulfur speciation and origin(s) in high-pressure fluids remain weakly constrained. Here, we use the four sulfur stable isotopes (^{32}S, ^{33}S, ^{34}S, ^{36}S) to decipher processes, e.g. mixing and/or open-system reaction occurring during sulfur devolatilization, and deduce the nature and origin of sulfur species in aqueous slab-derived fluids. We analyzed high-pressure metamorphic rocks from the Monviso massif Lower Shear Zone (Western Alps), recording evidence of an interface-parallel, channelized fluid flow near peak burial metamorphic conditions (~550°C and 2.7GPa). We sampled metasomatized Fe-Ti metagabbros, hybrid (talc) schists and serpentinites containing sulfides supposed to preserve the fluid composition, as well as other alpine metagabbro, metabasalt and serpentinites from greenschist to eclogite-facies devoid of the metasomatic overprint.

The sulfides found in abundance within LSZ lithologies are mainly pyrites that crystallized at peak conditions or during incipient exhumation. The sulfides in the non-metasomatic metagabbros and metabasalts cover a narrow range in δ'^{34}S of 0.89 ± 0.63‰ and Δ'^{33}S of 0.017 ± 0.01‰ (n = 6) that could be inherited from hydrothermal sulfides formed on the seafloor. In contrast, the LSZ sulfides from the Fe-Ti metagabbros, serpentinites and hybrid schists (n = 24) are enriched in ^{34}S and ^{33}S (δ'^{34}S from 4.91 to 21.32‰ and Δ'^{33}S from 0.02 to 0.06‰) compared to the sulfides from the non-metasomatic rocks. Mixing between reduced sulfur species would require an end-member composition as high as δ^{34}S ~ 22‰ and Δ^{33}S ~ 0.06‰, values rarely recorded by sulfides in the literature. Instead, we propose that this ^{33}S-^{34}S-enrichment results from open-system sulfate reduction to sulfide in aqueous fluids at 550°C, with an initial δ^{34}S ~ 12 ± 3‰ and Δ^{33}S ~ 0.025 ± 0.010‰. Because this signature is different from seawater sulfate, we suggest that sulfate was produced by hydrothermal sulfide oxidation during antigorite breakdown at 650°C, in agreement with Mg, Ni and Cr enrichments in the metasomatized HP mafic lithologies. This model is consistent with oxidized ^{34}S-enriched arc lavas and could explain recycled sulfur with positive δ^{34}S recorded by ocean island basalts.