



Tracing the Recycling of Water, Carbon, and Sulfur during Subduction Metamorphism of Seafloor Serpentinites:

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We use the contents and isotope compositions of water, carbon, and sulfur in serpentinites and their dehydration products in order to trace the cycling of volatiles during subduction. Antigorite serpentinites of the Cerro del Almirante complex, Spain, contain 9-12 wt% H₂O and 910 ± 730 ppm sulfur, have bulk $\delta^{18}\text{O}$ values of 8.6 ± 0.4‰, $\delta\text{D} = -54 \pm 5\text{‰}$ and $\delta^{34}\text{S} = 5.0\text{‰}$ consistent with serpentinization at temperatures of ~200°C by seawater hydrothermal fluids on the Tethyan seafloor. The serpentinites were dehydrated to chlorite-harzburgite (olivine + orthopyroxene + chlorite) at 700°C and 1.6-1.9 GPa during subduction metamorphism, resulting in loss of water, carbon, and sulfur. The chlorite-harzburgite contains 5.7 ± 1.9 wt% H₂O, have bulk $\delta^{18}\text{O} = 8.0 \pm 0.9\text{‰}$ and $\delta\text{D} = -77 \pm 11\text{‰}$. The rocks contain 650 ± 620 ppm sulfur having $\delta^{34}\text{S} = 1.2\text{‰}$. Dehydration of serpentinite resulted in loss of 5 wt% H₂O having $\delta^{18}\text{O} = 8-10\text{‰}$ and $\delta\text{D} = -27$ to -65‰ and loss of 260 ppm sulfur as sulfate, having $\delta^{34}\text{S} = 14.5\text{‰}$. The contents and $\delta^{13}\text{C}$ of total carbon in the two rock types overlap, with a broad trend of decreasing carbon contents and $\delta^{13}\text{C}$ from ~1300 to 200 ppm and -9.6 to -20.2‰. This reflects mixing between reduced carbon in the rocks (210 ppm, $\delta^{13}\text{C} \approx -26\text{‰}$) and seawater-derived carbonate ($\delta^{13}\text{C} \approx -1\text{‰}$). Our results indicate: 1) Serpentinized oceanic peridotites carry significant amounts of isotopically fractionated water, carbon and sulfur into subduction zones; 2) Subduction of serpentinites to high P and T results in loss of water, carbon, and sulfur, which can induce melting and contribute to ¹⁸O, D, and ³⁴S enrichments and oxidation of the sub-arc mantle wedge; and 3) Isotopically fractionated water, carbon, and sulfur in serpentinite dehydration products are recycled deeper into the mantle where they can contribute to isotope heterogeneities and may be significant for volatile budgets of the deep Earth.