



Is the CO₂ content of subduction-zone fluids higher than previously thought?

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Subduction of the oceanic lithosphere and its sedimentary cover is accompanied by devolatilization processes. CO₂ removal through dissolution of carbonates occurring in altered oceanic lithosphere and its sedimentary cover, along with diapirism of slab rocks and/or melts, provides an efficient way to recycle carbon back to the mantle wedge and, ultimately, to the Earth's surface. However, other forms of carbon, often closely associated with silicates, have been reported in slab rocks and in particular in subduction mélanges. For instance, graphite has been described in blueschist-facies mafic rocks, metasediments and hybridized peridotites at Santa Catalina, and in ophiolitic serpentinites from the Western Alps, where also diamond has been found in UHP metasediments.

Estimates of dissolved CO₂ in subduction-zone fluids are based on thermodynamic models, relying on a very sparse experimental data base. We provide comprehensive experimental constraints on the composition of high-pressure graphite-saturated COH fluids in terms of dissolved CO₂, SiO₂ and MgO in increasingly complex petrological systems at controlled redox conditions, buffered by using the double-capsule technique and both the nickel-nickel oxide (NNO) and the fayalite-magnetite-quartz (FMQ) buffers ($\Delta\text{FMQ} \approx -0.5$), in order to develop a model for the interaction between deep aqueous fluids and silicates in subduction mélanges. A carbonate-free compositional range has been explored at P = 1 GPa, T = 800°C and P = 3 GPa, T = 800°C in order to focus on the role of graphite and silicates in the investigated processes. We synthesized COH fluids in equilibrium with graphite and other minerals representative of subduction mélanges, i.e. Mg-silicates (forsterite and enstatite), representative of the mantle component, and quartz/coesite, representative of the sedimentary component. Experimental products were analyzed for their volatile COH composition by quadrupole mass spectrometry (QMS) and for their Mg and Si solute load by cryogenic laser ablation inductively coupled plasma mass spectrometry (ICP-MS). Measured data were compared to thermodynamic modeling results. Our results suggest that the interaction of deep aqueous fluids with silicates in the presence of graphite in a subduction mélange promotes the dissolution of graphite and enhances the CO₂ contents of the fluids. This provides a new mechanism for controlling the volatile composition of COH fluids already at depths of ~30 km, promoting the deep CO₂ transfer from the slab–mantle interface to the overlying mantle wedge, in particular where fluids are stable over melts.