High-pressure experimental and thermodynamic constraints on the solubility of carbonates in subduction zone fluids

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Carbon on the Earth's surface can be carried into the mantle by the subducting slabs in the form of carbonates and organics. However, not all input carbon in the slab could be finally recycled into the deep Earth's mantle because parts of carbon will release from the subducting slabs by different decarbonation processes (e.g., metamorphism and dissolution). In comparison with metamorphism decarbonation, the dissolution of carbonates in the subduction zone fluids is more essential but not well constrained previously. In this study, we systematically constrained the solubility of different carbonates (e.g., calcite/aragonite, dolomite, and magnesite) at high-pressure and high-temperature conditions relevant to subduction zones by combining high-pressure experimental and thermodynamic simulations. We simultaneously compared the dissolution processes of calcite/aragonite, dolomite, and magnesite in aqueous fluids at the same high-pressure (high-P) and high temperature (high-T) conditions by applying the four-hole gasket technique in the diamond anvil cell (DAC) experiments. In situ Raman spectra of fluids in DAC clearly showed that dolomite has a similar solubility to aragonite in aqueous fluids at the same P-T conditions, while the solubility of magnesite is much lower than that of both aragonite and dolomite. Notably, we also found that the dissolution of dolomite in aqueous fluid is an incongruent process via reaction: CaMg(CO3)2(solid) in H2O = $MgCO_{3(solid)} + Ca^{2+}_{(aqueous)} + CO_{3}^{2-}_{(aqueous)}$. Based on our highpressure experimental results, we modified the carbon species in the original Deep Earth Water (DEW) model and quantitatively calculated the speciation and solubility of calcite/aragonite, dolomite, and magnesite in aqueous fluid at pressure up to 5 GPa and temperature up to 800 °C. Then, we modeled and compared the decarbonation fluxes between metamorphism and dissolution from typical marine sediment and altered oceanic crust (AOC) in subduction zones and found dissolution of carbonates is ignored but more important decarbonation process than metamorphism in the cold and intermediate subduction zones.