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Carbonate stability during serpentinite dehydration in subduction zones – insights from open system thermodynamic modeling

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Aqueous fluids containing dissolved carbonate, silicate and salt components are essential to understanding mass transfer from the slab to the plate interface and the mantle wedge in subduction zones. Thermodynamic models show that prograde devolatilization reactions of carbonate-rich slab lithologies at subarc depths cannot account for the carbon flux of arc volcanoes. This imbalance persists when considering metal ion complexes that enhance carbon solubility in aqueous fluids [1], indicating that open-system fluid processes play a critical role in the subsolidus mobilization of carbon in subduction zones. Here, we present the results of open system thermodynamic modeling implementing the DEW aqueous database in Perple_X [1] to calculate the solubility, fluid speciation, and phase equilibria of serpentinite-hosted carbonates (meta-ophicalcite and carbonate-talc rocks) in fluids derived from the antigorite breakdown. These lithologies are more appropriate than pure aragonite to investigate carbonate solubility in subduction zones. For a given time-integrated fluid flux, the carbon released by dissolution of meta-ophicalcites and carbonate-talc rocks is markedly higher than for pure aragonite. The stability of $CaHCO_3^+$(aq) promotes Ca loss and increases pH, resulting in fluids more alkaline than those derived from serpentinite dehydration. The calculated mass-loss, phase assemblages, and fluid speciation allow identifying key open-system buffer reactions between carbonates, silicates, and fluid. The dissolution of serpentinite-hosted carbonates depends on the serpentinite and carbonate volume in the slab, the thermal regime of the subduction zone, and the fluid pathways. Mass balance calculations and natural case studies show that meta-ophicalcites can be preserved beyond the antigorite breakdown even in hot subduction zones, carrying carbon into the deep mantle. In contrast, fluid infiltration readily dissolves carbonate-talc rocks and transforms them into orthopyroxenite in most subduction zone regimes.

[1] Connolly & Galvez, 2018, *EPSL* 501, 90-102.