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High Solubility of Aragonite in Aqueous Fluids: Experiments and Models

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Carbonate dissolution in subduction zone fluids is critical to assess the carbon budget in subduction zones. Depending on the solubility of carbonate minerals in aqueous fluids, the subducting lithosphere may be either strongly depleted and the mantle metasomatized if the solubility is high, as suggested by natural samples or it could transport carbon deeper into the Earth's mantle if the solubility is low enough.

Dissolution of carbonate minerals strongly depends on pressure and temperature as well as on the chemistry of the fluid, leading to a highly variable speciation of aqueous carbon. Combined experimental and theoretical efforts allowed the investigation of speciation and solubility of carbonate minerals in aqueous fluids at high P,T conditions. In this study, we present in situ Raman and X–ray fluorescence spectroscopic measurements of aragonite dissolution to 5 GPa and 500°C and the subsequent thermodynamic model of aragonite solubility in aqueous fluids thanks to the Deep Earth Water model. The amount of dissolved aragonite in the fluid was calculated from challenging and unprecedented measurements of the Ca fluorescence K–lines at low-energy. Experiments were performed at the ESRF, using a dedicated design of an externally-heated diamond anvil cell and an incident high-flux and highly focused monochromatic X–Ray beam at 20 keV. The results show a spectacularly high solubility of aragonite at HP-HT in water, reaching molar concentrations and further enhanced in presence of NaCl in the solution. Such a high solubility requires the formation of ion pairs and a new model of speciation in equilibrium with aragonite under oxidizing conditions.