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Experimental determination of plagioclase dissolution rates at near equilibrium conditions utilizing an isotopic tracer

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Alteration of oceanic basalts is an important sink on the global carbon cycle and over the last two decades they have been targeted as a potential sink of anthropogenic carbon emissions by mineral carbonation. The dissolution of primary minerals rich in Ca, Fe and Mg is likely the rate-limiting step to achieve mineral carbonation. As minerals dissolve carbonate forming cations are released which can react with dissolved CO_2 to form Ca-carbonate minerals. Plagioclase feldspar, the most volumetrically abundant mineral in basalts, is targeted for mineral carbonation as it represents most of the reactive surface area available for alkalinity generation and carbonate precipitation in basalts. However, plagioclase dissolution rates have historically been determined at far from equilibrium conditions to avoid secondary mineral precipitation and significant changes in effluent solutions. As a result, we have few quantitative constraints on plagioclase dissolution and coupled clay and carbonate mineral precipitation kinetics at near-equilibrium conditions. Here, we react ^{29}Si-doped synthetic seawater solutions with plagioclase at 60°C and low and high partial pressures of CO_2. These experiments allow us to quantify feedbacks between dissolution and precipitation reactions and estimate dissolution rates as the solutions approach equilibrium with respect to plagioclase. Our results show a rate reduction of around two orders of magnitude over experimental timescales up to several months when dissolving plagioclase in seawater at 60°C and both at low and high pressures of CO_2. This newly determined dependence of rates on solution saturation state provides a new framework to guide efforts to both model oceanic crust alteration and evaluate the persistence of injected free CO_2 during mineral carbonation simulations.