

Silicon isotopes as a new method of measuring silicate mineral reaction rates in the critical zone

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²⁹Si doped experiments demonstrated that Si isotope doping is a robust new experimental technique for measuring silicate reaction kinetics. Albite dissolution batch experiments were conducted under ambient temperature and pH 3-7.5. In our experiments, initial solutions were doped with artificial ²⁹Si, resulting in a Si isotopic composition highly anomalous to natural Si isotope compositions. The isotopic contrast and analytical precision of ± 0.05 (unit in %) allow detection of the dissolution of a minute amount of albite in the aqueous solutions. Results show that the temporal evolution of ²⁸Si and ²⁹Si abundances in batch experiments (3-270 days) tightly bracketed the steady state albite dissolution rates, and the measured rates are consistent with literature data. Because the precipitation of secondary phases consumes silica but leaves the Si isotope ratios unchanged in experimental solutions, dissolution rates were still measurable when secondary phase precipitation took place in experiments. Meanwhile, the combination of Si isotopes and Si elementary concentrations, precisely determined with the Si isotope dilution method, allowed albite dissolution and secondary phase precipitation rates to be determined simultaneously. Experimental data illustrate that this method allows measurements of silicate reaction rates at precisions and conditions never before possible. The high detection sensitivity allows measuring steady state rates extending to longer experimental duration than previously possible at ambient temperatures. Freedom from interferences of secondary phase precipitation means we can now measure rates at circumneutral pH and near equilibrium conditions under which precipitation likely takes place. Particularly, measurements of rates close to equilibrium would help resolve the long-standing problem of field-lab gap in silicate dissolution rates.