Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.17143

Rapid secondary phase formation and associated Si isotope fractionation during lab weathering experiments of andesite

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Isotopic proxies (such as Si, Li, Ge isotopes) are frequently used to qualitatively track and interpret chemical weathering processes in the Critical Zone. While it has been shown that they are primarily fractionated by the formation of secondary weathering phases, the kinetic rates and isotopic fractionation factors associated with these reactions are rarely calibrated in controlled lab experiments, especially ones that are designed to simulate in-situ weathering. Such calibrations are needed to enable more quantitative use of isotopic weathering tracers in complex natural systems.

Here, we present the results of a batch reactor weathering experiment, where crushed andesitic bedload (representative of partly weathered regolith) from the Capesterre river (Guadeloupe island, West Indies) was reacted under 10% pCO_2 for up to 150 days. During the experiment, dissolved Si reached a steady state concentration of 420-460 μmol/L, while dissolved δ^{30}Si evolved from -0.4 to 1.7-2.0‰. This silicon isotope fractionation is driven by the rapid formation of secondary phases, visible in suspension (compare Fig. 1 and 2) and and likely to be amorphous Al-Si oxides like allophane (Al/Si = 0.8-2.5 mol/mol).

In addition, dissolved cation concentrations were strongly modified by cation exchange onto negatively charged solid surfaces (likely a small fraction of secondary phases present in the river bedload). We used Cs^+ tagging and cation leaching procedures to correct the dissolved cation concentrations for exchange onto solids, allowing the calculation of primary mineral dissolution rates close to equilibrium, independently of Si concentrations.

Overall, we are able to directly calibrate andesite regolith reaction rates under elevated CO_2 concentration, including the kinetics of secondary phase formation and associated Si isotope fractionation. These experimental data enable a more robust modeling of weathering processes and the interpretation of δ^{30}Si signatures in the studied catchment.

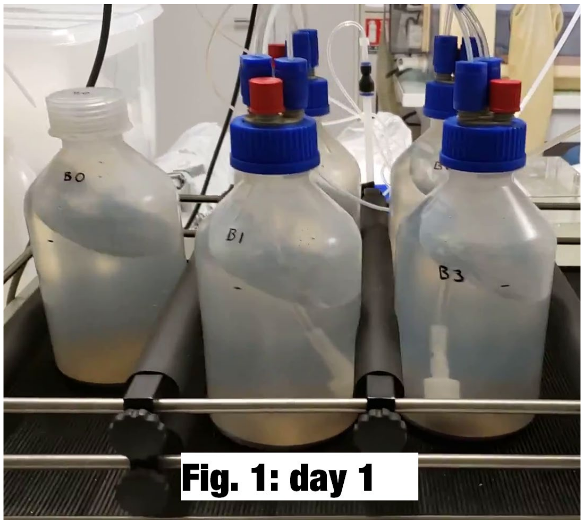

Fig. 1: day 1

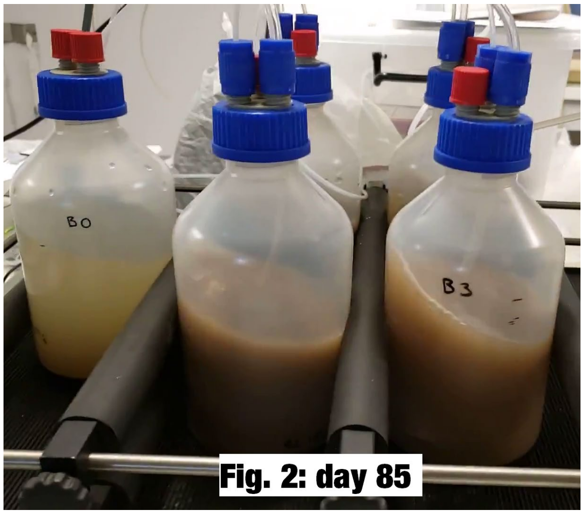

Fig. 2: day 85