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Theoretical evaluation of equilibrium triple silicon isotope fractionation in Earth-surface processes

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Chemical weathering and reverse weathering are central components of Earth-surface processes, linking the global silicon and carbon cycles and exerting long-term control on atmospheric CO_2 and planetary habitability. Although conventional stable isotope systems have been widely applied to investigate weathering reactions, their interpretation is commonly limited by biological cycling, hydrothermal inputs, and the non-uniqueness of single-isotope signals generated by distinct mineral–fluid pathways. These limitations reflect a fundamental challenge in chemical weathering studies: multiple overlapping processes can produce similar isotopic signatures.

High-precision triple silicon isotope measurements, expressed as $\Delta'^{29}Si = \delta'^{29}Si - C \times \delta'^{30}Si$ (where C is a reference slope), provide an additional dimension for constraining mass-dependent fractionation and offer new potential to distinguish among silicate weathering, authigenic clay formation, and biologically mediated Si cycling[1-2]. However, quantitative interpretation of $\Delta'^{29}Si$ in Earth-surface environments has been hindered by the lack of fundamental constraints on the silicon isotope fractionation exponent, $\theta(\equiv \ln^{29}\alpha/\ln^{30}\alpha)$, particularly at low temperatures.

Here we present a theoretical evaluation of equilibrium triple silicon isotope fractionation using quantum mechanical calculations to determine temperature-dependent fractionation exponents (θ_{eq}) between aqueous silicate species and major silicon-bearing minerals. The resulting θ_{eq} values establish the equilibrium framework required to translate measured $\Delta'^{29}Si$ signatures into specific mineral–fluid processes. Based on these results, we recommend a standardized reference slope (C = 0.5155) for low-temperature terrestrial environments(Fig. 1), enabling systematic assessment of deviations from equilibrium associated with kinetic isotope effects during weathering and authigenic clay formation. This work provides the theoretical foundation necessary to apply triple silicon isotopes as a process-sensitive proxy for disentangling overlapping pathways in Earth-surface silicon and thus carbon cycling.

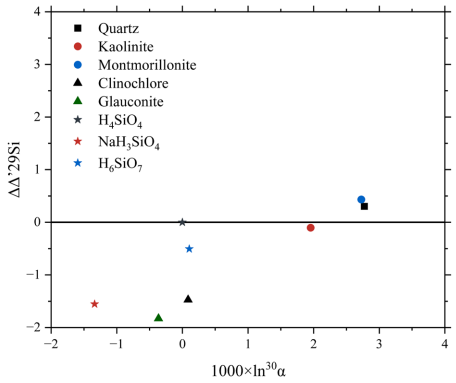

Fig. 1. Equilibrium triple silicon isotope fractionation plot ($\Delta\Delta'^{29}Si$ vs. $1000\times\ln^{30}\alpha$) calculated relative to aqueous H_4SiO_4 using the recommended 0.5155 reference slope. These results define the theoretical equilibrium baseline for identifying kinetic and biological signals in global silicon cycling.

Reference

[1] Triple silicon isotope insights into the formation of Precambrian cherts, Sun et al., (2023), *Earth and Planetary Science Letters* 607, 118069.

[2] A new method for silicon triple isotope analysis with application to siliceous sponge spicules, Pack et al., (2023), *Geochemistry, Geophysics, Geosystems* 24, e2023GC011243.