



Silicon isotope fractionation during silica precipitation from hot-spring waters

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Hot-spring systems in the Geysir geothermal area, Iceland, have been studied to explore silicon isotope fractionation in a natural setting where sinter deposits are actively formed over a temperature interval between 20° and 100°C. The $\text{SiO}_{2(aq)}$ concentrations in spring and stream waters range between 290 and 560ppm and stay relatively constant along downstream trajectories, irrespective of significant cooling gradients. The waters are predominantly oversaturated in amorphous silica at the temperatures measured in the field. Correlations between the saturation indices, temperature and amounts of evaporative water loss suggest that cooling and evaporation are the main causes of subaqueous silica precipitation. The $\delta^{30}\text{Si}$ values of dissolved silica in spring water and outflowing streams average around +1‰ probably due to the small quantities of instantaneously precipitating silica relative to the dissolved amount. Siliceous sinters, in contrast, range between -0.1‰ to -4.0‰ consistent with a preferred incorporation of the light silicon isotope and with values for precipitated silica becoming more negative with downstream decreasing temperatures. Larger fractionation magnitudes are inversely correlated with the precipitation rate, which itself is dependent on temperature, saturation state and the extent of a system. The resulting magnitudes of solid-fluid isotopic fractionation generally decline from -3.5‰ at 10°C to -2.0‰ at 90°C. These values confirm a similar relationship between fractionation magnitude and temperature that we found in laboratory-controlled silica-precipitation experiments. However, a relatively constant offset of ca. -2.9‰ between field and experimental fractionation values indicates that temperature alone cannot be responsible for the observed shifts. We infer that precipitation kinetics are a prominent control of silicon isotope fractionation in aqueous environments, whereby the influence of the extent of the system on the precipitation rate is a central factor. As an important corollary, the fractionation behavior during precipitation of silica from saturated solutions in natural environments should be regarded as a system-dependent feature.