

Silicon isotopic fractionation during water-rock interaction

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The Si isotopic composition of natural waters is a promising proxy to gain insight into the processes that govern the global biogeochemical Si cycle. We measured the isotopic Si fractionation of opal, diatomite, quartz and amorphous silica during dissolution at pH 3 and 9 at 70 °C in an attempt to help quantify these processes.

The isotopic composition of the Si released from quartz is identical to that of the dissolving mineral. In contrast, the Si released initially by opal-CT, diatomite, and amorphous silica dissolution at pH 3 is isotopically light, but with time, the Si isotopic composition of the fluid phase becomes heavier and eventually exceeds that of the dissolving solids. In contrast, at pH 9, the fluids are initially enriched in isotopically heavy Si; these fluids become isotopically lighter with time, converging in the case of diatomite and opal towards the composition of the dissolving solids. This trend continues despite the fact that bulk equilibrium between fluid and solid has been attained. This observation confirms that minerals and their co-existing fluids can alter isotopic composition even after bulk equilibrium between fluid and solid is attained. Moreover the temporal pattern of the isotopic evolution in our experiments suggests that the two-way-transfer of material to and from the mineral surface is responsible for the observed release of Si isotopes during dissolution.