

Silicon isotope fractionation during the formation of amorphous hydroxyaluminosilicates and hydrous ferric silicate phases

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Silicon (Si) is a key element during coupled mineral dissolution and neo-formation processes, as it is mobilized during silicate weathering, and transported as silicic acid (Si(OH)₄) towards the ocean, before being sequestered by silicifying organisms (e.g., diatoms) and diverse silicate minerals i.e., authigenic clay minerals. The Si cycle is of particular interest within the critical zone (CZ), where Si is suspected to first precipitate as amorphous, gel-like phases such as short-range ordered hydroxyaluminosilicates (HAS: e.g. allophane) or as hydrous ferric silicates (HFS: e.g. hisingerite). These highly reactive minerals are acting as precursors for the formation of important soil clay minerals e.g., within the smectite and kaolinite group. To decode the reaction paths and the environmental controls underlying HAS and HFS formation, the use of Si isotope fractionation can be a powerful tool. For this purpose, a series of allophane-hisingerite precipitation experiments at high temporal resolution has been performed to investigate and assess the kinetic and equilibrium Si isotope fractionation between the reactive fluid and the precipitated solid phase. Isotope exchange mechanisms are investigated using the three-isotope method, which is a novel approach to trace the direction and the progress of low-temperature water-mineral/rock interactions.