Formation and environmental significance of short-range order allophane hisingerite solid-solutions

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The chemical weathering of silicates and the subsequent formation of clay minerals are important processes within Earth's critical zone, affecting, for instance, the pH value, the water-bearing capacity, the ion exchange properties and the availability and mobility of nutrients in soils. Short-range ordered (SRO) minerals such as allophane ($\sim Al_2O_3 \cdot (SiO_2)_{1.3–2} \cdot (H_2O)_{2.5–3}$) and hisingerite ($\sim Fe^{3+}_2Si_2O_5(OH)_4 \cdot H_2O$) are common secondary weathering products in soils, but the relationships between the ambient environmental conditions, SRO-mineral formation paths and their resulting crystal-chemical properties are poorly understood. Therefore, end-members and solid-solutions of the allophane-hisingerite series were precipitated at different molar ratios of Al/Si, Fe/Si and (Al+Fe)/Si at ambient temperature using batch experiments in order to study the nature, chemical composition and nanostructure of the precipitates. The obtained SRO-minerals were characterized by XRD, FTIR, (E)SEM and TEM techniques. The reactive aqueous solutions were continuously sampled over a period of 2 weeks and analyzed using ICP-OES to investigate the temporal evolution of Al, Fe and Si during SRO-mineral precipitation. Preliminary results indicate that solid solutions with homogenous products were obtained and that allophane-hisingerite formation occurs within seconds to few minutes under the chosen experimental conditions. This might proceed via an instantaneous precipitation of Al/Fe-bearing octahedral template sheets onto which silicate tetrahedrons are getting attached through condensation and polymerization reactions. Silicon isotope analyses of the reactive fluids and solids will provide further insights into the specific physicochemical conditions and pathways of SRO-mineral formation at low temperature.