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Prediction and validation of coupled mineral dissolution and precipitation along a reactive front in porous rocks

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Understanding reactive transport in porous rocks is often challenging when multiple reactions involve the same constituents. For example, primary silica-aluminium minerals such as feldspar and chlorite can dissolve close to an acidic source and secondary silica-aluminium minerals in form of kaolinite, zeolites and smectites can precipitate further downstream of the reactive front. In this case, the outflow concentrations only reflect the net balance between dissolution and precipitation reactions. The type of reactions and the respective reaction rates, however, remain unknown. This problem is addressed through flow-through experiments where the column is filled with scoria, a pyroclastic rock mainly composed of volcanic glass, pyroxene, olivine and plagioclase and nitric acid is used as the inflow reagent. The fluid flow velocity and the respective residence time was varied and the steady-state outflow fluid composition analysed. The data was initially analysed using combinatorial inverse modelling implemented in R coupled to PHREEQC [1]. Here, the fluid outflow composition and the solubility of minerals is used to calculate the statistical likelihood of minerals involved in dissolution and precipitation reactions [2]. In parallel, high resolution mineral maps were derived from SEM-EDX images and registered to micro-CT images with the aim to visualise the distribution of different mineral phases and groups in the flow-through column. A time series of micro-CT images then allowed the identification of minerals and their locations in the column involved in mineral dissolution and precipitation. The inverse modelling and the experimental results suggested volcanic glass is the main primary phase to dissolve while a combination of kaolinite, smectite and zeolites are likely to precipitate. This information was used to develop a 1D reactive transport model simulating the reactive front with mineral dissolution and precipitation in flow-through column experiments.

[1] De Lucia, Kühn (2021) *Advances in Geosciences* 56, doi.org/10.5194/adgeo-56-33-2021.

[2] Manu, De Lucia, Kühn (2023) *Minerals* 13 (7), doi.org/10.3390/min13070899.