Effects of microstructural changes associated with silicate dissolution on (bio)weathering rates

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Our study aims at evaluating the effects of microstructural and textural changes occurring at the fluid-mineral interface from the mesoscale down to the atomic scale on the (bio)weathering rates of silicates such as labradorite feldspar or olivine under both laboratory and field-relevant conditions.

A combined approach involving vertical scanning interferometry (VSI) and aqueous chemistry analyses revealed that the evolution of feldspar dissolution rates at 80°C and pH = 3 is not captured by current kinetics rate laws, because of microstructural changes occurring at the fluid-mineral interface. Feldspar dissolution is suggested to be controlled by a coupled dissolution-precipitation mechanism at the mineral–fluid interface, as evaluated with focused ion beam-transmission electron microscopy (FIB-TEM) observations combined with cellular automaton modelling. The decline of labradorite dissolution rates may be related to poor transport properties of the amorphous silica surface layers (ASSLs) formed at the reacting interface, as inferred from ion probe (nanoSIMS) and synchrotron-based X-ray reflectivity measurements.

The passivating properties of ASSLs were further tested under field-relevant conditions, using soil solutions collected at the Strengbach critical zone observatory (Aubure, France). Our data show that the reaction rate is controlled by ASSLs, with a modest increase of olivine dissolution rate resulting from bacterial activity. However, microorganisms did not allow to overcome the passivation barrier in the earlier stages of mineral colonization.

Overall, our results indicate that the formation and microstructural evolution of ASSLs act as a likely candidate for the decline in time of silicate dissolution rates frequently reported at the field scale.