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Quantifying the influence of heterogeneity and preferential flow on the scale and time dependence of weathering rates

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Numerous previous laboratory and field observations and inferences of weathering rates suggest significant scale dependence (laboratory rates » field rates) and time dependence (rates appear to decrease progressively with time of exposure). Preferential flow induced by heterogeneity, manifest as permeability variations, macropores, or discrete fractures, has been suggested as one class of extrinsic mechanisms responsible for the observed scale and time dependence. Additional intrinsic mechanisms proposed include the decrease in reactive surface area with weathering. In this research, we present a quantitative evaluation of the influence of heterogeneity and preferential flow on weathering rates using high-resolution reactive transport modeling. We employ PFLOTRAN, an open source subsurface flow and reactive transport code that utilizes parallelization over multiple processors and provides a robust framework for simulating the complex weathering patterns resulting from preferential flow. Simulations were performed in discrete fracture networks (DFNs) and correlated random permeability fields (CRPF). The behavior in these simulations was compared to that in homogeneous permeability fields. The simulations reproduce to some extent the scale and time dependence of weathering rates, although the modeled scale and time dependence are less pronounced than indicated by observations. The simulations in DFNs indicated a systematic time-dependence related to the formation of diffusion-controlled weathering fronts that propagate into matrix blocks. However, the decline of system-averaged weathering rates does not follow a 1/sqrt(time) dependence characteristic of diffusion, due to network scale effects and depletion of matrix blocks. The behavior in CRPF was akin to that in homogeneous permeability fields with enhanced dispersion, with a time-dependence that reflects the advective sweeping of the weathering front from the simulation domain. Our results suggest that structured preferential flow, as resulting from DFNs can readily explain an order-of-magnitude disparity between laboratory and field rates, and a similar decrease with time over $\sim$ 1 million years. However, to explain the nearly four orders of magnitude decrease indicated by observations, preferential flow and diffusion limitations must act in consort with other intrinsic mechanisms.