Vadose Zone Weathering Rates Inferred from U-Series Disequilibrium

Kate Maher[1], Donald J DePaolo[1,2], Carl Steefel[3], John N Christensen[2]

[1]University of California, Berkeley, Department of Earth and Planetary Science, Berkeley, CA 94720-4746, United States

[2]Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, Ca 94720, United States

[3]Geosciences and Environmental Technology Division, Lawrence Berkeley National Laboratory, Livermore, CA 94559, United States

The U-series isotope system can be used to quantify reaction rates in aquifers and thick vadose zone environments. The approach is based on the α-recoil of ^{234}Th atoms across grain boundaries, which enriches the pore fluid in ^{234}U. Dissolution of the solid phase releases mainly 238U to the pore fluid, so that the ^{234}U/^{238}U activity ratio of the pore fluid is a measure of the local ratio of the dissolution U flux to the α-recoil flux. The interpretation of U-series disequilibrium in weathering environments depends on the α-recoil flux, which is highly dependent on both the distribution of U and the grain size distribution of the soil. The recoil length of the ^{234}Th atom in a silicate is approximately 700 $\AA$, therefore the fine-grained material, with a greater surface area to volume ratio, contributes a significant amount of the ^{234}U to the porewater. As a result, estimates of the recoil flux based on the mean grain size may under predict the supply of ^{234}U by over an order of magnitude. In order to quantify the reaction rate, this recoil flux must be accurately measured by consideration of the recoil contribution of the fine-grained material, despite the fact that it may only comprise a small fraction of the total mass of the sediment. U concentrations and high precision isotope measurements (^{234}U/^{238}U) of pore fluids, mineral separates, and grain size intervals are used to constrain reaction rates for a 70 m vadose zone core from eastern Washington State. The pore water ^{234}U/^{238}U activity ratios range from 1.04 to 1.20 in the pore fluids, and from 0.94 to 1.0 in the various size fractions. Data from the solid phases is used to construct a predictive model for α-recoil flux in heterogeneous soil. The calculated recoil loss, which considers each grain size interval, is up to 7 times greater than for a geometric prediction based on the mean grain size. The measured ^{234}U/^{238}U ratios for the vadose zone, in conjunction with the estimates for the a-recoil flux, yield weathering rates of approximately 10$^{-6.4}$ yr^{-1}, which agree with estimates derived from measurements in granitic soils that are based on mineral abundances and soil age. The data suggest that recoil loss from the fine-grained size fractions in a heterogeneous soil can have a significant impact on the interpretation of U isotopic data.