IN-SITU REACTION RATES IN THE VADOSE ZONE AT THE HANFORD SITE INFERRED FROM U SERIES DISEQUILIBRIUM

Katherine Maher (kmaher@uclink4.berkeley.edu) [1,2], Carl Steefel[2], Donald J. DePaolo[1, 3], John N. Christensen[3]. (1) The Department of Earth and Planetary Science, University of California, Berkeley, CA 94720, Fax: 510-642-9520, (2) Environmental Science Division, Lawrence Livermore National Laboratory, (3) Earth Sciences Division, E.O. Lawrence Berkeley National Laboratory.

At the Hanford site, processes within the vadose zone play an important role in determining the chemistry of groundwater and the rate of transport of contaminants to the water table. Models used to describe chemical transport processes in the vadose zone, however, depend on the reaction kinetics—this is particularly true for naturally derived species like U and Sr. However, dissolution rates of sediments are difficult to quantify due to uncertainties associated with both the amount of reactive surface area available in the sediment and the thermodynamic driving forces for the chemical reactions. Mineral dissolution rates measured in the laboratory typically predict rates that are 2 to 4 orders of magnitude faster than estimates based on field measurements. Such experimental rates, which are commonly measured under far-from-equilibrium conditions, are not broadly applicable to many natural systems. Unfortunately, relatively few field measurements of in situ reaction rates are available, and most of those are on silicate soils. Almost no field measurements exist for deep vadose zone materials or for rocks in the saturated zone.

The U-series isotope system can be used to quantify reaction rates in aquifers and thick vadose zone environments. The approach is based on α-recoil of ^{234}Th atoms across grain boundaries, which enriches the pore fluid in ^{234}U. Dissolution of the solid phase releases mainly ^{238}U to the pore fluid, so that the ^{234}U/^{238}U ratio of the pore fluid is a measure of the local ratio of the dissolution U flux to the α-recoil flux.

U concentrations and isotope measurements (U-234/U-238) are combined with a multicomponent reactive transport model with kinetic treatment of reaction rates in order to constrain reaction rates in the vadose zone at the Hanford Site. U measurements for pore waters, bulk solids, exchangeable and selected mineral phases were conducted on an uncontaminated 70 meter vadose zone core within the 200 West area. The pore water ^{234}U/^{238}U activity ratios range from 1.04 to 1.20 in the pore fluids and exchangeable fractions, 1.0 in the bulk solids and from 0.94 to 1.0 in the various size fractions. The measured ^{234}U/^{238}U ratios for the vadose zone core yield weathering rates of approximately $10^{-6.4}$ yr^{-1}, in general agreement with estimates based on other methods.