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Limitations on using U/Th series nuclide systematics for studying subsurface radionuclide transport and groundwater flow

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U- and Th- series nuclides have provided essential tools for studying weathering and subsurface element transport processes. The radionuclides U, Th, Ra, Rn, and Pb have a range of half-lives and contrasting chemical behaviours, and their distribution between subsurface solids and water can be used to quantify rates of soil formation, chemical and mechanical weathering of watersheds, and potentially, subsurface water flow rates. Decay systematics clearly connect the different isotopes, although transfer between different phases and through subsurface systems are generally defined through a series of assumptions that have not been experimentally substantiated. Modification of these assumptions may have significant effects on the conclusions of U/Th series studies. Areas of uncertainty include:

Uniformity of conditions. There are several scales of possible local heterogeneity involving different adsorptive conditions, water redox chemistry, and matrix compositions and grain sizes, from intimately distributed microenvironments to distinct layers. Hydrologic properties control the effects of these zones on surrounding groundwater (or even samples withdrawn by well pumping) that may complicate intepretations of sample chemistry.

Nuclide inputs. Comparing the different isotopes requires relating the input rates from recoil and weathering. The common assumption is that nuclides are released at similar rates by recoil and congruently by weathering. Various theories have been proposed for preferential release of Rn and leaching of radionuclides, although such effects, as well as possible radionuclide fractionation during weathering release, have not been substantiated.

Surface interaction mechanisms. It is generally assumed that radionuclides are removed from water onto surfaces only by reversible adsorption. However, coprecipitation, incorporation into aging secondary mineral structures, and different binding mechanisms can inhibit isotope exchange with nuclides in solution.

Transport mechanisms. Radionuclides may be transported not only as dissolved species, but also on colloids. Colloid-bound species may have different isotope exchange rates as isotopes are released into the water and different transport rates through the subsurface, resulting in separate, distinct populations.

Many of the potential complications to U/Th series systematics may be harnessed for greater understanding of the processes involved in the evolution of soils, aquifers, and watersheds, although sampling procedures, site evaluation techniques, and transport models may need modification. The greatest challenge is in determining how to unravel the evolution of long-lived isotopes that integrate conditions over broader regions.