Variability in porewater $^{224}Ra/^{228}Ra$ ratio and its implications for groundwater tracer studies

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Radium (Ra) forms naturally from the decay of uranium and thorium. There are four naturally occurring Ra isotopes: ^{226}Ra, ^{228}Ra, ^{224}Ra, and ^{223}Ra. These isotopes have extensively been used to infer groundwater flow paths, mixing rates and groundwater discharge to coastal bodies. Due to previously reported spatial and temporal variability in Ra isotope activities in the subsurface, Ra-based hydrogeological applications are prone to be associated with high uncertainties. Numerous studies have invoked the role of geochemical processes and alpha recoil process to explain the observed variability in Ra isotope activities. However, it is difficult to decouple both these processes during the study of variability in Ra isotope activities in the subsurface. Here, we overcame this difficulty by shifting the focus to understanding variability in Ra isotope activity ratios (AR) in the subsurface. Using the ratios cancels the effect of geochemical processes. In this study, we describe a transient box model approach to simulate $^{224}Ra/^{228}Ra$ AR profiles in groundwater. Moreover, both ^{224}Ra and ^{228}Ra are members of the same decay series, and therefore their activities are unaffected by natural distribution of parent nuclide(^{232}Th) in the aquifer. We systematically formulate temporal activity profiles of ^{232}Th decay chain radionuclides within the groundwater and aquifer solid. Our model assumes that the radionuclides between the solid and aqueous phases interact by multiple processes including alpha-recoil, radioactive decay, and adsorption-desorption. The recoil input functions associated with ^{228}Ra and ^{224}Ra based on spatial distribution of their respective parent nuclide were modeled. We simulated the impact of non-equilibrium conditions perturbing the groundwater equilibrium on $^{224}Ra/^{228}Ra$ activity ratios. The simulated AR were compared to field observations of $^{224}Ra/^{228}Ra$ AR to derive insights into groundwater dynamics. Our findings demonstrate that non-equilibrium conditions prevailing in groundwater will always result in $^{224}Ra/^{228}Ra$ AR>1, while $^{224}Ra/^{228}Ra$ AR<1 is a consequence of variability in recoil supply of ^{224}Ra and ^{228}Ra along a flow path. The model results have significant implications for establishing accurate constraints on end member Ra activity, which is the key to deriving accurate estimate of groundwater flux to the coastal oceans.