A reactive transport model for the isotopic patterns recording uranium accumulation and mobiliy

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Reactive transport models are commonly utilized in contaminant hydrology and other short term, accelerated reactive systems, such as the remediation of uranium contaminated groundwater through subsurface biostimulation experiments. These simulations accurately reproduce observed trends in concentration and stable isotope data, and allow researchers to quantify and predict the complex biogeochemical reaction networks governing accumulation, fate, transport and isotopic signatures of heavy metals in the subsurface. This capability implies the potential to quantify and predict shifts in the distribution and magnitude of stable metal isotope ratios beyond the context of short-term, artificially stimulated events. Here we extend reactive transport simulations of uranium redox cycling developed for biostimulation experiments to investigate the stable isotope signatures recording long term uranium contamination and the formation of uranium ore deposits.

The first application focuses on accumulation of uranium in naturally reducing zones, which slowly leach uranium and prolong contamination in a shallow subsurface aquifer. The simulation is run over a 50-year period, representing the time since elevated uranium concentrations first occurred in the aquifer. This decadal scale model generates a clear spatial distribution in accumulated solid phase uranium within the reduced section of the aquifer resulting from ambient rates of biogenic reduction, and a corresponding pattern of solid phase stable isotope ratios indicating spatial heterogeneity from the fringe to the core of the naturally reducing zone. The same reaction network is then extended to millennial timescales to consider the formation of a roll front deposit, illustrating distinct patterns in the spatial distribution and zoning of accumulated uranium stable isotopes. In total these simulations offer a predictive forward modelling capability for the isotopic signatures of metal redox cycling in a wide range of temporal and spatial scales