

Mapping Uranium (U) Movement in Northern Plains Groundwater using Isoscapes

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Uranium (U) is an often-overlooked contaminant in public drinking water systems in the United States, with ~63% of drinking water records from US Environmental Protection Agency (EPA) show detectable U, while average U concentration of ~2% of community water exceeds the EPA's maximum contamination limit (MCL) of 30 µg/L of U¹. High U in public water supplies likely originates from groundwater, known to derive its U from geological sources and potentially contamination from mining. Particularly vulnerable are Native American communities residing in the Northern Plains, reliant on groundwater as one of their primary drinking water source, which has a high prevalence of U above MCL^{2,3}. In collaboration with the Native American communities, we aim to understand the redox processes within aquifers to track the evolution of U concentrations around private wells, and to identify safe zones within the aquifer. Reduction of U(VI) partitions and enriches ²³⁸U isotopes in reduced U(IV) solids, leaving the remaining dissolved U(VI) in groundwater enriched in ²³⁵U. This enrichment can be quantified by measuring the ²³⁸U/²³⁵U ratio, usually expressed as δ²³⁸U. Additionally, groundwater near U deposits inherits its low U activity ratio (²³⁴U/²³⁸U < 1; typically, ~0.9) from localized oxidation of U minerals, which can be used to distinguish local vs. distal U sources. Using this concept, we developed δ²³⁸U and U activity ratio isoscapes in groundwater collected from private wells from three reservations in North and South Dakota for mechanistic understanding about the source proximity and cycling of U. Our preliminary results indicate that U(VI) concentrations ranged from 0.05 to 82 µg/L. In a subset of these samples (n=118), δ²³⁸U ranged from -0.94 to +0.90‰ indicating hotspots of U dissolution and accumulation through a range of redox reactions. The U activity ratios (²³⁴U/²³⁸U) in the same subset of samples vary from 1.53 to 3.07 suggesting distal U source(s) relative to sampled wells indicated by the enrichment of ²³⁴U due to α-decay and U leaching from minerals along the groundwater flow path.