High-precision uranium isotopic studies at the Hanford Site, Washington using MC-ICPMS

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Uranium from nuclear industrial activities covers a wide range of ²³⁵U/²³⁸U and ²³⁶U/²³⁸U due to variable combinations of isotopic enrichment and use in nuclear reactors. In addition, the irradiation of ²³²Th produces ²³³U and thus a signature separate from variable burn-up of different U fuel types. Natural background uranium in groundwater and porewater has essentially constant ²³⁵U/²³⁸U, virtually zero ²³⁶U/²³⁸U and ²³³U/²³⁸U, but variable ²³⁴U/²³⁸U due to alpha recoil effects. The contrasts in isotopic composition between natural and processed uranium, as well as the wide compositional range of processed uranium, provides the means to trace contaminant uranium in the environment and delineate the sources and history of contamination.

We have developed techniques of high precision measurement of uranium isotopes using an ICP source multiple collector magnetic sector mass spectrometer (MC-ICPMS) (IsoProbe, GV Instruments Ltd.).
U isotopic compositions are measured simultaneously using a combination of Faraday cups (for ²³⁵U and ²³⁸U) and a Daly photomultiplier ion counting system (for ²³⁴U, ²³⁶U and ²³³U in separate measurements). U is separated from samples prior to introduction to the MC-ICPMS via a desolvation system. A single analysis of a 20ppb U solution uses ~10ng of sample U. We use bracketing analyses of a natural secular equilibrium U standard to correct instrumental mass fractionation, establish Daly/Faraday gain, and account for peak-tailing on ²³⁶U. This allows us to avoid the use of a ²³³U -²³⁶U double spike for mass fractionation correction that would compromise our ability to measure ²³⁶U and ²³³U. The lower limit for ²³⁶U/²³⁸U measurement is about 2x10⁻⁸. For 1ppb U in a water sample, this represents 5x10⁷ atoms ²³⁶U per liter.

As demonstrations of our techniques we will present data from several ongoing studies at the Hanford Site, where decades of nuclear related activities have left significant local U contamination, including: (1) investigation of the connection between groundwater and vadose zone contamination in the B-BX-BY Waste Management Area (WMA) (Christensen et al. (2004) Env. Sci. Tech., 38:3330) (2) behavior of vadose zone U contamination in a core from the T WMA (3) sourcing, apportioning and tracing the contribution of the Hanford Site to the U flux of the Columbia River.

High-precision uranium isotopic studies for environmental forensics at the Hanford Site, Washington using MC-ICPMS