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Demonstration studies at the Hanford Site, Washington

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

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The contrasts in isotopic composition between natural and anthropogenic uranium and the wide variation in the composition of different processed uranium sources, promotes the measurement of uranium isotopic composition as a fingerprint and tracer of uranium contamination in the environment. Previous studies mainly have focused on the use of only one of the isotope ratios of U, e.g. $^{234}\text{U}/^{238}\text{U}$, $^{238}\text{U}/^{235}\text{U}$ or $^{236}\text{U}/^{238}\text{U}$. We measure all three of these ratios in environmental samples in order to better distinguish, characterize, identify and apportion U sources. For our U isotopic measurements, we employ an IsoProbe (GV Instr. Inc.) multiple-collector ICP source magnetic sector mass spectrometer. U isotopic compositions are measured simultaneously using a combination of Faraday cups (for ^{235}U and ^{238}U) and a Daly photomultiplier ion counting system (for ^{234}U and ^{236}U in two separate analyses). U is separated from samples (e.g. vadose zone pore water, groundwater, rock/soil samples) prior to introduction to the MC-ICPMS via a desolvation system. At 7E-11 amps of ^{238}U ions, a single analysis of a 20ppb U solution uses ~10ng of sample U. For correction of instrumental mass fractionation, we use bracketing analyses of a natural secular equilibrium U standard. This allows us to avoid the use of a double ^{233}U - ^{236}U spike for mass fractionation correction that would compromise our ability to measure $^{236}\text{U}/^{238}\text{U}$. We also use the standard analyses for Daly/Faraday gain and for peak-tail correction of the ^{236}U analyses. Typical precision for $^{238}\text{U}/^{235}\text{U}$ is $\leq 0.05\%$ 2s, while for $^{234}\text{U}/^{238}\text{U}$ it is $\leq 0.15\%$ 2s. Precision for $^{236}\text{U}/^{238}\text{U}$ is $\leq 0.15\%$ 2s down to the 10^{-7} range where precision degrades by a factor of ten. The limit for $^{236}\text{U}/^{238}\text{U}$ measurement is about 2×10^{-8} , only ~five times higher than accelerator MS. For 1ppb U, this represents 5×10^7 atoms ^{236}U per liter water.

We will present three ongoing studies at the Hanford site as a demonstration of our techniques: (1) investigation of groundwater and vadose zone contamination in the B-BX-BY WMA (Christensen et al. (2004) Env. Sci. Tech., in press) (2) signatures of vadose zone contamination in waste cribs near U-Plant and (3) investigation of U contamination of the Columbia River.

New approach and technique for tracing uranium in the environment.