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Heavy-element accelerator mass spectrometry (AMS) provides a high sensitivity, accurate, and robust technique for the measurement of actinide elements at ultra-low concentrations. The development of heavy element AMS at the Lawrence Livermore National Laboratory (LLNL) has centered on plutonium and uranium isotope applications in human health and exposure, long-term environmental stewardship, and source-term characterization. These studies have shown that AMS has a sufficiently high abundance sensitivity to measure low abundance isotopes in the presence of mass "m–1" and "m+1" ions generated from principal isotopes of the element of interest. In many cases the isotopic signature ratios of the low-abundance isotopes provide more sensitive and accurate *fingerprint* information for assessing sources of contamination in either the workplace or the environment. This is especially true of uranium where the major isotopes associated with an anthropogenic source will be rapidly diluted by the natural uranium background. Uranium-238 (^{238}U) and uranium-235 (^{235}U) are primordial radionuclides with half-lives of 4.5×10^9 and 7.04×10^8 years, respectively. Uranium-234 (^{234}U) (half-life = 2.45×10^8 years) is usually present in the environment in radioactive equilibrium with ^{238}U, with a relative natural abundance of about 0.005 atom %. Uranium-236 (^{236}U) (half-life = 2.34×10^7 years) also occurs in nature but at ultra-low concentrations of about 10^{-16} atom %. However, enhanced levels of ^{236}U are produced in nuclear reactors by neutron irradiation of ^{235}U. Within the United States a significant fraction of the uranium used in industry or nuclear weapons production is derived from reprocessed nuclear fuel and, as such, potentially carries signature ratios of ^{236}U/^{238}U or ^{236}U/^{235}U that are readily distinguishable from the natural uranium background. We report on the results of an external interlaboratory exercise involving low-level uranium isotope measurements in synthetic urine, and the preliminary findings of an initial study showing the presence of ^{236}U in bioassay samples collected from LLNL workers potentially exposed to uranium.

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