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DETECTION AND QUANTIFICATION OF ACTINIDES BY ACCELERATOR MASS SPECTROMETRY

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Accelerator mass spectrometry (AMS) is a highly sensitive and robust atom counting technique. It is ideal to measure very small amounts of long-lived radionuclides that otherwise could not be analyzed by conventional radiometric methods. AMS is characterized by high rejection of molecular interferences and low susceptibility to matrix components. At LLNL, we have developed a routine AMS capability for the measurement of plutonium concentrations and isotopic ratios in environmental samples including soils, sediments, waters, and human urine. The high-throughput design of the LLNL facility combined with AMS reduced demands on sample preparation result in a cost-effective method with rapid turn around of results. Hundreds of environmental and bioassay samples have already been measured for Pu-239 and Pu-240. The lower limit for quantification is about 1E+06 atoms/sample with a linear response expanding to >1E+11 atoms/sample; recent tests on U-236 and Np-237 have produced similar results. Current studies indicate that acute exposure to plutonium results in trace quantities in urine that are detectable by AMS for months to years following exposure. In particular, archived samples for one plutonium worker exposed by a puncture wound, and another exposed via inhalation were used to reconstruct elimination kinetics. Results obtained by AMS for the first worker revealed an unexpected chronic, nearly linear increase in urinary Pu-239 activity consistent with leakage from the original accidental puncture site. Plutonium was not detected in either individual by conventional methods, whereas AMS clearly established the existence and an approximate wound date for the puncture-wound plutonium source. This study shows that urine samples can be used to establish exposure to plutonium many years following exposure. Daily urinary excretions of plutonium in the general population should be about 1E+06 atoms, i.e., AMS current quantification limit. Accordingly, incidental exposure to plutonium is expected to result in concentrations in urine that can be detected with existing AMS capabilities.

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