

Analysis of High-Fired Plutonium Oxide and Other Actinides in MAPEP Soil Samples

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Recently, the Radiological and Environmental Sciences Laboratory (RESL) presented results of the Mixed Analyte Performance Evaluation Program (MAPEP) for actinides in soil. In this laboratory evaluation test, substantial number of participants failed on ²³⁹Pu analysis. It has been concluded that incomplete sample digestion and analyte dissolution was a primary cause of failure. The United States Transuranium and Uranium Registries (USTUR) performed a study to verify USTUR standard procedures used for actinide analysis in human tissues. MAPEP-11-MaS24 soil was received by the USTUR. This reference soil was certified for ²³⁹Pu, ²⁴¹Am, ²³⁴U, and ²³⁸U concentrations and plutonium was presented in the form of 'high-fired' ²³⁹PuO₂. Samples of 0.5, 1.0, and 2.0 g were collected in triplicate and were microwave digested at a control temperature of 200 °C and monitored pressure of 40 bar for 20 minutes using concentrated HNO₃-HCl-HF or HNO₃-HCl reagent mixture. After a sample was spiked with tracers, actinide (Pu/U/Am) separation was carried out on TEVA-TRU-DGA extraction chromatographic column consisting of 1-ml cartridges. Following electrodeposition, activities of Pu, U, and Am fractions were measured by α-spectrometry. Regardless of sample size, complete soil digestion was achieved with HNO₃-HCl-HF, while HNO₃-HCl yielded insoluble residue. The average chemical tracer recoveries were 34.3±6.2%, 100.8±1.7%, and 98.3±4.9% for ²⁴³Am, ²⁴²Pu, and ²³²U, respectively. Low ²⁴³Am recovery was attributed to the soil matrix effect. Measured average concentrations of ²³⁹Pu, ²⁴¹Am, ²³⁴U, and ²³⁸U in samples digested with HNO₃-HCl-HF were 2.54±0.16, 1.64±0.12, 4.41±0.09, and 4.61±0.11 pCi g⁻¹, respectively. These values were not statistically different from certified ²³⁹Pu, ²⁴¹Am, ²³⁴U, and ²³⁸U concentrations with negative bias of the mean of 4.0%, 1.3%, 7.2%, and 7.0%, respectively. Using HNO₃-HCl, only 5% (0.14±0.01 pCi g⁻¹) of the initial ²³⁹Pu was dissolved and recovered from the solution. The remaining residue was digested with HNO₃-HCl-HF and ²³⁹Pu concentration was calculated to be 2.42±0.10 pCi g⁻¹ resulting in a combined value of 2.56±0.10 pCi g⁻¹ (bias -3.4%). Negative bias of 77.6%, 72.1%, and 71.9% for ²⁴¹Am, ²³⁴U, and ²³⁸U, respectively, was also observed with HNO₃-HCl. Incomplete dissolution of the analyte using HNO₃-HCl results in a low actinide recovery from the sample, while HNO₃-HCl-HF allows for complete dissolution of all actinides present in soil, including high-fired plutonium oxide.

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