

ANALYSIS OF HIGH-FIRED PLUTONIUM OXIDE AND OTHER ACTINIDES IN MAPEP SOIL SAMPLES

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Topics: Radiochemical separations; Other topics relevant to radiobioassay and radiochemical measurements

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 ^{239}Pu 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 ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U concentrations and plutonium was presented in the form of 'high-fired' $^{239}\text{PuO}_2$. 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 $\text{HNO}_3\text{-HCl-HF}$ or $\text{HNO}_3\text{-HCl}$ 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 $\text{HNO}_3\text{-HCl-HF}$, while $\text{HNO}_3\text{-HCl}$ yielded insoluble residue. The average chemical tracer recoveries were $34.3\pm 6.2\%$, $100.8\pm 1.7\%$, and $98.3\pm 4.9\%$ for ^{243}Am , ^{242}Pu , and ^{232}U , respectively. Low ^{243}Am recovery was attributed to the soil matrix effect. Measured average concentrations of ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U in samples digested with $\text{HNO}_3\text{-HCl-HF}$ 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 ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U concentrations with negative bias of the mean of 4.0%, 1.3%, 7.2%, and 7.0%, respectively. Using $\text{HNO}_3\text{-HCl}$, only 5% (0.14 ± 0.01 pCi g⁻¹) of the initial ^{239}Pu was dissolved and recovered from the solution. The remaining residue was digested with $\text{HNO}_3\text{-HCl-HF}$ and ^{239}Pu 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 ^{241}Am , ^{234}U , and ^{238}U , respectively, was also observed with $\text{HNO}_3\text{-HCl}$. Incomplete dissolution of the analyte using $\text{HNO}_3\text{-HCl}$ results in a low actinide recovery from the sample, while $\text{HNO}_3\text{-HCl-HF}$ allows for complete dissolution of all actinides present in soil, including high-fired plutonium oxide.