Accelerator Mass Spectrometric (AMS) Measurements of Plutonium Activity Concentrations and $^{240}\text{Pu}/^{239}\text{Pu}$ Atom Ratios in Soil Extracts Supplied by the Carlsbad Environmental Monitoring & Research Center

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Final Report

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Accelerator Mass Spectrometric (AMS) measurements of plutonium activity concentrations and $^{240}$Pu/$^{239}$Pu atom ratios in soil extracts supplied by the Carlsbad Environmental Monitoring & Research Center.

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Summary

Plutonium-239 ($^{239}$Pu) and plutonium-239+240 ($^{239+240}$Pu) activities concentrations and $^{240}$Pu/$^{239}$Pu atom ratios are reported for a series of chemically purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center (CEMRC) in New Mexico. Samples were analyzed without further purification at the Lawrence Livermore National Laboratory (LLNL) using accelerator mass spectrometry (AMS). This report also includes a brief description of the AMS system and internal laboratory procedures used to ensure the quality and reliability of the measurement data.
1.0 Materials and Methods

Each sample (N=20) received for analysis consisted of a dried residue contained in a small quartz vial supplied by Lawrence Livermore National Laboratory (LLNL). The samples were supplied blind to LLNL researchers but were known to be prepared from large volume soil extracts after acid leaching and ion-exchange chromatography. At the request of LLNL researchers, 2 samples were prepared and identified as process blanks. All samples contained a known amount of plutonium-242 (242Pu) (~ca. 3.5×10^9 atoms) added as an isotope dilution spike. The spike information was used to calculate the amount of plutonium present in each sample split. These data should not be confused with any measurement data on the total amount of plutonium present in the original soil samples.

The dried sample residues were rehydrated in nitric acid and the material transferred to plastic centrifuge tubes. Plutonium was coprecipitated on a small quantity of Fe hydroxide, the precipitate transferred to a quartz vial and ignited at 800°C. The material was then packed into an aluminum AMS target with about 3 mg of added niobium (Nb) metal powder to enhance ion production. The analytical scheme included analysis of Fe-Nb target blanks (N=6) as well as Certified Reference Materials, (CRM) 128 (239Pu/242Pu = 0.9993 ± 0.0003) and CRM 138 (240Pu/239Pu = 0.0863 ± 0.0001), traceable to the U.S. National Institute of Standards and Technology (NIST).

The heavy ion AMS system used for low-level plutonium isotope measurements at the Center for Accelerator Mass Spectrometry (CAMS) at LLNL is shown in Figure 1.

![Figure 1. Heavy isotope accelerator mass spectrometry system at LLNL (Brown et al., 2004)](image)

Negative plutonium ions (e.g., 239-244Pu16O-) are produced from sample targets using a cesium splutter source and injected into the accelerator through a 90° electrostatic analyzer. Negative ions entering the accelerator are stripped, converted to positive ions and accelerated into the high-energy spectrometer of the AMS system.

The high-energy spectrometer was designed to selectively transport 5+ ions to the detector using electrostatic deflection of desired ions through image slits coupled with fast mass switching of the low-energy
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spectrometer and quasi-continuous normalization to the reference isotope (e.g., $^{242}\text{Pu}^{5+}$). Ions are counted on a two-anode, longitudinal field gas ionization detector with sufficient resolution to allow clean rejection of interfering ions at 4+ and lower charge states.

2.0 RESULTS

$^{239}\text{Pu}$ and $^{239+240}\text{Pu}$ activity concentrations and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in each of the 20 samples received from CEMRC are reported in Table 1. The results are expressed in units of μBq per sample (±1 standard deviation) based on the $^{242}\text{Pu}$ spike information supplied to LLNL researchers. Some samples produced unusually high residual ion-currents but this phenomenon does not appear to have affected data quality.

The total measurement efficiency for the analysis was about $\sim 5 \times 10^{-5}$ with observed plutonium background concentrations based on Fe-Nb blank targets falling below $\sim 1 \times 10^5$ atoms. The Minimal Detectable Activity (MDA) for the measurement system at the time of analysis was estimated to be around $4 \times 10^{-4}$ μBq and $2 \times 10^{-3}$ μBq of $^{239}\text{Pu}$ and $^{239+240}\text{Pu}$, respectively.

Measurement data for AMS targets prepared from certified CRMs were all within stated uncertainties and showed no evidence of mass-dependent biases. For example, the mean weighted measured versus expected (theoretical) ratios of $^{239}\text{Pu}/^{242}\text{Pu}$ (N=12) in CRM 128 and $^{240}\text{Pu}/^{239}\text{Pu}$ (N=6) in CRM 138 were $1.006 \pm 0.004$ and $1.014 \pm 0.014$, respectively.

3.0 CONCLUSION

A series of purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center were analyzed by accelerator mass spectrometry at the Lawrence Livermore National Laboratory. All test samples contained measurable quantities of plutonium in excess of that contained in quality control blank targets. Based on supplementary quality control analyses, it is expected that the accuracy and precision of all measurements performed under this agreement are within the stated uncertainties.

4.0 ACKNOWLEDGEMENT

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REFERENCES


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Accelerator Mass Spectrometric (AMS) Measurements of Plutonium Activity Concentrations and $^{240}$Pu/$^{239}$Pu Atom Ratios in Soil Extracts Supplied by the Carlsbad Environmental Monitoring & Research Center.

Table 1. $^{239}$Pu and $^{239+240}$Pu activity concentrations and $^{240}$Pu/$^{239}$Pu atom ratios in purified soil extracts received from the Carlsbad Environmental Monitoring & Research Center (CEMRC).

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Sample Description</th>
<th>$^{239}$Pu (mBq, sample received)</th>
<th>$^{239+240}$Pu (mBq, sample received)</th>
<th>$^{240}$Pu/$^{239}$Pu atom ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>RFC631</td>
<td>CEMRC NMSU; 105851</td>
<td>1.364 ± 0.017</td>
<td>1.464 ± 0.018</td>
<td>0.074 ± 0.003</td>
</tr>
<tr>
<td>RFC632</td>
<td>CEMRC NMSU; 105852</td>
<td>0.424 ± 0.010</td>
<td>0.456 ± 0.010</td>
<td>0.075 ± 0.006</td>
</tr>
<tr>
<td>RFC633</td>
<td>CEMRC NMSU; 105853</td>
<td>3.373 ± 0.038</td>
<td>3.609 ± 0.039</td>
<td>0.070 ± 0.002</td>
</tr>
<tr>
<td>RFC634</td>
<td>CEMRC NMSU; 105854</td>
<td>0.283 ± 0.008</td>
<td>0.315 ± 0.008</td>
<td>0.113 ± 0.009</td>
</tr>
<tr>
<td>RFC635</td>
<td>CEMRC NMSU; 105855</td>
<td>0.246 ± 0.006</td>
<td>0.270 ± 0.007</td>
<td>0.100 ± 0.008</td>
</tr>
<tr>
<td>RFC636</td>
<td>CEMRC NMSU; 1345_TB</td>
<td>0.034 ± 0.003</td>
<td>0.036 ± 0.003</td>
<td>0.084 ± 0.021</td>
</tr>
<tr>
<td>RFC637</td>
<td>CEMRC NMSU; 105856</td>
<td>0.364 ± 0.009</td>
<td>0.421 ± 0.010</td>
<td>0.156 ± 0.010</td>
</tr>
<tr>
<td>RFC638</td>
<td>CEMRC NMSU; 105857</td>
<td>0.317 ± 0.008</td>
<td>0.366 ± 0.009</td>
<td>0.154 ± 0.011</td>
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<tr>
<td>RFC639</td>
<td>CEMRC NMSU; 105858</td>
<td>0.510 ± 0.010</td>
<td>0.595 ± 0.011</td>
<td>0.168 ± 0.008</td>
</tr>
<tr>
<td>RFC640</td>
<td>CEMRC NMSU; 105859</td>
<td>1.447 ± 0.023</td>
<td>1.617 ± 0.025</td>
<td>0.118 ± 0.006</td>
</tr>
<tr>
<td>RFC641</td>
<td>CEMRC NMSU; 1346_LCS</td>
<td>1.593 ± 0.019</td>
<td>1.607 ± 0.019</td>
<td>0.009 ± 0.001</td>
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<tr>
<td>RFC642</td>
<td>CEMRC NMSU; 105860</td>
<td>0.447 ± 0.009</td>
<td>0.526 ± 0.010</td>
<td>0.177 ± 0.009</td>
</tr>
<tr>
<td>RFC643</td>
<td>CEMRC NMSU; 105861</td>
<td>0.508 ± 0.011</td>
<td>0.607 ± 0.012</td>
<td>0.193 ± 0.010</td>
</tr>
<tr>
<td>RFC644</td>
<td>CEMRC NMSU; 105862</td>
<td>0.727 ± 0.013</td>
<td>0.863 ± 0.014</td>
<td>0.186 ± 0.008</td>
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<tr>
<td>RFC645</td>
<td>CEMRC NMSU; 105863</td>
<td>0.258 ± 0.007</td>
<td>0.302 ± 0.007</td>
<td>0.172 ± 0.011</td>
</tr>
<tr>
<td>RFC646</td>
<td>CEMRC NMSU; 105864</td>
<td>0.589 ± 0.011</td>
<td>0.690 ± 0.012</td>
<td>0.173 ± 0.008</td>
</tr>
<tr>
<td>RFC647</td>
<td>CEMRC NMSU; 105865</td>
<td>0.318 ± 0.009</td>
<td>0.365 ± 0.010</td>
<td>0.146 ± 0.012</td>
</tr>
<tr>
<td>RFC648</td>
<td>CEMRC NMSU; 105866</td>
<td>0.371 ± 0.008</td>
<td>0.431 ± 0.008</td>
<td>0.164 ± 0.009</td>
</tr>
<tr>
<td>RFC649</td>
<td>CEMRC NMSU; 105867</td>
<td>0.418 ± 0.011</td>
<td>0.496 ± 0.012</td>
<td>0.188 ± 0.013</td>
</tr>
<tr>
<td>RFC650</td>
<td>CEMRC NMSU; 1348_TB</td>
<td>0.002 ± 0.001</td>
<td>0.002 ± 0.001</td>
<td>0.003 ± 0.001</td>
</tr>
</tbody>
</table>

Note: RFC647 and RFC648 have very low activity concentrations and may not be significant.

LLNL Sample ID: Carlsbad Environmental Monitoring & Research Center (CEMRC); NMSU: National美s University; LCS: Livermore. Sample ID numbers are 105851 to 105867.