Rapid Separation Method for Actinides in Emergency Soil Samples

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Abstract

A new rapid method for the determination of actinides in soil and sediment samples has been developed at the Savannah River Site Environmental Lab (Aiken, SC, USA) that can be used for samples up to 2 grams in emergency response situations. The actinides in soil method utilizes a rapid sodium hydroxide fusion method, a lanthanum fluoride soil matrix removal step, and a streamlined column separation process with stacked TEVA, TRU and DGA Resin cartridges. Lanthanum was separated rapidly and effectively from Am and Cm on DGA Resin. Vacuum box technology and rapid flow rates are used to reduce analytical time. Alpha sources are prepared using cerium fluoride microprecipitation for counting by alpha spectrometry. The method showed high chemical recoveries and effective removal of interferences. This new procedure was applied to emergency soil samples received in the NRIP Emergency Response exercise administered by the National Institute for Standards and Technology (NIST) in April, 2009. The actinides in soil results were reported within 4-5 hours with excellent quality.

Introduction

There is an increasing need to develop faster analytical methods for emergency response, including emergency soil and air filter samples [1,2,3]. There are a number of analytical methods reported that use ion exchange/extraction chromatography plus alpha spectrometry to determine actinides in soil. Hou et al surveyed a wide range of separation methods for Pu in waters and environmental solid samples [4]. Methods included varied combinations of ion exchange and/or extraction chromatographic techniques. Chemical recoveries for Pu typically varied between 40-85%.

Vajda et al reported an interesting method for actinides in soil in which 0.5 g soil samples are fused using lithium metaborate. After preconcentration of actinides using calcium fluoride precipitation, a TRU Resin separation is performed [5]. Tests on IAEA soil standards showed very good results, but tracer recoveries showed some significant variation, as high as 80% for plutonium, but notably as low as 20-30% for uranium. The method could, however, be completed rapidly, but still required ~ 24 hours. The authors discuss the challenge of performing the separation of multiple actinides on a single resin. Using only one resin to separate multiple actinides can result in fraction contamination for difficult sample matrices, especially if high levels of actinides are present. The overall results were very good relative to soil reference values, however, two of the IAEA soil samples tested showed relatively low chemical recoveries for Pu and U. The authors concluded that the lower chemical recoveries were caused by the soil matrix. This may have resulted from using only 1 ml of TRU Resin to recover all the actinides from the soil sample, even though the soil aliquot was only 0.5 g.

Wang et al reported a sequential method to determine actinides and strontium in soil samples [6]. The samples were digested in nitric acid and hydrogen peroxide, and redissolved in a large volume of 3M nitric acid. A large anion resin column (Dowex 1x8) was used to collect and separate Pu and Th. The rinse fractions from the anion resin were treated further and processed individually for Am, U and Sr. Several sequential precipitations were carried out. An oxalate precipitation was performed at pH 4.2 on the anion resin rinse solution followed by a Sr Resin separation. A separate oxalate precipitation at pH 1.5 was performed on the supernatant after the first oxalate precipitation to recover Am and separate on TRU Resin. The supernatant from the second oxalate precipitation was passed through a large amount of Chelex 100 resin to collect uranium and purify uranium. Strontium was counted using Cerenkov counting, while all actinide fractions were electrodeposited for counting by alpha spectrometry. The chemical recoveries using this method on NRIP soil were as follows: plutonium (60-76%), americium (40-59%), uranium (57-76%), and Strontium (63-77%). A large number of sequential steps were required, but the accuracy of the actinide and strontium results versus the NIST reference values was very good.

Eikenberg et al compared three different separation methods to determine actinides in soil samples [7]. Samples were leached in 8M nitric acid, filtered and a calcium oxalate precipitation technique that settles overnight was used to preconcentrate actinides. Leaching samples does not fully recover uranium and thorium in the soil and does not effectively digest refractory particles. Sill et al have emphasized the need for total dissolution soil methods [8]. One of the separation methods tested was anion resin (AG-1X2) plus DGA Resin (Eichrom Technologies, Lyle, IL, USA). Am, Cm, and U were collected on DGA Resin, which has a *k*’ of approximately 30,000 for Am [9]. The sample was loaded in 3M nitric acid, and uranium was eluted in 0.25M nitric acid, prior to stripping Am and Cm with 0.2M HCL. It was not clear what the uranium tracer recoveries were for these samples, but DGA resin is not typically used to recover and purify uranium because the *k*’ in 3M HNO3 for U on DGA Resin is only about 20. Electrodeposition was used to prepare alpha sources for measurement by alpha spectrometry.

We have previously used TEVA Resin plus TRU Resin plus DGA Resin in the SRS Environmental Laboratory to determine actinides in 5 gram soil samples. TRU Resin is used in tandem with DGA Resin to effectively recover uranium and provide very high chemical recoveries for Am and Cm. The method first uses, nitric acid –hydrofluoric acid to digest samples and remove silica, then uses an alkaline fusion to digest the samples, an iron hydroxide precipitation, followed by a cerium fluoride preconcentration step to remove the sample matrix and preconcentrate the actinides. Cerium fluoride microprecipitation is used to prepare the alpha sources for counting by alpha spectrometry. This method, after initial heating of samples to 550C in a furnace, requires about 12 hours of sample preparation work to get samples ready for alpha counting. Chemical recoveries for Pu, Am and U of greater than 90% were reported [10]. A version of this method to determine Pu and Am in 200 gram soil samples using acid leaching only was also reported [11].

A new method has been developed in the Savannah River Site Environmental Lab (Aiken, SC, USA) that has reduced the sample preparation time for 1-2 gram samples to ~3 hours. A new rapid method for the determination of actinides in soil and sediment samples has been developed that can be used for samples up to 2 grams in emergency response situations.

The Savannah River Site Environmental Lab participated in the 2009 NRIP Emergency Response program administered by the National Institute for Standards and Technology (NIST) in April, 2009. Previous work had concentrated on rapid actinide and ^{89,90}Sr methods for emergency water and urine samples [12]. The NRIP soil samples were analyzed using a rapid sodium hydroxide fusion, followed by precipitation steps including a lanthanum fluoride matrix removal step, followed by a stacked column consisting of TEVA Resin + TRU Resin + DGA Resin. Lanthanum, which follows Am on TRU Resin and DGA Resin, was removed on DGA Resin using a dilute nitric acid rinse. Lanthanum was separated rapidly and effectively from Am and Cm on DGA Resin. Vacuum box technology and rapid flow rates are used to reduce analytical time. Alpha sources are prepared using cerium fluoride microprecipitation for counting by alpha spectrometry. The method showed high chemical recoveries and effective removal of interferences.

Experimental

Reagents

The resins employed in this work are TEVA Resin® (Aliquat ™336), TRU-Resin® (tri-n-butylphosphate (TBP) and octyl (phenyl) N,N diisobutylcarbamoylmethylphosphine oxide (CMPO)), and DGA Resin (N,N,N',N' tetraoctyldiglycolamide), available from Eichrom Technologies, Inc., (Darien, Illinois, USA). Nitric, hydrochloric and hydrofluoric acids were prepared from reagent-grade acids (Fisher Scientific, Inc.). All water was obtained from a Milli-Q2™ water purification system. All other materials were ACS reagent grade. Radiochemical isotope tracers ^{242}Pu, ^{243}Am, and ^{232}U that were obtained from Analytics, Inc. (Atlanta, GA, USA) and diluted to the approximately 0.37 Bq ml^{-1} level were employed to enable yield corrections. ^{232}U tracer was prepared to be self-cleaning, removing its ^{228}Th daughter using barium sulfate precipitation [13].

Procedures

Column preparation. TEVA, TRU, and DGA-Resin columns were obtained as cartridges containing 2 ml of each resin from Eichrom Technologies, Inc.. Small particle size (50-100 micron) resin was employed, along with a vacuum extraction system (Eichrom Technologies). Flow rates of 1-2 ml min^{-1} are typically used in the SRS Environmental Laboratory, but flow rates ~ 2 times faster were used for this work.

Sample Preparation. The NRIP soil samples (1g) were added to 250 ml zirconium crucibles (Metal Technology, Inc., Albany, OR, USA). Tracers were added to each crucible and the samples were dried briefly on a hot plate.

After removing the crucibles and allowing them to cool, 15 grams of sodium hydroxide were added to each crucible. The crucibles were covered with a zirconium lid and placed into a furnace at 600C for ~ 10 minutes.

After removing the crucibles from the furnace, they were cooled for about 10 minutes, transferred to a hot plate and water was added to transfer the solids to 225 ml centrifuge tubes. The residual solids were removed from the crucibles by adding water and heating the crucibles on the hot plate as needed. One hundred and twenty-five milligrams of iron (added as ferric nitrate) and six milligrams of lanthanum (as lanthanum nitrate) were added to 225 ml centrifuge tubes prior to transferring the alkaline solids from the crucibles into the tubes. The samples were diluted to 180 ml with water. Five milliliters of 20% titanium chloride were added to each tube, followed by 1 ml of 10% barium nitrate to complex any carbonate present. The tubes were centrifuged at 3500 rpm for 5 minutes and the supernatant was discarded. The remaining solids were dissolved in a total volume of ~60 ml of 1.5 M HCl. This solution was diluted to ~170 ml with 0.01M HCL. Two milligrams of lanthanum as lanthanum nitrate were added to each sample. To ensure no actinides were in the hexavalent state and facilitate complete precipitation, 5 milliliters of 20% titanium chloride were added to each sample. Twenty milliliters of 28M hydrofluoric acid were added to each sample. The samples were placed on ice for ~10 minutes to reduce solubility and centrifuged for 10 minutes at 3500 rpm. The supernatant was removed and the residual solids containing the actinides were dissolved in 5 ml of warm 3M HNO3-0.25M boric acid, 6 ml of 7M HNO3 and 7 ml of 2 M aluminum nitrate. The solids were transferred to 100 ml teflon beakers during this step and warmed to redissolve the solids. The aluminum nitrate was previously scrubbed to remove trace uranium by passing approximately 250 ml of 2M aluminum nitrate through a large column (Environmental Express, Mount Pleasant, SC, USA) containing 7 ml of UTEVA Resin (Eichrom Technologies) at ~10 to 15 ml per minute. The columns were prepared from a water slurry of the UTEVA Resin.

Column separation. TEVA, TRU, and DGA-Resin cartridges were stacked on the vacuum box from top to bottom, in that order. Fifty milliliter centrifuge tubes were used to collect rinse or final purified fractions. Column load solutions were loaded at ~1-2 drops per second, rinse solutions at ~3 drops per second and column strip solutions were added at ~2 drops per second using vacuum.

A valence adjustment was performed by adding 0.5 ml 1.5M sulfamic acid and 1.25 ml 1.5M ascorbic acid with a three minute wait step to reduce plutonium to Pu^{3+}. Np was not determined for these NRIP samples, but it should be noted that if ^{237}Np measurement is needed, it can assayed along with Pu in the purified Pu fraction if ^{236}Pu tracer is used

[14]. If ^{237}Np separation is desired, 0.4 ml 5 mg/ml Fe as ferric nitrate can be added to facilitate ^{237}Np reduction to Np^{4+}. The ferric ions are reduced to ferrous ions by the ascorbic acid, which reduces Np effectively to Np^{4+}. Following the reduction step, 1 ml 3.5M sodium nitrite was added to oxidize plutonium to Pu^{4+}. After the valence adjustment, the sample solution was loaded onto the stacked column at approximately ~2 drops per second. After the sample was loaded, a beaker rinse of ~5 ml 3M HNO_3 was transferred to the stacked column and a rinse of 10 ml 3M HNO_3 was added directly to the stacked column. The TRU Resin and DGA-Resin cartridges were removed and the TEVA cartridges were kept on the vacuum box. The TEVA cartridge was rinsed with 10 ml 3M nitric acid to remove sample matrix components. To elute thorium from TEVA Resin, 20 ml 9M hydrochloric acid were added and discarded. A 5 ml volume of 3M HNO3 was added to TEVA Resin (and discarded) to ensure complete removal of sample matrix components.

The plutonium was stripped from TEVA Resin with 20 ml 0.1M hydrochloric acid-0.05M hydrofluoric acid –0.01M titanium (III) chloride (freshly prepared)**.** Fifty micrograms of cerium as cerium nitrate were added to the tubes, along with 1 ml of concentrated hydrofluoric acid (49%), prior to elution of the plutonium to reduce microprecipitation wait times. A 0.5 ml volume of 30 wt% hydrogen peroxide was added after the plutonium was eluted to oxidize any residual uranium to U^{6+} as a precaution. After waiting 10 minutes, the solutions were filtered onto 0.1 micron 25 mm polypropylene filters and counted by alpha spectrometry.

The DGA Resin cartridges were placed on a separate vacuum box and processed at the same time as the TEVA Resin cartridges to save time. The DGA Resin cartridges were rinsed with 5 ml of 0.1M HNO_3 at ~2 drops per second to remove any residual uranium that may have passed through TRU Resin. The TRU Resin cartridges were placed above each DGA Resin cartridge. Am was stripped from TRU Resin with 15 ml 3M HCl at ~1-2 drops per second onto DGA Resin. The TRU Resin cartridges were removed. DGA Resin cartridges (alone) were rinsed with 5 ml 3M HCL, 3 ml 1M HNO_3, and 20 ml 0.05M HNO_3 at 1-2 drops per second to remove lanthanum. Am was stripped from DGA Resin with 10 ml 0.25M HCL into clean tubes at ~1-2 drops per second. Cerium was added as previously described to the tubes, along with 1 ml of concentrated hydrofluoric acid (49%), prior to elution. After waiting 10 minutes, the solutions were filtered onto 0.1 micron 25 mm polypropylene filters (Resolve® filter-Eichrom Technologies) and counted by alpha spectrometry.

TRU Resin was rinsed with 15 ml 4M HCl-0.2M HF-0.002M $TiCl_3$ to remove any residual thorium and polonium that may have passed through TEVA and been retained on TRU Resin at ~2-3 drops per second. After the 4M HCl-0.2M HF-0.002M $TiCl_3$ rinse was added to TRU Resin, 5 ml 8M HNO3 was added to TEVA Resin and this rinse was discarded.

Uranium was stripped from TRU Resin using 15 ml 0.1M ammonium bioxalate at ~2 drops per second**.** Cerium was added to the tubes as previously described, along with 1 ml of concentrated hydrofluoric acid (49%), prior to elution. A 0.5 ml volume of 20 wt% titanium chloride was also added to each tube also prior to elution to reduce uranium to U^{+4}. After waiting 10 minutes, the solutions were filtered onto 0.1 micron 25 mm polypropylene filters (Resolve® filter-Eichrom Technologies) and counted by alpha spectrometry.

Actinide filters were counted by alpha spectrometry for 30-40 minutes due to the relatively high levels of actinides in these emergency response samples.

While alpha spectrometry was used in this work, previous work in this lab has shown that if ICP-MS measurement is desired, alternate strip solutions may be used that are compatible with direct introduction without any significant signal suppression [15]. Alternately, Pu may be stripped from TEVA Resin using 15 ml 0.25M HCL-0.005M HF-0.0001M titanium (III) chloride solution. The 0.25M HCL solution used to strip Am from DGA Resin is already compatible with ICP-MS measurements. Uranium may be stripped from TRU Resin using 15 ml 0.01M ammonium bioxalate. A combination of ICP-MS and alpha spectrometry may be used as needed.

Apparatus

Plutonium, americium, and uranium measurements were performed by alpha-particle pulse-height measurements using Passivated Implanted Planar Silicon (PIPS) detectors. Polycarbonate vacuum boxes with 24 positions and a rack to hold 50 ml plastic tubes were used. Two boxes were connected to a single vacuum source by using a T-connector and individual valves on the tubing to each box.

Results and Discussion

Table 1 shows turnaround times for actinides in NRIP 2009 soil samples. The SRS Environmental Bioassay Lab reported $^{239/240}$ Pu, Pu-238, ^{234}U, ^{235}U, ^{238}U, and ^{241}Am in soil samples well within the 8 hour target time. The uranium and americium were reported in 4.15 and 4.42 hours respectively, while the Pu was reported in just over 5 hours.

Table 2 shows the average difference of the SRS measured values for NRIP-2009 soil samples versus the NIST reference values. The average difference from NIST reference values for the average results from five samples (N=5) containing approximately 3 different levels of activity is shown for each analyte. Considering the short count time of ~30-50 minutes, the accuracy of the average measured values (N=5) was good, more than adequate for emergency response screening. The same samples were also recounted later for 4 hours to determine the effect of a longer count time. The average difference did not show any significant differences with the longer count times.

The average difference for Pu results vs. the NIST reference values was less than 5% for ^{238}Pu and ^{240}Pu isotopes. There is a slight negative bias for uranium and americium, but this bias is acceptable, particularly for emergency response screening. The differences are within the ~ ±30% uncertainties reported for these results.

Table 3 shows the SRS reported values compared with the NIST reference values for ^{240}Pu for each soil sample analyzed. The differences, which range from -14% to +9.1%, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{240}Pu was only 0.4%. The average tracer recovery for ^{242}Pu was 95.2%. Table 4 shows the SRS reported values compared with the NIST reference values for ^{238}Pu for each soil sample analyzed. The differences, which range from -12.7% to +9.3%, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{238}Pu was only -3.6%.

Table 5 shows the SRS reported values compared with the NIST reference values for ^{241}Am for each soil sample analyzed. The differences, which range from -20.7% to -1.7%, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{241}Pu was only -11.5%. The average tracer recovery for ^{243}Am was 97.4%.

Table 6 shows the SRS reported values compared with the NIST reference values for ^{234}U for each soil sample analyzed. The differences, which range from -27.6% to +0.5%, fall within the reported uncertainty ranges for each reported result at the 95% confidence level (except samples 16 and 25, which are slightly outside that range). For emergency response screening purposes, this bias is typically not a problem. The average bias for ^{234}U was -15.0%. The average tracer recovery for ^{232}U was 53.9%. Table 7 shows the SRS reported values compared with the NIST reference values for ^{238}U for each soil sample analyzed. The differences, which range from -23.1% to -9.1%, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{238}U was -15.9%.

Additional MAPEP reference soil samples were processed and some slight adjustments were made to determine if uranium recoveries could be improved. It was found that increasing the titanium chloride level in the initial precipitation using iron hydroxide improved uranium chemical yields significantly. The amount of 20% titanium chloride added was increased from 5ml to 10ml. This enhanced uranium precipitation likely resulted from improved reduction of the uranium to U^{4+}. The column load solution acidity was increased slightly by adding 1.5ml concentrated nitric acid after the valence adjustment with sodium nitrite. This increases the total nitrate concentration to ~6M HNO_3. The beaker rinse was also changed from 5ml 3M HNO_3 to 5ml 6M HNO_3. This increase in total nitrate concentration enhances uranium retention slightly on TRU Resin and americium/curium retention on DGA Resin. The increased nitrate level also reduces potential calcium interference on DGA Resin because calcium ions have less retention on DGA Resin at higher nitric acid concentrations. It should be noted that total nitrate level was not increased much greater than 6M total nitrate to avoid interference on TRU Resin from any residual iron present, since iron retention increases significantly on TRU Resin at very high nitrate levels. Figures 1 and 2 shows the soil sample preparation and actinide column separation respectively, including the improvements made.

MAPEP-07-MAS18 soil standards (Department of Energy (DOE) – Radiological and Environmental Sciences Laboratory (RESL), Idaho Falls, ID, USA) were analyzed. MAPEP-07-MAS18 contain Pu, U and Am isotopes with reference values.. It should be noted that the Pu-239 isotope was made refractory by DOE-RESL to test the ruggedness of laboratory sample digestion methods. In addition, ^{244}Cm was spiked into the MAPEP-07-MAS18 samples to also test for ^{244}Cm.

Table 8 shows the results on MAPEP 18 samples for Pu isotopes. The average ^{242}Pu tracer recovery was 99.3%. The ^{238}Pu average bias was only 2.85%, with an RSD of 3% and the $^{239/240}$Pu average bias was -4.86%, with an RSD of 5.39%.

Table 9 shows the results on MAPEP 18 samples for Am and Cm isotopes. The average ^{243}Am tracer recovery was 101.2%. The ^{241}Am average bias was only -7.79%, with an RSD of 5.22% and the ^{244}Cm average bias was 2.89%, with an RSD of 3.79%.

Table 10 shows the results on MAPEP 18 samples for U isotopes. The average ^{232}U tracer recovery was 86.4%, indicating the improvements to the method increased the uranium chemical yield. The ^{234}U average bias was only -2.48%, with an RSD of 1.57% and the ^{238}U average bias was -1.32%, with an RSD of 3.21%.

Figure 3 shows an example of the plutonium spectra for the NRIP 2009 soil samples. The ^{242}Pu tracer recovery was 91.36% and the Full Width Half Maximum (FWHM) was 44.9keV, showing acceptable alpha peak resolution and minimal reduction in tracer recoveries even with rapid column flow rates. The ^{239}Pu peak labeled on the spectra represents ^{239}Pu plus ^{240}Pu, since these isotopes have essentially the same alpha energy.

Figure 4 shows an example of the americium spectra for the NRIP 2009 soil samples. The ^{243}Am tracer recovery was 102.2% and the Full Width Half Maximum (FWHM) was 56.4 keV, showing acceptable alpha peak resolution. Lanthanum was used instead of cerium as a preconcentration step because La retention is slightly less than Ce on DGA Resin [16]. The rapid removal of La was sufficient, as evidenced by the acceptable alpha peak resolution.

Figure 5 shows an example of the uranium spectra for the NRIP 2009 soil samples. The ^{232}U tracer recovery was 68.3% and the Full Width Half Maximum (FWHM) was 80.5 keV, showing acceptable alpha peak resolution.

Conclusions

For emergency response screening, the SRS NRIP 2009 soil data quality is sufficient, but samples may be counted longer as needed for routine sample analysis. The column chemistry is very rapid and flexible, and the soil matrix (including any refractory actinide particles) are completely digested using the rapid fusion. This method has high tracer recoveries and effectively removes interferences. The report times in the NRIP-2009 program by the SRS Environmental laboratory demonstrate the speed and effectiveness of this new method. Improvements were made to significantly improve uranium chemical yields. This new rapid method can also been applied to routine samples to reduce labor costs and analysis time.

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Table Captions

Table 1 Turnaround times on NRIP-09 soil samples

Table 2 NRIP-2009 Soil Analysis Average Results

Table 3 NRIP-2009 Soil Analysis Results for ^{240}Pu

Table 4 NRIP-2009 Soil Analysis Results for ^{238}Pu

Table 5 NRIP-2009 Soil Analysis Results for ^{241}Am

Table 6 NRIP-2009 Soil Analysis Results for ^{234}U

Table 7 NRIP-2009 Soil Analysis Results for ^{238}U

Table 8 MAPEP Soil Analysis Results for Pu Isotopes

Table 9 MAPEP Soil Analysis Results for Am and Cm Isotopes

Table 10 MAPEP Soil Analysis Results for U Isotopes

Figure Captions

Fig. 1 Rapid Soil Sample Preparation

Fig. 2 Rapid Actinide Column Separation

Fig. 3 Alpha spectra showing Pu Isotopes in NRIP 2009 Soil Sample

Fig. 4 Alpha spectra showing Am Isotopes in NRIP 2009 Soil Sample

Fig. 5 Alpha spectra showing U Isotopes in NRIP 2009 Soil Sample

Table 1 Turnaround times on NRIP-09 soil samples

Nuclide	Turnaround Times
^{241}Am	4.42 hrs
$^{238/239}$Pu	5.40 hrs
$^{234,235.238}$U	4.15 hrs

Table 2 NRIP-2009 Soil Analysis Average Results

Nuclide	Avg. Difference Reported vs NIST	Avg. Difference Longer Recounts
^{238}Pu	-3.6 %	-7.3%
^{240}Pu	0.4%	-4.5%
^{241}Am	-11.5%	-11.0%
^{234}U	-15.0%	-14.6%
^{238}U	-15.9%	-14.7%

Actinides: 30-50 minute count time / Recounts: 4 hour count time

Table 3 NRIP-2009 Soil Analysis Results for ^{240}Pu

Sample ID	Pu Yield (%)	NIST Value (Bq Smp^{-1})	SRS Reported Value (Bq Smp^{-1} ± %, k=2)		Difference (±%)
2	84.9	0.272	0.234 ±29%		-14
9	101	0.278	0.300 ±27%		+7.9
16	85.2	0.205	0.224 ±29%		+9.1
25	91.4	0.207	0.219 ±29%		+5.8
36	114	0.0848	0.079 ±37%		-6.8
Avg,	95.2 %			Avg.	+0.4%

Table 4 NRIP-2009 Soil Analysis Results for ^{238}Pu

Sample ID	Pu Yield (%)	NIST Value (Bq Smp^{-1})	SRS Reported Value (Bq Smp^{-1} ± %, k=2)		Difference (±%)
2	84.9	0.217	0.201 ±30%		-7.3
9	101	0.222	0.218 ±28%		-1.7
16	85.2	0.1638	0.179 ±30%		+9.3
25	91.4	0.1651	0.156 ±32%		-5.5
36	114	0.0676	0.059 ±40%		-12.7
Avg,	95.2 %			Avg.	-3.6%

Table 5 NRIP-2009 Soil Analysis Results for ^{241}Am

Sample ID	Am Yield (%)	NIST Value (Bq Smp^{-1})	SRS Reported Value (Bq Smp^{-1} ± %, k=2)		Difference (±%)
2	102	0.639	0.507 ±21%		-20.7
9	92.7	0.653	0.560 ±21%		-14.3
16	107	0.483	0.435 ±21%		-9.9
25	102	0.487	0.433 ±21%		-11.0
36	83.3	0.1992	0.196 ±28%		-1.6
Avg,	97.4 %			Avg.	-11.5%

Table 6 NRIP-2009 Soil Analysis Results for ^{234}U

Sample ID	U Yield (%)	NIST Value (Bq Smp^{-1})	SRS Reported Value (Bq Smp^{-1} ± %, k=2)		Difference (±%)
2	40.0	0.725	0.739 ±26%		0.5
9	43.2	0.750	0.671 ±26%		-10.5
16	60.1	0.563	0.422 ±25%		-25.1
25	57.4	0.569	0.412 ±25%		-27.6
36	68.9	0.226	0.196 ±27%		-12.2
Avg,	53.9 %			Avg.	-15.0%

Table 7 NRIP-2009 Soil Analysis Results for ^{238}U

Sample ID	U Yield (%)	NIST Value (Bq Smp^{-1})	SRS Reported Value (Bq Smp^{-1} ± %, k=2)	Difference (±%)
2	40.0	0.761	0.619 ±27%	-18.6
9	43.2	0.776	0.664 ±26%	-14.5
16	60.1	0.582	0.448 ±24%	-23.1
25	57.4	0.589	0.535 ±24%	-9.1
36	68.9	0.265	0.227 ±27%	-14.2
Avg,	53.9 %		Avg.	-15.9%

Table 8 MAPEP Soil Analysis Results for Pu Isotopes

MAPEP 18	Pu-242	Pu-238	Pu-239
	% Rec	Bq Kg-1	Bq Kg-1
1	96.8	75.7	88.4
2	97.2	77.1	90.3
3	99.8	77.3	85.5
4	100.7	72.5	87.0
5	107.6	72.2	77.0
6	93.5	74.4	86.2
Avg.	99.3	74.9	85.7
RSD	4.84	3.00	5.39
	Reference	72.80	90.1
	% diff	2.85	-4.86

Table 9 MAPEP Soil Analysis Results for Am and Cm Isotopes

MAPEP 18	Am-243	Am-241	Cm-244
	% Rec	Bq Kg-1	Bq Kg-1
1	104	116.6	32.4
2	98.3	124.0	32.7
3	103.9	114.0	31.6
4	96.7	125.1	34.6
5	103.4	115.1	34.3
6	101.1	109.2	31.9
Avg.	101.2	117.3	32.9
RSD	3.08	5.22	3.79
	Reference	127.20	32.0
	% diff	-7.79	2.89

Table 10 MAPEP Soil Analysis Results for U Isotopes

MAPEP 18		U-232	U-234	U-238
		% Rec	Bq Kg-1	Bq Kg-1
1		93.2	135.6	140.4
2		85.8	140.2	153.6
3		87.1	138.0	143.2
4		86.6	137.3	143.6
5		91.6	138.0	147.3
6		74.1	141.7	148.4
	Avg.	86.4	138.5	146.1
	RSD	7.77	1.57	3.21
		Reference	142	148
		% diff	-2.48	-1.32

Figure 1 Rapid Soil Sample Preparation

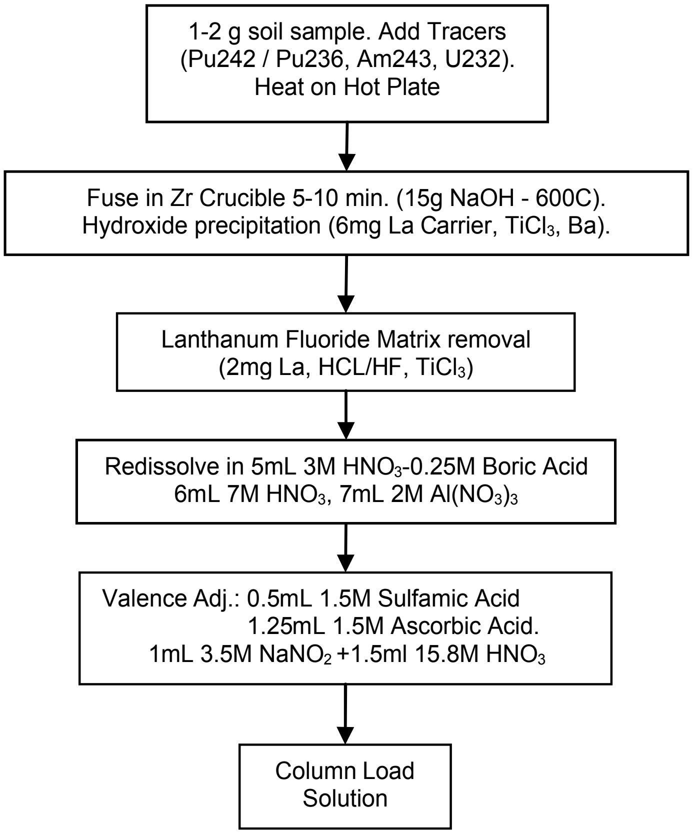


Figure 2 Rapid Actinide Column Separation

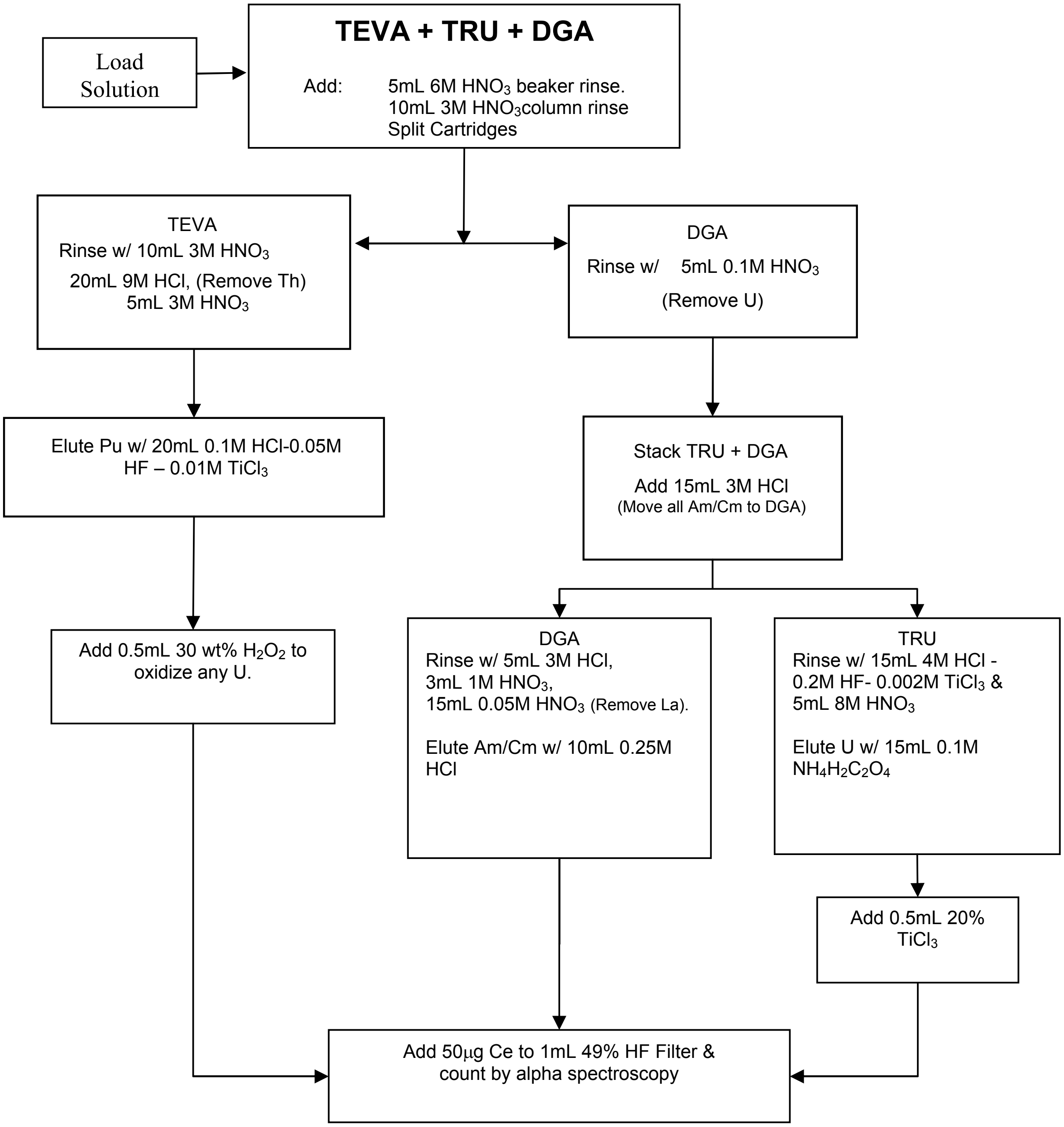
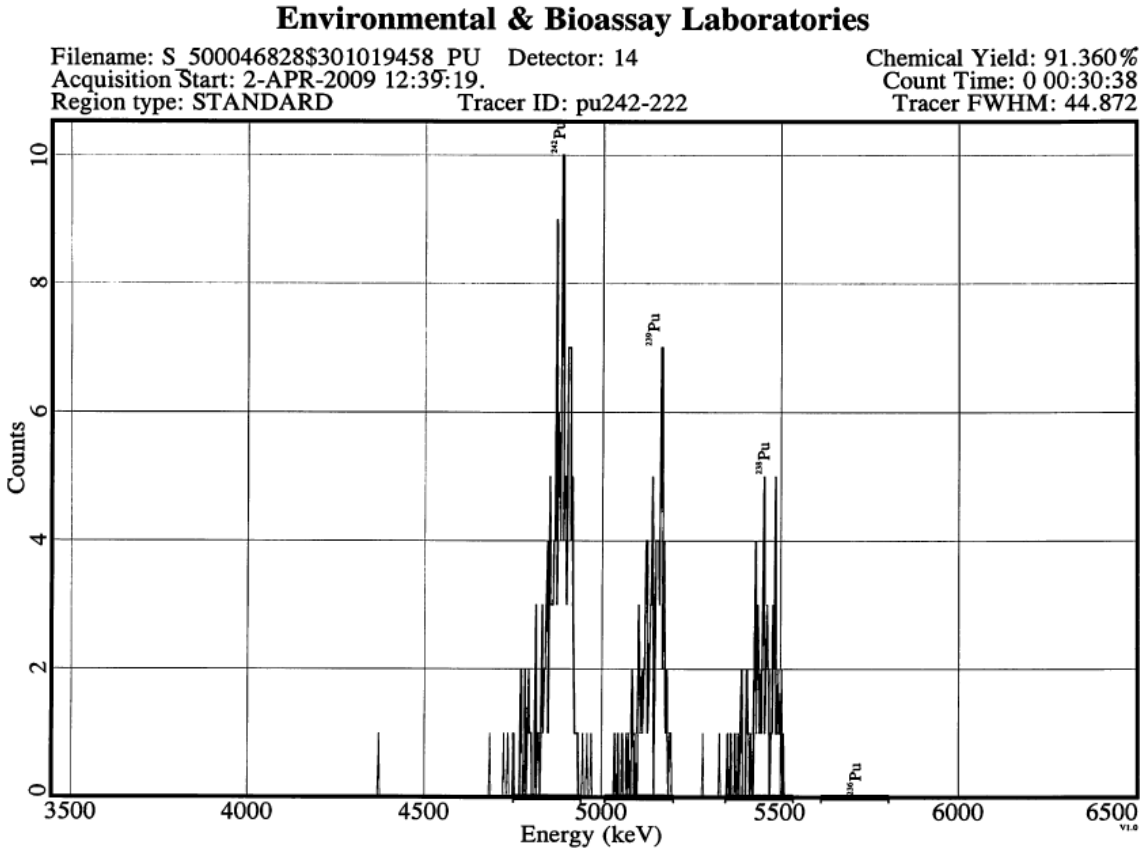


Figure 3 Alpha spectra showing Pu Isotopes in NRIP 2009 Soil Sample



Environmental & Bioassay Laboratories

Filename: S_500046828$301019458_PU   Detector: 14   Chemical Yield: 91.360%
Acquisition Start: 2-APR-2009 12:39:19.   Count Time: 0 00:30:38
Region type: STANDARD   Tracer ID: pu242-222   Tracer FWHM: 44.872

Figure 4 Alpha spectra showing Am Isotopes in NRIP 2009 Soil Sample

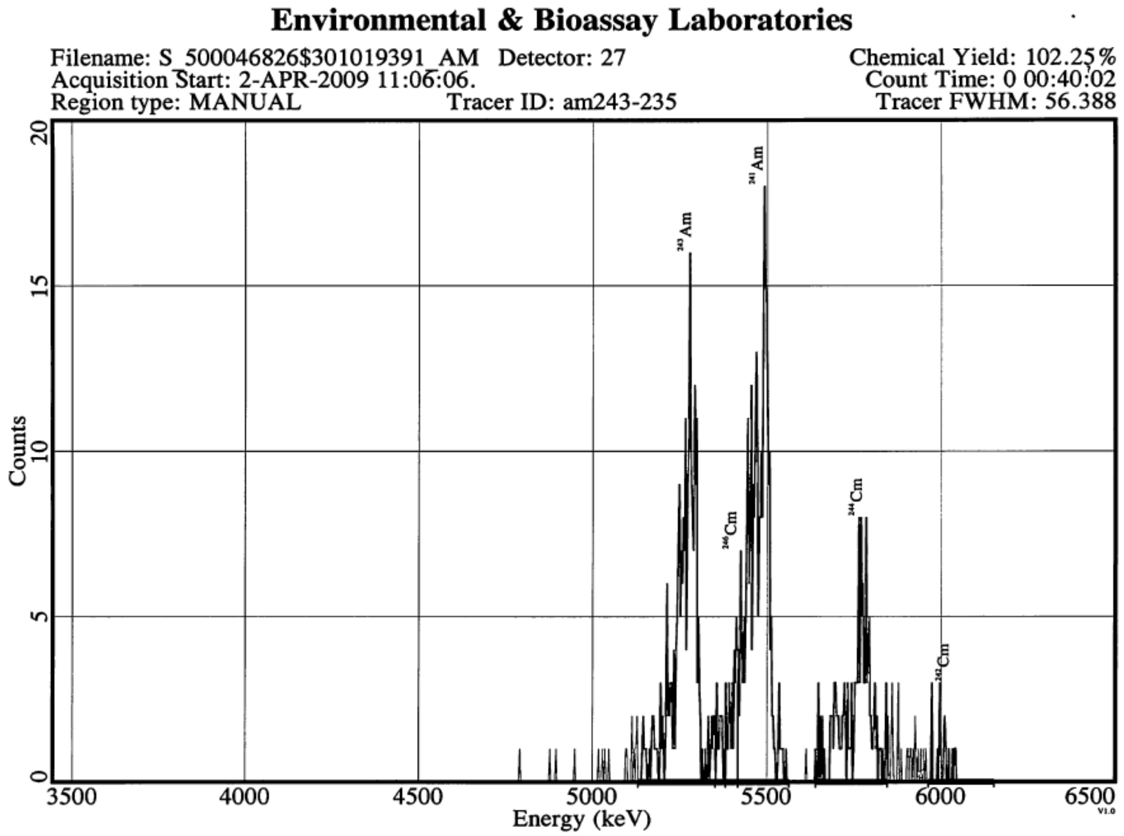
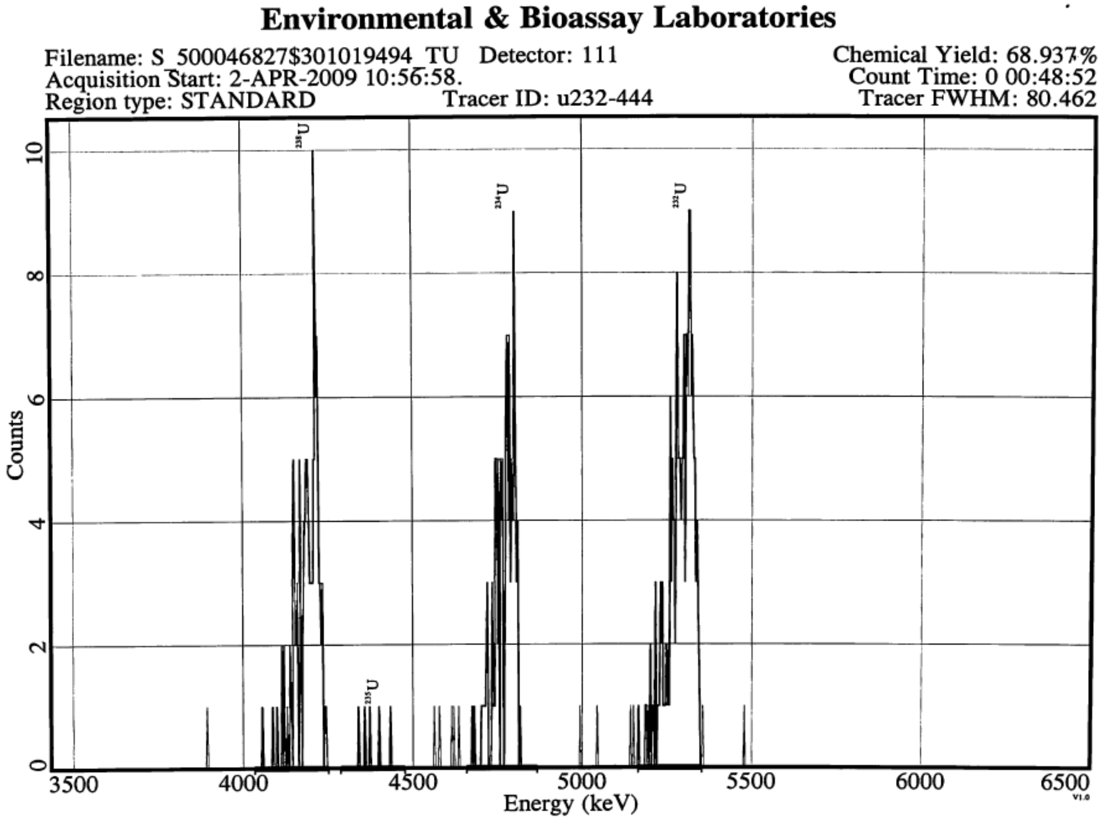


Figure 5 Alpha spectra showing U Isotopes in NRIP 2009 Soil Sample



Environmental & Bioassay Laboratories

Filename: S_500046827$301019494_TU Detector: 111
Acquisition Start: 2-APR-2009 10:56:58.
Region type: STANDARD Tracer ID: u232-444

Chemical Yield: 68.937%
Count Time: 0 00:48:52
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