

Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied: 1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or 2. representation that such use or results of such use would not infringe privately owned rights; or 3. endorsement or recommendation of any specifically identified commercial product, process, or service. Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Rapid Separation Method for Actinides in Emergency Air Filter Samples

Sherrod L. Maxwell, III, Brian K. Culligan and Gary W. Noyes

Sherrod L. Maxwell

Savannah River Nuclear Solutions, LLC

Building 735-B

Aiken, SC 29808, USA

phone #: 803-952-7473

Fax#: 803-952-7881

Email: sherrod.maxwell@srs.gov

Brian K. Culligan

Savannah River Nuclear Solutions, LLC

Building 735-B

Aiken, SC 29808, USA

phone #: 803-952-7242

Fax#: 803-952-7881

Gary W. Noyes

Savannah River Nuclear Solutions, LLC

Building 735-B

Aiken, SC 29808, USA

phone #: 803-952-8585

Abstract

A new rapid method for the determination of actinides and strontium in air filter samples has been developed at the Savannah River Site Environmental Lab (Aiken, SC, USA) that can be used in emergency response situations. The actinides and strontium in air filter method utilizes a rapid acid digestion method and a streamlined column separation process with stacked TEVA, TRU and Sr Resin cartridges. Vacuum box technology and rapid flow rates are used to reduce analytical time. Alpha emitters are prepared using cerium fluoride microprecipitation for counting by alpha spectrometry. The purified ^{90}Sr fractions are mounted directly on planchets and counted by gas flow proportional counting. The method showed high chemical recoveries and effective removal of interferences. This new procedure was applied to emergency air filter samples received in the NRIP Emergency Response exercise administered by the National Institute for Standards and Technology (NIST) in April, 2009. The actinide and ^{90}Sr in air filter results were reported in ~4 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. Wang et al reported a sequential method to determine actinides and strontium in environmental samples [4]. 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 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 air filters were as follows: plutonium (61-85%), americium (42-65%), uranium (56-73%), and Strontium (67-83%). 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.

Salminen et al reported a method for determining plutonium in air filters using UTEVA Resin plus TRU Resin [5]. The organic filters were ashed overnight at 400C. Samples were wet-ashed with nitric acid, filtered, evaporated and separated for plutonium and uranium analysis. The authors note the importance of high chemical recoveries since the Pu activity levels on air filter samples are typically very low. The Pu recoveries on real samples varied between 40-100% with an average yield of 78%. The authors were

interested in leachable plutonium so less rigorous acid leach was used. Refractory particles that could be present from a RDD (Radiological Dispersive Device) would not likely be digested using this method in an emergency.

A new method has been developed in the Savannah River Site Environmental Lab (Aiken, SC, USA) that has reduced the sample preparation time for air filter samples to ~2 hours. A new rapid method for the determination of actinides and radiostrontium in air filter samples has been developed that can be used 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 $^{89,90}\text{Sr}$ methods for emergency water and urine samples [6]. The NRIP air filter samples were analyzed using a streamlined acid digestion method using followed by a stacked column consisting of TEVA Resin + TRU Resin + Sr Resin. Vacuum box technology with simultaneous column extraction and rapid flow rates are used to reduce analytical time. Alpha emitters are prepared using cerium fluoride microprecipitation for counting by alpha spectrometry. The purified ^{90}Sr fractions are mounted directly on planchets and counted by gas flow proportional counting. 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 diisobutylcarbamoymethylphosphine

oxide (CMPO)), and Sr Resin (4, 4', (5') di-t-butylcyclohexane-18-crown-6), available from Eichrom Technologies, Inc., (Lyle, Illinois, USA). Nitric 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 Analytix, Inc. (Atlanta, GA, USA) and diluted to approximately 0.37 Bq ml^{-1} were employed to enable yield corrections. ^{232}U tracer was prepared to be self-cleaning, removing its ^{228}Th daughter using barium sulfate precipitation [7].

Procedures

Column preparation. TEVA, TRU, and Sr-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 $1\text{-}2 \text{ ml min}^{-1}$ are typically used in the SRS Environmental Laboratory, but flow rates ~ 2 times faster were used for this work.

Sample Preparation. The NRIP air filter samples were added to 250 ml Teflon beakers. Tracers were added to each crucible, 3ml 15.8M HNO_3 , 3ml 30wt% H_2O_2 and 5ml 49% HF was added to each beaker and the samples were heated to dryness on a hot plate. The following was done three times: 3ml 15.8M HNO_3 and 3ml 30wt% H_2O_2 were added to each sample and heated to dryness on the hot plate. Finally, 3ml 3M HNO_3 -0.25M boric acid was added and the samples were evaporated to dryness. The samples

were redissolved in 6ml HNO₃ and 6 ml 2M aluminum nitrate to prepare a 3M HNO₃-1M aluminum nitrate load solution.

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 Sr-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 Pu³⁺. Following the reduction step, 1.25 ml 3.5M sodium nitrite was added to oxidize plutonium to Pu⁴⁺. 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 ~3 ml 3M HNO₃ was transferred to the stacked column and a rinse of 10 ml 3M HNO₃ was added directly to the stacked column. The TRU Resin and Sr-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.

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, dried, and counted by alpha spectrometry.

The TRU Resin cartridges were placed on a separate vacuum box. Am was stripped from TRU Resin with 15 ml 4M HCl at ~1-2 drops per second. Cerium was added as previously described to the tubes, along with 3 ml of concentrated hydrofluoric acid (49%), prior to elution. The solutions were diluted to ~30 ml with water to reduce solution acidity. 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 12 ml 4M HCl-0.2M HF to remove any residual thorium that may have passed through TEVA and been retained on TRU Resin at ~2-3 drops per second. 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), dried and

counted by alpha spectrometry. It should be noted that ^{210}Po , which can be present in air filter samples, can be removed very effectively using 4M HCl-0.2M HF-0.002M TiCl_3 if high levels of ^{210}Po are expected.[8]

Actinide filters were counted by alpha spectrometry for 30-40 minutes due to the relatively high levels of actinides in these emergency response samples. Longer count times may be used as needed to meet required detection limits.

The Sr-Resin cartridges were placed on a vacuum box and rinsed with 15 ml 8M HNO_3 at ~2-3 drops per second. The ^{90}Sr was stripped from the Sr-Resin using 10 ml 0.05M HNO_3 into 50 ml tubes at ~2 drops per second. This solution was transferred to preweighed planchets and evaporated on a hot plate to dryness. A 2 ml volume of 0.05M HNO_3 was used to rinse each tube and then was transferred to each planchet and dried. The dried planchets were allowed to cool and then were weighed to determine gravimetric carrier recovery. The planchets were counted by gas flow proportional counting.

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 [9]. 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 air filter samples. The SRS Environmental Bioassay Lab reported $^{239/240}\text{Pu}$, ^{238}Pu , ^{234}U , ^{238}U , ^{241}Am and ^{90}Sr in air filter samples well within the 8 hour target time. The americium and uranium results were reported in 3.6 and 3.7 hours respectively, while the Pu isotope results was reported in 3.9 hours. ^{90}Sr results were reported in only 3.3 hours.

Table 2 shows the average difference of the SRS measured values for NRIP-2009 air filter 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 average difference for Pu results vs. the NIST reference values was less than 8% for all actinide isotopes, and the bias for ^{90}Sr was -9.9%. The quality of the analytical

results is very good, even with the rapid sample processing and short count times. The differences are well within the $\sim \pm 30\%$ uncertainties reported for these results and the results are acceptable for emergency response screening as well as for routine analysis.

Table 3 shows the SRS reported values compared with the NIST reference values for ^{240}Pu for each air filter sample analyzed. The differences, which range from -13.3% to $+0.6\%$, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{240}Pu was only -7.3% . The average tracer recovery for ^{242}Pu was 94.1% . Table 4 shows the SRS reported values compared with the NIST reference values for ^{238}Pu for each air filter sample analyzed. The differences, which range from -10.3% to $+12.8\%$, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{238}Pu was only 3.3% .

Table 5 shows the SRS reported values compared with the NIST reference values for ^{241}Am for each air filter sample analyzed. The differences, which range from -6.3% to $+17.1\%$, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{241}Am was only $+7.6\%$. The average tracer recovery for ^{243}Am was 86.7% .

Table 6 shows the SRS reported values compared with the NIST reference values for ^{234}U for each air filter sample analyzed. The differences, which range from -15.6% to $+2.5\%$, fall within the reported uncertainty ranges for each reported result at the 95% confidence level. The average bias for ^{234}U was -3.36% . The average tracer recovery for ^{232}U was 66.8% . Table 7 shows the SRS reported values compared with the NIST reference values for ^{238}U for each air filter sample analyzed. The differences, which range from -14.1% to $+10.0\%$, fall within the reported uncertainty ranges for each reported

result at the 95% confidence level. The average bias for ^{238}U was -3.10%.

Table 8 shows the SRS reported values compared with the NIST reference values for ^{90}Sr for each air filter sample analyzed. The differences, which range from -16.8% to -3.1%, were acceptable for emergency as well as routine environmental analyses. The average bias for ^{90}Sr was only -9.8%. The average stable strontium carrier recovery was 85.5%. The collection and separation of ^{90}Sr for analysis along with the actinides results in significant time savings. If $^{89/90}\text{Sr}$ differentiation is needed, there are Čerenkov counting techniques for more rapid determination of ^{89}Sr and ^{90}Sr . ^{89}Sr can be measured directly by Čerenkov counting, employing methodology that takes advantage of the high Čerenkov counting efficiency of ^{89}Sr relative to ^{90}Sr . [10]

Additional reference air filter samples were processed using this new analytical method. Figures 1 and 2 shows the air filter sample preparation and column separation respectively. Titanium may be added to the 4M HCL-0.2M thorium removal rinse step on TRU Resin to provided enhanced ^{210}Po removal as needed. MAPEP air filter standards (Department of Energy (DOE) – Radiological and Environmental Sciences Laboratory (RESL), Idaho Falls, ID, USA) were analyzed. Tables 9 and 10 shows the MAPEP air filter sample results for Pu isotopes. The average ^{242}Pu tracer recovery was 110.5% (3.97% RSD). The chemical recovery slightly higher than 100% is explained by the fact that the sample alpha source filter disks are just slightly closer to the alpha detectors than the electrodeposited stainless steel calibration disks. The ^{238}Pu bias results range from -7.2% bias to +7.6%, with an average bias of only 1.2%. The $^{239/240}\text{Pu}$ bias results range from -12.3% bias to +5.6%, with an average bias of -2.7%.

Table 11 shows the results on MAPEP air filter samples for Am isotopes. The average ^{243}Am tracer recovery was 105.6% (6.48%RSD). The slightly positive bias in the tracer recoveries was explained earlier to be from a slight calibration bias. The ^{241}Am bias results range from -7.8% bias to +1.8%, with an average bias of -4.7%. Samples 1,2 and 7 did not contain added ^{241}Am and were met false positive test acceptance limits.

Tables 12 and 13 shows the results on MAPEP air filter samples for U isotopes. The average ^{232}U tracer recovery was 107.9% (3.51% RSD). The chemical recovery slightly higher than 100% is explained by the fact that the sample alpha source filter disks are just slightly closer to the alpha detectors than the electrodeposited stainless steel calibration disks. The ^{234}U bias results range from -4.4% bias to +3.2%, with an average bias of only -0.2%. The ^{238}U bias results range from -6.4% bias to +2.4%, with an average bias of -2.4%.

Table 14 shows the results on MAPEP air filter samples for Am isotopes. The average stable strontium carrier recovery was 91.5% (0.81% RSD). The ^{90}Sr bias results range from -3.7% bias to +6.72%, with an average bias of 2.7%.

Figure 3 shows an example of the plutonium spectra for the NRIP 2009 air filter samples. The ^{242}Pu tracer recovery was 98.7% and the Full Width Half Maximum (FWHM) was 36.8 keV, 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 air filter samples. The ^{243}Am tracer recovery was 90.2% and the Full Width Half Maximum (FWHM) was 37.9 keV, showing acceptable alpha peak resolution.

Figure 5 shows an example of the uranium spectra for the NRIP 2009 air filter samples. The ^{232}U tracer recovery was 92.6% and the Full Width Half Maximum (FWHM) was 62.3 keV, showing acceptable alpha peak resolution.

The MDA (Minimum Detectable Activity) for the actinides is calculated according to the following equation [11]:

$$\text{MDA} = [3 + 4.65\sqrt{B}] / (\text{CT} * \text{R} * \text{V} * \text{Eff} * 0.060)$$

Where B = Total Background counts, = BKG (rate) * BKG Count time

CT = sample count time (min)

R = Chemical Recovery

V = Sample Volume (Liters)

EFF = Detector Efficiency

0.060 = conversion from dpm to mBq

In low-level counting, where a zero background count is quite common, the constant 3 is used to prevent an excessively high false positive rate.

The MDA can be adjusted as needed, depending on count time. For an air filter sample aliquot, the MDA for a 2 hour count time is 1.63 mBq filter⁻¹. For a 22 hour count time, the MDA is 0.15 mBq filter⁻¹. The MDA for ^{90}Sr is calculated in a similar fashion and was determined to be equal to 104 mBq filter⁻¹ for an air filter sample counted for 10

minutes. Longer count times may be used to reduce analytical uncertainty or lower the MDA as needed.

Conclusions

The SRS NRIP 2009 air filter data quality is more than sufficient for emergency response screening, but samples may be counted longer as needed for routine sample analysis to improve data quality as needed. The stacked cartridge column chemistry is very rapid and flexible, and the glass fiber air filter matrix is completely digested using the nitric acid-hydrofluoric acid digestion. This method has high tracer recoveries and effectively removes interferences. The report times in the NRIP-2009 program by the SRS Environmental laboratory (~4 hours) demonstrate the speed and effectiveness of this new method. This rapid method can also be applied to routine samples to reduce labor costs and analysis time. The column chemistry can also be adapted to measurements of actinide isotopes using ICP-MS.

Acknowledgment

This work was performed under the auspices of the Department of Energy, DOE Contract No. DE-AC09-96SR18500. The authors wish to acknowledge Christy Posey, Dan Stewart, Jack Herrington and Becky Chavous for their assistance with this work.

References

1. Inn, K.G.W., Proceedings of the 50th Annual Conference on Bioassay, Analytical and Environmental Radiochemistry, Cincinnati, OH, (2004) 113
2. Stricklin, D.L., Tjarnhage, A, and Nygren, U.: Application of low energy gamma-spectrometry in rapid actinide analysis for emergency preparedness, J. Radioanal. Nucl.Chem. **251** (1), 69 (2002)
3. D. Larivere, Cumming, T.A. , Kiser S., Li C. and Cornett R., Automated flow injection system using extraction chromatography for the determination of plutonium in urine by inductively coupled plasma mass spectrometry, J. Anal. At. Spectrom., **23**, 352 (2008),
4. Wang, J., Chen, I, and Chiu, J.: Sequential isotopic determination of plutonium, thorium, americium, strontium and uranium in environmental and bioassay samples, Applied Radiation and Isotopes, **61**, 299 (2004)
5. Salminen, S., Outola, I, Jaakkola, T, Pulli, S., Zillacus, R., and Lehto, J., Radiochim. Acta, **92**, 467 (2004)
6. Maxwell, S. and Culligan, B.: Rapid separation method for emergency water and urine samples, J. Radioanal. Nucl. Chem, **279** (No.3), 901, (2009)
7. Sill, C., Purification of radioactive tracers for use in high sensitivity alpha spectrometry: Analytical Chemistry, **46**(no.11), 1426, (1974)
8. Maxwell, S. and Culligan, B.: Rapid separation method for emergency water and urine samples, J. Radioanal. Nucl. Chem, **279** (No.3), 901, (2009)

9. Maxwell III, S.L. and Jones, V. D. : Rapid determination of actinides in urine by inductively coupled plasma mass spectrometry and alpha spectrometry: A hybrid approach: *Talanta*, **80** (No.1), 143, (2009)
10. Martin, J. P. and Odell, K. J. : The Development of Emergency radioanalytical techniques for the Determination of Radiostrontium and Transuranic radioisotopes in Environmental Material, *Radioact. and Radiochem.*, 9(3), 49, (1998)
11. Currie., L.A. : Limits for Qualitative Detection and Quantita-tive Determination: Application to Radiochemistry *Anal. Chem.* No. 40, 586, (1968)

Table Captions

Table 1	Turnaround times on NRIP-09 air filter samples
Table 2	NRIP-2009 Air Filter Analysis Average Results
Table 3	NRIP-2009 Air Filter Analysis Results for ^{240}Pu
Table 4	NRIP-2009 Air Filter Analysis Results for ^{238}Pu
Table 5	NRIP-2009 Air Filter Analysis Results for ^{241}Am
Table 6	NRIP-2009 Air Filter Analysis Results for ^{234}U
Table 7	NRIP-2009 Air Filter Analysis Results for ^{238}U
Table 8	NRIP-2009 Air Filter Analysis Results for ^{90}Sr
Table 9	MAPEP Air Filter Analysis Results for ^{238}Pu
Table 10	MAPEP Air Filter Analysis Results for ^{239}Pu
Table 11	MAPEP Air Filter Analysis Results for ^{241}Am
Table 12	MAPEP Air Filter Analysis Results for ^{234}U
Table 13	MAPEP Air Filter Analysis Results for ^{238}U

Figure Captions

Figure 1 Rapid Air Filter Sample Preparation

Figure 2 Rapid Air Filter Column Separation

Figure 3 Alpha spectra showing Pu Isotopes in NRIP 2009 Air Filter Sample

Figure 4 Alpha spectra showing Am Isotopes in NRIP 2009 Air Filter Sample

Figure 5 Alpha spectra showing U Isotopes in NRIP 2009 Air Filter Sample

Table 1 Turnaround Times on NRIP-09 Air Filter Samples

Nuclide	Turnaround Times
^{241}Am	3.6 hrs
$^{238/239}\text{Pu}$	3.9 hrs
$^{234,238}\text{U}$	3.7 hrs
^{90}Sr	3.3 hrs

Table 2 NRIP-2009 Air Filter Analysis Average Results

Nuclide	Avg. Difference Reported vs NIST
^{238}Pu	3.3%
^{240}Pu	-7.3%
^{241}Am	7.6%
^{234}U	-3.4%
^{238}U	-3.1%
^{90}Sr	-9.9%

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

Sample ID	Pu Yield (%)	NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹ ± %, k=2)	Difference (±%)
s10	96.4	0.284	0.260 ±24%	-8.4
s13	76.6	0.331	0.333 ±25%	+0.6
s20	106.6	0.200	0.175 ±25%	-12.5
s39	98.7	0.089	0.086 ±32%	-3.0
s60	92.2	0.091	0.079 ±33%	-13.3
Avg,	94.1 %			Avg. -7.3%

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

Sample ID	Pu Yield (%)	NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹ ± %, k=2)	Difference (±%)
s10	96.4	0.226	0.231 ±24%	+2.0
s13	76.6	0.264	0.269 ±26%	+1.9
s20	106.6	0.160	0.143 ±26%	-10.3
s39	98.7	0.071	0.078 ±32%	+10.3
s60	92.2	0.073	0.082 ±33%	+12.8
Avg,	94.1 %			Avg. +3.3%

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

Sample ID	Am Yield (%)	NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹ ± %, k=2)	Difference (±%)
s10	92.5	0.667	0.654 ±22%	-2.0
s13	86.8	0.778	0.729 ±22%	-6.3
s20	82.9	0.470	0.550 ±24%	+17.0
s39	90.2	0.208	0.244 ±27%	+17.1
s60	81.0	0.214	0.240 ±28%	+12.1
Avg,	86.7 %			Avg. +7.6%

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

Sample ID	U Yield (%)	NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹ ± %, k=2)	Difference (±%)
s10	52.5	0.727	0.710 ±28%	-2.3
s13	43.6	0.847	0.650 ±29%	+0.3
s20	59.0	0.512	0.432 ±29%	-15.6
s39	92.6	0.227	0.223 ±28%	-1.7
s60	86.2	0.233	0.239 ±28%	+2.5
Avg,	66.8 %			Avg. -3.36%

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

Sample ID	U Yield (%)	NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹ ± %, k=2)	Difference (±%)
s10	52.5	0.755	0.788 ±28%	+4.4
s13	43.6	0.879	0.884 ±28%	+0.5
s20	59.0	0.531	0.445 ±29%	-16.3
s39	92.6	0.236	0.259 ±27%	+10.0
s60	86.2	0.242	0.208 ±29%	-14.1
Avg,	66.8 %			Avg. -3.10%

Table 8 NRIP-2009 Air Filter Analysis Results for ^{90}Sr

Sample ID	Carrier Yield (%)	^{90}Sr NIST Value (Bq Smp ⁻¹)	SRS Reported Value (Bq Smp ⁻¹)	Difference (%)
s10	89.8	6.12	5.50	-10.1
s13	79.0	7.13	6.45	-9.5
s20	87.9	4.31	3.89	-9.7
s39	85.9	1.91	1.59	-16.8
s60	84.9	1.96	1.90	-3.1
Avg	85.5			-9.8
RSD	4.79			

Table 9 MAPEP Air Filter Analysis Results for ^{238}Pu

Sample ID	^{242}Pu Yield (%)	^{238}Pu Reference Value (Bq Smp $^{-1}$)	SRS Measured Value (Bq Smp $^{-1}$)	Difference (%)	Reference
1	106.0	0.091	0.095	4.5	MAPEP 21
2	108.1	0.091	0.094	3.5	MAPEP 21
3	106.1	0.176	0.189	7.6	MAPEP 20
4	116.4	0.176	0.178	1.1	MAPEP 20
5	117.6	0.176	0.163	-7.3	MAPEP 20
6	109.1	0.176	0.189	7.6	MAPEP 20
7	111.3	0.118	0.118	-0.1	MAPEP 19
8	109.4	0.067	0.062	-7.2	MAPEP 17
Avg	110.5			1.2	
% RSD	3.97				

Table 10 MAPEP Air Filter Analysis Results for ^{239}Pu

Sample ID	^{242}Pu Yield (%)	^{239}Pu Reference Value (Bq Smp $^{-1}$)	SRS Measured Value (Bq Smp $^{-1}$)	Difference (%)	Reference
1	106.0	0.138	0.134	-2.9	MAPEP 21
2	108.1	0.138	0.137	-0.5	MAPEP 21
3	106.1	0.157	0.166	5.6	MAPEP 20
4	116.4	0.157	0.153	-2.4	MAPEP 20
5	117.6	0.157	0.149	-5.3	MAPEP 20
6	109.1	0.157	0.157	-0.1	MAPEP 20
7	111.3	0.152	0.146	-3.9	MAPEP 19
8	109.4	0.084	0.074	-12.3	MAPEP 17
Avg	110.5			-2.7	
% RSD	3.97				

Table 11 MAPEP Air Filter Analysis Results for ^{241}Am

Sample ID	^{243}Am Yield (%)	^{241}Am Reference Value (Bq Smp $^{-1}$)	SRS Measured Value (Bq Smp $^{-1}$)	Difference (%)	Reference
1	102.8	NA	0.0003	NA	MAPEP 21
2	110.5	NA	0.0001	NA	MAPEP 21
3	116.2	0.205	0.190	-7.2	MAPEP 20
4	109.4	0.205	0.192	-6.2	MAPEP 20
5	108.2	0.205	0.189	-7.8	MAPEP 20
6	102	0.205	0.197	-4.0	MAPEP 20
7	94.1	NA	0.001	NA	MAPEP 19
8	101.5	0.098	0.100	1.8	MAPEP 17
Avg	105.6			-4.7	
% RSD	6.48				

NA is shown when no ^{241}Am was present. Acceptable false positive test results.

Table 12 MAPEP Air Filter Analysis Results for ^{234}U

Sample ID	^{232}U Yield (%)	^{234}U Reference Value (Bq Smp $^{-1}$)	SRS Measured Value (Bq Smp $^{-1}$)	Difference (%)	Reference
1	102.3	0.300	0.301	0.4	MAPEP 21
2	106.8	0.300	0.307	2.5	MAPEP 21
3	109.3	0.198	0.204	3.2	MAPEP 20
4	104.9	0.198	0.192	-2.8	MAPEP 20
5	112.2	0.198	0.196	-1.1	MAPEP 20
6	105.3	0.198	0.202	2.1	MAPEP 20
7	108.5	0.262	0.258	-1.4	MAPEP 19
8	113.5	0.098	0.094	-4.4	MAPEP 17
Avg	107.9			-0.2	
% RSD	3.51				

Table 13 MAPEP Air Filter Analysis Results for ^{238}U

Sample ID	^{232}U Yield (%)	^{238}U Reference Value (Bq Smp-1)	SRS Measured Value (Bq Smp-1)	Difference (%)	Reference
1	102.3	0.312	0.312	0.0	MAPEP 21
2	106.8	0.312	0.320	2.4	MAPEP 21
3	109.3	0.210	0.207	-1.3	MAPEP 20
4	104.9	0.210	0.201	-4.5	MAPEP 20
5	112.2	0.210	0.197	-6.4	MAPEP 20
6	105.3	0.210	0.207	-1.5	MAPEP 20
7	108.5	0.272	0.257	-5.4	MAPEP 19
8	113.5	0.102	0.100	-2.3	MAPEP 17
Avg	107.9			-2.4	
% RSD	3.51				

Table 14 MAPEP Air Filter Analysis Results for ^{90}Sr

Sample ID	Carrier Yield (%)	^{90}Sr Reference Value (Bq Smp $^{-1}$)	SRS Measured Value (Bq Smp $^{-1}$)	Difference (%)	Reference
1	91.5	0.7650	0.7932	3.7	MAPEP 21
2	90.6	0.7650	0.7370	-3.7	MAPEP 21
3	92.5	0.6400	0.6611	3.3	MAPEP 20
4	92.5	0.6400	0.6438	0.6	MAPEP 20
5	90.5	0.6400	0.6440	0.6	MAPEP 20
6	91.5	0.6400	0.6697	4.6	MAPEP 20
7	91.5	1.1200	1.1945	6.7	MAPEP 19
8	91.5	0.6077	0.6417	5.6	MAPEP 17
Avg	91.5			2.7	
% RSD	0.81				

Figure 1 Rapid Air Filter Sample Preparation

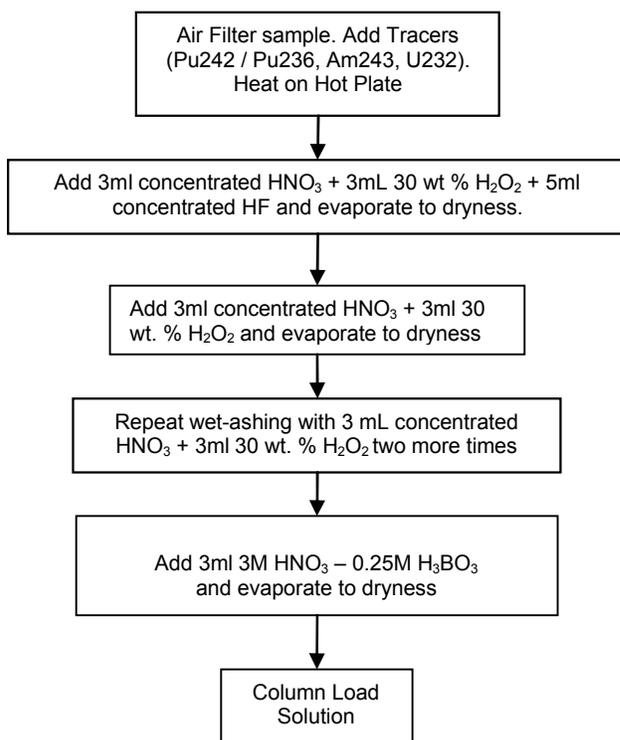


Figure 2 Rapid Air Filter Column Separation

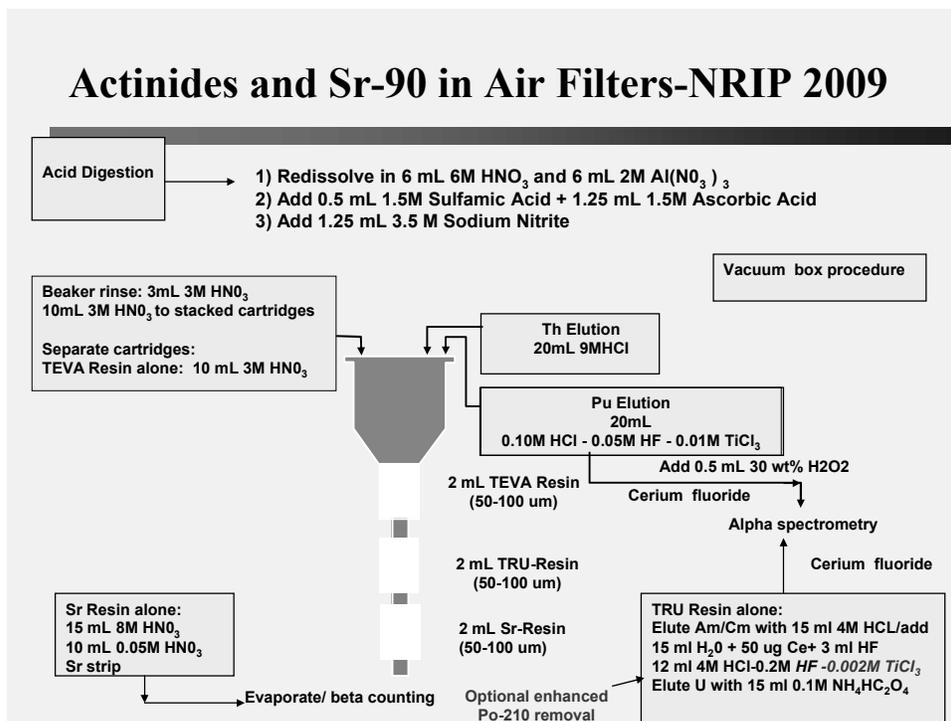


Figure 3 Alpha spectra showing Pu Isotopes in NRIP 2009 Air Filter Sample

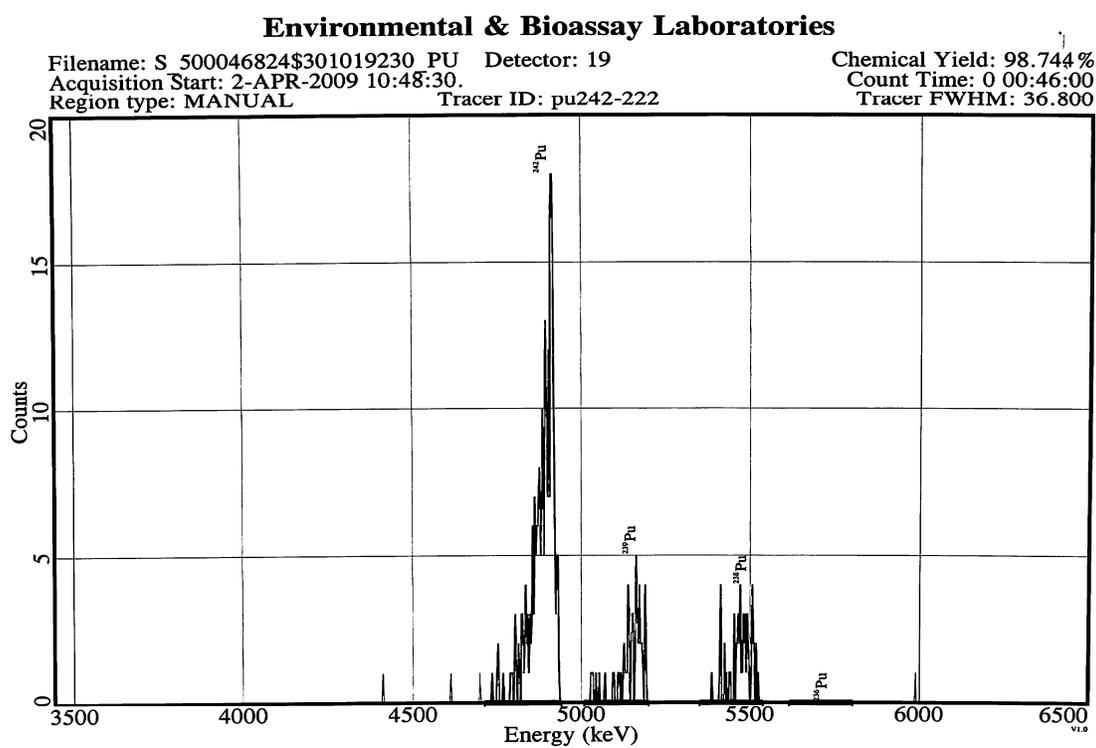


Figure 4 Alpha spectra showing Am Isotopes in NRIP 2009 Air Filter Sample

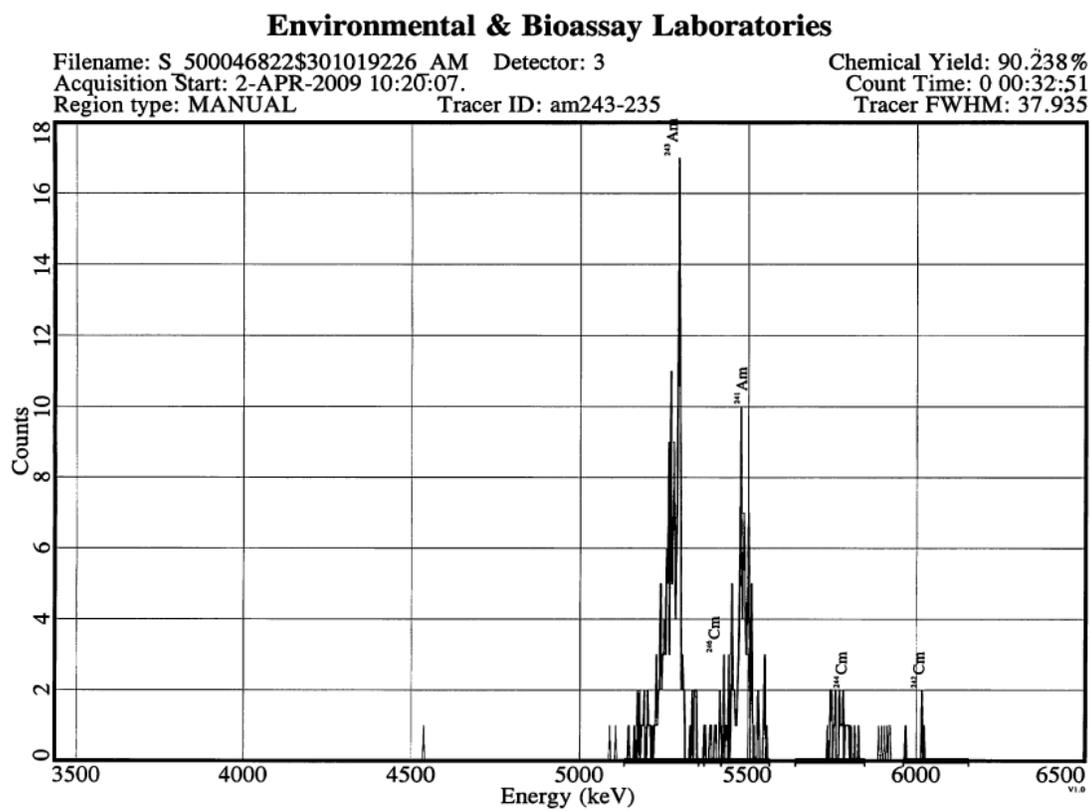


Figure 5 Alpha spectra showing U Isotopes in NRIP 2009 Air Filter Sample

