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Benchmarking Ortec ISOTOPIC Measurements and Calculations

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Introduction

This report represents a description of compiled benchmark tests conducted to probe and to demonstrate the extensive utility of the Ortec ISOTOPIC[1] γ-ray analysis computer program. The ISOTOPIC program performs analyses of γ-ray spectra applied to specific acquisition configurations in order to apply finite-geometry correction factors and sample-matrix-container photon absorption correction factors. The analysis program provides an extensive set of preset acquisition configurations to which the user can add relevant parameters in order to build the geometry and absorption correction factors that the program determines from calculus and from nuclear γ-ray absorption and scatter data.

The Analytical Development Section field nuclear measurement group of the Savannah River National Laboratory uses the Ortec ISOTOPIC analysis program extensively for analyses of solid waste and process holdup applied to passive γ-ray acquisitions. Frequently the results of these γ-ray acquisitions and analyses are to determine compliance with facility criticality safety guidelines.[2] Another use of results is to designate 55-gallon drum solid waste as qualified TRU waste[3] or as low-level waste.[4,5] Other examples of the application of the ISOTOPIC analysis technique to passive γ-ray acquisitions include analyses of standard waste box items and unique solid waste configurations.[6]

In many passive γ-ray acquisition circumstances the container and sample have sufficient density that the calculated energy-dependent transmission correction factors have intrinsic uncertainties in the range 15% - 100%. This is frequently the case when assaying 55-gallon drums of solid waste with masses of up to 400 kg and when assaying solid waste in extensive unique containers. Often an accurate assay of the transuranic content of these containers is not required, but rather a good defensible designation as >100 nCi/g (TRU waste) or <100 nCi/g (low level solid waste) is required. In these cases the ISOTOPIC analysis program is especially valuable because it allows a rapid, defensible, reproducible analysis of radioactive content without tedious and repetitive experimental measurement of γ-ray transmission through the sample and container at multiple photon energies.

The ISOTOPIC analysis technique is also especially valuable in facility holdup measurements where the acquisition configuration does not fit the accepted generalized geometries where detector efficiencies have been solved exactly with good calculus.[7,8] Generally in facility passive γ-ray holdup measurements the acquisition geometry is only

approximately reproducible, and the sample (object) is an extensive glovebox or HEPA filter component. In these cases accuracy of analyses is rarely possible, however demonstrating fissile Pu and U content within criticality safety guidelines yields valuable operating information. Demonstrating such content can be performed with broad assumptions and within broad factors (e.g. 2 – 8) of conservatism. The ISOTOPIC analysis program yields rapid defensible analyses of content within acceptable uncertainty and within acceptable conservatism without extensive repetitive experimental measurements.

In addition to transmission correction determinations based on the mass and composition of objects, the ISOTOPIC program performs finite geometry corrections based on object shape and dimensions. These geometry corrections are based upon finite element summation to approximate exact closed form calculus. In this report we provide several benchmark comparisons to the same technique provided by the Canberra In Situ Object Counting System (ISOCS)[9] and to the finite thickness calculations described by Russo in reference 10.

This report describes the benchmark comparisons we have performed to demonstrate and to document that the ISOTOPIC analysis program yields the results we claim to our customers. The subsequent eight sub-sections describe sequentially benchmarks of the ISOTOPIC program via comparison with

- separate calculations of Pu-239 content in a 55-gallon drum performed by the MicroShield[11] analysis program,
- shielded versus unshielded results using a stainless-steel-clad SRS Burial Ground glovebox HEPA filter unit,
- distinct acquisition configurations of an SRNL HEPA filter using the ISOTOPIC code,
- separate analysis using the Canberra Q^2 instrument[12],
- distinct analysis of unknown Np-237 nuclear material using multiple detectors and with generalized geometry acquisition configurations,
- analyses of 55-gallon drum γ-ray spectra using the ISOCS code,
- γ-ray analysis of a known U-235 standard,
- comparison of in-situ holdup results with off-line ISOTOPIC analyses of contained solid waste.

Benchmark Comparisons

Comparison with MicroShield Calculations

One of our first tests of the ISOTOPIC calculation program involved simple calibration of three high purity germanium detectors to perform holdup analyses of a 55-gallon drum filled uniformly with a known content of Pu-239. Each of the three detectors was calibrated in the point source acquisition configuration with a mixed source of radioactivity from a range of twelve inches.[13] All three have been used to perform analyses of TRU content in 55-gallon drum solid waste with masses up 200 kg. The

ISOTOPIC program and the MicroShield point kernel γ-ray transport program are both able to take drum dimensions, drum masses, and acquisition parameters to determine geometry and γ-ray absorption correction factors to transform acquired γ-ray data to measured content. A determination of Pu-239 content is an especially useful test, because that species has multiple passive γ-ray emissions over the energy range 100 – 415 keV.

The ISOTOPIC program is able to quickly perform analyses for each of eight Pu-239 transitions. A unique and important feature of ISOTOPIC is that it provides a graphical display of these results for each gamma-ray from a radionuclide. Figure 1 is an example taken from reference 13 where the blue points demonstrate excellent agreement with the resulting Pu-239 content based on eight γ-ray peaks in the spectrum. This kind of interactive analysis strongly reinforces the user's confidence in his result.

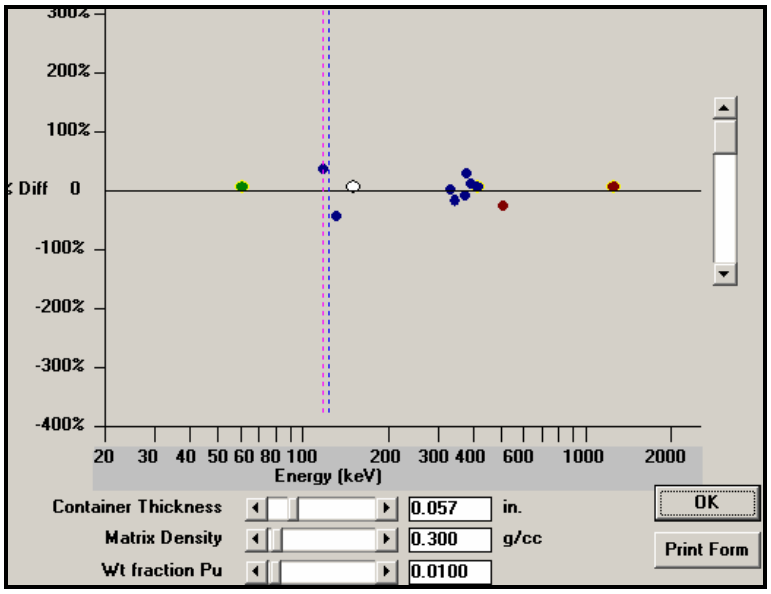

Figure 1. Screen shot of ISOTOPIC's plot of output values as a function of gamma-ray energy. The points shown in blue are for various energies characteristic of Pu-239.

Table 1 shows a comparison of ISOTOPIC's geometry and absorption corrections based on the Pu-239 375-keV γ-ray for the same sample analysis with the corrections determined by MicroShield. The correction factors from the two programs are in excellent agreement. This initial benchmark test represents a very strong endorsement of both programs.

Table 1. An independent comparison of ISOTOPIC's geometry and attenuation correction factors to those given by MicroShield for Pu-239 dispersed in a 55-gallon drum.

Correction Factors	MicroShield 375 keV	Isotopic 375 keV
Air Gap Attenuation	1.011	1.012
Drum Wall Attenuation	1.150	1.132
Matrix Attenuation	1.213	1.217
Attenuation C.F.	1.410	1.394
Geometry	16.012	16.600
Combined C.F.	31.844	32.266

HEPA Filter Measurements

The next benchmark test we discuss is a demonstration of ISOTOPIC's capability to perform γ-ray attenuation corrections from the point source standard placed inside of a Burial Ground glove-box facility HEPA filter and housing unit. The objective in the HEPA filter holdup measurements was to use the point source efficiency calibration and to rely upon the ISOTOPIC program to perform transmission correction and finite geometry corrections to conform to the HEPA filter. These experiments were performed in reference 14, and we reproduce those results here. A photo of the acquisitions is shown in Figure 2, and a comparison of the γ-ray spectra acquired from inside the filter and from the same distance not inside the filter is shown in Figure 3.

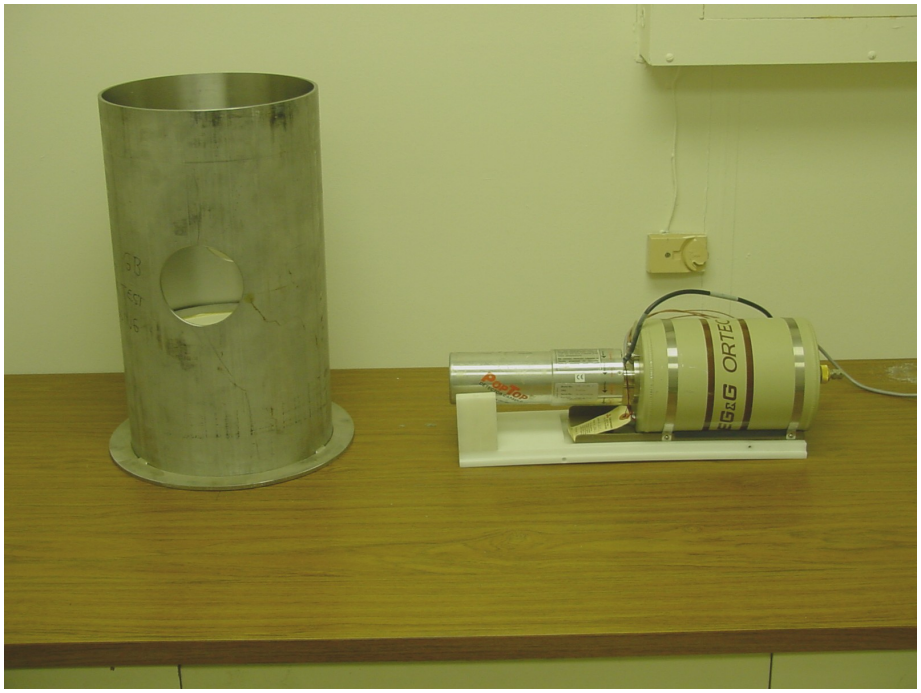

Figure 2. Detector viewing a HEPA filter in a Pad 19 filter housing at a source-to-HEPA centerline distance of 12 inches.

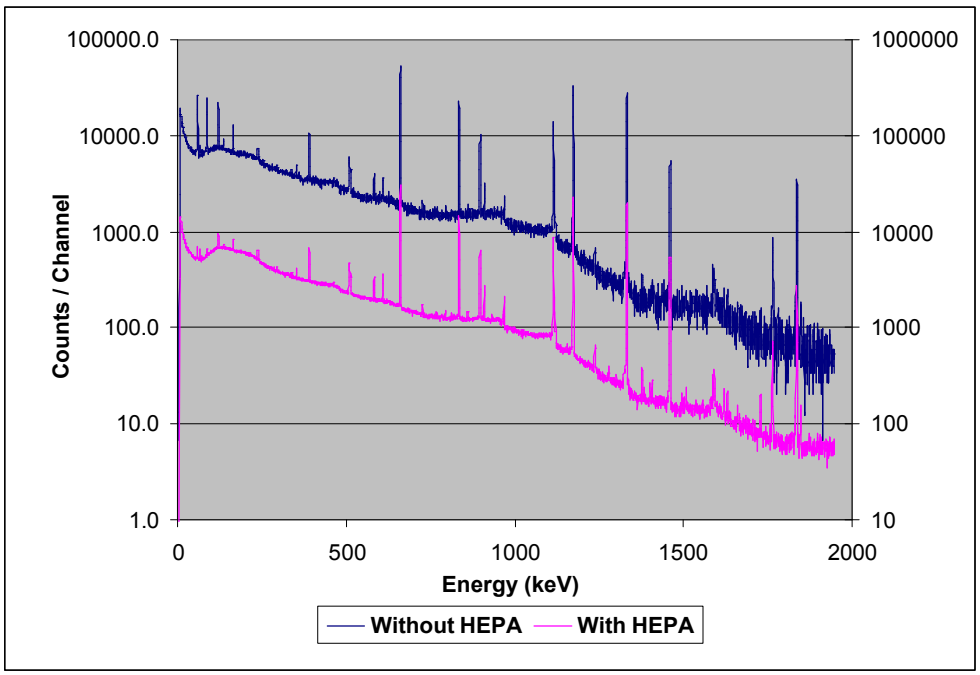

Figure 3. Spectra of a mixed g-ray standard measured twelve inches from the face of the HPGe detector with and without the HEPA filter and its housing. Data are normalized to the same count time.

The authors of 14 applied ISOTOPIC's user interface to account for the thickness of stainless steel (HEPA filter's shell and the filter housing). After the standard was counted in the housing, the data were analyzed using ISOTOPIC and the efficiency curve for the unattenuated standards. After ISOTOPIC applies attenuation corrections, the ratio of shielded to the unshielded result at each energy should be near unity. Table 2 shows that while the attenuation corrections can be large at low energies, the activity ratio is near unity in every case after applying attenuation corrections.

Table 2. Shielded to unshielded activity ratios after applying attenuation corrections determined by ISOTOPIC. Shielded results are for measurements in a HEPA filter and its housing. Unshielded results are for the measurements at the same distance but without the HEPA filter and housing.

ENERGY (KEV)	ATTENUATION CORRECTION	SHIELDED / UNSHIELDED ACTIVITY RATIO	RATIO UNCERTAINTY (%)	
88.03	24.9	1.619	14.1	
122.1	**5.98**	1.108	**6.5**	**Pu-238**
165.9	**3.305**	1.059	**8.1**	
244.7	2.336	0.905	2.9	
344.3	**2.014**	0.922	**1.1**	
391.7	**1.938**	0.960	**4.9**	**Pu-239**
411.1	**1.911**	1.037	**6.5**	
444.0	1.875	0.907	6.1	
661.7	1.714	0.943	6.8	
779.9	1.653	0.957	2.4	
834.8	1.628	0.973	0.9	
867.4	1.614	1.014	4.6	
898.1	1.600	0.947	1.7	
964.1	1.574	0.951	2.1	
1086	1.532	0.950	2.1	
1112	1.524	0.950	2.3	
1116	1.523	0.920	1.3	
1333	1.464	0.988	0.9	
1409	1.447	0.940	1.6	
1836	1.373	0.985	2.3	

Comparison of two distinct acquisition configurations of an SRNL HEPA filter using the ISOTOPIC code

From August 2006 through January 2007 the AD field nuclear measurement group performed eight holdup measurements of the SRNL drum remediation glovebox facility. Each of these sets of measurements involved eight acquisitions of the glovebox floor and walls and two HEPA filter acquisitions. The HEPA filter acquisitions were configured to view the cylindrical filter from approximately the midpoint of the vertical axis and from approximately 12 inches from the symmetry axis. The ISOTOPIC representation of the in-situ acquisition is shown in Figure 4. The filter has a steel container (housing) 3/8" thick and a combustible matrix with a density of 0.3 g/cc. With each measurement the program determined transmission and geometry corrections and TRU contents for each of Pu-238,239,241; as well as Am-241; Cm-243,244,245; and Np-237/Pa-233.

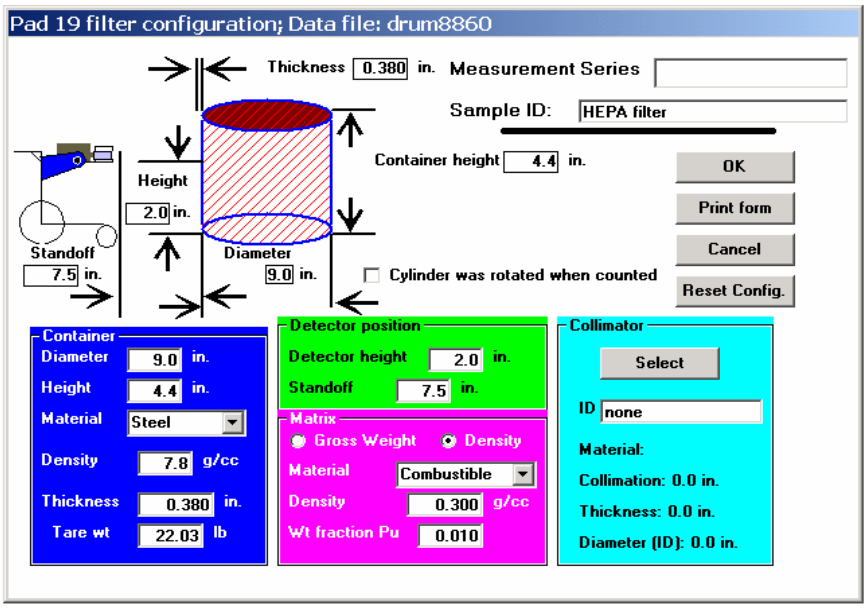

Figure 4. ISOTOPIC representation of an SRNL HEPA filter acquisition configuration.

After the final assay of the SRNL HEPA filter TRU contents, the filters were removed and placed into two separate 55 gallon drums. We recognized an opportunity to count these filters inside the drums using an alternate acquisition configuration and also using the SRNL Q^2 55-gallon drum counter. We believed that an assay of the isolated HEPA filters would yield an excellent confirmation of our original modeling of them. Such a confirmation would be especially valuable to benchmark the technical merit of all of the ISOTOPIC calculations.

For an alternate acquisition configuration, we lay the drums on their sides and viewed the HEPA filter down the filter symmetry axis from inside the drum. These two acquisitions of the two 55-gallon drums were performed with both a unique acquisition configuration and a detector different from the one used in the in-situ acquisitions of the HEPA filters. So, while the technique of analysis is still γ-PHA, the configuration and detector are both unique and independent of the in-situ analyses. We used a standoff distance of 47 inches in one case and of 53 inches for the other. The data were acquired, analyzed, and reported in reference 15. We do not show the off-line ISOTOPIC acquisition configuration, but trust it is easy for the reader to visualize.

Our geometry factors in the off-line configurations were 20.7 for the 47-inch standoff and 25.3 for the 53-inch standoff, which are both considerably larger than the in-situ geometry factors of 1.2. The measured quantities in the two HEPA filters from the off-line acquisitions are listed in Table 3, where we compare them with the final January 2007 in-situ measurements. We believe the agreement between the ISOTOPIC results from the two acquisition configurations is well within the uncertainty of each calculation. The data are plotted in Figure 5 in the next sub-section.

Table 3. Comparison of measured content (mCi) of the final in-situ assay of the SRNL HEPA filters to the measured values off-line using a unique detector in the cylindrical configuration as described in the text.

Species	HEPA E	HEPA E in drum	Species	HEPA W	HEPA W in drum
^{238}Pu	**1.31E+05**	1.24E+05	^{238}Pu	**2.62E+05**	**1.79E+05**
^{239}Pu	**1.62E+05**	**2.00E+05**	^{239}Pu	**1.71E+05**	**1.30E+05**
^{241}Pu	**6.68E+05**	**5.08E+05**	^{241}Pu	**7.19E+05**	**3.92E+05**
^{237}Np	**3.27E+01**	**3.17E+01**	^{237}Np	**2.99E+01**	**1.85E+01**
^{243}Cm	**2.89E+01**	**3.84E+01**	^{243}Cm	**2.35E+01**	**2.10E+01**
^{244}Cm	**1.31E+05**	1.28E+05	^{244}Cm	**2.62E+05**	**1.79E+05**
^{241}Am	**3.11E+05**	**7.41E+05**	^{241}Am	**4.84E+05**	**5.08E+05**
Direct Ratio		**0.92±0.28**	**Direct Ratio**		**1.39±0.30**

Comparison of analysis of an SRNL HEPA filter using the Canberra Q^2 instrument

Using the same SRNL HEPA filters as immediately above, we compared the ISOTOPIC analytical results to the analysis obtained of the HEPA filter drums by the Canberra Q^2 instrument.[12] The Q^2 was designed by Canberra specifically to perform transmission corrected γ-PHA analysis of uniformly distributed 55-gallon drum content. The instrument performs an energy-dependent matrix and container transmission correction taken directly from the mass of the item. The instrument has three HpGe γ-ray detectors stacked vertically so that, taken together, the instrument obtains a good vertical profile of the item. The empirical efficiency calibration contains the instrument geometry correction internally.

Each of the two drums that we described above was analyzed by the Q^2 instrument using a 600-sec count time. The results are reported in reference 15, and we reproduce them in Table 4 in a format very similar to that of Table 3. The Q^2 performs transmission corrected analysis of Am-241 based entirely on the 59-keV γ-ray, which is very strongly absorbed by the sample and container. Comparison of this output with the ISOTOPIC results that are based on multiple higher energy transitions from Am-241 decay is not a good practice. So we have deleted the Am-241 results from Table 4. The Q^2 measurements in Table 4 do not report ^{244}Cm. In our in-situ measurements we were not able to distinguish ^{244}Cm from ^{238}Pu, so we reported measured contents where all of the 153-keV γ-ray activity was assigned separately to each. Since the Q^2 output has no algorithm to deal with Cm-244, the comparison of the Pu-238 analyses is not valid. We have therefore deleted both Cm-244 and Pu-238 from the comparison in Table 4.

Note the disagreement between the Pu-241 analyses is significant in Table 4. We can not explain this difference based on counting geometry or transmission correction, though we note that the Pu-241 analyses are based on a lower energy transition (148-keV) than that of the remaining three species. We have included Pu-241 in the comparison of Table 4, but tabulate the species by species ratios of measured values both including and excluding Pu-241. Like the comparisons of Table 3, we believe Table 4 represents a strong endorsement of the ISOTOPIC analysis technique.

Table 4. Comparison of measured content (mCi) of the final in-situ assay of the SRNL HEPA filters to the measured values off-line using the SRNL Q^2 instrument.

Species	HEPA E	Q^2	species	HEPA W	Q^2
^{239}Pu	**1.62E+05**	1.20E+05	^{239}Pu	**1.71E+05**	1.09E+05
^{241}Pu	**6.68E+05**	3.84E+05	^{241}Pu	**7.19E+05**	3.72E+05
^{237}Np	**3.27E+01**	28.3	^{237}Np	**2.99E+01**	20.6
^{243}Cm	**2.89E+01**	25.58	^{243}Cm	**2.35E+01**	17.2
Direct ratio		**1.34±0.28**	**Direct ratio**		**1.58±0.25**
Excluding ^{241}Pu		**1.21±0.58**	**Excluding ^{241}Pu**		**1.46±0.10**

In Figure 5 we have plotted a histogram of the results of Tables 3 and 4. This figure is taken from reference 15 and includes seven nuclides. With some hedging on the Pu-241 comparisons, these results are in very good agreement.

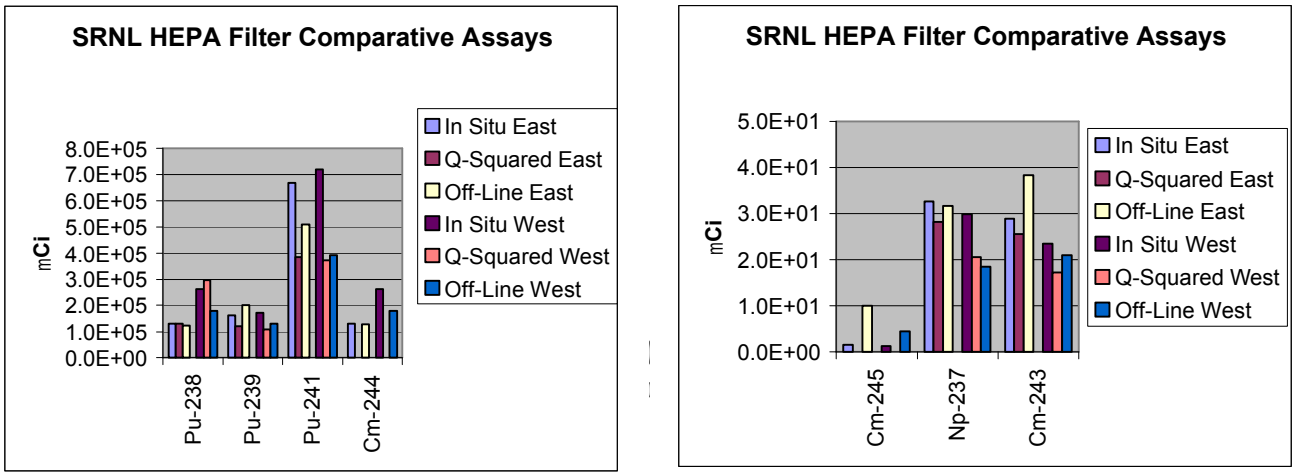

Figure 5. Comparison of measured contents (or MDA's) between the final in-situ assays, the Q^2 assays, and the off-line assays for the SRNL East and West glovebox HEPA filters.

Comparison of distinct analyses of unknown Np-237 nuclear material using multiple detectors and with generalized geometry acquisition configurations

A further benchmark comparison involved determination of Np-237 content in an unknown sample of special nuclear material received into the Actinide Technology Section (ATS) of SRNL. The ADS field nuclear measurement group was requested by ATS and by SRNL Material Control and Accountability (MC&A) to perform an analysis of this material for Np-237 content. Because SRNL had no analyzer system MC&A-qualified for Np-237 analysis, the groups agreed to a measurement plan that included well defined γ-ray analyses using two separate detectors and using three distinct analytical determinations.[16]

The plan agreed was that the AD field group would acquire passive γ-ray data from the unknown source, and the spectra would be analyzed using

1. the ISOTOPIC analysis program with data from both detectors and
2. an experimental point source calibration constant determined for one of the detectors.

The analyses of 1 would include the transmission correction and geometry correction calculated by the program. The analysis of 2 would include a generalized geometry point source acquisition configuration corrected for finite geometry by the technique of Russo.[10] The calibration constant of method 2 would be backed by a theoretical determination of Np-237 calibration constant combined with the technical review of a refereed journal.[17] Therefore the SRNL MC&A group was satisfied that AD and ATS could provide a firm, defensible measure of Np-237 content suitable for booking special nuclear material.

The details of all of the analyses of the passive γ-ray spectra acquired with the two independent detectors are described in reference 16, and we do not reproduce those details here. The results of the comparison are reproduced in Table 5. We tabulate results from two spectra acquired with each detector. In reference 16 the two groups reported an Np-237 content of 8.90±0.62 g, where the uncertainty reported is two-sigma. In this report we emphasize the outstanding agreement of the six results.

Table 5. Results of the comparison of methods 1 and 2 as described in text and in reference 16. Uncertainties are propagated from one-sigma counting statistics only.

HPGe Spectrum	cps	^{237}Np (g) Method 1	^{237}Np (g) Method 2
Detector 1-1	126.32±0.91	9.26±0.07	
Detector 1-2	123.62±0.90	9.06±0.06	
Detector 2-1	103.36±0.58	8.59±0.05	
Detector 2-2	109.04±0.59	9.09±0.05	
Detector 2-1	103.36±0.58		8.46±0.05
Detector 2-2	109.04±0.59		8.92±0.05

Comparison of analyses of 55-gallon drum γ-ray spectra using the ISOCS code

While the SRNL AD nuclear measurement group prefers the ISOTOPIC code for γ-ray analyses, a separate nuclear measurement group in F-Area Analytical Laboratories uses the Canberra ISOCS[9] code preferentially. The two codes essentially perform the same function of correcting for photon absorption and correcting for finite geometry, so it is imperative that they two codes provide results in good agreement. To test that imperative, the two groups have performed analysis of six common 55-gallon drum spectra that the AL group acquired using Canberra Genie2K software. The AL group analyzed the six spectra for TRU content and Cs-137 content using the ISOCS technique,[18] and the SRNL group analyzed the identical spectra using the ISOTOPIC technique. We compare results, which are presented for the first time in Table 6 and in Figure 6 of this report.

In columns four and eight of Table 6 we provide the ratio of ISOCS output to that of ISOTOPIC output including uncertainty. Generally the ISOCS results are 10% to 20% higher, but the measured ratios with uncertainty include 1.00 in almost every instance. We believe this is outstanding agreement for waste material packaged in drums with masses near 40 kilograms. The overall un-weighted average ratio species by species is 1.20±0.25. Note it is the Pu-239 results that deviate the most from unity. The ISOCS results in reference 18 for Pu-239 are based on the 129-keV transition, while the ISOTOPIC results of Table 6 are based on the 414-keV transition. Using the common γ-ray at 414 keV would very likely improve agreement further still. The ratios are plotted in Figure 6 without uncertainty.

Table 6. Comparison of results obtained for six F-area Analytical Laboratory drums g-ray spectra analyzed by the ISOCS and by the ISOTOPIC techniques.

Drum swd071806				**Drum swd071808**			
Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio	Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio
Cs-137	< 0.02	0.0532	N/A	Cs-137	2.06	1.59	1.30±0.43
Np-237	1.66	1.35	1.23±0.42	U-235	< 0.7	0.635	N/A
Pu-238	8.70E5	6.62E5	1.31±0.24	Np-237	76.4	65.9	1.16±0.35
Pu-239	7234	7910	0.91±0.49	Pu-238	1.72E7	1.28E7	1.34±0.55
				Pu-239	2.92E5	2.07E5	1.41±0.48
Drum swd071810				**Drum swd071812**			
Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio	Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio
Cs-137	0.497	0.662	0.75±0.75	Cs-137	1.27	1.01	1.26±0.45
U-235	< 0.6	< 0.5	1.2±1.2	U-235	1.51	1.29	1.17±0.44
Np-237	32.1	29.1	1.10±0.33	Np-237	34.7	30.0	1.16±0.34
Pu-238	2.15E7	1.70E7	1.26±0.45	Pu-238	1.51E7	1.13E7	1.34±0.57
Pu-239	4.08E4	2.66E4	1.53±0.60	Pu-239	7.05E4	4.69E4	1.50±0.53
Drum swd071813				**Drum swd081115**			
Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio	Nuclide	µCi ISOCS	µCi ISOTOPIC	Ratio
Cs-137	0.319	0.264	1.21±0.83	Cs-137	0.303	0.314	0.96±0.79
U-235	< 703	79.6	N/A	U-235	1.02	1.02	1.00±0.74
Np-237	91.8	78.5	1.17±0.35	Np-237	429	779	0.55±0.17
Pu-238	2.77E7	2.10E7	1.32±0.41	Pu-238	5.52E7	4.03E7	1.37±0.31
Pu-239	1.44E5	9.03E4	1.59±0.34	Pu-239	9.36E4	2.34E5	0.40±0.38

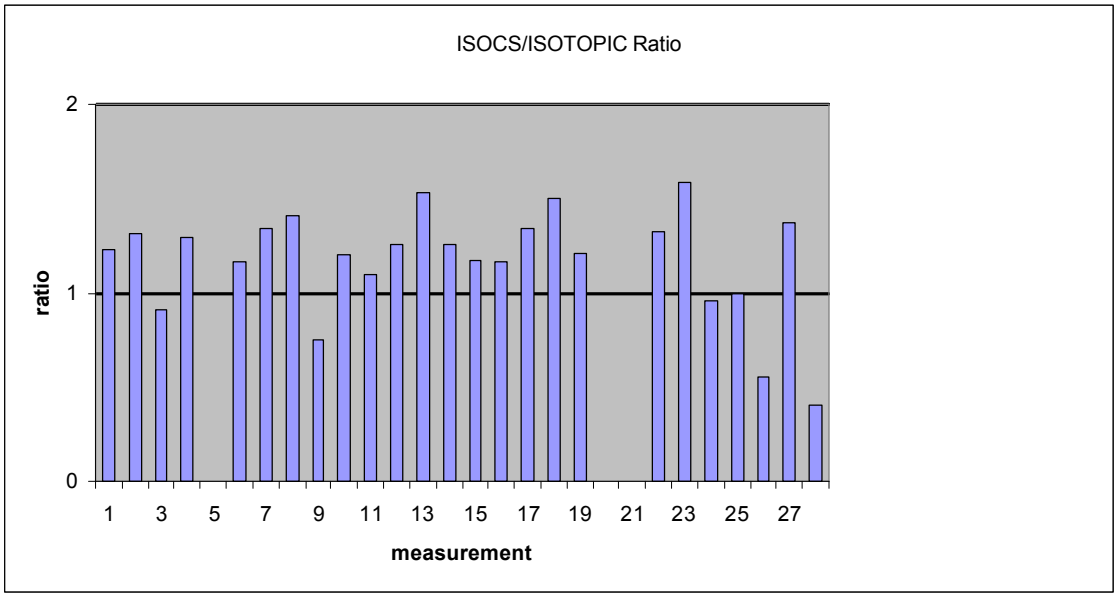

Figure 6. Comparison of measured contents of six 55-gallon drums of solid waste analyzed by ISOC and by ISOTOPIC techniques.

ISOTOPIC Analysis of a Known U-235 Standard

All of the previous tests described have utilized the species Np-237 or Pu-238,239. Each of these has γ-rays above 300 keV that penetrate better than low energy γ-rays. A measurement of a known U-235 standard would offer a useful test, as that species has multiple passive γ-rays, but none is above 205-keV. This species presents the opportunity to more rigorously test the transmission correction capabilities of the ISOTOPIC program.

In the experiments and results of reference 19 the authors performed an ISOTOPIC analysis of γ-ray spectra acquired from a known item of mass 36 g U-235 powder distributed in a 500 ml plastic bottle. The analysis of U-235 content was reported based solely on the 57% branch transition at 186 keV from U-235 decay, however the program was used to perform analysis with five other lower energy γ-rays and with the 105.7-keV $K_{\beta 1}$ x-ray. The analysis is depicted in Figure 7, and the results are shown in Table 7.

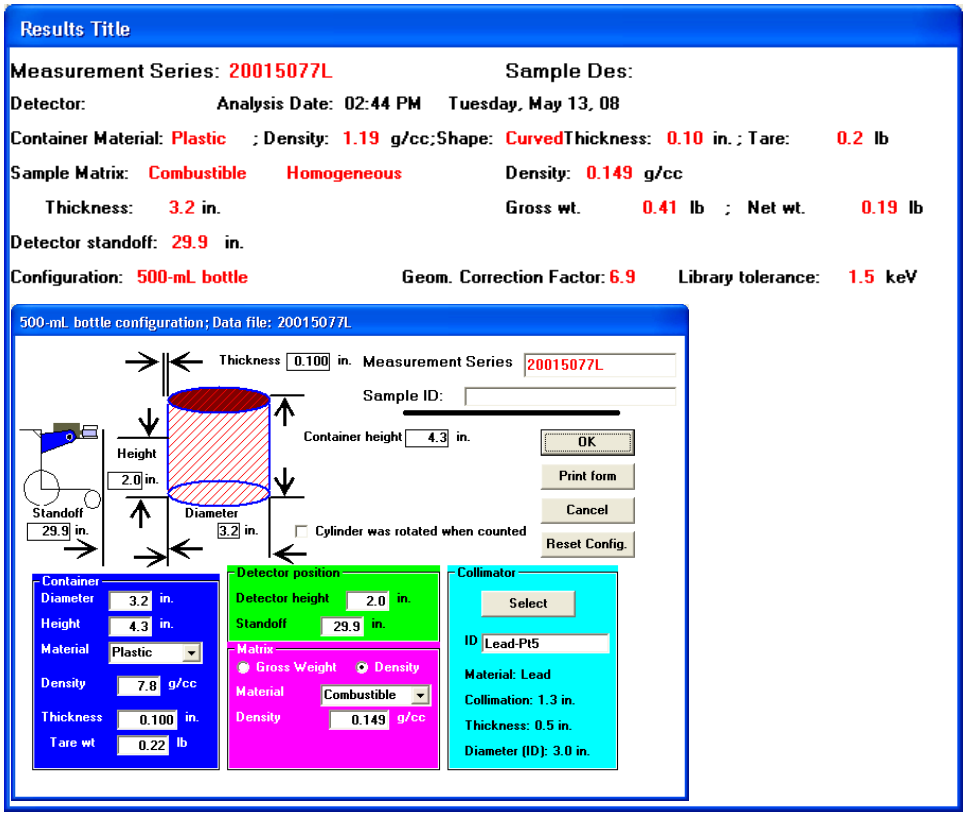

Figure 7. ISOTOPIC model of the 500-ml polyethylene bottle.

Table 7. ^{235}U Activities from ISOTOPIC analysis.

Detected γ-ray (keV)	Peak area (300 s)	^{235}U decay branch	Overall transmission factor	Equation (3) Result (dps)	Result (g)
105.7 x-ray	1350	0.0198	1.220	2.578×10^{6}	32.6
144.1	6094	0.105	1.281	1.928×10^{6}	24.3
163.5	3438	0.047	1.234	2.355×10^{6}	29.7
185.6	46721	0.53	1.199	2.813×10^{6}	**35.5**
202.1	912	0.010	1.180	3.216×10^{6}	40.6
205.2	3400	0.047	1.178	3.112×10^{6}	39.3
			Average(s)	$2.67(48) \times 10^{6}$	**33.7(6.1)**

Using the 186-keV transition in the analyses, the authors reported a measured content of 35.5 g U-235 – in excellent agreement with the known content. Using all six γ-rays and the x-ray, the measured average value reported was (33.7±6.1) – also in excellent agreement.[19] These results demonstrated the capability of the ISOTOPIC to perform transmission correction over a factor of two in energy in the low energy range of (100 – 205) keV. An important observation by the

authors was that the program was even successful to accurately implement the K absorption edge discontinuity in the transmission correction calculation. We believe this represents an especially strong endorsement of ISOTOPIC's capability.

Comparison of in-situ holdup results with off-line ISOTOPIC analyses of contained solid waste.

The last comparison included in this report involves results from the final in-situ holdup measurement performed on the Pad 19 Modular Remediation System glovebox and HEPA filter units. After this final in-situ holdup measurement was performed and technically reviewed results reported in May 2008,[20] the facility was completely decontaminated and decommissioned (D&D). The contents of both gloveboxes and of all five HEPA filter units were packaged as low level solid waste into five 55-gallon drums and one standard waste box (SWB).

In June of 2008 the AD field nuclear measurement group assayed these five drums and the SWB using close field and far field passive γ-ray PHA. We analyzed results using the ISOTOPIC program and reported the technically reviewed results in reference 21. In Table 8 we present the summed Np-237 content and summed Pu-238 content in the six waste containers and from the May 2008 MRS holdup measurement. We do not include the summed Pu-239 nor Pu-241 contents in Table 8. Both the in-situ holdup measurement and the assay of the six waste containers yielded limits of detection only for these two species. The limits from the six waste containers are far greater than the limits in the in situ holdup measurement, so these two sums have no chance of agreement.

We did obtain measured values of Np-237 in all of the in situ holdup acquisitions of the last MRS assay and in all of the six solid waste containers. Summing the Np-237 content was a simple process that does however require some explanation. We obtained measured values of Pu-238 in most of the in situ holdup acquisitions and in five of the six solid waste containers. Our sum of the two Pu-238 contents also requires an explanation.

As described in reference 15, the in situ holdup measurements of the MRS glovebox content were deliberately configured to over-calculate the TRU contents. The in situ HEPA filter measurements were configured to be as accurate as possible. The glovebox acquisitions were set up to have vastly overlapping views and to be viewed from opposite sides. Thus most of the glovebox floor was viewed by the detector more than twice. The walls were viewed on average more than once. We estimate that each individual glovebox acquisition over-calculates the TRU content by a factor f of $1 < f < 4$. In the in situ sum of Table 8, the eight glovebox results reported in 20, the measured values are arbitrarily divided by 2, and the single limit of detection obtained for Pu-238 is arbitrarily divided by four. The five HEPA filter measurements are included with no correction factor. Likewise the measured values of each of the six solid waste containers are summed with no correction factor, but the single limit of detection obtained for Pu-238 is arbitrarily divided by two.

Table 8. Summed Np-237 and Pu-238 Content obtained in the final set of in situ MRS holdup assays and obtained in the six solid waste containers from it D&D as described in text.

Species	Summed in-situ MRS mass (g)	Summed Solid Waste Containers mass (g)
Np-237	**(0.087±0.032)**	**(0.062±0.008)**
Pu-238	**(0.15±0.09)**	**(0.22±0.04)**

Note the two sums represented in Table 8 for Np-237 agree completely within our estimates of the one-sigma uncertainties and agree within 40% overall. The two Pu-238 sums also agree completely within our estimates of the one-sigma uncertainties and agree within 32% overall. Though both of these sums were calculated using the OSOTOPIC program, we believe this comparison ranks among our strongest benchmark tests of all.

Conclusion

In this technical report we describe eight independent benchmark tests of the Ortec ISOTOPIC γ-ray analysis program for determination of TRU content in unique waste configurations. Four of the tests are simple analysis of known standards in non-generalized counting geometries with variable container and sample self-absorption. One of those four tests also involved comparison with results obtained by the MicroShield photon transport program. Each of these four benchmark tests yielded favorable results for known U-235, Np-237, or Pu-239 content.

One of the eight benchmark tests involved comparison of ISOTOPIC results of TRU content in SRNL HEPA filter units with results obtained by the Canberra Q^2 instrument. A separate test involved those same SRNL HEPA filter units measured in a distinct acquisition configuration and analyzed again by ISOTOPIC. These extremely favorable results represent a powerful test of the ISOTOPIC program's analytical capabilities.

Another similar benchmark test involved comparing ISOTOPIC holdup results obtained in the final in-situ holdup assay of the SRS MRS glovebox facility with ISOTOPIC results of the contents of the RMS glovebox obtained off-line packaged separately into five 55-gallon drums and one standard waste box container. Both sets of measurements contained near 30% uncertainty, but still yielded good agreement.

Finally we performed a very important comparison of five 55-gallon drums analyzed by the ISOCS and ISOTOPIC programs. These two analytical tools are intended to perform identical corrections for energy dependent γ-ray absorption and for finite geometry correction. Demonstrating that these two program yield results in close agreement upon analysis of common γ-ray spectra represents a very important benchmark of both programs capabilities.

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