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A Technical Review of the SWEPP Gamma-Ray Spectrometer System

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**A Technical Review of the SWEPP Gamma-Ray
Spectrometer System**

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Published March 1996

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ABSTRACT

The SWEPP Gamma-ray Spectrometer (SGRS) was developed by INEL researchers as a nonintrusive method of determining the isotopic ratios of TRU and U materials in 208-liter waste drums. The SGRS has been in use at SWEPP since mid-1994. Enough questions have been raised regarding the system reliability and technical capabilities, that, coupled with a desire to procure an additional gamma-ray spectrometry system in order to increase the drum throughput of SWEPP, have prompted an independent technical review of the SGRS. The author was chosen as the reviewer, and this report documents the results of the review.

While the SGRS is accurate in its isotopic ratio results, the system is not computationally robust. The primary reason for this lack of calculational reliability is the implementation of the attenuation corrections. Suggested changes may improve the system reliability dramatically.

The SGRS is a multiple detector spectrometry system. Tests were conducted on various methods for combining the four detector results into a single drum representative value. No clear solution was reached for the cases in which the isotopic ratios are vertically segregated; however, some methods showed promise. These should be investigated further.

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A Technical Review of the SWEPP Gamma-Ray Spectrometer System

INTRODUCTION

The SWEPP Gamma-ray Spectrometer (SGRS) was developed by INEL researchers as a nonintrusive method of determining the isotopic ratios of TRU and U materials in 208-liter waste drums. The SGRS has been in use at SWEPP since mid-1994. Enough questions have been raised regarding the system reliability and technical capabilities, that, coupled with a desire to procure an additional gamma-ray spectrometry system in order to increase the drum throughput of SWEPP, have prompted an independent technical review of the SGRS. The author was chosen as the reviewer, and this report documents the results of the review. Certain of the concerns about this system are more operational than technical in nature.

The accurate and rapid determination of TRU isotopic ratios in waste drums is a challenging task. During my examination of this system I have reviewed almost one-hundred SGRS results, and most of them appear to be correct. However, the system lacks a certain "robustness" in its operation and in its results. I have divided my observations into operational and technical concerns.

DESCRIPTION OF THE SGRS

Details of the SGRS hardware and software configuration are found in the project file,¹ and in several reports,² and engineering design files³ (EDF's). The description here will be brief and will highlight the features important to later discussions.

Hardware

The SGRS incorporates four high purity germanium (HPGe) detectors installed in a specially-fabricated shield. In keeping with nomenclature established at the INEL's Test Reactor Area labs, these detectors are named A1 through A4. (A fifth detector, A5, is a spare.) The detectors are closed-end coaxial detectors with a high diameter-to-length ratio. They are well matched and are nominally 10% relative efficiency detectors with a nominal diameter of 45 mm. The nominal detector resolution at 122 keV is 0.63 keV full width at half maximum (FWHM).

The detectors are installed in a vertical array in a 15-cm thick low background steel shield designed to accommodate a 208 liter waste drum. The detectors are vertically positioned about 22 cm apart starting at an elevation about 8 cm above the bottom of a drum to be assayed. The drum to be assayed rests on a rotator pedestal driven by a stepping motor. The SGRS operator can choose to enable or disable drum rotation during any count. Obviously unknown drums are always rotated, while some mockup drums may not be.

Each SGRS detector channel incorporates an INEL-developed pulse injection with subsequent removal sub system. Each detector is equipped with a stable dual pulser, and each analog to digital converter (ADC) incorporates a special interface that recognizes the pulser events, separates them from the gamma-ray events, and digitally shifts the pulser pulses to store

in a reserved buffer region at the extreme high end of the spectrum. The resultant pulser peaks are used for energy calibration and count-rate effects correction. They also allow for powerful quality assurance procedures.

Detector output pulses are processed through an analog electronics chain that includes a commercial linear amplifier that was modified to improve its stability at high input rates. The ADCs address 8192 channel memory blocks in Canberra AIM modules. Acquisition control and data retrieval are under the control of a VAX work station communicating over an ETHERNET communications link.

Software

The VAXGAP⁴ code provides the basic acquisition control software and the routines used to fit and analyze the peaks of the gamma-ray spectrum. Ambient backgrounds are corrected using a concurrent background routine that carefully determines the appropriate background corrections using the two background spectra most contemporaneous to the time of analysis. Peaks to be fit can be located by three different means — a peak search routine, an "include" library, and a limit library. Peaks located by search or listed in the include library are fit by nonlinear least squares. The limit library specifies a set of peak energies, generally free from interferences. If they have not been analyzed either through search or include, the limit-specified peaks are fit using a linear fit routine optimized for very low (or even nonexistent) peak areas. The VAXGAP code accesses user-supplied files of counting efficiency data, nuclear decay information, etc.

The SGRS-specific portion of the software calculates a set of isotopic mass ratios from the relative intensity of one or more pairs of gamma-ray peaks. The calculational method is discussed in Reference 3. The peak pairs used under different sample conditions are reproduced in Tables 1 through 3.³

The SGRS uses a unique approach to correction of the peak area ratios for differences in relative counting efficiency. While other Pu isotopic analysis programs use a set of free-standing peaks to derive a relative efficiency curve, the SGRS uses a similar set of four ²³⁹Pu peaks to calculate gamma-ray attenuation corrections. In conjunction with mass attenuation coefficients determined by the drum's content code, the relative peak intensities are processed to compute the effective path length through the matrix material and through Pu (self absorption). The unattenuated activity values for ²³⁹Pu calculated from peaks at 129.3, 203.6, 345.0, and 413.7 keV, and the matrix and Pu mass attenuation coefficients are used to develop a set of four linear equations that are solved by standard linear least-squares for the effective path length through the matrix (x_{eff}) and the Pu self absorption path length (t_{eff}). These calculated attenuation parameters are then used to correct an unattenuated efficiency curve developed from point sources located at the drum centerline. These attenuation-corrected efficiencies are then used in the isotopic ratio calculation. This approach has the advantage of allowing an estimate of the actual activity of a radionuclide in addition to the calculation of mass ratios between nuclides. However, as discussed later in this report, its present implementation contributes to the lack of robustness in the SGRS calculations.

Table 1. Standard set of peaks used for the mass ratio calculations when neither ^{241}Am nor ^{235}U concentrations are excessive

<u>Isotopic Mass Ratio</u>	<u>Energies (kev)</u>
<u>Pu-240</u>	160.31
Pu-239	161.48
 <u>Pu-241</u>	 148.54
Pu-239	144.20
 <u>Am-241</u>	 146.55
Pu-239	144.20
 <u>Pu-238</u>	 152.72
Pu-241	148.54

Table 2. Peaks used for the mass ratio calculations when ^{235}U concentrations are excessive

<u>Isotopic Mass Ratio</u>	<u>Energies (keV)</u>
<u>Pu-240</u>	160.31
Pu-239	161.48
 <u>Pu-241</u>	 148.54
Pu-239	161.48
 <u>Am-241</u>	 125.29
Pu-239	129.30
 <u>U-235</u>	 163.36
Pu-239	161.48
 <u>U-238</u> ¹	 1001
U-235	185.72

¹ This ratio is reported in the event the drum contains uranium only.

Table 3. Peaks used for the mass ratio calculations when ^{241}Am concentrations are excessive

Isotopic Mass Ratio	Energies (keV)
<u>Pu-241</u>	148.54
Pu-239	129.30
<u>Am-241</u>	125.29
Pu-239	129.30
<u>Pu-240</u>	160.31
Pu-239	129.30
<u>Pu-238</u>	152.72
Pu-239	129.30
<u>Pu-238</u>	152.72
Pu-239	144.20
<u>U-235</u>	185.72
Pu-239	203.55
<u>U-235</u>	185.72
Pu-239	129.30
<u>Am-241</u>	125.29
U-235	185.72
<u>Am-241</u>	165.92
U-235	163.36
<u>Am-241</u>	169.56
U-235	163.36
<u>Am-241</u>	169.56
U-235	185.72
<u>Am-241</u>	169.56
U-235	163.36

After isotopic ratios are calculated from each of the four detectors, the results are inspected for outliers. In order for the set of results to be accepted, the reduced Chi-squared value must be less than or equal to four, otherwise an outlier is identified and deleted from the average. When a consistent set of ratios has been determined, the overall result on a given drum is calculated as the error weighted average of the individual detector results. This approach reflects the assumption that the isotopic contents of the sample drums are homogenous. That is, while the activity in the drum may be non-homogeneously distributed, all of the active material has the same isotopic ratio throughout the drum. This underlying assumption is clearly identified in the system references.

OPERATIONAL CONCERNS

Spectral Quality Assurance

The SGRS incorporates a system of pulse injection with subsequent removal that facilitates powerful methods of spectral quality control. At any time during data acquisition, the controlling work station can interrogate the accumulating spectrum and determine if both of the required pulser peaks are in about the right position in their buffer, have about the right magnitude and are of the right width. Alternatively, the controlling station can read the acquiring spectrum and analyze it. Either of these techniques allow real time feedback on the spectral quality. The software system to implement either of these checks is not installed on the SGRS but should be.

An inspection of the backgrounds stored on the SGRS identified several background spectra that could not be analyzed. Some of these defective spectra had only one pulser peak in the pulser storage buffer, indicating that the system gain was set too high. At least one spectrum was completely empty, and had been acquired with no input to the ADC. All of these defects could have been avoided by implementation of a spectral quality check.

The SGRS system should interrogate the acquiring spectrum shortly after a count is started and inspect the number, approximate height and the shape of the pulser peaks, or actually transmit and analyze the spectrum. If an abnormality is detected, the operator should be prominently alerted. Had such a system been in place, non-conformance reports (NCRs) EO-3206 and EO-3207 could have been avoided.

Background and Calibration-Check Frequency

SGRS operating procedures initially required weekly background checks and daily spectrometer calibration checks. The disposition of NCR EO-3201 now requires monthly background checks and weekly calibration checks when the system is in use. This frequency of checks appears adequate and prudent.

I investigated the results of analysis of a set of backgrounds drawn more-or-less randomly from those stored on the SGRS. The only gamma-ray used in the SGRS ratio analysis

that was consistently detected in all spectra in all detectors was the 185 keV gamma-ray line of ^{235}U . All of the background results at 185 keV were statistically similar ($\chi^2/\text{df} \leq 4.0$). Peaks at 129 keV and 413 keV were less consistently detected but the agreement among the detected lines was similar. The only TRU-related gamma-ray result that was consistently detected and showed greater than random variation was the 662 keV line from ^{241}Am (and ^{137}Cs). This is not surprising since this is the only gamma-ray of interest that has sufficient energy to penetrate the steel shield to any significant degree. However, the significance of the variation in the 662 keV line is more due to the small relative error on each result rather than the magnitude of the peak count results. Given the relatively small differences between system backgrounds acquired over the last year, monthly check intervals appear adequate.

I have similarly inspected the results from the periodic ^{152}Eu -calibration drum counts. Here the agreement between results is so good that new values that deviate even slightly from the established running average can generate warning notes to the operator that the new result is outside the control limits. Over the course of several months, total microCurie results deviated less than 1%, and measured pulser equivalent values normally differed by less than 0.15 keV. The weekly frequency is more than adequate; however, the control limits on these results should be expanded. I am concerned that if we place the SGRS operators in the position of rather routinely disregarding warning messages about calibration results being outside the established limits, we are an accident waiting to happen.

Required Counting Times

The SGRS implements a "Pu Monitor" that determines the required system counting time. When selected, the Pu monitor checks the total number of stored counts in a spectral region that extends from 144.2 keV to 148.5 keV. The system counts until the total count in this region equals about 30,000 or until the default maximum counting interval of 10 hours (36,000 seconds) is reached. The monitored spectral region includes two moderate yield peaks from ^{239}Pu (at 144.2 and 146.1 keV), a moderate yield ^{241}Am peak at 146.5 keV, and a similar yield peak from ^{241}Pu (148.5 keV). The choice of 30,000 total counts is empirical, and is roughly designed to allow the assay of the $^{240}/^{239}\text{Pu}$ ratio to a precision of about 10% in weapons grade (WG) material.

Given the number of drums that must be assayed, there is a desire to decrease the counting interval for most drums, particularly those with very low TRU content. However, there is a conflicting desire to improve the "accuracy" of the $^{240}/^{239}\text{Pu}$ results. (See for example NCR EO-3196.)

The system manufacturer quotes minimum detectable activities (MDAs) on similar systems of less than 100 nCi/g for certain TRU isotopes in counting times of 10 to 15 minutes (600 to 900 seconds). Many would interpret the short counting times reported by these manufacturers for detection of certain nuclides, and the lengthy counting times used by the SGRS as a deficiency in the SGRS. This is not entirely true.

The SGRS is used to establish TRU mass ratios that with the quantitative ^{239}Pu assay from passive neutron counting establish a total TRU content for an item. In order to decrease the effect of possible poor corrections for attenuation, these ratios are computed from pairs of

closely spaced peaks (See Tables 1, 2, and 3). Many of these peaks have only moderate branching ratios. This technique of deriving isotopic ratios from only moderately intense peaks requires long counting intervals to obtain any reasonable precision in the final ratio.

To illustrate this point I have calculated a number of illustrative cases. First, to provide a comparison between the detection sensitivity of each SGRS detector and the published sensitivity of the Q² unit marketed by Canberra, I have estimated MDAs for the SGRS using the standard formula derived by Currie⁵ and recommended in Section 9.0 of the WIPP QAPP.⁶ This same formula was used to compute the sensitivity of the Canberra Q² unit. The Canberra unit has calculated MDAs for ²³⁹Pu (at 413 keV) of 16.7 nCi/g at 0.1 g/cc, 9.1 nCi/g at 0.3 g/cc, and 14.0 nCi/g at 0.8 g/cc using a 600s counting time.

Table 4. Estimated detectable limits for the SGRS in a 600 second counting interval

Isotope	Energy	MDA in nCi/g @0.2g/cc	MDA in nCi/g @0.45g/cc	MDA in nCi/g @0.8g/cc
²³⁸ Pu	152.7	136	245	390
²³⁹ Pu	129.3	26	84	101
	144.2	442	939	1401
	161.5	956	1565	2574
	203.6	170	194	342
	345	167	135	217
	414	68	52	79
²⁴⁰ Pu	160.31	294	481	792
²⁴¹ Pu	148.5	689	1322	2079
²⁴¹ Am	125.29	42	165	181
	146.55	283	570	874
	662.4	295	627	295
²³⁵ U	185.7	1.8E-03 (0.9μg/g) ^a	2.3E-03 (1.2μg/g) ^a	4.0E-03 (2.1μg/g) ^a

^a ²³⁵U estimated detectable limits expressed in μg ²³⁵U per gram of matrix.

Note that the estimated MDAs for the SGRS are within a factor of 4 to 6 of those for the Canberra Q² unit. The Q² system is quoted with 20% relative efficiency (RE) Ge detectors while the SGRS uses nominally 10% RE detectors. The efficiency difference is roughly related to the detector diameter. This diameter difference alone would provide about a factor of 4 in the relative response of the two units.

Some notes are in order relating to these detectable limit calculations. Estimated MDAs are sensitive to the assumed background counting rate and to the expected detector efficiency. In order to provide a more direct comparison to the published Canberra values, the assumed background counting rates were determined from an ambient background spectrum. In later calculations the background counting rates are calculated from the continuum of a spectrum containing about the concentration of TRU to be detected. Since the presence of sample activity results in an elevated continuum level, these calculations result in higher, and more realistic detectable limits.

The expected net count rates were estimated using the unattenuated efficiency tables and applying attenuation corrections calculated from the content code-related matrix mass absorption coefficients, the stated matrix densities, and the radius of a 208 l drum. These were not calculated as distributed sources, and to do so would increase the detection efficiency, particularly at the higher matrix densities, and would result in lower (and more correct) estimated MDAs. Since this attenuated point source approximation was used for the SGRS MDA calculations, the quoted comparison values for the Canberra Q² system are those for the "worst case" source distribution, which is a similar attenuated point source assumption. An estimate of the importance of this approximation to the estimate SGRS MDAs can be obtained from the published Canberra data. Their MDAs calculated assuming a homogenous source distribution are generally a factor of four to seven lower than comparable "worst case" numbers. Similar adjustment factors are probably appropriate for the SGRS.

Also, no credit was taken in the SGRS calculations for "cross talk" between detectors. In other words, Detector A1 was presumed to view only the quarter of the drum volume located in front of A1. Photon transport calculations show that, in low density drums, Detector A1 is nearly as efficient at detecting material in front of Detector A2 as it is in detecting material in its own drum region. Detector cross talk in a combustible matrix drum approximately doubles the actual detection efficiency, and if accounted for would result in lower (and more correct) MDAs than those estimated in Table 4.

In light of the low detectable limits that can be reached in 600 seconds of counting time, why such lengthy counting intervals for actual SGRS assays of waste drums? The answer lies in the need to detect moderate yield peaks with sufficient precision to allow the calculation of meaningful ratios. Note in Table 4 that the detectable limit for the 160.3 keV line of ²⁴⁰Pu is about ten-times greater than that of ²³⁹Pu. Couple this with the fact that in weapons grade (WG) Pu the activity of ²⁴⁰Pu is much less than that of ²³⁹Pu, and it becomes easier to see why establishing precise mass ratios requires surprisingly long counting times.

I have completed several required counting time calculations. One of these is reported in Table 5, while others are reproduced in Appendix A. Table 5 reports the estimated number of seconds one would need to count in order to detect each of the relevant lines from a waste drum containing 1000 nCi α activity per gram if the TRU was aged weapons grade Pu. This is a source containing 13 nCi/g ²³⁸Pu, 710 nCi/g ²³⁹Pu, 144 nCi/g ²⁴⁰Pu, 854 nCi/g ²⁴¹Pu, and 133 nCi/g ²⁴¹Am. The background counting rates were determined from the relevant continuum levels in a spectrum of a drum having about 1000 nCi/g ²³⁹Pu. All of the previous *caveats* regarding the efficiency and attenuation approximations apply. Note that while the ²³⁹Pu can be detected in as little as 17 seconds, certain of the lines required to establish ratios are not detected in counting times of nearly a month. Even at the 1000 nCi/g level long counting times are required just to detect certain lines needed for the ratio calculations.

In order to calculate meaningful and precise mass ratios, it is not enough that the required lines be "detected," they must be quantified to a precision that propagates to a meaningful ratio precision. Table 6 reports the estimated minimum counting time required to determine, from the indicated gamma-ray pairs, isotopic mass ratios to precisions of 5, 10, and 20%. The source is again assumed to be the 1000 nCi α /g WG source previously described. These calculations consider ONLY the contribution of random statistical variation in the

counting results. Certain error components, such as the uncertainty in the attenuation corrections, have not been considered and can under some conditions be very important. Given

Table 5. Estimated counting times required to detect specific gamma-ray peaks from a 1000 nCi α /g source of WG material.

Isotope	energy	Counting time in seconds to detect at densities of:		
		0.2g/cc	0.45 g/cc	0.8 g/cc
²³⁸ Pu	152.7	8.6E+05	2.8E+06	7.1E+06
²³⁹ Pu	129.3	1.7E+01	1.4E+02	2.0E+02
	144.2	3.3E+03	1.5E+04	3.3E+04
	161.5	1.5E+04	4.0E+04	1.1E+05
	203.6	4.8E+02	6.2E+02	1.9E+03
	345	2.6E+02	1.8E+02	4.2E+02
	414	3.7E+01	2.5E+01	4.6E+01
	160.31	3.4E+04	9.2E+04	2.5E+05
²⁴⁰ Pu	148.5	5.6E+03	2.0E+04	5.0E+04
²⁴¹ Am	125.29	9.3E+02	1.4E+04	1.7E+04
	146.55	3.8E+04	1.5E+05	3.6E+05
	662.4	7.3E+03	3.1E+04	7.3E+03

the simplifying assumptions, these results should thus be viewed as "ballpark" counting intervals under the conditions specified.

Table 6. Estimated counting times required to determine mass ratios to relative precision levels of 5, 10, and 20%. Source is 1000 nCi α /g WG Pu.

Ratio	Peak 1	Peak 2	Counting time in sec for error of:		
	keV	keV	5%	10%	20%
²⁴⁰ / ²³⁹ Pu	160.3	161.5	4.92E+06	1.23E+06	3.07E+05
	160.3	129	3.43E+06	8.57E+05	2.14E+05
²⁴¹ / ²³⁹ Pu	148.5	144.2	1.32E+06	3.30E+05	8.25E+04
	148.5	161.48	2.26E+06	5.65E+05	1.41E+05
²³⁸ / ²³⁹ Pu	148.5	129	7.67E+05	1.92E+05	4.79E+04
	152.7	129	1.04E+08	2.60E+07	6.50E+06
²⁴¹ Am/ ²³⁹ Pu	152.7	144.2	1.05E+08	2.62E+07	6.54E+06
	125.3	129	5.32E+05	1.33E+05	3.33E+04
	146.6	144.2	6.30E+06	1.58E+06	3.94E+05

These calculated counting times are demoralizing to people who need to assay thousands of waste drums; however, there are some approaches that might improve the waste stream throughput. First, one could use the estimated total activity of ²³⁹Pu, ²⁴¹Am, and ²³⁵U to limit the counting time on drums containing less than 100 nCi/g TRU to no more than about 5000 seconds. [Detection limits for ²³⁵U do not appear in the tabular output. Because the gamma-ray

emission probability for the 186 keV ^{235}U gamma ray is very high at 0.572 γ/d , ^{235}U at levels of about 100 nCi/g (a substantial mass concentration of about 50 mg ^{235}U per gram of matrix) is detectable in counting times of less than 1 second.] Although the accuracy of these total activity results is questionable, they should be defensible as a sorting technique. Spectral summing techniques could be implemented (carefully) to decrease the required counting times by a factor of about 2. Since the principal problem is the determination of the $^{241}\text{Am}/^{239}\text{Pu}$ ratio, consideration should be given to estimation of this mass ratio from the 662 keV ^{241}Am line and the 414 keV ^{239}Pu line in high density drums. Although the resultant mass ratio will be somewhat uncertain because of errors in the relative attenuation correction, they may be acceptable.

TECHNICAL CONCERNS

Nuclear Data

One of the first items that I reviewed during this work was the nuclear data parameters (gamma-ray energies, branching ratios and half-lives) in the SGRS isotope library. All of the nuclear data are the best available, being drawn from the most accepted international source.⁷

Detector Resolution and Peak Shape

The SGRS relies on several very closely spaced gamma-ray lines for the calculation of isotopic mass ratios. The mass ratios are derived from multiplet peak fitting in the VAXGAP⁴ gamma-ray analysis code. VAXGAP fits the spectral peak data to a pure Gaussian function. While a Gaussian curve provides an excellent approximation to the shape of gamma-ray spectral peaks, all detectors produce actual peak shapes that differ to some extent from a pure Gaussian shape. The most pronounced difference is a slightly increased amount of "tailing" on the low energy side of each peak. Certain other gamma-ray analysis codes for Pu isotopic ratio determination, such as MGA,⁸ account for this tailing by determining peak shape parameters that modify the pure Gaussian peak shape by the inclusion of low side tailing parameter. Any low-side tail not accounted for by the fitted peak shape in the highest energy peak in a multiplet is assigned (inappropriately) to the area of the next lowest energy peak, and so forth on down the line.

The effect of this perturbation in peak shapes can nearly be eliminated by the purchase of detectors that produce peak shapes in the region of interest that have very close to Gaussian shapes. Detector resolutions are by convention specified by defining the maximum allowable full peak width at half the maximum peak height (FWHM) at 1332 keV and at 122 keV. The allowable amount of tailing can be controlled by specifying that the ratio of the full peak width at one-tenth the maximum height (FW0.1M) to the FWHM and the full peak width at one-fiftieth the maximum height (FW0.02M) be within a few percent of the values for a Gaussian function. The theoretical values for FW0.1M/FWHM and FW0.02M/FWHM for a pure Gaussian function are 1.823 and 2.376 respectively. Detectors can be supplied with peak shape parameters within 1 to 2% of theoretical.

The specification sheets provided by the manufacturer for the SGRS detectors quote the FWHM and FW0.1M values at 1332 and 122 keV. For our purposes the important energy range is at 122 keV. In all cases the specified FW0.1M/FWHM ratios are no more than 2% greater than theoretical, thus, the detectors as supplied are capable of peak shape performance that eliminate any concern about low energy tailing. However, this does not appear to be the actual performance of these units as installed.

The ^{152}Eu source that is used routinely as the SGRS calibration source emits a 122 keV gamma-ray. I have measured the actual FW0.1M/FWHM and the FW0.02M/FWHM ratios for the four SGRS detectors from a recent calibration run. These are compared to the theoretical values in Table 7.

Table 7. Comparison of measured peak width parameters for the SGRS detectors with those of a Gaussian function

Detector	FW0.1M/ FWHM	Measured/Theory (%)	FW0.02M/ FWHM	Measured/Theory (%)
A1	1.91	105%	2.84	120%
A2	2.28	125%	3.12	131%
A3	2.03	111%	2.74	115%
A4	2.04	112%	2.70	113%

The effect of detector tailing at the levels noted on the calculated isotopic ratios is not particularly severe. The effect may cause a slight high bias in, for example, the $^{240}\text{Pu}/^{239}\text{Pu}$ mass ratios, since this ratio is computed from very closely spaced peaks. It is however, somewhat disturbing that the peak shape of all the detectors seems much poorer than their specifications. There are many reasons why spectrometry systems installed outside of the laboratory can have degraded performance, and many of these cannot be corrected. I would, however, ask for electronics support in checking all of the system adjustments and generally "tuning up" the spectrometer system.

Comparison to Other Pu Isotopic Codes

In addition to the INEL-developed SGRS isotopic analysis technique, there are several other Pu isotopic codes developed at other national laboratories. These include MGA⁸ and its predecessor GRPANL⁹ (from LLNL), GRPAUT¹⁰ (a cooperative Mound lab/ LLNL product), and FRAM¹¹ (LANL). Only the SGRS technique was specifically developed for waste drum assay. The other techniques were originally developed for plutonium safeguards measurements, and have now been adapted to the TRU waste measurement problem.¹²

As part of the development and checkout of the SGRS system, limited cross comparisons were conducted between the SGRS and two other Pu isotopic codes, FRAM and MGA. The following sections briefly describe the FRAM and MGA analysis techniques and present the cross comparison results.

FRAM

The FRAM code develops Pu isotopic ratios from an analysis of generally the same energy region as is used in the SGRS. The code requires no content code nor estimated matrix mass attenuation information. Each spectrum is analyzed to develop a relative efficiency curve that is later applied to specific peak pairs in order to determine isotopic ratios. A "bootstrap" approach is used to develop a comprehensive relative efficiency curve. Initial values are determined from the ^{239}Pu lines at 129.3, 143.4+144.2, 171.3, 179.2, 189.3, 195.7, 203.5, 255.4, 297.5, 345.0, 375.0, and 413.7 keV. The curve is normalized to 1.0 at 413.7 keV. Limited curve fitting and extrapolation are used to interpolate to energies for other Pu, Am, and ^{235}U lines. Certain identified interferences are then corrected iteratively, refining the initial efficiency values. When an adequate relative efficiency curve has been determined, isotopic ratios are determined relative to ^{241}Pu and then converted to ratios relative to ^{239}Pu by simple math. The peak pairs used for the ratio calculations (under equilibrium rather than "freshly separated" conditions) are presented in Table 8. These peak pairs, when rearranged to ratios versus ^{239}Pu , are similar to those used in the SGRS. However, FRAM uses different lines for the $^{239}\text{Pu}/^{241}\text{Pu}$ ratio, including some from the ^{237}U daughter of ^{241}Pu . U-237 lines are not used by the SGRS.

The FRAM code does not use a peak fitting algorithm, but rather does a region-of-interest summation. This approach also places great emphasis on good detector resolution and peak shape.

The FRAM code and the SGRS were used to analyze a series of 6 INEL waste drums.¹³ Both codes analyzed the same spectra; however, only one of the SGRS detectors was used. The selected waste drums had varying relative amounts of ^{241}Am and ^{235}U in addition to WG Pu. The document included in Appendix B presents details on the waste drums and the results of all analyses. The plots of Figures 1 through 5 present the comparative results. Error

Table 8. Peak pairs used by the FRAM code for isotopic ratio calculations

Isotopic Ratio	Numerator (keV)	Denominator (keV)
$^{238}\text{Pu}/^{241}\text{Pu}$	152.7	148.6
$^{239}\text{Pu}/^{241}\text{Pu}$	345.0	332.4 (^{237}U)
	203.5	208.0 (^{237}U)
$^{240}\text{Pu}/^{241}\text{Pu}$	160.3	164.6
$^{241}\text{Am}/^{241}\text{Pu}$	125.3	129.3
	169.5	171.3

bars are plotted at the 1σ level. As an aide in viewing these plots, note that the horizontal bars delimiting the error bars are longer on the INEL results.

Figure 1. INEL and FRAM results comparison for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio

Figure 2. INEL and FRAM results comparison for the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio

Figure 3. INEL and FRAM results comparison for the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio

Figure 4. INEL and FRAM results comparison for the $^{241}\text{Am}/^{239}\text{Pu}$ ratio

Figure 5. INEL and FRAM results comparison for the $^{235}\text{U}/^{239}\text{Pu}$ ratio

MGA

The MGA code, developed at LLNL has been adapted by Canberra, Inc. for waste drum analyses.¹² MGA determines Pu isotopic ratios from the careful analysis of four complex peak grouping in the 110 to 225 keV region. Additionally, two additional peak groupings at 330 keV and 370 keV can be used if the detection system allows. Each of these groups is complex and is analyzed by fitting to tailed Gaussian peak shapes. Table 9¹⁴ details the peak groupings analyzed.

After all peak groups are fit, a relative efficiency curve is determined by fitting the relative results of the peaks marked with an asterisk (*) in Table 9. Iterative fits are used to correct peaks for known interferences. The curve is normalized to ^{239}Pu . The normalization factors required for the ^{241}Pu and ^{241}Am data points determine the ^{241}Pu and ^{241}Am to ^{239}Pu ratios. Pu-238 and ^{240}Pu ratios are determined from the 152.68 and 160.28 keV peaks respectively. U-235 ratios come from the 185.7 keV line.

Spectra used for MGA analyses are acquired at a relatively high gain (about 0.1 keV/channel). Thus during the cross comparison with the SGRS, the same spectra were not analyzed, rather a single MGA spectrum was taken on each of six waste drums and analyzed by Canberra personnel. These were then compared with the results obtained from the multiple detector SGRS analysis. Figures 6 through 10 compare these results.

Table 9. Gamma-ray peak groups analyzed in MGA

Isotope	Energy (keV)	Intensity	Isotope	Energy (keV)	Intensity
Group 1			Group 2		
239Pu	122.994	1.00 x 10 ⁻⁵	239Pu	141.567	3.2 x 10 ⁻⁷
239Pu	123.62	1.97 x 10 ⁻⁷	239Pu	144.211	2.83 x 10 ⁻⁶
239Pu	124.51	6.13 x 10 ⁻⁷	239Pu	146.077	1.19 x 10 ⁻⁶
239Pu	125.21	7.11 x 10 ⁻⁷	241Am	146.557	4.61 x 10 ⁻⁶
241Am	125.292*	4.08 x 10 ⁻⁵	241Pu	148.567*	1.87 x 10 ⁻⁶
239Pu	129.294*	6.26 x 10 ⁻⁵	241AM	150.11	7.40 x 10 ⁻⁷
Group 3			Group 4		
241Pu	159.955	6.74 x 10 ⁻⁸	239Pu	203.537*	5.60 x 10 ⁻⁶
239Pu	160.19	6.20 x 10 ⁻⁸	241Pu-237U	208.0*	5.33 x 10 ⁻⁶
240Pu	160.28	4.02 x 10 ⁻⁶	241Am	208.0	7.91 x 10 ⁻⁶
239Pu	161.45	1.20 x 10 ⁻⁶			
241Pu-237U	164.58*	4.53 x 10 ⁻⁷			
241Am	164.58	6.67 x 10 ⁻⁷			
241Am	165.93	2.32 x 10 ⁻⁷			
Group 5			Group 6		
241Pu-237U	332.401	2.98 x 10 ⁻⁷	239Pu	367.050	8.65 x 10 ⁻⁷
241Am	332.401	1.49 x 10 ⁻⁶	239Pu	368.546	9.03 x 10 ⁻⁷
239Pu	332.853	5.06 x 10 ⁻⁶	241Pu-237U	368.603	1.05 x 10 ⁻⁸
241Pu-237U	335.438	2.39 x 10 ⁻⁸	241Am	368.603	2.17 x 10 ⁻⁶
241Am	335.438	4.96 x 10 ⁻⁶	241Pu-237U	370.933	2.71 x 10 ⁻⁸
239Pu	336.104	1.134 x 10 ⁻⁶	241Am	370.933	5.23 x 10 ⁻⁷
239Pu	341.522	6.62 x 10 ⁻⁷	239Pu	375.042	1.57 x 10 ⁻⁵
239Pu	345.011*	5.59 x 10 ⁻⁶	241Am	376.595	1.38 x 10 ⁻⁶
			239Pu	380.166	3.05 x 10 ⁻⁶
			239Pu	382.751	2.59 x 10 ⁻⁶

* Indicates that a peak is used for determining the relative efficiency curve. The 238Pu 152.68-keV gamma-ray peak and the 235U 185.72-keV peak are analyzed individually.

Figure 6. INEL and MGA results comparison for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio

Figure 7. INEL and MGA results comparison for the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio

Again the comparison between the two techniques is excellent. However, the estimated error limits on the SGRS values exceed those of MGA.

Summary

The cross comparisons of the SGRS results with those of FRAM and MGA show clearly that the SGRS approach is valid and the system is capable of producing isotopic mass ratios equally as accurate as those of other accepted techniques. The SGRS errors are overestimated; however, this can largely be corrected by more careful error analysis.

The ability of an NDA system to provide accurate results doesn't completely validate the system capabilities. One must also ask how reliably the SGRS can produce these accurate values. While the SGRS performs well most of the time it lacks a robustness in its results. From time-to-time certain pathological cases cause unacceptable results to be generated by the SGRS, and it is this lack of *consistent* accuracy that is disturbing. I believe that this lack of robustness is primarily related to the method and implementation of the attenuation correction scheme used in the SGRS. This is discussed in the next section.

Attenuation Corrections

Rather than developing a spectrum-specific relative efficiency curve from the measured gamma-ray spectral data, the SGRS applies a spectrum and drum-specific attenuation correction to an unattenuated absolute efficiency curve. This technique has the advantage of allowing estimates of the total drum activity from the attenuation corrected gamma-ray values. However, it may contribute to the lack of robustness that has been noted in the SGRS results. In either case, since the gamma-rays chosen for ratio determinations are quite close in energy, the relative attenuation effect should be small, and the impact of the attenuation corrections on the absolute value of the mass ratio is slight.

The SGRS technique is as follows.³ The general equation for attenuation corrections is:

$$A^* = A_i e^{\mu_i x_{\text{eff}}} e^{s_i t_{\text{eff}}}$$

where:

- A^* = the attenuation-corrected absolute Pu activity in d/s
- A_i = the uncorrected Pu activity calculated from the line at energy i
- μ_i = the matrix mass attenuation coefficient in cm^2/g at energy i
- x_{eff} = the effective attenuation distance in the matrix in g/cm^2
- s_i = the mass attenuation coefficient for Pu at energy i
- t_{eff} = the effective attenuation distance in Pu in g/cm^2

Rearranging to a linear equation one obtains:

$$\ln A_i = \ln A^* - \mu_i x_{\text{eff}} - s_i t_{\text{eff}}$$

a standard linear equation with three unknowns.

The measured Pu counting rates at 129.3, 203.55, 345.01, and 413.71 keV are converted to uncorrected activity values using the unattenuated efficiency table values, and the set of four equations solved by standard least squares techniques to determine the attenuation distances x_{eff} and t_{eff} . The system documentation does not indicate whether this is an error-weighted fit. If it is not, it should be.

When this approach works, it works quite well. However, in certain pathological cases it results in erroneous and even negative values for one of the distance parameters. This is not entirely unexpected for the following reasons.^a

An old "rule-of-thumb" is that for least squares fitting one should have about twice as many data points as one has parameters to determine. In the SGRS analysis we have four data points from which we are determining three parameters. If any single value is in error the effect can be catastrophic. For example, when the spectrum taken on Drum 003181 using detector A3 was analyzed, an unresolved interference caused the value of $A_{203.55}$ to be higher than any of the other three lines used in the attenuation analysis. (If all is correct, the effect of attenuation

^a This section draws heavily on discussions with R. G. Helmer and Y. D. Harker. Their assistance is gratefully acknowledged.

should be greatest at the lowest energy of 129.3 keV and least at 413.7 keV.) This anomaly caused the x_{eff} to be calculated as $-10 \pm 30 \text{ g/cm}^2$. The attenuation correction determination might be rendered more robust by the inclusion of additional ^{239}Pu lines in the fitted set. The FRAM code, for example, uses the four lines that are presently used in the SGRS along with the ^{239}Pu lines at 143.4+144.2, 171.3, 179.2, 189.3, 195.7, 255.4, 297.5, and 375.0 keV to determine a relative efficiency curve. Two warnings are in order. First, there may be unresolved interferences with some of these lines (for example, ^{235}U has a gamma-ray line at 143.76 keV). Any such interferences would have to be identified and decontaminated using an iterative calculation. Second, these additional lines are relatively weak, and requiring them to be found will increase the required assay times.

Particularly in high density matrices, there may be little difference in the attenuation effect of the matrix and the Pu sample self absorption. In such cases, the measured data may not be able to distinguish clearly between the effect of the two attenuators. In the mathematical calculations, this shows up as large relative uncertainties in each of the calculated values and a large negative correlation coefficient between the two. This is the same as saying that the concerted effect of the two parameters can be well determined, but the individual contribution of either to the total effect can't be well determined. In the present SGRS implementation, the cross-correlation coefficient between the estimated x_{eff} and t_{eff} is ignored during the propagation of errors to the final ratio values and to the estimated activity concentrations. This is incorrect and accounts for the higher than expected error limits on the SGRS ratios. The cross correlation coefficient should be available from the fit, and it should be propagated to the final ratio values and the estimated activity concentrations. If the mass ratio (y) is computed as:

$$y = \frac{C_{ij}}{C_{kl}} \cdot \frac{S_l}{S_j} \cdot \frac{BR_{kl}}{BR_{ij}} \cdot \frac{\epsilon_k}{\epsilon_i} \cdot \frac{e^{\mu_i x_{\text{eff}}} e^{s_i t_{\text{eff}}}}{e^{\mu_k x_{\text{eff}}} e^{s_k t_{\text{eff}}}}$$

where the measured counting rate (C), the specific activity (S), the gamma-ray branching ratios (BR), the unattenuated efficiency values (ϵ), and the attenuation parameters are designated for peak energies i and k from isotopes j and l .

Designating the absolute efficiency ratio as R_ϵ then the estimated variance [$\sigma^2(y)$] can be computed as:

$$\frac{\sigma^2(y)}{y^2} = \left(\frac{\sigma_{C_{ij}}}{C_{ij}} \right)^2 + \left(\frac{\sigma_{C_{kl}}}{C_{kl}} \right)^2 + \left(\frac{\sigma_{BR_{kl}}}{BR_{kl}} \right)^2 + \left(\frac{\sigma_{BR_{ij}}}{BR_{ij}} \right)^2 + \left(\frac{\sigma_{R_\epsilon}}{R_\epsilon} \right)^2 +$$

$$(\mu_i - \mu_k)^2 \sigma_{x_{\text{eff}}}^2 + (s_i - s_k)^2 \sigma_{t_{\text{eff}}}^2 + 2(\mu_i - \mu_k)(s_i - s_k) \text{covar}(x_{\text{eff}}, t_{\text{eff}})$$

where the final term propagates the covariance.

One must also be concerned about the uncertainty in the uncertainty values. The three major contributors to the uncertainty in the computed attenuation correction will be the uncertainty in the x_{eff} , the uncertainty in the t_{eff} , and the correlation between these two. If, for example, the squares of these three terms are 100, 150, and -235 respectively, the square of the combined uncertainty is simply computed as $100+150-235 = 15$. However, the cancellation is so

strong that one must be concerned about the accuracy of the uncertainties. Even if the mathematics is carefully done, the result may not be particularly useful.

To illustrate the effect of the covariance term on the final mass ratio uncertainties, I have calculated a few cases to illustrate the effect of the covariance between x_{eff} and t_{eff} . The first was a combustible matrix drum (spectrum A1520916530). This drum contained no ^{235}U . The relative uncertainty in the mass ratio of $^{241}\text{Am}/^{239}\text{Pu}$ calculated from the 125 and 129 keV lines without the covariance term was 4.55%. When the covariance term is considered this error drops very slightly to 4.48%. The small effect is due to the light matrix and the closely spaced line energies. This drum had no ^{235}U , but if it had, and the counting statistics for the 185 and 129 keV lines had been the same as for the 125 and 129, the relative error in the $^{235}\text{U}/^{239}\text{Pu}$ ratio based on these two widely-spaced lines would drop from 4.55% to 3.6% when the covariance term was considered. The effect is more pronounced in more attenuating matrices. Another case was computed for RF Drum 000210319, a content code 480 (mixed metals) drum (spectrum A11521610510). This drum contained ^{235}U ; however, the 185 keV peak had a statistical counting error of 42%. The relative mass ratio uncertainty for the $^{235}\text{U}/^{239}\text{Pu}$ ratio (again from the 185 and 129 keV lines) when the covariance term is ignored calculates as 67%. When the covariance term is correctly propagated, the relative error drops to 57%.

It is very interesting to note that in both of these cases the variance in the t_{eff} parameter is a powerful contributor to the overall variance. For example, in the mixed metals drum, the largest contributor to the relative variance was the 42% uncertainty in the uranium peak area. This uncertainty contributes 18% (0.42^2) to the total variance. The variance contribution from t_{eff} is 21%. A portion (about half) of this contribution is subtracted when the covariance term is properly considered, but it remains a powerful contributor to the calculated system uncertainties. The x_{eff} parameter is much better defined.

The SGRS should make an attempt to identify pathological cases that will result in poor attenuation thickness estimates. One approach is to identify those fits that result in negative values and/or calculated relative errors on x_{eff} and t_{eff} that exceed some chosen threshold value (perhaps 80%). When either of these problems occur, the obvious alternative is to pick one distance, probably x_{eff} and one attenuation coefficient, probably μ_i , and perform the fit with one unknown.

The SGRS could be perhaps be retrofit with a transmission-source system for the measurement of the matrix attenuation. Such a retrofit would require four sources mounted probably in the door of the existing system with a system to expose or shield the sources from the detector's view. A source such as ^{152}Eu that emits gamma-rays in relatively even spaced energy intervals from 122 keV to 1408 keV might be the best choice of source nuclide. An SGRS measurement with the sources exposed could provide an estimate of the overall matrix attenuation, and might allow the SGRS to function as a psuedo-segmented gamma scanner, generating better absolute activity values.

In summary, a great deal of the lack of robustness in the SGRS measurements is probably due to the attenuation correction technique. Careful consideration should be given to increasing the number of ^{239}Pu gamma-ray lines used in this analysis. In cases where either of the estimate attenuation distances are negative or are determined with an unacceptably high error, the case should be recalculated with just the matrix attenuation as the unknown. In cases where

both x_{eff} and t_{eff} are determined, the cross correlation must be calculated and propagated to the final mass ratios and to the concentration estimates. A retrofit to install transmission sources on the SGRS should be discussed with the manufacturer.

Combining Multiple Detector Results

The discussion so far has considered only the results of a single detector. Determining the best way to combine these individual results to a single set of mass ratio values representative of the entire drum was also investigated during this review.

The underlying assumption of not only the SGRS analysis, but also the analysis of the other codes such as FRAM and MGA, is that the radioactive material in the drum is isotopically homogenous. This does not mean that the radioactive material is necessarily homogeneously distributed in the drum matrix, but that when Pu, for example, is present anywhere in the drum it has the same isotopic composition as Pu located elsewhere in the drum. In theory, if different attenuation corrections were calculated for ^{241}Am and ^{235}U , using a set of isotope-specific peaks, the case of separate elemental distributions could be correctly handled. However, in the present SGRS, an attempt to do so for ^{235}U gives very inconsistent results.

The activity distribution in many of the SWEPP drums is vertically distributed. Although the Pu isotopic composition is generally constant, the ^{241}Am and ^{235}U activities can vary substantially from top to bottom in a given drum. There are a number of possible calculational schemes that can be used to determine the overall drum content ratios. None of these alternative schemes appear foolproof. The various techniques are discussed in the following subsections.

The Present SGRS Weighted Average

The SGRS computes the final drum ratios as the inverse variance-weighted average of the four detector results. This is consistent with the assumption that the isotopic ratios of the fissile material remain fixed regardless of its location in the waste drum. The set of four detector results is first tested for "outliers" using the same test that is used in the VAXGAP code to inspect the set of results from a number of different peaks for the same isotope.⁴ The test is based on the significance of the Chi-squared over the degrees of freedom (χ^2/df) parameter (the "reduced" Chi-square). If the value of χ^2/df exceeds four then an outlier is identified, removed from the set, and the remaining values retested. (While the outlier test in the SGRS code is correctly formulated, there is an error in the coded technique to identify the outlier value to remove.) With four values, the probability that random data will result in $\chi^2/\text{df} \geq 4$, is $\approx 0.7\%$ and with three values is $\approx 1.8\%$. When a consistent set of data is obtained, the grand weighted average is calculated as:

$$R_{\text{ave}} = \frac{\left(\sum_i \frac{R_i}{\sigma_i^2} \right)}{\left(\sum_i (1/\sigma_i^2) \right)}$$

and its uncertainty estimated as the larger of:

$$\sigma_{ave} = \left(\frac{1}{\sum_i 1/\sigma_i^2} \right)^{1/2} \quad \text{or} \quad \sigma_{ave} = \left(\frac{1}{\sum_i 1/\sigma_i^2} \right)^{1/2} \cdot (\chi^2/df).$$

The SGRS weighted averaging scheme generally works quite well; however, as might be expected, when the drum contents are fractionated it produces questionable results. For example, if only the bottom detector (A4) measures a $^{241}\text{Am}/^{239}\text{Pu}$ ratio, this ratio is accepted as applicable to the total drum; an approach that results in an elevated estimate of the total drum content. This condition was identified as a problem in NCR 3196. Additional discussion of this approach will be presented after the alternatives are discussed.

The applicability of the outlier test is questionable. An INEL statistician once told me, "Outlier tests generally do a better job of testing your assumptions about your data than they do of testing your data."^b Since we expect the Pu isotopics to be similar throughout the drum, perhaps some form of the outlier test should be preserved for these measurements in order to protect against gross blunders and errors. However, this is not our expectation for the non-Pu isotopic ratios of $^{241}\text{Am}/^{239}\text{Pu}$ and $^{235}\text{U}/^{239}\text{Pu}$. We could choose not to eliminate outliers from the set of $^{241}\text{Am}/^{239}\text{Pu}$ and $^{235}\text{U}/^{239}\text{Pu}$ ratios prior to averaging. Note, however, that choosing not to eliminate these possible outliers means that we do not accept the homogeneity assumption on which this averaging scheme relies. It would be more correct to continue to test all of the ratio sets for outliers (perhaps expanding the acceptable value of the χ^2/df parameter to 5 or even 6), and if outliers are detected abandon this averaging scheme in favor of one of the alternates.

Relative Pu Content Weighting

To avoid the problem of a single mass ratio being propagated to the entire drum contents, Y. D. Harker initially suggested a weighting scheme based on the relative activity of the drum ^{239}Pu detected by each of the four SGRS detectors.^c The proposed technique divides the drum into four vertical segments each viewed by a detector. The mass density and isotopic composition are assumed to be uniform within a zone, but can vary from zone to zone. The activity registered in a given detector is presumed to come only from that detector's viewed zone; there is no "cross talk" between the detectors. The approach then determines a "drum total" mass ratio that, under some conditions can better represent the overall drum contents. The drum total mass ratio can be expressed as:

$$\frac{M_j}{M_o} = \sum_{i=1}^{i=4} \left(\frac{m_{ij}}{m_{io}} \cdot \frac{a_{io}}{A_o} \right)$$

^b Larry Blackwood, *Private Communication*, December, 1995.

^c Y. D. Harker to G. W. Twedell, *Gamma Spec. Averaging Review*, Notegram December 1995

where: M_j/M_o = the drum total mass ratio of isotope j to isotope o
 m_{ij}/m_{io} = the mass ratio of isotope j to isotope o determined by detector i
 a_{io} = the activity of isotope o determined by detector i
 A_o = the total drum activity of isotope o (a_{io} summed over the four detectors)

Most logically, isotope o would be chosen to be ^{239}Pu . Thus each detector's contribution to the total drum ratio is weighted according to the portion of the total ^{239}Pu activity that it detects.

While this technique gives good results in some cases, detector cross talk can result in drum total mass ratios that are too high. A detailed discussion is again deferred until all the alternatives have been discussed.

Spectral Summation

One technique for combining the four detector results into a single result is to sum the four individual spectra, report the total counting time as the sum of the individual times, and then perform the analysis on the summed result. Each spectrum would have to be shifted to a common energy calibration, and there is a concern that this process of "gain shifting" would degrade the peak resolution in the final summed spectrum. During our initial testing of this approach, with a non-optimized algorithm, we noted peak broadening sufficient to be of concern, but probably insufficient to compromise the analysis results. Others have reported^d excellent results using certain summation algorithms.

For the SGRS application, an efficiency table must be developed for the summation spectrum. Mathematically, an efficiency table that is the average of the appropriate efficiencies from the four summed detectors is the correct table; however, this needs to be confirmed by experiment. Additionally, proper calculation of the attenuation correction lengths for the summed spectrum requires careful consideration.

This approach has the significant advantage of markedly decreasing the required counting times. If the SGRS were to implement spectral summing, required counting times could be decreased by a factor of 2 to 4 (depending on the relative effect of the continuum). The importance of this summation effect on the detection of weak spectral peaks is illustrated in the plots reproduced in Appendix C. The first four plots present the individual SGRS detector results (A1-A4) in a spectral region near the 1001 keV peak of ^{238}U (the actual area is not important). In each of these plots, the only noticeable peak is the large peak at 1001 keV. The fifth plot presents the summation spectrum. Two additional peaks are now discernible at 1010 and 1015 keV. The three peaks in the summation spectrum have been successfully fit to a Gaussian function, and the results of these fits are graphically displayed. This feature of significant sensitivity improvement suggests that a detector summation technique should be seriously investigated.

^d B. Gillespie, Canberra, Inc., *Private Communication*, January, 1996.

For the production of a drum total result, the spectral summation technique could have the same difficulty as expected from the relative Pu weighting technique. Detector cross talk, unless properly considered in the efficiency results, might cause elevated results. Detailed discussion is postponed until the evaluation section.

The Matrix Solution

A fourth possible technique for combining the results of the four detectors has also been suggested.[°] It has the disadvantage of requiring a fair amount of modeling work, and a significant change in the SGRS code. However, it might result in significant improvements in both the overall drum ratios and the absolute concentration measurements. Given a set of efficiency values that quantify the efficiency of detectors A1, A2, A3 and A4 for each of their respective drum volumes and for all of the "cross talk" efficiencies (for example the efficiency with which Detector A1 responds to activities in the volume in front of A2, A3, and A4) this approach solves a set of simultaneous equations for the requisite mass ratios and absolute activities.

Defining:

A_i = the measured activity for nuclide "1" in detector i

B_i = the measured activity for nuclide "2" in detector i

ϵ_{ij} = the efficiency response matrix element that connects nuclide 1 source activity in zone j with the detector response of detector i

λ_{ij} = the efficiency response matrix element that connects nuclide 2 source activity in zone j with the detector response of detector i

S_j = nuclide 1 source activity in zone j

P_j = nuclide 2 source activity in zone j

S = the total drum activity of nuclide 1 (sum of S_j over all four zones)

P = the total drum activity of nuclide 2 (sum of P_j over all four zones)

$$A_i = \sum_j \epsilon_{ij} S_j \quad \text{and} \quad B_i = \sum_j \lambda_{ij} P_j$$

or in matrix notation

$$\mathbf{A} = \boldsymbol{\epsilon} \bullet \mathbf{S} \quad \text{and} \quad \mathbf{B} = \boldsymbol{\lambda} \bullet \mathbf{P}$$

Assuming that $\mathbf{A}$ and $\mathbf{B}$ have inverse matrices:

$$\mathbf{S} = \boldsymbol{\epsilon}^{-1} \bullet \mathbf{A} \quad \text{and} \quad \mathbf{P} = \boldsymbol{\lambda}^{-1} \bullet \mathbf{B}$$

$$S_i = \sum_j \epsilon_{ij}^{-1} A_j \quad \text{and} \quad P_i = \sum_j \lambda_{ij}^{-1} B_j$$

$$S = \sum_i S_i = \sum_i \sum_j \epsilon_{ij}^{-1} A_j \quad \text{and} \quad P = \sum_i P_i = \sum_i \sum_j \lambda_{ij}^{-1} B_j$$

[°] This approach is also Y. D. Harker's

Reversing the order of the summations:

$$S = \sum_j A_j \sum_i \varepsilon_{ij}^{-1} \quad \text{and} \quad P = \sum_j B_j \sum_i \lambda_{ij}^{-1}$$

Then, with the summation indices changed for clarity, the drum ratio of nuclide 1 to nuclide 2 is:

$$R = \frac{S}{P} = \frac{\sum_j A_j \sum_i \varepsilon_{ij}^{-1}}{\sum_n B_n \sum_m \lambda_{mn}^{-1}} \quad \text{or} \quad R = \sum_j A_j \left[\frac{\sum_i \varepsilon_{ij}^{-1}}{\sum_n B_n \sum_m \lambda_{mn}^{-1}} \right]$$

$$R = \sum_j \frac{A_j}{B_j} \left[\frac{B_j \sum_i \varepsilon_{ij}^{-1}}{\sum_n B_n \sum_m \lambda_{mn}^{-1}} \right]$$

Now, defining a detector ratio as $R_j = \frac{A_j}{B_j}$

$$\therefore R = \sum_j R_j w_j \quad \text{Where:} \quad w_j = \frac{B_j \sum_i \varepsilon_{ij}^{-1}}{\sum_n B_n \sum_m \lambda_{mn}^{-1}}$$

$\sum_i \varepsilon_{ij}^{-1}$ is the j^{th} column sum of matrix ε^{-1} and $\sum_m \lambda_{mn}^{-1}$ is the n^{th} column sum of λ^{-1} . Defining these as ε_j^{-1} and λ_n^{-1} then:

$$w_j = \frac{B_j \varepsilon_j^{-1}}{\sum_n B_n \lambda_n^{-1}}$$

Evaluation of the Weighting Alternatives

Consider the assumptions that underlay each of the weighting alternatives. First, let's examine the presently implemented "grand weighted averaging" technique. The underlying assumption is that the material viewed by each detector has the same isotopic composition. Even when the material is actually vertically segregated, cross talk between the separate detectors can average out these differences, and make the spectral results appear more uniform. (Cross talk measurements in a low density combustibles drum indicate that detector A3 is, for example, as efficient in viewing the detector A4 volume as it is in viewing its own.) Since detector cross talk is highest in low density drums, the grand weighted average methodology might provide acceptable results in vertically segregated low density drums.

Alternately, the relative Pu content weighting approach requires that detector cross talk be zero. This assumption is clearly incorrect in low density drums, but could be reasonable in very high density drums. This approach should give elevated mass ratios in low density drums, and more accurate results at high densities. This approach appears to be complementary to the present error-weighting scheme.

It is not clear that the average of all detector efficiencies will provide a result on the summation spectra that properly compensates for detector cross talk. Peak broadening during the spectral summation may impact certain calculated ratios.

The only restrictive assumption in the matrix solution approach is that the isotopic content be homogenous within a volume element. The number of volume elements defined cannot exceed the number of detectors. This approach does require efficiency matrix information for each drum content code and density, thus adding substantially to the calibration complexity. However, this method did not perform well in a test case.

A simple test of these weighting scenarios was conducted. Sources were configured in the combustible matrix calibration drum. In order to decrease the required counting times the sources were placed in source tube 2. This is the tube located halfway between the drum wall and the drum center. The mock-up drum was placed on the SGRS rotator pad with source tube 2 (the 180° position) at the back of the shield facing the detectors, and the rotator was turned off. To acquire the efficiency matrix information a set of four measurements were acquired with ZPPR plate 1932 at four different elevations. These elevations were calculated to center the source plate at each detector elevation. A spectrum was acquired on all four detectors for each source position. The counting time for each of these measurements was 3600 seconds. The source holder was then configured with two Pu- and U-bearing plates (SEFOR 1-032 and 1-029) in the bottom volume segment (centered on detector A4), and one Pu-bearing plate (ZPPR 1932) in the highest volume (centered on detector A1). Using the same drum orientation, this mock-up arrangement was counted for about 8 hours (28,800 seconds). Table 10 details the isotopic loading of this test drum by volume segment.

Table 10. Isotopic loading of the mock-up test drum by volume segment

Isotope	A1		A2		A3		A4		Total	
	Mass(g)	Ratio to ²³⁹ Pu	Mass(g)	Ratio to ²³⁹ Pu	Mass(g)	Ratio to ²³⁹ Pu	Mass(g)	Ratio to ²³⁹ Pu	Mass(g)	Ratio to ²³⁹ Pu
²³⁹ Pu	30.81	100%					26.99	100%	57.80	100%
²⁴⁰ Pu	1.432	4.6%					2.57	9.5%	4.00	6.92%
²⁴¹ Pu	0.016	0.05%					0.06	0.28%	0.076	0.13%
²³⁵ U							0.276	1.02%	0.276	0.48%
²³⁸ U							118.6	440%	118.6	205%
²⁴¹ Am	0.074	0.24%					0.202	0.75%	0.276	0.48%

The spectra acquired during this test were analyzed on the SGRS. The isotopic results calculated by the SGRS from each detector and the overall grand weighted average are presented in Table 11. Additionally, the ²³⁵U/²³⁹Pu mass ratios for the mock-up drum were computed using the relative Pu weighting, spectral summation, and the matrix solution approaches. The detector efficiency values required for the matrix solution were calculated from the individual ZPPR plate

runs.^f The gain shifting algorithm used for the spectral sums was a simple ratio technique and was not optimized to decrease peak broadening. The efficiency values used in the summation calculation were the average values from the individual ZPPR plate runs. These results are compared with the known value in Table 12.

While both the present SGRS error-weighted average and the relative Pu weighting techniques give somewhat elevated results, the spectral summation and matrix methods give low results. This may be partially due to the efficiency value choice for the spectral summation technique; however, this is not of dominant importance. It is noteworthy, the effect that spectral summation has on the precision of the calculated ratio. While the SGRS technique has a

Table 11. SGRS results from the mock-up drum

	A1		A2		A3		A4		Average	
Ratio	Ratio	Error	Ratio	Error	Ratio	Error	Ratio	Error	Ratio	Error
²³⁸ Pu/ ²³⁹ Pu	6.90E-05	9.00E-06	5.30E-05	8.00E-06	1.00E-04	2.00E-05	—	—	6.33E-05	5.73E-06
²⁴⁰ Pu/ ²³⁹ Pu	5.20E-02	2.00E-03	5.50E-02	3.00E-03	7.30E-02	4.00E-03	1.20E-02	3.00E-03	4.68E-02	1.37E-03
²⁴¹ Pu/ ²³⁹ Pu	8.40E-04	3.00E-05	9.00E-04	3.00E-05	1.52E-03	5.00E-05	—	—	9.69E-04	1.95E-05
²⁴¹ Am/ ²³⁹ Pu	3.20E-03	4.00E-04	3.30E-03	2.00E-04	5.00E-03	3.00E-04	6.80E-03	3.00E-04	4.37E-03	1.37E-04
²³⁵ U/ ²³⁹ Pu	—	—	—	—	5.40E-03	8.00E-04	8.00E-03	2.00E-03	5.76E-03	7.43E-04

	Weighted Average		Correct Value	Meas /Corr
Ratio	Ratio	Error	Ratio	
²³⁸ Pu/ ²³⁹ Pu	6.33E-05	5.73E-06	nd	
²⁴⁰ Pu/ ²³⁹ Pu	4.68E-02	1.37E-03	6.92E-02	67.6%
²⁴¹ Pu/ ²³⁹ Pu	9.69E-04	1.95E-05	1.31E-03	73.7%
²⁴¹ Am/ ²³⁹ Pu	4.37E-03	1.37E-04	4.78E-03	91.5%
²³⁵ U/ ²³⁹ Pu	5.76E-03	7.43E-04	4.78E-03	120.6%

Table 12. The results from alternative weighting systems for the ²³⁵U/²³⁹Pu ratio on the mock-up drum

Weighting Method	Calculated		Calculated/Actual
	Ratio	Error	
SGRS (1/σ ²)	5.76E-3	7.4E-4	120%
Relative Pu Content	5.46E-3	8.1E-4	114%
Spectral Summing	3.20E-3	2.8E-4	66%
Matrix solution	2.10E-3	nd	44%

calculated relative error of about 12%, the summation result has an estimated relative error of about 8%.

^f These calculations were done by Y. D. Harker

The performance of the matrix solution approach on this particular problem is also disappointing. This is probably due to poor statistical definition of the ^{235}U activities reported from detectors A1 and A2. Since this simple matrix calculation is not error-weighted, it must derive an answer that satisfies the reported result from each detector without regard to statistical accuracy. A more sophisticated matrix approach that incorporated error-weighting might have far better performance.

Since this is a highly segregated, low-density drum, it is not surprising that SGRS and Relative Pu weighting schemes give elevated results. This may be a problem with no ready solution. We should certainly revisit the matrix solution approach since it theoretically should allow accurate calculations. The technique of spectral summation should be vigorously pursued, not only because it might solve the dilemma of properly combining the individual detector results to an acceptable value, but also because it could substantially decrease the required counting times. Pending the successful development of either the summation or matrix solution techniques, I would suggest that the present weighted averaging scheme be used. Data reviewers need to be aware of the pitfalls in this approach, and prepared to use alternative calculational means on certain problem cases.

CONCLUSIONS

The major observations of this technical review are summarized in this section. The SGRS is capable of accurate performance. This is illustrated by the excellent agreement with the results of the FRAM and MGA codes, two widely accepted Pu isotopic analysis techniques. However, it is less computationally reliable than desired, particularly when assaying low activity drums.

The SGRS detection sensitivity for ^{239}Pu (using the 413 keV line) is about 60 nCi/g in 600 seconds. This detection sensitivity is within a factor of four to seven of the advertised sensitivity of commercial units. However, since isotopic ratio calculations use low to moderate yield gamma-ray peaks, substantial counting times (10^4 to 10^5 seconds) can be required to quantify these ratios to reasonable precision. The counting times could be dramatically decreased (factors of 2 to 4) if an acceptable spectral summation routine was developed and implemented.

The spectral quality assurance on the SGRS should be improved. This can be accomplished either by monitoring the pulser buffers or by periodically transmitting and analyzing the acquiring spectrum.

The SGRS detectors exhibit peak shapes that are poorer than their purchase specifications. While this is not a serious problem, the spectrometer system should be carefully "tuned up" with the intent of improving the peak widths at one-tenth and one-fiftieth the maximum height.

The attenuation correction calculation is the Achilles heel of the system. If things go wrong in the analysis of a single spectrum, this is where they nearly always goes awry. Thus, a

great deal of work focused on the attenuation correction technique. The following points summarize my suggestions:

- If the least squares fit that determines the attenuation lengths is not an error-weighted fit, it should be.
- Look carefully at adding more peaks to the set that are least-squares fit. A rule of thumb for least squares would suggest six rather than four peaks.
- The covariance term between x_{eff} and t_{eff} (the attenuation distances) is ignored in the error propagation. This is a large negative error term that can cancel up to half of the variance contribution from this entire procedure. The failure to consider this error term is probably the reason why SGRS ratio errors (often calculated from the same data) are larger than errors from FRAM or MGA.
- In each of the limited number of variance calculations that I did, the variance contribution from t_{eff} was surprisingly high. If the covariance cancellation was not considered, this was the largest error component.
- The SGRS code must attempt to identify poor results in this very sensitive least squares fitting procedure. I would suggest at a minimum that if either x_{eff} or t_{eff} go negative during the fit process, the data should be refit determining only x_{eff} .

The SGRS combines the individual detector results into a single result using an inverse variance-weighted average. While this technique gives acceptable results most of the time, it overestimates drum contents in cases where only one detector detects an isotope. Some alternate approaches were tested, but none of these significantly out performed the weighted averaging approach in estimating the drum contents.

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-
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- ¹³ L. V. East, *Comparison of INEL and LANL Analyses of Waste Drum Gamma-Ray Spectra*, DRAFT 6/2/92 (reproduced in Appendix B).
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APPENDIX A

Required Counting Time Details

The following spreadsheet outputs detail the required counting time calculations performed for this study. While each output is annotated, the following results are included in the following order.

1. A calculation of the estimated counting times and MDAs for a drum with an activity of weapons grade (WG) at a concentration of 100 α nCi/g with the background data drawn from the ambient background results.
2. A calculation of the estimated counting times and MDAs for a drum with an activity of weapons grade (WG) at a concentration of 100 α nCi/g with the background data drawn from the continuum values in a spectrum of a drum containing about 100 α nCi/g.
3. A calculation of the estimated counting times and MDAs for a drum with an activity of 100 nCi/g of all nuclides with the background data drawn from the continuum values in a spectrum of a drum containing about 100 α nCi/g.
4. A calculation of the estimated counting times and MDAs for a drum with an activity of weapons grade (WG) at a concentration of 1000 α nCi/g with the background data drawn from the continuum values in a spectrum of a drum containing about 1000 α nCi/g.
5. A calculation of the estimated counting times and MDAs for a drum with an activity of 1000 nCi/g of all nuclides with the background data drawn from the continuum values in a spectrum of a drum containing about 1000 α nCi/g.

[illegible]

		239-Pu	129.3	0.031596	0.101475	0.12808	68.7528	2.46E+00	1.80E+01	3.85E+01	1.99E+00			
			144.2	0.030714	0.085608	0.11984	52.4412	2.40E+00	1.15E+01	3.04E+01	1.69E+00			
			161.5	0.029808	0.075555	0.1132	39.834	2.34E+00	8.61E+00	2.52E+01	1.49E+00			
			203.6	0.027636	0.06066	0.1008	22.3002	2.20E+00	5.63E+00	1.77E+01	1.25E+00			
			345	0.02288	0.043875	0.08072	6.831	1.92E+00	3.49E+00	9.98E+00	1.07E+00			
			414	0.021468	0.040491	0.07544	4.93056	1.84E+00	3.17E+00	8.59E+00	1.05E+00			
		240-Pu	160.31	0.029808	0.0756	0.11328	40.4046	2.34E+00	8.62E+00	2.52E+01	1.50E+00			
		241-Pu	148.5	0.030392	0.081675	0.11776	4.869	2.38E+00	1.03E+01	2.87E+01	1.05E+00			
		241-Am	125.29	0.0314	0.10818	0.13152	68.76	2.45E+00	2.18E+01	4.24E+01	1.99E+00			
			146.55	0.0304	0.083385	0.11856	50.274	2.38E+00	1.08E+01	2.93E+01	1.65E+00			
			662.4	0.017612	0.07257	0.06628	0.1306	1.65E+00	7.91E+00	6.61E+00	1.00E+00			
		235-U	186	0.028486	0.065475	0.10544		2.25E+00	6.46E+00	2.02E+01				
		estimated counts/sec at each peak region												
		Isotope	energy	0.2g/cc	0.45g/cc	0.8g/cc								
		238-Pu	152.7	3.25E-04	1.80E-04	1.13E-04								
		239-Pu	129.3	9.19E-02	2.82E-02	2.35E-02								
			144.2	5.35E-03	2.52E-03	1.69E-03								
			161.5	2.47E-03	1.51E-03	9.19E-04								
			203.6	1.25E-02	1.10E-02	6.22E-03								
			345	9.29E-03	1.15E-02	7.15E-03								
			414	2.14E-02	2.80E-02	1.84E-02								
		240-Pu	160.31	1.63E-03	9.97E-04	6.06E-04								
		241-Pu	148.5	4.13E-03	2.15E-03	1.37E-03								
		241-Am	125.29	1.06E-02	2.68E-03	2.45E-03								
			146.55	1.56E-03	7.77E-04	5.07E-04								
			662.4	6.81E-04	3.20E-04	6.80E-04								
		Now we need to estimate the continuum counting rate (Rb). Interferences will be ignored. The ambient bkg rate is roughly 1/5 of the continuum rate.												
		Rates will be determined from the "eyeballed" c/s/ch times the peak width (2*FWHM)												
		data from ambient bkg spec												
				continuum counting rates in c/s			MDA (Ld) in c/s for t=5000s			MDA (Ld) in c/s for t=600s				
	a1513019330	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc				
		238-Pu	152.7	2.40E-02	2.40E-02	2.40E-02	0.01072964	0.01072964	0.01072964	0.033925849				
		239-Pu	129.3	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
			144.2	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
			161.5	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
			203.6	1.80E-02	1.80E-02	1.80E-02	0.009364755	0.009364755	0.009364755	0.029985766				
			345	8.30E-03	8.30E-03	8.30E-03	0.006533106	0.006533106	0.006533106	0.0218115				
			414	7.00E-03	7.00E-03	7.00E-03	0.006043954	0.006043954	0.006043954	0.02039944				
		240-Pu	160.31	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
		241-Pu	148.5	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
		241-Am	125.29	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
			146.55	2.30E-02	2.30E-02	2.30E-02	0.010515139	0.010515139	0.010515139	0.033306639				
			662.4	3.10E-03	3.10E-03	3.10E-03	0.004203414	0.004203414	0.004203414	0.015086257				
		235-U	186	0.018	0.018	0.018	0.009364755	0.009364755	0.009364755	0.029985766				
		bottom line: calc time required to reach MDA from est Rb and est Rn. Equation is:												
		let $x=1/(t)^{.5}$												
		then $2.71*x^2+4.65*(Rb)^{.5}*x-Rn=0$												
		solve for x from quadratic												
		x values in s ^(-.5)												
		counting time in seconds to detect												

[illegible]

[illegible]

[illegible]

data from about 180nCi/g combustible .2g/cc=a1520916530			continuum counting rates in c/s			MDA (Ld) in c/s for t=5000s				
	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc		
	238-Pu	152.7	9.27E-02	9.27E-02	9.27E-02	2.06E-02	2.06E-02	2.06E-02		
	239-Pu	129.3	1.06E-01	1.06E-01	1.06E-01	2.20E-02	2.20E-02	2.20E-02		
		144.2	9.36E-02	9.36E-02	9.36E-02	2.07E-02	2.07E-02	2.07E-02		
		161.5	9.60E-02	9.60E-02	9.60E-02	2.09E-02	2.09E-02	2.09E-02		
		203.6	6.60E-02	6.60E-02	6.60E-02	1.74E-02	1.74E-02	1.74E-02		
		345	1.80E-02	1.80E-02	1.80E-02	9.36E-03	9.36E-03	9.36E-03		
		414	1.10E-02	1.10E-02	1.10E-02	7.44E-03	7.44E-03	7.44E-03		
	240-Pu	160.31	9.60E-02	9.60E-02	9.60E-02	2.09E-02	2.09E-02	2.09E-02		
	241-Pu	148.5	9.40E-02	9.40E-02	9.40E-02	2.07E-02	2.07E-02	2.07E-02		
	241-Am	125.29	1.06E-01	1.06E-01	1.06E-01	2.20E-02	2.20E-02	2.20E-02		
		146.55	9.36E-02	9.36E-02	9.36E-02	2.07E-02	2.07E-02	2.07E-02		
		662.4	4.06E-03	4.06E-03	4.06E-03	4.73E-03	4.73E-03	4.73E-03		
bottom line: calc time required to reach MDA from est Rb and est Rn. Equation is:										
	let $x=1/(t)^{.5}$									
	then $2.71*x^2+4.65*(Rb)^{.5}*x-Rn=0$									
	solve for x from quadratic									
			x values in s ^(-.5)			counting time in seconds to detect				
	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc		
	238-Pu	152.7	2.30E-04	1.27E-04	8.00E-05	1.89E+07	6.20E+07	1.56E+08		
	239-Pu	129.3	5.52E-02	1.81E-02	1.51E-02	3.28E+02	3.07E+03	4.38E+03		
		144.2	3.73E-03	1.76E-03	1.18E-03	7.17E+04	3.21E+05	7.14E+05		
		161.5	1.71E-03	1.05E-03	6.37E-04	3.42E+05	9.13E+05	2.47E+06		
		203.6	1.02E-02	9.01E-03	5.15E-03	9.54E+03	1.23E+04	3.77E+04		
		345	1.40E-02	1.71E-02	1.09E-02	5.08E+03	3.40E+03	8.36E+03		
		414	3.64E-02	4.57E-02	3.20E-02	7.54E+02	4.79E+02	9.79E+02		
	240-Pu	160.31	1.13E-03	6.91E-04	4.20E-04	7.81E+05	2.09E+06	5.67E+06		
	241-Pu	148.5	2.88E-03	1.51E-03	9.58E-04	1.21E+05	4.41E+05	1.09E+06		
	241-Am	125.29	6.94E-03	1.77E-03	1.62E-03	2.08E+04	3.20E+05	3.83E+05		
		146.55	1.10E-03	5.46E-04	3.56E-04	8.31E+05	3.36E+06	7.88E+06		
		662.4	2.25E-03	1.07E-03	2.25E-03	1.97E+05	8.74E+05	1.97E+05		
Estimate the activity measurable (MDA) for each nuclide at each line in a 5000 second count										
100nCi/g continuum										

				MDA (Ld) in c/s for t=5000s	MDA in nCi/g @0.2g/cc in 5000s	MDA in nCi/g @0.45g/cc in 5000s	MDA in nCi/g @0.8g/cc in 5000s				
		Isotope	energy		0.2	4.50E-01	0.8				
		238-Pu	152.7	2.06E-02	82	149	236				
		239-Pu	129.3	2.20E-02	17	55	66				
			144.2	2.07E-02	274	583	869				
			161.5	2.09E-02	600	983	1617				
			203.6	1.74E-02	99	113	199				
			345	9.36E-03	72	58	93				
			414	7.44E-03	25	19	29				
		240-Pu	160.31	2.09E-02	184	302	497				
		241-Pu	148.5	2.07E-02	429	821	1292				
		241-Am	125.29	2.20E-02	27	109	119				
			146.55	2.07E-02	176	354	542				
			662.4	4.73E-03	92	197	92				
Now lets calculate the counting time t required to compute certain ratios to a given fractional error.											
To do this, some important error components will be ignored! These include errors in the attenuation correction, the effeciency, and the BR's. Thus the errors are based on count stats onlyand are for 0.45g/cc drums											
		$t = \frac{1}{(FE)^2} (\sum_i ((R_{ni} + R_{bi})) / (R_{ni})^2)$									
		where FE=fractional error									
Ratio	Peak1			Peak2			Counting time for error of:				
	keV	Rn1 (c/s)	Rb1 (c/s)	keV	Rn2 (c/s)	Rb2 (c/s)	5%	10%	20%		
240/239Pu	160.3	9.97E-04	9.60E-02	161.5	1.51E-03	9.60E-02	1.12E+08	2.79E+07	6.97E+06		
	160.3	9.97E-04	9.60E-02	129	2.82E-02	1.06E-01	7.78E+07	1.95E+07	4.86E+06		
241/239Pu	148.5	2.15E-03	9.40E-02	144.2	2.52E-03	9.36E-02	2.84E+07	7.10E+06	1.77E+06		
	148.5	2.15E-03	9.40E-02	161.48	1.51E-03	9.60E-02	5.03E+07	1.26E+07	3.15E+06		
	148.5	2.15E-03	9.40E-02	129	2.82E-02	1.06E-01	1.65E+07	4.13E+06	1.03E+06		
238/239Pu	152.7	1.80E-04	9.27E-02	129	2.82E-02	1.06E-01	2.29E+09	5.74E+08	1.43E+08		
	152.7	1.80E-04	9.27E-02	144.2	2.52E-03	9.36E-02	2.31E+09	5.77E+08	1.44E+08		
241Am/239P	125.3	2.68E-03	1.06E-01	129	2.82E-02	1.06E-01	1.20E+07	3.01E+06	7.53E+05		
	146.6	7.77E-04	9.36E-02	1.44E+02	2.52E-03	9.36E-02	1.36E+08	3.41E+07	8.53E+06		

[an 23 96 Estimate of required counting times									
Activity Estimate from 100nC/g of each alpha nuclide									
total activity at density (in d/s)									
isotope									
nCi/g									
0.2g/cc									
0.45g/cc									
0.8g/cc									
0.2g/cc									
0.45g/cc									
0.8g/cc									
the g/s rates are total unattenuated emission rates									
g/sec from each quarter at each density									
BR									
energy									
isotope									
238-Pu									
239-Pu									
240-Pu									
241-Pu									
241-Am									
gamma/sec per quarter at ratio energies									
the g/s rates are total unattenuated emission rates									
g/sec from each quarter at each density									
BR									
energy									
isotope									
238-Pu									
239-Pu									
240-Pu									
241-Pu									
241-Am									
Efficiencies from point source eff with cross-talk neglected from table 110									
values here are unattenuated									
Efficiency (c/gamma)									
isotope									
energy									
238-Pu									
239-Pu									
240-Pu									
241-Pu									
241-Am									
125.29									
1.29E-04									

			146.55	1.64E-04							
			662.4	6.35E-05							
		the values in the following table are the mass attenuation coef from EDF-722									
		converted to linear coef in cm ⁻¹ . Also included then is the att correction assuming									
		a drum radius of 28.6 cm									
						t=	0.01	cm			
	x eff=	28.5	cm				Pu for grins				
			table=	11	19	14	21				
				linear atten coeff (cm ⁻¹)				attenuation correction matrix			Pu
		Isotope	energy	0.2	0.45	0.8	18	0.2g/cc	0.45g/cc	0.8g/cc	self abs corr
		238-Pu	152.7	0.030392	0.07965	0.11608	45.5706	2.38E+00	9.68E+00	2.73E+01	1.58E+00
		239-Pu	129.3	0.031596	0.101475	0.12808	68.7528	2.46E+00	1.80E+01	3.85E+01	1.99E+00
			144.2	0.030714	0.085608	0.11984	52.4412	2.40E+00	1.15E+01	3.04E+01	1.69E+00
			161.5	0.029808	0.075555	0.1132	39.834	2.34E+00	8.61E+00	2.52E+01	1.49E+00
			203.6	0.027636	0.06066	0.1008	22.3002	2.20E+00	5.63E+00	1.77E+01	1.25E+00
			345	0.02288	0.043875	0.08072	6.831	1.92E+00	3.49E+00	9.98E+00	1.07E+00
			414	0.021468	0.040491	0.07544	4.93056	1.84E+00	3.17E+00	8.59E+00	1.05E+00
		240-Pu	160.31	0.029808	0.0756	0.11328	40.4046	2.34E+00	8.62E+00	2.52E+01	1.50E+00
		241-Pu	148.5	0.030392	0.081675	0.11776	4.869	2.38E+00	1.03E+01	2.87E+01	1.05E+00
		241-Am	125.29	0.0314	0.10818	0.13152	68.76	2.45E+00	2.18E+01	4.24E+01	1.99E+00
			146.55	0.0304	0.083385	0.11856	50.274	2.38E+00	1.08E+01	2.93E+01	1.65E+00
			662.4	0.017612	0.07257	0.06628	0.1306	1.65E+00	7.91E+00	6.61E+00	1.00E+00
		estimated counts/sec at each peak region									
		Isotope	energy	0.2g/cc	0.45g/cc	0.8g/cc					
		238-Pu	152.7	2.50E-02	1.38E-02	8.71E-03					
		239-Pu	129.3	1.29E-01	3.97E-02	3.31E-02					
			144.2	7.53E-03	3.55E-03	2.38E-03					
			161.5	3.48E-03	2.13E-03	1.29E-03					
			203.6	1.76E-02	1.55E-02	8.76E-03					
			345	1.31E-02	1.62E-02	1.01E-02					
			414	3.01E-02	3.94E-02	2.58E-02					
		240-Pu	160.31	1.13E-02	6.92E-03	4.21E-03					
		241-Pu	148.5	4.83E-03	2.52E-03	1.60E-03					
		241-Am	125.29	8.00E-02	2.02E-02	1.84E-02					
			146.55	1.18E-02	5.84E-03	3.81E-03					
			662.4	5.12E-03	2.41E-03	5.12E-03					
		Now we need to estimate the continuum counting rate (Rb). Interferences will									
		be ignored. The ambient bkg rate is roughly 1/5 of the continuum rate.									
		Rates will be determined from the "eyeballed" c/s/ch times the peak width (2*FWHM)									

data from about 180nCi/g combustible			continuum counting rates in c/s			MDA (Ld) in c/s for t=5000s				
.2g/cc=a1520916530	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc		
	238-Pu	152.7	9.27E-02	9.27E-02	9.27E-02	2.06E-02	2.06E-02	2.06E-02		
	239-Pu	129.3	1.06E-01	1.06E-01	1.06E-01	2.20E-02	2.20E-02	2.20E-02		
		144.2	9.36E-02	9.36E-02	9.36E-02	2.07E-02	2.07E-02	2.07E-02		
		161.5	9.60E-02	9.60E-02	9.60E-02	2.09E-02	2.09E-02	2.09E-02		
		203.6	6.60E-02	6.60E-02	6.60E-02	1.74E-02	1.74E-02	1.74E-02		
		345	1.80E-02	1.80E-02	1.80E-02	9.36E-03	9.36E-03	9.36E-03		
		414	1.10E-02	1.10E-02	1.10E-02	7.44E-03	7.44E-03	7.44E-03		
	240-Pu	160.31	9.60E-02	9.60E-02	9.60E-02	2.09E-02	2.09E-02	2.09E-02		
	241-Pu	148.5	9.40E-02	9.40E-02	9.40E-02	2.07E-02	2.07E-02	2.07E-02		
	241-Am	125.29	1.06E-01	1.06E-01	1.06E-01	2.20E-02	2.20E-02	2.20E-02		
		146.55	9.36E-02	9.36E-02	9.36E-02	2.07E-02	2.07E-02	2.07E-02		
		662.4	4.06E-03	4.06E-03	4.06E-03	4.73E-03	4.73E-03	4.73E-03		
bottom line: calc time required to reach MDA from est Rb and est Rn. Equation is:										
	let $x=1/(t)^{.5}$									
	then $2.71*x^2+4.65*(Rb)^{.5}*x-Rn=0$									
	solve for x from quadratic									
			x values in s ^(-.5)			counting time in seconds to detect				
	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc		
	238-Pu	152.7	1.71E-02	9.60E-03	6.08E-03	3.41E+03	1.09E+04	2.70E+04		
	239-Pu	129.3	7.53E-02	2.51E-02	2.11E-02	1.76E+02	1.58E+03	2.25E+03		
		144.2	5.24E-03	2.48E-03	1.67E-03	3.64E+04	1.62E+05	3.61E+05		
		161.5	2.41E-03	1.47E-03	8.97E-04	1.73E+05	4.61E+05	1.24E+06		
		203.6	1.43E-02	1.26E-02	7.22E-03	4.90E+03	6.31E+03	1.92E+04		
		345	1.93E-02	2.35E-02	1.51E-02	2.67E+03	1.81E+03	4.36E+03		
		414	4.86E-02	6.04E-02	4.28E-02	4.24E+02	2.74E+02	5.46E+02		
	240-Pu	160.31	7.76E-03	4.76E-03	2.90E-03	1.66E+04	4.41E+04	1.19E+05		
	241-Pu	148.5	3.37E-03	1.76E-03	1.12E-03	8.82E+04	3.22E+05	7.95E+05		
	241-Am	125.29	4.86E-02	1.30E-02	1.19E-02	4.23E+02	5.90E+03	7.03E+03		
		146.55	8.14E-03	4.08E-03	2.67E-03	1.51E+04	6.02E+04	1.41E+05		
		662.4	1.52E-02	7.59E-03	1.52E-02	4.34E+03	1.73E+04	4.35E+03		
Estimate the activity measurable (MDA) for each nuclide at each line in a 5000 second count										
100nCi/g continuum										

				MDA (Ld) in c/s for t=5000s	MDA in nCi/g @0.2g/cc in 5000s	MDA in nCi/g @0.45g/cc in 5000s	MDA in nCi/g @0.8g/cc in 5000s				
		Isotope	energy		0.2	4.50E-01	0.8				
		238-Pu	152.7	2.06E-02	82	149	236				
		239-Pu	129.3	2.20E-02	17	55	66				
			144.2	2.07E-02	274	583	869				
			161.5	2.09E-02	600	983	1617				
			203.6	1.74E-02	99	113	199				
			345	9.36E-03	72	58	93				
			414	7.44E-03	25	19	29				
		240-Pu	160.31	2.09E-02	184	302	497				
		241-Pu	148.5	2.07E-02	429	821	1292				
		241-Am	125.29	2.20E-02	27	109	119				
			146.55	2.07E-02	176	354	542				
			662.4	4.73E-03	92	197	92				
All at 100 nCi/g											
Now lets calculate the counting time t required to compute certain ratios to a given fractional error.											
To do this, some important error components will be ignored! These include errors in the attenuation											
correction, the effeciency, and the BR's. Thus the errors are based on count stats onlyand are for 0.45g/cc drums											
		$t=(1/(FE)^2)(SUMi((Rni+Rbi)/(Rni)^2))$									
		where FE=fractional error									
Ratio	Peak1			Peak2			Counting time for error of:				
	keV	Rn1 (c/s)	Rb1 (c/s)	keV	Rn2 (c/s)	Rb2 (c/s)	5%	10%	20%		
240/239Pu	160.3	6.92E-03	9.60E-02	161.5	2.13E-03	9.60E-02	1.88E+07	4.70E+06	1.18E+06		
	160.3	6.92E-03	9.60E-02	129	3.97E-02	1.06E-01	1.72E+06	4.31E+05	1.08E+05		
241/239Pu	148.5	2.52E-03	9.40E-02	144.2	3.55E-03	9.36E-02	1.81E+07	4.52E+06	1.13E+06		
	148.5	2.52E-03	9.40E-02	161.48	2.13E-03	9.60E-02	2.91E+07	7.29E+06	1.82E+06		
	148.5	2.52E-03	9.40E-02	129	3.97E-02	1.06E-01	1.21E+07	3.02E+06	7.54E+05		
238/239Pu	152.7	1.38E-02	9.27E-02	129	3.97E-02	1.06E-01	4.80E+05	1.20E+05	3.00E+04		
	152.7	1.38E-02	9.27E-02	144.2	3.55E-03	9.36E-02	6.48E+06	1.62E+06	4.05E+05		
241Am/239P	125.3	2.02E-02	1.06E-01	1.29E+02	3.97E-02	1.06E-01	2.92E+05	7.30E+04	1.82E+04		
	146.6	5.84E-03	9.36E-02	144.2	3.55E-03	9.36E-02	8.33E+06	2.08E+06	5.21E+05		

Activity Estimate from 1000nCi/g WG Pu (alpha activity)										Activity Estimate from 1000nCi/g WG Pu (alpha activity)									
total activity at density (in d/s)										total activity at density (in d/s)									
nCi/g										nCi/g									
Isotope										Isotope									
238-Pu										238-Pu									
13										13									
1.92E+07										1.92E+07									
4.33E+07										4.33E+07									
0.45g/cc										0.45g/cc									
0.2g/cc										0.2g/cc									
0.45g/cc										0.45g/cc									
0.8g/cc										0.8g/cc									
activity per quarter degment										activity per quarter degment									
gamma/sec per quarter at ratio energies										gamma/sec per quarter at ratio energies									
the g/s rates are total unattenuated emission rates										the g/s rates are total unattenuated emission rates									
g/sec from each quarter at each density										g/sec from each quarter at each density									
BR										BR									
energy										energy									
Isotope										Isotope									
238-Pu										238-Pu									
152.7										152.7									
129.3										129.3									
6.31E-05										6.31E-05									
3.04E-06										3.04E-06									
144.2										144.2									
161.5										161.5									
203.6										203.6									
5.69E-06										5.69E-06									
1460.6										1460.6									
3286.4										3286.4									
8688.8										8688.8									
15446.8										15446.8									
856.7										856.7									
214.2										214.2									
481.9										481.9									
1321.7										1321.7									
2349.6										2349.6									
8070.4										8070.4									
905.5										905.5									
708.6										708.6									
Efficiencies from point source eff with cross-talk neglected from table 1110										Efficiencies from point source eff with cross-talk neglected from table 1110									
values here are unattenuated										values here are unattenuated									
Efficiency										Efficiency									
(c/gamma)										(c/gamma)									
Isotope										Isotope									
238-Pu										238-Pu									
152.7										152.7									
129.3										129.3									
1.36E-04										1.36E-04									
1.61E-04										1.61E-04									
1.79E-04										1.79E-04									
1.84E-04										1.84E-04									
203.6										203.6									
345										345									
1.22E-04										1.22E-04									
414										414									
1.02E-04										1.02E-04									
160.31										160.31									
1.78E-04										1.78E-04									
1.67E-04										1.67E-04									
125.29										125.29									
1.29E-04										1.29E-04									
146.55										146.55									
662.4										662.4									
6.35E-05										6.35E-05									
the values in the following table are the mass attenuation coef from EDF-722										the values in the following table are the mass attenuation coef from EDF-722									
converted to linear coef in cm ⁻¹ . Also included then is the att correction assuming										converted to linear coef in cm ⁻¹ . Also included then is the att correction assuming									

		a drum radius of 28.6 cm											
						t=	0.01	cm					
	x eff=	28.5	cm				Pu for grins						
		table=		11	19	14	21						
				linear atten coeff (cm ⁻¹)				attenuation correction matrix			Pu		
		Isotope	energy	0.2	0.45	0.8	18	0.2g/cc	0.45g/cc	0.8g/cc	self abs corr		
		238-Pu	152.7	0.030392	0.07965	0.11608	45.5706	2.38E+00	9.68E+00	2.73E+01	1.58E+00		
		239-Pu	129.3	0.031596	0.101475	0.12808	68.7528	2.46E+00	1.80E+01	3.85E+01	1.99E+00		
			144.2	0.030714	0.085608	0.11984	52.4412	2.40E+00	1.15E+01	3.04E+01	1.69E+00		
			161.5	0.029808	0.075555	0.1132	39.834	2.34E+00	8.61E+00	2.52E+01	1.49E+00		
			203.6	0.027636	0.06066	0.1008	22.3002	2.20E+00	5.63E+00	1.77E+01	1.25E+00		
			345	0.02288	0.043875	0.08072	6.831	1.92E+00	3.49E+00	9.98E+00	1.07E+00		
			414	0.021468	0.040491	0.07544	4.93056	1.84E+00	3.17E+00	8.59E+00	1.05E+00		
		240-Pu	160.31	0.029808	0.0756	0.11328	40.4046	2.34E+00	8.62E+00	2.52E+01	1.50E+00		
		241-Pu	148.5	0.030392	0.081675	0.11776	4.869	2.38E+00	1.03E+01	2.87E+01	1.05E+00		
		241-Am	125.29	0.0314	0.10818	0.13152	68.76	2.45E+00	2.18E+01	4.24E+01	1.99E+00		
			146.55	0.0304	0.083385	0.11856	50.274	2.38E+00	1.08E+01	2.93E+01	1.65E+00		
			662.4	0.017612	0.07257	0.06628	0.1306	1.65E+00	7.91E+00	6.61E+00	1.00E+00		
		estimated counts/sec at each peak region											
		Isotope	energy	0.2g/cc	0.45g/cc	0.8g/cc							
		238-Pu	152.7	3.25E-03	1.80E-03	1.13E-03							
		239-Pu	129.3	9.19E-01	2.82E-01	2.35E-01							
			144.2	5.35E-02	2.52E-02	1.69E-02							
			161.5	2.47E-02	1.51E-02	9.19E-03							
			203.6	1.25E-01	1.10E-01	6.22E-02							
			345	9.29E-02	1.15E-01	7.15E-02							
			414	2.14E-01	2.80E-01	1.84E-01							
		240-Pu	160.31	1.63E-02	9.97E-03	6.06E-03							
		241-Pu	148.5	4.13E-02	2.15E-02	1.37E-02							
		241-Am	125.29	1.06E-01	2.68E-02	2.45E-02							
			146.55	1.56E-02	7.77E-03	5.07E-03							
			662.4	6.81E-03	3.20E-03	6.80E-03							
		Now we need to estimate the continuum counting rate (Rb). Interferences will be ignored. The ambient bkg rate is roughly 1/5 of the continuum rate.											
		Rates will be determined from the "eyeballed" c/s/ch times the peak width (2*FWHM)											
	data from ambient bkg spec			continuum counting rates in c/s			MDA (Ld) in c/s for t=5000s						
	a1513019330	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc				
		238-Pu	152.7	4.20E-01	4.20E-01	4.20E-01	4.32E-02	4.32E-02	4.32E-02				
		239-Pu	129.3	4.60E-01	4.60E-01	4.60E-01	4.51E-02	4.51E-02	4.51E-02				
			144.2	4.30E-01	4.30E-01	4.30E-01	4.37E-02	4.37E-02	4.37E-02				
			161.5	4.20E-01	4.20E-01	4.20E-01	4.32E-02	4.32E-02	4.32E-02				
			203.6	3.20E-01	3.20E-01	3.20E-01	3.77E-02	3.77E-02	3.77E-02				

			345	8.20E-02	8.20E-02	8.20E-02	1.94E-02	1.94E-02	1.94E-02			
			414	3.30E-02	3.30E-02	3.30E-02	1.25E-02	1.25E-02	1.25E-02			
		240-Pu	160.31	4.20E-01	4.20E-01	4.20E-01	4.32E-02	4.32E-02	4.32E-02			
		241-Pu	148.5	4.30E-01	4.30E-01	4.30E-01	4.37E-02	4.37E-02	4.37E-02			
		241-Am	125.29	4.60E-01	4.60E-01	4.60E-01	4.51E-02	4.51E-02	4.51E-02			
			146.55	4.30E-01	4.30E-01	4.30E-01	4.37E-02	4.37E-02	4.37E-02			
			662.4	1.40E-02	1.40E-02	1.40E-02	8.32E-03	8.32E-03	8.32E-03			
bottom line: calc time required to reach MDA from est Rb and est Rn. Equation is:												
			let $x=1/(t)^{.5}$									
			then $2.71 \cdot x^2 + 4.65 \cdot (Rb)^{.5} \cdot x - Rn = 0$									
			solve for x from quadratic									
				x values in $s^{(-.5)}$			counting time in seconds to detect					
		Isotope	energy	.2 g/cc	.45g/cc	.8g/cc	.2 g/cc	.45g/cc	.8g/cc			
		238-Pu	152.7	1.08E-03	5.97E-04	3.76E-04	8.59E+05	2.81E+06	7.09E+06			
		239-Pu	129.3	2.41E-01	8.35E-02	7.03E-02	1.72E+01	1.44E+02	2.03E+02			
			144.2	1.73E-02	8.20E-03	5.51E-03	3.35E+03	1.49E+04	3.30E+04			
			161.5	8.15E-03	4.99E-03	3.04E-03	1.51E+04	4.01E+04	1.08E+05			
			203.6	4.54E-02	4.01E-02	2.31E-02	4.84E+02	6.22E+02	1.87E+03			
			345	6.20E-02	7.49E-02	4.88E-02	2.60E+02	1.78E+02	4.19E+02			
			414	1.65E-01	2.01E-01	1.47E-01	3.66E+01	2.47E+01	4.60E+01			
		240-Pu	160.31	5.40E-03	3.30E-03	2.01E-03	3.43E+04	9.19E+04	2.49E+05			
		241-Pu	148.5	1.34E-02	7.02E-03	4.47E-03	5.59E+03	2.03E+04	5.01E+04			
		241-Am	125.29	3.28E-02	8.45E-03	7.73E-03	9.30E+02	1.40E+04	1.68E+04			
			146.55	5.11E-03	2.54E-03	1.66E-03	3.84E+04	1.55E+05	3.63E+05			
			662.4	1.17E-02	5.66E-03	1.17E-02	7.30E+03	3.12E+04	7.31E+03			
Estimate the activity measurable (MDA) for each nuclide at each line in a 5000 second count												
1000nCi/g												
				MDA (Ld) in c/s for t=5000s	MDA in nCi/g @0.2g/cc in 5000s	MDA in nCi/g @0.45g/cc in 5000s	MDA in nCi/g @0.8g/cc in 5000s					
		Isotope	energy		0.2	0.45	0.8					
		238-Pu	152.7	4.32E-02	172	312	496					
		239-Pu	129.3	4.51E-02	35	114	136					
			144.2	4.37E-02	580	1231	1837					
			161.5	4.32E-02	1239	2028	3336					
			203.6	3.77E-02	214	244	431					
			345	1.94E-02	148	120	192					
			414	1.25E-02	42	32	48					
		240-Pu	160.31	4.32E-02	380	623	1026					
		241-Pu	148.5	4.37E-02	904	1732	2725					
		241-Am	125.29	4.51E-02	56	224	245					

			146.55	4.37E-02	371	747	1145					
			662.4	8.32E-03	163	346	163					
Now lets calculate the counting time t required to compute certain ratios to a given fractional error.												
To do this, some important error components will be ignored! These include errors in the attenuation correction, the efficiency, and the BR's. Thus the errors are based on count stats onlyand are for 0.45g/cc drums												
			t=(1/(FE)^2)(SUMii((Rni+Rbi))/(Rni)^2)									
			where FE=fractional error									
Ratio	Peak1			Peak2			Counting time in sec for error of:					
	keV	Rn1 (c/s)	Rb1 (c/s)	keV	Rn2 (c/s)	Rb2 (c/s)	5%	10%	20%			
240/239Pu	160.3	9.97E-03	4.20E-01	1.62E+02	1.51E-02	4.20E-01	4.92E+06	1.23E+06	3.07E+05			
	160.3	9.97E-03	4.20E-01	1.29E+02	2.82E-01	4.60E-01	3.43E+06	8.57E+05	2.14E+05			
241/239Pu	148.5	2.15E-02	4.30E-01	1.44E+02	2.52E-02	4.30E-01	1.32E+06	3.30E+05	8.25E+04			
	148.5	2.15E-02	4.30E-01	1.61E+02	1.51E-02	4.20E-01	2.26E+06	5.65E+05	1.41E+05			
	148.5	2.15E-02	4.30E-01	1.29E+02	2.82E-01	4.60E-01	7.67E+05	1.92E+05	4.79E+04			
238/239Pu	152.7	1.80E-03	4.20E-01	1.29E+02	2.82E-01	4.60E-01	1.04E+08	2.60E+07	6.50E+06			
	152.7	1.80E-03	4.20E-01	1.44E+02	2.52E-02	4.30E-01	1.05E+08	2.62E+07	6.54E+06			
241Am/239P	125.3	2.68E-02	4.60E-01	1.29E+02	2.82E-01	4.60E-01	5.32E+05	1.33E+05	3.33E+04			
	146.6	7.77E-03	4.30E-01	1.44E+02	2.52E-02	4.30E-01	6.30E+06	1.58E+06	3.94E+05			
			counting time in seconds to detect				Ratio	Peak1	Peak2	Counting time in sec for error of:		
	Isotope	energy	.2 g/cc	.45g/cc	.8g/cc			keV	keV	5%	10%	20%
	238-Pu	152.7	8.6E+05	2.8E+06	7.1E+06	240/239Pu		160.3	161.5	4.92E+06	1.23E+06	3.07E+05
	239-Pu	129.3	1.7E+01	1.4E+02	2.0E+02			160.3	129	3.43E+06	8.57E+05	2.14E+05
		144.2	3.3E+03	1.5E+04	3.3E+04	241/239Pu		148.5	144.2	1.32E+06	3.30E+05	8.25E+04
		161.5	1.5E+04	4.0E+04	1.1E+05			148.5	161.48	2.26E+06	5.65E+05	1.41E+05
		203.6	4.8E+02	6.2E+02	1.9E+03			148.5	129	7.67E+05	1.92E+05	4.79E+04
		345	2.6E+02	1.8E+02	4.2E+02	238/239Pu		152.7	129	1.04E+08	2.60E+07	6.50E+06
		414	3.7E+01	2.5E+01	4.6E+01			152.7	144.2	1.05E+08	2.62E+07	6.54E+06
	240-Pu	160.31	3.4E+04	9.2E+04	2.5E+05	241Am/239P		125.3	129	5.32E+05	1.33E+05	3.33E+04
	241-Pu	148.5	5.6E+03	2.0E+04	5.0E+04			146.6	144.2	6.30E+06	1.58E+06	3.94E+05
	241-Am	125.29	9.3E+02	1.4E+04	1.7E+04							
		146.55	3.8E+04	1.5E+05	3.6E+05							
		662.4	7.3E+03	3.1E+04	7.3E+03							

Activity calculations at 1000nC/g for everything										total activity at density (in d/s)		activity per quarter degment							
Isotope		nCi/g	0.2g/cc	0.45g/cc	0.8g/cc	0.2g/cc	0.45g/cc	0.8g/cc											
238-Pu	238-Pu	1000	1.48E+09	3.33E+09	5.92E+09	3.70E+08	8.33E+08	1.48E+09											
239-Pu	239-Pu	1000	1.48E+09	3.33E+09	5.92E+09	3.70E+08	8.33E+08	1.48E+09											
240-Pu	240-Pu	1000	1.48E+09	3.33E+09	5.92E+09	3.70E+08	8.33E+08	1.48E+09											
241-Pu	241-Pu	1000	1.48E+09	3.33E+09	5.92E+09	3.70E+08	8.33E+08	1.48E+09											
241-Am	241-Am	1000	1.48E+09	3.33E+09	5.92E+09	3.70E+08	8.33E+08	1.48E+09											
gamma/sec per quarter at ratio energies										the g/s rates are total unattenuated emission rates									
Isotope	energy	BR	0.2g/cc	0.45g/cc	0.8g/cc	g/sec from each quarter at each density													
238-Pu	152.7	9.37E-06	3466.9	7800.5	13867.6														
239-Pu	129.3	6.31E-05	23347.0	52530.8	93388.0														
	144.2	3.04E-06	1123.0	2526.6	4491.8														
	161.5	1.23E-06	455.1	1024.0	1820.4														
	203.6	5.69E-06	2105.3	4736.9	8421.2														
	345	5.56E-06	2057.2	4628.7	8228.8														
	414	1.47E-05	5439.0	12237.8	21756.0														
240-Pu	160.31	4.02E-06	1487.4	3346.7	5949.6														
241-Pu	148.5	1.86E-06	687.8	1547.6	2751.3														
241-Am	125.29	4.10E-05	15170.0	34132.5	60680.0														
	146.55	4.60E-06	1702.0	3829.5	6808.0														
	662.4	3.60E-06	1332.0	2997.0	5328.0														
Efficiencies from point source eff with cross-talk neglected from table 1110																			
Values here are unattenuated																			
	Isotope	energy																	
	238-Pu	152.7	1.72E-04																
	239-Pu	129.3	1.36E-04																
		144.2	1.61E-04																
		161.5	1.79E-04																
		203.6	1.84E-04																
		345	1.22E-04																
		414	1.02E-04																
	240-Pu	160.31	1.78E-04																
	241-Pu	148.5	1.67E-04																
	241-Am	125.29	1.29E-04																

			146.55	1.64E-04							
			662.4	6.35E-05							
		the values in the following table are the mass attenuation coef from EDF-722									
		converted to linear coef in cm ⁻¹ . Also included then is the att correction assuming									
		a drum radius of 28.6 cm									
						t=	0.01	cm			
	x eff=	28.5	cm				Pu for grins				
			table=	11	19	14	21				
				linear atten coeff (cm ⁻¹)				attenuation correction matrix			Pu
		Isotope	energy	0.2	0.45	0.8	18	0.2g/cc	0.45g/cc	0.8g/cc	self abs corr
		238-Pu	152.7	0.030392	0.07965	0.11608	45.5706	2.38E+00	9.68E+00	2.73E+01	1.58E+00
		239-Pu	129.3	0.031596	0.101475	0.12808	68.7528	2.46E+00	1.80E+01	3.85E+01	1.99E+00
			144.2	0.030714	0.085608	0.11984	52.4412	2.40E+00	1.15E+01	3.04E+01	1.69E+00
			161.5	0.029808	0.075555	0.1132	39.834	2.34E+00	8.61E+00	2.52E+01	1.49E+00
			203.6	0.027636	0.06066	0.1008	22.3002	2.20E+00	5.63E+00	1.77E+01	1.25E+00
			345	0.02288	0.043875	0.08072	6.831	1.92E+00	3.49E+00	9.98E+00	1.07E+00
			414	0.021468	0.040491	0.07544	4.93056	1.84E+00	3.17E+00	8.59E+00	1.05E+00
		240-Pu	160.31	0.029808	0.0756	0.11328	40.4046	2.34E+00	8.62E+00	2.52E+01	1.50E+00
		241-Pu	148.5	0.030392	0.081675	0.11776	4.869	2.38E+00	1.03E+01	2.87E+01	1.05E+00
		241-Am	125.29	0.0314	0.10818	0.13152	68.76	2.45E+00	2.18E+01	4.24E+01	1.99E+00
			146.55	0.0304	0.083385	0.11856	50.274	2.38E+00	1.08E+01	2.93E+01	1.65E+00
			662.4	0.017612	0.07257	0.06628	0.1306	1.65E+00	7.91E+00	6.61E+00	1.00E+00
		estimated counts/sec at each peak region									
		Isotope	energy	0.2g/cc	0.45g/cc	0.8g/cc					
		238-Pu	152.7	2.50E-01	1.38E-01	8.71E-02					
		239-Pu	129.3	1.29E+00	3.97E-01	3.31E-01					
			144.2	7.53E-02	3.55E-02	2.38E-02					
			161.5	3.48E-02	2.13E-02	1.29E-02					
			203.6	1.76E-01	1.55E-01	8.76E-02					
			345	1.31E-01	1.62E-01	1.01E-01					
			414	3.01E-01	3.94E-01	2.58E-01					
		240-Pu	160.31	1.13E-01	6.92E-02	4.21E-02					
		241-Pu	148.5	4.83E-02	2.52E-02	1.60E-02					
		241-Am	125.29	8.00E-01	2.02E-01	1.84E-01					
			146.55	1.18E-01	5.84E-02	3.81E-02					
			662.4	5.12E-02	2.41E-02	5.12E-02					
		Now we need to estimate the continuum counting rate (Rb). Interferences will									
		be ignored. The ambient bkg rate is roughly 1/5 of the continuum rate.									
		Rates will be determined from the "eyeballed" c/s/ch times the peak width (2*FWHM)									

[illegible]

				MDA (Ld) in c/s for t=5000s	MDA in nCi/g @0.2g/cc in 5000s	MDA in nCi/g @0.45g/cc in 5000s	MDA in nCi/g @0.8g/cc in 5000s				
		Isotope	energy		0.2	0.45	0.8				
		238-Pu	152.7	4.32E-02	172	312	496				
		239-Pu	129.3	4.51E-02	35	114	136				
			144.2	4.37E-02	580	1231	1837				
			161.5	4.32E-02	1239	2028	3336				
			203.6	3.77E-02	214	244	431				
			345	1.94E-02	148	120	192				
			414	1.25E-02	42	32	48				
		240-Pu	160.31	4.32E-02	380	623	1026				
		241-Pu	148.5	4.37E-02	904	1732	2725				
		241-Am	125.29	4.51E-02	56	224	245				
			146.55	4.37E-02	371	747	1145				
			662.4	8.32E-03	163	346	163				
All activities at 1000 nCi/g											
Now lets calculate the counting time t required to compute certain ratios to a given fractional error.											
To do this, some important error components will be ignored! These include errors in the attenuation											
correction, the effeciency, and the BR's. Thus the errors are based on count stats onlyand are for 0.45g/cc drums											
		$t = (1/(FE)^2) \sum ((R_{ni} + R_{bi}) / (R_{ni})^2)$									
		where FE=fractional error									
Ratio	Peak1			Peak2			Counting time in sec for error of:				
	keV	Rn1 (c/s)	Rb1 (c/s)	keV	Rn2 (c/s)	Rb2 (c/s)	5%	10%	20%		
240/239Pu	160.3	6.92E-02	4.20E-01	1.62E+02	2.13E-02	4.20E-01	8.37E+05	2.09E+05	5.23E+04		
	160.3	6.92E-02	4.20E-01	1.29E+02	3.97E-01	4.60E-01	7.92E+04	1.98E+04	4.95E+03		
241/239Pu	148.5	2.52E-02	4.30E-01	1.44E+02	3.55E-02	4.30E-01	8.42E+05	2.11E+05	5.26E+04		
	148.5	2.52E-02	4.30E-01	1.61E+02	2.13E-02	4.20E-01	1.32E+06	3.30E+05	8.24E+04		
	148.5	2.52E-02	4.30E-01	1.29E+02	3.97E-01	4.60E-01	5.61E+05	1.40E+05	3.50E+04		
238/239Pu	152.7	1.38E-01	4.20E-01	1.29E+02	3.97E-01	4.60E-01	2.38E+04	5.94E+03	1.49E+03		
	152.7	1.38E-01	4.20E-01	1.44E+02	3.55E-02	4.30E-01	3.05E+05	7.63E+04	1.91E+04		
241Am/239P	125.3	2.02E-01	4.60E-01	1.29E+02	3.97E-01	4.60E-01	1.44E+04	3.59E+03	8.98E+02		
	146.6	5.84E-02	4.30E-01	1.44E+02	3.55E-02	4.30E-01	3.92E+05	9.81E+04	2.45E+04		

APPENDIX B

Comparison of INEL and LANL Analyses of Waste Drum Gamma-Ray Spectra

Draft
L. V. East
June 2, 1992

**Comparison of INEL and LANL Analyses of
Waste Drum Gamma-Ray Spectra**

L. V. East
June 2, 1992

This note compares the results obtained from the INEL gamma-ray assay code for determining plutonium isotopic ratios and the Los Alamos "FRAM" code on spectra obtained from some sludge drums at SWEPP. Both codes analyzed the same data.

1. Drum ID 022790

This drum contains sludge contaminated with weapons grade plutonium having "normal" ^{241}Am content. INEL and LANL analysis results are shown in the following table.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(1.19 \pm 0.06) \times 10^{-4}$	$(0.95 \pm 0.04) \times 10^{-4}$
$^{240}\text{Pu}/^{239}\text{Pu}$	$(6.12 \pm 0.19) \times 10^{-2}$	$(6.34 \pm 0.07) \times 10^{-2}$
$^{241}\text{Pu}/^{239}\text{Pu}$	$(2.38 \pm 0.07) \times 10^{-3}$	$(2.1104 \pm 0.0015) \times 10^{-3}$
$^{241}\text{Am}/^{239}\text{Pu}$	$(1.87 \pm 0.14) \times 10^{-3}$	$(2.031 \pm 0.004) \times 10^{-3}$
$^{235}\text{U}/^{239}\text{Pu}$	---	---

2. Drum ID 003854

This drum contains a small amount of depleted uranium in addition to WG Pu with "normal" Am content. Analysis results are shown in the following table.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(0.98 \pm 0.08) \times 10^{-4}$	$(0.95 \pm 0.08) \times 10^{-4}$
$^{240}\text{Pu}/^{239}\text{Pu}$	$(6.0 \pm 0.2) \times 10^{-2}$	$(5.90 \pm 0.14) \times 10^{-2}$
$^{241}\text{Pu}/^{239}\text{Pu}$	$(2.22 \pm 0.08) \times 10^{-3}$	$(2.129 \pm 0.003) \times 10^{-3}$
$^{241}\text{Am}/^{239}\text{Pu}$	$(1.98 \pm 0.09) \times 10^{-3}$	$(1.707 \pm 0.012) \times 10^{-3}$
$^{235}\text{U}/^{239}\text{Pu}$	$(1.08 \pm 0.08) \times 10^{-2}$	$(1.19 \pm 0.02) \times 10^{-2}$

3. Drum ID 002542

This drum contains some depleted uranium in addition to WG plutonium with normal Am content. Analysis results are shown in the table below.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(1.5 \pm 0.1) \times 10^{-4}$	$(1.5 \pm 0.1) \times 10^{-4}$
$^{240}\text{Pu}/^{239}\text{Pu}$	$(5.8 \pm 0.3) \times 10^{-2}$	$(6.09 \pm 0.19) \times 10^{-2}$
$^{241}\text{Pu}/^{239}\text{Pu}$	$(2.27 \pm 0.07) \times 10^{-3}$	$(2.176 \pm 0.004) \times 10^{-3}$
$^{241}\text{Am}/^{239}\text{Pu}$	$(1.7 \pm 0.2) \times 10^{-3}$	$(2.056 \pm 0.016) \times 10^{-3}$
$^{235}\text{U}/^{239}\text{Pu}$	$(1.34 \pm 0.09) \times 10^{-2}$	$(1.60 \pm 0.04) \times 10^{-2}$

4. Drum ID 014010

Drum contains a large amount of depleted uranium in addition to WG Pu with elevated Am content. Analysis results are shown in the table below.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(2.4 \pm 0.3) \times 10^{-4}$	$(2.4 \pm 0.3) \times 10^{-4}$
$^{240}\text{Pu}/^{239}\text{Pu}$	$(6.8 \pm 0.4) \times 10^{-2}$	$(7.0 \pm 0.4) \times 10^{-2}$
$^{241}\text{Pu}/^{239}\text{Pu}$	$(1.9 \pm 0.1) \times 10^{-3}$	$(2.044 \pm 0.007) \times 10^{-3}$
$^{241}\text{Am}/^{239}\text{Pu}$	$(5.2 \pm 0.3) \times 10^{-3}$	$(5.85 \pm 0.04) \times 10^{-3}$
$^{241}\text{Am}/^{235}\text{U}$	$(3.4 \pm 0.2) \times 10^{-3}$	not measured
$^{235}\text{U}/^{239}\text{Pu}$	1.64 ± 0.03	1.549 ± 0.004

5. Drum ID 000593

Drum contains a large amount of depleted uranium in addition to WG Pu with elevated Am content. Analysis results are shown in the table below.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(8 \pm 2) \times 10^{-4}$	$(6 \pm 3) \times 10^{-4}$
$^{240}\text{Pu}/^{239}\text{Pu}$	not measured	$(8 \pm 5) \times 10^{-2}$
$^{241}\text{Pu}/^{239}\text{Pu}$	$(1.8 \pm 0.2) \times 10^{-3}$	$(2.67 \pm 0.06) \times 10^{-3}$
$^{241}\text{Am}/^{239}\text{Pu}$	$(4.0 \pm 0.5) \times 10^{-3}$	$(3.8 \pm 0.2) \times 10^{-3}$
$^{241}\text{Am}/^{235}\text{U}$	$(5 \pm 1) \times 10^{-4}$	not measured
$^{235}\text{U}/^{239}\text{Pu}$	7.5 ± 0.5	8.53 ± 0.05

6. Drum ID 021442

Drum contains a large amount of depleted uranium and a small amount of WG Pu with greatly elevated Am content. Results are shown below.

Mass Ratio	INEL Results	LANL Results
$^{238}\text{Pu}/^{239}\text{Pu}$	$(9 \pm 4) \times 10^{-4}$	not measured
$^{240}\text{Pu}/^{239}\text{Pu}$	not measured	not measured
$^{241}\text{Pu}/^{239}\text{Pu}$	not measured	not measured
$^{241}\text{Am}/^{239}\text{Pu}$	1.18 ± 0.05	1.004 ± 0.005
$^{241}\text{Am}/^{235}\text{U}$	$(4.27 \pm 0.11) \times 10^{-2}$	$(4.218 \pm 0.005) \times 10^{-2}$
$^{235}\text{U}/^{239}\text{Pu}$	30 ± 5	not measured

7. General Observations

$^{238}\text{Pu}/^{239}\text{Pu}$ values: The agreement between the two analysis codes for this mass ratio is excellent, both in magnitude and relative error values.

$^{240}\text{Pu}/^{239}\text{Pu}$ values: Agreement is also excellent; the FRAM code tends to report a slightly smaller uncertainty value. Since there is only one usable gamma ray from ^{240}Pu , the smaller uncertainty is probably due to the use of multiple (essentially all that are measurable) gamma lines for ^{239}Pu determination.

$^{241}\text{Pu}/^{239}\text{Pu}$ values: Reasonable agreement is present. However, the FRAM code reports much smaller (by an order of magnitude) uncertainty values. This is probably due to the use of all measurable gamma rays for this ratio determination in the FRAM code.

$^{241}\text{Am}/^{239}\text{Pu}$ values: The situation is similar to the $^{241}\text{Pu}/^{239}\text{Pu}$ values.

$^{241}\text{Am}/^{235}\text{U}$ values: The INEL code determines this ratio as a consistency check for the $^{241}\text{Am}/^{239}\text{Pu}$ and $^{235}\text{U}/^{239}\text{Pu}$ values; it was not measured by FRAM except for the high content Am drum. This one value is in excellent agreement with the INEL code's value, but again, a smaller uncertainty was reported by FRAM.

$^{235}\text{U}/^{239}\text{Pu}$ values: The FRAM code did not measure this ratio for the two lowest uranium content drums (not clear why; maybe an operator initiated option) or for the highest content drum, in which case all ratios were measured relative to ^{241}Am . When both codes

measured this ratio, agreement was reasonably good, with FRAM reporting significantly smaller uncertainty values.

In general, the agreement between the two codes is quite good. FRAM reports much smaller uncertainty values for the 241 isotopes and ^{235}U (when measured), and slightly smaller uncertainty values for ^{240}Pu .

APPENDIX C

Individual and Summed Spectral Plots

The five plots included in this appendix include spectral sections from detectors A1 through A4 and a plot of the same section of the summation spectrum. The peaks in the summed spectrum have been fit to highlight their presence.

COUNT M1 ZPPR 1932 AND SEFORS 1-032 AND 1-029

A1612310180

COUNT M1 ZPPR 1932 AND SEFORS 1-032 AND 1-029

A2612310180

COUNT M1 ZPPR 1932 AND SEFORS 1-032 AND 1-029

A3612310180

A4612310180

A1

SUM MATRIX SPECTRA

CENTROID(s):	5155.7	5200.1	5224.6
ENERGY(s):	1001.7	1010.3	1015.0
AREA(s):	24400	85	280
% ERROR(s):	1	48	17

A16123 10180