

To Sample or Not to Sample – An Investigation Into the Uncertainty from Sampling Followed by Laboratory Analysis Versus *In-Toto* Gamma Spectroscopy for Situations of Non-Uniform Radioactivity Distribution - 8211

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ABSTRACT

During the decommissioning of structures and the remediation of soil, material that is known or suspected to be contaminated with radioactivity must be assayed. For many, if not most, of the cases, the radioactive material concentration can be determined or inferred by gamma spectroscopy. In most, if not all, of these cases, the radioactivity is not uniformly distributed. For these situations, is it better to perform *in-toto* measurements of the entire item, or is it better to extract a sample and analyze that sample? For the *in-toto* gamma spectroscopy measurement, the results will not be accurate unless the calibration method perfectly represents the measured item. And since the calibrations normally assume uniformly distributed radioactivity, any non-uniform distribution of radioactivity in the measured material will cause an error in the results. A mathematical program using probabilistic methods has been developed to estimate the uncertainty in *in-toto* gamma spectroscopy measurements. This program has the capability of determining the efficiency of various types of non-uniform radioactivity distributions and to compare it to the uniform distribution efficiency. Repeating this for a large number of random tries allows the estimation of the measurement uncertainty. A new feature has been added to the program to simulate the extraction of a sample from these same non-uniform distributions, to compute the average concentration in this extracted sample, and to compare it to the average concentration in the original item to be measured. Repeating this for a large number of random tries allows the estimation of the sampling uncertainty. These two computations were run on several different “typical” contamination measurement scenarios [200 liter drums, 27 cubic meter “truck” containers, 0.01-10 liter “laboratory” containers, and 3600 square meter soil “*in-situ*” plots]. In all cases examined, the sampling uncertainty was larger than the *in-toto* gamma measurement uncertainty – typically about an order of magnitude larger. This means that *in-toto* measurements are likely better for situations where the activity is not homogeneous, and where gamma spectroscopy is appropriate. If sampling is to be done, then the best results will be when the largest mass of sample is extracted and when it is made up from the largest number of discrete sub-samples. The more extreme the non-uniformity, the more important this is.

INTRODUCTION

It is frequently desired to know the total radioactive content or concentration of materials to support decisions made in D&D and ER projects. For example – “Is the radioactive content of this container [drum, box, truck] such that the material can be left on site, or go to a low cost

radioactive disposal site, or must it go to a high cost radioactive disposal site ?” Or, “Is the concentration of this nuclide in the soil sufficiently low that it can be left in place, or must it be removed and treated as radioactive waste?” In most cases, these containers or this *in-situ* soil is not homogeneous in radioactive concentration – it is more likely characterized by “small” volumes of elevated radioactivity distributed in a matrix that is much lower concentration or non-radioactive.

The common and “acceptable” method to determine the average concentration or the total radioactivity is to extract a sample, and to send that sample to a qualified laboratory for analysis. Normally, a much larger quantity of material than needed is extracted and sent to the laboratory, where it is further sub-sampled, and this sub-sample is what is actually analyzed by the [assumed] high-quality method.

It is well recognized that laboratory analysis of the final sub-sample has some measurement uncertainty. There are many well-documented methods specifying how to compute and report the total uncertainty from the laboratory analysis. But it is generally assumed that the item that was measured was homogeneous.

It is well recognized that for the case of gamma spectroscopy that if the measured item is not homogeneous, and if a homogeneous calibration is assumed [as it normally is] then there will be a bias in the individual result, which will cause an additional uncertainty in a group of individual results. This applies to both the laboratory case where a “small” sample is measured via gamma spectroscopy, as well as the field case where a large item or container is measured *in-situ*. This “well known problem” of inaccuracy in gamma spectroscopy measurements of large items, and the frequent requirement to answer “what is the error if all the radioactivity is at the worst spot ?” has greatly limited the acceptability by regulatory agencies of *in-situ* gamma spectroscopy as the “final official” measurement. These field *in-toto* measurements are frequently referred to as “field screening” measurements, however, the same quality of gamma spectroscopy instrument is normally used in the field as is used in the laboratory for “real” measurements. It is also interesting to note that this same non-uniformity problem so commonly recognized with *in-situ* gamma spectroscopy is generally ignored in the laboratory when sub-sampling is performed followed by gamma spectroscopy.

Whenever the item being measured does not have a uniform distribution of radioactivity, then any assessment of the activity must include all sources of uncertainty. For *in-toto* gamma spectroscopy measurements, this must include the uncertainty in the representativeness of the efficiency calibration. If there is a sampling process involved, then this must include the uncertainty in the representativeness of the sub-sample that is eventually analyzed. Only then can the final decision about the measurement be made with the appropriate degree of confidence.

Both of these types of uncertainty were evaluated in this paper for a series of measurement situations commonly encountered in D&D ER projects, and operational waste assay measurements.

METHOD TO ESTIMATE UNCERTAINTIES

Performing physical experiments to determine uncertainty is not practical due to the difficulty of creating large radioactive non-homogeneous items, and to the inaccuracy of creating large homogeneous items. However these are easily simulated mathematically.

Canberra has developed mathematical efficiency calibration software to accurately determine the counting efficiency of a wide variety of sample configurations. This software has been validated from 45-7000 keV, from zero to 500 meters distance, and in all 360 degrees around the detector. It is widely used, with over 1000 Ge and NaI detectors having been characterized for use with the software [1, 2, 3]. ISOCS is the name of the software version for *in-situ* use, and LabSOCS is the name of the version normally used for laboratory counting situations. This software has been commercially available for nearly 10 years, and approximately 1000 detectors have been delivered to customers for use with this software.

The ISOCS and LabSOCS software when used correctly use can be quite accurate [5-10%] for situations where the physical parameters describing the item being measured are well known, but in the real world of DD and ER and waste assay measurements, that is rarely the case. For *in-situ* measurements it is commonly assumed that the largest component of the total uncertainty is the efficiency calibration uncertainty. Users have long performed “worst case” analyses and used these to estimate an uncertainty, but this is a tedious process and highly variable from user to user. Recently, an accessory to the ISOCS software [ISOCS Uncertainty Estimator, or IUE] has been developed to make this task easier [4, 5, 6]. The user enters into the software those parameters describing the object that are known, and for parameters that are not well known [e.g. density, container wall thickness, container fill height] enters both the range of these parameters [e.g. 0.5 to 1.3 g/cc density] and the nature of the distribution [e.g. uniform, triangular, Gaussian]. The IUE software will then compute the uncertainty of the calibration efficiency, using probabilistic methods.

IUE allows the simulation of common measurement instruments or techniques, e.g. scanning detectors, rotating sample, and multiple detectors counting the same object from different locations or different sides.

IUE can also compute the uncertainty from the non-uniform distribution of radioactivity, which is the portion of the software that will be used here. Several of the ISOCS sample shapes allow the introduction of “hot-spots” of radioactivity. IUE allows these hotspots to be of random size, and allows user to specify either a fixed number of them, or a random number of hotspots. The software can also place the hotspots in defined locations or in random locations.

An additional feature of the IUE software allows it to simulate the extraction of a “representative” sample from the material with the non-uniform radioactivity. The user defines how many individual sub-samples are taken from each container, and the size of each sub-sample. The Sampling program assumes that the user will combine the individual sub-samples from each container for laboratory analysis, and computes the activity concentration in the laboratory composite sample. The difference between the laboratory composite sample concentration and the original material concentration is the sampling bias. This process is repeated a large number of times. The mean sampling bias is then computed, which should approach zero. The standard deviation of these individual sampling biases is the sampling

uncertainty, which should be combined with the sample measurement uncertainty for the Total Propagated Uncertainty of the activity in the original item.

In the following sections, we examine several common measurement scenarios using the IUE software. Each of those evaluations will cover a wide range of matrix volumes, non-uniform distribution factors, and sub-sample sizes. These evaluations will compute the uncertainty expected from the incorrect efficiency calibration in an *in-toto* measurement of a non-uniform distribution where the efficiency calibration is for a uniform distribution. This will be compared with the sampling uncertainty from extraction of various numbers and volumes of sub-samples from the same *in-toto* measurement scenarios.

The 4 scenarios to be considered are:

- 1) 10cc – 10 liters of soil containing from 1 – 1000 radioactive particles, measured with a single detector in a laboratory-type situation
- 2) 200 liter containers of soil with a few [1-5] 10cm diameter lumps of radioactivity; measured with a typical drum waste assay system
- 3) 27 m³ containers of soil with 10 – 1000 20x20x20cm lumps of radioactivity, measured with a typical large container “box” assay system
- 4) 60m x 60m plot of soil with 1 – 100 1m² areas of radioactivity; measured in a typical *in-situ* measurement process

SCENARIO 1 – LABORATORY ASSAY OF SOIL

There are many complexities in radioassay of environmental particulate contamination in soil. It is well known that nuclear plant emissions or weapons testing fallout depositions are not uniformly distributed within the soil matrix, and are extremely non-homogeneous. The laboratory has the choice of measuring the entire quantity received using gamma spectroscopy, or extracting a small sub-sample of it for subsequent measurement. So, is gamma spectroscopy of a large quantity adequately precise, given the associated uncertainty from the non-uniform distribution? Or is extracting a sample followed by homogenization and a more sensitive analysis better?

For this evaluation, it is assumed that the laboratory receives samples of soil that contain very small radioactive particles randomly distributed in otherwise non-radioactive soil. The incoming sample volumes assumed were 10, 100, 1000, and 10000cc. The number of radioactive particles with each volume of soil was 1, 3, 10, 30, 100, 300, and 1000. One assay option is gamma spectroscopy of the total sample. The other assay option is to extract a small random sample for some other kind of assay. Both were simulated here in this computational experiment.

Assay of the total sample via gamma spectroscopy

The sample was assumed to be placed in a cubical sample container with equal sides, and counted directly on the endcap of a mid-sized (40-50% relative efficiency) coaxial HPGe detector. The IUE program was used to determine the uncertainty from the random number and location of the radioactive particles within each assay volume. The energies considered were 60

keV, 200 keV, and 1000 keV, intended to be representative of a wide range of nuclides. A large number of models were created – typically 1000 for the situations with very few particles down to around a hundred models for those with many particles. Each model had an independent random distribution of particles. The standard deviation of the group of models for each volume-particle combination was computed and is presented in the center section of Table 1.

Table 1 Uncertainty for *In-Toto* gamma spectroscopy and sampling

Both Arithmetic standard deviation [ASD] and Geometric standard deviation [GSD] results were computed. Geometric values are used since in gamma spectroscopy, those particles that are far from the detector have lower efficiency due to attenuation in the soil matrix; therefore the distribution of results is not symmetric on a linear scale, but approaches a logarithmic distribution. A GSD is merely an ASD computed using the logarithm of the data. Both can be expressed

as a fraction or as a percentage or as absolute values. While an ASD of 25% means + and - 25%, a GSD of 25% means multiply by 1.25 and divide by 1.25; conversely, 200% GSD means +100% and -50% ASD. GSDs are asymmetric on a linear scale, but at small values this asymmetry is of little consequence.

volume	particles	particles/cc	<i>in-toto</i> % Geometric SD			subsampling	
			60 keV	200 keV	1000 keV	dia cylinder	%ArithSD
10	1	0.1	57	52	42	6.08 mm	115
10	3	0.3	22	20	17	6.08 mm	71
10	10	1	14.6	13.5	11.4	6.08 mm	38
10	30	3	8.4	7.8	6.6	6.08 mm	20
10	100	10	3.6	3.4	2.8	6.08 mm	12
10	300	30	2.6	2.4	2	6.08 mm	7
10	1000	100	1.5	1.4	1.2	6.08 mm	3.8
100	1	0.01	186	117	83	4.14 mm	377
100	3	0.03	76	49	36	4.14 mm	226
100	10	0.1	39	27	20	4.14 mm	121
100	30	0.3	22.6	16.5	12.7	4.14 mm	71
100	100	1	11	8	6	4.14 mm	39
100	300	3	6	4.5	3.5	4.14 mm	21
100	1000	10	2.6	2	1.6	4.14 mm	12
1000	1	0.001	515	228	144	2.82 mm	1525
1000	3	0.003	265	160	84	2.82 mm	686
1000	10	0.01	112	61	42	2.82 mm	394
1000	30	0.03	44	26	18	2.82 mm	229
1000	100	0.1	30	18.5	13	2.82 mm	126
1000	300	0.3	11.4	7	5	2.82 mm	74
1000	1000	1	8.6	5.5	4	2.82 mm	41
10000	1	0.0001	2280	515	242	1.92 mm	
10000	3	0.0003	976	264	133	1.92 mm	
10000	10	0.001	214	99	64	1.92 mm	1127
10000	30	0.003	105	57	39	1.92 mm	750
10000	100	0.01	63	34	23	1.92 mm	435
10000	300	0.03	34	18	12	1.92 mm	221
10000	1000	0.1	20	10	6.5	1.92 mm	118
10000	3000	0.3				1.92 mm	79

The *in-toto* analysis uncertainty results are shown graphically in Figure 1 for each energy and for each assay volume. The *in-toto* data are the various shorter segment lines at the lower portion of the graph. For each particular energy and volume, the data follows the expected square-root trend – a 100-fold increase in particles/cc results in a 10-fold reduction in the standard deviation.

The data also shows two other trends. As the energy gets higher, the uncertainty from non-uniformity gets smaller for the same assay volume. This should be obvious, as the low energy photons are affected by both the random distribution and matrix attenuation, whereas the high energy photons have the same random distribution impact but much less attenuation. But the other trend is somewhat less obvious; as the assay volume gets larger, the standard deviation gets

smaller for the same particle density. This is because there are more total number of particles in the larger assay volumes.

These values are only that component of the uncertainty that is due to non-uniformity. This must be combined with other sources of uncertainty (e.g. counting statistics, calibration uncertainty, ...) to get the total measurement uncertainty of the incoming sample; however with good calibration techniques and when the non-uniformity component is 10-20% or greater, it is probably the dominant component of the TPU.

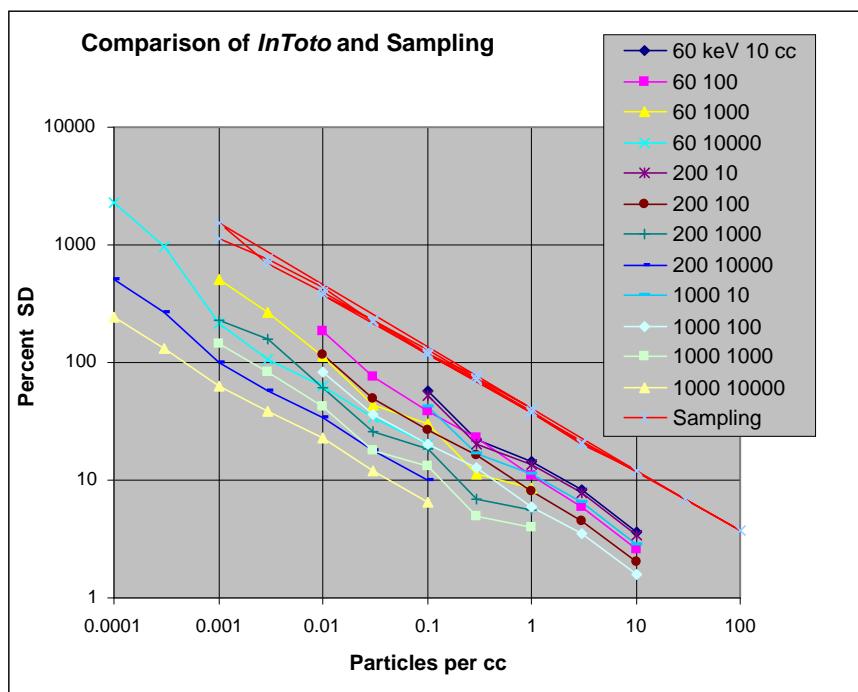


Figure 1 Uncertainty in Laboratory Assay of Soil from *In-toto* Measurement and Sample Extraction

Assay of a portion of the total volume

In this portion of the analysis a random sample is extracted from the incoming total volume and homogenized, as would be the case if alpha spectroscopy was to be performed, for example. Ten grams of soil was assumed to be extracted from the incoming volume. It is reasonable to assume that the chemical preparation and radioactive counting of the prepared sample have rather small and well determined uncertainties. But since the purpose is to know what is in the incoming sample volume, then the sampling uncertainty must also be determined and propagated.

The Sampling function of IUE was used for this task. The sample shape can be spherical, simulating a “spoon-full”, or it can be cylindrical simulating a top-bottom core sample. The user can specify the sample volume, and whether this volume is made up of a single sample or a number of sub-samples. If sub-samples are used, they are assumed to be composited for analysis. In this experiment, 10 cylindrical sub-samples were used. The location of each sample or sub-sample is randomly determined. Each volume-particle/cc group shown in Table 1 was treated separately. One laboratory sample was extracted from each of the 100-1000 models previously created for the *in-toto* simulated gamma spectral analysis. For each laboratory aliquot, the ratio of the activity concentration in the sample to the activity concentration in the incoming total volume was computed and is converted to percent. The relative sampling uncertainty is the standard deviation of these ratios and is presented in right hand columns of Table 1. The upper straight line in Figure 1 shows graphically the uncertainty results from the sampling. The results are what should have been expected. In this case, the sample size is always 10 grams; therefore as the particle concentration gets larger, there are more particles per

sample, and consequently the uncertainty in the sampling error decreases as the square root of the number of particles.

But another perhaps unexpected thing happens. For those incoming volumes with particle densities less than 1 per cc, many of the 10g samples have zero radioactivity – the sample that was extracted for the “better” analysis did not contain any radioactive particles. At 0.1 particles/cc, about 50% of the samples have zero particles, and thus zero [or MDA] activity would be reported. Consequently, the example alpha spectroscopy report would infer that half of the containers would also have zero [or MDA] radioactivity, if the sampling uncertainty is ignored.

Comparison between In-toto assay and Sampling uncertainties

So, which is better -- *In-toto* assay or Sampling and assaying a portion? For all cases in this experiment, the *in-toto* measurement error component from non-uniform distribution was smaller than the sampling error component for the subsequent “better” analysis.

SCENARIO 2 – 200 LITER DRUM OF SOIL

Assay of the total sample via gamma spectroscopy

This is another common measurement scenario. In this scenario, there exist a large number of 200 liter drums filled with soil, at an average density of 1.2 g/cc. The radioactivity in the soil is known to be quite non-uniform. Based upon field scanning data, it is believed that the radioactive soil is contained in grapefruit-sized nodules [hotspots] which are interspersed in non-radioactive soil of the same composition and density. The nuclides of interest have energies of 60 keV and 1000 keV.

What is the optimum counting geometry if the purpose is to minimize the total uncertainty of the drum assay? The largest contribution to the uncertainty is the number and location of the radioactive hotspots in the drum. Therefore all other items were considered “well-known” and were not varied. The variables were simply the number of radioactive sources per drum. Situation one assumed that there were 1-5 radioactive hotspots per drum, all values equally probable, and all sources randomly distributed. Situation two assumed that there were 10-20 hotspots per drum.

The counting geometry variables that were investigated were distance from the side of the drum [20cm, and 100cm], counting from a single side or from two sides of the drum, fixed or vertically scanning detectors, and stationary or rotating drum. These represent techniques that could be done in the field with a single detector, and also represent commonly used geometries in drum assay systems.

A detailed investigation was done in reference X and is summarized here in Table 2. In these analyses, especially at the 60 keV energy, the data are disproportionately distributed on the low energy side of the mean. A skewness evaluation indicates that the geometric standard deviation is the more proper one to use. As the standard deviation is improved, either by better geometry

or higher energy or more hotspots, the skewness decreases and the two standard deviation measures approach each other. Both are presented here for comparison.

Table 2 Uncertainty for 200 Liter Drum with Hotspots						
distance	motion	hotspots	60 keV		1000 keV	
			%sdA	sdG	%sdA	sdG
20cm	stationary	1-5	256	28.00	81	2.44
20cm	scanning	1-5	300	27.00	93	2.50
100cm	stationary	1-5	184	18.50	60	2.02
20cm	rotate 180deg	1-5	167	6.33	43	1.49
100cm	rotate 180deg	1-5	115	3.94	25	1.28
20cm	rotating	1-5	88	2.80	22	1.24
20cm	scan+rotate	1-5	85	3.24	24	1.27
100cm	rotating	1-5	89	3.30	23	1.26
20cm	stationary	10-20	71	2.10	20	1.23
20cm	scanning	10-20	70	2.31	22	1.25
100cm	stationary	10-20	48	1.73	15	1.17
20cm	rotate 180deg	10-20	46	1.63	11	1.12
100cm	rotate 180deg	10-20	37	1.50	8	1.09
20cm	rotating	10-20	30	1.40	8	1.08
20cm	scan+rotate	10-20	28	1.36	8	1.08
100cm	rotating	10-20	20	1.24	5	1.05
			+ -	× ÷	+ -	× ÷

Several trends can be seen from the data.

- Low energies have considerably higher standard deviation than high energies.
- A detector up close at 20cm has the highest standard deviation;
- Scanning the detector up and down the full drum height doesn't improve the standard deviation very much for this situation where the radioactivity is randomly distributed, but might be useful if there were the potential for the hotspots to settle;
- Moving the detector back to 100 cm definitely helps, but also reduces the efficiency a factor of 2 at low energies and 4 at high energies, and therefore will increase the counting statistic component of the total propagated uncertainty;
- Keeping the detector at 20cm and rotating it 180 degrees half-way through the count is even better and retains the high efficiency;
- Rotating the drum 180 degrees half-way through the count with the detector at 100cm is somewhat better;
- Continuously rotating the drum during the count is the best, and it is somewhat better at 100cm than 20cm, but doesn't matter very much if the detector is scanning;
- Increasing the number of hotspots dramatically reduces the standard deviation for all geometries and for all energies.

The biggest impact on the efficiency uncertainty estimate is having more hotspots in the drum. At 60 keV, if there are 1-5 hotspots, the uncertainty is a factor of 28, while if a reasonable assumption can show that there are 10-20 hotspots in the drum, then the uncertainty is only a factor of 2 for the simple and efficient 20cm stationary count, and down to 30-40% with the better geometries. At high energies, even when up close, the uncertainty is a factor of 2-3 for the simple up-close stationary count, reducing down to a 5-10% with the better geometries.

As a commentary – is it really necessary to have a very low standard deviation? No – but what IS required is to accurately present the quality of the result so that the proper interpretation can be made. Using the above case as an example, if the measurement result for Am-241 at 60 keV was a factor of 100 below the “limit” then even the quick simple 20cm stationary measurement would be adequate to prove that the item is “acceptable”. If most of the samples are like this, then this simple geometry is a good one to use. If then a few of the samples have results closer to the limit, then those few could be recounted in a more precise method – perhaps on a rotating platform at 100cm.all-sized clumps of contamination exist.

Assay of a portion of the total volume

An alternate assay method is to extract a “representative” sample from each container for a subsequent laboratory assay. The two examples in Table 2 [1-5 hotspots, 10-20 hotspots] were used with IUE to evaluate various sampling regimes.

A typical sampling process was simulated using a 2cm diameter coring tool, which extracts a top-bottom section of the drum. The number of cores taken per drum was varied from 1 per drum to 100 cores per drum. A composite of all the cores for that drum was assumed to be analyzed. The left graph in Figure 2 plots the total mass of sample extracted from each container [277 grams per core] vs. the arithmetic standard deviation of the sampling process. The graph shows the sampling uncertainty for the 1-5 hotspot case and the 10-20 hotspot case. It also shows the sampling uncertainty for the situation where only 1 sub-sample was taken but where the diameter was the variable. In all three cases, the larger the mass of sample extracted from the container for subsequent homogenization and analysis, the smaller the sampling uncertainty. And, the larger the number of hotspots in the drum, the smaller the sampling uncertainty.

The graph on the right side of Figure 2 shows the situation where the amount of sample extracted was held at a constant mass [10kg and 1 kg], but the number of sub-samples making up that total was varied. This shows that the more sub-samples that make up the total sample extracted, the better the sampling uncertainty. However, while this works in theory, there is a practical limit as taking core samples much smaller than 1cm is very difficult. A 1cm diameter core lower limit means 3 cores maximum for a 1kg sample and 30 cores maximum for a 10kg sample.

Comparison between In-toto assay and Sampling uncertainties

Probably the most common field assay method for DD/ER measurements is to a rotating drum assay or 2-sided assay with the detector at 1 meter. This would have a calibration component of the measurement uncertainty at about 25% for high energy photons [e.g. U-238, Cs-137, Co-60] and much higher for low energy photons [several hundred percent for Am-241]. For waste assay

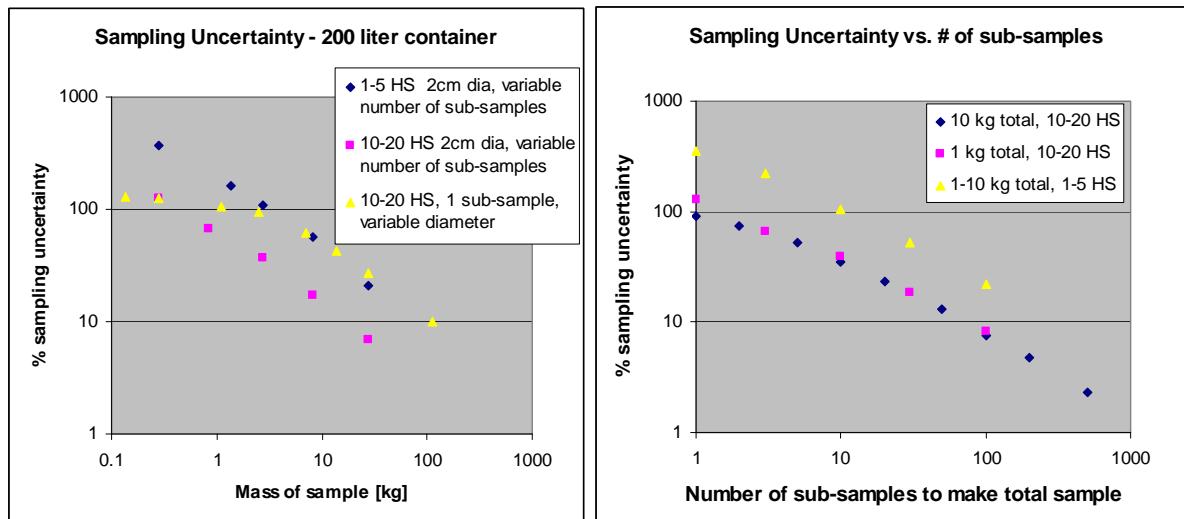


Figure 2 Sampling Uncertainty for 200 liter Drums

systems, which use scanning detectors and a rotating sample, these uncertainties are considerably lower [<10% for high energy, 20-30% for Am-241 at 60 keV].

A common sampling method is to extract several core samples per drum, totaling approximately 1 kg. For the 1-5 hotspot per drum example this would mean a sampling uncertainty of approximately 200%; for the 10-20 hotspot per drum example, the uncertainty improves to about 100% if only 1 core is taken and about 80% if 4 cores are taken.

In order to approach the 20-30% low energy *in-toto* measurement uncertainty, the sampling would have to extract and subsequently homogenize between 10-30 kg of sample. Note that merely extracting a large sample does not reduce the sampling uncertainty unless the subsequent analysis can analyze the total sample without further sub-sampling, unless the full 10-30 kg is homogenized before any further sub-sampling.

Alternately, take a very large number of small sub-samples. If only 1 kg of sample can be homogenized, and 20-30% sampling accuracy is desired then either 30 [10-20 hotspots] or 100 subsamples [1-5 hotspots] must be taken.

But with the practical limit of 1cm diameter for a coring tool, the best that can be done is 60% uncertainty for the 1kg sample case down to 20% for the 10kg sample case, when there are 10-20 hotspots per drum. For 1-5 hotspots per drum, the best that can be done is 200% at 1 kg down to 50% at 10 kg sample size.

So, like the previous scenario, common *in-toto* measurements have lower uncertainties than practical sampling strategies.

SCENARIO 3 – TRUCK OR LARGE CONTAINER OF SOIL

Here we assumed that field screening measurements have detected soil contamination that must be removed for disposal. These measurements also indicated that while most of the soil is not contaminated, basketball-sized clumps of contamination exist. The soil has been excavated and

placed into a large vehicle that holds 27 cubic meters [32 metric tons]. The size of the container assumed here was 2.4m [8'] wide x 6.1m [20'] long and filled to a depth of 1.8m [6']. Two disposition alternatives exist – a low cost one for low concentration material, otherwise a high cost one. The purpose of the assay is to determine which disposition alternative to use. Which is the better method to assay the vehicle contents – *in-situ* measurements or sending a sample to an excellent laboratory?

In-situ gamma spectroscopy assay method of total vehicle

The field assay was assumed to be conducted using gamma spectroscopy. The detector was placed 1 meter from the vehicle. Six spectra were taken along the 20' side of the vehicle, 3 on one side of the vehicle and 3 on the other. The spectra were added together for a single analysis. The vehicle was assumed to contain 10, 30, 100, 300, or 1000 hotspots, with the rest of the soil uncontaminated. The radioactive volume percentage of the total volume thus varied from 0.31% to 31%. In a similar manner as in the preceding analysis, many randomly generated calibration models were generated and the efficiency uncertainty evaluated at 100, 200, 600, and 1000 keV. Table 3 shows the results. For the 10 hotspot per vehicle situation, the measurement uncertainty is very high [a factor of 13] for low energy nuclides, as expected, but decreases to a factor of 1.6 for high energy nuclides.

As the number of hotspots and therefore the percentage volume occupied by those hotspots increases, the uncertainty decreases, to where it is negligible compared to other uncertainties at the 300-1000 hotspot range.

Table 3 Calibration Uncertainty from *In-toto* Measurement of Very Large Container

Number of hotspots	% Geometric SD				Hotspot %volume
	100 keV	200 keV	600 keV	1000 keV	
10	1300	660	280	160	0.31
30	213	144	88	63	0.92
100	48	40	31	25	3.1
300	22	18	14	11	9.2
1000	7.2	5.1	3.4	2.5	31

Vehicle sampling uncertainty

For laboratory analysis, it was assumed that 40 lbs of soil would be extracted and sent back to the laboratory for homogenization and analysis. Several different sampling strategies were evaluated. These results are presented in Table 4. Four different sampling strategies were evaluated: 15 each 1" diameter cores, 4 each 2" diameter cores, 15 each 5" diameter spheres, and 4 each 8" diameter spheres. The samples were randomly taken and the concentration of radioactivity in the composite sample compared to the concentration of radioactivity in the container. This process was repeated a large number of times and the uncertainty computed, as presented in Table 4. Increasing the number of hotspots reduces the

Table 4 Sample Extraction Uncertainty from Very Large Container

Number of hotspots	% Arithmetic SD			
	1"Cx15	2"Cx4	5"Sx15	8"Sx4
10	156	298	368	539
30	88	158	185	292
100	49	82	109	180
300	25	46	57	86
1000	11.5	19.5	28.2	34.4

uncertainty by approximately the square root of the increase.

As expected, the sampling uncertainty is high for the vehicles with few hotspots and gets much lower when there are more hotspots. The data also indicates that the more samples per vehicle that are composited together, the lower the uncertainty. Increasing the number of sub-samples from 4 to 15 reduces the uncertainty approximately a factor of 2. Somewhat unexpectedly, going from a spherical sampling method to a cylindrical full-height sampling method, but keeping the number of samples the same also reduces the sampling uncertainty a factor of 2.

Comparison between *in-situ* assay and sampling uncertainties

So, which is better -- *in-situ* assay of the vehicle, or extracting and assaying a portion? For most all situations at 30 hotspots and more, the *in-situ* uncertainty is less than the sampling uncertainty. This is even more so for high energies, and for the worse-performing sampling strategies. However, for the 10-30 hotspot range, both methods have a quite high uncertainty [hundreds of percent] and the choice might depend upon sampling strategy and gamma energy. However, any practical sampling strategy of a sparsely populated non-uniform material will frequently fail to extract any part of the radioactivity, and therefore give a result of zero for the laboratory analysis – falsely indicating that the container has no radioactivity. Those figures are shown in Figure 3. For the 10-30 hotspot range, the samples have 50% or more chance of showing zero radioactivity. This should probably lead one to conclude that the *in-situ* technique is to be preferred.

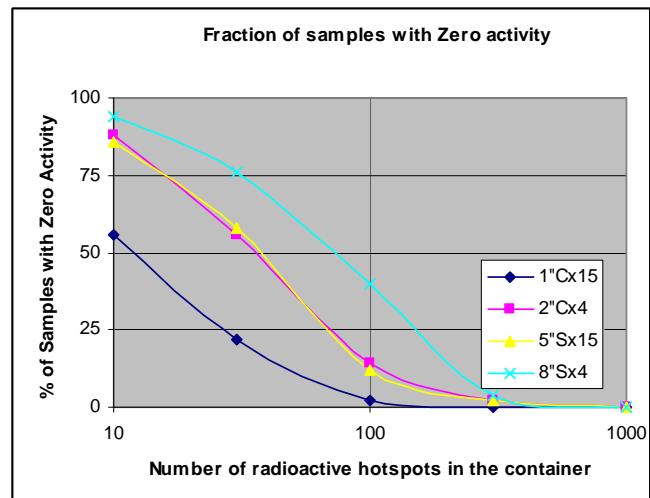


Figure 3 Fraction of samples with zero radioactivity

SCENARIO 4 – 3600 M² OF CONTAMINATED SOIL

For this scenario, there is an area of soil that could potentially be considered as contaminated. The soil area assumed here was 60m x 60m for a total area of 3600 square meters. Due to past actions at the site, there are small areas [assumed to be 1m² here] that are contaminated, however the bulk of the land area is not contaminated. The contamination is assumed to be in a layer extending from the surface down to 15cm.

Measurements are needed to assess the average radioactivity of the 3600 m² area for the purpose of comparison of the average activity to release limits. One method of assay is to perform *in-situ* gamma spectroscopy, positioning a gamma spectroscopy detector several meters from the ground and repeating this measurement in a grid pattern over the entire area. An alternate method would be to extract a group of samples from random locations for subsequent laboratory analysis.

Assay of the total area via *in-situ* gamma spectroscopy

A typical set of *in-situ* measurements involves placing the detector at a fixed distance from the ground and acquiring a spectrum. This process is then repeated over a grid pattern to cover the entire survey area. The average concentration for the survey unit is determined by the average of the concentrations from each detector. The individual readings can also be used to locate the general area of elevated activity, if needed. For the average activity, two primary variables are suspected to affect the measurement uncertainty – detector grid spacing, and detector-surface spacing.

If the detectors are spaced 30 meters apart in a rectangular grid pattern, then 9 measurements will cover the survey area. If the grid spacing is reduced to 20 meters, then 16 measurements will be made. If the grid spacing is reduced to 10 meters, then it will take 49 measurements to cover the area.

Computations of the *in-situ* uncertainty were performed at gamma energies of 60, 200, and 1000 keV and the results are presented in Figure 4. For this measurement scenario, the uncertainty is dominated by geometrical factors, rather than attenuation, therefore there was very little difference between the 3 energies. The 200 keV results are shown; the 60 keV uncertainty is about 1.2x and the 1000 keV uncertainty is about 0.8x the values presented here.

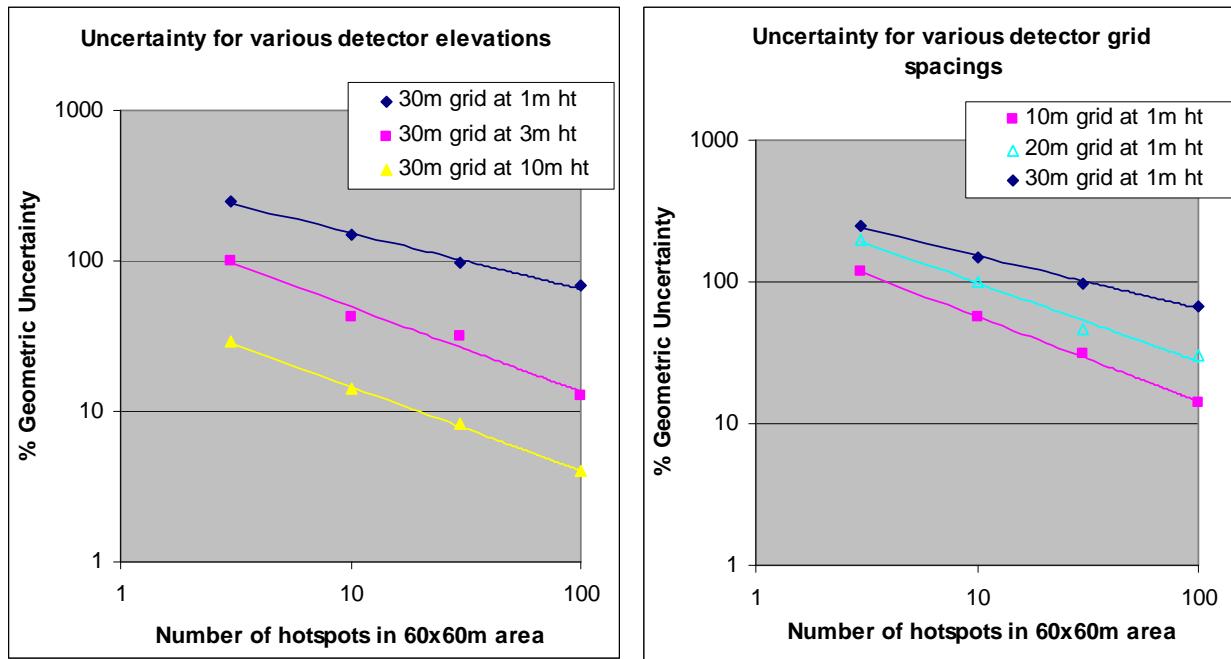


Figure 4 *In-situ* soil measurement uncertainty under various detector and distance configurations

The left graph of Figure 4 shows the uncertainty vs. the number of hotspots in the 3600 square meter area for the 30 m grid spacing at 3 different detector elevations – 1, 3, and 10 meters. The graph on the right shows the uncertainty vs. the number of hotspots for a detector at 1 meter elevation and for 3 different grid spacings – 10m, 20m, and 10m.

Observations from these *in-situ* simulated measurements:

- The uncertainty as a function of the number of hotspots generally follows the square-root function – as the number of hotspots per unit area increases a factor of 4, the uncertainty is reduced a factor of 2.
- Decreasing the grid spacing from 30m to 20m [from 9 to 16 measurements] only reduces the uncertainty an average factor of 1.6, and decreasing the grid spacing from 30m to 10m [from 9 to 49 measurements] reduces the uncertainty just about a factor of 4.
- Elevating the detector from the ground is very helpful; going from 1m to 3m reduces the uncertainty a factor of 3-4 and going to 10m elevation reduces the uncertainty a factor of 10-15.

Assay of a portion of the total volume

The typical soil sampling method involves using a coring tool to extract a cylinder of soil from the surface down to the desired depth. Here we assumed it was a 10cm diameter core sampling down to 15cm depth. Multiple cores were taken and composited for laboratory analysis. Figure 5 shows the results. There is a significant reduction in the uncertainty as the number of cores increased and as the number of hotspots increased, both following the expected square root function – a four fold increase in samples or hotspots is a 2x decrease in uncertainty.

Computations were done for cores of 1cm and 10cm diameter, and there was no statistical difference. Taking 1cm diameter cores is not mechanically practical, and it should be noted that 1000 cores of a common 2" [5 cm] diameter would weigh nearly 1000 lbs.

The graph at the right of Figure 5 indicates that nearly 1000 samples are needed to have a reasonably high probability of finding any of the hotspot in the laboratory samples – at 10 hotspots and 300 samples, nearly half of the samples will have zero activity even when present.

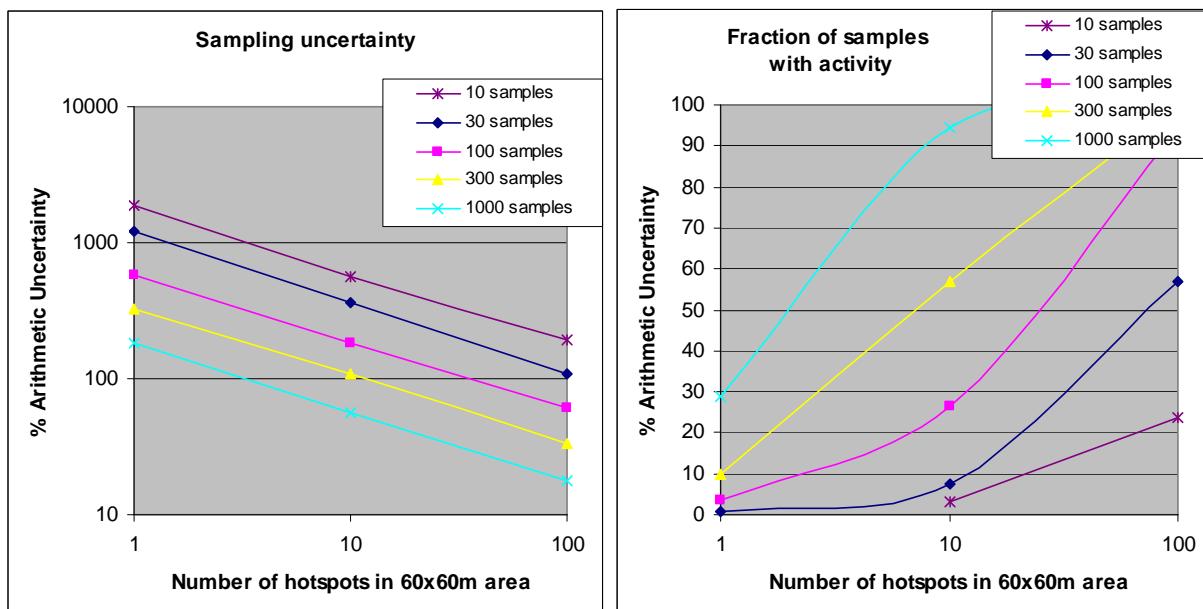


Figure 5 Sample extraction uncertainty for various numbers of samples

Comparison between *in-toto* assay and Sampling uncertainties

So, which is better -- *in-situ* assay of the soil, or extracting and assaying a portion? For the case with 10 hotspots in the survey unit, the *in-situ* measurements can achieve approximately 50% uncertainty with either 30m grid spacing and with the detector at 3m, or 10m grid spacing with the detector at 1m elevation.

Compare that to the sampling uncertainty of 180% when 100 samples are taken. 1000 samples is about 60% uncertainty, and will weigh nearly $\frac{1}{2}$ ton, and must be totally homogenized in order for there not to be further increase in the sampling uncertainty from subsequent sub-sampling in the laboratory.

A further drawback of sampling for sparse distribution like this is that unless a large number of samples is taken [1000 here] there is a high probability that no portion of any of the samples will actually hit the hotspots. For this 10 hotspot scenario, when 30 samples are taken 93% of the time there will be zero activity, and when 300 samples are taken, 46% of the time there will be zero activity shown in the laboratory analysis.

CONCLUSION

When determining the total uncertainty of an item there are many components that must be considered and propagated to the final Total Uncertainty. One of those items is the uncertainty in the efficiency calibration, primarily from the non-uniformity uncertainty. It is shown that the IUE portion of the ISOCS efficiency calibration software is a useful tool to examine the non-uniform contribution to uncertainty for various hypothetical analysis scenarios in order to choose the most appropriate method for the job under consideration.

When the radioactivity in an item to be assayed is not uniformly distributed, then not only will the *in-toto* assay uncertainty be elevated, so will the uncertainty due to any process that attempts to extract a representative portion of the material for a subsequent assay.

For four different typical measurement scenarios, the uncertainty is calculated for both a total sample gamma spectroscopy measurement, and for a sampling process. It is shown that the larger the volume analyzed by gamma spectroscopy, and the larger the sample extracted, the lower the respective uncertainty.

For the four scenarios analyzed, it is shown that the sampling uncertainty is either greater than the total sample gamma spectroscopy uncertainty, or that sampling has other undesirable characteristics such as a high probability of not containing any radioactivity; therefore the total measurement method is preferred. That is not to say that total gamma spectroscopy measurements are always better than sampling and careful laboratory assay of the sample, but it was in these 4 cases, and these 4 situations encompass a wide range of measurement conditions.

It is concluded that for situations where the material cannot be assured to be totally homogeneous, careful consideration must be given to determination and reporting of the sampling error and the total measurement uncertainty to the end user of the data. A large number

of replicate samples and analysis would be one way of determining the sampling error. Multiple total sample measurements of the same sample in a random manner would be a way to evaluate the total sample measurement uncertainty.

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