Optimum Method to Determine Gamma Activity in Large Boxes of Radioactive Material – *In-toto* Measurement vs. Sample Extraction
9246

Frazier Bronson CHP *; Kara Morris
Canberra Industries, 800 Research Parkway, Meriden CT 06450, USA
*Corresponding author: fbronson@canberra.com.

ABSTRACT

During the operations of nuclear facilities and during the environmental remediation and decommissioning of nuclear facilities, radioactive waste is generated which must be assayed. To save on labor and transportation costs, this material is commonly placed in large containers of typically several cubic meters in size [e.g. B-25]. The most common choices of assay are to either extract a representative sample of the contents for laboratory assay, or to use in-situ gamma spectroscopy of the total container. Both of these methods have strengths and weaknesses. InSitu methods determine container activity directly and typically quantify gamma emitters which are then correlated to total activity. Gamma measurement accuracy can suffer from an inadequate efficiency calibration, which can be caused by variations in container size, shape, matrix fill height, or matrix density, but primarily by non-homogeneous distribution of the radioactivity. Laboratory assay methods using a small sample extracted from the container are generally quite accurate for the sample analyzed; but the determination of total container activity requires the assumption that the sample analyzed in the laboratory is truly representative of the total container, which is also heavily influenced by the non-homogeneous distribution of the radioactivity. This evaluation attempts to determine the best way to estimate the activity within the container and gives quantitative estimates of measurement uncertainty for various conditions of radioactivity contained within the container and for various *in-toto* and sampling strategies. A new feature of the ISOCS [*In-Situ* Object Counting System] software called IUE [ISOCS Uncertainty Estimator] was used. First, the various parameters were examined which cause uncertainty in the *in-toto* measurement to evaluate those which are the major contributors and to asses the measurement uncertainty for a uniformly distributed sample. Next, a series of levels of non-homogeneous distributions were analyzed with a variety of potential detector placement strategies. These variables included number of detectors, placement of the detectors, and movement of the detectors. The uncertainty due to non-homogeneity is reduced when the detectors are moved away from the container, when more detector positions are used, and when the detectors are scanned. When the contents of the container are not homogeneous, the sampling uncertainty is likely to be larger than the in-toto measurement uncertainty. For those same non-homogeneous sample situations, various sampling strategies were tried, including type of sample extraction method, size of sample extracted, and number of samples extracted. The conclusion is that if the contents of the container are not homogeneous, the uncertainty due to the sampling process is likely to be larger than the *in-toto* measurement uncertainty.

KEYWORDS: Gamma spectroscopy, in-situ, in-toto, uncertainty, box, container, sampling

INTRODUCTION

A common measurement task is to determine the radioactivity concentration or content within boxes of 1-10 m3 in size. If the contents of the container are well known, and if both the matrix within the container and the radioactivity within the matrix are uniform, then it is a rather conventional task to evaluate the best way to measure it. Extracting a sample for laboratory measurement or performing *in-toto* measurements of the entire container are well known and common methods.

But in practice, this convenient assumption of homogeneity is rarely the case. In both cases (*in-toto* measurement and sampling followed by laboratory measurement) this non-uniform distribution of radioactivity greatly compromises the accuracy of the assay and makes uncertainty estimates much more complicated than simple propagation of counting statistics. It is the purpose of this paper to show which of these methods is more accurate and to show the appropriate use of each.

In-toto quantitative assay of the containers can be performed by a variety of devices ranging from automatic multi-detector scanning systems to simple portable single detector systems. There are many choices of counting protocol that can be made, depending upon the level of accuracy needed and depending upon the sensitivity and counting time desired. These include: detectors close to the container or far away; single or multiple detectors; fixed detectors or scanning; and detectors on single or multiple sides of the box.

With uniform distribution of radioactivity in a known matrix, the efficiency is easily and accurately determined with radioactive sources or with mathematical calibrations. Since calibrations are most always made with uniform distributions, measurements where the radioactivity in the container is not uniform will be in error. For non-uniform distribution of radioactivity, the largest source of uncertainty is likely to be because a uniform reference calibration does not accurately represent the true efficiency. However, certain counting protocols which will be explored in this document can be shown to greatly reduce that uncertainty.

Alternatively, the container may be opened and a sample extracted and sent to the laboratory. If the sample is subsequently homogenized and properly prepared and counted in a geometry where the calibration is well known, the laboratory measurement uncertainty should be small. However, the accuracy of the radioactivity in the original container depends upon how well the laboratory sample concentration represents the container concentration. When the radioactivity in the container is not uniformly distributed, the largest source of uncertainty will very likely be the sampling uncertainty. However, certain sampling protocols discussed later can be shown to greatly reduce that uncertainty.

So, given the choice of *in-toto* measurement of a container with an imperfect gamma assay method, or extracting a sample followed by a [assumed perfect] laboratory assay: which will give a more accurate determination of the contents of the container?

MEASUREMENT OBJECTIVES

The first step in the process is to find out the objectives of the measurement, and therefore the quality of the measurement that is required. This is a frequently overlooked step in the process. Rarely is the objective as simple as “how much radioactivity is in that box ?”. More likely it is something like “give me enough information to reliably categorize these containers as A, B, or C”.

The project manager planning the measurement campaign must try to find out as much as possible about the desired measurement objectives so he can develop a measurement strategy to best achieve it.

Examples of things to inquire about:

- Desired accuracy of each individual container, or of a group of containers; frequently a function of the level of activity and the type of nuclide
- Decision points where the containers are categorized into the appropriate group
- Acceptable Confidence Level for placing the containers in the correct category
- Timing – when are the results needed

- Cost – how much labor can be expended, and what kind of equipment can be used; the cost penalty for an incorrect decision
- Accessibility – are the boxes easy to access, can they be moved, is moving equipment available
- Past measurements or other knowledge – do you know the nuclides to be expected, the range of activity expected, the level of uniformity expected, ...

MEASUREMENT STRATEGY

The proper development of a measurement strategy can only happen after first knowing the Measurement Objectives, and by then understanding how the uncertainty in the measurement result changes due to the many possibilities of performing it.

The two primary measurement possibilities are *in-toto* measurement of the entire intact container via gamma spectroscopy, and extracting a sample of the container for subsequent laboratory analysis. Each one of these methods has both good and bad points, which the program manager must understand well enough to design the most practical method that meets the agreed-upon Measurement Objectives.

In-Toto Measurements

- Generally restricted to only gamma measurements
- Detection levels generally quite good since the sample quantity is large
- Low energy or low abundance gammas may be problematic at low levels
- Can be performed with NaI or LaBr scintillators or Ge semiconductor detectors
    - Scintillation instruments with poor energy resolution may have interference problems from NORM or other radioactivity in the container or nearby
- Opening of the container is not required
- Instantaneous results can be used to guide future measurements on that container or others
- Expensive and sometimes fragile equipment must be used under field conditions
- Efficiency calibrations of large items problematic if using radioactive sources, but easily done with mathematical methods
- No radioactive waste generated
- Taking multiple measurements of a container from different perspectives can reduce the uncertainty and provide evidence of uniformity, or the lack of it.

Sampling of Container followed by Laboratory Measurements

- Opening of the container is required, with the appropriate level of safety protection
- Obtaining a representative sample from locations other than the top of the container is difficult
- Sample must be packaged and shipped to laboratory
- Chain of custody of sample and matching it to the original container must be established
- Can perform chemical separation on sample for removal of interferences and optimum sample geometry for alpha, beta, and gamma assay
- Sample preparation other than for simple gamma spectroscopy is generally rather expensive
- Detection levels can be quite good since sample is close to detector and counting times can be long
- Results not generally available for weeks to months
- Samples must be disposed as radioactive waste

- Taking multiple samples from a container can reduce the uncertainty and if analyzed separately can provide evidence of uniformity, or the lack of it.

Uncertainty Estimation

If the contents of the container are well known and uniform, then the measurement is a simple task. Either use a simple calibration method for the field *in-toto* method, or open the container and scoop some of the contents from the top and take it back to the laboratory. Under these conditions the measurement uncertainty is well understood and easily handled by conventional methods.

For uniform and simple matrices, efficiency calibrations are rather easy to perform. Mixtures of radioactive calibration sources distributed in a simulated uniform manner have long been used successfully. More recently, mathematical calibrations like MCNP [1] and ISOCS® [2,3,4] have been used and accepted by the user community for both calibration of large geometries like here and for laboratory geometries. They are especially convenient to use and when the container is large, or when the matrix is something other than water – e.g. soil, concrete chunks, metal shavings, ...

But this idealistic situation of a uniform matrix and a uniform distribution of radioactivity within the matrix is rarely the case. It is unusual for the matrix within a specific container to be well known in composition, but rather it is commonly a mixture of various materials in unknown concentrations where the matrix varies within the container, and where the density is not well known [NWK] and varies within the container. It is even more unusual for the radioactivity to be homogeneously distributed within the container.

Consequently any efficiency calibration, source-based or mathematics-based, will be wrong when any of these conditions exist. This is true in both laboratory measurements of the sample and *in-toto* measurements. For *in-toto* measurements, this deficiency of the generic calibration used to accurately represent the exact conditions in the container under measurement is most likely the major contribution to the uncertainty of the *in-toto* measurement. For laboratory measurements, the sample preparation normally should transform the sample into the proper form to assure validity of the measurement instrument calibration. However to translate those laboratory results into container results requires propagation of laboratory measurement uncertainty [normally small] and the sampling uncertainty [can be quite large].

For the conditions expected to be encountered, what is the *in-toto* measurement uncertainty? And if samples are extracted, what is the sampling uncertainty ?

To help answer questions like this, a new feature to the ISOCS efficiency calibration software has been developed that allows uncertainty computations to be performed where there is incomplete knowledge of an accurate representation of the sample. The ISOCS Uncertainty Estimator [IUE] software [6, 7, 8, 9] can also simulate various non-uniform distributions in containers and then compute the resulting uncertainty in the efficiency calibration. The software also has a feature that simulates various sampling methods. The IUE software can then be used to create various non-homogeneous distributions in drums, pipes, and boxes and then estimate both the calibration uncertainty and the sampling uncertainty.

ISOCS Uncertainty Estimator – Overall Operation

IUE has been developed to improve the quality of the gamma spectroscopy uncertainty estimate, to improve the ease of generating these uncertainty estimates, and to document how they were generated.

The user first runs the ISOCS efficiency calibration software in the normal manner to compute the normal reference efficiency for the sample being measured. This efficiency file has encoded within it the

inherent uncertainty in the ISOCS efficiency calibration method – i.e. 4-8%. As with most efficiency calibrations, this assumes the calibration model is a perfect representation of the sample.

Some of the ISOCS data entry parameters are well known and do not vary appreciably; e.g. the container is always known to be type 304 stainless steel. Other [or most in many situations] parameters are not well known [NWK], e.g. the wall thickness of the container or the density of the contents. It is these "NWK" parameters that contribute to the uncertainty in the calibration efficiency. This is equally true for source-based calibrations and mathematical calibrations. For each NWK parameter, the user is required to provide to the IUE software an estimate as to how much that parameter varies. This can be determined, for example, by measuring a group of containers, or by consulting the manufacturing specifications for the containers, or just by making educated guesses. The parameters that can be varied include dimensional parameters [diameter, distance, thickness, density, …], as well as material composition of each item in the model.

For each NWK parameter, the user provides upper and lower limits [e.g. maximum and minimum density] and a distribution form that the parameter values within those limits are assumed to follow. As an example of this distribution form, if the values were determined by a series of measurements, then the limits can be assigned to represent 1 standard deviation, 2 standard deviations, or 3 standard deviations. If the values are just known as limits, then they could be assigned a uniform distribution function [all values equally probably] or a triangular distribution function [zero probability beyond the limits increasing linearly to the maximum probability in the middle].

The IUE software then assigns a value to each of these NWK parameters following the probabilities defined by the distribution function for that parameter. The efficiency is computed for that model. This process is repeated a large number of times until adding more models doesn't change the result. The software then computes the model-to-model uncertainty for each energy, which will then be combined with the calibration uncertainty and the counting statistics uncertainty to estimate that portion of the Total Measurement Uncertainty [TMU].

IUE Data Entry Method

The user first points the software to one of the intermediate files created in the normal process of performing an ISOCS efficiency calibration. This file contains all the physical parameters of the normal [assumed perfect] calibration model.

The user is then presented with a series of screens showing all the parameters from the calibration model and given an opportunity to make each of them a variable parameter. If that parameter is to be varied, then the user enters for each parameter the minimum value, the maximum value, and the distribution function to be used. Two examples of these screens are shown below as Figures 1 and 2.

In the case where the variable parameter is a material, the user enters a series of discrete materials, along with a weighting factor denoting the likelihood of that particular material being present.
All input parameters are stored in a file and in a printed report for the project record.

Calculational Methodology

The method used in this software is Probabilistic – all variables are assumed to vary randomly, but in a manner as described by their individual probability distribution function. All variables [except a few that are noted elsewhere] are assumed to vary independently from others, to the extent that it is physically possible.

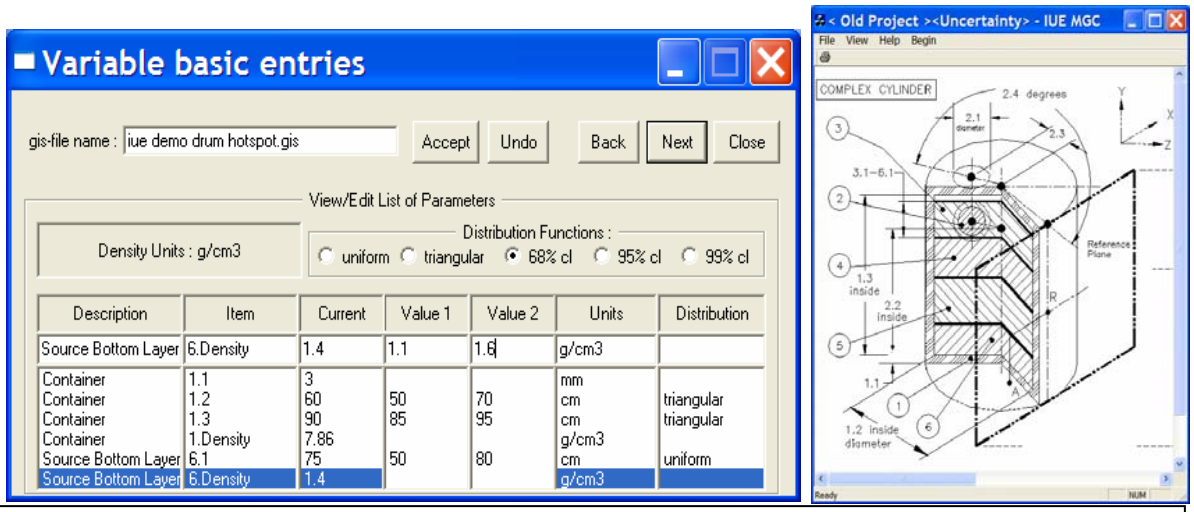


Figure 1: Typical IUE data input screen [left]. Parameters are entered here to describe the amount and type of variation for the model. Entries correspond to the numbers on the graphic on the Figure 2 [at right].

Using these rules, the IUE software creates the files for a series of ISOCS calibration models. A random process is used to generate values for each NWK parameter, according to the probability distribution function rules and limits defined by the user. These values are combined to create an ISOCS model. A large number of these models are created and checked for validity.

The IUE software then computes the efficiency for a large number of energies using each of the valid random models. The IUE software now contains an array of efficiency vs. energy values. For each energy, the IUE software then computes the mean efficiency and the standard deviation of the efficiency values. This standard deviation now represents the uncertainty from the combined effect of all the NWK parameters at each energy.

This uncertainty is then combined with the basic ISOCS calibration uncertainty and embedded within the efficiency calibration. When this efficiency calibration is used to analyze a sample, this total calibration uncertainty is propagated with counting statistics uncertainty and other uncertainties for the final total measurement uncertainty.

Other Software Features

For measurements situations that use multiple detectors, the software allows the user to specify the number of detectors and their placement around the object. Therefore it can be used to calibrate or estimate uncertainty from common field measurement systems like drum, box or truck counters.

For measurement situations that use rotating samples [e.g. drums], the software allows the user to define how many discrete steps are used to simulate a continuous rotation. For measurement situations that use scanning detectors, the software allows the user to define how many discrete steps are used to simulate a continuous scan. This allows the software to be used to calibrate and estimate uncertainty for common box and drum measurement systems.

Some measurement situations have non-uniform sample concentration. Several of the ISOCS sample shapes [templates] allow non-uniform distributions, including discrete “hot spots”. The IUE software

expands that by allowing a multiple [or variable] number of hotspots and the size of the hotspots [fixed or variable] to be included in the model.

Although most of the variables are treated as independent variables, a few of them can be inter-dependent. A common example is sample height in a container, sample density, and sample weight. The weight is typically the most well known parameter, as it is rather easy to determine. The IUE software lets the user enter the weight as a variable parameter and then it computes either sample height or sample density.

The software computes the arithmetic mean efficiency and standard deviation, as well as the geometric mean efficiency and standard deviation. For measurement situations where attenuation is the dominant factor, the values are more likely to be in a log-normal pattern, where the geometric values are more relevant.

The IUE software also operates in a Sensitivity Mode, where only 1 parameter is varied at a time. This provides the user with feedback as to which of the parameters are the major contributors to the total uncertainty, thus allowing the user to concentrate data collection resources on those dimensions that are most important.

The IUE software can also simulate the extraction of a portion of the container and compute the "representativeness" or uncertainty of that sample. This process only works on those containers that have "hot spots" of non-uniform radioactivity. The user specifies the sample type [core or grab], specifies the sample volume to be extracted, and specifies how many sub-samples will make up the total sample extraction volume. The IUE Sampling program then simulates extraction of these samples from the same containers that were used for the previously described *in-toto* uncertainty analysis. This is done multiple times using random sampling locations from each of the containers. The software computes the fraction of the radioactive source within each sample as compared to the amount in the container. Then the uncertainty is computed from the total population of samples.

Large Box Assay Uncertainty Calculations

Homogeneous Distribution Example

An in-situ Ge gamma spectroscopy system is being used to assay a group of 2 cubic meter boxes filled with debris from a facility demolition task.

The boxes are nominally 1m tall and 1m x 2m in size. From the manufacturing specifications, the maximum variation in the 2m length is 2cm, and is 1cm variation in the width and height. The walls of the container are 3mm steel, but can vary between 2 and 4mm. Since no other information about the nature of this variation is known, a triangular distribution was assumed.

The bulk of the container was filled with low density low Z material like wood, asphalt, or small metal pieces. The bulk matrix was assumed to be composed of iron 20% of the time, cellulose 40% of the time, and a 50-50 mixture of iron and cellulose the rest of the time. Surveys before the building was demolished detected no radioactivity, therefore it is reasonable to expect that most of the container contents are approximately uniform in radioactivity concentration.

The fill height of each box is not known, and it is neither practical nor desirable to open the containers for inspection. But from discussions and procedures during the fill operations, the containers were filled until they were approximately 70-100% full.

The composition and density of each individual box is not known, but the total weight of each box has been determined. The weight of the containers varied from 480 to 960 kg, with most of them in the 700-750 kg range. In this example, the weight and fill-height were used to estimate container density for use by the calibration software.

The ISOCS cart and detector were wheeled up next to the boxes, at approximately 100cm from the side of the drum. That distance was crudely measured, and the cart repositioned if the distance was not between 90 and 110 cm. The detector in the ISOCS cart is nominally 50 cm from the ground, and aimed approximately at the center of the box. But since the ground is not flat, there could be a 10cm variation in the detector height, a 10cm variation in horizontal detector position, and a 10cm variation in detector aiming position.

The nuclides of interest for this site are Am-241 at 60 keV, Uranium-235 at 185 keV, and Uranium-238, using the Pa-234m daughter at 1001 keV.

There are 10 uncontrolled variables in this problem. What uncertainty should be assigned to the combination of all these variables when counting an individual drum? To answer this question the above data were used with IUE, first in the Sensitivity Analysis mode, and next in the Uncertainty Analysis mode.

The Sensitivity Analysis mode first computes the efficiency for the reference conditions, and then varies each of the parameters one at a time, computing the efficiency using the maximum and then the minimum value of that parameter. The program then reports the minimum and maximum efficiency caused by changes in that parameter, at each energy analyzed. A summary of those results are presented in Table 1.

Table 1 Sensitivity Analysis – important parameters shown

Parameter	Parameter range	Eff range at 60keV	Eff range at 1000 keV
Wall thickness	3 – 4 mm	0.75 – 1.63	0.98 – 1.02
Box–detector distance	90 – 110 cm	0.91 – 1.10	0.89 – 1.14
Matrix material	Cellulose – iron	0.59 – 3.33	0.96 – 1.05
Matrix density	480 – 960 kg	0.98 – 1.34	0.98 – 1.19

For low energies, the dominant contributor is the matrix material, and next is the container wall thickness. For high energies, the contributions are much less, with box-detector distance being the dominant one. The variability in box size, detector elevation, and detector aiming point were trivial, and therefore not reported here. If accurate measurements at low energies are important, then the user could consider inspecting the contents for more accurate determination of material visually, or with a transmission source, for those boxes that are close to the decision point; and might also consider a device like a portable ultrasonic probe to determine the exact wall thickness.

Next, the IUE software was used in the Uncertainty Analysis mode. The program created several hundred mathematical calibrations which were analyzed for standard deviation. Table 2 shows the 95% CL uncertainty estimate. The first row in the data is when all the parameters previously described were allowed to vary.

From the IUE Sensitivity Analysis results, the user knew that wall thickness was a big factor at low energies, and wanted to see what would happen to the uncertainty if that parameter was accurately measured. The next row shows the result. Still not satisfied, the user evaluated the improvement if the density were made well known, e.g. by the use of a transmission source or by weight and fill-height determination.

Table 2 Uncertainty analysis results for 2 cubic meter box		
Condition	**%SD at 60keV**	**%SD at 1000keV**
All items variable	88 %	12 %
After fixing the container wall	68 %	11.5 %
After fixing the density	67 %	8.3 %

Non-homogeneous distribution of radioactivity in 2 cubic meter box – in-toto measurement

This exercise will illustrate the usefulness of the IUE software to optimize a counting geometry for the boxes described previously, but this time with the presence of radioactive hotspots, and then to assign an uncertainty to the efficiency calibration for that optimum geometry.

Due the activities in the buildings, the presence of small areas of contamination were discovered, and removed when found, but the existence of others still remaining cannot be excluded. These would most likely be higher in density and higher in Z, e.g. chunks of metallic items. For this modeling, a 50-50 mixture of iron and cellulose of a density of 0.5 g/cc was assumed for the bulk matrix, and the radioactive "hotspots" were assumed to be iron at 0.7 g/cc. The nominal size of these hotspots was estimated to be 10x10x10cm. They are randomly distributed within the box.

The nuclides of interest have energies of 60 keV, 200, and 1000 keV. What is the optimum counting geometry if the purpose is to minimize the total uncertainty of the box assay?

In this scenario, 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 box. The first situation assumed that there were 1-5 radioactive hotspots per box. All values were equally probable. Other scenarios were 10-15, 20-30, 100, and 150 hotspots per box. All hotspot sources were randomly distributed throughout the box matrix.

The counting geometries that were investigated were

- distance from the side of the box [20cm and 100cm]
- number of detectors [1, 2, 4, and 6]
- various detector placements
    - 1 detector; at the center of the 2m side
    - 2 detectors; 1 at the center of the 2m side, 1 on the opposite side
    - 4 detectors; 1 on each of the 2m sides, 1 on each of the 1m sides
    - 4 detectors; 2 on each of the long sides each 70cm from the Center Line
    - 6 detectors; as above plus 1 on each of the 1m sides
    - 2 detectors; 1 on each of the 2m sides, scanning -1.2m to +1.2m from CL

Table 3 presents the results. For both energies, there are two different standard deviation values. The column labeled "%sdA" is the "normal" or arithmetic standard deviation of the efficiency values, expressed as a percent of the mean efficiency. The column labeled "sdG" is the geometric standard deviation, expressed as a factor of the geometric mean efficiency value. Whereas arithmetic standard deviations are added and subtracted from the mean, geometric standard deviations are multiplied and divided by the mean to yield the upper and lower confidence intervals. Figure 3 shows these results graphically.

In these analyses, especially at 60 keV, 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 results from the two different standard deviation measures approach each other. Both are presented in Table 3 for comparison.

Several trends can be seen from the data.

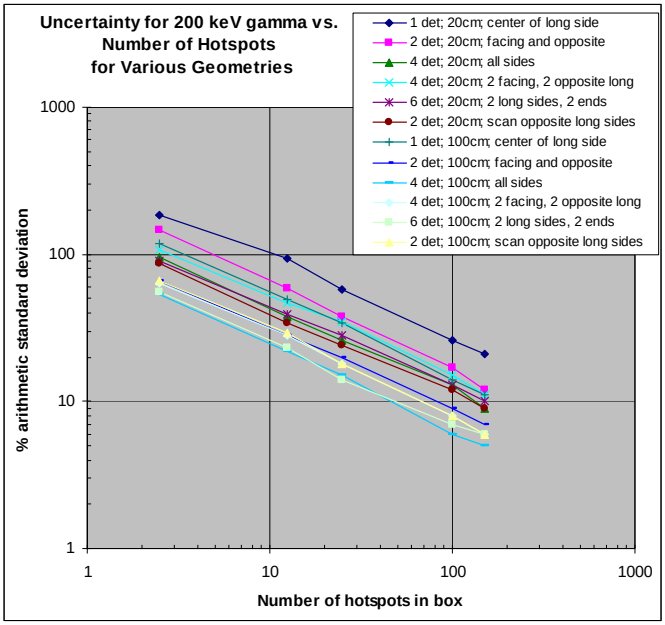
- Low energies have considerably higher standard deviation than high energies
    - 60 keV has a 4x larger sd than 200 keV while 1000 keV is 1.5x smaller
- Detectors at 20cm distance from the box have higher standard deviation that detectors at 100cm
    - 20cm uncertainty is about 2x higher than the 100cm uncertainty
- Moving the detector back to 100 reduces the efficiency, and therefore will increase the counting statistic component of the total propagated uncertainty; alternatively, the measurement time could be increased
    - At 60 keV the efficiency is 1.5x lower; for the same sd increase count time 2.2x
    - At 200 and 1000 keV, efficiency is 3x lower; for same sd increase count time 9x
    - However, if non-uniformity uncertainty is high, then a low counting statistic component isn't very helpful in reducing the total propagated uncertainty, as long as the Minimum Detectable Activity is acceptable
- Adding more detectors reduces the uncertainty.
    - 4 detectors reduces the sd a factor of 2 over a single detector, as long as they are viewing substantially different regions of the container
    - 6 detectors was best [factor of 2.2 lower sd than 1 detector], but not much better than 4 detectors
    - 2 scanning detectors was equivalent in sd to 6 stationary detectors, although efficiency will be about 3x lower and therefore higher sd for counting statistic component.
- Adding more detectors increases the efficiency, and therefore reduces the counting statistic component or counting time and MDA; a single detector moved to each of those positions can achieve the same, but with a longer total assay time
- Increasing the number of hotspots dramatically reduces the standard deviation for all geometries and for all energies
    - a 10-fold increase in the number of hotspots is a 3-fold reduction in uncertainty

Non-homogeneous distribution of radioactivity in boxes – sampling

In this exercise, the same containers with random hotspots used in the previous section were sampled using the IUE sampling function. Both "grab" sampling and core sampling strategies were investigated. For grab samples, a spherical sample was "extracted" from random locations in each of the containers. The diameter of the sphere used was 2", 4", 6", and 8". The number of samples was also varied – from 1 to 100 per container. For core samples a cylinder from the top to the bottom of the container was simulated. Core diameters used were 1", 2", and 4". Again, the number of cores per container varied from 1 to 100. Figures 5 and 6 show those results.

Table 3 Uncertainty for 1m x 1m x 2m Box with Hotspots

				60 keV		200 keV		1000 keV	
Dist	**Det's**	**Geometry**	**Hotspots**	**%sdA**	**sdG**	**%sdA**	**sdG**	**%sdA**	**sdG**
20	1	Center of long side	1-5	677	2581	185	6.6	119	3.0
100	1	Center of long side	1-5	262	1471	119	4.8	78	2.3
20	2	Facing and opposite long	1-5	505	111	147	3.1	84	2.0
100	2	Facing and opposite long	1-5	184	40	66	2.1	34	1.4
20	4	All four sides	1-5	446	32	96	2.2	54	1.6
100	4	All four sides	1-5	147	19	53	1.8	28	1.4
20	4	2 on facing, 2 on opposite	1-5	370	78	107	2.8	63	1.8
100	4	2 on facing, 2 on opposite	1-5	166	35	64	2.0	31	1.4
20	6	2 facing, 2 opposite, 2 on	1-5	319	81	90	2.9	58	1.8
100	6	2 facing, 2 opposite, 2 on	1-5	143	25	56	1.9	29	1.3
20	2	scanning, 1 facing, 1	1-5	265	41	87	2.4	50	1.6
100	2	scanning, 1 facing, 1	1-5	165	37	66	2.0	33	1.4
20	1	Center of long side	10-15	384	37	93	2.6	61	1.8
100	1	Center of long side	10-15	121	10	49	1.8	31	1.4
20	2	Facing and opposite long	10-15	272	10	59	1.7	35	1.4
100	2	Facing and opposite long	10-15	81	3.1	28	1.3	15	1.2
20	4	All four sides	10-15	176	5.1	38	1.5	21	1.2
100	4	All four sides	10-15	64	2.1	22	1.2	11	1.1
20	4	2 on facing, 2 on opposite	10-15	155	7.6	47	1.6	27	1.3
100	4	2 on facing, 2 on opposite	10-15	73	3.1	28	1.3	14	1.2
20	6	2 facing, 2 opposite, 2 on	10-15	151	4.8	39	1.5	25	1.3
100	6	2 facing, 2 opposite, 2 on	10-15	61	2.3	23	1.3	12	1.1
20	2	1 facing, 1 opposite, full	10-15	117	4.0	34	1.4	19	1.2
100	2	2 facing, 1 opposite, full	10-15	72	2.9	29	1.3	14	1.2
20	1	Center of long side	20-30	243	10.7	58	1.8	37	1.4
100	1	Center of long side	20-30	84	3.5	34	1.4	22	1.3
20	2	Facing and opposite long	20-30	157	5.6	38	1.5	23	1.3
100	2	Facing and opposite long	20-30	59	2.0	20	1.2	10	1.1
20	4	All four sides	20-30	121	3.3	26	1.3	15	1.2
100	4	All four sides	20-30	44	1.7	15	1.2	8	1.1
20	4	2 on facing, 2 on opposite	20-30	121	4.1	35	1.4	20	1.2
100	4	2 on facing, 2 on opposite	20-30	51	1.9	18	1.2	9	1.1
20	6	2 facing, 2 opposite, 2 on	20-30	102	2.8	28	1.3	18	1.2
100	6	2 facing, 2 opposite, 2 on	20-30	37	1.5	14	1.2	7	1.1
20	2	scanning, 1 facing, 1	20-30	69	2.9	24	1.3	14	1.2
100	2	scanning, 1 facing, 1	20-30	47	1.9	18	1.2	9	1.1
20	1	Center of long side	100	101	2.9	26	1.3	17	1.2
100	1	Center of long side	100	38	1.5	14	1.2	9	1.1
20	2	Facing and opposite long	100	77	2.3	17	1.2	10	1.1
100	2	Facing and opposite long	100	27	1.5	9	1.1	4	1.1
20	4	All four sides	100	61	1.9	13	1.1	7	1.1
100	4	All four sides	100	19	1.2	6	1.1	3	1.0
20	4	2 on facing, 2 on opposite	100	56	1.8	15	1.2	8	1.1
100	4	2 on facing, 2 on opposite	100	23	1.3	8	1.1	4	1.0
20	6	2 facing, 2 opposite, 2 on	100	45	1.6	13	1.1	8	1.1
100	6	2 facing, 2 opposite, 2 on	100	19	1.2	7	1.1	4	1.0
20	2	scanning, 1 facing, 1	100	40	1.5	12	1.1	7	1.1
100	2	scanning, 1 facing, 1	100	24	1.3	8	1.1	4	1.0
20	1	Center of long side	150	98	1.2	21	1.2	13	1.1
100	1	Center of long side	150	29	1.4	11	1.1	7	1.1
20	2	Facing and opposite long	150	59	1.9	12	1.1	7	1.1
100	2	Facing and opposite long	150	20	1.2	7	1.1	3	1.0
20	4	All four sides	150	45	1.6	9	1.1	5	1.1
100	4	All four sides	150	15	1.2	5	1.1	3	1.0
20	4	2 on facing, 2 on opposite	150	47	1.7	11	1.1	6	1.1
100	4	2 on facing, 2 on opposite	150	18	1.2	6	1.1	3	1.0
20	6	2 facing, 2 opposite, 2 on	150	37	1.5	10	1.1	6	1.1
100	6	2 facing, 2 opposite, 2 on	150	16	1.2	6	1.1	3	1.0
20	2	scanning, 1 facing, 1	150	28	1.3	9	1.1	5	1.1
100	2	scanning, 1 facing, 1	150	19	1.2	6	1.1	3	1.0
				+ or -	**x or ÷**	**+ or -**	**x or**	**+ or -**	**x or**



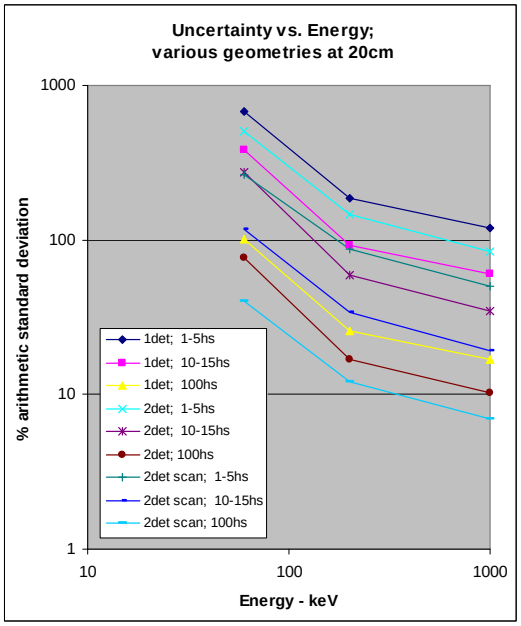
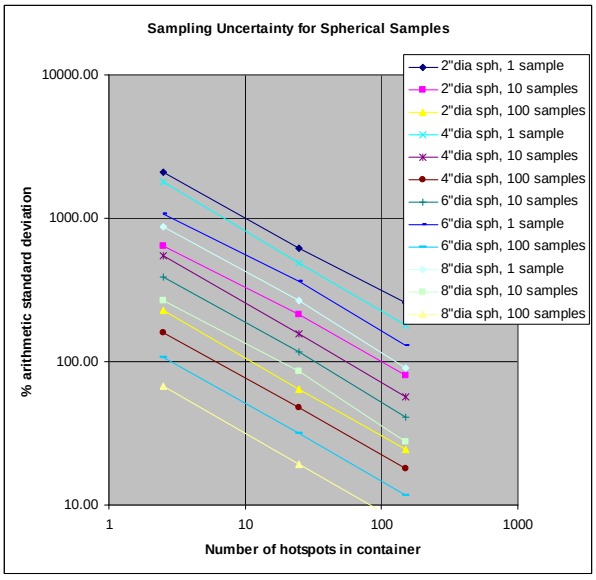


Figure 3 [left]: Uncertainty at 200 keV for various number of hotspots and various geometries.
Figure 4 [right]: Uncertainty for various geometries at 60, 200, and 1000 keV.



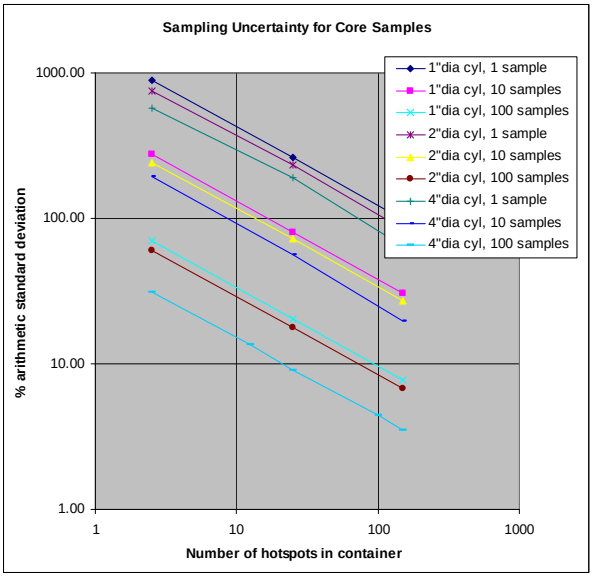


Figure 5 [left] Sampling uncertainty for various sizes and number of spherical samples.
Figure 6 [right]: Sampling uncertainty for various sizes and number of cylindrical core samples

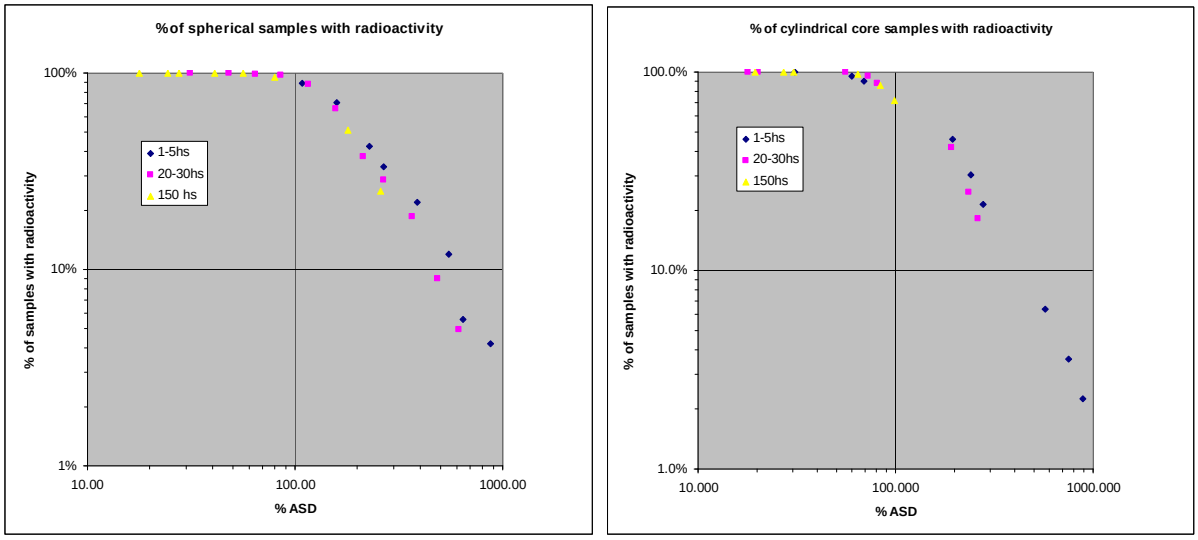
For comparison of sampling to i*n-Toto* measurements, consider a situation where there are 10-15 hotspots in a 2 cubic meter container.

From Table 3 and Figure 3 for *in-toto* measurements at 200 keV, it can be seen that for situations where there are 10-15 hotspots in the 2 cubic meter box, that all measurement methods yield results with an arithmetic relative standard deviation of 20-90%, and that most of them are in the 20-50% range. Lower energies have higher sd, and higher energies have lower sd.

A “typical” sampling campaign of containers this size will rarely be even as many as 10 samples per container. If they are spherical samples, then the uncertainty for 10-15 hotspots will be about 300% for 10 2” diameter samples down to about 120% for 10 8” diameter samples. If they are top-bottom core samples, then the uncertainty will be about 120% for 10 1” diameter samples down to about 80% for 10 4” diameter samples.

To achieve a 20-50% arithmetic standard deviation from sample extraction will require about 100 separate spherical samples of 6” or 8” in diameter, or over 100 cores of 1”, 2”, or 4” diameter.

Furthermore, unless there are quite a few samples taken [more than 10 when there are 10-20 hotspots] or a large number of hotspots in the container [50-100], then most of the laboratory assay results will report zero activity, even if it is present inside the box, as shown in Figures 7 and 8. That is because of the rather high probability that none of the samples extracted from the container actually contained a portion of the hotspot of radioactivity. This can falsely indicate that the particular box is “clean” when in fact it is not.



Figures 7 [left] and 8 [right]: Percent of laboratory assays that will show positive radioactivity

CONCLUSION

It has been shown that for rather extreme non-homogeneous distributions, there are several reasonable easy to implement *in-toto* measurement strategies that can reduce the measurement uncertainty down to the 10-100% sd range and lower.

It has also been shown that for these same non-homogeneous distributions, any practical sampling strategy will have a considerably higher uncertainty. Therefore even a perfect laboratory measurement when propagated with the sampling uncertainty will have a total uncertainty higher than the *in-toto* result.

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 decision for that container can be made. For example: if the measurement result for U-235 at 185 keV was a factor of 20 below the “decision limit,” then even the quick and simple 20cm single detector location stationary measurement with a measurement uncertainty of a factor of 7 would be adequate to prove that the item is “acceptable”, even if there are as few as 1-5 hotspots in the 2 cubic meter container. If most of the containers are like this, then this simple geometry is an economical one to use. If then a few of the containers have results closer to the limit [perhaps within 2sd or a factor of 13], then those few could be recounted in a more

precise method – perhaps with 20cm measurements on all 4 sides, which reduced the uncertainty to a factor of 4 for the 1-5 hotspot case.

REFERENCES

[1] J. F. BRIESMEISTER (ed), MCNP- A General Monte Carlo N-Particle Transport Code, Report LA-12625-M, Los Alamos National Laboratory
[2] F. BRONSON, B YOUNG, Mathematical Calibrations of Ge Detectors and the Instruments that Use Them; Proceedings of 5th Annual NDA/NDE Waste Characterization Conference, Salt Lake City, UT, Jan 11, 1997.
[3] R. VENKATARAMAN, F. BRONSON, V. ATRASHKEVICH, B. YOUNG, AND M. FIELD; Validation of In-Situ Object Counting System (ISOCS) Mathematical Efficiency Calibration Software; Nuclear Instruments and Methods in Physics Research (A), 422 (1999), pp. 450-454.
[4] F. BRONSON, V. ATRASHKEVICH, G GEURKOV, AND B. YOUNG; Probabilistic Uncertainty Estimator for Gamma Spectroscopy Measurements. Journal of Radioanalytical and Nuclear Chemistry [in press], Presented at MARC VII Conference, April 2006, Kona HI
[5] Model S573 ISOCS Calibration Software, Canberra Industries 800 Research Parkway Meriden CT USA, www.canberra.com
[6] F. BRONSON; The Uncertainty from Inhomogeneity; A Comparison between Gamma Spectroscopy of a Large Sample and Sampling Error; [in press] Presented at HPS Mid-Year Symposium, Jan 2007, Knoxville KY
[7] F. BRONSON, and V. ATRASHKEVICH; Measurement Uncertainty from *In-Situ* Gamma Spectroscopy of Non-homogeneous Containers and from Laboratory Assay; Proceedings of Waste Management 2007, Tucson AZ
[8] F. BRONSON; 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; Proceedings of Waste Management 2008, Phoenix AZ
[9] D. NAKAZAWA, S. CROFT, M. HENRY, W. MUELLER, R. VENKATARAMAN, M. VILLANI, H. ZHU; Determination of Non-Uniform Source Distribution Uncertainty of Waste Assay Systems Using ISOCS Uncertainty Estimator; Proceedings of International Symposium on Radiation Safety Management: Daejeon, Korea, pp. 59-95 (2007).