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Probabilistic uncertainty estimator for gamma-spectroscopy measurements

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To properly interpret the quality of a gamma-spectroscopy measurement, an uncertainty estimate must be made. The uncertainty in the efficiency calibration is the dominant component to the total propagated measurement uncertainty for many types of measurements. Any deviations between the as-calibrated geometry and the as-measured geometry contribute to the total uncertainty. A mathematical technique has been developed to evaluate the variations between calibration and measurement conditions. A sensitivity analysis mode identifies those variables with the largest contribution to the uncertainty. The uncertainty mode uses probabilistic techniques for the combined variables to compute average efficiency and uncertainty, and then to propagate those values with the gamma-spectroscopic analysis into the final result for that sample.

Introduction

Gamma-spectroscopy is a very useful tool to quantify the activity of various items, such as samples in the laboratory, or waste assay containers, or large items in-situ. In addition to the activity or concentration of the sample, an uncertainty estimate is also needed to fully understand the quality of the measurement. This uncertainty estimate should include the uncertainty in the efficiency calibration of the instrument, as well as many other parameters. For many kinds of samples, especially those that have not been carefully prepared for laboratory gamma-spectroscopy, it is the efficiency uncertainty that is the dominant component to the total propagated measurement uncertainty.

Efficiency vs. energy calibrations have been traditionally determined using well-known radioactive sources in carefully prepared geometries to best represent the sample being measured. For small samples typical in the laboratory and simple samples like water this is relatively easy. But for large and/or non-water samples mathematical calibrations are very common, can be quite accurate, and are much quicker and more convenient. One such mathematical calibration tool is the Canberra ISOCS (In-Situ Object Calibration Software). This software can quickly compute an efficiency calibration formula for a wide range of sample types and shapes.[1,2] However, this calibration software, and essentially all other mathematical or source-based calibration methods, assumes that the source to be measured is exactly like the calibration source model. This is rarely the case.

When building a calibration source or creating a mathematical calibration model, exact discrete values for the physical dimensions and materials of the source or model are used. But the real sources being measured are not exactly the same. Differences between the real source and the calibration source or model include: container wall thickness; container diameter; sample height in the container; sample density; sample matrix composition; sample uniformity; source–detector distance, etc.

A typical method to address these is to assume worst-case values for each of these, either singly or all together. In most situations, all of these differences between the reference calibration and the measured sample exist concurrently. Therefore, the simplistic methods of considering only one item at a time will most likely have too small uncertainty values, and using the worst-case values of all of them together will most likely have too large uncertainty values.

ISOCS Uncertainty estimator

A new tool called ISOCS Uncertainty Estimator (IUE) has been developed to improve the quality of the gamma-spectroscopy uncertainty estimate, to improve the ease of generating it, and to document how it was generated.

The user first runs the ISOCS 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 most efficiencies, this assume of the calibration model is a perfect representation of the sample.

The IUE software is then used to create the model uncertainty which will then be combined with the calibration uncertainty and the counting statistics uncertainty. The software can also be used for a sensitivity analysis, to find those parameters which contribute the most to the uncertainty.

Data required by user

To create an ISOCS calibration file, the user needs to know the physical parameters of the object, such as

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dimensions of the container and sample, and composition of the container and sample. Depending upon the item's complexity, and assumptions the user might feel comfortable, this could be as few as 5 parameters, or as many as 50 or more parameters.

Some of those parameters are well known and do not vary appreciably; e.g., the container is always known to be type 304 stainless steel. Other parameters are not well known, e.g., the wall thickness of the container or the density of the contents. It is these not well known parameters that contribute to the uncertainty in the calibration efficiency. For each not well known parameter, the user is required to provide an estimate as to how much it varies; e.g., by measuring a group of containers or 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, etc.), as well as material composition of each item of the model.

For each not well known 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).

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 in Figs 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.

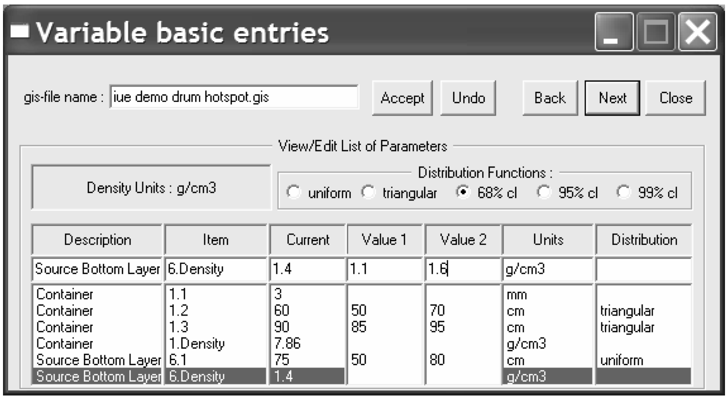
All input parameters for the project record are stored in a file, and in a printed report.

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 which is logically possible.

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 not well known 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 values at each energy.



gis-file name : iue demo drum hotspot.gis

Density Units : g/cm3

Distribution Functions : ○ uniform ○ triangular ● 68% cl ○ 95% cl ○ 99% cl

Description	Item	Current	Value 1	Value 2	Units	Distribution
Source Bottom Layer	6.Density	1.4	1.1	1.6	g/cm3	
Container	1.1	3			mm	
Container	1.2	60	50	70	cm	triangular
Container	1.3	90	85	95	cm	triangular
Container	1.Density	7.86			g/cm3	
Source Bottom Layer	6.1	75	50	80	cm	uniform
Source Bottom Layer	6.Density	1.4			g/cm3	

Fig. 1. Typical IUE data input screen. Parameters are entered here to describe the amount and type of variation for the model. Entries correspond to the numbers on the graphic in Fig. 2

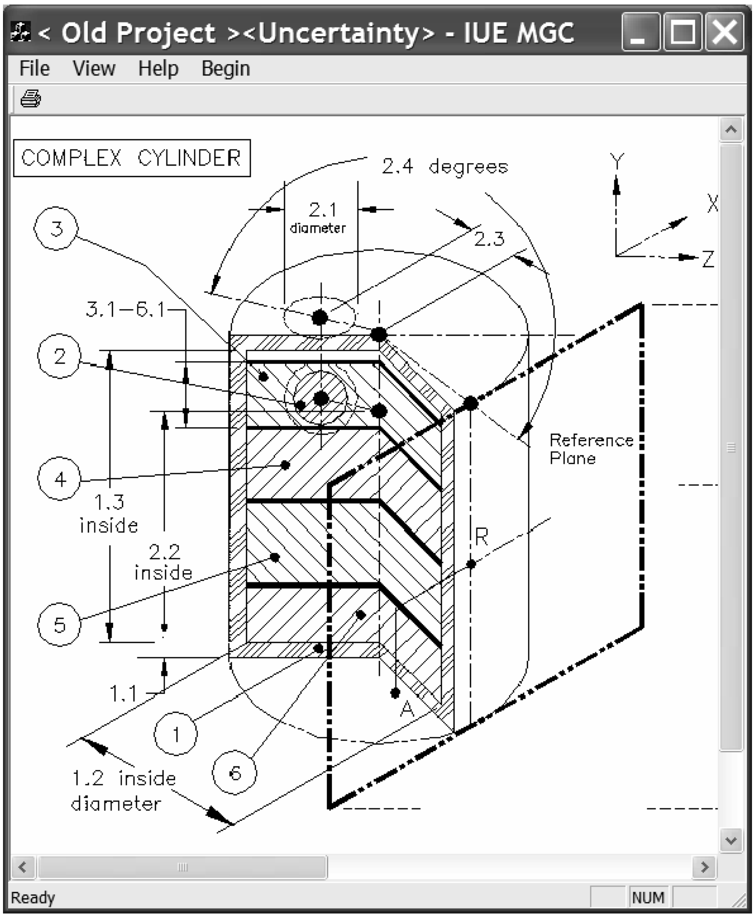


Fig. 2. One of 19 different generic shapes allowed in ISOCS and IUE. The user provides the "current" dimensions for each item in the ISOCS software. These show up in the IUE screens, where the user enters the variable parameters

For each energy, the IUE software then computes the mean efficiency, and the standard deviation. This standard deviation now represents the uncertainty of the combined effect of all the not well known parameters.

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 with multiple detectors, the software allows the user to specify the number of detectors, and determine their placement around the object.

For measurement with rotating samples, the software allows the user to specify this, and to define how many discrete steps are used to simulate a continuous rotation.

For measurement with scanning detectors, the software allows the user to specify this, and define how many discrete steps are used to simulate a continuous scan.

Some measurement uses non-uniform sample concentration. Several of the ISOCS sample types allow non-uniform distributions, including "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) are included in the model.

Although most of the variables are treated as independent variables, a few of them can be 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 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 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 to know which of the parameters are the major contributors to the total uncertainty, thus allowing the user to concentrate data collection resources for those dimensions that are most important.

Examples of IUE calculations

200-liter drum assay under field conditions

This is a common field measurement situation. An in-situ Ge gamma-spectroscopy system is being used to assay a group of 200-liter drums filled with radioactive soil from an environmental remediation project. The site had contaminated soil of many different types, and consequently many different densities. These ranged from wet sandy material at density of around 1.8 g/cm^3 down to soil mixed with decayed vegetation at densities of 1.0. The soil composition of the containers was estimated to be normal soil approximately 50% of the time, mostly sand approximately 25% of the time, and soil and decayed vegetation about 25% of the time. The drums were filled with material that had been stored in piles and rather well mixed, therefore, it is reasonable to assume that the radioactivity in each individual container is homogeneous.

The composition and density of each individual drum is not known, but the total weight of each drum is known. The weight of the containers varied from 400 to 800 lbs. A random sampling of the weights showed that 95% of them were between 450 and 750 lbs.

The fill height of each drum is not known, and it is neither practical nor desirable to open each drum for inspection. But from discussions and procedures during the filling operations, the containers were filled until they were approximately 70–90% full.

The ISOCS cart and detector were wheeled up next to the drums, at approximately 100 cm from the side of the drum. That distance was measured, and the cart repositioned if the distance was not between 90 and 110 cm. The detector in the ISOCS cart is 26 cm from the ground, and directed to the center of the drum. But since the ground is not flat, there could be a 10 cm variation in the detector height.

The drum specifications from the manufacturer claim that the diameter and height of the container are to be controlled within 1 cm of the nominal value, and the wall thickness within a 20% range.

The nuclides of interest for this site are ^{241}Am at 60 keV and depleted uranium, using the ^{234m}Pa daughter at 1001 keV.

There are 8 uncontrolled variables in this problem. Which ones will cause the largest variation in the efficiency? To answer that question the program was first run in the Sensitivity Analysis mode. The upper and lower boundary of each parameter is entered. The software varies these one at a time and computes the change from the base efficiency. The results are shown in Table 1. They are expressed as a percent variation from the reference position. In this case, it is the sample density that is the worst offender for both low and high energies, followed by container thickness but only for the low energy.

What uncertainty is to be assigned to the combination of all these variables when counting an individual drum? It might appear to be quite bad. To answer this question the same data were used with the addition of the distribution parameter, with the software 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 were allowed to vary as described before. From the sensitivity analysis, the user knew that density was a big factor, and will hypothetically explore what would happen if he would more accurately determine it. The next row shows the result. Still not satisfied, he could use an ultrasonic probe to accurately measure the wall thickness, which removes that variable and gives the results in the last row.

Laboratory sample assay

This is a common laboratory sample measurement problem. The sample container is a 115-cm^3 container, nominally 5 cm diameter and 6 cm in height. However, since this is a standard injection molded container, the bottom is not flat but concave, and varies ±2 mm of concavity from container to container.

Table 1. Sensitivity analysis results for 200-l drum

Variable	Variation at 60 keV, %	Variation at 1001 keV, %
Drum diameter	±2	±2
Drum height	0	0
Drum wall thickness	±29	±2
Sample height	±3	±1
Sample density	+39 –28	+31 –20
Sample composition	+9 –6	±1
Detector distance	+18 –14	+18 –15
Detector height	+0 –4	+0 –2

Table 2. Uncertainty analysis results for 200-l drum

Condition	95% CL at 60 keV	95% CL at 1001 keV
All items variable	36%	20%
After fixing the density	30	14
After fixing the container wall	16	14

The sample is filled with 100 g of soil. The soil type varies from sand, to "normal" soil (most likely) to peaty soil, however, only a single calibration is used. The soil density varies from sample to sample (ranges between 1.1 and 1.4 g/cm^3) because of the different compositions of soil counted, therefore, the fill height in the container also varies. The base calibration assumes a density of 1.25 g/cm^3. Since the soil is dried and ground, it is reasonable to assume that the radioactivity is uniform.

There are 4 uncontrolled variables in this problem. Which of these are the most important contributors to the overall uncertainty at 60 keV and at 1001 keV? Again, the Sensitivity mode of IUE was used, giving the results in Table 3.

What additional uncertainty estimate should be applied as a result of these variables? Table 4 shows the 95% CL error estimate for all 4 of the variables in the first row. The user is not satisfied and wants better accuracy. He then decides to determine the density by using the fill height and the weight and then uses the LabSOCS software (the laboratory version of the ISOCS software) to compute the efficiency for that exact combination of density and fill height. So those are no longer variables, but the sample composition and container distortion variables are still left. The last row shows the improvement in accuracy.

200-liter non-homogeneous drum assay

This exercise will illustrate the usefulness of the IUE software to optimize a counting geometry, and then to assign an uncertainty to the efficiency calibration for that optimum geometry. In this scenario, there exist a large number of 200-liter drums filled with soil, at an average density of 1.2 g/cm^3. The radioactivity in the soil is known to be quite non-uniform. 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. The assumed situation is that there are 1–5 radioactive hotspots per drum, all values equally probably, and all sources randomly distributed. Situation two assumed that there are 10–20 hotspots per drum.

The counting geometries that were investigated have distances from the side of the drum (20 cm, and 100 cm), counting from a single side or from two sides of the drum, fixed or vertically scanning detectors, and stationary or rotating drum.

Table 5 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.

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.

Several trends can be seen from the data.

- Low energies have considerably higher standard deviation than high energies;
- A detector close at 20 cm has the highest standard deviation;
- Scanning the detector up and down, the full drum height does not 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 by 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 20 cm 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 100 cm is somewhat better;

Table 3. Sensitivity analysis for laboratory sample

Variable	Variation at 60 keV, %	Variation at 1001 keV, %
Container bottom curvature	+7 –7	+9 –8
Sample fill height	+13 –7	+10 –6
Sample density	+9 –5	+9 –8
Sample composition	+7 –0	0

Table 4. Uncertainty analysis for laboratory sample

Condition	95% CL at 60 keV	95% CL at 1001 keV
All items variable	13%	9%
After fixing the density and height	6	3

Table 5. Uncertainty for 200-liter drum with hotspots

Distance, cm	Motion	Hotspots	60 keV		1000 keV	
			%sdA	sdG	%sdA	sdG
20	Stationary	1–5	256	28.00	81	2.44
20	Scanning	1–5	300	27.00	93	2.50
100	Stationary	1–5	184	18.50	60	2.02
20	Rotate 180 deg	1–5	167	6.33	43	1.49
100	Rotate 180 deg	1–5	115	3.94	25	1.28
20	Rotating	1–5	88	2.80	22	1.24
20	Scan+rotate	1–5	85	3.24	24	1.27
100	Rotating	1–5	89	3.30	23	1.26
20	Stationary	10–20	71	2.10	20	1.23
20	Scanning	10–20	70	2.31	22	1.25
100	Stationary	10–20	48	1.73	15	1.17
20	Rotate 180 deg	10–20	46	1.63	11	1.12
100	Rotate 180 deg	10–20	37	1.50	8	1.09
20	Rotating	10–20	30	1.40	8	1.08
20	Scan+rotate	10–20	28	1.36	8	1.08
100	Rotating	10–20	20	1.24	5	1.05
			+ –	× ÷	+ –	× ÷

- Continuously rotating the drum during the count is the best, and it is somewhat better at 100 cm than 20 cm, but does not 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 has a factor of 28, while if a reasonable assumption can show that there are 10–20 hotspots in the drum, then the uncertainty has only a factor of 2 for the simple and efficient 20 cm stationary count, and down to 30–40% with the better geometries. At high energies, even when up close, the uncertainty has 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 a proper interpretation could be made. Using the above case as an example, if the measurement result for ^{241}Am at 60 keV has a factor of 100 below the “limit” then even the quick simple 20 cm 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 100 cm.

Conclusions

This paper presents a brief description of the soon-to-be released ISOCS Uncertainty Estimator (IUE). The software builds upon the basic ISOCS software, which calculates the gamma-ray efficiency for a particular geometry based on input parameters (e.g., sample dimensions, densities, etc.) provided by the user. The IUE estimates the contribution to the efficiency uncertainty due to the uncertainties on the individual input parameters. It performs this uncertainty propagation numerically by probabilistically varying the input values. Two modes of operation have been discussed. The first mode estimates the total efficiency uncertainty for the measurement geometry by simultaneously varying all of the input values. The second mode estimates the uncertainty contribution from each of the input parameters separately. The latter mode is especially useful as a diagnostic tool to determine where to concentrate effort towards reducing the overall measurement uncertainty. These functions are very powerful tools; they allow quick and easy uncertainty analyses that were previously very time-consuming and costly. They also allow hypothetical counting conditions to be evaluated for assay quality during the design process. At present, the software is undergoing internal Quality Assurance testing and validation. When complete, the software will be available as a part of the ISOCS mathematical efficiency calibration software suite.

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