The Role of Mathematical Methods in Efficiency Calibration and Uncertainty Estimation in Gamma Based Non-Destructive Assay - 12311

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ABSTRACT

Mathematical methods are being increasingly employed in the efficiency calibration of gamma based systems for non-destructive assay (NDA) of radioactive waste and for the estimation of the Total Measurement Uncertainty (TMU). Recently, ASTM (American Society for Testing and Materials) released a standard guide for use of modeling passive gamma measurements. This is a testimony to the common use and increasing acceptance of mathematical techniques in the calibration and characterization of NDA systems. Mathematical methods offer flexibility and cost savings in terms of rapidly incorporating calibrations for multiple container types, geometries, and matrix types in a new waste assay system or a system that may already be operational. Mathematical methods are also useful in modeling heterogeneous matrices and non-uniform activity distributions. In compliance with good practice, if a computational method is used in waste assay (or in any other radiological application), it must be validated or benchmarked using representative measurements. In this paper, applications involving mathematical methods in gamma based NDA systems are discussed with several examples. The application examples are from NDA systems that were recently calibrated and performance tested. Measurement based verification results are presented.

INTRODUCTION

In recent years, mathematical methods that calculate gamma ray or X-ray detector responses have become useful in the non-destructive assay of items containing radioactive waste. A properly validated mathematical methodology can indeed be advantageous since it can yield results of desired accuracy, save time, and be cost-effective. Computer codes using a Monte-Carlo approach or a ray tracing methodology, or a combination of both, can be used for calculating detector efficiencies for a wide variety of source geometries. These codes can also be programmed to run efficiency computations by varying the “not well known” (NWK) parameters of a waste item to within given tolerances, and determine the TMU budget for the measurement campaign. The computed TMU can then be verified by performing a representative set of measurements.

The applications discussed in the current paper are based on the Monte Carlo computer code MCNP [1], and Canberra Industries’ In Situ Object Calibration Software (ISOCS) [2-3]. The mathematical methodology embedded in ISOCS is a combination of an MCNP-based response characterization of a gamma ray detector in 4π and a ray tracing approach to compute photon attenuation along path lengths to the detector, through absorbers that are internal and external to the radioactive source.

Nakazawa et al have previously published on the subject of efficiency calibration of NDA systems using semi-analytical mathematical approaches [4]. The current paper builds on the previous publication and includes newer applications and TMU case studies.

MATHEMATICAL METHODS IN EFFICIENCY CALIBRATION OF GAMMA WASTE ASSAY SYSTEMS

For a container matrix of a given density, and a given counting geometry, efficiency data points are generated at several gamma ray energies within the desired range (for e.g. 60 keV to 3000 keV), using a mathematical method of choice. The calculations are repeated for multiple matrix densities, typically, in the 0 to 3 $g.cm^{-1}$ range. An efficiency versus energy calibration curve has to be created for each matrix density. The efficiency curves for the different matrix densities will have to be merged to create a "multi-density" curve or the "multi-curve" for the specific container and counting geometry. If an item is assayed over multiple segments, then a separate multi-curve must be generated for each segment, as well as a multi-curve for analyzing the summed spectrum. If a waste stream consists of multiple container types and/or geometries, the multi-curve calibration must be generated for each container type and geometry.

Monte Carlo computer codes such as MCNP can be used to model the source and the detector, along with the collimator and shielding that may be used in the measurement. However, it could take several hours or days to generate the efficiency data with acceptable precisions.

Ray tracing methodology is another approach to computing efficiency responses at different gamma ray energies. One drawback of a ray tracing code is that it does not take into account the multiple interactions that take place inside the gamma ray detector. Typically when a photon with an energy of several hundred keV enters the active volume of the detector, it undergoes multiple Compton scattering interactions since those interactions are more probable, lose energy, and either escape the active volume or eventually be absorbed photo-electrically when the photon energy falls to a low value (less than 100 keV for e.g.). Thus multiple interactions increase the probability of full energy deposition within the active volume. These interactions have a strong dependence on detector construction, geometry, and active volume, which are frequently non-standard. If not benchmarked to measurements, assumptions in full energy detector response can lead to large errors. A validated Monte Carlo simulation does indeed take into account the multiple interactions within a detector active volume. Nevertheless, ray tracing codes can be effectively employed to calculate photon attenuation through absorbers.

A third mathematical approach is a combination of a Monte Carlo, and a ray tracing method. The ISOCS mathematical method is indeed a hybrid of Monte Carlo and ray tracing methods. In the ISOCS method, the gamma ray detector, typically HPGe, is response characterized at the factory by Canberra Industries. The characterization is performed assuming that the detector is in free space. It is based on an MCNP model developed for the specific HPGe detector, with the model validated using NIST (National Institute of Standards and Techniques) traceable multi-line gamma ray standard sources. The characterization is a “look-up table” consisting of MCNP generated detector efficiencies as a function of energy and spatial coordinates. The Monte Carlo calculations run during the response characterization truthfully simulate the photon interactions that occur within the detector. The characterization extends up to a radius of 500 meters and in 4π directions. The response characterization is used in conjunction with the ISOCS software to generate efficiency data points for a given source geometry. The ISOCS software voxelizes the active regions of the container matrix, and calculates photon attenuation at a given energy through absorbers internal and external to the sources. A ray tracing methodology is followed in calculating the attenuation factors. The attenuation is factored into the free space response and the attenuation corrected efficiency is computed on a voxel by voxel basis. The voxelized efficiencies are integrated over the source volume to yield the overall efficiency for the container matrix. Since the attenuation calculations proceed using a ray tracing method, they are fast and can be completed within a few seconds.

Automated ISOCS System and Calibration of Complex Geometries

Mathematical methods are especially useful in the calibration of complex container geometries. For example, high activity waste may be packed into containers with thick layers of concrete or steel. To perform a measurement based calibration, high activity (greater than 1 mCi or 37 MBq) gamma ray standard sources would be needed in order to obtain reasonable counting precisions ($\pm 5\%$ at 1σ). High activity sources would in turn entail a higher level of safety precautions, proper storage and compliance with license requirements. A mathematical methodology validated by benchmark measurements will alleviate these logistics.

The “Automated ISOCS System” calibration is one such application where the ISOCS mathematical method was used. The system is intended to assay nuclear power plant waste, and therefore activation products and fission products can be expected to be present in the waste stream. The Automated ISOCS system consists of a Broad Energy Germanium (BEGe) model BE5030 with front surface area of 5000 mm^2 and thickness of 30 mm that is integrated with a portable cryostat [5]. The detector was housed with a high activity collimator that had the ability to raise or lower additional shielding based upon container dose rate measurements. The additional shielding consisted of a 16 cm thick lead block that contains three slits (1.25 mm wide, 10 cm high). Pictures of the system are shown in Figures 1.

Figure 1. Front (left) and side (right) view of the Automated ISOCS System.

Waste containers were designed to be placed on a rotating platform that is on rails and moved using a motor. The platform distance and collimator were adjusted to one of three preset configurations, based on dose rate measurements: a) 1 meter distance, collimator open, b) 4 meter distance, open collimator, and c) 4 meter distance, closed collimator. Exact container-to-detector distances vary slightly depending on the container geometry.

All containers are continuously rotated during the assay except for the rectangular concrete container. The concrete container has inner dimensions of 1100 mm x 1100 mm x 1000 mm (length x width x height) and a concrete wall thickness is 120 mm. In the mathematical modeling, the radioactive material is approximated to have a homogeneous source distribution inside the rectangular container. To average out non-uniformities, measurements are made on each side of the box, the spectra were summed, and analyzed using summed efficiencies. The detector is shielded using a cylindrical lead shield of length 152.4 mm and thickness of 94.2 mm. For low and medium dose rates (< 0.1 Sv/hr), an "open" collimator is used. The "open" collimator is a square lead collimator of width 101.6 mm and a thickness of 94.2 mm. The inner surfaces of the collimator and the shield are lined using copper and tin filters to attenuate the lead X-rays. The detector front surface was located flush with the cylindrical shield and consequently immediately behind the collimator. The distance from the front surface of the collimator to the detector was 203.2 mm. For high activity waste (dose rates > 0.1 Sv/hr) an additional 156 mm thick lead attenuator with three slots drilled through it, was introduced in front of the square collimator. The slots had a slant angle of 3.8 degrees. The ISOCS software allowed for accurate modeling of all of the collimator details, including the tin-copper liners and the slotted absorber. The multi-density efficiency curve, calculated using ISOCS for the concrete container geometry and the "open" collimator is shown in Figure 2. The solid lines are sixth-order, log-log polynomial fits.

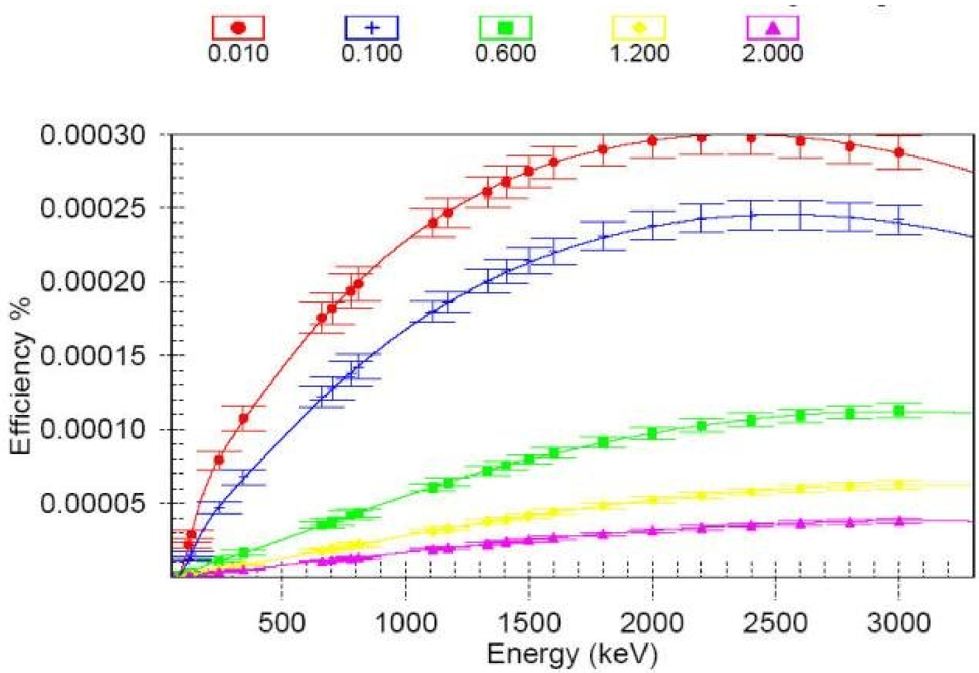

Figure 2. Multi-density Efficiency Curves for the Rectangular Concrete Container assayed with the "open" collimator; the values in the legend at the top of the figure are densities in units of g.cm^{-3}.

Since the concrete container was not available at the factory, a cardboard container filled with foam matrix (density of 0.02 g.cm^{-3}) was used to verify the ISOCS computation method. The dimensions of the cardboard box were 800 mm x 1000 mm x 1520 mm (length x width x height). Four Eu-152 gamma ray rod source standards were inserted into holes in the matrix. The gamma line sources are each comprised of nominally 5 µCi of Eu-152. The positions of the source rods are vertically centered within the box, and the 229 and 102 mm horizontal displacements are symmetric around the box for the four rods. Figure 3 shows the positions of the rod sources. The box was located 1 meter away from the detector. The "open" square collimator was used. The box was manually rotated by 90º every 900 seconds during the assay, and the spectra were summed.

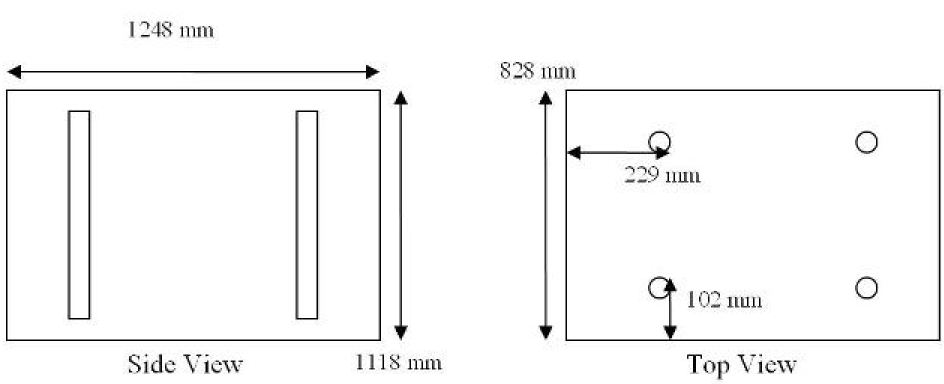

Figure 3. Position of Source Rods within the Simulated Cardboard Box.

The multi-density efficiency curve computed using ISOCS is shown in Figure 4 below. For this geometry, the solid lines are fifth-order, log-log polynomial fits.

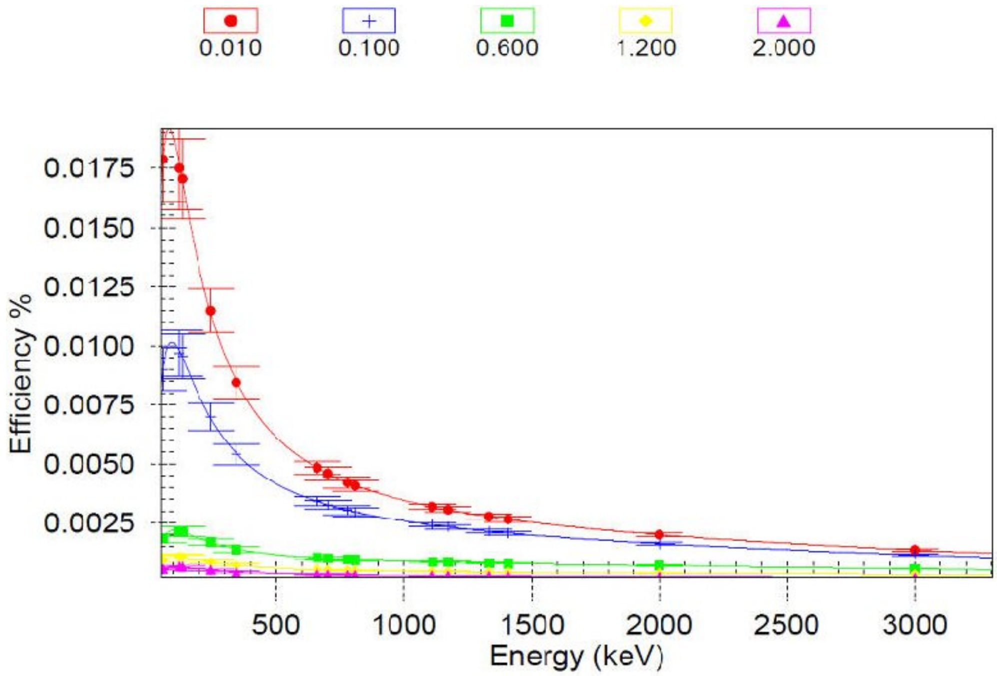

Figure 4. Multi-Density Efficiency Curve for the Cardboard Box Geometry used for ISOCS Verification; the values in the legend at the top of the figure are densities in units of $g.cm^{-3}$.

The measured activities as a function of gamma ray energy are given in Table 1 below and are compared to the expected activities. The TMU for the cardboard box was adapted from a similar calculation performed [6] on a Standard Waste Box (SWB) and a Standard Large Box (SLB). The TMU budget is given in Table 2.

Table 1. Comparison of Measured to True Activities for the 800 mm x 1000 mm x 1520 mm Cardboard Box

Energy (keV)	**Measured Activity (μCi)**	**TMU (%)**	**Expected Activity (μCi)**	**Deviation (number of σ)**
121	17.64	23	18.3	-0.163
244	24.62	22	18.3	1.167
344	25.03	22	18.3	1.222
778	24.84	21	18.3	1.254
1112	23.71	21	18.3	1.087
1408	21.87	21	18.3	0.777

Table 2. Estimates of 1-σ Uncertainty contributions for an assay of Calibration Verification Box Count in a waste container that has a matrix density of 0.015 g/cc.

Uncertainty Source / Energy (keV)	121	244	344	778	1112	1408
Counting Statistics	< 5%	< 5%	< 5%	< 5%	< 5%	< 5%
ISOCS / Multicurve Calibration	10%	8%	8%	6%	4%	4%
Matrix Inhomogeneity	0 – X% subject to each assay					
Rate Loss Correction	< 0.5%	< 0.5%	< 0.5%	< 0.5%	< 0.5%	< 0.5%
Non-Uniform Source Distribution	20%	20%	20%	20%	20%	20%
Background	< 1%	< 1%	< 1%	< 1%	< 1%	< 1%
Nuclide Interferences	< 1%	< 1%	< 1%	< 1%	< 1%	< 1%
Self Attenuation / Lump	0 – X% subject to each assay					
Fill Height	5%	5%	5%	5%	5%	5%
System Stability / Other	0 – X% subject to each assay					
TMU	23%	22%	22%	21%	21%	21%

The measured activities given in Table 1 agree with the certified activities to within 1σ level of uncertainties on average.

AN EXERCISE TO ESTIMATE TMU COMPONENTS FOR A WASTE ITEM

As a case study, TMU components were estimated for the 1100 mm x 1100 mm x 1000 mm concrete box discussed previously. Three of the important components that contribute to TMU, namely matrix density, matrix fill height, and non-uniform distribution of source activity, were explored computationally using the "ISOCS Uncertainty Estimator" (IUE) software package. The "not well known" (NWK) parameters of a waste item are input into the IUE, with a lower and an upper bound value for the given parameter. A probability distribution for generating values of the given parameter within these bounds is also indicated (for example, Gaussian, Uniform, Triangular etc). The IUE software then creates a desired number of input models of the given item by generating different NWK parameters per the probability distribution indicated. ISOCS computations are performed using each input model, and quantities such as the average efficiencies and standard deviation among the computed models are calculated. The efficiencies for each model are tabulated and are available to be compared to the efficiency that was used in the calibration of the waste item. The calibration efficiency is designated as <R> in the figures and discussion below.

Variation of Matrix Density

One of the data points for the multi-density efficiency calibration of the concrete box was 0.6 g.cm^{-3}. In the TMU estimation, density of the item was varied from 0.2 g.cm^{-3} to 1.0 g.cm^{-3}. The concrete box was assumed to be 100% filled (1000 mm) with radioactivity distributed uniformly throughout the volume. One hundred (100) ISOCS models were generated using a Gaussian distribution function, and the efficiencies were calculated using each model at several energies between 121 keV and 1408 keV. The ratio of the efficiency (or response) for each model was taken with respect to the calibration data point at 0.6 g.cm^{-3}. This analysis aims at estimating non-uniformities, errors, or unknowns in the matrix material, where the only observable is the bulk density. Figures 5a and 5b show the distribution of the response ratios at 121.8 keV and 662 keV, respectively.

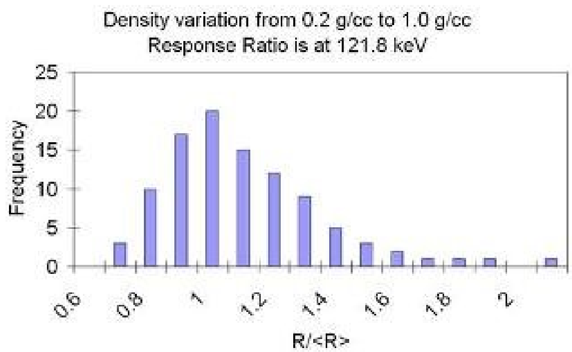

Figure 5a. Density variation (121.8 keV)

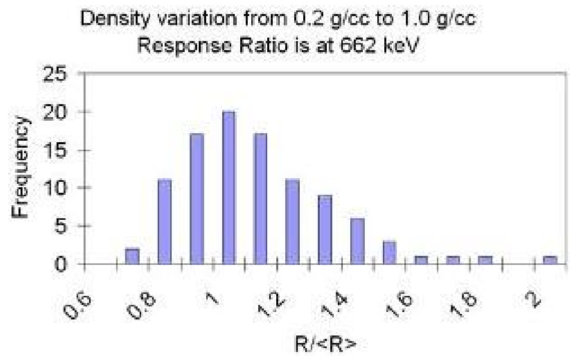

Figure 5b. Density variation (662 keV)

The plots show that the distributions are reasonably symmetric about the calibration density of 0.6 g.cm^{-3} at the 1σ level. The data points at the extremes of the distribution were assumed to correspond to ±3σ limits, and TMU component was determined as (R_{max} – R_{min})/6σ. Within the limits explored, the "Gaussian" distribution has captured almost all of the probability of variation in response. Therefore this assumption seems reasonable for uncertainty estimation to a first order despite the sparse number of models simulated. The intent is not to accurately determine the standard deviation of the uncertainty distribution.

Variation of Fill Height

The matrix density of the waste item was maintained at 0.6 g.cm^{-3} and the fill height was varied from 50% to 100%. A uniform source distribution was assumed. The response ratios with respect to the calibration condition of a fill height of 100% were calculated at various energies. Figure 6a and 6b give the distribution of the response ratios at 121.8 keV and 662 keV, respectively, for fill height variation.

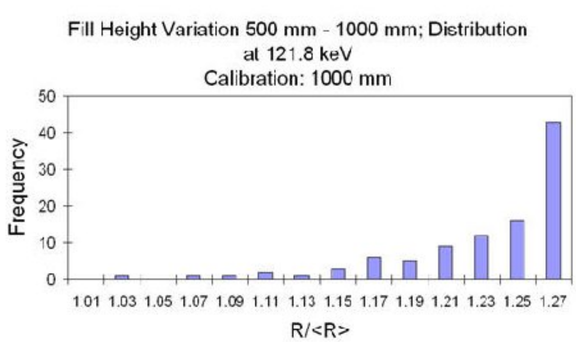

Figure 6a. Fill height variation (121.8 keV)

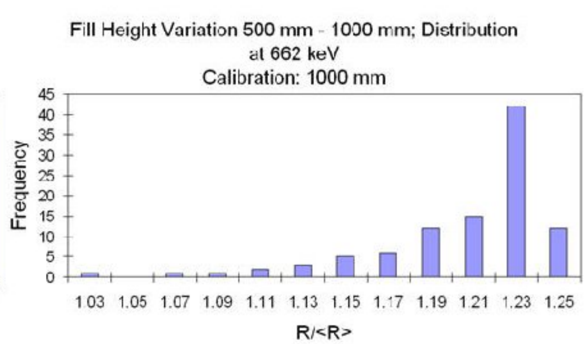

Figure 6b. Fill height variation (662 keV)

From figures 6a and 6b, it is evident that the response ratios with respect to the calibration response (100% full) are all greater than unity and that the ratio distributions are strongly asymmetric. The R/<R> ratios for the 121.8 keV gamma ray energy tend to be clustered into the bin at 1.27, while the ratios for the 662 keV gamma ray energy tend to be clustered around the bin at 1.23. If there is an energy dependence, it is very mild. In the examined fill height scenario (50% to 100%) the efficiencies for the less than full geometries are all greater than the efficiency for the 100% full geometry. The reason for this can be understood by examining the counting geometry for this system (Figure 7).

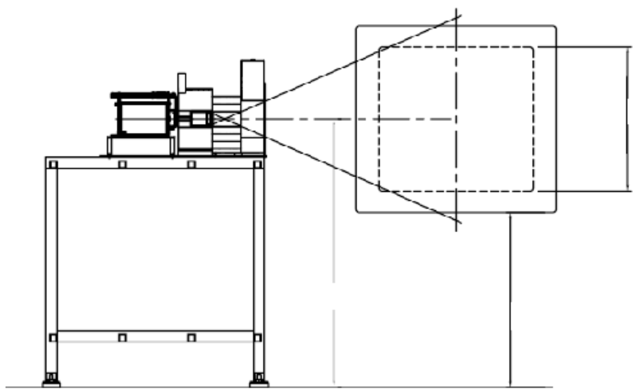

Figure 7. Field of View with the Concrete Box at a 1 meter distance (open collimator).

When the box is 100% filled, the gamma emissions from volume elements (or voxels) closer to the top or the bottom of the matrix are shadowed by 94.2 mm thick collimator. Also, the gamma rays emitted closer to the top or the bottom have a greater path length through the concrete liner of the box, than the gamma rays emitted from the mid-section of the box. By configuring a fill height that is less than 100%, the voxels that contribute less to the efficiency are not included, and therefore the overall efficiency of the source region is weighted in favour of the higher efficiency voxels. The plots shown in 6a and 6b indicate that under the current calibration conditions, the uncertainty component due to the fill height introduces a high (positive) bias. The relative bias is taken to be the average value of the R/<R> ratio at a given energy. If one had done a calibration based on, say a 50% full box, then the bias would have been a low (negative) bias. This exercise shows that not all uncertainty components are symmetric, and one cannot simply take the approach of (max-min)/nσ. At increasing distances, these effects might be less dramatic at the expense of sensitivity. In addition, simple ray-tracing from the active volume of the detector to active area of the assay items, as in Figure 7, may not be optimal for collimator design. With mathematical methods, it is now possible to explore these scenarios more thoroughly and determine uncertainties that are realistic.

Non-Uniform Source Distribution

The uncertainty component due to a non-uniform source distribution was explored by modeling hot spots of radioactivity within the container matrix. The activity was assumed to be present only at the hot spots and not in other portions of the matrix. The concrete box was arbitrarily assumed to contain five (5) hotspots. Five hundred (500) random distributions of the 5-hot spot configurations were modeled in the IUE software. The hot spots were 30 mm x 30 mm x 30 mm cubes distributed inside the item of inner dimensions 1100 mm x 1100 mm x 1000. The 5 hot spots totaled up to approximately 11% by volume of the container matrix. The histograms for 121.8 keV and 662 keV are shown below in Figures 8 and 9, respectively.

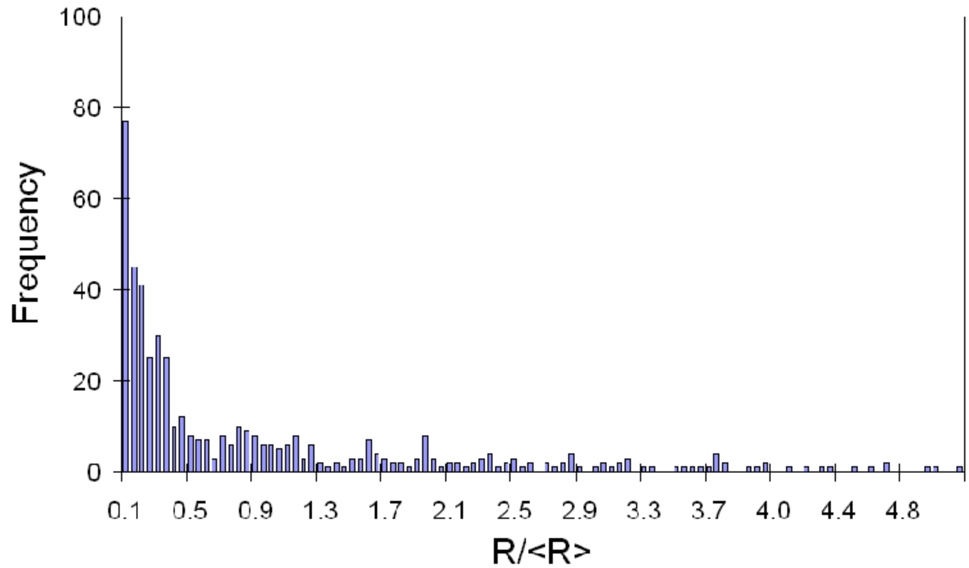

Figure 8. Response Ratio distribution for 5 Hot Spots inside the Concrete Box (1210 liters) at 121.8 keV. Each hot spot in 27 liters, and density of matrix is 0.6 $g.cm^{-3}$.

The distribution for the 121.8 keV gamma ray energy is highly skewed towards the low response values. In a cellulose matrix of density of 0.6 $g.cm^{-3}$, the mean free path of 121.8 keV photons is 11 cm approximately. The inner dimensions of the concrete box are 110 cm, in other words, ten times the mean free path of the 121.8 keV photons. The high attenuation suffered by the 121.8 keV gamma rays for sampled locations that are deeper into the matrix out weighs any efficiency gains from sampled locations that have shallower path lengths.

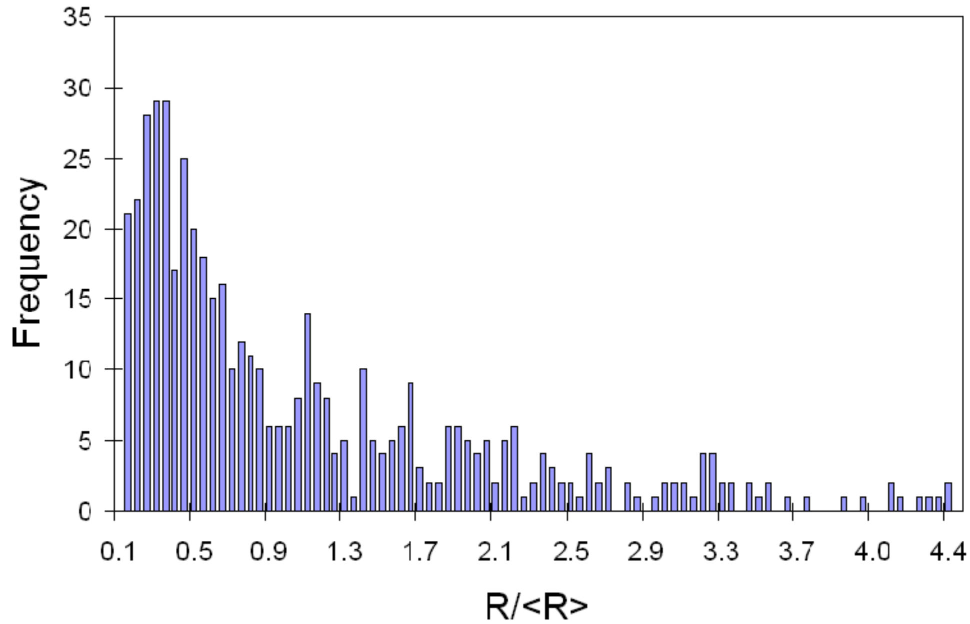

Figure 9. Response Ratio distribution for 5 Hot Spots inside the Concrete Box (1210 liters) at 662 keV. Each hot spot in 27 liters, and density of matrix is 0.6 $g.cm^{-3}$.

For 662 keV photons, the distribution is less skewed, but still non-Gaussian.

For these non-Gaussian distributions, biases that are asymmetric about the uniform distribution (R/<R> = 1) are calculated. Equations (1) and (2) are used to determine an average response ratio R/<R> by weighting the histogram bins by the corresponding frequencies f. Equation (1) gives the weighted average of the response ratios for bins ≤ 1, and equation (2) gives the weighted average for bins ≥1:

$$\left\langle \frac{R}{\langle R \rangle} \right\rangle_{\leq 1} = \frac{\sum_{0}^{1} \left(R / \langle R \rangle \right)_i \cdot f_i}{\sum_{0}^{1} f_i}, \qquad \text{(Eq. 1)}$$

$$\left\langle \frac{R}{\langle R \rangle} \right\rangle_{\geq 1} = \frac{\sum_{\geq 1} \left(R / \langle R \rangle \right)_i \cdot f_i}{\sum_{\geq 1} f_i}. \qquad \text{(Eq. 2)}$$

The weighted averages for the response ratios can be taken as the relative biases. Other approaches, such as using theoretical standard deviations from exponential and rectangular distributions when appropriate, have also been investigated and explored [8].

The magnitudes of TMU components calculated in this exercise are given in Table 3. It must be noted that the TMU estimates presented are for a narrow regime of parameter variations. For example, one can increase the number and/or the size of the hot spots to determine an optimum number of hot spots for a given container size. Mathematical methods offer the flexibility to undertake such studies. The intent in this paper is to demonstrate that mathematical methods are are not only a viable approach but for many reasons the preferred approach for TMU estimation. Ray tracing methods such as the ones employed in ISOCS tend to be computationally faster when compared to Monte Carlo codes such as MCNP. However, in compliance with good practice, any TMUs obtained using mathematical methods must be validated using a representative set of measurements.

Table 3. Results of the example TMU calculation using the IUE

Energy (keV)	Density Variation	Fill Height Variation	Non-Uniform Source (5 Hot Spots)
121.8	± 24.20%	+22.4%	-70.5%, +128.2%
662	± 21.51%	+19.6%	-56.6%, +101.8%
1332	±1 8.66%	+17.6%	-45.6%, +79.6%

ACCEPTANCE OF MATHEMATICAL METHODS BY NDA PRACTITIONERS

As a symbol of the growing acceptance of mathematical methods, the ASTM sub-committee C26.10 on NDA Techniques released a "Standard Guide for Passive Gamma Measurements Using Modeling" in the year 2010 [7]. The standard guide, C1726-10 is consensus guide, and is a testimony to the fact that modeling using mathematical methods has become common practice. The ASTM guide is applicable to assay of radionuclides in containers, whose gamma-ray absorption properties can be measured or estimated, for which representative certified standards are not available. It can be applied to in situ measurements or to laboratory measurements. The guide goes on to state that the methods discussed in it assist in demonstrating regulatory compliance in areas such as nuclear safeguards (SNM), waste disposal, criticality control, inventory control, and decontamination and decommissioning. The ASTM guide C1726-10 covers a wide variety of modeling techniques including Generalized Geometry Hold-up (GGH), Far-field approximation, Monte-Carlo Radiation Transport codes, and Hybrid methods that combine Monte Carlo and ray tracing approaches.

Mathematical methods such as ISOCS that have the built-in capability to estimate TMU components, and that have a large body validation and verification data, are candidates for assaying transuranic waste and have the potential to satisfy Waste Acceptance Criteria (WAC) for the Waste Isolation Pilot Plant (WIPP).

CONCLUSION

Mathematical methods play an important role in the efficiency calibration of gamma based NDA systems. This is especially true when the measurement program involves a wide variety of complex item geometries and matrix combinations for which the development of physical standards may be impractical. Mathematical methods offer a cost effective means to perform TMU campaigns. Good practice demands that all mathematical estimates be benchmarked and validated using representative sets of measurements.

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