EVALUATING THE DEPENDENCE OF GAMMA-ASSAY RESULTS ON SOURCE AND MATRIX DISTRIBUTIONS

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ABSTRACT

Quantitative high resolution gamma-ray spectroscopy is extensively deployed to non-destructively characterize waste containers. Traditionally the assay result is reported as if the activity is uniformly distributed within the matrix and assumes the matrix is homogeneous and fills the container. Deviations from these conditions result in a bias which is traditionally allowed for in an ad hoc fashion by expanding the total measurement uncertainty reported. Detailed experimental study is tedious and hampered by practical problems such as the production of a large number of test cases. For this reason we have elected to use the Monte Carlo technique to investigate some salient aspects of the problem so that a better treatment might be formulated.

In this work we concentrate on the measurement of the standard waste box (SWB) in a standard gamma box counter configuration. We explore the impact of non-uniform source distribution, matrix fill height and matrix inhomogeneity by running a large number of scenarios. We show how the reported uncertainty may be reduced when there is reason to believe multiple hot spots are present.

Key Words: HRGS, waste assay, TMU

INTRODUCTION

Non-destructive high resolution gamma-ray spectroscopy (HRGS) assay is a workhorse technique within the DOE complex for the sentencing of transuranic and other radioactive wastes. Understanding the capabilities and fundamental limitations of the technique is key to improving the application and reporting protocol. It is well known, for example, that for segmented drum scanners [1,2] matrix correction factors are a dominant source of uncertainty at low energies/high densities. In this work we consider a generic version of a general purpose gamma box counter [e.g. 3] applied to the measurement of standard waste boxes (SWBs) and ask what the impact of matrix inhomogeneity, matrix fill height and activity distribution have on the assay result based on a calibration referred to uniform matrix and uniform activity distribution. These uncertainties are often large and uncontrollable compared to, say, counting precision and the emission rate of certified reference materials used to establish the absolute full energy peak efficiency under reference conditions.

To map out the spatial response and explore the parameter space of matrix variability experimentally would require an excessively large number of trails. It is more cost effective and a better use of resources to use Monte Carlo simulation techniques to generate the sought after probability density functions (PDFs). These can then be combined with the other significant sources of uncertainty to form the overall or total measurement uncertainty.

SIMULATION METHOD

The simulation tool used in this was developed in-house especially for carrying out such evaluations. It has been described elsewhere [4,5]. In essence, a series of point detectors may be distributed around the item to be measured emulating the effective pattern resulting when the item is scanned passed the detector array. The dimensions and structure of the container are input and the code maintains incoherent mass attenuation coefficients for a selection of materials. The matrix inside the container is represented as a 20x20x20 3-D array of volume elements (voxels). In a given trial each the linear attenuation coefficient of each of the 8000 voxels is picked according to the user specified material-density-voidage distribution. Point gamma emitters are placed at random in the matrix and the summed response, R, is generated by applying Beer's Law of exponential photon attenuation from the source(s) to all detectors along the ray segments through each voxel and container wall. Many trials (typically 1-10 million source positions) are used to generate the spatial PDF for a given matrix.

The scan configuration adopted here is typical of the Canberra WM-2500 Series Box Counter applied to SWB containers (approximately 2000 ℓ). A pair of opposing detectors are mounted on either side of the box and the box is counted at three positions along its length making for a total of 12 views. The scan pattern is chosen to provide partially overlapping field fields of view so as to obtain a reasonably even areal coverage.

A common practice in waste assay is to calibrate the efficiency of the assay instrument by measuring a known, uniform, calibration standard container. Therefore, in the Monte Carlo calculations, the efficiency for each nonuniform source / matrix geometry was calculated and the ratio was taken against the efficiency for a container that is truly uniform both in activity distribution and absorbing matrix. Thus, a ratio of unity indicates that the assay result is "accurate", that is, it would reproduce the true activity in the container. A ratio greater than unity indicates that the assay result for that geometry would be an overestimate of the true activity, and vice versa.

SWB CONTAINER AND WASTE FORMS

The SWB is approximately 156 cm L x 131cm H x 93 cm W. It is constructed from steel with a mean wall thickness of about 0.36 cm (support braces result in some variation point to point). The detector positions where located vertically at 1/3 and 2/3 of the internal height and the horizontal scan positions were 1/6, 3/6 and 5/6 along the internal length. The collimation of the detector (45 degree opening) was such that with the detectors 80 cm from the container wall the viewing cone was approximately 288 cm across at the mid-plane of the box.

Calculations were run for all combinations (182 in total) of the seven gamma-ray energies and 26 material-density-void fraction combinations listed in Table 1 below. For a given material-density combination, the bulk density was held constant for various void fractions. This was done by scaling the density of each material appropriately. For example, consider the "Concrete, 1.0 $g \cdot cm^{-3}$" combination. For a void fraction of 0%, the matrix was completely filled with concrete at 1.0 $g \cdot cm^{-3}$. For a void fraction of 20%, one fifth of the matrix voxels were filled with air (density roughly 0 $g \cdot cm^{-3}$), and four fifths of the voxels were filled with concrete at a density of 1.25 $g \cdot cm^{-3}$. Thus the bulk density of the matrix ($0.2 * 0\ g \cdot cm^{-3} + 0.8 * 1.25\ g \cdot cm^{-3}$) remained unchanged.

For each combination (i.e. for each linear attenuation coefficient (μ) value and void fraction), 500,000 configurations were run assuming five point sources randomly placed within the container volume. The counting geometry for the SWB was modeled as described above. The overall behavior of the ratio distributions versus sample bulk density and void fraction has been presented in detail in earlier work for some other sample containers [4,5].

Energy (keV)		Material, Density ($g \cdot cm^{-3}$)	Void Fractions (%)
59	(^{241}Am)	Air, 0.0012	0 (Air *is* void)
129	(^{239}Pu)	Cellulose, 0.1	0, 10, 20, 30
186	(^{235}U)	Cellulose, 0.4	0, 10, 20, 30
414	(^{239}Pu)	Concrete, 0.7	0, 10, 20, 30, 40
662	(^{137}Cs)	Concrete, 1.0	0, 10, 20, 30, 40
1001	(^{238}U)	Concrete/Steel, 1.6	0, 10, 20, 30, 40, 60, 80
1333	(^{60}Co)		

Table 1. Summary of γ-lines (nuclides) and matrix materials modeled. The concrete/steel combination was an 0.8:0.2 ratio by volume.

RESULTS AND DISCUSSION

As discussed elsewhere [4], for the case of a uniform matrix (i.e. 0% void fraction), the distribution of assay results is determined solely from the random placement of sources within the matrix, and the average calculated ratio is unity. The presence of void space has a tendency to bias the assay results high [5]; this is due to the increasing availability of low-attenuation shine paths. The mean assay ratios for the SWB containers are depicted in Figure 1 below. While a dramatic bias is evident for higher attenuation (μ, *'Mu'*) values, typical assay situations for containers as large as SWB's are limited to linear attenuation coefficient values from 0.01 to 0.1 cm^{-1} and void fractions no more than about 40%. Within this range the bias is no more than roughly a factor of 1.3 for the highest attenuations.

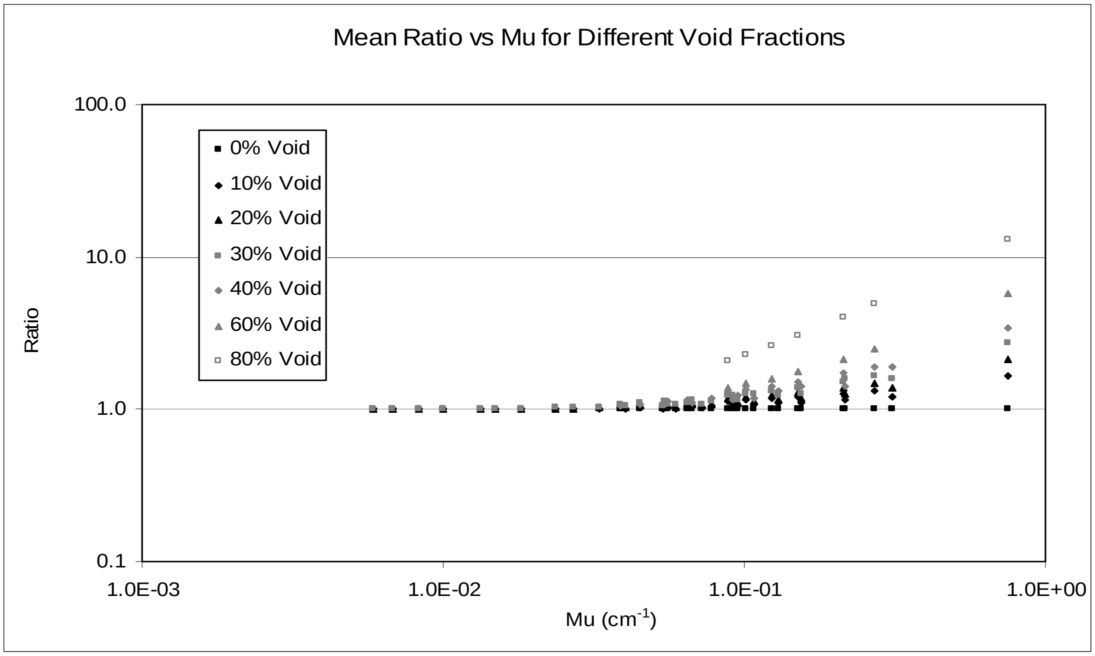

Figure 1.

It is also possible to estimate confidence bands from these distributions. For estimating total measurement uncertainty (TMU) values, a common starting point is to utilize $\pm 3\sigma$ bounds. These calculated limits for the SWB container are presented below in Figures 2 and 3 below.

It should be noted that, since the probability distributions are generally quite asymmetric, it doesn't seem sensible to simply calculate the root of the second moment about the mean of each distribution and report that as the "standard deviation". Instead it makes more sense to approach these distributions from a probabilistic standpoint. Specifically, for a pure Gaussian distribution, the $\pm 3\sigma$ bounds encompass ~ 99.730% of the probability under the distribution; or alternatively ~ 0.135% of the probability lies outside the bounds in each direction. Understanding this, one can then look for similar probability bounds for the above asymmetric ratio distributions. Namely, one can define the probabilistic equivalent to "$\pm 3\sigma$" bounds as those that encompass 99.730% of the probability. This is the approach that was used to obtain the data shown in Figures 2 and 3.

For linear attenuation coefficient values below roughly 0.01 cm^{-1}, the $\pm 3\sigma$ boundaries flatten out to roughly ± 30%. This includes all of the empty box (i.e. air-filled) cases, which have attenuation coefficient values in the 10^{-5}-10^{-4} cm^{-1} range, and so these cases are thus below the plotted μ-range in the figure.

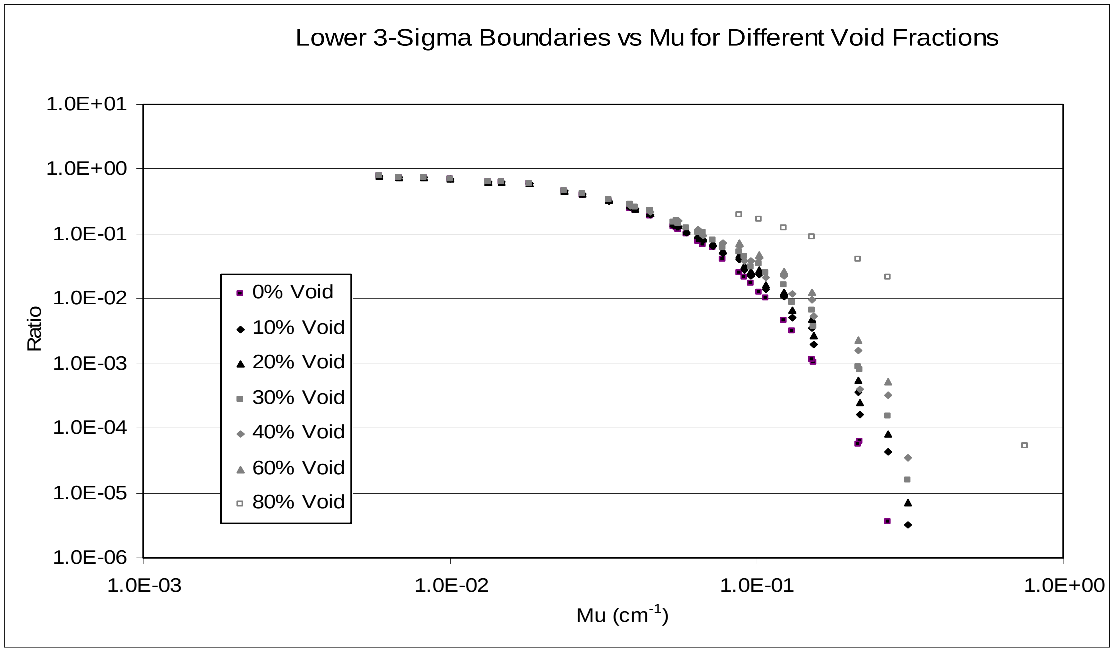

Figure 2. Lower 3-sigma confidence limits for the SWB.

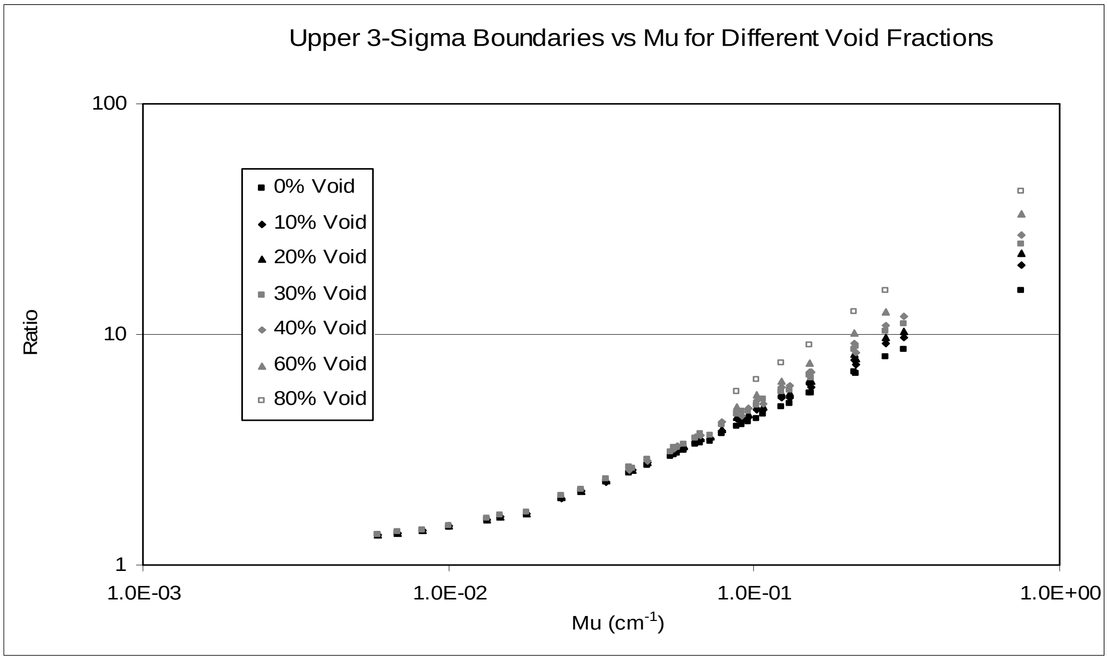

Figure 3. Upper 3-sigma confidence limits for the SWB.

As with earlier calculations with other containers [4,5], the bounds deviate further from unity with increasing attenuation. Also, both the upper *and* lower bounds increase with void fraction, as is consistent with the entire distribution shifting upwards with increasing void fraction.

Assuming the same “typical” assay conditions as above – linear attenuation coefficient values between 0.01 and 0.1 cm^{-1} and void fractions no more than 40% – the conservative “worst case” upper and lower 3σ bounds correspond to roughly a factor of 5 overestimate and a factor of 75 underestimate for the highest attenuations considered.

CONCLUSIONS

A physical measurement is incomplete without an associated statement of uncertainty. For waste measurements generating uncertainty and bias estimates is often quite challenging because of the wide variation in the waste forms encountered. In this work we approached the evaluation problem, for the case of a SWB container in a generic HRGS box counter, by Monte Carlo simulation. The importance of source non-uniformity and matrix heterogeneity are difficult and tedious to estimate except by simulation and traditionally allowances for them have been propagated into the assay report in an ad hoc manner. This work is part of an effort to set this problem onto a better foundation.

We find that the response to point source position within the matrix is dramatic and results in an asymmetric PDF. When multiple randomly located ‘hot spots’ may be assumed the PDF narrows and becomes more symmetric so that central region become Gaussian like. For a few point sources deviations persist out into the tail so that, for example, large excursions (‘several sigma’) are more common than a simple Gaussian distribution model would suggest. These observations would argue for the reporting of asymmetric uncertainty bands and in the most demanding situations of full PDFs rather than of a simple confidence interval.

Fill height is an example of a gross deviation from uniform matrix filling the container. Results not shown in this paper indicate it is an important variable. If radiography data is available then a calculated fill height correction factor can be applied to remove the bias. The detector heights could also be adjusted. If the fill height is not known then in general a one sided bias will result. It may be better in such cases to calibrate to a partially filled reference condition (e.g. 70% full) so that variations both above and below might be expected so that the waste-stream averaged result is more accurate. The best course would require understanding of the nature of the waste stream.

Matrix attenuation is generally less for a heterogeneous fill than for a uniform matrix of equal mean density. This is because gamma rays stream through the voids unimpeded. This is a manifestation of an effect seen in shielding walls constructed of ‘pebbles’. For assays based on a correction factor derived from net weight (mean density) and a uniform matrix calibration this will result in a tendency for over-reporting. The degree bias depends on the level of structure in the matrix and in general will be difficult to estimate even if radiography data is to hand – although this is an intriguing avenue for future exploration. However, this realization argues for the use of calibration reference materials which have a degree of voidage so that they are somewhat more representative of solid sundry wastes. The voidage would need to be taken into account is any benchmark modeling designed to verify the model to permit it to be used to extend the calibration and to generate uncertainty budgets.

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