The Special Properties of Massimetric Efficiency Calibrations as Compared to the Traditional Efficiency Calibration for D&D and ER Gamma Spectroscopy Measurements -- 10021

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ABSTRACT

The traditional method of representing the efficiency calibration for a gamma spectroscopy system is peak counts detected in the spectrum per unit time, divided by the number of gammas at that energy emitted from the source in the same unit of time – i.e. (counts per second emitted) ÷ (gammas per second detected), or counts/gamma. Massimetric efficiency is merely the product of traditional efficiency and the mass of the calibration source – i.e. (counts · grams) ÷ gamma. A mathematically equivalent way of expressing it is (counts per second detected) ÷ (gammas per second emitted per gram of sample).

Common characteristics that exist in assays of an item [e.g. container of waste, large area of soil, …] for purposes of Decontamination and Decommissioning [D&D] and Environmental Remediation [ER] include: computing the concentration of radioactivity [Activity/gram] for the item for comparison to a control concentration; measuring large items; not knowing the exact size of the item; not knowing the exact density of the item; not knowing the exact atomic composition of the item.

For traditional calibrations, these variations in size, density, and composition cause large variations in the efficiency. Therefore for accurate results, the size, density, and composition of the item must be well known and a proper calibration for these conditions must be obtained. This is very difficult to do for most D&D and ER projects. But if the item is large, as is normally the case for this application, then the use of Massimetric calibrations can give reasonably accurate results even when the size, density, and composition of the items is not known. Massimetric efficiency is shown to reduce the standard deviation by a factor of 3 or more, and is shown to result in standard deviations of 10-30% for a wide range of unknown sample conditions commonly encountered in D&D and ER projects.

INTRODUCTION

It is traditional in gamma spectroscopy to calibrate detectors in terms of absolute efficiency, i.e. counts in the spectral photopeak per gamma of that energy emitted from the sample during the counting time. The unknown sample is weighed, and placed into the calibrated counting geometry for assay. The net peak counts are then converted to activity using the traditional efficiency calibration to determine activity of the sample [e.g. Bq]. That activity is then divided by mass to get Activity Concentration [e.g. Bq/g].

$$\text{Activity in Bq} = \frac{(\text{counts})}{(\text{efficiency}) \, (\text{counting time})} \qquad [1]$$

$$\text{Concentration in Bq per gram} = \frac{\text{Bq}}{\text{mass}} \qquad [2]$$

Where: counts = background corrected counts in the photopeak of interest
efficiency = peak counts detected per gamma of that energy emitted
time = seconds
mass = grams of the item

This efficiency calibration has a few well known characteristics:
- As the item diameter increases, the efficiency decreases
- As the item thickness increases, the efficiency decreases
- As the item density increases, the efficiency decreases

All of these effects are because the part of the item that is being added as that parameter increases is further away from the detector [lower efficiency] and because the added part is attenuated by the closer parts of the item [lower efficiency].

For most kinds of waste assay or environmental measurements the end result is concentration – e.g. Bq/g. This value is used to compare against release limits, to compute dose, and for comparison to natural levels of radioactivity.

Equations [1] and [2] can be combined to compute the concentration in a single step as follows:

$$\text{Concentration in Bq per gram} = \frac{(\text{counts})}{(\text{efficiency}) \, (\text{grams}) \, (\text{counting time})} \qquad [3]$$

The quantity "(efficiency) (grams)" in the denominator of equation [3] is what we call Massimetric efficiency. The Massimetric efficiency is simply the traditional efficiency multiplied by the mass of the sample.

Another way of representing the Massimetric efficiency is the counts per second detected per gamma per second emitted **per gram of sample**, or cps/γps/g.

MASSIMETRIC EFFICIENCY FOR LARGE ITEMS

For many common measurement scenarios, the item being measured can be considered infinitely large. It is infinitely large when adding more sample (bigger diameter, more thickness, higher density) doesn't result in any higher count-rate in the detector. When approaching these situations, the Massimetric efficiency can be shown to offer significant advantages.

Observe in figures 1-3 how the behavior of the Massimetric efficiency differs from the traditional efficiency:

- As the sample diameter increases, the efficiency increases, and approaches a maximum
- As the sample thickness increases, the efficiency increases, and approaches a maximum
- As the sample density increases, the efficiency increases, and approaches a maximum.

Consequently, as long as the sample is big enough and dense enough, then a single efficiency calibration can be used. The actual size of "infinite" is a function of the source-detector geometry. Distant items must be quite large, but close items in favorable geometries need only be a few liters in size.

The Canberra ISOCS efficiency calibration software [1] was used for these computations. A standard feature of the ISOCS software since its inception has been the ability to do Massimetric efficiency calibrations [efficiency · mass]. In addition, the software can also compute the efficiency calibration as [efficiency · area] and [efficiency · length]. These types of calibrations also have similar special properties of the Massimetric efficiency, in that they are also infinite size calibrations, and will allow the direct computation of results in units of Bq per meter of length, or Bq per square meter of area, should that be useful.

All calibrations in this document were done with a typical mid-sized p-type coaxial detector, however the results are applicable to all sizes and types of gamma spectroscopy detectors. All calibrations were done with the detector at 1 meter from the surface of the source. Efficiency calibrations were done for all conditions at energies of 20 keV, 60 keV, 200 keV, and 1000 keV.

Figure 1 shows how the Massimetric Efficiency changes as the diameter of the source is changed, but all other parameter are held constant. The efficiency is normalized to the efficiency at the largest diameter (720m) for that energy. The source matrix is water, the source thickness is 1 meter, and the density is 1 g/cc. Near-maximum efficiency is reached at about 100 meters. Lower energies reach this at smaller diameters than higher energies. If 10% accuracy is acceptable, then a calibration at ~15m diameter can be used for all sources greater than ~9m diameter when the detector is 1m from the source. If the detector was only 10cm from the source then the same accuracy can be reached when the source diameter is ~1m.

Figure 2 shows how the Massimetric Efficiency changes as the thickness of the source is changed, but all other parameter are held constant. The efficiency is normalized to the efficiency at the largest thickness (1m) for that energy. The source matrix is water, the source diameter is 100m, and the density is 1 g/cc. Maximum efficiency is reached at ~10cm for 20 keV and ~50cm for 1000keV. If 10% accuracy is acceptable, then a calibration at ~30cm thickness diameter can be used for all thickness values greater than ~20cm and for all energies less than ~2000 keV.
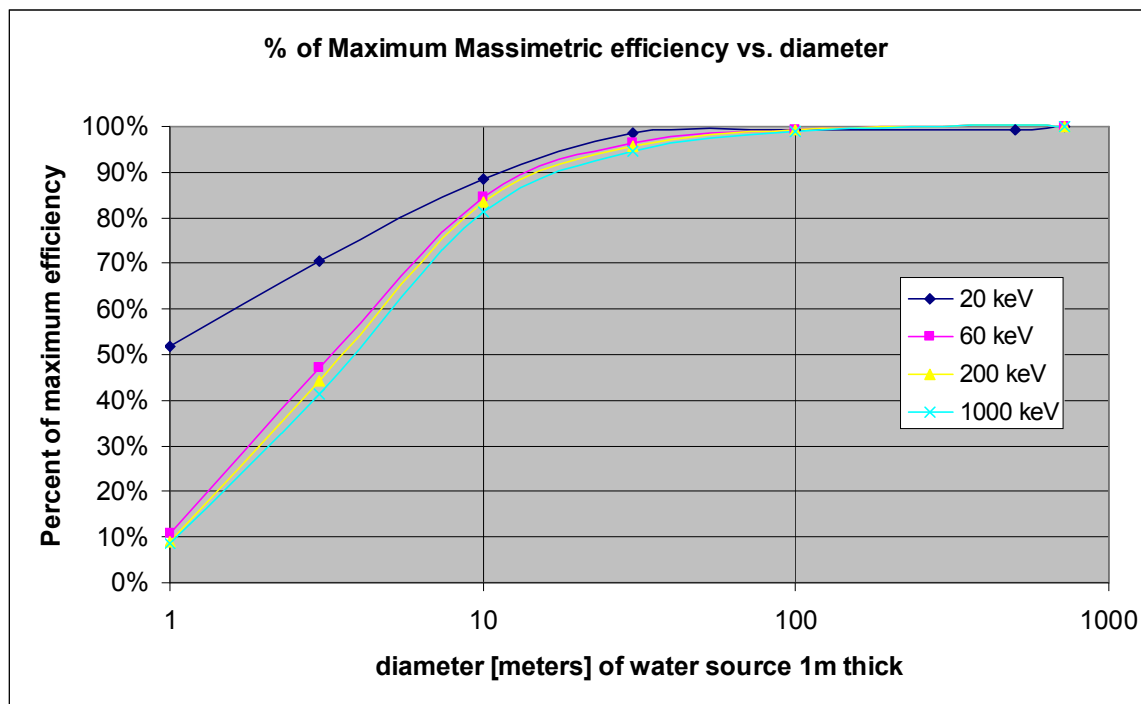
% of Maximum Massimetric efficiency vs. diameter

Figure 1 Massimetric Efficiency vs. diameter

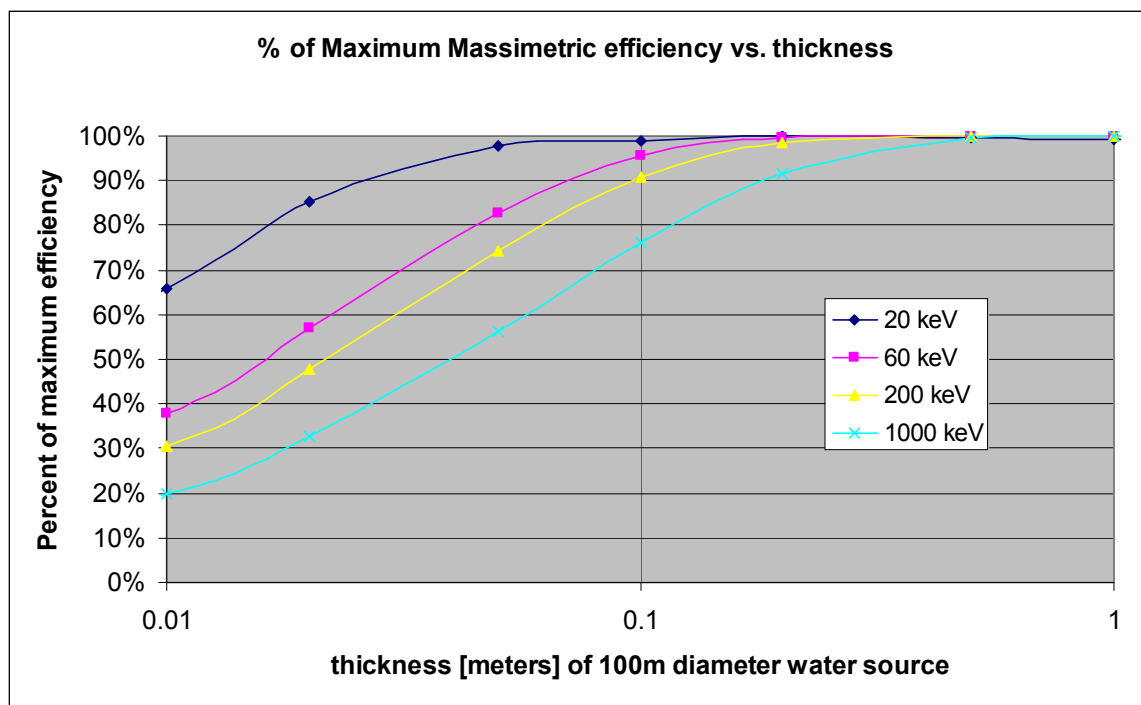**% of Maximum Massimetric efficiency vs. thickness**

Figure 2   Massimetric efficiency vs. thickness

Figure 3 shows how the Massimetric Efficiency changes as the density of the source is changed, but all other parameter are held constant. The efficiency is normalized to the maximum density of 20g/cc. The source matrix is water, the source diameter is 100m, and the thickness is 1m.

Maximum efficiency is reached at ~0.05g/cc. If 10% accuracy is acceptable, then a calibration at ~0.1g/cc density can be used for all density values greater than ~0.02g/cc and for all energies.
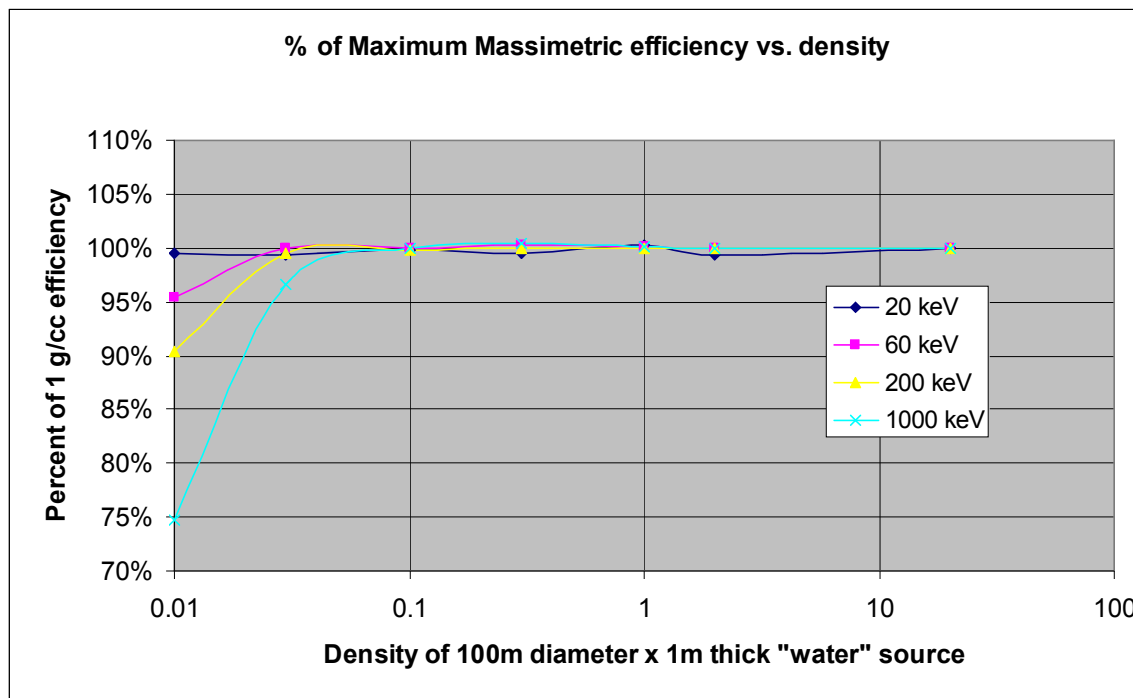

Figure 3 Massimetric efficiency vs. density

From the data presented so far, it seems to be clear that the use of Massimetric efficiency has significant advantages over the traditional efficiency when the sample diameter, thickness, and density are above some minimum values. In the measurements typical for waste assay and environmental remediation, these parameters are rarely known accurately. But as long as it is likely that they are above the minimum values stated here, then reasonably accurate measurements can be made.

But those are frequently not the only unknown parameters. In many cases the sample matrix is not well known. It is common to assay containers where the contents are variable and uncertain. Even when the matrix is visible [e.g. soil] the exact composition of that is quite variable and not likely to be known. Figure 4 shows how the efficiency varies for different elements. The atomic number of the sample was varied from Hydrogen to Uranium. The rest of the sample was held constant at 100m diameter, 1m thick, and 1 g/cc density. The individual efficiency values have been normalized to the efficiency of water.

For the case of variable sample matrix, the traditional efficiency and the Massimetric efficiency behave the same. Here, there is no special advantage of the Massimetric efficiency. But since this is a realistic measurement condition, this effect must be included also in this evaluation. It is obvious that for the 20 keV measurements that the element makes a very large difference. If water was assumed as the calibration material, then Li would over-respond by a factor of 4, Al would under-respond a factor of 4, and Fe would be factor of 30 low. Fortunately, these low energies are rarely assayed through the walls of a container or under other conditions where the

item can't be closely examined. For 60 keV (e.g. Am241) the situation is much better. All elements between He and Si are within 30% of the water calibration, and Fe is only a factor of 5 low. For 200 keV, the 30% under-response region extends up to Sn, and for 1000 keV the 30% under-response region extends all the way to Uranium. The jumps in the 20 keV relative efficiency below Sn and in the 200 keV relative efficiency below W are due to the K-edges.
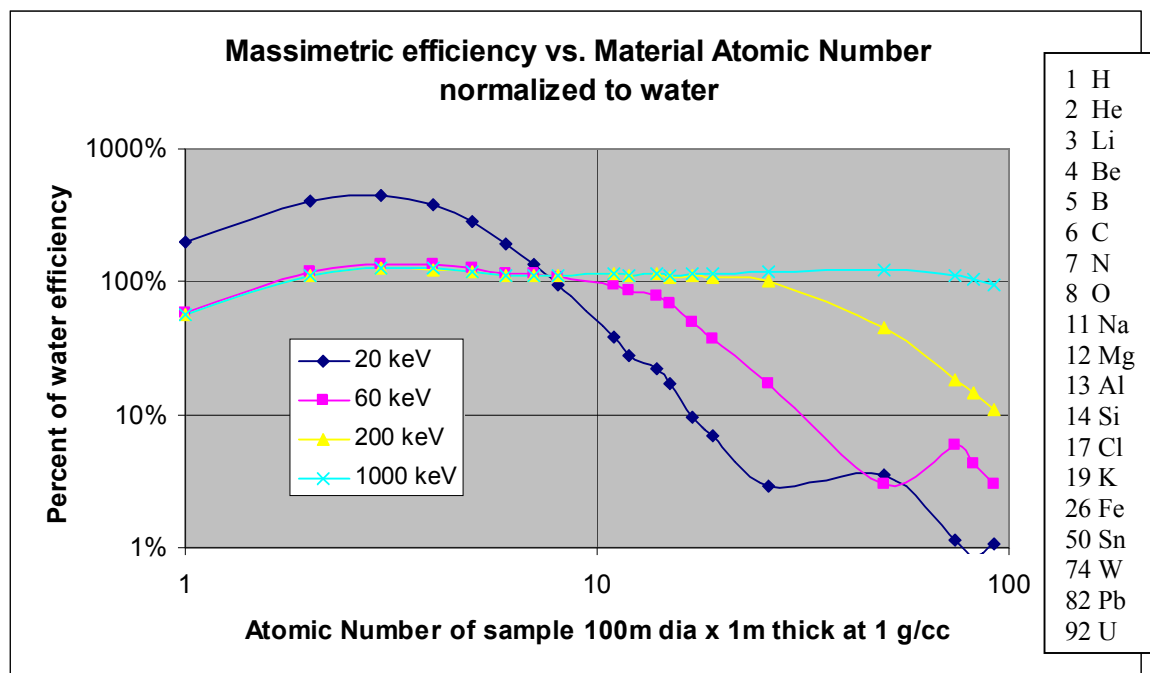

Figure 4   Massimetric efficiency vs. Atomic Number of matrix element

Figure 5 is an attempt to show how the efficiency varies for materials commonly measured. These are the same conditions are in Figure 4 - large sample at 1 g/cc. The various soil materials [drydirt, dirt1, dirt2, dirt3, dirt4] are all from the ISOCS material library, and represent different elemental compositions of soil from various literature references. The one labeled "dirt3" is highly vegetative and behaves much like cellulose; the one labeled "dirt4" is highly mineralized with more of high atomic number materials with a high photoelectric cross-section. Since the range of materials isn't as large as the elements in Figure 4, the variation is much less. For example in the 200 keV and 1000 keV case, all results are within -3% to +15% of the water calibration. And at 60 keV, all materials are within -50% to +7%. If Am241 was an important nuclide, and material was segregated into three categories [organic, inorganic except iron, and iron], then 3 separate calibrations are all that are needed to keep the material portion of the calibrations all within ±15%
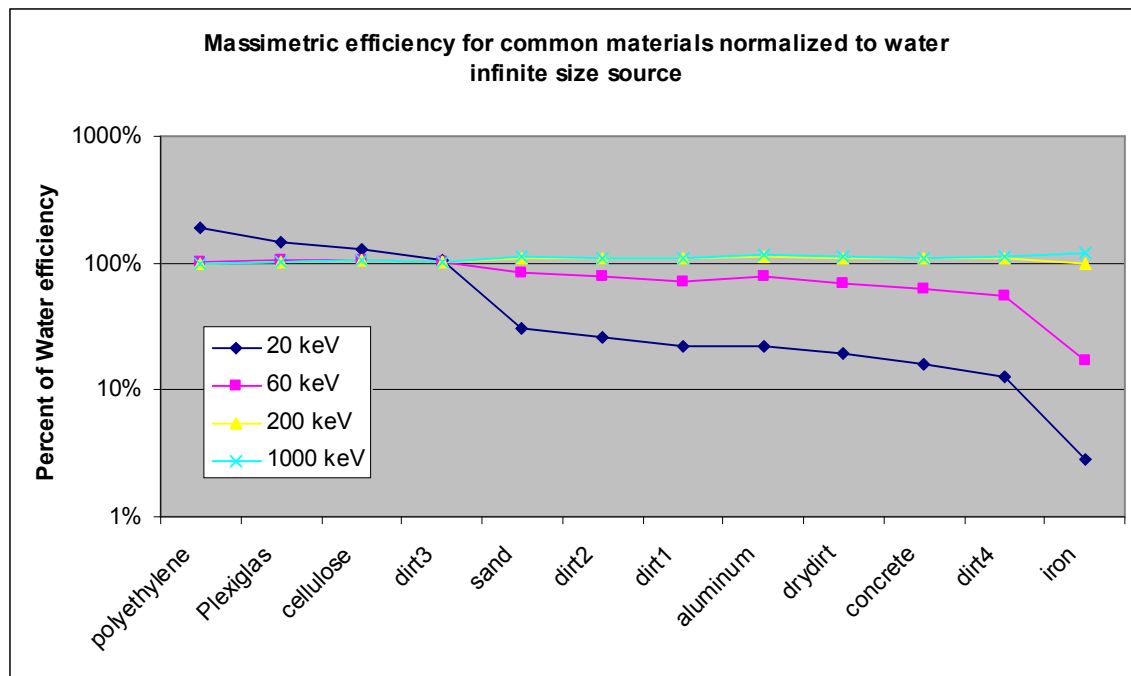
Figure 5 Massimetric efficiency variation for common materials, all at density = 1g/cc

Combining all of these items is a useful exercise to indicate the potential accuracy for a "generic" calibration for "large" items. The generic calibration for this example would be water that is 9m diameter and 30cm thick at a distance of 1 meter. If one assumes that during the field measurements, these source conditions are all unknown, and all that they are all independent variables, then the uncertainties can be combined in quadrature. If the values here are considered ranges, then a reasonable assumption is that the standard deviation is 1/3 of the range. These conditions are summarized in Table 1.

Table 1 Summary of calibration accuracy and conditions

Parameter	Condition	Efficiency Accuracy Range
Source diameter	>10m for 1m detector separation or >1m for 0.1m separation	±10%
Source thickness	> 0.2m	±10%
Source density	> 0.1 g/cc	±10%
Common materials as source matrix	~ 200 – 2000 keV energy	-3% +15%
	~ 60 – 2000 keV energy	-50% +10%

Converting these ranges to standard deviations and combining them as independent variables leads to a standard deviation of about 10% for energies in the ~200-2000 keV range. This assumes a judicious choice of reference calibration material and geometry that represents a median efficiency of the population of materials.

Another method to make this comparison was to use the new ISOCS Uncertainty Estimator (IUE) function [2]. The IUE software uses a probabilistic estimation method. An ISOCS efficiency calibration model is used as the starting parameters. This model contains all the relevant dimensions of the item necessary to accurately compute the efficiency. The IUE

software allows the user to determine which of those parameters are "not well known" (NWK). For each NWK parameter the user enters a minimum value, a maximum value and a probability distribution function (PDF). The PDF can be rectangular [all values between minimum and maximum are equally probably] or triangular [the central value the most probable decreasing linearly to zero probability at minimum and maximum] along with 3 Gaussian shapes where minimum and maximum represent the 68%, 95% and 99% confidence limits, respectively. IUE then creates a large number of efficiency calibration models, where each NWK parameter in each model is randomly chosen based upon the PDF. The efficiency at each energy is computed for each of these large number of models, either the normal or traditional efficiency or the Massimetric efficiency. Efficiency*length and Efficiency*area are also possible selections. Finally, the mean and standard deviation is computed from the large population of efficiency values for each energy.

IUE was used as an independent method to derive the same parameters as estimated in Table 1. The materials were the same ones used in Figure 5. The diameter range was 10 – 50 meters. The thickness range was 20 – 100 cm. The density range was 0.1 – 2.3 g/cc. The first scenario used the Uniform or Rectangular PDF for all NWK parameters. The second scenario assumed a 95% confidence level (CL) Gaussian distribution for the density and for the thickness, and uniform for the diameter and the matrix content.

Table 2 Comparison of efficiency methods using ISOCS IUE function

Distribution	% sd Normal efficiency				% sd Massimetric efficiency			
	20 keV	60 keV	200 keV	1000 keV	20 keV	60 keV	200 keV	1000 keV
Uniform	190	250	240	214	80	20	7	12
Gaussian	150	115	108	100	97	30	6	7

The precision in the Massimetric efficiency method is significantly better than the Normal method at all energies, and by factors of 10 or more in the important 200-1000 keV range. And the IUE results are consistent with the manually computed results from the independently derived parameters.

MASSIMETRIC EFFICIENCY FOR 200 LITER DRUMS

The previous section was for samples large enough to be assumed nearly infinite in geometric angle subtended, thickness, and density. In this next section we will investigate the uncertainty for measurements of a more constrained sample – materials inside a 200 liter drum. It is very common to assay these drums without knowing the nature of the matrix inside the drum, e.g. the material, fill height, and density. All of these affect the normal efficiency calibration, but affect the massimetric efficiency calibration to a much smaller degree.

For these measurement conditions a nominal 200 liter [55 US gallon] container was used. It had inside dimensions of 57cm diameter x 84cm high, and had a 2mm thick steel wall. The matrix in the container was filled to 76cm [90% full] for the nominal condition, unless otherwise stated. The detector was viewing the side of the drum, at about 35cm from the base of the drum – slightly below the middle of the nominal mid fill-height.

Several conditions were investigated at both 15cm and 100cm container-detector distance. The close measurements were also tried with the ISOCS 30degree collimator and with a very narrow field-of-view collimator. While the 15cm distance showed slight improvements in the Massimetric efficiency uniformity, as did the collimated measurements, the difference is so small that other factors [counting time favoring close distances, non-uniform efficiency response favoring far distances] would be the determining factor for the method to use. The following data is applicable to both distances and collimator conditions.

The first parameter investigated was the impact of material density. The matrix was water. The fill height was held constant at 76cm and the density of the "water" was varied. The efficiency values were normalized to that of water at 1 g/cc density. Figure 4 shows the Massimetric efficiency ratios on the left and the normal efficiency ratios on the right. In all cases, the Massimetric efficiency is better – ranging from factors of 2 to factors of 10 improvement.
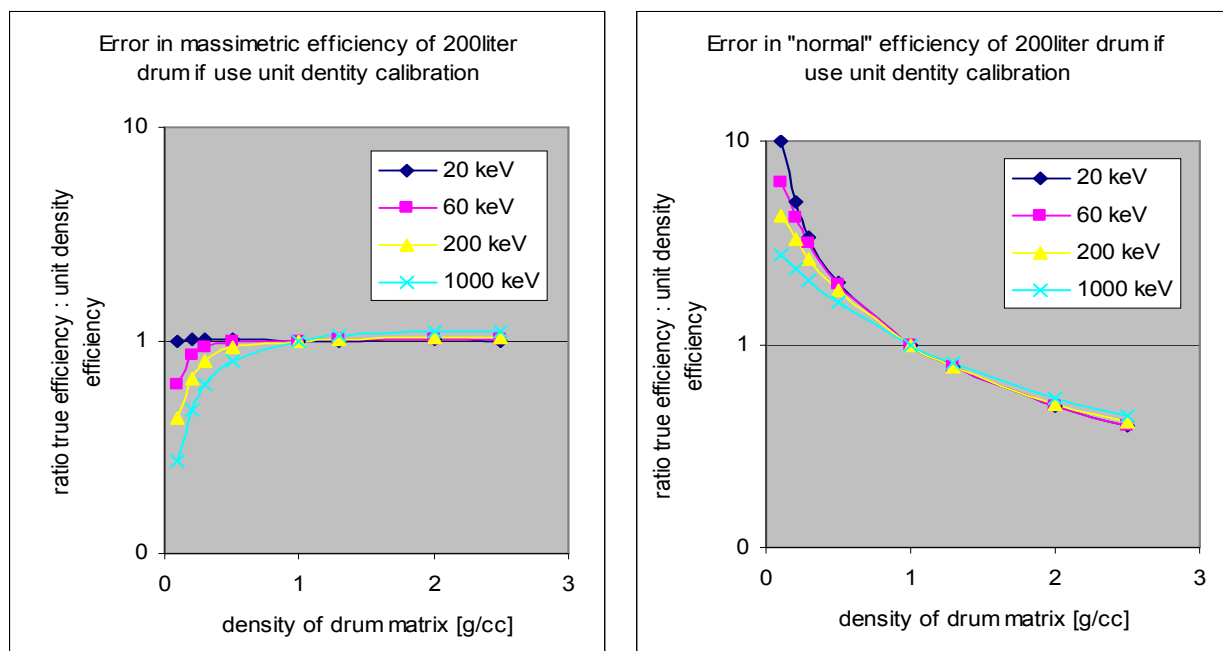

Figure 6   Massimetric efficiency [left] and traditional efficiency [right] for various container densities
The next set of tests was with the fill height varying from 45% full [15"] to 100% full. The results are shown in Figure 7. The left graph holds the density constant but varies the fill height; consequently the mass of the contents varied from 95 kg to 195 kg. The right graph holds the mass constant; consequently the density of the contents varies from 0.6 g/cc to 1.11 g/cc. The data are normalized to unit density and 61 cm [24"] fill height.

Each situation has about a ±40% maximum error. For this kind of variation there is very little difference between the Massimetric and the normal efficiency.
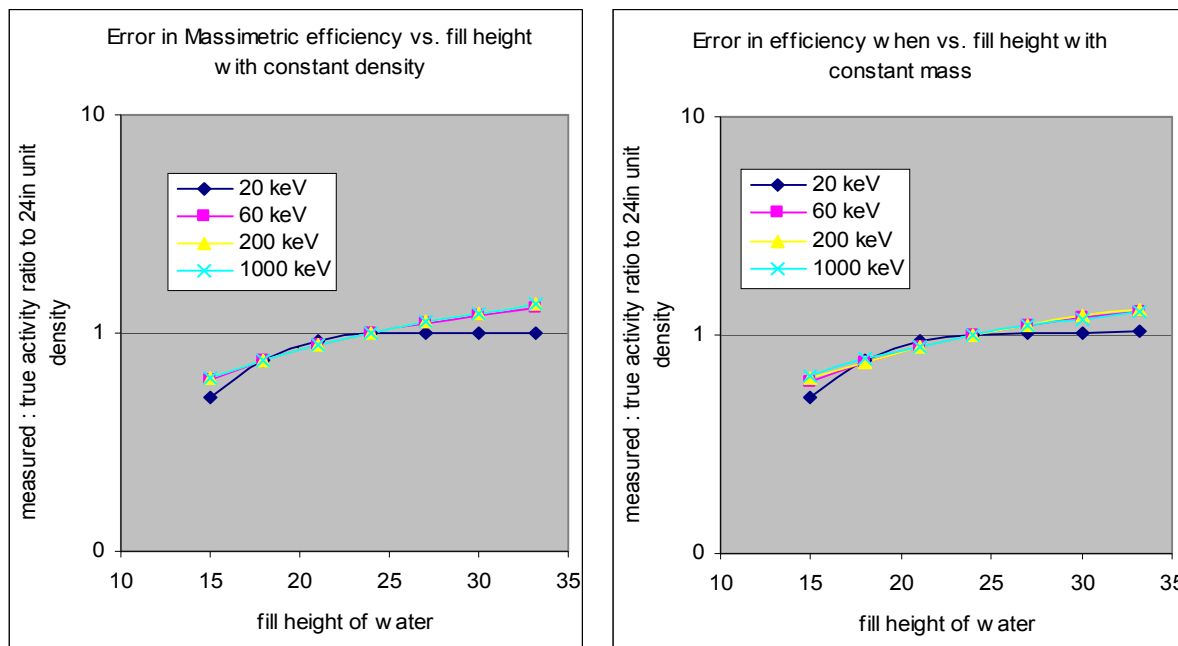
Figure 7   Massimetric efficiency for varying fill heights with constant density [left] and constant drum mass [right]

The above conditions were for homogeneous density throughout the matrix. But in normal containers, especially those that have undergone much handling, the heavy items sink to the bottom, making the bottom of the container more dense than the top. A series of calculations was done to evaluate the impact of this on the accuracy of the efficiency calibration. The container was divided into 4 layers of equal height. The density of each layer was adjusted so that the topmost layer had the lowest density and the bottom layer had the highest density. The total weight of the container was kept the same. The matrix was water. Each efficiency was normalized to the efficiency for uniform concentration. Table 3 shows the results. In this case the efficiency error from assuming a uniform concentration is not very large. Even in the most extreme condition, the maximum error was only 16%. For this situation there is little difference between the normal and the Massimetric efficiency.

Table 3   Efficiency error when matrix vertical density is not uniform

Layer density – g/cc [from top to bottom]	20 keV	60 keV	200 keV	1000 keV
1.0 1.0 1.0 1.0	1.00	1.00	1.00	1.00
0.7 0.9 1.1 1.3	1.01	1.00	0.99	0.99
0.4 0.8 1.2 1.6	1.01	0.98	0.97	0.94
0.1 0.7 1.3 1.9	1.01	0.89	0.86	0.84

For an evaluation of how all these variations apply to a drum assay campaign of unknown, but realistic, contents, the following conditions in Table 4 were examined. Eight different drum contents were used. Each matrix had 2 different density possibilities, which were chosen to represent the limits that might be encountered. Each combination was also examined at fill heights of 24" and 30", representing 72% and 90% of capacity. Efficiencies were computed at the 4 energies for these 28 conditions. The efficiency values were normalized to that of water.

Table 4   Efficiency evaluation conditions for drum assay campaign with unknown contents

Matrix	Density [g/cc]		Fill Height [in]	
cellulose	0.2	0.6	30	24
aluminum	0.3	1.0	30	24
iron	0.5	1.5	30	24
drydirt	1.0	1.6	30	24
concrete	1.4	2.3	30	24
sand	1.4	2.0	30	24
oil	0.6	1.0	30	24
water	1		27	

The results are presented in Figure 7 for the normal or traditional efficiency mode, and in Figure 8 for the Massimetric efficiency method.
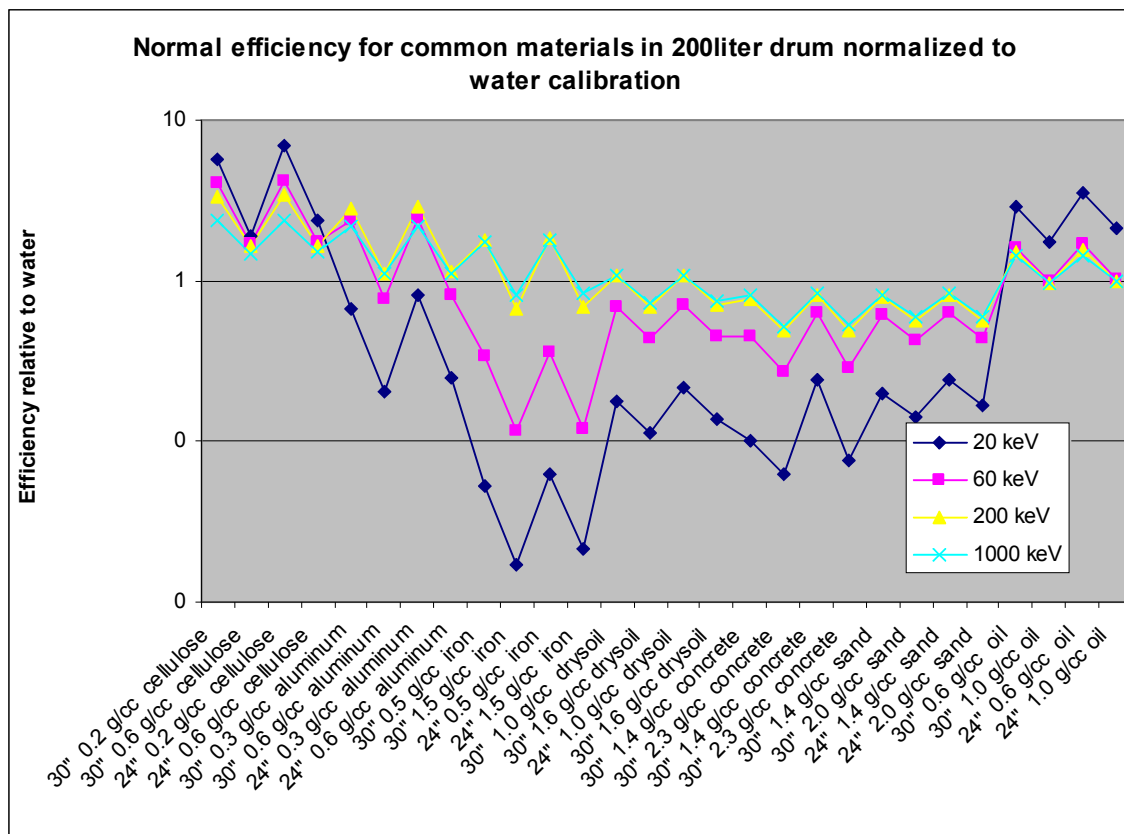

Figure 8   Normal efficiency for common assay conditions, relative to water calibration
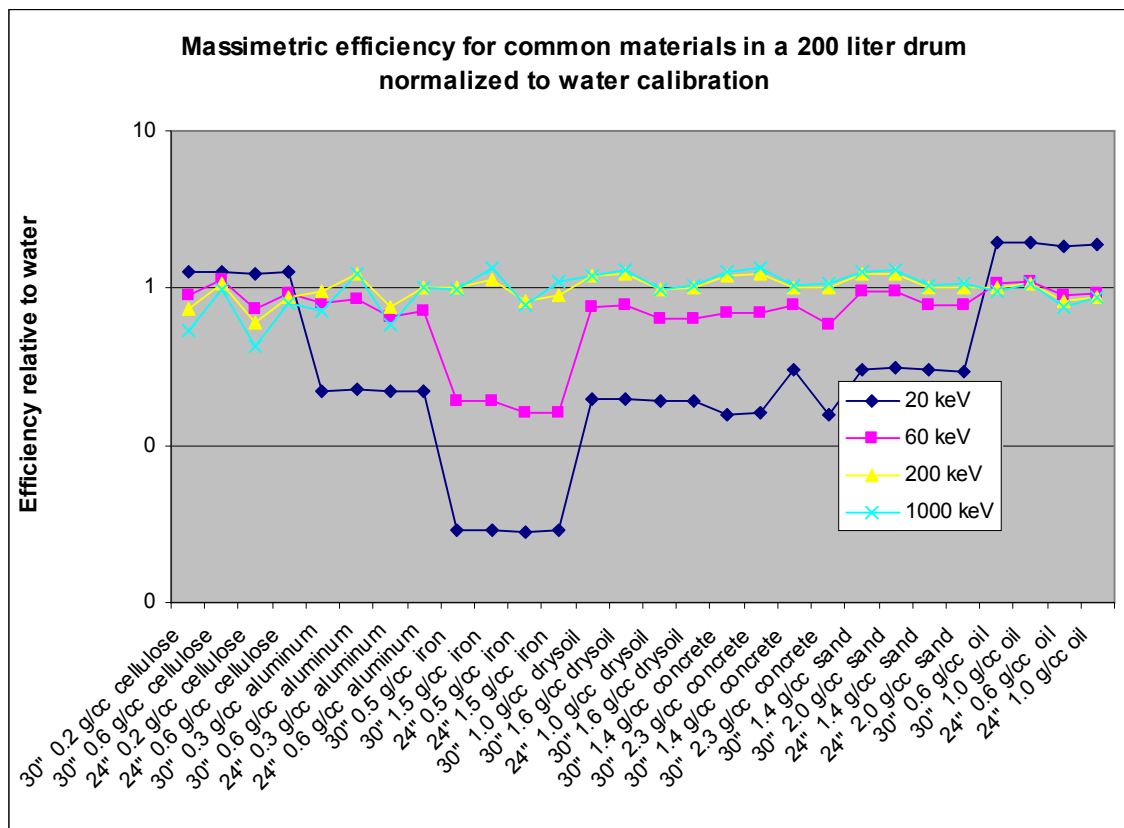
Figure 9   Massimetric efficiency for common assay conditions, relative to water calibration

Table 5 shows a comparison of the two efficiency calibration methods. Using the Massimetric efficiency has greatly improved the ability of a single calibration to be used for all of these different matrices and densities and fill heights. The mean of all the drums is much closer to the expected value of 1 for the 60 – 1000 keV values. The standard deviation for the Massimetric efficiency is around 25% as compared to 60% to over 100% for the normal method.

Table 5   Comparison of Massimetric and Normal efficiency for unknown drum campaign

	Massimetric Efficiency				Normal Efficiency			
	20 keV	60 keV	200 keV	1000 keV	20 keV	60 keV	200 keV	1000 keV
mean	0.60	0.74	1.01	1.00	1.11	1.08	1.30	1.19
std dev'n	0.66	0.27	0.17	0.24	1.74	1.05	0.84	0.56
% rel std deviation	**110**	**36**	**17**	**24**	**157**	**97**	**64**	**47**
maximum	1.94	1.12	1.25	1.36	6.89	4.16	3.38	2.40
minimum	0.03	0.16	0.60	0.43	0.02	0.12	0.48	0.52
max w/o Fe	1.94	1.12	1.25	1.33	6.89	4.16	3.38	2.40
min w/o Fe	0.15	0.58	0.60	0.43	0.06	0.27	0.48	0.52

The ISOCS IUE feature was used again to provide a different approach to compare the two methods of efficiency calibration. For these IUE calculations, the parameters in Table 4 were

used to construct a range of possible configurations. All the materials were given an equal probability of occurrence. In IUE the density is an independent variable, not one dependent upon the matrix as in the Table 5 calculations. So the entire range from 0.2 to 2.3 g/cc was used. The same is true for the fill height where 24" and 30" were used as min and max values. In the first run, both the density and fill height were assigned uniform probability for all values between the limits. This is the most conservative choice. In the second run, the density values were used as 99% CL limits, and the fill height values were used as 95% CL limits – a somewhat more realistic situation. Table 6 shows those results. The IUE rectangular distribution agrees quite well with the individual calculations from Table 5. The Gaussian distributions are lower and probably more realistic. But both methods demonstrate that the Massimetric method can reduce the standard deviation (std dev, sd) to ~40% of the value of the normal or traditional efficiency method, for the 60, 200, and 1000 keV energies.

Table 6   Comparison of Standard Deviations to IUE calculations

	Massimetric Efficiency %sd				Normal Efficiency %sd			
	20 keV	60 keV	200 keV	1000 keV	20 keV	60 keV	200 keV	1000 keV
IUE – rectangular distributions	102	36	11	18	144	83	63	48
IUE – Gaussian distributions	102	35	7.5	10	109	50	35	28
St dev from Table 5	110	36	17	24	157	97	64	47

CONCLUSION

The use of the Massimetric efficiency calibration has been shown to allow a single calibration to be used for a wide range of item assay conditions and also have lower uncertainty than the traditional efficiency calibration. The Massimetric efficiency calibration allows sample concentration [Bq/g] to be determined without weighing the item. Massimetric efficiency calibration was always more precise for this wide range of conditions evaluated here, except for one case where it was only slightly worse. For very large "infinite" size objects, the uncertainty for Massimetric efficiency is of the order of 10% for a wide range of measurement conditions. For 200 liter drums the uncertainty increases to 20% for the conditions studied. This is nearly a factor of 3 improvement over the traditional efficiency calibration method. The creating of Massimetric efficiency is no more difficult than the creation of normal efficiency calibrations, and is a standard feature of the ISOCS mathematical efficiency calibration software. The estimation of the standard deviation of the concentration [Bq/g] from a population of items covering a wide range of compositions can be easily determined with the IUE feature of the ISOCS software.

REFERENCES

1. F. Bronson, *ISOCS, a laboratory quality Ge gamma spectroscopy system that you can take to the source for immediate high quality results*, Proceedings of the Rapid Radioactivity

Measurements in Emergency and Routine Situations Conference, London UK, 15-17 October 1997

2. F. Bronson, V. Atrashkevich, G. Geurkov, B. Young, *Probabilistic uncertainty estimator for gamma-spectroscopy measurements*, Journal of Radioanalytical and Nuclear Chemistry, Vol. 276, No. 3, (2008) 589-594