ASSAY ERRORS INTRODUCED BY MATRIX CORRECTION USING TRANSMISSIONS DETERMINED BY THE MATERIAL BASIS SET METHOD

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ABSTRACT

Nondestructive assay instruments such as segmented gamma scanners and tomographic gamma scanners generally use measured transmission values to calculate correction factors for attenuation effects. Current practice is to use one or more transmission sources with peak energies close to the assay energy and to use interpolation techniques to obtain transmissions at each assay energy. This practice limits the source radioisotopes that are useful for each assay radioisotope and can lead to using isotopes that are expensive or have half-lives so short as to require frequent replacement. Use of the Material Basis Set (MBS) approach to perform the transmission interpolation or extrapolation provides more flexibility in choice of transmission sources and provides transmission values over a wider range of assay energies. A previous computational study by the authors [1] quantified assay errors introduced by the MBS method for four radioactive sources and eight diverse matrix types assuming exact values of measured transmissions. The present study includes the effect of statistical uncertainties in measured transmissions. To determine the effect of these uncertainties on the MBS-determined values, measured transmission uncertainties were corrected for intensity and attenuation to estimate measured transmission uncertainties for the different assay scenarios. Using these estimated uncertainties, the assay bias and random error introduced by the MBS method plus transmission measurement uncertainties was determined. Particular attention was given to extrapolation to low transmission values at low gamma ray energy (60 keV), where the energy variation is the steepest.

INTRODUCTION

The measurement of the quantity of one or more radioisotopes in a container by detection of the emitted gamma rays generally requires that a correction be made for attenuation of the gamma rays by the material containing the radioisotopes. The correction can be calculated directly if the composition, density, and uniformity of the material are known along with the activity distribution. Most often this is not the case, and the correction is made based on measured transmissions of gamma rays through the container. The usual method is to use a separate "transmission" gamma ray source that emits gamma rays at one or more energies near the assay energy. Transmissions through the container are measured at the transmission source energies, and then these values are interpolated or extrapolated to the assay energy or energies using exponential, linear, or quadratic functions or polynomials in logarithm-logarithm space. Sources with energies most appropriate for assay of some important radioisotopes, such as ^{235}U and ^{239}Pu, have relatively short half lives and require frequent replacement. Also, a single transmission source used with traditional interpolation and extrapolation methods is not well suited for attenuation correction over a wide range of energies, as needed for an assay instrument that quantifies many radioisotopes with each assay sequence. Use of the MBS method determines the transmission through a container over a wide range of energies based on measured transmissions at only a few strong, disparate energies. Also, this method can be applied using transmission sources having multi-year half-lives and needing replacement at no less than 7-year intervals. The calculation-based study presented here quantifies the assay bias and random errors for each assay scenario due to attenuation-correction using the MBS and provides guidance for its use.

SUMMARY
This study is an extension of a previous one [1] by the authors to simulate the effect of random errors in measured transmissions on MBS-derived assay results. This study used only limited experimental data for guidance and is based almost entirely on computations. Four gamma ray sources, eight assay materials, and seven radioisotopes had been chosen for the initial study and are treated in the current study. Their description will be repeated here so this paper is self-sufficient. The gamma ray energies most useful for transmission measurements were chosen from those emitted by each of the four sources. For each assay material, the gamma ray transmission through the material and the associated detection efficiency at each assay energy were calculated. Also, the transmission at each of the chosen transmission source energies was calculated. Uncertainties at the 1-σ level were estimated for the source transmissions by adjustment of empirical data. Pseudo-normally distributed source transmissions were generated using each calculated source transmission as the mean value and the estimated uncertainty as the standard deviation. For each assay scenario, forty thousand calculations using the (pseudo-) normally distributed transmissions as input were performed. These calculation results and the directly-calculated efficiencies were used to determine the assay bias and random error introduced by attenuation correction using the MBS Method for each assay energy for that scenario. The assay bias and random errors shown in this report are calculated errors in *assay results*. The assay bias and random error using the MBS method (as configured for this study) is summarized for a range of assay energies (seven assay radioisotopes) for each of the eight assay materials for each of the four transmission sources.

MBS METHOD
The uncollided gamma ray transmission, $T(E)$, at energy, E, through a thickness, Δx, of material having a mass attenuation coefficient $\mu_m(E)$ and density ρ is given by:

$$T(E) = e^{-\mu_m(E)\cdot\rho\cdot\Delta x} \quad \text{which can be rewritten as} \quad \mu_m(E)\cdot\rho\cdot\Delta x = -\ln(T(E)). \qquad (1)$$

If the energy dependence of the transmission of a homogeneous sample is approximated by a linear combination of the mass attenuation coefficients of a low-Z and a high-Z element, and, if needed, a mid-Z element (where Z is the atomic number), Equation 1 can be written as:

$$[C_1'\cdot\ \mu_L(E) + C_2'\cdot\ \mu_M(E) + C_3'\cdot\ \mu_H(E)]\ \cdot\rho\cdot\Delta x = -\ln(T(E)) \qquad (2)$$

where μ_L, μ_M, and μ_H are the mass attenuation coefficients of low-Z, mid-Z, and high-Z elements, respectively, and are known functions of gamma ray energy [2]. In representing the energy dependence of the mass attenuation coefficient of the material assayed μ_A as an expansion in terms of the known energy dependent attenuation coefficients of two or three other materials, a complex energy dependence is captured in terms of only a few coefficients $\{C_i'\}$. The energy dependent mass attenuation coefficients are thus used to define a physical or materials-derived algorithm for transmission interpolation and extrapolation instead of using a fit to a linear or quadratic function. Indeed, rather than elements, any convenient compounds or materials could be chosen depending on the nature of the problem.

A method for determining the coefficients in equation (2) above has been reported [1] and is not repeated here. If the mid-Z term is omitted ($C_2 \equiv 0$), the MBS approach defines a 2 x 2 system of simultaneous equations. Otherwise, it is a 3 x 3 system. The resulting system can be solved by a determinant or matrix method. Once the $\{C_i\}$ are determined, transmissions T_j at other energies E_j are calculated using the Equation (3).

$$T(E_j) = \exp[C_1\cdot\ \mu_L(E_j) + C_2\cdot\ \mu_M(E_j) + C_3\cdot\ \mu_H(E_j)] \qquad (3)$$

The mass attenuation coefficients can be represented as polynomials in $ln(E)$-$ln(\mu)$ space. For the elements whose mass attenuation coefficients have a K-edge in the energy range of interest, the edge can be handled by defining one set of coefficients for points less than the energy of the discontinuity and another set for higher energies. The attenuation coefficients of carbon and tin, for example, can be used as the basis elements for many assay materials of practical interest at energies greater than about 120 keV. At energies below about 120 keV, care must be taken in the selection of the basis elements and measured transmission energies. This is because of the rapid increase in attenuation as the energy decreases below this energy, and also because of the potentially confounding effects of the K-edges of elements in the assay material or used as basis elements in the MBS algorithm. An organic compound (e.g., cellulose) containing H_2 may also be used for known low-Z matrices.

The number of measured transmission values must always exceed the number of basis elements. If the number of measured transmissions equals the number of basis elements (mathematically, three equations and three unknowns with, at most, one solution), the MBS equations will always have a solution as long as the measured transmissions have no error. However, statistical errors in real world transmission measurement can cause MBS equations to have no solution. The method fails. Having one or two more measured transmission values than basis elements will make it unlikely that the MBS calculation will fail. Despite this, failures can occasionally occur.

CALCULATIONS

Calculations determined assay bias and random errors due to the MBS method for the thirty-two transmission source-assay matrix scenarios described below. Each assay error was determined as follows: From the defined properties of each matrix, the transmission and counting efficiency ε_D at each assay energy were calculated; these directly calculated efficiencies were taken to be the reference values for the assay computations. The transmissions of source gamma rays were also calculated. In the previous study, these calculated transmissions were input to MBS calculations to determine interpolated or extrapolated assay transmission values, these transmission values were used to determine the counting efficiencies, and these values were then compared to the direct calculations to determine the error introduced by the MBS method. In the present study, the source transmissions were used as the mean values of pseudo-normally distributions of transmission values to mimic the behavior of measured transmissions in an actual assay instrument. The standard deviations used for the distributions were estimated based on adjustment of emperical data for source line intensity and the transmission value. We used experience from measuring a large number of real waste drums on a Q^2 instrument as guidance as to a reasonable amount of scatter to include. Each T-line was treated as independent. For each of the thirty-two scenarios, 40,000 efficiency calculations were performed using the (pseudo-) normally distributed transmission values as input to MBS calculation of assay transmissions that were then used to calculate counting efficiencies. The resulting efficiencies were collected in histrograms, from which the mean, mode (most frequent value), and standard deviation of the efficiency population for each assay energy of that scenario were determined. The mode was taken to be the assay efficiency ε_{MBS} to be compared to the reference (directly calculated) value. The radioisotope quantity resulting from an assay is proportional to the inverse of the efficiency, so that the assay bias introduced by the MBS method is:

$$assay\ error\ (\%) = 100 \cdot [(1 / \varepsilon_{MBS}) / (1 / \varepsilon_D) - 1] \ . \qquad (4)$$

The standard deviation of the results provides an estimate of the random error introduced by the normal distributions (from counting statistics) of the measured source transmission values and the effect of these on the MBS and efficiency calculations.

Efficiency calculations were performed using a point kernel approach assuming homogeneous assay material and with the attenuation coefficient of the material derived from the container diameter and specified transmission. Two container sizes were defined: a 5-gal pail and a 55-gal drum (where 1 US gallon equals approximately 3.7854 liters). For each material type, the container size that resulted in the directly calculated 129 keV transmission closer to 0.001 was selected. The 55-gal drum was selected as the container only for Job-Control Waste. The geometry defined for each of the containers was that for assay of a typical SGS segment. For the 5-gal pail, a 2-inch-high segment of an 11-inch diameter pail with the pail wall 6 inches from the detector was modeled. For the 55-gal drum, a 4-inch-high segment of a 22-inch diameter drum with the drum wall 12 inches from the detector was modeled. The calculations applied only to the assay material. The container walls were not a factor in the study.

The gamma ray transmission sources selected for the study and their half lives are as follows: ^{125}Sb, 2.76 years; ^{133}Ba, 10.52 years, ^{152}Eu, 13.54 years, and ^{166m}Ho, 1200 years. Gamma energies suitable for transmission measurements were selected based on the following criteria: among highest intensity gamma ray energies emitted by the source; energies above (about) 150 keV to avoid interference from x-ray lines and also to more likely have transmission values high enough for routine measurement; a low-energy line in the neighborhood of 150-200 keV; a mid-energy line in the neighborhood of 300-400 keV; and a high energy line in the vicinity of 750-1000 keV. Some transmission gamma energies lower than specified by the criteria were included to test their effects. A total of 22 transmission gamma ray energies were chosen for the study. For each transmission source, the available energies and their suitability to the MBS method are:

^{125}Sb: 177.3, 427.9, and 636.0 keV. This, as it turns out, is a near-ideal energy set for use with the MBS method pegging as it does both the low and high energy dependence.

^{133}Ba: 81.0, 160.6, 276.4, 356.0, and 383.8 keV. This set is deficient at higher energies for the range of assay nuclides included in the study.

^{152}Eu: 121.8, 244.7, 344.3, and 778.9 keV. This is a good set if the transmission at 121.8 keV can be measured. If not, the lower energy lines are closer than ideal. Also, ^{152}Eu has high-energy gamma rays that are difficult to shield and which also contribute continuum to the spectrum.

^{166m}Ho: 80.6, 184.4, 215.9, 280.6, 300.8, 452.3, 529.8, 711.7, 752.3, 951.0 keV. This is a near-ideal set except for the difficulty in shielding high-energy source gamma rays and potential interference with the 184.4 keV line if ^{235}U is present in the material assayed.

Eight diverse "real-world" assay materials were defined for the study. Mixture fractions are in terms of weight. These materials were all defined to be homogeneous and uniform. The assay materials and densities are as follows:

Job-Control Waste:	Paper (45%) / Plastic (45%) / Aluminum (10%), density = 0.5 g/cc
Building Rubble:	Concrete (95%) / Steel (5%) with 50% void space, density = 1.3 g/cc
Dirt:	density = 1.7 g/cc
Sand:	density = 1.7 g/cc
Process Scrap:	PuO_2 (2.25%) / CaF_2 (50%) / CaO (47.75%), density = 0.7 g/cc
Incinerator Ash:	UO_2 (2.25%) / Ash (97.75%), density = 0.8 g/cc

Raschig Rings:	Borosilicate glass with 0.1% Pu, density = 0.8 g/cc
Lead Gloves:	density = 0.38 g/cc

The assay isotopes and associated assay energies selected for the study are primarily of importance to DOE facilities, but the assay energies also nicely span the range 60-1000 keV where the assay energies of most radioisotopes of measurement interest are found.

^{241}Am	59.6 keV	^{238}Pu	152.7 and 766.4 keV
^{235}U	143.8 and 185.7 keV	^{239}Pu	129.3 and 413.7 keV
^{238}U	1001 keV	^{137}Cs	662 keV
^{237}Np	212.3 and 311.9 keV		

Mass attenuation values were obtained from the XCOM database [2]. The database is accessed via the internet, and it includes gamma ray (photon) attenuation coefficients for elements, compounds, and mixtures at energies from a standard grid or those requested by a user. The "Total Attenuation without Coherent Scattering" values were used since, to first order, coherent scattering does not remove primary photons from the beam. All of the MBS calculations were initially done with carbon as the low-Z and tin as the high-Z basis element, and for many scenarios those were also the final choices. Transmission source energies were chosen that met the criteria given above to the extent possible. All calculations for a given scenario used the same basis elements and transmission lines. MBS-calculated transmissions at energies of 129 keV and higher were not very sensitive to the energies and basis elements used and were accurately fit with any reasonable set of parameters. Efforts were made to improve the results for the 60 keV assay energy, but the results of the improvement attempts were mixed. The final results shown below are based primarily on "generic" sets of source energies and choices of basis elements appropriate for the assay material. This is the way the MBS method would be applied in routine assays. Prior knowledge of a particular matrix may permit a more appropriate choice of basis materials to be made on a case by case analysis.

RESULTS

Most histograms resulting from the sets of calculations have the shape of a normal distribution. Most of the histograms for the 60 keV cases are skewed, some strongly so. Figure 1 shows sample histograms for different skewness coefficients J = 1 – Mode / Mean. For a symmetric histogram, the mode equals the mean, and J = 0. Histograms for 144, 186, 212, 313, and 662 keV assay energies are not shown because they fall within the curves shown. The non-symmetry of some histograms is correlated to extrapolated transmissions that are extremely low, less than 10^{-12} generally. Cases with such extremely low transmissions are not physically valid. An idea of the symmetries of the histograms is given in the following: For the 60 keV assay peak, skewness values

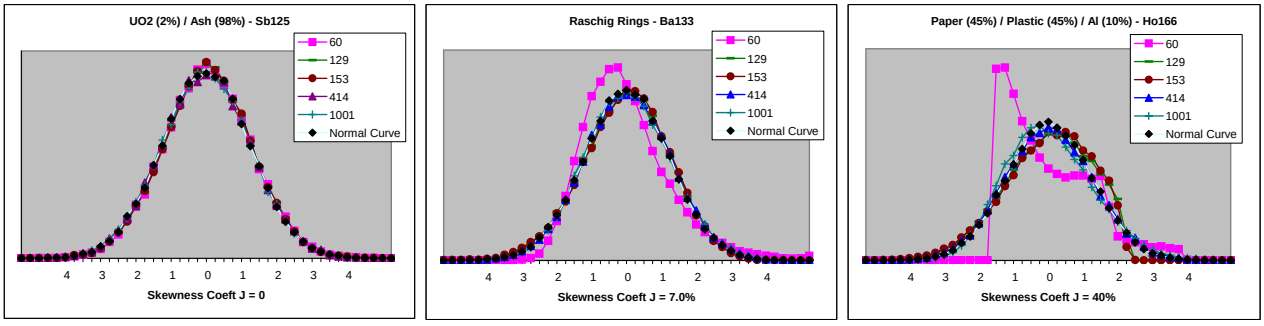

Figure 1. Histograms for three scenarios, showing symmetric (J = 0), mildly skewed (J = 7%), and strongly skewed (J = 40%) histograms.

in the range J = 20 to 40% occurred 7 times; in the range J = 10 to 20%, 4 times; in the range J = 2 to 10%, 7 times, and J ≤ 2%, 14 times. For all other peak results, skewness values in the range J = 0.6 to 1.1% occurred 7 times, and and J < 0.6%, 313 times. Cases using ^{166}Ho transmission energies had the highest skewness magnidudes, and cases using ^{125}Sb had the lowest. The information that the random errors are normally distributed means that usual error propagation techniques can be used with MBS analysis.

Twenty-two of the 32 scenarios did not have any MBS calculations resulting in no solution. The ten scenarios that did have solution failures used two basis elements and three transmission energies. Five ^{166m}Ho scenarios showed the lowest (0.007%) and highest (11.6%) solution failure rates. Three ^{152}Eu scenarios had failure rates of 0.29% to 5%. Two ^{133}Ba scenarios had failure rates of 0.007% and 0.04%. The other scenarios, including all ^{125}Sb scenarios, had no solution failures in 45,000 calculations each. Among the 5-gal Pail scenarios that had solution failures, calculated MBS solutions gave transmission values of 10^{-12} to 10^{-25}, lower than the directly calculated values. These unreasonably low transmission values seem to be a consequence of the less-than ideal set of gamma ray energies from all sources except ^{125}Sb. The minimum transmissions for the Job Control Waste Scenarios (55-gal Drum) were in the 10^{-6} to 10^{-11} range, not in a measurable range.

Assay bias results for the four sources and 11 assay energies for assay of the Dirt material type are displayed in Figure 2. This graph shows that the different transmission source energy sets lead to somewhat different assay biases. MBS-method-introduced biases at 129 keV and higher energies are much less than at 60 keV, and are generally less than 1%. This is true for the other materials included in the study, as well.

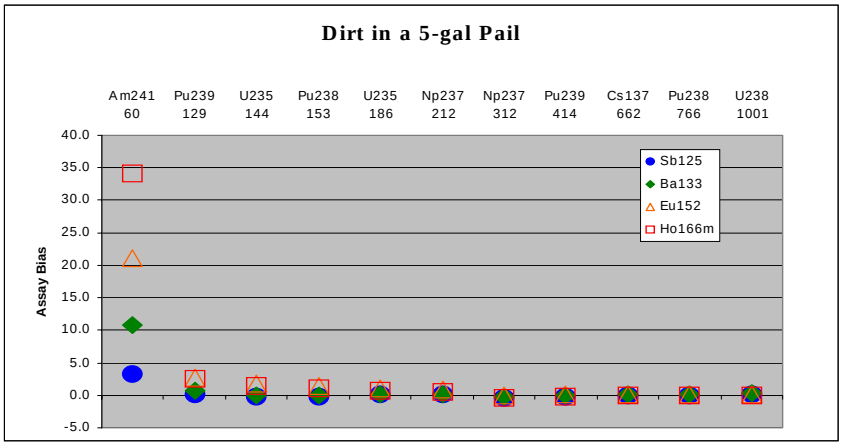

Figure 2. MBS-method assay biases for four transmission sources and for eleven assay energies.

A summary of the study results is given in Tables 1 through 4, with each table providing results for one transmission source. Each table shows the source gamma ray energies that were used, the basis elements used for each of the eight assay materials, and a summary of the MBS-method assay result biases and random errors. Note that the 60 keV results are generally the worst case. The column titled “Max Abs Others” is the maximum absolute value of the assay biases for all nine assay energies greater than 129 keV. This quantity bounds the magnitude of the biases for the highest nine assay energies. These bounds are generally less than 1% and are not higher than 2.2%. The

random errors depend on the standard deviations assigned to the source transmissions. These assigned values were estimates based on adjustments to emperical

Transmission Source: ^{125}Sb **Source Energies (keV): 176, 428, 636**

Material Assayed	Basis	Elements	MBS Bias (%) at Assay Energy			Random Error (Standard Deviation, %)				
			60 keV	129 keV	Max Abs Others	60 keV	129 keV	153 keV	186 keV	Max Others
Job-Control Waste	C	Sn	0.8	-0.1	0.2	11.4	1.4	0.9	0.5	0.3
Rubble	C	Sn	-1.2	-0.2	0.3	7.2	1.5	0.9	0.5	0.4
Dirt	C	Sn	3.2	0.2	0.3	7.5	1.3	0.8	0.5	0.3
Sand	C	Sn	-11.1	-1.0	0.6	11.9	1.7	1.0	0.6	0.4
Uranium Oxide and Ash	C	U	-31.1	-0.5	0.3	1.1	0.8	0.5	0.3	0.2
Plutonium Scrap	C	Pu	-37.0	-1.3	0.6	1.2	0.8	0.6	0.4	0.3
Raschig Rings	C	Sn	3.5	-0.2	0.2	13.3	2.1	1.3	0.8	0.5
Lead-Lined Gloves	C	Pb	2.1	1.5	0.4	1.6	1.5	1.3	1.1	0.9

Table 1. Summary table of assay biases and random errors for ^{125}Sb scenarios.

Transmission Source: ^{133}Ba **Source Energies (keV): 161, 276, 384**
Lead-Lined Gloves only: 276, 356, 384

Material Assayed	Basis	Elements	MBS Bias (%) at Assay Energy			Random Error (Standard Deviation, %)				
			60 keV	129 keV	Max Abs Others	60 keV	129 keV	153 keV	186 keV	Max Others
Job-Control Waste	C	Sn	4.6	-0.2	0.2	22.9	2.5	1.5	0.8	0.5
Rubble	C	Sn	-0.3	-0.1	0.4	14.2	2.7	1.7	0.9	0.6
Dirt	C	Sn	10.8	0.6	0.2	24.7	4.4	2.7	1.4	0.9
Sand	C	Sn	-5.3	-1.1	0.6	30.8	4.2	2.5	1.3	0.8
Uranium Oxide and Ash	C	U	-31.4	-1.3	0.7	3.8	2.5	1.7	0.9	0.6
Plutonium Scrap	C	Pu	-36.7	-1.1	0.5	1.0	0.7	0.4	0.3	0.2
Raschig Rings	C	Sn	7.6	0.2	0.3	17.8	2.8	1.7	0.9	0.5
Lead-Lined Gloves	C	Pb	1.7	1.1	0.6	2.1	1.9	1.6	1.3	1.0

Table 2. Summary table of assay biases and random errors for ^{133}Ba scenarios.

Transmission Source: ^{152}Eu **Source Energies (keV): 245, 344, 779**
Plutonium Scrap only: Added 122 keV

Material Assayed	Basis	Elements	MVS Bias (%) at Gamma Energy			Random Error (Standard Deviation, %)				
			60 keV	129 keV	Max Abs Others	60 keV	129 keV	153 keV	186 keV	Max Others
Job-Control Waste	C	Sn	12.6	-0.2	0.2	27.4	4.1	2.5	1.4	0.9
Rubble	C	Sn	16.5	3.0	2.2	14.2	3.3	2.1	1.2	0.8
Dirt	C	Sn	21.1	2.8	1.7	20.3	4.1	2.6	1.4	0.9
Sand	C	Sn	-7.5	-2.7	1.8	36.6	6.2	3.8	2.1	1.4
Uranium Oxide and Ash	C	U	-31.9	-1.2	0.7	3.3	2.2	1.5	0.9	0.6
Plutonium Scrap	C	Pu Sn	-10.1	-1.4	1.0	1.5	2.1	1.5	0.9	0.6
Raschig Rings	C	Sn	23.2	1.3	1.0	29.4	5.6	3.5	1.9	1.2
Lead-Lined Gloves	C	Pb	1.9	1.3	0.4	1.4	1.3	1.1	0.9	0.7

Table 3. Summary table of assay biases and random errors for ^{152}Eu scenarios.

Transmission Source: ^{166m}Ho **Source Energies (keV): 216, 280, 712**
Plutonium Scrap only: Added 81 keV

Material Assayed	Basis	Elements	MBS Bias (%) at Gamma Energy			Random Error (Standard Deviation, %)				
			60 keV	129 keV	Max Abs Others	60 keV	129 keV	153 keV	186 keV	Max Others
Job-Control Waste	C	Sn	19.4	-0.4	0.5	31.8	7.0	4.1	2.1	1.3
Rubble	C	Sn	10.1	0.7	0.3	26.3	5.8	3.6	2.0	1.2
Dirt	C	Sn	34.2	2.5	1.4	34.6	7.5	4.7	2.6	1.6
Sand	C	Sn	18.2	0.1	0.7	39.3	7.7	4.7	2.6	1.6
Uranium Oxide and Ash	C	U	-31.3	-1.1	0.8	7.8	5.2	3.6	2.1	1.4
Plutonium Scrap	C	Pu Sn	-5.1	-0.7	0.4	2.1	4.3	3.0	1.8	1.2
Raschig Rings	C	Sn	26.1	0.5	0.4	36.2	6.8	4.2	2.3	1.5
Lead-Lined Gloves	C	Pb	2.0	1.4	0.3	3.1	2.9	2.4	1.9	1.5

Table 4. Summary table of assay biases and random errors for ^{166m}Ho scenarios.

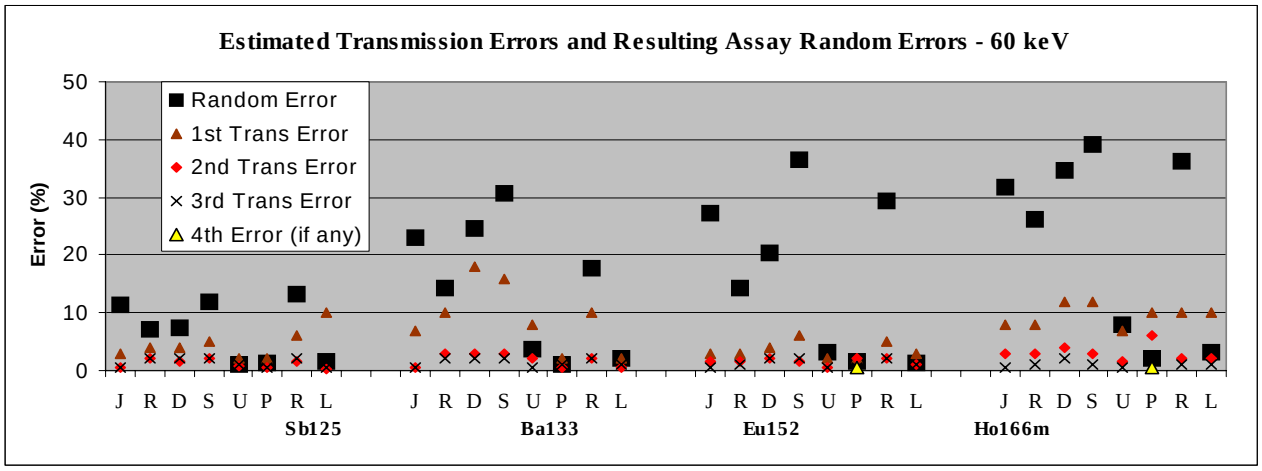

Figure 3. Estimated error fractions assigned to "measured" transmissions and the resulting random errors in the 60 keV assay results. Transmission errors are lowest to highest energy. For this study ^{125}Sb outperforms the other transmission sources.

data. The relative errors assigned to the transmission gammas for each scenario, in order lowest to highest energy are shown in Figure 3, along with the resulting calculated 60 keV assay random errors due to transmission measurement errors and the MBS method. Assay materials are in the same order as in the tables, and the first letter of each is shown on the graph. It is clear from the figure that these random errors are strongly dependent on the transmission source used, hence the energies of the transmission gammas.

CONCLUSIONS

The MBS method allows the complex energy dependence of transmission factors governed primarily by the behavior of the photoelectric and Compton interaction processes (in the SNM range) to be represented simply using only two or three free parameters. The method works very well for the materials included in this study and which encompass a broad range of measurement scenarios. Since the assay random errors were found to be normally distributed for most scenarios and assay energies, typical error propagation techniques can be used for MBS analyses. However, the potential for skewed error distributions with extrapolation to low energies should be kept in mind. Also, MBS analysis must be set up properly to avoid MBS solution failures. The most important measure to reduce the failure rate is to use a number of measured transmission energies that exceeds the number of basis elements. Failures can be reduced or avoided by use of a transmission source having lines which can be well determined in the 150-200 keV range and in the 650-1000 keV range. Ideally the source should also be long-lived, readily available, low cost and have a simple spectrum. The most nearly ideal single nuclide transmission source of those included in the study is ^{125}Sb. The other sources have minor deficiencies, but also provided acceptable results.

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