Covariances for Gamma Spectrometer Efficiency Calibrations

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Abstract. An essential part of the efficiency calibration of gamma spectrometers is the determination of uncertainties on the results. Although this is routinely done, it often does not include the correlations between efficiencies at different energies. These can be important in the subsequent use of the detectors to obtain activities for a set of dosimetry reactions. If those values are not mutually independent, then obviously that fact could impact the validity of adjustments or of other conclusions resulting from the analysis. Examples are given of detector calibrations in which the correlations are calculated and propagated through an analysis of measured activities.

1. Introduction

Calibration of the full-energy detection efficiency of gamma spectrometers is an essential step in radiometric analysis, used for nearly all reactor surveillance dosimetry. The calibration is usually performed using multi-element standard sources, or a combination of these together with sources of some isotopes that produce many gamma lines. The sources used do not include all isotopes which the detectors are later used to measure, and therefore the calibration has to include an energy-dependent fitting to obtain the efficiencies at energies other than those in the calibration measurements. This fitting process introduces strong correlations between gamma lines of nearby energies.

An essential part of the calibration is the determination of uncertainties on the results. Although this is routinely done, it often does not include the correlations between efficiencies at different energies. These can be important in the subsequent use of the detectors to obtain activities for a set of dosimetry reactions [1]. If these results are not mutually independent, then obviously that fact could impact the validity of adjustments [2] or of other conclusions resulting from the analysis, unless the correlations are fully traced and included in the analysis. The calculation of the correlations is straightforward, particularly when the class of functions used to fit the energy-dependent efficiency is linear in the unknown parameters. This includes cases in which multiple spectrometers are used, after calibration with the same or similar sources [3].

Examples are given in this paper of detector calibrations in which the correlations are calculated and propagated through an analysis of measured activities. Methods of calculating the correlations are described in detail, and numerical examples are presented in which strong correlations are present between activities measured for different activation detectors. The effect of uncertainties arising from

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other causes than source calibration and counting errors is also briefly discussed, so that the results of this work can be placed in context with other errors arising in radiometric dosimetry.

2. Previous Work

Many papers have been published on the topic of efficiency calibrations for germanium spectrometers. Only a few will be mentioned here. McNelles and Campbell used a variety of analytical functions [4]. Most had more than four variable parameters, and most successful was the 8-parameter expression

\[
\varepsilon = (a_1/E)^{a_2} + a_3 \exp(-a_4 E) + a_5 \exp(-a_6 E) + a_7 \exp(-a_8 E). \tag{1}
\]

Using calibrated sources with gamma energies in the energy range 160–1333 keV, the reduced, weighted chi-squared value was 0.69 and the mean deviation 0.5%. The uncertainty reported in the fitted efficiency curve varied from 1% to 2.3% over that energy range.

Yoshizawa et al. [5] measured gamma ray intensities over the energy range 279 to 2754 keV, and claimed uncertainties in the range 0.3% to 0.5%. None of the analytical expressions tested for detector efficiency could fit the curve satisfactorily over that range, so the authors used piecewise fitting in different energy ranges and also had to apply “graphical corrections”. It may be that the classes of functions usually employed are not be adequate for fitting data with uncertainties much less than 1%, which may be needed when determining emission probabilities for reference standards. For activation dosimetry, however, uncertainties of 1% are usually considered more than adequate.

Debertin [6] introduced the following class of functions:

\[
\varepsilon = a_1 \ln(E) + a_2 \ln(E)/E + a_3 (\ln(E))^2 / E + a_4 (\ln(E))^4 / E + a_5 (\ln(E))^5 / E. \tag{2}
\]

These “proved to result in a sufficiently good fit for our Ge(Li) detectors” for the energy region 120–1400 keV. This paper [6] is interesting because it introduces a class of fitting functions that is linear in the unknown parameters \(a_i\), and also because of observations made about correlations in the activities used for calibration when multi-energy sources are used: “correlations can greatly affect calculated standard deviations and should be taken into account for strongly correlated experimental data.”

Gray and Ahmad [7] also selected a linear class of fitting function, defined by:

\[
\varepsilon = (a_1 + a_2 \ln(E) + a_3 (\ln(E))^2 + a_4 (\ln(E))^3 + a_5 (\ln(E))^5 + a_6 (\ln(E))^7) / E. \tag{3}
\]

Their paper discusses at length the benefits of using linear fitting functions. The authors concluded that there is strong evidence that this class of functions “adequately represents the efficiency versus energy relationship for Ge(Li) detectors over the energy range (120–1850) keV, for efficiency measurements with standard deviations greater than 1%.”

Another class of linear fitting functions has been used to fit the logarithm of the measured efficiencies:

\[
\ln(\varepsilon) = \sum_{i} a_i \left( \ln \left( \frac{E_i}{E} \right) \right)^{i-1}, \tag{4}
\]

where \(E_c\) is an arbitrary energy chosen near the middle of the energy range (to simplify the problem numerically). The number of parameters, \(I\), was chosen by Lin et al. [8] to be 6, for the energy range 300–9700 keV. Kis et al. [9] used up to 9 parameters for the range 50–11000 keV. Such a large number is not needed for a narrower energy range, and may be impractical when the number of measured efficiencies is limited, because the number of parameters must always be fewer than the number of measurements, ideally many fewer. It must be remembered that the purpose of fitting a continuous function to the data is so that efficiencies at energies in between those included in the calibration can be determined. Functions which do not sufficiently constrain the values in between the fitted points are useless for this purpose. The criterion stated in [9] for determining the number of needed parameters

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is as follows: “In order to justify the introduction of the (n+1)th parameter in the fitting the difference between the chi-squares for n parameters and n+1 parameters has to be greater than 3.841, which comes from the tabulated values of the chi-square distribution with one parameter.” The functions of the class (4) were built into the code Hypermet-PC [10], and are also now commonly used in commercially available software sold with gamma spectrometry systems.

3. Generalized Least Squares Model for Linear Fitting Formulae

In cases where the efficiency is represented by a model that is linear in the unknown parameters, the generalized least squares model is applicable.

Let \( \mathbf{A} \) represent a column vector of the parameters \( a_i \), for \( i = 1, I \). Then the efficiencies measured for each photon energy of the calibration source can be represented by:

\[
\mathbf{e} = \mathbf{S}\mathbf{A} + \mathbf{\delta},
\]

where \( \mathbf{e} \) is a column vector of the measured value for the efficiencies, \( \mathbf{S} \) is the design matrix for the model, and \( \mathbf{\delta} \) is a vector representing the experimental deviations from the ideal model. If we assume that \( \mathbf{\delta} \) is distributed as a multivariate normal distribution with zero mean and covariance matrix \( \mathbf{V}_\mathbf{e} \), then the least-squares solution for the expectation values of the parameters is given by:

\[
\hat{\mathbf{A}} = (\mathbf{S}'\mathbf{V}^{-1}_\mathbf{e}\mathbf{S})^{-1}\mathbf{S}'\mathbf{V}^{-1}_\mathbf{e}\mathbf{e},
\]

where the symbol indicates the transpose of a matrix. The covariance matrix of this estimate is

\[
\mathbf{V}_{\hat{\mathbf{A}}} = (\mathbf{S}'\mathbf{V}^{-1}_\mathbf{e}\mathbf{S})^{-1}.
\]

Notice that this matrix is necessarily full rank, equal to the dimension of the parameter vector, and that this requires the number of the measured efficiencies to be greater than the number of parameters.

Now, if we need to calculate the efficiencies for the photon energies of a set of activation detectors, represented by a column vector \( \mathbf{e}_d \), then

\[
\hat{\mathbf{e}}_d = \mathbf{D}\hat{\mathbf{A}},
\]

where \( \mathbf{D} \) is the design matrix for the same model as \( \mathbf{S} \), but with its elements calculated for the new set of energies (an example will be given later).

Making use of Eq. (7), the covariance matrix \( \mathbf{V}_d \) for this estimate of \( \mathbf{e}_d \) is given by,

\[
\mathbf{V}_d = \mathbf{D}(\mathbf{S}'\mathbf{V}^{-1}_\mathbf{e}\mathbf{S})^{-1}\mathbf{D}'.
\]

Now in this case the rank of the matrix \( \mathbf{V}_d \) can be no greater than the number of parameters in \( \mathbf{A} \), so the matrix will be degenerate if the number of activation detectors is larger than that. This means, for example, that if there are 6 parameters in the efficiency model but one evaluates efficiency at 18 detector energies, then there must be 12 linear equality constraints on the efficiency variations, because only 6 components can vary independently.

It should be noted that the covariance matrices given in Eqs. (7) and (9) do not depend on the measured values of the efficiencies, but only on their uncertainties. This is convenient because, as we shall see, those may sometimes be estimated without even specifying any particular measurement, or even any particular detector.

The preceding analysis assumes a model in which the efficiency function is linear in the parameters, but it can be readily, and advantageously, adapted for the case in which the natural logarithm of the efficiency is represented by a model that is linear in its parameters. In that case, we have to assume that the log of the measurement has a normal distribution of its errors, equivalent to assuming that the distribution of the measurements is log normal. This may be a good assumption, but the point is moot
Table 1. Calibration source specifications.

<table>
<thead>
<tr>
<th>radionuclide</th>
<th>Cd-109</th>
<th>Co-57</th>
<th>Ce-139</th>
<th>Hg-203</th>
<th>Sn-113</th>
<th>Sr-85</th>
<th>Cs-137</th>
<th>Y-88</th>
<th>Co-60</th>
<th>Co-60</th>
<th>Y-88</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gamma energy (keV)</td>
<td>88.03</td>
<td>122.1</td>
<td>165.9</td>
<td>279.2</td>
<td>391.7</td>
<td>514.0</td>
<td>661.6</td>
<td>898.0</td>
<td>1173</td>
<td>1333</td>
<td>1836</td>
</tr>
<tr>
<td>uncertainty % (1σ)</td>
<td>2.9</td>
<td>0.8</td>
<td>0.8</td>
<td>2.1</td>
<td>0.8</td>
<td>0.9</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>correlation coefficient</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.95*</td>
<td>0.999*</td>
<td>0.999*</td>
<td>0.95*</td>
<td></td>
</tr>
<tr>
<td>% counting uncertainty</td>
<td>2.68</td>
<td>1.06</td>
<td>0.80</td>
<td>0.60</td>
<td>1.07</td>
<td>2.19</td>
<td>1.23</td>
<td>0.71</td>
<td>0.56</td>
<td>0.69</td>
<td>0.80</td>
</tr>
</tbody>
</table>

for standard deviations of 5% or less, expected in this case, because then the difference between log normal and normal distributions is negligible.

In order to adapt the analysis for a model in which log(efficiency) is given by a linear model in \( A \), we have only to define \( V_{l_{e}} \) and \( V_{ld} \) as the covariances of the logarithms of the measured efficiencies and the calculated activation detector efficiencies, respectively. Then from (9) we can write immediately

\[
V_{ld} = D.(S'.V_{l_{e}}^{-1}.S)^{-1}.D',
\]

where the matrices \( D \) and \( S \) are now the ones that apply to the new model that is analogous to Eqs. (5) and (8), except that \( \ln(\epsilon) \) appears on the left side, and \( \delta \) represents the fractional deviations from the ideal model, equal to the deviations of their logarithms. This result is very satisfactory, because the matrices that specify the log covariances are the same as the fractional covariances of the efficiencies themselves! These quantities are more convenient to deal with than the actual variances for several reasons: their magnitudes are more similar for all the gamma emission lines, they are more readily understandable in terms of percentage errors, and finally the numerical matrix inversions are quicker and more accurate when the diagonal element values do not vary by orders of magnitude.

When correlation matrices are shown later, they were calculated using Eq. (10), and then dividing each element of the covariance matrix by the square roots of the diagonal elements corresponding to the row and column of that element. These square roots represent the fractional standard deviations. This standard operation is based on the definition of the correlation coefficient.

If several detectors are calibrated with the same standard source, then naturally there will be a cross correlation between their evaluated efficiencies. This situation can be dealt with in a simultaneous evaluation using an expanded model in which the vectors \( A \) for the parameters are appended into a single longer vector, with a similar extension of the vector of measured efficiency values and of the other matrices. A much simpler method is available, however. If only the source strength uncertainties are considered, then the covariance matrix calculated using (10) for each detector is identical if the same source is used, and the cross covariance matrix between detectors is also the same, because for each individual gamma energy the detectors are perfectly correlated and with equal uncertainties. Now if the random error from counting statistics is found for each detector, it may be represented in fractional form as a diagonal covariance matrix. This can be simply added to the previous fractional covariance matrix for that detector, and similarly for each detector. The counting errors are independent between detectors, and so do not contribute to the cross covariance matrices, which are left unchanged. The cross correlation is thus diluted by the random errors because of the increased total uncertainty.

4. Numerical Examples

A commonly used type of calibration source has 11 gamma energies from 88 keV to 1836 keV, from 9 radioisotopes, as shown in Table 1.

The uncertainties listed in row 3 of Table 1 are based on the calibration certificate of a source (Isotrak QCD1) calibrated according to the requirements of the United Kingdom Accreditation Service (UKAS).
The certificate lists combined Type A and Type B uncertainties, and an “Expanded Uncertainty”. The latter is “based on a standard uncertainty multiplied by a coverage factor $k = 2.00$.” The uncertainties in Table 1 are half of those expanded uncertainties (rounded up to one decimal place) and thus correspond to 1 standard deviation ($1\sigma$). The correlation coefficients given in row 4 are not from the calibration certificate, but are assigned by inspecting the decay schemes of Co-60 and Y-88. The values listed apply to the correlations between the members of the two pairs of lines, other correlations being assumed to be zero. Thus the covariance matrix is diagonal, except for those two pairs of lines. These two pairs of lines might also require attention to be paid to summing corrections, unless the geometrical efficiency is small. The counting uncertainties in row 5 represent statistical errors ($1\sigma$) of an actual calibration count. The value higher than 2% for the Sr-85 gamma line is a consequence of its short half life, because the source had undergone some decay before the count.

It is a common practice to use calibration sources containing isotopes with many lines, for example Eu-152, Eu-154 and Ag-110m. This should only be done if a reliable full covariance matrix for the emission lines is available, which is often not the case. See the previously quoted comment on this by Debertin [6]. Even when the coincidence summing corrections are assumed to be correctly handled, the lack of a proper covariance matrix for the source data will invalidate any fitting based on statistical criteria, and give incorrect uncertainties for the derived efficiencies. This paper does not consider further the use of multi-energy sources, other than Co-60 and Y-88, for that reason.

Using the model of equation (4), repeated here for convenience,

$$\ln (\varepsilon) = \sum_{i} a_i \left( \ln \left( \frac{E_c}{E} \right) \right)^{i-1}, \quad (11)$$

and selecting $I = 6$, we have a six parameter model that can be used to fit a set of 11 efficiency measurements for a source of the type specified in Table 1. A convenient choice for $E_c$ is then 962 keV, the mean of the highest and lowest energies in the set.

The covariance matrix for the measurements includes the covariance matrix of the calibrated emission rates of the source, derived from Table 1, and it can be represented in fractional form. In that case the entries represent the expectation values of the paired products of fractional deviations from the mean. Added to this is a covariance matrix representing counting errors in the efficiency measurements. These include statistical counting errors, and possibly also bias errors from the peak area algorithm used. Positioning errors for the calibration source itself need not necessarily be considered part of the calibration, but when an activation monitor is counted, we have to consider the discrepancy between its position and that of the calibration source. This is liable to have a random component that can be observed by repetition experiments, and also a bias (Type B) uncertainty attributable to the geometrical differences of the activation detectors from the calibration source. It is assumed here that all these counting errors can be represented by diagonal matrices of uncorrelated random components, and that these can be conveniently represented as percentage or fractional errors. Fractional covariance matrices may be simply added together as needed, when the product of random variables is found and their different sources of error are represented in separate matrices.

For a given input covariance matrix, the covariance matrix for the fractional errors of the efficiencies at any set of activation detector gamma energies can be calculated using Eq. (10). We need the matrices $S$ and $D$. Table 2 illustrates the matrix $S$ for the energies of the sources in Table 1, and for the linear model described by equation (4), using $I = 6$ and

$$x = \ln \left( \frac{E_c}{E} \right) \quad \text{and} \quad E_c = 962 \text{ keV}. \quad (12)$$

The matrix $D$, for the activation detector energies, is constructed in exactly the same manner, and so need not be illustrated. For the examples used here, the following activation detector gamma energies were used (Fig. 1).
Table 2. The Design Matrix, $S$, for the Fit. Elements of $S$ are within the heavy outline.

<table>
<thead>
<tr>
<th>$E$ (keV)</th>
<th>$x^0$</th>
<th>$x^1$</th>
<th>$x^2$</th>
<th>$x^3$</th>
<th>$x^4$</th>
<th>$x^5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>88.03</td>
<td>1.0</td>
<td>2.3913</td>
<td>5.7185</td>
<td>13.6748</td>
<td>32.7011</td>
<td>78.1995</td>
</tr>
<tr>
<td>122.06</td>
<td>1.0</td>
<td>2.0645</td>
<td>4.2622</td>
<td>8.7993</td>
<td>18.1661</td>
<td>37.5039</td>
</tr>
<tr>
<td>165.85</td>
<td>1.0</td>
<td>1.7579</td>
<td>3.0903</td>
<td>5.4326</td>
<td>9.5501</td>
<td>16.7884</td>
</tr>
<tr>
<td>279.19</td>
<td>1.0</td>
<td>1.2371</td>
<td>1.5305</td>
<td>1.8934</td>
<td>2.3423</td>
<td>2.8978</td>
</tr>
<tr>
<td>391.69</td>
<td>1.0</td>
<td>0.8985</td>
<td>0.8074</td>
<td>0.7255</td>
<td>0.6519</td>
<td>0.5857</td>
</tr>
<tr>
<td>513.99</td>
<td>1.0</td>
<td>0.6268</td>
<td>0.3929</td>
<td>0.2463</td>
<td>0.1544</td>
<td>0.0968</td>
</tr>
<tr>
<td>661.65</td>
<td>1.0</td>
<td>0.3743</td>
<td>0.1401</td>
<td>0.0524</td>
<td>0.0196</td>
<td>0.0073</td>
</tr>
<tr>
<td>898.02</td>
<td>1.0</td>
<td>0.0688</td>
<td>0.0047</td>
<td>0.0003</td>
<td>0.0000</td>
<td>0.0000</td>
</tr>
<tr>
<td>1173.22</td>
<td>1.0</td>
<td>0.1985</td>
<td>0.0394</td>
<td>−0.0078</td>
<td>0.0016</td>
<td>0.0003</td>
</tr>
<tr>
<td>1332.49</td>
<td>1.0</td>
<td>0.3258</td>
<td>0.1061</td>
<td>−0.0346</td>
<td>0.0113</td>
<td>−0.0037</td>
</tr>
<tr>
<td>1836.01</td>
<td>1.0</td>
<td>0.6463</td>
<td>0.4177</td>
<td>−0.2700</td>
<td>0.1745</td>
<td>−0.1128</td>
</tr>
</tbody>
</table>

Figure 1. Activation detector radioisotopes and gamma energies in keV.

Figure 2. Efficiency standard deviations and cross correlations for 18 gamma energies used in radiometric monitors, when counting statistics are not included. Lines shown on the plot are eye-guides.
Figure 3. Efficiency standard deviations and cross correlations for 18 gamma energies used in radiometric monitors, when the calibration counting statistics are included.

Figure 2 shows the standard deviations and correlation matrix for efficiencies calculated using Eq. (9) for the 18 activation detector energies of Fig. 1. The data series shown in the lower part of Fig. 2 are simply the rows of the correlation matrix. The diagonal entries, identically equal to 1, are omitted. In this example, only the source uncertainties from row 3 of Table 1 were considered, with the correlations shown in row 4. This represents the situation when counting statistic are negligible. Very strong correlations are present between activities measured for different activation detectors for all gamma lines from 658 keV and up. The correlations between the two pairs of gamma lines for Y-88 and Co-60 cause highly correlated results above 900 keV.

Figure 3 shows the standard deviations and correlation matrix for efficiencies calculated using equation (9) when the statistical counting errors from the calibration, shown in row 5 of Table 1, are included together with the source strength uncertainties. The counting errors dilute the correlation between the pairs of gamma lines for Y-88 and Co-60, and have a remarkable effect on the correlations for the high energy part of the matrix (compare with Fig. 2). There are still many strong correlations, however, and this is true no matter how large are the counting errors in the calibration, because the correlation matrix can only be of rank 6, equal to the number of parameters in the fitting.

Values for uncertainties arising from detector calibration for some fast/thermal neutron spectral index ratios are: $^{58}\text{Ni}(n,p)^{58}\text{Co}(811\text{ keV})/^{45}\text{Sc}(n,g)^{46}\text{Sc}(889\text{ keV})$, ± 0.3%; $^{115}\text{In}(n,n')^{115m}\text{In}(336\text{ keV})/^{197}\text{Au}(n,g)^{198}\text{Au}(412\text{ keV})$, ±0.4%. When the correlations are ignored, the values found for the same ratios are ±1.1%; and ±1.3%, respectively. These values include statistical errors for the calibrations, but not for the activation detector counting. In practice the latter might dominate.

Figure 4 shows the standard deviations and correlation matrix for activities measured using a detector calibrated as described for the data of Fig. 3. Statistical errors for the activity counting are included, and these are assumed to have a standard deviation of 1%, which is typical of many reactor dosimetry experiments. The correlations are much lower than those of Fig. 3, but still significant.
Figure 4. Activity standard deviations and cross correlations for 18 gamma energies used in radiometric monitors, when random errors of 1% are included for the activity counting.

5. Conclusions

The analysis presented shows that activities measured in radiometric monitors need not have uncertainties larger than 1.5%, and that those uncertainties are strongly correlated between detectors. Because of these correlations, The examples given show that detector efficiency uncertainties can be almost negligible in reactor spectrum determination when the correlations are correctly accounted for.

Larger uncertainties are often quoted, and may be present because of filter corrections, flux perturbation corrections, and positioning errors during irradiation. Some of those uncertainties properly belong to the calculations with which the measurements are compared. The composition of the monitor materials may also add uncertainties, but if pure natural elements are used the uncertainty in isotopic abundance ought not to be included when the activities are compared with calculated ones, if the activation cross section measurement and evaluation also used natural elements. The same applies to gamma intensities, if the gamma lines used in the cross section measurement are known.

References


