Determining the probability of locating peaks using computerized peak-location methods in gamma-ray spectra as a function of the relative peak-area uncertainty

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Abstract

The probabilities of ~~locating~~^{detecting} peaks with a high relative peak-area uncertainty were determined empirically with nine types of peak-location software used in laboratories engaged in gamma-ray spectrometry measurements. It was found that it is not possible to locate the peaks with a probability of 0.95, for those peaks that have a relative peak-area uncertainty in excess of 50%. ~~Locating~~^{Identifying} peaks at these relative peak-area uncertainties with a probability greater than 0.95 is only possible in the library-driven mode, where the peak positions are supposed a-priori. The deficiencies of the library-driven mode and the possibilities to improve the probabilities of locating peaks are briefly discussed.

Keywords: Gamma-ray spectrometry; Relative peak-area uncertainty; Peak location; Decision threshold;

Introduction

In gamma-ray spectrometry measurements it is frequently necessary to evaluate peaks that cannot be easily distinguished from fluctuations in the continuous background on which they are superimposed. Since the presence of peaks in the spectrum implies the presence of gamma-ray emitters, the obvious criteria to be used, when considering a small peak in order to signal the presence of a gamma-ray emitter, are related to the position of the peak in the spectrum. This should correspond to the energy where the gamma-rays from the emitter are registered, and to its width, which should resemble the resolution of the spectrometer.

However, since statistical fluctuations in the continuous background can arise at any position in the spectrum and assume any width, those two criteria are not sufficient to differentiate between statistical fluctuations and small signals.

Therefore, it is necessary to establish a criterion based on the expressiveness of the peak. If the peak is small enough, compared to the statistical fluctuations that can be reasonably expected, it is supposed to represent a statistical fluctuation and not a signal due to the presence of a gamma-ray emitter. It is obvious that in the case of a quantitative criterion, the standard deviation of the number of counts in an energy interval, corresponding to the resolution of the spectrometer, should be used. This number of counts represents the uncertainty of a peak area having a value of zero, i.e., the null measurement uncertainty (ISO/IEC 2007). This criterion, formulated by Currie in 1968, states that excursions of the continuous background having an area exceeding the critical limit, n^* , defined as

$$n^* = k_{1-\alpha} \cdot u(n=0) \quad (1)$$

are classified as peaks, signalling the presence of gamma-ray emitters. In Eq. (1) $u(n=0)$ denotes the null measurement uncertainty and the proportionality factor $k_{1-\alpha}$ describes the probability of making errors of type I, i.e., that the peak is actually caused by a statistical fluctuation. Usually, critical limits are calculated for a probability of 5%, implying that $k_{1-\alpha}=1.65$.

In the standard ISO 11929 (ISO, 2010) the critical limit, which refers to the number of counts, i.e., the indication, is substituted by the decision threshold, which expresses the probability of the occurrence of type-I errors with the value of the measurand y^* as follows:

$$y^* = k_{1-\alpha} \cdot u(y=0) , \quad (2)$$

where $k_{1-\alpha} \cdot u(y=0)$ denotes the null measurement uncertainty of the measurand.

The standard requires documenting whether the indication, signalling the presence of the measurand, was recognized or not and documenting the corresponding decision threshold in reports on measurements of test items. The decision threshold should be reported, regardless of whether a peak signalling the presence of the gamma-ray emitter was located in the spectrum or not. However, the method of the calculation differs in both cases, since in the first case the height of the continuous background is determined from the spectral regions that do not overlap with the peak region and in the second case from a region of interest comprising the region where the peak is expected (Canberra, 2004 and DIN 2014). Therefore, the decision threshold can be different for the located and for the not-located peaks residing on equal backgrounds and having equal areas. Alternatively, for located peaks, the decision threshold can be calculated from the peak data (Korun et al, 2016).

It is obvious that to comply with the requirement of the standard, the location of the peaks, denoting the presence of the measurand, should be performed with a reliability that is defined by the probability α of making errors of type I. The

expressiveness of the peak that should be recognized with this probability is determined by the definition of the decision threshold. Since the measure for the expressiveness of a peak is the relative uncertainty of its area, from

$$\frac{u(y=0)}{y} = \frac{1}{k_{1-\alpha}} , \quad (3)$$

it follows that for a 0.05 probability of making type-I errors, peaks with a relative uncertainty of 61% should be recognized with a probability of 0.95.

To examine the ability of the software that is currently used in laboratories engaged in gamma-ray spectrometry measurements, an action was initiated within the Low-Level Working Group of the ICRM (International Committee for Radionuclide Metrology), with the aim being to determine the probability of locating peaks in gamma-ray spectra as a function of their relative peak-area uncertainty. The action represents the follow-up of an action already performed with the purpose of determining the systematic influences on peak areas when the peaks are not expressive (Bruggeman et al, 2018). The analyses within the follow-up action were performed on the same set of spectra as the action on the systematic influences, available at the URL address

<https://www.dropbox.com/sh/f9ws2t6g5mn0nqd/AABi05-yq1ICf76EYdvMCZq6a?dl=0> .

It is the purpose of this article to present the results of the action and to draw some conclusions about the software and on the way the laboratories use it.

Methods

The participating laboratories were asked to analyze the set of 35 spectra, to enter the results of the analyses in an Excel file available at the same URL address as the spectra, and then to send the Excel file to the proposer of the

action for comparison and analyses. Twenty laboratories reported twenty-five sets of results. Thirteen sets were obtained with Canberra's peak-location Genie 2000 2nd Differences Program (Canberra, 2004). These analyses were performed with the values of the sensitivity parameter between 2.07 and 2.55. Other types of software that were used were the Genie 2000 VMS Standard Peak Search Program (Canberra, 2006), two sets; APTEC (Aptec 2000), two sets; ANGES (Mishev 2016), two sets; Gamma Vision (Ortec 2016), two sets; Maestro (Ortec 2008), one set; Silena (Silena, 2000), one set; Gamma-W (IAEA 1998), one set; and SPUNAL (Mitsios, 2018), one set.

The probabilities of locating a peak at the energy E , $p(E)$ were calculated as

$$p(E) = n(E)/N \quad (4)$$

where $n(E)$ denotes the number of spectra within a specific set of results, where a peak at the energy E was located, and N is the number of spectra in the set. The uncertainty of this probability was calculated as (Gilmore, 2008 and Korun et al., 2015)

$$u[p(E)] = \sqrt{\frac{p(E) \cdot [1 - p(E)]}{N}} \quad (5)$$

The relative uncertainty, attributed to the mean peak area of a peak occurring at an energy E , was calculated as the mean relative uncertainty of the peak areas calculated at the energy E within a set

$$\bar{u}_{rel}[n_p(E)] = \frac{1}{n(E)} \sum_{i=1}^{n(E)} u_{rel}[n_{pi}(E)] \quad , \quad (6)$$

where $n_{pi}(E)$ denotes the number of counts in the peak at the energy E in the i -th spectrum within a set of results and $u_{rel}[n_{pi}(E)]$ is its relative uncertainty. The uncertainty was calculated as the a-posteriori uncertainty from the widths of the

distributions of the relative uncertainties occurring at the energy E within the set:

$$u[\bar{u}_{rel}[n_p(E)]] = \sqrt{\frac{\sum_{i=1}^{n(E)} [u_{rel}[n_{pi}(E)] - \bar{u}_{rel}[n_p(E)]]^2}{[n(E) - 1] \cdot n(E)}} . \quad (7)$$

Results

The results reported by the participants are presented as the probabilities of the location as functions of the mean relative uncertainty in Figs. 1 and 2 for the various types of software used for the peak analysis. It is a common feature of all the data sets received that at small relative peak-area uncertainties the probability of detection tends to unity. However, it is difficult to compare the various data sets, because the uncertainties attributed to the peak areas vary significantly. Fig. 3 presents the distribution of the data sets reported over the mean relative uncertainty reported for this set. It is obvious that when comparing the results from different sets the mean relative peak-area uncertainties reported cannot be used directly, but relative to the data-set-specific mean relative peak-area uncertainty $\bar{u}_{rel}$

$$\bar{u}_{rel} = \frac{1}{M} \sum_{j=1}^M \bar{u}_{rel}[n_p(E_j)] , \quad (8)$$

where M denotes the number of energies within a data set where the peaks were located and $\bar{u}_{rel}[n_p(E_j)]$ is the mean relative peak-area uncertainty of the peaks located at the energy E_j .

The variability of the data-set-specific mean relative peak-area uncertainties cannot be explained using the methods of evaluation for the peak areas only, i.e.,

depending on whether the peak areas were calculated using the ROI method or using the least-squares method. It is obvious, therefore, that the variability originates in the variability of the spectral regions used for determining the background. However, all the relative uncertainties reported exceeded the lower limit for the relative uncertainty implied by the Poisson statistics for the case of a very wide spectral interval used for determining the height of the continuous background, i.e., $u_{\text{rel}} > n_g^{-1/2}$, where n_g denotes the gross number of counts in the peak region.

Since the correlation between the location probability and the relative peak-area uncertainty is rather scattered, it is difficult to establish a quantitative criterion to express the upper limit of the interval of the relative uncertainties where the probability for the location falls below 0.95. It can be observed that a region of relative peak-area uncertainties exists, where the probabilities for the location being equal to unity are contiguous, and that another region of relative uncertainties exists where the probabilities for location, being smaller than unity, are contiguous. The upper and lower boundaries of these two intervals are denoted by $u_{\text{rel,Max}}$ and $u_{\text{rel,Min}}$, respectively. Figs. 4a and 4b present the correlation between $u_{\text{rel,Max}}/\bar{u}_{\text{rel}}$ and the number of peaks having $u_{\text{rel}} < u_{\text{rel,Max}}$ and the correlation between the number of peaks with $u_{\text{rel}} > u_{\text{rel,Min}}$ and $u_{\text{rel,Min}}/\bar{u}_{\text{rel}}$, respectively. It is obvious that the correlations describe the robustness of the peak analysis. Fig. 4a presents the upper limits of the ranges of the relative uncertainties comprising only peaks with a probability of unity for location. The wider this range is, the smaller is the probability of type-II errors, i.e., the more robust is the peak-location algorithm for locating expressive peaks and the more peaks encompass the interval $[0, u_{\text{rel,Max}}]$. Fig. 4b presents the largest mean relative peak-area uncertainty of a peak, located with a probability of unity. The larger this relative uncertainty is, the smaller is the probability of type-II errors in the case of unexpressive peaks, i.e., the more the peak-location algorithm is sensitive. Also, the larger $u_{\text{rel,Min}}$ is, the fewer peaks occur with the mean peak-area uncertainty exceeding $u_{\text{rel,Min}}$.

It is obvious that the relative peak-area uncertainty corresponding to the probability of locating a peak with the probability of 0.95 must lie in the interval $[u_{rel,Max}, u_{rel,Min}]$. To determine this uncertainty, the dependence of the probability for location as a function of the relative uncertainty was smoothed, and from the smoothed dependence the smallest relative uncertainty was determined, where the probability for detection drops below 0.95. The distribution of the datasets over this relative uncertainty is shown in Fig. 5.

Discussion

It can be seen in Fig. 5 that the mean relative peak-area uncertainties, needed for locating a peak with a probability of 0.95, are smaller than 60% in all the data sets, except one. The main parameter, which influences the probability for location and the data-set-specific mean relative peak-area uncertainty, is the value of the sensitivity parameter, which influences the location probability of the inexpressive peaks. High probabilities for location increase the incidence of the locations of unexpressive peaks, having a high relative peak-area uncertainty. Therefore, in the data sets they increase the mean peak-area uncertainty. The correlation among the number of energies, where the peaks were located within a data set, and $\bar{u}_{rel}$ is presented in Fig. 6. Here, the data sets where the peak areas were calculated with the least-square method and the data sets calculated with the ROI method are denoted with open triangles and full dots, respectively. It can be observed that the same number of located peaks, i.e., for data sets analysed with the same effective sensitivity, the mean relative uncertainties obtained with the LSQ method are smaller, as expected (Korun et al., 2018). For the data sets comprising more than 100 peaks, the type of software used is identified. In Fig. 7 the correlation among $\bar{u}_{rel}$ and the relative uncertainty needed to locate a peak with a probability of 0.95 is presented. It follows from Fig. 7 that some types of software achieve a larger mean relative peak-area uncertainty with a probability for detection of 0.95, despite a smaller $\bar{u}_{rel}$. By comparing Figs. 6 and 7 it is clear that this occurs although the number

of energies where the peaks were located was approximately the same. The reason for this is the better selectivity for recognizing inexpressive peaks. These data sets are, in Fig. 5, located at the mean relative peak-area uncertainties, which are recognized with a probability of 0.95, in excess of 30%.

It should be mentioned here that the data set obtained with the Gamma-Vision software differs qualitatively from the other data sets (Fig. 2). This set was calculated in the library-driven mode. Here, the spectrum is not scanned for the presence of peaks, but at energies given in a nuclide library the presence of a peak is supposed. Then, the spectrum in the vicinity of this energy is decomposed using the least-squares method into the continuous background and a peak. In this way, by supposing a-priori the presence of a peak, the software is “forced” to calculate the peak’s properties. The advantage of this method was recognized already, since here the probability of detection is not given by the value of the sensitivity parameter, but by the criteria for rejecting the supposed peak (e.g., on the basis of the energy tolerance or the sign of the peak area).

In the case of Gamma-Vision it can be seen that the testing of whether a peak is present at a specific energy was not preformed optimally. Fig. 8 presents the probabilities of locating the peaks, where the peaks located with a probability substantially less than unity are identified by their energy. It is obvious that the peaks at the highest energies were located with a comparatively low probability, which points to the criteria that reject high-energy peaks.

However, there is a price to be paid for the good performance in recognizing unexpressive peaks. By compelling the software to recognize peaks that would not be located otherwise the quality of the results is reduced, since in these cases the shape of the fitted spectrum is less compatible with a peak on a continuous background. This has a consequence that the uncertainties of the calculated peak parameters increase, and with them the relative peak-area uncertainties. Therefore, their dispersion increases and with it also the mean relative peak-area uncertainty and its reproducibility. It can be seen in Figs. 1 and 2 that the largest

mean relative peak-area uncertainties were obtained with the library-driven analysis. This deficiency could be improved by using the penalized least-squares method (Kanisch, 2017).

To increase the probability of locating unexpressive peaks, a smaller value of the peak-significance parameter should be used, but this can cause an unacceptable incidence of spurious peaks, i.e., type-I errors (Korun et al., 2017). To decrease the incidence of spurious peaks, the peak widths should be increased, i.e., the measured spectra should be longer. Then, to locate a peak, the contents of more consecutive channels must exceed the average channel content, which diminishes the probability of recognizing statistical fluctuations as peaks. In addition, by comparing the peak's FWHM with the calibrated FWHM the incidence of spurious peaks could be reduced as well (Korun et al, 2013).

Conclusion

It has been shown that peaks with relative peak-area uncertainties in excess of 50% are not located with a probability of 95%, except in the library-driven peak-analysis mode. However, the disadvantage of this mode is that in the case of unexpressive peaks the peak-area uncertainties increase substantially, making these peak areas of limited use for further analysis. An alternative possibility to increase the probability of locating small peaks is to use a smaller value of the sensitivity parameter, but this should be accompanied by introducing measures to decrease the incidence of spurious peaks.

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Figure captions

Fig. 1. Correlations between the mean relative peak-area uncertainties and the probabilities of [detecting-locating](#) the peaks for different [types-types](#) of software used in the peak analysis.

Fig. 2. Correlations between the mean relative peak-area uncertainties and the probabilities of [detecting-locating](#) the peaks for data sets comprising larger relative peak-area uncertainties.

Fig. 3. Distribution of data sets over the set-specific mean relative uncertainty $\bar{u}_{rel}$.

Fig. 4. Correlation between the number of peaks having $u_{rel} < u_{rel,Max}$ and $u_{rel,Max}/\bar{u}_{rel}$ (a) and the correlation between the number of peaks with $u_{rel} > u_{rel,Min}$ and $u_{rel,Min}/u_{rel,Max}/\bar{u}_{rel}$ (b).

Fig. 5. Distribution of data sets over the relative peak-area uncertainty needed to locate a peak with a probability exceeding 95%.

Fig. 6. Correlation between the number of energies, where peaks were located within a data set, and the set-specific mean relative uncertainty $\bar{u}_{rel}$. The data sets calculated with the ROI method are denoted with full dots and the data sets calculated with the LSQ method with empty triangles.

Fig. 7. Correlation between the set-specific mean relative uncertainty and the relative peak-area uncertainty needed to locate a peak with a probability of 0.95. The data sets calculated with the ROI method are denoted with full dots and the data sets calculated with the LSQ method with empty triangles.

Fig. 8. Correlation between the mean relative peak-area uncertainty and the probability of ~~recognizing~~locating a peak in the data set calculated in the library-driven mode.

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