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Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation — Fundamentals and application —

Part 3: Applications to unfolding methods

Détermination des limites caractéristiques (seuil de décision, limite de détection et extrémités de l'intervalle élargi) pour mesurages de rayonnements ionisants — Principes fondamentaux et applications —

Partie 3: Applications aux méthodes de déploiement

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Foreword

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The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular, the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

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For an explanation of the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT), see www.iso.org/iso/foreword.html.

This document was prepared by This document was prepared by ISO/TC 85, *Nuclear energy, nuclear technologies, and radiological protection*, Subcommittee SC 2, *Radiological protection*.

This second edition of ISO 11929-3 together with ISO 11929-1, ISO 11929-2, cancels and replaces ISO 11929:2010 which have been technically revised, specifically with reference to the type of statistical treatment of the data and extended with respect to the methodology of uncertainty assessment from the ISO/IEC Guide 98-3:2009, to the ISO/IEC Guide 98-3-1:2008.

A list of all the parts in the ISO 11929 series can be found on the ISO website.

Introduction

Measurement uncertainties and characteristic values, such as the decision threshold, the detection limit and limits of the coverage interval for measurements as well as the best estimate and its associated standard measurement uncertainty, are of importance in metrology in general and for radiological protection in particular. The quantification of the uncertainty associated with a measurement result provides a basis for the trust an individual can have in a measurement result. Conformity with regulatory limits, constraints or reference values can only be demonstrated by taking into account and quantifying all sources of uncertainty. Characteristic limits provide, at the end, the basis for deciding under uncertainty.

This standard provides characteristic values of a non-negative measurand of ionizing radiation. It is also applicable for a wide range of measuring methods extending beyond measurements of ionizing radiation.

The limits to be provided according to the ISO 11929 series for specified probabilities of wrong decisions allow detection possibilities to be assessed for a measurand and for the physical effect quantified by this measurand as follows:

— the "decision threshold" allows a decision to be made on whether or not the physical effect quantified by the measurand is present;

— the "detection limit" indicates the smallest true quantity value of the measurand that can still be detected with the applied measurement procedure; this gives a decision on whether or not the measurement procedure satisfies the requirements and is therefore suitable for the intended measurement purpose;

— the "limits of the coverage interval" enclose, in the case of the physical effect recognized as present, a coverage interval containing the true quantity value of the measurand with a specified probability.

Hereinafter, the limits mentioned are jointly called the "characteristic limits".

NOTE According to ISO/IEC Guide 99:2007 updated by JCGM 200:2012 the term "coverage interval" is used here instead of "confidence interval" in order to distinguish the wording of Bayesian terminology from that of conventional statistics.

All the characteristic values are based on Bayesian statistics and on the ISO/IEC 98-3 Guide to the Expression of Uncertainty in Measurement as well as on the ISO/IEC Guide 98-3-1 and ISO/IEC 98-3-2. As explained in detail in ISO 11929-2, the characteristic values are mathematically defined by means of moments and quantiles of probability distributions of the possible measurand values.

Since measurement uncertainty plays an important part in ISO 11929, the evaluation of measurements and the treatment of measurement uncertainties are carried out by means of the general procedures according to the ISO/IEC Guide 98-3 and to the ISO/IEC Guide 98-3-1; see also References [9] to [13]. This enables the strict separation of the evaluation of the measurements, on the one hand, and the provision and calculation of the characteristic values, on the other hand. The ISO 11929 series makes use of a theory of uncertainty in measurement [14] to [16] based on Bayesian statistics (e.g. [17] to [22]) in order to allow to take into account also those uncertainties that cannot be derived from repeated or counting measurements. The latter uncertainties cannot be handled by frequentist statistics.

Because of developments in metrology concerning measurement uncertainty laid down in the ISO/IEC Guide 98-3, ISO 11929:2010 was drawn up on the basis of the ISO/IEC Guide 98-3, but using Bayesian statistics and the Bayesian theory of measurement uncertainty. This theory provides a Bayesian foundation for the ISO/IEC Guide 98-3. Moreover, ISO 11929:2010 was based on the definitions of the characteristic values[9], the standard proposal[10], and the introducing article[11]. It unified and replaced all earlier parts of ISO 11929 and was applicable not only to a large variety of particular measurements of ionizing radiation but also, in analogy, to other measurement procedures.

Since the ISO/IEC Guide 98-3-1 has been published, dealing comprehensively with a more general treatment of measurement uncertainty using the Monte Carlo method in complex measurement

evaluations. This provided an incentive for writing a corresponding Monte Carlo supplement[12] to ISO 11929:2010 and to revise ISO 11929:2010. The revised ISO 11929 is also essentially founded on Bayesian statistics and can serve as a bridge between ISO 11929:2010 and the ISO/IEC Guide 98-3-1. Moreover, more general definitions of the characteristic values (ISO 11929-2) and the Monte Carlo computation of the characteristic values make it possible to go a step beyond the present state of standardization laid down in ISO 11929:2010 since probability distributions rather than uncertainties can be propagated. It is thus more comprehensive and extending the range of applications.

The revised ISO 11929, moreover, is more explicit on the calculation of the characteristic values. It corrects also a problem in ISO 11929:2010 regarding uncertain quantities and influences, which do not behave randomly in measurements repeated several times. Reference [13] gives a survey on the basis of the revision. Furthermore, this document gives detailed advice how to calculate characteristic values in the case of multivariate measurements using unfolding methods. For such measurements, the ISO/IEC Guide 98-3-2 provides the basis of the uncertainty evaluation.

Formulas are provided for the calculation of the characteristic values of an ionizing radiation measurand via the "standard measurement uncertainty" of the measurand (hereinafter the "standard uncertainty") derived according to the ISO/IEC Guide 98-3 as well as via probability density functions (PDFs) of the measurand derived in accordance with ISO/IEC Guide 98-3-1. The standard uncertainties or probability density functions take into account the uncertainties of the actual measurement as well as those of sample treatment, calibration of the measuring system and other influences. The latter uncertainties are assumed to be known from previous investigations.

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1 Scope

The ISO 11929 series specifies a procedure, in the field of ionizing radiation metrology, for the calculation of the "decision threshold", the "detection limit" and the "limits of the coverage interval" for a non-negative ionizing radiation measurand when counting measurements with preselection of time or counts are carried out. The measurand results from a gross count rate and a background count rate as well as from further quantities on the basis of a model of the evaluation. In particular, the measurand can be the net count rate as the difference of the gross count rate and the background count rate, or the net activity of a sample. It can also be influenced by calibration of the measuring system, by sample treatment and by other factors.

ISO 11929 has been divided into four parts covering elementary applications in ISO 11929-1, advanced applications on the basis of the ISO/IEC Guide 98-3-1 in ISO 11929-2, applications to unfolding methods in this document, and guidance to the application in ISO 11929-4.

ISO 11929-1 covers basic applications of counting measurements frequently used in the field of ionizing radiation metrology. It is restricted to applications for which the uncertainties can be evaluated on the basis of the ISO/IEC Guide 98-3 (JCGM 2008). In Annex A of ISO 11929-1:2019, the special case of repeated counting measurements with random influences is covered, while measurements with linear analogous ratemeters, are covered in Annex B of ISO 11929-1:2019.

ISO 11929-2 extends the former ISO 11929:2010 to the evaluation of measurement uncertainties according to the ISO/IEC Guide 98-3-1. ISO 11929-2 also presents some explanatory notes regarding general aspects of counting measurements and on Bayesian statistics in measurements.

This document deals with the evaluation of measurements using unfolding methods and counting spectrometric multi-channel measurements if evaluated by unfolding methods, in particular, for alpha- and gamma-spectrometric measurements. Further, it provides some advice on how to deal with correlations and covariances.

ISO 11929-4 gives guidance to the application of the ISO 11929 series, summarizes shortly the general procedure and then presents a wide range of numerical examples.

ISO 11929 Standard also applies analogously to other measurements of any kind especially if a similar model of the evaluation is involved. Further practical examples can be found, for example, in ISO 18589[7], ISO 9696[2], ISO 9697[3], ISO 9698[4], ISO 10703[5], ISO 7503[1], ISO 28218[8], and ISO 11665[6].

NOTE A code system, named UncertRadio, is available for calculations according to ISO 11929- 1 to ISO 11929-3. UncertRadio[35][36] can be downloaded for free from https://www.thuenen.de/en/fi/fields-of-activity/marine-environment/coordination-centre-of-radioactivity/uncertradio/. The download contains a setup installation file which copies all files and folders into a folder specified by the user. After installation one has to add information to the PATH of Windows as indicated by a pop-up window during installation. English language can be chosen and extensive "help" information is available.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 3534-1, *Statistics — Vocabulary and symbols — Part 1: General statistical terms and terms used in probability*

ISO 80000-1, *Quantities and units — Part 1: General*

ISO 80000-10, *Quantities and units — Part 10: Atomic and nuclear physics*

ISO/IEC Guide 98-3, *Uncertainty of measurement — Part 1: Guide to the expression of uncertainty in measurement, JCGM 100:2008*

ISO/IEC Guide 98-3-1, *Evaluation of measurement data — Supplement 1 to the "Guide to the expression of uncertainty in measurement" — a Propagation of distributions using a Monte Carlo method, JCGM 101:2008*

ISO/IEC Guide 98-3-2, *Evaluation of measurement data — Supplement 2 to the "Guide to the expression of uncertainty in measurement" — Models with any number of output quantities, JCGM 102:2011*

ISO/IEC Guide 99, *International vocabulary of metrology — Basic and general concepts and associated terms (VIM)*

3 Terms and definitions

For the purposes of this document, the terms and definitions given in ISO 80000-1, ISO 80000-10, ISO/IEC Guide 98-3, ISO/IEC Guide 98-3-1, ISO/IEC 98-3-2, ISO/IEC Guide 99 and ISO 3534-1 and the following apply.

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at http://www.electropedia.org/

3.1
quantity value
value of a quantity
value
number and reference together expressing magnitude of a quantity

[SOURCE: JCGM 200:2012, 1.19]

3.2
measurement
process of experimentally obtaining one or more quantity values that can reasonably be attributed to a quantity

[SOURCE: JCGM 200:2012, 2.1]

3.3
measurand
quantity intended to be measured

[SOURCE: JCGM 200:2012, 2.3]

3.4
coverage interval
interval containing the set of true quantity values of a measurand with a stated probability, based on the information available

[SOURCE: JCGM 200:2012, 2.36]

Note 1 to entry: A coverage interval does not need to be centred on the chosen measured quantity value (see JCGM 101:2008).

Note 2 to entry: A coverage interval should not be termed "confidence interval" to avoid confusion with the statistical concept.

3.5
measurement method
method of measurement
generic description of a logical organization of operations used in a measurement

[SOURCE: JCGM 200:2012, 2.4]

3.6
measurement procedure
detailed description of a measurement according to one or more measurement principles and to a given measurement method, based on a measurement model and including any calculation to obtain a measurement result

[SOURCE: JCGM 200:2012, 2.6]

3.7
measurement result
result of measurement
set of quantity values being attributed to a measurand together with any other available relevant information

[SOURCE: JCGM 200:2012, 2.9]

3.8
measured quantity value
value of a measured quantity
measured value
quantity value representing a measurement result

[SOURCE: JCGM 200:2012, 2.10]

3.9
true quantity value
true value of a quantity
true value
quantity value consistent with the definition of a quantity

[SOURCE: JCGM 200:2012, 2.11]

Note 1 to entry: In the Error Approach to describing measurement, a true quantity value is considered unique and, in practice, unknowable. The Uncertainty Approach is to recognize that, owing to the inherently incomplete amount of detail in the definition of a quantity, there is not a single true quantity value but rather a set of true quantity values consistent with the definition. However, this set of values is, in principle and in practice, unknowable. Other approaches dispense altogether with the concept of true quantity value and rely on the concept of metrological compatibility of measurement results for assessing their validity.

Note 2 to entry: When the definitional uncertainty associated with the measurand is considered to be negligible compared to the other components of the measurement uncertainty, the measurand may be considered to have an "essentially unique" true quantity value. This is the approach taken by the ISO/IEC Guide 98-3 and associated documents, where the word "true" is considered to be redundant.

3.10
measurement uncertainty
uncertainty of measurement
uncertainty
non-negative parameter characterizing the dispersion of the quantity values being attributed to a measurand, based on the information used

[SOURCE: JCGM 200:2012, 2.26]

Note 1 to entry: Measurement uncertainty includes components arising from systematic effects, such as components associated with corrections and the assigned quantity values of measurement standards, as well as the definitional uncertainty. Sometimes estimated systematic effects are not corrected for but, instead, associated measurement uncertainty components are incorporated.

Note 2 to entry: The parameter may be, for example, a standard deviation called standard measurement uncertainty (or a specified multiple of it), or the half-width of an interval, having a stated coverage probability.

Note 3 to entry: Measurement uncertainty comprises, in general, many components. Some of these may be evaluated by Type A evaluation of measurement uncertainty from the statistical distribution of the quantity values from series of measurements and can be characterized by standard deviations. The other components, which may be evaluated by Type B evaluation of measurement uncertainty, can also be characterized by standard deviations, evaluated from probability density functions based on experience or other information.

Note 4 to entry: In general, for a given set of information, it is understood that the measurement uncertainty is associated with a stated quantity value attributed to the measurand. A modification of this value results in a modification of the associated uncertainty.

3.11
model of evaluation
set of mathematical relationships between all measured and other quantities involved in the evaluation of measurements

Note 1 to entry: The model of evaluation does not need to be an explicit function; it can also be an algorithm realized by a computer code.

3.12
decision threshold
value of the estimator of the measurand, which when exceeded by the result of an actual measurement using a given measurement procedure of a measurand quantifying a physical effect, is used to decide that the physical effect is present

Note 1 to entry: The decision threshold is defined such that in cases where the measurement result, y, exceeds the decision threshold, y^*, the probability that the true value of the measurand is zero is less or equal to a chosen probability for a wrong decision, α.

Note 2 to entry: If the result, y, is below the decision threshold, y^*, it is decided to conclude that the result cannot be attributed to the physical effect; nevertheless it cannot be concluded that it is absent.

3.13
detection limit
smallest true value of the measurand which ensures a specified probability of being detectable by the measurement procedure

Note 1 to entry: With the *decision threshold* (3.12), the detection limit is the smallest true value of the measurand for which the probability of wrongly deciding that the true value of the measurand is zero is equal to a specified value, β, when, in fact, the true value of the measurand is not zero. The probability of being detectable is consequently $(1 - \beta)$.

Note 2 to entry: The terms detection limit and decision threshold are used in an ambiguous way in different standards (e.g. standards related to chemical analysis or quality assurance). If these terms are referred to one has to state according to which standard they are used.

3.14
limits of the coverage interval
values which define a coverage interval

Note 1 to entry: A coverage interval is sometimes known as a credible interval or a Bayesian interval. Its limits are calculated in the ISO 11929 series to contain the true value of the measurand with a specified probability $(1 - \gamma)$.

Note 2 to entry: The definition of a coverage interval is ambiguous without further stipulations. In this standard two alternatives, namely the probabilistically symmetric and the shortest coverage interval are used.

3.15
best estimate of the true quantity value of the measurand
expectation value of the probability distribution of the true quantity value of the measurand, given the experimental result and all prior information on the measurand

Note 1 to entry: The best estimate is the one, among all possible estimates of the measurand on the basis of given information, which is associated with the minimum uncertainty.

3.16
guideline value
value which corresponds to scientific, legal or other requirements with regard to the detection capability and which is intended to be assessed by the measurement procedure by comparison with the detection limit

Note 1 to entry: The guideline value can be given, for example, as an activity, a specific activity or an activity concentration, a surface activity or a dose rate.

Note 2 to entry: The comparison of the detection limit with a guideline value allows a decision on whether or not the measurement procedure satisfies the requirements set forth by the guideline value and is therefore suitable for the intended measurement purpose. The measurement procedure satisfies the requirement if the detection limit is smaller than the guideline value.

Note 3 to entry: The guideline value shall not be confused with other values stipulated as conformity requests or as regulatory limits.

3.17
background effect
measurement effect caused by radiation other than that caused by the object of the measurement itself

Note 1 to entry: The background effect can be due to natural radiation sources or radioactive materials in or around the measuring instrumentation and also to the sample itself (for instance, from other lines in a spectrum).

3.18
background effect in spectrometric measurement
number of events of no interest in the region of a specific line in the spectrum

3.19
net effect
contribution of the possible radiation of a measurement object (for instance, of a radiation source or radiation field) to the measurement effect

3.20
gross effect
measurement effect caused by the background effect and the net effect

3.21
shielding factor
factor describing the reduction of the background count rate by the effect of shielding caused by the measurement object

3.22
relaxation time constant
duration in which the output signal of a linear-scale ratemeter decreases to 1/e times the starting value after stopping the sequence of the input pulses

4 Quantities and symbols

The symbols for auxiliary quantities and the symbols only used in the annexes are not listed. Physical quantities are denoted by upper-case letters but shall be carefully distinguished from their values, denoted by the corresponding lower-case letters.

$\boldsymbol{A}$	response matrix of the spectrometer
A_{ik}	elements of the response matrix A
a_0, a_1	parameters in an algebraic expression of the standard uncertainty of a net counting rate
b	width of a gamma peak, in channels
c_j	position parameter of a peak j, in gamma-ray or alpha-ray spectrometry
diag	indicator for a diagonal matrix
$\boldsymbol{D}$	matrix converting measured activities to decay corrected activity concentrations
$\boldsymbol{d}$	set of statistically independent quantities
f_{B}	function representing the analogue of the total peak area method design factor $[1 + b/(2L)]$ for the peak fitting case (gamma-ray spectrometry)
$f_{\mathrm{att},i}$	self-attenuation correction factor for gamma-line i
$f_{\mathrm{TCS},i}$	true-coincidence-summing correction factor for gamma-line i
f_{d}	decay correction factor including the decay during the measurement
G_k	function of the input quantities X_i (i = 1, …, m)
$\boldsymbol{G}$	column matrix of the G_k
h	full width at half-maximum of a peak, in channels
$h(.)$	function as part of an implicit model
$H(\vartheta_i)$	functional relationship representing the spectral density at ϑ_i of a multi-channel spectrum
i	number of a channel in a multi-channel spectrum obtained by a spectrometric nuclear radiation measurement (i = 1,..1 m)
$\boldsymbol{J}$	matrix of partial derivatives of y^+ with respect to parameters y
L	width of a background region (in channels) adjacent to a gamma peak
L_k	k-th element of a system of functions describing spectral densities, which constitute by superposition the total fitting function
m	number of input quantities; or number of channels in the spectrum; number of lines per nuclide used for activity calculation; or a parameter index

N_i	Poisson-distributed random variable of events counted in channel, i, during the measuring time, t (i =, …, m)
n_i	number of events counted in a channel, i, during the measuring time, t (i =, …, m), estimate of N_i
n	number of output quantities in unfolding
n_g	gross counts in a peak region
n_0	average background counts per channel (spectrum)
p_i	estimate of an input quantity which is not subject to fitting (parameter); contained in the response matrix A
p	column matrix of the p_i
$\boldsymbol{p_c}$	values of non-linear parameters held fixed at their calibrated estimates
$p_{\alpha,i}$	alpha emission probability of gamma-line i
$p_{\gamma,i}$	gamma emission probability of gamma-line i
$\boldsymbol{q}$	column matrix of input quantities considered as parameters; mainly contained in the matrix D
$\boldsymbol{Q}$	matrix of partial derivatives of $\boldsymbol{y}$ with respect to parameters $\boldsymbol{p}$
$\boldsymbol{Q'}$	matrix of partial derivatives of $\boldsymbol{y^+}$ with respect to parameters $\boldsymbol{q}$
R_{ni}	net counting rate of the peak i of interest
$R_{ni,0}$	net counting rate of a background spectrum peak at the position of the peak i of interest
R_{gi}	gross counting rate of the peak i of interest
R_{Ti}	counting rate of the trapezoidal background continuum of the peak i of interest
t	duration of measurement
X_i	random variable of the rate of events counted in channel i during the measuring time, t, input quantity of the evaluation, $X_i = N_i/t$ (i = 1, …, m)
$\boldsymbol{X}$	column matrix of the X_i
x_i	rate of events counted in channel, i, during the measuring time, t, $x_i = n_i/t$ (i = 1, …, m), estimate of X_i
$\boldsymbol{x}$	column matrix of the x_i
x_{net}	column matrix of net counting rates
$u(x_i, x_j)$	covariance associated with x_i and x_j
$u(y_k)$	standard uncertainty associated with y_k
$\boldsymbol{U_x}$	uncertainty matrix of $\boldsymbol{X}$
$\boldsymbol{U_y}$	uncertainty matrix of $\boldsymbol{Y}$
$\boldsymbol{w}$	column matrix of input estimates; $\boldsymbol{w} = (x_1, \ldots, x_m, p_1, p_2, \ldots)^{\mathrm{T}}$ (transposed row matrix)

Y_k	output quantity (parameter) derived from the multi-channel spectrum by unfolding methods (k = 1, …, n)
$\boldsymbol{Y}$	column matrix of the Y_k
y_k	estimate of the output quantity Y_k (k = 1, …, n) resulting from (primary) unfolding
$\tilde{\boldsymbol{y}}$	column matrix $\boldsymbol{y}$ after replacement of y_1 with $\tilde{y}$
$\boldsymbol{Y}^+$	column matrix of final output quantity values after conversion to decay corrected activity concentrations
$\boldsymbol{Y_0}$	column matrix of background counting rates
$\boldsymbol{z}$	column matrix of values z_i fitted to the values x_i
Δ_j	fractional size of a parameter j, used for the parameter increment in partial derivatives with respect to this parameter
ϑ	continuous parameter, e.g. energy or time) related to the different channel numbers in a gamma-ray spectrum
ϑ_i	value of ϑ connected with channel (i = 1, …, m)
ε_i	detection efficiency of a nuclide i or of a gamma-line i
η_l	area fraction of tailing component l of an alpha peak, shape parameter in alpha spectrometry
τ_l	tailing parameter of tailing component l of an alpha peak, shape parameter in alpha spectrometry
σ	width of a Gaussian, parameter in alpha spectrometry
$\psi_k(\vartheta)$	function describing the shape of an individual spectral line or of a background contribution (k = 1, …, n)

5 Evaluation of a measurement using unfolding methods

5.1 General aspects

This clause is based on the ISO/IEC Guide 98-3 and the ISO/IEC Guide 98-3-2. The latter extends the ISO/IEC Guide 98-3 framework to any number of output quantities. Stipulations are made regarding the evaluation of nuclear radiation counting and spectrometric measurements by unfolding methods and the calculation of the characteristic values.

5.2 Models of unfolding and general uncertainty evaluation

When simultaneously measuring more than one output quantity, their individual probability distributions are superimposed with respect to an independent quantity such as radiation energy or time, which may yield (e.g. an energy spectrum or a time-dependent decay-curve) as the primary output of the measurement. Most often, the superposition is linear. A problem occurs if their individual probability distributions suffer from smearing or broadening (e.g. by a non-ideal detector response distribution function). The process of reconstructing the original probability density functions from the measured one, an energy spectrum or a decay-curve, and from the (known) detection response density function is termed as “unfolding”.

Thus, measuring values $\boldsymbol{y}$ of physical quantities $\boldsymbol{Y}$ (rank n), like radionuclide-specific activities or counting rates, starts from measuring values $\boldsymbol{x}$ of $\boldsymbol{X}$ (rank m) (e.g. which represent the channel contents of a multichannel spectrum (energy spectrum) or measured counting rates forming a time-dependent

decay-curve). In the context of this standard, such a measurement is treated as a linear superposition of the source activity and background related distribution functions (or contributions) $A_{k,i}$ of the radionuclide k to each of the components i of the measured $\boldsymbol{x}$: $x_i = \sum_k A_{k,i}\, y_k$.

Although functional representations of detector response functions $A_{k,i}$ (e.g. gamma line-shape) may depend non-linearly on parameters like the width parameter, their associated net areas are always linearly superimposed.

A measurement of more than one output quantity requires a multivariate measurement model. Such quantities are generally mutually correlated because they depend on common input quantities. Depending on how Formulas for evaluating the values of each Y_k can be formulated, two forms of such a model exist. The case of an explicit model is given, if it is possible to formulate separate functions $G_k(X)$, depending only on $\boldsymbol{X}$, for calculating any of the values of Y_k; $\boldsymbol{G}$ is the multivariate measurement function (see ISO/IEC Guide 98-3-2:2008, Clause 6). An implicit model is encountered, if components of $\boldsymbol{Y}$ are involved in such functions also, thereby requiring an iterative process for solving. Such a model for $\boldsymbol{Y}$ is specified by a set of n Formulas

$$h = \left(h_1, \ldots, h_n\right)^{\mathrm{T}} \text{ or } h\left(Y, X\right) = 0 \tag{1}$$

The explicit multivariate model is given by a set of n functional relationships

$$Y_k = G_k\left(X_1, \ldots, X_m\right); \left(k = 1, \ldots, n\right) \tag{2}$$

Estimates y_k of the n measurands Y_k are obtained from Formula (2) by inserting estimates x_i for the m input quantities X_i (i = 1,..., m)

$$y_k = G_k\left(x_1, \ldots, x_m\right); \left(k = 1, \ldots, n\right) \tag{3}$$

The standard uncertainties, $u(x_i)$, and covariances, $u(x_i, x_j)$, associated with the x_i are the elements of the symmetric uncertainty matrix $\boldsymbol{U_x}$ and meet the relations $u(x_i, x_i) = u^2(x_i)$ and $u(x_i, x_j) = u(x_j, x_i)$. If they are given, the analogous standard uncertainties $u(y_k)$ and covariances $u(y_k,y_l)$ associated with the y_k follow from

$$u\left(y_k, y_l\right) = \sum_{i,j=1}^{m} \frac{\partial G_k}{\partial x_i} \cdot \frac{\partial G_l}{\partial x_j} \cdot u\left(x_i, x_j\right); \left(k, l = 1, \ldots, n\right) \tag{4}$$

One obtains $u\left(y_k\right) = \sqrt{u\left(y_k, y_k\right)}$ and $u(y_k, y_l) = u(y_l, y_k)$ ($k \neq l$). For convenience, the partial derivatives $\partial G_k / \partial X_i$ with all the input quantities X_i substituted by their estimates x_i are briefly denoted by $\partial G_k / \partial x_i$ in Formula (4) and in the following.

The model functions G_k need not be explicitly available as arithmetical expressions. They can also be given as an algorithm, for instance, in form of a computer code. In such cases, or when more complicated model functions are involved, the partial derivatives possibly cannot be explicitly derived but can numerically be approximated sufficiently exactly using half of the standard uncertainty $u(x_i)$ as an increment of x_i

$$\frac{\partial G_k}{\partial x_i} = \frac{1}{u\left(x_i\right)} \left\{ G_k\left[x_1, \ldots, x_i + u\left(x_i\right)/2, \ldots, x_m\right] - G_k\left[x_1, \ldots, x_i - u\left(x_i\right)/2, \ldots, x_m\right] \right\} \tag{5}$$

NOTE 1 Formulas (2) to (4) apply for model functions G_k which can be taken as sufficiently linear in the uncertainty ranges between $x_i - u(x_i)$ and $x_i + u(x_i)$. Otherwise, more refined procedures can be applied as described in the ISO/IEC Guide 98-3:2009, 5.1.2.

NOTE 2 In practise, $u(x_i)$ in Formula (5) is replaced by a much smaller value Δ_j (e.g. $\Delta_j = 2 \cdot 10^{-6}\, x_j$) for improving the precision of the differential quotient.

It has to be emphasized that in multivariate measurements, it is more convenient to use matrix notation. Therefore, those quantities, values and functions being denoted by the same symbol are in the following combined to form a column matrix, written as a transposed row matrix and denoted by the same symbol, but in bold face. Examples are $x = (x_1,..., x_m)^T$ and $y = (y_1,..., y_n)^T$ and $\boldsymbol{G}(\boldsymbol{x}) = (G_1,..., G_n)^T$. In addition, the uncertainty matrices $\boldsymbol{U_x} = [u(x_i, x_j)]$ and $\boldsymbol{U_y} = [u(y_k, y_l)]$ and also the sensitivity matrix $G_x = \left(\partial G_k / \partial x_i\right)$ are introduced. Formulas (3) and (4) then read

$$\boldsymbol{y} = \boldsymbol{G}(\boldsymbol{x}); \quad \boldsymbol{U}_y = \boldsymbol{G}_x \boldsymbol{U}_x \boldsymbol{G}_x^T \tag{6}$$

An often-encountered situation in multivariate measurements with an explicit model is described by linear equations which can be combined into a matrix equation of the form $\boldsymbol{X} = \boldsymbol{A} \cdot \boldsymbol{Y}$. It is solved by the method of weighted linear least-squares, also called generalized least-squares, if the system of equations is over-determined.

5.3 Unfolding as a sub-model

The primary output quantities $\boldsymbol{Y}$ obtained from unfolding are activities or counting rates. Most often $\boldsymbol{Y}$ is not the desired measurand, but activity concentrations. The latter may need to be corrected for radionuclide dependent radioactive decay, chemical yield or other influences. Therefore, another measurand of interest, $\boldsymbol{Y}^+$, has often to be calculated

$$\boldsymbol{Y}^+ = \boldsymbol{D} \cdot \boldsymbol{Y} \quad \text{or} \quad \boldsymbol{Y}^+ = \boldsymbol{D} \cdot (\boldsymbol{Y} - \boldsymbol{Y_0}) \tag{7}$$

with a diagonal matrix $\boldsymbol{D}$. Its diagonal elements generally are functions $D_{j,j}$ $(\boldsymbol{q}, y)$ of input quantities $\boldsymbol{q}$; they may also depend on $\boldsymbol{Y}$, if any of the elements y_k is used in them. The latter occurs if for instance the design of the simultaneous measurement of activities of Strontium isotopes is extended such that the measurement of ^{85}Sr, added with a known activity to the sample as a radiochemical tracer, is included in unfolding in order to calculate from it the chemical Strontium yield. The second case of Formula (7) may occur for instance in fitting peak areas in gamma-ray spectrometry, where $\boldsymbol{Y}$ are counting rates of fitted peak areas from which possible peak contributions $\boldsymbol{Y_0}$ still are to be subtracted, which are determined from a separately measured background spectrum.

The extension to a two-step uncertainty propagation implied by utilizing the transformation $\boldsymbol{D}$ is outlined in 5.6.

5.4 Input quantities and their uncertainties

The input quantities encompass all quantities from which measured or other values are used in the unfolding and which have uncertainties associated with them. The count number or counting rate input quantities, denoted as X_i, are separated from the other input quantities, considered as parameters, the values of which are denoted as p_i (see 5.5).

Depending on the type of measurement, the count rate related input quantities, X_i, where n_i events are counted during a measuring time, t_c, may be linked to a common single channel analysis or to individual channel analyzers in the case of multiple counting channels or of even a multi-channel analysis, as in multi-window liquid-scintillation counting or in an alpha- or gamma-ray spectrum.

For a count rate $X_i = R_i$ with the given counting result, n_i, recorded during the measuring time, t_c, and if independent Poisson statistics can be assumed for the individual channels, the specifications $x_i = r_i = n_i / t_c$ and $u^2(x_i) = n_i / t_c^2 = x_i / t_c$ apply. In addition, the covariances often can be set at zero, i.e. $u(x_i, x_j) = 0$ $(i \neq j)$. The counting times may vary between measurements associated with different X_i. The components of uncertainty of measurement comprise uncertainty matrices $U_x = [u(x_i, x_j)]$ and $U_y = [u(y_k, y_l)]$. U_x is often diagonal with the diagonal elements $u^2(x_i) = n_i / t_c^2$.

It is useful to have Formulas by which the uncertainty matrix $\boldsymbol{U_x}$ of the input quantities $\boldsymbol{X}$ can be quantified. Assuming that $\boldsymbol{X}$ represents a vector of net counting rates, it is desirable to put the variances of $\boldsymbol{U_x}$ into the following general form

$$u^2(x_i) = \frac{x_i}{t_{ci}} + \sum_k \left[\frac{x_{0,k}}{t_{ci}} + u^2(x_{0,k}) \right] \tag{8}$$

where the symbols $x_{0,k}$ and t_{ci} denote possible background and interference counting rates, being subtracted from the gross counting rates associated with X_i, and the counting durations of the sample measurements, respectively. Covariances between any pair $(X_i, X_j)(i \neq j)$ of components of $\boldsymbol{X}$ exist if both of the quantities X_i and X_j, considered as functions, depend on the same of any of other input (background-related) quantities $\boldsymbol{d}$ associated with uncertainties. If the set of d_x is chosen to be statistically independent, such a covariance is given by (see Annex A)

$$u(x_i, x_j) = \sum_k \frac{\partial X_i}{\partial d_k} \frac{\partial X_j}{\partial d_k} u^2(d_k) \tag{9}$$

5.5 Parameters of unfolding

Examples of parameters are spectrum parameters such as the widths of spectral lines, or detection efficiencies, radionuclide-specific data, such as emission probabilities and half-lives, or correction factors. For parameters in unfolding, it is assumed that their values and standard uncertainties have been estimated in advance according to the rules of the ISO/IEC Guide 98-3.

The elements of the matrix $\boldsymbol{A}$ may contain other quantities which can be treated as a vector $\boldsymbol{p}$ of parameters, which may be associated with uncertainties. For convenience, the estimates $\boldsymbol{x}$ and $\boldsymbol{p}$ are combined to form the column matrix $\boldsymbol{w} = (x_1,\ldots, x_m, p_1, p_2,\ldots)^{\mathrm{T}}$.

For the unfolding, one needs the estimates, $\boldsymbol{x}$ and $\boldsymbol{p}$, of the input quantities and their associated uncertainty matrix, $\boldsymbol{U_w}(\boldsymbol{x},\boldsymbol{p})$. This uncertainty matrix has been calculated as a covariance matrix based on the ISO/IEC Guide 98-3 (see References [9] and [10]). The uncertainty matrix, $\boldsymbol{U_w}(\boldsymbol{x},\boldsymbol{p})$, is needed in form of its functional dependence on $\boldsymbol{x}$ since $\boldsymbol{x}$ shall be adjusted if decision threshold and detection limit are calculated while p stays constant. The uncertainty matrices $\boldsymbol{U_x}$ and $\boldsymbol{U_p}$ associated with $\boldsymbol{x}$ and $\boldsymbol{p}$ are partial matrices of $\boldsymbol{U_w}$. The rank of $\boldsymbol{U_x}$ shall not be smaller than the number n of model Formulas. If the data for $\boldsymbol{x}$ and $\boldsymbol{p}$ originate from different independent experiments, there is no correlation between $\boldsymbol{x}$ and $\boldsymbol{p}$ and the matrix elements of $\boldsymbol{U_w}$ related to pairs x_i and p_k vanish.

An explicit model of the linear unfolding then consists of n relationships between input and output quantities. These relationships can formally and most generally be written as a column matrix $\boldsymbol{H}(\boldsymbol{y},\boldsymbol{p})$ of model functions H_k which depend on all these quantities.

$$\boldsymbol{x} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p}) \tag{10}$$

Without combining $(\boldsymbol{x}, \boldsymbol{p})$ to $\boldsymbol{w}$ the uncertainty propagation would read as follows:

$$U_y = G_w U_w G_w^{\mathrm{T}} = G_x U_x G_x^{\mathrm{T}} + G_p U_p G_p^{\mathrm{T}} \tag{11}$$

It is shown in 5.6 that for the case of a linear and explicit model the expression $G_x U_x G_x^{\mathrm{T}}$ is evaluated by a different algebraic expression originating from a least-squares analysis. The second term in the previous Formula, however, is calculated in terms of uncertainty propagation with using the transformation matrix $\boldsymbol{G_p}$ with elements $G_{l,j} = \partial Y_i / \partial p_j$. To avoid possible confusion regarding the ambiguous symbol G, this matrix is designated as $\boldsymbol{Q}$ in the following. It has to be emphasized that

calculating this matrix is not an easily performed step, because it requires building the derivative of the output quantity values, considered as a function, with respect to the components of $\boldsymbol{p}$.

$$Q_{l,j} = \frac{\partial y_l}{\partial p_j} = \frac{1}{\Delta_j}\left\{ y_l(x, p_1, ..., p_j + \Delta_j/2) - y_l(x, p_1, ..., p_j - \Delta_j/2) \right\} \tag{12}$$

For Δ_j see the second note below Formula 5. The two terms in the curly bracket demonstrate that one value of a derivative requires two evaluations for an output quantity, that means two full least squares evaluations. It follows that a calculation algorithm has to be organized such that the full least squares analysis is organized as a function which by each call delivers a value y_l of an output quantity.

5.6 Procedure for unfolding

Unfolding in multivariate measurements means in essence fitting new values, $\boldsymbol{z}$, of the input quantities, $\boldsymbol{X}$, to the given estimates, $\boldsymbol{x}$ where $\boldsymbol{z} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p})$ depends on the measurand estimates, $\boldsymbol{y}$, to be determined and on fixed given estimates, $\boldsymbol{p}$, of further input quantities which are not subject to fit. The generalized least-squares method is highly recommended for use as a spectrum unfolding procedure since it can easily be combined with the uncertainty treatment and allows for a compact and transparent description as follows.

The measurand estimates, $\boldsymbol{y}$, are determined by minimizing the quantity

$$\chi^2 = (\boldsymbol{x} - \boldsymbol{z})^T \boldsymbol{U}_{\boldsymbol{x}}^{-1}(\boldsymbol{x})(\boldsymbol{x} - \boldsymbol{z}) = \min \tag{13}$$

with the constraint $\boldsymbol{z} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p})$ and the uncertainty matrix, $\boldsymbol{U}_{\boldsymbol{x}}(\boldsymbol{x})$, given as a function of $\boldsymbol{x}$ for finally obtaining the characteristic values. The results of this minimizing procedure are the functions:

$$\boldsymbol{y} = \boldsymbol{G}(\boldsymbol{x},\boldsymbol{p}) = \boldsymbol{G}(\boldsymbol{w}) \tag{14}$$

$$\boldsymbol{z} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p}) = \boldsymbol{H}\left[\boldsymbol{G}(\boldsymbol{w}),\boldsymbol{p}\right] = \boldsymbol{F}(\boldsymbol{w}) \tag{15}$$

and, similar to Formula (6) where $\boldsymbol{w}$ now plays the part of $\boldsymbol{x}$, the due uncertainty matrices

$$\boldsymbol{U}_{\boldsymbol{y}} = \boldsymbol{G}_{\boldsymbol{w}} \boldsymbol{U}_{\boldsymbol{w}} \boldsymbol{G}_{\boldsymbol{w}}^{\mathrm{T}}; \quad \boldsymbol{U}_{\boldsymbol{z}} = \boldsymbol{F}_{\boldsymbol{w}} \boldsymbol{U}_{\boldsymbol{w}} \boldsymbol{F}_{\boldsymbol{w}}^{\mathrm{T}} \tag{16}$$

Here, $\boldsymbol{x}$ and $\boldsymbol{p}$ are combined to form the column matrix, $\boldsymbol{w}$. The uncertainty matrices, $\boldsymbol{U}_{\boldsymbol{x}}$ and $\boldsymbol{U}_{\boldsymbol{p}}$, are likewise combined to form the uncertainty matrix, $\boldsymbol{U}_{\boldsymbol{w}}$. The sensitivity matrices, $\boldsymbol{F}_{\boldsymbol{w}}$ and $\boldsymbol{G}_{\boldsymbol{w}}$, denote the matrices of the partial derivatives of the functions $\boldsymbol{F}(\boldsymbol{w})$ and $\boldsymbol{G}(\boldsymbol{w})$, respectively. All the matrices on the right-hand side of Formula (16) are functions of $\boldsymbol{w}$.

NOTE 1 The right-hand side expressions of the Formulas (6) and (16) defining uncertainty matrices represent the common uncertainty propagation in matrix notation in which the partial derivatives form linear transformation matrices $\boldsymbol{G}$ or $\boldsymbol{F}$, respectively.

The results of the fit and the given data $\boldsymbol{x}$ conform if the standardized chi-square χ^2_s meets, with $m > n$ and with the obtained minimum χ^2_{min}, the chi-square condition

$$\chi^2_s = \frac{\left|\chi^2_{min} - (m-n)\right|}{\sqrt{2(m-n)}} \leq k_{1-\delta/2} \tag{17}$$

with $k_{1-\delta/2}$ being the quantile of the standardized normal distribution for the probability $1 - \delta/2$ of a wrong decision. A probability $\delta = 0{,}05$ is recommended.

NOTE 2 Formula (17) is a frequently used, but coarse approximation which takes only into account the expectation, $E(\chi^2) = \nu$, and the variance, $\mathrm{Var}(\chi^2) = 2\nu$, of the χ^2-distribution. A good test statistic would result in the criterion such as $\left(\left(\chi^2_{min}/(m-n)\right)^{1/3} - \left(1 - \frac{2}{9(m-n)}\right)\right) \Big/ \sqrt{\frac{2}{9(m-n)}} \leq k_{1-\delta/2}$.

In many cases, the function $\boldsymbol{H}(\boldsymbol{y},\boldsymbol{p})$ to be adapted to the given estimates $\boldsymbol{x}$ is linear in $\boldsymbol{y}$, i.e., $\boldsymbol{z} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p}) = \boldsymbol{A}\boldsymbol{y}$ where the matrix $\boldsymbol{A}$ does not depend on $\boldsymbol{p}$ and can represent the spectrometer response (explicit linear multivariate model). Then $\boldsymbol{x}$ and $\boldsymbol{w}$ are identical, and the minimizing procedure of the least-squares method can easily be carried out with results in

$$\boldsymbol{y} = \boldsymbol{U}_y \boldsymbol{A}^{\mathrm{T}} \boldsymbol{U}_x^{-1}(x)x\,; \quad \boldsymbol{U}_y = \left[\boldsymbol{A}^{\mathrm{T}} \boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{A}\right]^{-1} \tag{18}$$

$$\boldsymbol{z} = \boldsymbol{A}\boldsymbol{y}\,; \quad \boldsymbol{U}_z = \boldsymbol{A}\boldsymbol{U}_y\boldsymbol{A}^{\mathrm{T}} \tag{19}$$

$$\chi^2_{min} = (\boldsymbol{x}-\boldsymbol{z})^{\mathrm{T}} \boldsymbol{U}_x^{-1}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{z})\,; \quad \tilde{\boldsymbol{U}}_y = \left[\boldsymbol{A}^{\mathrm{T}} \boldsymbol{U}_x^{-1}(\boldsymbol{A}\tilde{\boldsymbol{y}})\boldsymbol{A}\right]^{-1} \tag{20}$$

If $\boldsymbol{A}$ contains parameters $\boldsymbol{p}$ associated with uncertainties $\boldsymbol{U}_p$, Formulas (18) to (20) are extended

$$\boldsymbol{U}_y(\boldsymbol{x}) = \left[\boldsymbol{A}^{\mathrm{T}} \boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{A}\right]^{-1}; \quad \boldsymbol{y} = \boldsymbol{U}_y(\boldsymbol{x})\,\boldsymbol{A}^{\mathrm{T}} \boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{x}\,; \quad \boldsymbol{U}_y = \boldsymbol{U}_y(\boldsymbol{x}) + \boldsymbol{Q}\boldsymbol{U}_p\boldsymbol{Q}^{\mathrm{T}} \tag{21}$$

$$\boldsymbol{z} = \boldsymbol{A}\boldsymbol{y};\ \boldsymbol{U}_z = \boldsymbol{A}\left(\boldsymbol{U}_y(\boldsymbol{x}) + \boldsymbol{Q}\boldsymbol{U}_p\boldsymbol{Q}^{\mathrm{T}}\right)\boldsymbol{A}^{\mathrm{T}} \tag{22}$$

$$\chi^2_{min} = (\boldsymbol{x}-\boldsymbol{z})^{T} \boldsymbol{U}_x^{-1}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{z})\,; \quad \tilde{\boldsymbol{U}}_y = \left[\boldsymbol{A}^{T} \boldsymbol{U}_x^{-1}(\boldsymbol{A}\tilde{\boldsymbol{y}})\boldsymbol{A}\right]^{-1} + \boldsymbol{Q}\boldsymbol{U}_p\boldsymbol{Q}^{\mathrm{T}} \tag{23}$$

If $\boldsymbol{Y}$ constitutes an unfolding sub-model, another measurand, $\boldsymbol{Y}^+$, is of interest. According to 5.3, it is calculated by Formula (7) with a diagonal matrix $\boldsymbol{D}$, i.e. $y_j^+ = D_{jj}\,y_j$, or $y_j^+ = D_{jj}(y_j - y_{0,j})$. Let the elements of $\boldsymbol{D}$ now contain further input quantities (parameters) q_m associated with uncertainties $u(q_m)$ combined into a vector and a covariance matrix. If there are some quantities with uncertainties (vector $\boldsymbol{p}$, covariance matrix $\boldsymbol{U}_p$) within the elements of the design matrix (see 5.5), it is recommended to simply merge $\boldsymbol{p}$ and $\boldsymbol{U}_p$ into $\boldsymbol{q}$ and $\boldsymbol{U}_q$, respectively. For calculating the uncertainty matrix associated with $\boldsymbol{U}_{y^+}$ the partial derivatives $\partial y_j^+ / \partial y_m = J_{jm}$ and $\partial y_j^+ / \partial q_m = Q'_{jm}$ forming the elements of transformation matrices $\boldsymbol{J}$ and $\boldsymbol{Q}'$, respectively, are needed. In the second case of Formula (7), the values and uncertainties of the components of y_0 are also included in $\boldsymbol{q}$ and $\boldsymbol{U}_q$; the partial derivatives are then obtained as $J_{jm} = \frac{\partial}{\partial y_m}\left(D_{jj}(\boldsymbol{q},\boldsymbol{Y})\,y_j\right)$ and $Q'_{jm} = \frac{\partial}{\partial q_m}\left(D_{jj}(\boldsymbol{q},\boldsymbol{Y})\left(y_j - y_{0,j}\right)\right)$; J_{jm} does not depend on

$\boldsymbol{y_0}$. The following matrix Formula is finally obtained for calculating the full uncertainty matrix $\boldsymbol{U}_{y^+}$ of the desired output quantity

$$\boldsymbol{U}_{y^+} = \boldsymbol{J}\boldsymbol{U}_y\boldsymbol{J}^T + \boldsymbol{Q'}\boldsymbol{U}_q\boldsymbol{Q'}^{\mathrm{T}} \tag{24}$$

with $\boldsymbol{U}_y = \left[\boldsymbol{A}^T\boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{A}\right]^{-1}$ according to Formula (18).

For an explicit model containing a least-squares sub-model (see 5.3), one therefore obtains by analogy to Formulas (18) to (20) when the second part of Formula (7) is considered (otherwise, $\boldsymbol{y_0}$ is set to zero)

$$\boldsymbol{U}_y(\boldsymbol{x}) = \left[\boldsymbol{A}^T\boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{A}\right]^{-1};\ \boldsymbol{y}^+ = \boldsymbol{D}\left[\boldsymbol{U}_y(\boldsymbol{x})\,\boldsymbol{A}^T\boldsymbol{U}_x^{-1}(\boldsymbol{x})\boldsymbol{x} - \boldsymbol{y_0}\right]$$
$$\boldsymbol{U}_{y^+} = \boldsymbol{U}_{y^+}(\boldsymbol{x}) + \boldsymbol{U}_{y^+}(\boldsymbol{q}) = \boldsymbol{J}\boldsymbol{U}_y(\boldsymbol{x})\boldsymbol{J}^T + \boldsymbol{Q'}\boldsymbol{U}_q\boldsymbol{Q'}^{\mathrm{T}} \tag{25}$$

$$\boldsymbol{z} = \left(\boldsymbol{A}\boldsymbol{D}^{-1}\right)\left(\boldsymbol{y}^+ + \boldsymbol{D}\boldsymbol{y_0}\right) = \left(\boldsymbol{A}\boldsymbol{D}^{-1}\right)\boldsymbol{y}^+ + \boldsymbol{A}\boldsymbol{y_0};\ \ \boldsymbol{U}_z = \left(\boldsymbol{A}\boldsymbol{D}^{-1}\right)\boldsymbol{U}_{y^+}(\boldsymbol{x})\left(\boldsymbol{A}\boldsymbol{D}^{-1}\right)^{\mathrm{T}} + \boldsymbol{Z}\boldsymbol{U}_q\boldsymbol{Z}^{\mathrm{T}} \tag{26}$$

$$\chi^2_{\min} = (\boldsymbol{x}-\boldsymbol{z})^{\mathrm{T}}\boldsymbol{U}_x^{-1}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{z});\ \ \tilde{\boldsymbol{U}}_{y^+} = \boldsymbol{J}\left[\boldsymbol{A}^{\mathrm{T}}\boldsymbol{U}_x^{-1}\left(\boldsymbol{A}\boldsymbol{D}^{-1}\tilde{\boldsymbol{y}}^+\right)\boldsymbol{A}\right]^{-1}\boldsymbol{J}^{\mathrm{T}} + \boldsymbol{Q'}\boldsymbol{U}_q\boldsymbol{Q'}^{\mathrm{T}} \tag{27}$$

with the transformation matrix $\partial z_j / \partial q_m = Z_{jm}$.

NOTE 3 When calculating values of $\tilde{\boldsymbol{U}}_y$ and $\tilde{\boldsymbol{U}}_{y^+}$ the terms $\boldsymbol{Q}\boldsymbol{U}_p\boldsymbol{Q}^{\mathrm{T}}$ and $\boldsymbol{Q'}\boldsymbol{U}_q\boldsymbol{Q'}^{\mathrm{T}}$ in Formulas (23) and (27), respectively, need to be re-calculated for each new value of $\tilde{y}$ or $\tilde{y}^+$ within the iteration for deriving the detection limit value, but not in the case of calculating only partial derivatives. *Alternatively:* To be quite safe, replace the terms $\boldsymbol{Q}\boldsymbol{U}_p\boldsymbol{Q}^{\mathrm{T}}$ and $\boldsymbol{Q'}\boldsymbol{U}_q\boldsymbol{Q'}^{\mathrm{T}}$ in Formulas (23) and (27) by $\tilde{\boldsymbol{Q}}\boldsymbol{U}_p\tilde{\boldsymbol{Q}}^{\mathrm{T}}$ and $\tilde{\boldsymbol{Q}'}\boldsymbol{U}_q\tilde{\boldsymbol{Q}'}^{\mathrm{T}}$, respectively.

NOTE 4 The similarity of the two Formulas $\boldsymbol{x} = \boldsymbol{A}\boldsymbol{y}$ and $\boldsymbol{x} = \boldsymbol{A}\boldsymbol{D}^{-1}\boldsymbol{y}^+$ could suggest to reduce the solution of the second one to that of the first one by replacing the second Formula by $\boldsymbol{x} = \boldsymbol{A'}\boldsymbol{y'}$. This only works if the elements of D do not depend on elements of y, otherwise the Formula becomes non-linear not allowing a solution corresponding to Formula (21). This is the reason why the contribution by $\boldsymbol{U_q}$, considering also parameters of D, was estimated by an additional uncertainty propagation step.

NOTE 5 The above-mentioned Formulas for models being linear in $\boldsymbol{y}$ require matrix algebra as a numerical tool, which are also available in common spreadsheets calculations. If the vector $\boldsymbol{x}$ represents channel counts, or channel counting rates, of a multi-channel spectrum, the components of $\boldsymbol{x}$ are not correlated. In this case it is not necessary to work with the full uncertainty matrix $\boldsymbol{U_x}$, which can be large; using standard routines for linear weighted least squares, such as LFIT[23], would be easier to handle and are significantly faster.

NOTE 6 If it would happen that the model is non-linear another standard technique could be used, such as the Levenberg-Marquardt method routine MRQMIN[23]. If the matrix $\boldsymbol{A}$ contains parameters with associated uncertainties, this would generally represent the case of total least-squares (e.g. References [24] and [25]). However, practical experience has shown to assume that it is sufficient to use standard least-squares to obtain $\boldsymbol{y}$ and to perform then full uncertainty propagation for obtaining $\boldsymbol{U_y}$.

5.7 Modification for Poisson distributed count numbers for unfolding

Counting rates are generally derived from measured counts which are Poisson distributed and thus deviate in the low-level case from the normal distribution (compare Annex A of ISO 11929-2:2019). Applying a linear least-squares procedure as presented above, which assumes normal distributed measured input data, is known to yield biased fitting parameters. This bias however can be reduced significantly by the following simple iteration scheme of the Formula (18) (e.g. References [27] and [26]). The steps taken are:

— assume that the Formula (18) have been evaluated once with the input data $\boldsymbol{x}$ (being noisy) to yield the vector $\boldsymbol{y}$;

— a new vector x_1 (not noisy) is calculated from $\boldsymbol{Ay} = x_1$;

— replace (only) the diagonal values of the matrix $\boldsymbol{U_x}$ according to $U_x(i,i) = x_1(i)/t$;

— perform now a re-evaluation by Formula (18), with the modified $\boldsymbol{U_{x,1}}$ but taking again the original input data vector $\boldsymbol{x}$

$$\boldsymbol{U}_{y;1}(x) = \left[\boldsymbol{A}^T \boldsymbol{U}_{x;1}^{-1}(x_1)\boldsymbol{A}\right]^{-1};\ y_1 = \boldsymbol{U}_{y;1}(\boldsymbol{x})\,\boldsymbol{A}^T \boldsymbol{U}_{x;1}^{-1}(x_1)\boldsymbol{x} \tag{28}$$

The last three steps may be repeated one or two times, not more. The basic idea of the modification applied here refers to the "Pearson" type of the Chi-square expression used for minimization instead of the "Neyman" or "modified Neyman" type which is behind the "Gaussian" least-squares method; see Reference [27].

Another technique is that of fitting by Poisson maximum-likelihood estimation (PMLE); see Reference [27]. It is a non-linear fitting method and is well suited for low numbers of counts. This can be achieved by using MRQMIN being adapted for PMLE; see Reference [28].

5.8 Evaluation of the primary results and their associated standard uncertainties

The calculation of partial derivatives of the output quantities with respect to input parameters can be put into a simpler form by introducing a function, which calculates the output vector by taking all actual values of the input quantities into account, such $\boldsymbol{x}$, $\boldsymbol{p}$, or $\boldsymbol{q}$; assume now that these input quantities form a vector $\boldsymbol{v}$. The following scheme shows, as a symbolic function ***Result***, the sequence of calculations necessary for establishing the vector $\tilde{y}^+$. Such a function would also be required for calculating an uncertainty budget.

The following scheme shows as a symbolic function ***Result*** the sequence of calculations necessary for establishing the vector $\tilde{y}^+$ and using the value its component k.

```
function Result (k, v, Ux, Up or Uq)
    calculate matrices A, D as functions of v ;
    invert (locally) the matrix Ux ;
    calculate the matrix Matmul(Transpose(A), Matmul(Ux^-1, A)) and invert it then to
    obtain Uy (x);
    obtain vector y = Matmul(Uy (x), Matmul(Transpose(A), Matmul(Ux^-1, x)));
    Result = y(k)
    if y is the sub-model: calculate the result vector as y+= Matmul(D, y) ;
                Result = y+(k)
end function Result
```

With using such a function, any partial derivative can be obtained as simply as follows

function value for unmodified input quantities:

```
Fv1 = Result(k, v, Ux, Up or Uq);
```

a loop now follows if partial derivatives are to be calculated for several input quantities:

loop

modify a single one of input quantities, $v(j)$, by adding a small amount Δ_j to $v(j)$:

```
Fv2 = Result(k, {v(1),..., v(j) + Δj,...}, Ux, Up or Uq);
```

partial derivative with respect to parameter $v(j)$ is the quotient:

$(Fv2 - Fv1) / \Delta_j$;

restore the unmodified parameter vector $\mathbf{v}$;

end loop

This represents an asymmetric form of the differential quotient and requires only one additional calculation per derivative. It is recommended to use small values of Δ_j, such as 2.E-6 $v(j)$) in double precision arithmetic.

Thus, one obtains the vector y_j^+ of primary measurement results. The standard uncertainties associated with y_j^+ are given by $u(y_j^+) = \sqrt{U_{y,jj}}$.

5.9 Standard uncertainty as a function of an assumed true value of the measurand

It is necessary to derive the uncertainty, $\tilde{u}(\tilde{y})$, of an assumed true value, $\tilde{y}$, as a function, which then is needed for calculating the decision threshold and the detection limit. In the multivariate case considered here, it is not possible to derive such an uncertainty function for all components of $\boldsymbol{Y}$ simultaneously; instead, they can be derived only for those individual components of $\boldsymbol{Y}$ which are of interest. Therefore, this function refers to a single component, the quantity of interest, e.g. y_j, of $\boldsymbol{Y}$. In the expression for $\tilde{u}(\tilde{y})$ the assumed true value $\tilde{y}$ is then obtained by replacing the component y_j in the vector $\boldsymbol{Y}$ by an assumed true value $\tilde{y}$ for j =1 yielding $\tilde{y} = (\tilde{y}, \max(0, y_2), ..., \max(0, y_n))^T$ .which is now used instead of $\boldsymbol{y}$.

NOTE Without the modification of restricting the other components of $\boldsymbol{Y}$ to positive values, a negative fitted y_k would imply for the output quantity of interest (e.g. y_j), with $k \neq j$, the effect of having lowered the subtracted background and interference to y_j; thereby, the uncertainty of y_j would be underestimated. This modification applies only if y_k represents an activity for which negative values are not meaningful.

By a single multivariate evaluation, this leads to $\tilde{z} = \boldsymbol{H}(\tilde{\boldsymbol{y}}, \boldsymbol{p})$, i.e. simultaneously modified input quantities replacing the column matrix $\boldsymbol{x}$. Thus, the column matrix $\tilde{w}$ follows as the combination of $\tilde{z}$ and $\boldsymbol{p}$ and, moreover, the uncertainties associated with the modified input quantity values, given by $\tilde{U}_w$, follow as the combination of $U_x(\tilde{z})$ and U_p. The matrices $\tilde{\boldsymbol{w}}$ and $\tilde{\boldsymbol{U}}_{\boldsymbol{w}}$ are used in Formula (11) instead of $\boldsymbol{w}$ and U_w to calculate $\tilde{U}_y$. Finally, the square root of the (1,1)-element of this uncertainty matrix is the needed function $\tilde{u}(\tilde{y})$.

Similarly, in the case of an explicit model which is linear with respect to $\boldsymbol{Y}$, the single multivariate evaluation leads according to Formula (22) to $\tilde{x} = \tilde{z} = A(p)\tilde{y}$, with $\boldsymbol{A}$ being dependent on parameters $\boldsymbol{p}$, while the uncertainty matrix $U_x(\tilde{z})$ associated with the modified input quantity values is calculated according to 5.6. Then, the matrices $\tilde{z}$ and $U_x(\tilde{z}) = U_x(A\tilde{y})$ are used in Formula (23) instead of $\boldsymbol{x}$ and U_x to calculate $\tilde{U}_y$ yielding the (k,k)-element of this uncertainty matrix as the needed function $\tilde{u}(\tilde{y}_k)$.

If the multivariate evaluation model has a sub-model (see 5.3), then the parameters $\boldsymbol{p}$ and $\boldsymbol{q}$ with uncertainties $\boldsymbol{U_p}$ and $\boldsymbol{U_q}$, respectively, are merged into a common vector $\boldsymbol{q}$ (ordering: $\boldsymbol{p}, \boldsymbol{q}$) and a common uncertainty matrix $\boldsymbol{U_q}$. Formula (26) then leads to $\tilde{x} = \tilde{z}$ while $U_x(\tilde{z})$ is calculated according to 5.4, $\tilde{\boldsymbol{U}}_y$ refers to the sub-model and is calculated as just described. $\tilde{\boldsymbol{U}}_y$ is then inserted into Formula (27) and the square root of the (k,k)-element of $\tilde{\boldsymbol{U}}_{y^+}$ represents a value of the uncertainty function $\tilde{u}\left(\tilde{y}_k^+\right)$.

The following scheme shows the sequence of calculations necessary for establishing the uncertainty function $\tilde{u}\left(\tilde{y}_k^+\right)$ as a symbolic function ***Utilde***.

function Utilde ($\tilde{y}^+$, k)

calculate matrices $\boldsymbol{A}$, $\boldsymbol{D}$ as a function of $\boldsymbol{q}$;

replace negative components $\neq k$ of $\tilde{y}^+$ by 0;

calculate $\tilde{x} = \boldsymbol{A}\boldsymbol{D}^{-1}\tilde{y}^+$;

calculate $\tilde{U}_{\tilde{x}}$ (see 5.4), e.g. by $u^2(x_i) = \frac{x_i}{t_c} + \frac{x_0}{t_c} + u^2(x_0)$ and $u(x_i, x_j) = u^2(x_0) = x_0 / t_0$;

invert matrix $\tilde{U}_{\tilde{x}}$

calculate the matrix Matmul(Transpose(**A**), Matmul($\tilde{\boldsymbol{U}}_{\tilde{x}}^{-1}$, **A**)) and invert it then to obtain $\tilde{\boldsymbol{U}}_{\tilde{y}}(\tilde{\boldsymbol{x}})$;

obtain y = Matmul($\tilde{\boldsymbol{U}}_{\tilde{y}}(\tilde{\boldsymbol{x}})$, Matmul (Transpose(**A**), matmul($\tilde{\boldsymbol{U}}_{\tilde{x}}^{-1}$, $\tilde{\boldsymbol{x}}$)))

calculate the matrix $\boldsymbol{Q}'$ of partial derivatives $\partial y_j^+ / \partial q_m = Q'_{jm}$;

calculate the matrix $\boldsymbol{J}$ of partial derivatives $\partial y_j^+ / \partial y_m = J_{jm}$;

calculate $\tilde{U}_{\tilde{y}^+}$ as

Matmul (**J**, Matmul($\tilde{U}_{\tilde{y}}(\tilde{x})$, Transpose (**J**)))+

Matmul (**Q′**, Matmul($\boldsymbol{U_q}$, Transpose(**Q′**)));

take $\sqrt{\tilde{U}_{\tilde{y}^+,k,k}}$ as the value ***Utilde*** for the component k.

end function Utilde

5.10 Decision threshold, detection limit and assessments

5.10.1 Specifications

The probability, α, of a wrong decision in favour of the presence of the physical effect investigated, the probability, β, of a wrong decision in favour of the absence of the physical effect investigated and the probability, $1 - \gamma$, for the coverage interval shall be specified. The choice depends on the application. A frequently cited choice is $\alpha = \beta$ and the value 0,05 for α and β. Then, $k_{1-\alpha} = k_{1-\beta} = 1{,}65$. For the coverage interval the probability $\gamma = 0{,}05$ is frequently chosen. If this is the case, then $k_{1-\gamma/2} = 1{,}96$.

If it is to be assessed whether or not a measurement procedure for the measurand satisfies the requirements to be fulfilled for scientific, legal or other reasons, a guideline value, y_{rj}, as a value of the measurand, for instance, an activity, shall also be specified.

5.10.2 Decision threshold

Decision threshold and detection limit in multivariate measurements always refers to a single component *j* of the vector y^+ (or y). If uncertainties can be evaluated according to the ISO/IEC Guide 98-3, the decision threshold for the component *j*, y_j^{+*}, is defined by (see ISO 11929-1:2019, 8.2)

$$y_j^{+*} = k_{1-\alpha}\sqrt{\tilde{U}_{\tilde{y}^+,jj}(\tilde{x})} \tag{29}$$

The value $\sqrt{\tilde{U}_{\tilde{y}^+,jj}(\tilde{x})}$ is derived with using the uncertainty function explained in 5.9 and setting $\tilde{\boldsymbol{x}} = (\boldsymbol{A}\boldsymbol{D}^{-1})\tilde{y}^+$, $\tilde{y}^+ = \left(\max\left(0, y_1^+\right), ..., \tilde{y}_j^+ = 0, ..., \max\left(0, y_n^+\right)\right)$.

5.10.3 Detection limit

By analogy to ISO 11929-1, the detection limit associated with the component *j* of the vector y^+ (or y), $y_j^{+\#}$, is defined as the solution of the implicit Formula

$$y_j^{+\#} = y_j^{+*} + k_{1-\beta}\sqrt{\tilde{U}_{\tilde{y}^+,j,j}(\tilde{x})} \tag{30}$$

with $\tilde{\boldsymbol{x}} = (\boldsymbol{A}\boldsymbol{D}^{-1})\tilde{y}^+$ and $\tilde{y}^+ = \left(\max\left(0, y_1^+\right), ..., \tilde{y}_j^+ = y_j^{+\#}, ..., \max\left(0, y_n^+\right)\right)$. Again, the square root value is estimated by the uncertainty function explained in 5.9. Formula (30) is an implicit Formula requiring solution by iteration because the value $y_j^{+\#}$ is also used within the term under the square root.

5.10.4 Assessments

The primary measurement result, *y*, has to be compared with the decision threshold, y^*. If the primary measurement result, *y*, exceeds the decision threshold, y^*, it is decided to conclude that the physical effect provided by the measurand is present, i.e. that a contribution from the sample has been recognized.

If the result, *y*, is below the decision threshold, y^*, it is decided to conclude that the result cannot be attributed to the physical effect. Nevertheless, it cannot be concluded that it is absent. If the physical effect is really absent, the probability of taking the wrong decision, that the effect is present, is equal to the specified probability, α.

The decision on whether or not a measurement procedure to be applied sufficiently satisfies the requirements regarding the detection of the physical effect quantified by the measurand is made by comparing the detection limit, $y^\#$, with the specified guideline value, y_r. If $y^\# > y_r$, the measurement procedure is not suitable for the intended measurement purpose with respect to the requirements.

5.11 Coverage interval and the best estimate and its associated standard uncertainty

5.11.1 General aspects

For more than one output quantity their uncertainties are correlated, and coverage ellipsoids may be constructed. They are, however, not considered in this standard. The coverage of a single output quantity is derived from its marginal distribution, which is obtained by integrating out the remaining output quantities in their joint distribution. Then, the coverage intervals described in the following are applicable.

The limits of the coverage interval are provided for a physical effect, recognized as present according to 5.10.2, limit the coverage interval in such a way that it contains the true value of the measurand with the specified probability $1 - \gamma$ (see ISO 11929-1:2019, 9.1). The limits of the coverage interval take into account the fact that the measurand is non-negative (see ISO 11929-2:2019, Annex B).

There is no unique definition for the coverage interval if only the condition $1 - \gamma$ is given. Further conditions are required which lead among others to the definitions of the probabilistically symmetric coverage interval and the shortest coverage interval. For the calculation of the limits of both types of coverage intervals the ISO 11929 series provides formulas for the case that uncertainties can be evaluated according to the ISO/IEC Guide 98-3.

NOTE For the purpose of radiation protection the regulator has to decide which type of coverage interval shall be used. When comparing upper limits of the two coverage intervals one has to take into account that they might have different probabilities.

A coverage interval in multivariate measurements always refers to a single component j of the measurand vector Y and is based on primary result y_j^+ and its associated standard uncertainty $u(y_j^+) = \sqrt{U_{y,jj}(x)}$ given the primary results of the other components of Y. Consequently, the calculation of a coverage interval has to be performed as stipulated in ISO 11929-1:2019, Clause 9 which is repeated in the following.

5.11.2 The probabilistically symmetric coverage interval

With a primary measurement result, y, of the measurand and the standard uncertainty, $u(y)$, associated with y, the lower limit of the probabilistically symmetric coverage interval, $y^{\triangleleft}$, and the upper limit of the probabilistically symmetric coverage interval, $y^{\triangleright}$, are calculated by

$$y^{\triangleleft} = y - k_p \cdot u(y) \quad \text{with} \quad p = \omega \cdot (1 - \gamma/2) \tag{31}$$

$$y^{\triangleright} = y + k_q \cdot u(y) \quad \text{with} \quad q = 1 - \omega \cdot \gamma/2 \tag{32}$$

where

$$\omega = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{y/u(y)} \exp\left(-\frac{v^2}{2}\right) \mathrm{d}v = \Phi\left[y/u(y)\right] \tag{33}$$

For the distribution function, $\Phi(t)$, of the standardized normal distribution and for its inversion, $k_p = \Phi^{-1}(p)$, see ISO 11929-1:2019, Table E.1. For methods for its calculation, see ISO 11929-1:2019, E.1 or, for instance, Reference [23].

In general, the limits of the probabilistically symmetric coverage interval are located neither symmetrical to y, nor to the best estimate, $\hat{y}$, but the probabilities of the measurand being smaller than $y^{\triangleleft}$ or larger than $y^{\triangleright}$ both equal $\gamma/2$. The relations $0 < y^{\triangleleft} < y^{\triangleright}$ apply.

$\omega = 1$ may be set if $y > 4u(y)$. In this case, the following approximations symmetrical to y apply:

$$y^{\triangleleft} = y - k_{1-\gamma/2} \cdot u(y) \quad \text{and} \quad y^{\triangleright} = y + k_{1-\gamma/2} \cdot u(y) \tag{34}$$

5.11.3 The shortest coverage interval

As described in detail in Reference [13], the lower limit of the shortest coverage interval, $y^{<}$, and the upper limit of the shortest coverage interval, $y^{>}$, are calculated from a primary measurement result, y, of the measurand and the standard uncertainty, $u(y)$, associated with y, either by

$$y^{<>} = y \pm k_p \cdot u(y);\ p = (1+\omega\cdot(1-\gamma))/2 \tag{35}$$

or if $y^{<} < 0$, by

$$y^{<} = 0;\ y^{>} = y + k_q \cdot u(y);\ q = 1-\omega\cdot\gamma \tag{36}$$

with ω given by Formula (33). The relations $0 \le y^{<} < y^{>}$ apply and the approximation of Formula (34) is valid.

5.12 Documentation

The content of the test report depends on the specific application as well as on demands of the customer or regulator.

Independently of this, information shall be retained in order to justify the data of the test report and to guarantee traceability. This applies in particular to:

a) a reference to this document, i.e. ISO 11929:2019;

b) the physical effect of interest, measurands and model of the evaluation;

c) the probabilities α and β of a false positive and a false negative decision, respectively, and, if necessary, the guideline values, y_r;

d) the vectors of primary measurement results, $\boldsymbol{y}$, and the standard uncertainties, $u(y)$, associated with $\boldsymbol{y}$;

e) the vector of decision thresholds, y^*;

f) the vector of detection limits, $y^\#$;

g) a statement, if necessary, as to whether or not the measurement procedure is suitable for the intended measurement purpose;

h) a statement as to whether or not the different physical effects are recognized as being present;

NOTE 1 If the physical effect is not recognized as being present given the probability α, i.e. if $y < y^*$ (see 8.4), it is occasionally demanded by the regulator to document $<y^\#$ instead of the measured result, y. Such documentation can be meaningful since it allows, by comparison with the guideline value, to demonstrate that the measurement procedure is suitable for the intended measurement purpose. It is, however, misleading because the mathematical meaning is not correct.

NOTE 2 Occasionally, it is requested by the customer or regulator to compare the primary measurement result, y, with the detection limit, $y^{\triangleleft}$, in order to decide whether the physical effect is recognized or not. Such stipulations are not in accordance with the ISO 11929 series. They have the consequence that it is decided too frequently that there the physical effect is absent when in fact it is not absent.

i) the physical effect, if recognized as being present, the lower limit of the symmetric coverage interval, $y^{\triangleleft}$, and the upper limit of the symmetric coverage interval, $y^{\triangleright}$, with the probability, $1 - \gamma$, for the coverage interval, best estimate, $\hat{y}$, of the measurand, and standard uncertainty, $u(\hat{y})$ associated with $\hat{y}$.

NOTE 3 Alternatively, the lower limit of the shortest coverage interval, $y^{<}$, and the upper limit of the shortest coverage interval, $y^{>}$, with the probability, $1 - \gamma$, for the coverage interval, the best estimate, $\hat{y}$, of the measurand, and the standard uncertainty, $u(\hat{y})$ associated with $\hat{y}$ can be documented.

Annex A
(informative)

Correlations and covariances

A.1 Definitions

Assume a measurement model with input quantities X_i, for which best estimates x_i and associated standard uncertainties $u(x_i)$ are available. If any pair X_i and X_j is related (dependent), the strength of this relation is specified as a covariance or a correlation. If the pair of quantities is unrelated (independent), their covariance is zero [ISO/IEC Guide 98-3, 4.4].

The covariance of best estimates x_i and x_j of X_i and X_j, respectively, is denoted as $u(x_i, x_j)$ or as $\mathrm{cov}(x_i, x_j)$. It may be expressed by the coefficient of correlation $r = r(x_i, x_j)$ as [ISO/IEC Guide 98-3:1995 5.2.2]

$$u\left(x_i, x_j\right) = r(x_i, x_j)\, u\left(x_i\right) u\left(x_j\right) \tag{A.1}$$

For two continuous random variables X_1 and X_2 for which a joint (multivariate) probability distribution function $g_X\left(\xi\right)$ exists, where $X = (X_1, X_2)^{\mathrm{T}}$ and $x = \left(\tilde{x}_1, \tilde{x}_2\right)^{\mathrm{T}}$, their covariance is given by [ISO/IEC Guide 98-3-1, 3.10]

$$\mathrm{Cov}\left(X_1, X_2\right) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left[\tilde{x}_1 - \mathrm{E}\left(X_1\right)\right]\left[\tilde{x}_2 - \mathrm{E}\left(X_2\right)\right] g_X(\tilde{x})\,\mathrm{d}\tilde{x}_1\,\mathrm{d}\tilde{x}_2 \tag{A.2}$$

A.2 Calculation of covariances

Consider now that a pair of variables is correlated by being dependent on other input quantities common to both. Looking for a way of deriving their covariance it is helpful to consider first the generalized uncertainty propagation for a pair of output quantities Y_l and Y_k, which depend on input quantities $\mathbf{X}$

$$u\left(Y_l, Y_k\right) = \sum_{i=1}^{n}\sum_{j=1}^{n}\frac{\partial Y_l}{\partial X_i}\frac{\partial Y_k}{\partial X_j}u\left(X_i, X_j\right) \tag{A.3}$$

This means that a covariance of output quantities Y_l and Y_k is expressed as a summation of variance and covariance terms of the input quantities $\mathbf{X}$. However, the values $u(X_i, X_j)$ are still unknown. Now, Formula (A.3) is used to derive this unknown variance for a pair X_l and X_k. The term $u(X_l, X_k)$ formally replaces the left side of Formula (A.3). Assuming then that X_l and X_k depend on a set of input quantities denoted as $\mathbf{Z}$ leads to the Formula

$$u\left(X_l, X_k\right) = \sum_{i=1}^{n}\sum_{j=1}^{n}\frac{\partial X_l}{\partial Z_i}\frac{\partial X_k}{\partial Z_j}u\left(Z_i, Z_j\right) \tag{A.4}$$

With a further assumption, that the components of $\mathbf{Z}$ are mutually independent, covariances $u(Z_i, Z_j)$ vanish for $i \neq j$ and this Formula reduces to

$$u\left(X_l, X_k\right) = \sum_{i=1}^{n}\frac{\partial X_l}{\partial Z_i}\frac{\partial X_k}{\partial Z_i}u^2\left(Z_i\right) \tag{A.5}$$

Formula (A.5) finally represents the common method of deriving covariances of pairs of dependent quantities X_l and X_k. Note that often some of the input quantities $\mathbf{X}$ also belong to $\mathbf{Z}$.

A.3 Example

As an example, consider the following Formula for an estimate of q being a ratio of two differences

$$q=\frac{x_1-x_\mathrm{T}}{x_2-x_\mathrm{T}}=\frac{y_1}{y_2} \tag{A.6}$$

It is often observed that the uncertainty propagation is based on y_1 and y_2 as pre-aggregated input quantity estimates, to which uncertainties $u(y_1)$ and $u(y_2)$ are assigned.

$$u_{\mathrm{rel}}^2(q)=u_{\mathrm{rel}}^2(y_1)+u_{\mathrm{rel}}^2(y_2) \tag{A.7}$$

This may introduce a "hidden" covariance; to check for this possibility one has to look for estimates of such input quantities, which could be common to both, y_1 and y_2. x_T represents an estimate of such a quantity. The uncertainty $u(x_\mathrm{T})$ is therefore used twice, in $u(y_1)$ and $u(y_2)$, respectively. The covariance $u(X_l, X_k)$ solves this problem. It is evaluated according to Formula (A.5) and formally yields

$$u(y_1,y_2)=\sum_{i=1}^{n}\frac{\partial y_1}{\partial z_i}\frac{\partial y_2}{\partial z_i}u^2(z_i) \tag{A.8}$$

As there is only one variable-estimate common to y_1 and y_2, i.e. x_T, the sum in Formula (A.7) has only one term and $z_1 = x_\mathrm{T}$. This yields

$$u(y_1,y_2)=\frac{\partial y_1}{\partial z_1}\frac{\partial y_2}{\partial z_1}u^2(z_1)=\frac{\partial(x_1-x_T)}{\partial x_T}\frac{\partial(x_2-x_T)}{\partial x_T}u^2(x_T)=(-1)(-1)u^2(x_T)=u^2(x_T) \tag{A.9}$$

Now, according to Formula (A.4) the correct version of Formula (A.7) is obtained by adding a third term to it

$$u_{\mathrm{rel}}^2(q)=u_{\mathrm{rel}}^2(y_1)+u_{\mathrm{rel}}^2(y_2)+2\frac{1}{q^2}\frac{\partial q}{\partial y_1}\frac{\partial q}{\partial y_2}u(y_1,y_2)$$

Evaluating the partial derivatives $\partial q/\partial y_1 = q/y_1$ and $\partial q/\partial y_2 = -q/y_2$, finally yields

$$u_{\mathrm{rel}}^2(q)=u_{\mathrm{rel}}^2(y_1)+u_{\mathrm{rel}}^2(y_2)-2\frac{u(y_1,y_2)}{y_1\,y_2}=u_{\mathrm{rel}}^2(y_1)+u_{\mathrm{rel}}^2(y_2)-2\frac{u^2(x_T)}{y_1\,y_2} \tag{A.10}$$

The same result would be obtained by doing the uncertainty propagation according to Formula (A.4) with the values of the primary input quantities x_1, x_2 and x_T, and without using a covariance.

This demonstrates that if the uncertainty propagation were based on the primary input values x_1, x_2 and x_T, its calculation would have been a bit longer but would lead to the correct uncertainty. Using instead functions of pre-aggregated primary input quantity values, like y_1 and y_2, and their derived uncertainties, $u(y_1)$ and $u(y_2)$, for uncertainty propagation, would have been easier but would include the uncertainty $u(x_\mathrm{T})$ erroneously two times.

A.4 Rules for covariances

Formula (A.5) can be used for deriving rules for covariance calculations if quantities X and Y are multiplied by constants a or b. Some basic rules are given by the following relations:

$$u(X,Y)=u(Y,X) \tag{A.11}$$

$$u(aX,bX)=abu(X,X)=abu^2(X) \tag{A.12}$$

$$u(aX,bY)=abu(X,Y) \tag{A.13}$$

$$u(X,Y+Z)=u(X,Y)+u(X,Z) \tag{A.14}$$

$$u(X+Y,Z)=u(X,Z)+u(Y,Z) \tag{A.15}$$

A.5 Uncertainty propagation and linear transformations

The concept behind estimating (unknown) uncertainties of output quantities $\boldsymbol{Y}$ from known uncertainties of the input quantities $\boldsymbol{X}$ may be considered as a linear transformation of variables.

For this purpose the variables are collected in an n-vector $\boldsymbol{X}$ and an m-vector $\boldsymbol{Y}$ and the (m x m)-covariance matrix $\boldsymbol{U_X}$ is required. A Taylor-series of the y_i around the means of x_i restricted to the first derivative results in a linear transformation

$$\boldsymbol{Y}=\boldsymbol{BX} \tag{A.16}$$

where $\boldsymbol{B}$ is an (m × n) -matrix with elements $B_{ik}=\partial Y_i/\partial X_k$. Elementary linear transformation mathematics leads to the following matrix relation[33][34]

$$\boldsymbol{U_Y}=\boldsymbol{BU_XB}^T \tag{A.17}$$

The T raised to the exponent indicates the transposed version of the matrix $\boldsymbol{B}$. With

$$\boldsymbol{B}=\begin{pmatrix} \partial Y_1/\partial X_1 & \partial Y_1/\partial X_2 & \cdots & \partial Y_1/\partial X_n \\ \partial Y_2/\partial X_1 & \partial Y_2/\partial X_2 & \cdots & \partial Y_2/\partial X_n \\ & \vdots & & \\ \partial Y_m/\partial X_1 & \partial Y_m/\partial X_2 & \cdots & \partial Y_m/\partial X_n \end{pmatrix} \tag{A.18}$$

the matrix Formula (A.17) turns out to be equivalent to Formula (A.3) when writing up the matrix multiplication summations

$$u(Y_l,Y_k)=\sum_{i=1}^{n}\sum_{j=1}^{n}\frac{\partial Y_l}{\partial X_i}\frac{\partial Y_k}{\partial X_j}u(X_i,X_j) \tag{A.19}$$

Formulas of the form of Formula (A.17), which represent the uncertainty propagation in matrix form, are often used in linear unfolding.

NOTE 1 See also ISO/IEC Guide 98-3-2:2008, Clause 3.

NOTE 2 The matrix with elements $u(X_i, X_j)$ may be termed uncertainty matrix (ISO/IEC Guide 98-3-1) or elements of the measurement covariance matrix (ISO/IEC Guide 98-3-2).

Annex B
(informative)

Spectrum unfolding in nuclear spectrometric measurement

B.1 General aspects

For the unfolding of a measured multi-channel spectrum, one fits functions $\boldsymbol{H}(\boldsymbol{y},\boldsymbol{p})$ according to Formula (10) to the estimates $\boldsymbol{x}$ of the m input quantities $\boldsymbol{X}$, for instance to the measured values $x_i = n_i/t$ of the spectral density calculated from the channel counts n_i. The model may contain values of parameters, $\boldsymbol{p}$, associated with uncertainties (e.g. location and line-shape parameters).

The calculation of the estimates y of the output quantities, $\boldsymbol{Y}$, of the uncertainty matrix, $\boldsymbol{U_y}$, associated with $\boldsymbol{y}$ and of the fitted values $\boldsymbol{z}$ (best estimate), of the input quantities, $\boldsymbol{X}$, from the given measured and estimated values of all input quantities, $\boldsymbol{w}$, with their associated uncertainty matrix, $\boldsymbol{U_w}$, requires in general a non-linear fitting procedure.

In the special case of a spectrum unfolding which is linear in the parameters $\boldsymbol{Y}$, the spectral density $H(\vartheta_i, \boldsymbol{p})$ is represented by the column matrix $\boldsymbol{X} = [H(\vartheta_i, \boldsymbol{p})]$. The ϑ_i are assumingly exact base points, for instance the energies or times assigned to the individual channels. In the linear case considered here, such values of parameters $\boldsymbol{p}$ which would result in a non-linear behaviour, if included in $\boldsymbol{y}$, need to be held fixed at values $\boldsymbol{p}_c$ obtained by preceding calibrations. The spectral density is approximated by a system of functions $L_k(\vartheta_i, \boldsymbol{p}_c)$

$$X_i = H\left(\vartheta_i, p_c\right) = \sum_{k=1}^{n} L_k\left(\vartheta_i, p_c\right) \cdot Y_k \;;\; (i = 1,\ldots,m) \text{ or } \boldsymbol{X} = \boldsymbol{AY} \tag{B.1}$$

With fixed parameter values, the constant response matrix A consists of the elements $A_{ik} = L_k(\vartheta_i, \boldsymbol{p}_c)$ which describe the shapes of the individual spectral lines and of the background contributions.

The output quantities, Y_k, to be determined are for instance the net peak areas of spectral lines, or a step-function height or the amplitudes of polynomial background contributions. They form the column matrix y of the measurand estimates.

Some of the parameters $\boldsymbol{p}$ may not be known exactly. Starting from estimates, they are likewise to be determined by the unfolding. Consequently, these unknown quantities are to be added to the output quantities, $\boldsymbol{y}$. Then, $\boldsymbol{A}$ depends also on y and one obtains the case, which is usual in complex gamma-spectrometry, of a non-linear model according to Formula (10) with $\boldsymbol{x} = \boldsymbol{H}(\boldsymbol{y},\boldsymbol{p}) = \boldsymbol{A}(\boldsymbol{y},\boldsymbol{p})\boldsymbol{y}$.

The functions $L_k(.)$ are the response functions of the spectrometer which can, for instance, be a semiconductor detector or a grid ionisation chamber in alpha-spectrometry, but also a semiconductor detector in gamma-spectrometry or a Bonner sphere in neutron spectrometry. Mathematically, they can be nearly arbitrarily chosen and therefore they can be set up as required for phenomenological or physical reasons. They can also be measured functions or calculated ones which reflect the underlying physical processes. They can be known as analytical expressions as well as numerical. With these response functions, it is not only possible to describe shapes of spectral lines. Also, the background under spectral lines can be modelled by superposition of such functions in any arbitrary way.

B.2 Gamma-ray spectrometry — Evaluation of a single peak by linear background subtraction

One elementary example of an evaluation of a gamma-spectrum is given here, in which events of a single undisturbed line with a known location in the spectrum are to be detected and a linear background

subtraction is sufficient to determine the net counts in the line. Such a linear background can often be assumed in gamma-spectrometry.

It is suitable for the background determination to introduce three adjacent channel regions, A1, B and A2, in the following way. The central region B comprises all the channels belonging to the line and has the total content, n_g, and the width, b. Assuming a Gaussian line shape with the full width, h, at half-maximum, region B shall be placed as symmetrically as possible over the line. For an evaluation by the common total peak area method, b is usually adjusted to the actual peak height and may require values larger than $b = 2{,}5h$, for large peak areas. Both, the net peak area and the background area, are determined from b. b is part of the peak evaluation model; the values of decision threshold and detection limit also depend on it.

NOTE 1 The method of peak fitting, however, uses the whole region, i.e., (A1, B, A2), for determining the net peak area and the background area simultaneously. While the net peak area does not depend on b, it is a matter of convention to define a value of b also in this case, from which a background area is determined, which leads to decision threshold and detection limit values. b is chosen as roughly comparable to that of the total peak area method, such as $b = 2{,}5h$, by which a portion of $f = 0{,}997$ of the peak area is covered. It is recommended to use this value also in the case of an absent peak, for which a detection limit is to be reported.

The full width h at half-maximum shall be determined under the same measurement conditions by means of a Reference sample emitting the line to be investigated strongly enough, or from neighbouring lines with comparable shapes and widths. Region B shall comprise an integer number of channels, so that b is rounded up accordingly.

Regions A1 and A2, bordering region B below and above, shall be specified with the same widths, $l = l_1 = l_2$. The total width, $l_1 + l_2 = 2l$, shall be chosen as large as possible, but at most so large that the background shape over all regions can still be taken as approximately linear. n_1 and n_2 are the total contents of all channels of regions A1 and A2, respectively. Expressing now the total counts of the linear background under the peak as the product of the width, b, given in channels, and the average background per channel, $(n_1 + n_2)/(2l)$ yields

$$n_0 = b\frac{(n_1 + n_2)}{2l} \tag{B.2}$$

The following calculations are based on ISO 11929-1:2019, Annex D. The counting rate, x_{net}, of the net peak area obtained during a counting duration t_g and its associated uncertainty are given as

$$x_{\mathrm{net}} = \frac{n_g}{t_g} - \frac{n_0}{t_g} \tag{B.3}$$

$$u^2(x_{\mathrm{net}}) = \frac{n_g}{t_g^2} + \frac{n_0}{t_g^2} = \frac{n_g}{t_g^2} + \left(\frac{b}{2lt_g}\right)^2 (n_1 + n_2) = \frac{n_g}{t_g^2} + \left(-\frac{n_0}{t_g^2} + \frac{n_0}{t_g^2}\right) + \frac{n_0}{t_g^2}\frac{b}{2l} = \frac{x_{\mathrm{net}}}{t_g} + \frac{n_0}{t_g^2}\left(1 + \frac{b}{2l}\right) \tag{B.4}$$

Comparing Formula (B.4) with the standardized form of a net count rate variance, see ISO 11929-1:2019, Formula (D.5)

$$u^2(x_{\mathrm{net}}) = a_1 x_{\mathrm{net}} + a_0 \tag{B.5}$$

yields the coefficients, a_0 and a_1, from which decision threshold and detection limit values can be evaluated directly (x_0 is now the trapezoidal background counting rate)

$$a_1 = \frac{1}{t_g},\ a_0 = \frac{n_0}{t_g^2}\left(1 + \frac{b}{2l}\right) = \frac{x_0}{t_g}\left(1 + \frac{b}{2l}\right) \tag{B.6}$$

NOTE 2 The expression in the bracket of Formula (B.6) is considered as a "design factor" of this classical peak evaluation method, i.e. of the Total Peak Area (TPA) method. This factor, which is independent of the peak area in the TPA method, changes, however, if the peak fitting method would be used. It is assumed that $(1 + b/(2l))$ then changes to a function $f_B(.)$.

Following ISO 11929-1:2019, Annex D and assuming a value w of the calibration factor, the value of the decision threshold can be calculated as follows

$$y^* = k_{1-\alpha}\tilde{u}(0) = k_{1-\alpha} w \sqrt{a_0} \tag{B.7}$$

$$y^* = k_{1-\alpha} w \sqrt{a_0} = k_{1-\alpha} w \sqrt{\frac{x_0}{t_g}\left(1+\frac{b}{2l}\right)} \tag{B.8}$$

Under the assumption $k_{1-\alpha} = k_{1-\beta} = k$ the detection limit value is calculated according to Annex D as

$$y^{\#} = \frac{2y^*\psi}{\theta} = \frac{2y^* + k^2 w/t_g}{1-k^2 u_{\mathrm{rel}}^2(w)} \tag{B.9}$$

where the two parameters used from ISO 11929-1:2019, Annex D are

$$\theta = 1 - k_{1-\beta}^2 u_{\mathrm{rel}}^2(w)\ ,\ \psi = 1 + \frac{k_{1-\beta}^2}{2y^*}(wa_1) \tag{B.10}$$

B.3 Gamma-ray spectrometry — Fitting a single peak

B.3.1 General aspects

In gamma-ray spectrometry with high-purity Germanium detectors the detection process is energy-dispersive. Therefore, an energy spectrum in the form of a multi-channel pulse height spectrum is obtained by a measurement. It consists of a background continuum (Compton continuum), slowly varying with energy, and superimposed to it several rather narrow peaks due to gamma-rays leaving their full energy by the photo effect in the Germanium crystal. The peaks in most cases are well isolated from each other; however, some of them may also overlap. It appears that their line shape is quite well described by a Gaussian where the width parameter h, usually characterized as the full width at half-maximum (FWHM), increases with the gamma-ray energy. The centre of a peak j, c_j (or E_j if converted to energy), estimated as channel number, corresponds to the gamma-ray energy, and can be used for identifying the radionuclide.

The net area of the Gaussian peak, converted to a net counting rate, is a measure of the activity of this radionuclide. The net peak area may be obtained by subtracting, for example, an integral of a linear background function from the integral of counts within the peak region; this is the "trapezoidal" method for estimating the background. It may, however, also be estimated by fitting the sum of a background function, if necessary also an asymmetric so-called step-function, and a (Gaussian) peak function to a suitably selected part of the spectrum which safely encompasses the peak.

The latter case is considered below. Linear fitting is applied with fixing the non-linear peak shape parameters for peak position and width to calibrated values. This is considered as appropriate for small peaks.

NOTE For larger peaks, however, generally non-linear fitting is used. If this is also applied to small peaks, care has to be taken of preventing the non-linear parameter values from leaving their meaningful ranges during fitting. This may be achieved by, for example, the method of penalized fitting[29], which corresponds to applying normal distributions of the fitting parameters as their Bayesian priors[30]. The parameters of these prior distributions are to be taken from preceding calibrations of the non-linear shape parameters.

B.3.2 Model consideration

Let n_F be the number of channels taken as fitting region; it may for example be defined by taking the asymmetric interval ($c_1 - 4h$, $c_1 + 3h$), in channels, corresponding to $n_F = 7h$; it is chosen as asymmetric because of the step function. In practice, the choice of this region may often depend on how close to c_1 other peaks occur. Assuming a polynomial background and a step-function beneath the peak, the

following functional representation can be used to describe the superposition of contributions from spectral lines and background in a part of the spectrum under investigation:

$$L_1(i) = \exp\left[-(i-c_1)^2/(2\sigma^2)\right]\Big/\sqrt{2\pi\sigma^2}$$

$$L_2(i) = \frac{1}{1+\exp\left[(i-c_1)/(a\sigma)\right]} \qquad \text{(B.11)}$$

$$L_j(i) = (i-c_1)^{j-3}\ ;\ (j=3,4,5)$$

where σ in Formula (B.11) is defined as $\sigma = h/2{,}355$; in these Formulas, all values are given in channels. Choosing the linear energy calibration coefficient as less than 0,5 keV channel^{-1} assures that the FWHM (h) is larger than a minimum of 4 channels. The number of output quantities subject to fitting is equal to the number of different indices j of Lj; however, only the first output quantity is that of interest, at this stage the net peak area.

The first function in Formula (B.11) describes the shape of a spectral line by a Gaussian function. The second line of Formula (B.11) represents the "step function" under a spectral line which shall be explained by the two effects of i) incomplete charge collection and ii) Compton-scattering in the source, which dominate at higher (i) or lower energies (ii), respectively. a is a parameter characterizing the steepness of the step function and shall be known beforehand. The residual functions in the third line of Formula (B.11) are used to model phenomenologically the background by a polynomial of up to second order.

Although the step-function area originates from the peak and is connected to its amplitude, it is common practise to assign its area to the background. When considering, for example, the case of estimating the decision threshold associated with the peak area, the assumed peak and thereby also the step-function vanish.

The assumption implicitly made by $L_2(i)$ in Formula (B.11) that the height of the step-function is independent of the peak height, is not generally usable. It may be applied only to larger peaks used for peak shape calibration. For small peaks the fitted height of L_2 might be dubious. Normally, the step-function height is coupled in an energy-dependent form to the peak height. If this is taken into account, however, the Formula (B.11) is replaced by the following Formula:

$$L_1(i) = \frac{1}{\sqrt{2\pi\sigma^2}}\left\{\exp\left[-(i-c_1)^2/(2\sigma^2)\right] + \frac{d}{1+\exp\left[(i-c_1)/(a\sigma)\right]}\right\} \qquad \text{(B.12)}$$

$$L_j(i) = (i-c_1)^{j-3}\ ;\ (j=2,3,4)$$

The new parameter introduced, d, is the relative step-function height, relative to the amplitude of the peak, the latter being $y_1\Big/\sqrt{2\pi\sigma^2}$; d then is a further shape parameter. A definite reason for using Formula (B.12) arises if the expected step function amplitude is less than about $\sqrt[2]{x_i}$ (x_i in counts) because fitting according to Formula (B.11) otherwise may yield also negative step-function heights which are not meaningful.

Except for the channel number i, all quantities in Formula (B.11) are parameters p_c of the spectral line or of the background step function the values of which have been determined by calibration; the channel position of the peak centre, c_1, can be estimated by other algorithms if the peak is not overlapped by another one. The elements of the response matrix are $A_{ij} = L_j(i)$, with i being the channel number covering the fitting region.

In actual cases, more complicated line shapes may be used, introducing, for instance, low-energy exponential tailings, which increases the number of parameters of the peak shape.

In the Formula system of Formulas (B.11) and (B.12), the value y_1, i.e. the net peak area counting rate, associated with L_1 (equivalently the counting rate), usually is not yet the one being of interest, but for instance the decay-corrected activity concentration. Therefore, these two Formulas describe the sub-model which is solved for y_1 by unfolding y_1 has to be multiplied with an extended calibration

factor, called w_1 here, which depends on the details of gamma-spectrometric measurement, $y_1^+ = w_1\left(y_1 - y_{1,0}\right) = \frac{e^{+\lambda_1 t_A} f_1}{\varepsilon_{\gamma 1}\, p_{\gamma 1} V f_d\left(\lambda_1, 0, t_c\right)}\left(y_1 - y_{1,0}\right)$. It is assumed also, as in the case of measuring ^{40}K, that a counting rate contribution, $y_{1,0}$, due to a background peak has to be subtracted, the counting rate of which has been determined from a separately measured background spectrum. A typical evaluation of such a measurement then is given as

$$y_1^+ = w_1\left(y_1 - y_{1,0}\right) = \frac{e^{+\lambda_1 t_A} f_1}{\varepsilon_{\gamma 1}\, p_{\gamma 1} V f_d\left(\lambda_1, 0, t_c\right)}\left(y_1 - y_{1,0}\right) \tag{B.13}$$

The other input quantities are: $\varepsilon_{\gamma 1}$ and $p_{\gamma 1}$, detection efficiency and gamma emission probability, respectively, for the energy of the gamma line; $f_d(\lambda_1,0,t_c)$, correction for the decay during the measurement with counting duration t_c and radionuclide decay constant λ_1; f_1, a correction for self-attenuation; t_A, the time elapsed between sampling and the start of the measurement; V, sample volume (in L); t_c and t_0, counting durations for sample and background measurement.

The background peak subtraction indicated in Formula (B.13) means in practise that the corresponding components of y and U_y have to be modified

$$y_{1,\mathrm{bc}} = y_1 - y_{1,0}\ ;\ U_{y,\mathrm{bc}}\left(1,1\right) = U_y\left(1,1\right) = u^2\left(y_{1,0}\right) \tag{B.14}$$

Then Formula (B.13) shortly reads: $y_1^+ = w_1\, y_{1,\mathrm{bc}}$ (the index bc means background peak corrected).

B.3.3 Uncertainties of input quantities

The primary input quantities belonging to the sub-model, w_i, are channel counting rates of the selected fitting region around the peak center c_1. The uncertainty matrix U_x is diagonal with elements $U_x(i,i) = x_i/t_c$. The values and uncertainties of the fixed parameters, p_c and U_{pc}, i.e. the peak position c_1 and σ as shape-parameter ($n_p = 2$), are given from preceding calibrations or obtained by other means. This includes also the net peak counting rate from a separate background measurement.

B.3.4 Evaluation of the primary result and its associated standard uncertainty

The equation system (B.1) represents an explicit and linear model with $A(p_c)$ being a $n \times m$ matrix the elements of which are – for each row i – are the function values of $L_j(i)$, $j = 1,\ldots, 5$. Its first column contains the values of the Gaussian peak function (normalized to 1) plus the step-function, see Formula (B.13).

The Formula system (B.12) with Formula (B.13) has now become a sub-model and leads, depending on the existence of a corresponding background peak to a two-step or three-step solution, the first according to Formula (18)

$$\boldsymbol{y} = \boldsymbol{U_y}\left(\boldsymbol{x}\right)\boldsymbol{A}^T\boldsymbol{U_x^{-1}}\left(\boldsymbol{x}\right)\boldsymbol{x};\ \ \boldsymbol{U_y}\left(\boldsymbol{x}\right) = \left[\boldsymbol{A}^T\boldsymbol{U_x^{-1}}\left(\boldsymbol{x}\right)\boldsymbol{A}\right]^{-1} \tag{B.15}$$

If a corresponding background peak exists, the next step is to calculate Formula (25). As a pre-requisite, it is recommended to construct a vector $\boldsymbol{q}$, associated with uncertainties $\boldsymbol{U_q}$, which includes the peaks shape parameters $\boldsymbol{p}_c$ and those parameters contained in the factor w_1 (see Formula (B.13)); the background counting rate $y_{1,0}$ being subtracted in Formula (B.14) is also included in $\boldsymbol{q}$ and $\boldsymbol{U_q}$. Then, with taking the second part of Formula (7) into account and using $D_{1,1} = w_1$ one obtains

$$\boldsymbol{U_y}\left(\boldsymbol{x}\right) = \left[\boldsymbol{A}^T\boldsymbol{U_x^{-1}}\left(\boldsymbol{x}\right)\boldsymbol{A}\right]^{-1};\ \boldsymbol{y}^+ = \boldsymbol{D}\left[\boldsymbol{U_y}\left(\boldsymbol{x}\right)\boldsymbol{A}^T\boldsymbol{U_x^{-1}}\left(\boldsymbol{x}\right)\boldsymbol{x} - \boldsymbol{y_0}\right] \tag{B.16}$$

$$\boldsymbol{U}_{\boldsymbol{y}^+} = \boldsymbol{U}_{\boldsymbol{y}^+}\left(\boldsymbol{x}\right) + \boldsymbol{U}_{\boldsymbol{y}^+}\left(\boldsymbol{q}\right) = \boldsymbol{J}\boldsymbol{U_y}\left(\boldsymbol{x}\right)\boldsymbol{J}^{\mathrm{T}} + \boldsymbol{Q'}\boldsymbol{U_q}\boldsymbol{Q'}^{\mathrm{T}} \tag{B.17}$$

NOTE If the peak-shape parameters as given in L_1 of Formula (B.12) are included in $\boldsymbol{q}$ and $\boldsymbol{U_q}$ their uncertainties if given are taken into account by the uncertainty propagation according to Formula (B.17).

B.3.5 Standard uncertainty as a function of an assumed true value of the measurand

In the decision threshold case of the activity concentration (k=1; see 5.9) a modified vector $\tilde{y}^+$ is prepared by replacing the first component of $\boldsymbol{q}$ and $\tilde{y}^+$, associated with the output quantity of interest, by $y_1^+ = 0$. This value can be converted to $\tilde{y}_1 = w^{-1}\tilde{y}_1^+ + y_{1,0}$. Following 5.9, the vector $\tilde{z}$ of modified input quantities x, i.e. of modified net counting rates of the spectrum channels, is obtained as $\tilde{z} = \tilde{x} = A\tilde{y}$. The components of $\tilde{U}_{\tilde{x}}$ are calculated as follows

$$u^2(\tilde{x}_i) = \frac{\tilde{x}_i}{t_c} \quad u(\tilde{x}_i, \tilde{x}_j) = 0,\ i \neq j \tag{B.18}$$

Now a complete set of modified measured input values, $\tilde{x}$ and $\tilde{U}_{\tilde{x}}$, is found which then by applying Formulas (B.13) to (B.17) leads to modified values y^+ and $\tilde{U}_{\tilde{y}^+}$, from which finally the square root of the (1,1)-element of $\tilde{U}_{\tilde{y}^+}$ is taken as the desired value of the uncertainty function for the activity concentration.

NOTE This means the actual spectrum region is modified and re-fitted in each evaluation of this uncertainty function. Using this re-fitting avoids finding an appropriate expression for the peak-fitting "design factor" function $f_B(.)$; see also the note below Formula (B.6).

For the calculation of the characteristic limits one has to proceed as stipulated in 5.10 to 5.11.

B.4 Gamma-ray spectrometry — Fitting double peaks

This case is quite similar to that of B.3 apart from assuming that the gamma-peak of interest, located at channel c_1, is now partly overlapped by a second one located at c_2. Referring to the model consideration, the width of the full fitting region is now extended to ($c_1 - 4h$,$c_2 = 3h$) corresponding to a number of channels $n_F = 7h + (c_1 - c_2)$. The model Formulas now include a second function $L_2(i)$ for the peak and the step-function of the second peak

$$L_1(i) = \frac{1}{\sqrt{2\pi\sigma^2}}\left\{\exp\left[-(i-c_1)^2/(2\sigma^2)\right] + \frac{d}{1+\exp\left[(i-c_1)/(a\sigma)\right]}\right\}$$

$$L_2(i) = \frac{1}{\sqrt{2\pi\sigma^2}}\left\{\exp\left[-(i-c_2)^2/(2\sigma^2)\right] + \frac{d}{1+\exp\left[(i-c_2)/(a\sigma)\right]}\right\} \tag{B.19}$$

$$L_j(i) = (i-c_1)^{j-3};\ (j = 3,4,5)$$

The same shape parameter values of σ, d and a are used for the two peaks. This means that the matrix $A_{ij} = L_j(i)$ is extended by one column (inserted before the second column). Then, solving this sub-model for y and U_y follows the Formula (B.16). With treating the second peak as the origin of interference, one is again interested only in the value of the first output quantity, y_1. Therefore, the steps now following are the same as in 6.2, with taking Formula (B.14) into account, if necessary.

It follows that this case is quite similar to that of B.3, only the dimensions of vectors and matrices within the sub-model are extended.