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Determination of the detection limit and decision threshold for ionizing radiation measurements —

Part 8: Fundamentals and application to unfolding of spectrometric measurements without the influence of sample treatment

Détermination de la limite de détection et du seuil de décision des mesurages de rayonnements ionisants —

Partie 8: Principes fondamentaux et leur application à la déconvolution des spectres des mesurages de rayonnements ionisants négligeant l'influence de la préparation d'un échantillon

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

International Standards are drafted in accordance with the rules given in the ISO/IEC Directives, Part 2.

The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights.

ISO 11929-8 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Subcommittee SC 2, *Radiation protection*.

ISO 11929 consists of the following parts, under the general title *Determination of the detection limit and decision threshold for ionizing radiation measurements*:

— *Part 1: Fundamentals and application to counting measurements without the influence of sample treatment*

— *Part 2: Fundamentals and application to counting measurements with the influence of sample treatment*

— *Part 3: Fundamentals and application to counting measurements with high resolution gamma spectrometry*, *without the influence of sample treatment*

— *Part 4: Fundamentals and applications to measurements by use of linear-scale analogue ratemeters*, *without the influence of sample treatment*

— *Part 5: Fundamentals and applications to counting measurements on filters during accumulation of radioactive material*

— *Part 6: Fundamentals and applications to measurements by use of transient mode*

— *Part 7: Fundamentals and general applications*

— *Part 8: Fundamentals and applications to unfolding of spectrometric measurements without the influence of sample treatment*

Introduction

This part of ISO 11929 gives basic information on the statistical principles for the determination of the detection limit, of the decision threshold and of the limits of the confidence interval for general applications of nuclear radiation measurements.

ISO 11929-1 and ISO 11929-2 deal with integral counting measurements with or without consideration of the sample treatment. High-resolution spectrometric measurements, which can be evaluated without unfolding techniques, are covered in ISO 11929-3 while evaluations of spectra via unfolding have to be treated according to this part of ISO 11929. ISO 11929-4 deals with measurements using linear scale analogue ratemeters, ISO 11929-5 with monitoring of the concentration of aerosols in exhaust gas, air or waste water, and ISO 11929-6 with measurements by use of a transient measuring mode.

Parts 1 to 4 were elaborated for special measuring tasks in nuclear radiation measurements based on the principles defined by Altschuler and Pasternack [1], Nicholson [2], Currie [3]. ISO 11929-7 gives a general Bayesian-statistical approach for the determination of decision thresholds, detection limit and confidence intervals by separating the determination of these characteristic quantities from the evaluation of the measurement. Consequently ISO 11929-7 is generally applicable and can be applied to any suitable procedure for the evaluation of a measurement. Parts 5, 6 and 7 and this part of ISO 11929 are based on methods of Bayesian statistics (see [5] in the Bibliography) for the determination of the characteristic limits (see [6] and [7] in the Bibliography) as well as for the unfolding (see [8] in the Bibliography).

This part of ISO 11929 makes consequent use of the general approach of ISO 11929-7 and describes explicitly the necessary procedures to determine decision thresholds, detection limits and confidence limits for physical quantities which are derived from the evaluation of nuclear spectrometric measurements by unfolding techniques, without taking into account the influence of sample treatment (see [4] in the Bibliography). There are many types of such quantities, for example, the net area of a spectral line in gamma- or alpha-spectrometry.

Since the uncertainty of measurement plays a fundamental role in this part of ISO 11929, evaluations of measurements and the determination of the uncertainties of measurement have to be performed according to the Guide for the Expression of Uncertainty in Measurement.

For this purpose, Bayesian statistical methods are used to specify statistical values characterized by the following given probabilities:

— The *decision threshold*, which allows a decision to be made for each measurement with a given probability of error as to whether the result of a measurement indicates the presence of the physical effect quantified by the measurand.

— The *detection limit*, which specifies the minimum true value of the measurand which can be detected with a given probability of error using the measuring procedure in question. This consequently allows a decision to be made as to whether a measuring method checked using this part of ISO 11929 satisfies certain requirements and is consequently suitable for the given purpose of measurement.

— The *limits of the confidence interval*, which define an interval which contains the true value of the measurand with a given probability, in the case that the result of the measurement exceeds the decision threshold.

Determination of the detection limit and decision threshold for ionizing radiation measurements —

Part 8:
Fundamentals and application to unfolding of spectrometric measurements without the influence of sample treatment

1 Scope

This part of ISO 11929 specifies a method for determination of suitable statistical values which allow an assessment of the detection capabilities in spectrometric nuclear radiation measurements, and of the physical effect quantified by a measurand (for example, a net area of a spectrometric line in an alpha- or gamma-spectrum) which is determined by evaluation of a multi-channel spectrum by unfolding methods, without the influence of sample treatment. For this purpose, Bayesian statistical methods are used to specify characteristic limits.

2 Normative references

The following referenced documents are indispensable for the application 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.

BIPM/IEC/IFCC/ISO/IUPAC/IUPAP/OIML, *Guide to the expression of uncertainty in measurement*, Geneva, 1993

BIPM/IEC/IFCC/ISO/IUPAC/IUPAP/OIML, *International vocabulary of basic and general terms in metrology.* 2nd edition, Geneva, 1993.

ISO 11929-3:2005, *Determination of the detection limit and decision threshold for ionizing radiation measurements — Part 3: Fundamentals and application to counting measurements with high resolution gamma spectrometry, without the influence of sample treatment*

ISO 11929-7:2005, *Determination of the detection limit and decision threshold for ionizing radiation measurements — Part 7: Fundamentals and general applications*

3 Terms and definitions

For the purposes of this document, the following terms and definitions apply.

3.1
measuring method
any logical sequence of operations, described generically, used in the performance of measurements

NOTE Adapted from the International Vocabulary of Basic and General Terms in Metrology:1993.

3.2
measurand
particular quantity subject to measurement

[International Vocabulary of Basic and General Terms in Metrology:1993]

NOTE In this part of ISO 11929, a measurand is non-negative and quantifies a nuclear radiation effect. The effect is not present if the value of the measurand is zero. It is characteristic of this part of ISO 11929 that the measurand is derived from a multi-channel spectrum by unfolding methods. An example of a measurand is the intensity of a line in a spectrum above the background in a spectrometric measurement.

3.3
uncertainty (of measurement)
parameter, associated with the result of a measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measurand

[Guide for the expression of uncertainty in measurement:1993]

NOTE The uncertainty of a measurement derived according to the Guide for the expression of uncertainty in measurement comprises, in general, many components. Some of these components may be evaluated from the statistical distribution of the results of series of measurements and can be characterized by experimental standard deviations. The other components, which also can be characterized by standard deviations, are evaluated from assumed or known probability distributions based on experience and other information.

3.4
mathematical model of the evaluation
a set of mathematical relationships between all measured and other quantities involved in the evaluation of measurements

3.5
decision quantity
random variable for the decision whether the physical effect to be measured is present or not

3.6
decision threshold
fixed value of the decision quantity by which, when exceeded by the result of an actual measurement of a measurand quantifying a physical effect, one decides that the physical effect is present

NOTE The decision threshold is the critical value of a statistical test to decide between the hypothesis that the physical effect is not present and the alternative hypothesis that it is present. When the critical value is exceeded by the result of an actual measurement, this is taken to indicate that the hypothesis should be rejected. The statistical test will be designed such that the probability of wrongly rejecting the hypothesis (error of the first kind) is at most equal to a given value α.

3.7
detection limit
smallest true value of the measurand which is detectable by the measuring method

NOTE The detection limit is the smallest true value of the measurand which is associated with the statistical test and hypotheses according to 3.6 by the following characteristics: if in reality the true value is equal to or exceeds the detection limit, the probability of wrongly not rejecting the hypothesis (error of the second kind) will be at most equal to a given value β.

3.8
confidence limits
values which define confidence intervals to be specified for the measurand in question which, if the result exceeds the decision threshold, includes the true value of the measurand with the given probability (1 - γ)

3.9
guideline value
value which corresponds to scientific, legal or other requirements for which the measuring procedure is intended to assess

EXAMPLE Activity, specific activity or activity concentration, surface activity, or dose rate.

4 Quantities and symbols

$\hat{\xi}$ Random variable as estimator for a non-negative measurand quantifying a physical effect

ξ Value of the estimator; true value of the measurand

$\tilde{u}(\xi)$ Standard uncertainty of the decision quantity X as a function of the true value ξ of the measurand

X Random variable as decision quantity; estimator of the measurand

x Result of a determination of the decision quantity X

$u(x)$ Standard uncertainty of the measurand associated with the measurand result x of a measurement

z Best estimate of the measurand

$u(z)$ Standard uncertainty of the measurand associated with the best estimate z

x^* Decision threshold for the measurand

ξ^* Detection limit for the measurand

ξ_l, ξ_u Respectively, lower and upper limit of the confidence interval for the measurand

i Number of a channel in a multi-channel spectrum obtained by a spectrometric nuclear radiation measurement; ($i = 1, ..., m$)

ϑ Continuous parameter (for example, energy or time) related to the different channels in a multi-channel spectrum

ϑ_i Value of ϑ connected with channel i; ($i = 1, ..., m$)

t Measuring time

m Number of channels in the spectrum

N_i Independent Poisson-distributed random variables of events counted in a channel i during the measurement of duration t; ($i = 1, ..., m$)

n_i Number of events counted in a channel i during the measuring time t; ($i = 1, ..., m$)

X_i Independent random variable of the rate of events counted in a channel i during a measurement of duration t, input quantities 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 a channel i during a measurement of duration t; $x_i = n_i / t$; ($i = 1, ..., m$)

$\boldsymbol{x}$ Column matrix of the x_i

$\boldsymbol{x}'$ Column matrix $\boldsymbol{x}' = \boldsymbol{A}\boldsymbol{y}'$

$u(x_i, x_j)$ Covariance associated with x_i and x_j

Y_k Output quantity Y_k derived from the multi-channel spectrum by unfolding methods; $k = 1, ..., n$

$\boldsymbol{Y}$ Column matrix of the Y_k

y_k Estimate of an output quantity (parameter), Y_k; ($k = 1, ..., n$)

$u(y_k)$ Standard uncertainty of y associated with y_k

$\boldsymbol{y}'$ Column matrix $\boldsymbol{y}$ after replacement of y_1 by ξ

$H(\vartheta_i)$ Functional relationship representing the spectral density at ϑ_i of a multi-channel spectrum; $H(\vartheta_i) = \sum_{k=1}^{n} \Psi_k(\vartheta_i) \cdot Y_k$

p Number of input quantities t_i which are not subject to fit

$\Psi_k(\vartheta)$ Function describing the shapes of the individual spectral lines and of the background contributions; ($k = 1, ..., n$)

n Number of output quantities

$\boldsymbol{v}$ Column matrix of input quantities; $\boldsymbol{v} = (x_1, ..., x_m, t_1, ..., t_p)$

t_i Input quantities which are not subject to fit

$\boldsymbol{M(Y)}$ Column matrix of the $H(\vartheta_i)$

$\boldsymbol{A}$ Response matrix of the spectrometer

A_{ik} Elements of the response matrix $\boldsymbol{A}$

$\boldsymbol{U_x}$ Uncertainty matrix of $\boldsymbol{X}$

$\boldsymbol{U_y}$ Uncertainty matrix of $\boldsymbol{Y}$

G_k Function of the input quantities X_i, ($i = 1, ..., m$)

$\boldsymbol{G}$ Column matrix of the G_k

α Probability of the error of the first kind; the probability of rejecting the hypothesis if it holds true

β Probability of the error of the second kind; the probability of accepting the hypothesis if it is false

$1 - \gamma$ Probability attributed to the confidence interval of the measurand; probability that the true value of the measurand is included by the confidence interval

k_p Quantiles of the standardized normal distribution for the probability p (see Table 1); $p = 1 - \alpha$, $1 - \beta$, $1 - \gamma$

E Operator for the formation of the expectation of a random variable

Var Operator for the formation of the variance of a random variable

diag Indicator for a diagonal matrix

5 Statistical values and confidence interval

5.1 Principles

5.1.1 General aspects

For a particular task involving nuclear radiation measurements, first the particular physical effect which is the objective of the measurement has to be described. Then a non-negative measurand has to be defined which quantifies the physical effect and which assumes the value zero if the effect is not present in an actual case.

A random variable, called a decision quantity X, has to be attributed to the measurand. It is also an estimator of the measurand. It is required that the expectation value EX of the decision quantity X equals the true value ξ of the measurand. A value x of the estimator X derived from measurements is a primary estimate of the measurand. The primary estimate x of the measurand, and its associated standard uncertainty $u(x)$, have to be calculated as the primary complete result of the measurement according to the Guide for the expression of uncertainty in measurement, by evaluation of measured quantities and of other information using a mathematical model of the evaluation which takes into account all relevant quantities. Generally, the fact that the measurand is non-negative will not be explicitly made use of. Therefore, x may become negative, in particular, if the true value of the measurand is close to zero.

NOTE The model of the evaluation of the measurement need not necessarily be given in the form of explicit mathematical formulas. It can also be represented by an algorithm or a computer code.

For the determination of the decision threshold and the detection limit, the standard uncertainty of the decision quantity has to be calculated, if possible, as a function $\tilde{u}(\xi)$ of the true value ξ of the measurand. In the case that this is not possible, approximate solutions are described below.

ξ s the value of another, non-negative estimator $\hat{\xi}$ of the measurand. The estimator $\hat{\xi}$, in contrast to X, makes use of the knowledge that the measurand is non-negative. The limits of the confidence interval to be determined refer to this estimator $\hat{\xi}$ (compare 5.4). Besides the limits of the confidence interval, the expectation value E$\hat{\xi}$ of this estimator as a best estimate z of the measurand, and the standard deviation $[\mathrm{Var}(\hat{\xi})]^{1/2}$ as the standard uncertainty $u(z)$ associated with the best estimate z of the measurand, have to be calculated (see 6.3).

For the numerical calculation of the decision threshold and the detection limit, the function $\tilde{u}(\xi)$ is needed, which is the standard uncertainty of the decision quantity X as a function of the true value ξ of the measurand. The function $\tilde{u}(\xi)$ generally has to be determined by the user of this part of ISO 11929 in the course of the evaluation of the measurement according to the Guide for the expression of Uncertainty in Measurement. For examples see Annex A. This function is often only slowly increasing. Therefore, it is justified in many cases to use the approximation $\tilde{u}(\xi) = u(x)$. This applies, in particular, if the primary estimate x of the measurand is not much larger than its standard uncertainty $u(x)$ associated with x. If the value x is calculated as the difference (net effect) of two approximately equal values y_1 and y_0 obtained from independent measurements, that is $x = y_1 - y_0$, one gets $\tilde{u}^2(0) = u^2(y_1) + u^2(y_0)$ with the standard uncertainties $u(y_1)$ and $u(y_0)$ associated with y_1 and y_0, respectively.

If only $\tilde{u}^2(0)$ and $u(x)$ are known, an approximation by linear interpolation is often sufficient for $x > 0$ according to:

$$\tilde{u}^2(\xi) = \tilde{u}^2(0) \cdot (1 - \xi / x) + u^2(x) \cdot \xi / x \tag{1}$$

NOTE In many practical cases, $\tilde{u}^2(\xi)$ is a slowly increasing linear function of ξ. This justifies the approximations above, in particular the linear interpolation of $\tilde{u}^2(\xi)$ instead of $\tilde{u}(\xi)$ itself.

For setting up the mathematical model of the evaluation of the measurement, one has to distinguish two types of physical quantities, input and output quantities. The output quantities Y_k ($k = 1, ..., n$) are viewed as measurands (for example, the parameters of an unfolding or fitting procedure) which have to be determined by the evaluation of a measurement. The decision quantity X is one of them. They depend on the input

quantities X_i ($i = 1, ..., m$) which are the quantities obtained by repeated measurements, influence quantities and results of previous measurements and evaluations. (Compare chapter 4.1.2 of the Guide for the expression of uncertainty in measurement:1993). One has to calculate the estimates y_k of the output quantities (measurands) as the results of the measurement and the standard uncertainties $u_c(y_k)$ associated with the y_k.

The model of the evaluation is given by a set of functional relationships:

$$Y_k = G_k\ (G_1, ..., X_m); (k = 1, ..., n) \tag{2}$$

Estimates of the measurands Y_k, denoted y_k, are obtained from Equation (2) using input estimates $x_1, ..., x_m$ for the values of the m quantities $X_1, ..., X_m$. Thus, the output estimates y_k and the standard uncertainties $u(y_k)$ associated with y_k are given by

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

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

where x_i and x_j are the estimates of X_i and X_j and $u(x_i, x_j) = u(x_j, x_i)$ is the estimated covariance associated with x_i and x_j. From these covariances one obtains:

$$u^2(y_k) = u\,(y_k, y_k) \tag{5}$$

In cases when the partial derivatives are not explicitly available, they can be numerically approximated in a sufficiently exact way using the standard uncertainty $u(x_k)$ as an increment of x_k by:

$$\frac{\partial G_k}{\partial x_i} = \frac{1}{u(x_i)} \left\{ G_k\,[x_1, ..., x_i + u(x_i)/2, ..., x_m] - G_k\,[x_1, ..., x_i - u(x_i)/2, ..., x_m] \right\} \tag{6}$$

See A.1 for further details and explanations.

5.1.2 Model

For the purpose of determining a decision quantity X (for example, a net peak area of a line) by unfolding spectra obtained by spectrometric nuclear radiation measurements, the following model is used. The input quantities are estimated by $x_i = n_i/t$, with n_i being the independent counted events in the ($i = 1, ..., m$) channels of a multi-channel spectrum. The n_i obey Poisson statistics and hence one obtains $u^2(x_i) = n_i/t^2 = x_i/t$ and $u(x_i, x_j) = 0$. Any influences of sample treatment on the uncertainties of the x_i and other non-Poisson sources of uncertainties are neglected in this part of ISO 11929.

NOTE If one or several n_i are zero, the problem occurs that unrealistically $u(n_i)$ is zero and consequently one gets the problem of division by $u^2(n_i) = 0$ when using the least-square methods for the unfolding. This problem can be avoided by replacing all n_i by $n_i + 1$ or by a suitable combination of channels in a multi-channel spectrum.

In the following, matrix notation is used for quantities, values and functions abbreviated by the same symbol. For instance, the column matrix $\boldsymbol{x} = (x_1, ..., x_m)^T$ is used for the estimates of the input quantities $\boldsymbol{X} = (X_1, ..., X_m)^T$. (Column matrices are written here as transposed row matrices.)

For the unfolding of a measured multi-channel spectrum, a set of functional relationships $\boldsymbol{M}(\boldsymbol{Y})$ according to Equation (3) is fitted to the estimates $\boldsymbol{x}$ of the m input quantities $\boldsymbol{X}$, which are the measured results $x_i = n_i/t$ calculated from the channel counts n_i of the spectral density. The input quantities may comprise further quantities from which measured data or other data are used in the unfolding and which have uncertainties associated with them. Further, there are input quantities t_i for which estimates exist, but which are not subject to fit. For convenience, the vectors $\boldsymbol{x}$ and $\boldsymbol{t}$ are combined in a vector $\boldsymbol{v}$.

Then the model of the unfolding is given by:

$$\boldsymbol{M}(\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{t}) = \boldsymbol{M}(\boldsymbol{v}, \boldsymbol{y}) = \mathbf{0} \tag{7}$$

with **0** being the zero vector.

The components of the measurement uncertainties are combined to uncertainty matrices $\boldsymbol{U}_{\boldsymbol{x}} = [u(x_i, {}_j)]$ and $\boldsymbol{U}_{\boldsymbol{y}} = [u(y_k, {}_l)]$. $\boldsymbol{U}_{\boldsymbol{x}}$ is a diagonal matrix with the diagonal elements $u^2(x_i) = n_i/t^2$ and consequently $\boldsymbol{U}_{\boldsymbol{x}} = \mathrm{diag}(x_i/t)$.

The calculation of the output quantities $\boldsymbol{y}$, of the uncertainty matrix $\boldsymbol{U}_{\boldsymbol{y}}$ associated with $\boldsymbol{y}$, and of the best estimates $\boldsymbol{z}$ of the input quantities $\boldsymbol{x}$ from the given measured and estimated data is, in general, a non-linear fitting procedure which mostly is done using the method of least squares as described, for instance, in DIN 1319-4. Formally, it is sufficient for the unfolding that given functions $\boldsymbol{F}$ and $\boldsymbol{G}$ or respective algorithms exist so that:

$$\boldsymbol{y} = \boldsymbol{F}\,(\boldsymbol{x}, \boldsymbol{t}) = \boldsymbol{F}(\boldsymbol{v}) \tag{8}$$

$$\boldsymbol{z} = \boldsymbol{G}(\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{t}) = \boldsymbol{H}(\boldsymbol{v}) \tag{9}$$

with $\boldsymbol{H}(\boldsymbol{v}) = \boldsymbol{G}\,[\boldsymbol{x},\ \boldsymbol{F}(\boldsymbol{x}, \boldsymbol{t}), \boldsymbol{t}]$

The uncertainty matrices $\boldsymbol{U}_{\boldsymbol{y}}$ and $\boldsymbol{U}_{\boldsymbol{z}}$ associated with the output quantities $\boldsymbol{y}$ and the best estimates $\boldsymbol{z}$ of the input quantities $\boldsymbol{x}$ have to be calculated according to the Guide for the expression of uncertainty in measurement. This yields

$$\boldsymbol{U}_{\boldsymbol{y}} = \boldsymbol{F}_{\boldsymbol{v}}\,\boldsymbol{U}_{\boldsymbol{v}}\,\boldsymbol{F}_{\boldsymbol{v}}^{\mathsf{T}};\ \ \boldsymbol{U}_{\boldsymbol{z}} = \boldsymbol{H}_{\boldsymbol{v}}\,\boldsymbol{U}_{\boldsymbol{v}}\,\boldsymbol{H}_{\boldsymbol{v}}^{\mathsf{T}} \tag{10}$$

with $\boldsymbol{F}_{\boldsymbol{v}}$ and $\boldsymbol{H}_{\boldsymbol{v}}$ denoting the matrices of the partial derivatives of the model functions: $\boldsymbol{F}_{\boldsymbol{v}} = (\partial F_i/\partial x_k)$ and $\boldsymbol{H}_{\boldsymbol{v}} = (\partial H_i/\partial x_k)$.

Let, for example, Y_1 be the decision quantity X. In order to calculate $\tilde{u}(\xi)$, one has to replace y_1 by ξ and obtain $y' = (\xi, y_2, ..., y_n)$ or and $\boldsymbol{x}' = \boldsymbol{M}(\boldsymbol{y}') = \boldsymbol{A}\boldsymbol{y}'$ with a suitable set of functions. From this, one forms the changed uncertainty matrix $\boldsymbol{U}_{\boldsymbol{x}'} = \mathrm{diag}(x_i'/t)$ and replaces $\boldsymbol{U}_{\boldsymbol{x}}$ by $\boldsymbol{U}_{\boldsymbol{x}'}$ in Equation (10). The (1,1)-element of the thus-changed uncertainty matrix $\boldsymbol{U}_{\boldsymbol{y}}$ then gives $\tilde{u}^2(\xi)$.

For explicit examples of model functions in alpha- and gamma-spectrometry and further explanations, see A.3.

5.2 Decision threshold

The decision threshold x^* of a non-negative measurand quantifying the physical effect, according to 5.1, is a value of the decision quantity X which, when it is exceeded by a result x of a measurement indicates that the physical effect is present. If $x \leqslant x^*$ one decides that the physical effect is not present. If this decision rule is observed, a wrong decision in favour of the presence of the physical effect occurs with the probability not greater than α (error of the first kind).

The decision threshold is given by:

$$x^* = k_{1-\alpha} \cdot \tilde{u}\,(0) \tag{11}$$

Values of the quantiles $k_{1-\alpha}$ of the standardized normal distribution are given in Table 1. It is $\Phi(k_{1-\alpha}) = 1 - \alpha$.

If the approximation $\tilde{u}\,(\xi) = u(x)$ is sufficient, one gets

$$x^* = k_{1-\alpha} \cdot u\,(x)$$

5.3 Detection limit

The detection limit ξ^*, which is the smallest true value of the measurand detectable with the measuring method, is so much larger than the decision threshold that the probability of an error of the second kind equals β. The detection limit is given by:

$$\xi^* = x^* + k_{1-\beta} \cdot \tilde{u}(\xi^*) \tag{12}$$

Equation (12) is an implicit one. The detection limit can be calculated from it by iteration using, for example, the starting approximation $\xi^* = 2x^*$. The iteration converges in most cases. Equation (12) may have multiple solutions. In this case, the detection limit is the smallest one. If Equation (12) has no solution, the measuring procedure is not suited for the measuring purpose.

If the approximation $\tilde{u}(\xi) = u(x)$ is sufficient, then $\xi^* = (k_{1-\alpha} + k_{1-\beta}) \cdot u(x)$ is valid.

If $\tilde{u}(\xi)$ is not explicitly known for $\xi > 0$, one gets with $\tilde{u}(0)$ and with a result x of a measurement and its associated uncertainty $u(x)$, an approximation of ξ^* using the interpolation formula according to Equation (1)

$$\xi^* = a + \sqrt{a^2 + (k_{1-\beta}^2 - k_{1-\alpha}^2) \cdot \tilde{u}^2(0)} \tag{13}$$

with $a = k_{1-\alpha} \cdot \tilde{u}(0) + \frac{1}{2}(k_{1-\beta}^2 / x) \cdot [u^2(x) - \tilde{u}^2(0)]$

For $\alpha = \beta$ one obtains $\xi^* = 2\alpha$.

When using the approximation of Equation (13) to calculate the detection limit ξ^* and when type B uncertainties are not negligible, a measurement result $x > \approx 2x^*$ shall be chosen. If $x \gg 2x^*$ holds, one obtains an unreasonably high detection limit. In this case, the approximation yields only an upper limit of ξ^*. If type B uncertainties are negligible, Equations (12) and (13) converge to the same result for the detection limit.

Values of the quantiles $k_{1-\alpha}$, $k_{1-\beta}$ of the standardized normal distribution are given in Table 1. It is $\Phi(k_{1-\alpha}) = 1-\alpha$ and $\Phi(k_{1-\beta}) = 1-\beta$.

5.4 Confidence limits

For a result x of a measurement which exceeds the decision threshold x^*, the confidence interval includes the true value of the measurand with the given probability $1 - \gamma$. It is enclosed by the confidence limits ξ_l and ξ_u according to:

$$\xi_l = x - k_p \cdot u(x) \text{ with } p = \kappa \cdot (1 - \gamma/2) \tag{14}$$

$$\xi_u = x + k_q \cdot u(x) \text{ with } q = 1 - (\kappa \cdot \gamma/2) \tag{15}$$

κ is given by:

$$\kappa = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x/u(x)} \exp(-z^2/2)\ \mathrm{d}z = \Phi[x/u(x)] \tag{16}$$

Values of the function $\Phi(t)$ are tabulated (see [9] in the Bibliography) and given in Table 1. It is $\Phi(k_p) = p$ and $\Phi(k_q) = q$.

The confidence limits are not symmetrical around the expectation $E\hat{\xi}$. The probabilities of $\hat{\xi} < \xi_l$ and $\hat{\xi} > \xi_u$, however, are both equal to $\gamma/2$ and the relationship $0 < \xi_l < \xi_u$ is valid. For $x \gg (x)$, the approximation

$$\xi_{l,u} = x \pm k_{1-\gamma/2} \cdot u(x) \tag{17}$$

is applicable if $x > 2 \cdot k_{1-\gamma/2} \cdot u(x)$

6 Application of this part of ISO 11929

6.1 Specific values

The probabilities α, β and $(1 - \gamma)$ shall be specified in advance by the user of this part of ISO 11929. Commonly used values are $\alpha = \beta = 0{,}05$ and $\gamma = 0{,}05$.

6.2 Assessment of a measuring method

To check whether a measuring method (see 3.1) is suitable for the measurement of a physical effect, the detection limit shall be compared with a specified guideline value (e.g. specified requirements on the sensitivity of the measuring procedure for scientific, legal or other reasons; see 3.9).

The detection limit shall be calculated by means of Equation (12). If the detection limit thus determined is greater than the guideline value, the measuring procedure is not suitable for the measurement.

6.3 Assessment of measured results

A measured result has to be compared with the decision threshold calculated by means of Equation (11). If the result of the measurement x is larger than the decision threshold x^*, it is decided that the physical effect quantified by the measurand is present.

If this is the case, the best estimate z of the measurand is calculated using κ from Equation (16) by:

$$z = \mathrm{E}\hat{\xi} = x + \frac{u(x) \cdot \exp\left\{-x^2 / [2u^2(x)]\right\}}{\kappa \cdot \sqrt{2\pi}} \tag{18}$$

with the standard uncertainty $u(z)$ associated with z:

$$u(z) = \sqrt{\mathrm{Var}(\hat{\xi})} = \sqrt{u^2(x) - (z - x) \cdot z} \tag{19}$$

The following relationships: $z \geqslant x$ and $z \geqslant 0$, as well as $u(z)$ $u \leqslant (x)$, are valid and for $x \gg u(x)$, i.e. $x > 4 \cdot u(x)$, the approximations $z = x$ and $u(z) = u(x)$ hold true.

6.4 Documentation

The documentation of measurements in accordance with this part of ISO 11929 shall contain details of the probabilities α, β and $(1 - \gamma)$, the decision threshold x^*, the detection limit ξ^*, and the guideline value.

For a result x of the measurement exceeding the decision threshold x^*, the standard uncertainty $u(x)$ associated with x and the limits of the confidence interval $\xi_{l,u}$ have to be given. If the result x of the measurement is below the decision threshold ξ^*, it shall be documented as “below the decision threshold”.

If the detection limit exceeds the guideline value, it shall be documented that the method is not suitable for the measurement purpose.

In addition, the best estimate z of the measurand and the standard uncertainty $u(z)$ associated with z may be specified if $x/u(x) < 4$.

7 Values of the distribution function of the standardized normal distribution

Values $\Phi(t) = \int_{-\infty}^{t} \varphi(z)\ \mathrm{d}z$ with $\varphi(z) = (1/\sqrt{2\pi}) \cdot \exp(-z^2/2)$ are given in Table 1. For the distribution function of the standardized normal distribution, $\Phi(-t) = 1 - \Phi(t)$ is valid. Quantiles of the standardized normal distribution can also be obtained from Table 1, since $t = k_p$ for $p = \Phi(t)$, i.e. $\Phi(k_p) = p$.

For $t \geqslant 0$, the approximation (see [14] in the Bibliography):

$$\Phi(t) = 1 - \frac{\exp(-t^2/2)}{\sqrt{2\pi}} \cdot (\alpha_1 \cdot \zeta + \alpha_2 \cdot \zeta^2 + \alpha_3 \cdot \zeta^3) + \varepsilon;\ \ \zeta = \frac{1}{1 + \alpha_0 \cdot t}$$

is valid with $|\varepsilon| < 10^{-5}$ and

$\alpha_0 = 0{,}332\,67$; $\alpha_1 = 0{,}436\,183\,6$; $\alpha_2 = -0{,}120\,167\,6$; $\alpha_3 = 0{,}937\,298\,0$

For $t < 0$, one obtains $\Phi(t)$ from the relationship $\Phi(t) = 1 - \Phi(-t)$.

For $0{,}5 \leqslant p < 1$, the approximation (see [14] in the Bibliography):

$$k_p = t - \frac{b_0 + b_1 \cdot t + b_2 \cdot t^2}{1 + c_1 \cdot t + c_2 \cdot t^2 + c_3 \cdot t^3} + \varepsilon;\ t = \sqrt{-2 \cdot \ln(1 - p)}$$

is valid with $|\varepsilon| < 4{,}5 \times 10^{-4}$ and

$b_0 = 2{,}515\,517$; $b_1 = 0{,}802\,853$; $b_2 = 0{,}010\,328$

$c_1 = 1{,}432\,788$; $c_2 = 0{,}189\,269$; $c_3 = 0{,}001\,308$

For $0 < p < 0{,}5$, one obtains k_p from the relationship $k_p = -k_{1-p}$.

Table 1 — Values of the distribution function of the standardized normal distribution $\Phi(t)$
(see [7] in the Bibliography)

t	$\Phi(t)$	t	$\Phi(t)$	t	$\Phi(t)$	t	$\Phi(t)$	t	$\Phi(t)$
0,00	0,500 0	0,70	0,758 0	1,40	0,919 2	2,10	0,982 1	2,80	0,997 4
0,02	0,508 0	0,72	0,764 2	1,42	0,922 2	2,12	0,983 0	2,90	0,998 1
0,04	0,516 0	0,74	0,770 4	1,44	0,925 1	2,14	0,983 8	3,00	0,998 6
0,06	0,523 9	0,76	0,776 4	1,46	0,927 8	2,16	0,984 6	3,10	0,999 0
0,08	0,531 9	0,78	0,782 3	1,48	0,930 6	2,18	0,985 4	3,20	0,999 3
0,10	0,539 8	0,80	0,788 1	1,50	0,933 2	2,20	0,986 1	3,30	0,999 5
0,12	0,547 8	0,82	0,793 9	1,52	0,935 7	2,22	0,986 8	3,40	0,999 7
0,14	0,555 7	0,84	0,799 6	1,54	0,938 2	2,24	0,987 4	3,50	0,999 8
0,16	0,563 6	0,86	0,805 1	1,56	0,940 6	2,26	0,988 1	3,60	0,999 8
0,18	0,571 4	0,88	0,810 6	1,58	0,943 0	2,28	0,988 7	3,80	0,999 9
0,20	0,579 3	0,90	0,815 9	1,60	0,945 2	2,30	0,989 3	4,00	1,000 0
0,22	0,587 1	0,92	0,821 2	1,62	0,947 4	2,32	0,989 8		
0,24	0,594 8	0,94	0,826 4	1,64	0,949 5	2,34	0,990 4		
0,26	0,602 0	0,96	0,831 5	1,66	0,951 5	2,36	0,990 9		
0,28	0,610 3	0,98	0,836 5	1,68	0,953 5	2,38	0,991 3		
0,30	0,617 9	1,00	0,841 3	1,70	0,955 4	2,40	0,991 8		
0,32	0,625 5	1,02	0,846 1	1,72	0,957 3	2,42	0,992 2		
0,34	0,633 1	1,04	0,850 8	1,74	0,959 1	2,44	0,992 7		
0,36	0,640 6	1,06	0,855 4	1,76	0,961 0	2,46	0,993 0		
0,38	0,648 0	1,08	0,859 9	1,78	0,962 5	2,48	0,993 4		
0,40	0,655 4	1,10	0,864 3	1,80	0,964 1	2,50	0,993 8		
0,42	0,662 8	1,12	0,868 6	1,82	0,965 6	2,52	0,994 1		
0,44	0,670 0	1,14	0,872 9	1,84	0,967 1	2,54	0,994 5		
0,46	0,677 2	1,16	0,877 0	1,86	0,968 6	2,56	0,994 8		
0,48	0,684 4	1,18	0,881 0	1,88	0,970 0	2,58	0,995 1		
0,50	0,691 5	1,20	0,884 9	1,90	0,971 3	2,60	0,995 3		
0,52	0,698 5	1,22	0,888 8	1,92	0,972 6	2,62	0,995 6		
0,54	0,705 4	1,24	0,892 5	1,94	0,973 8	2,64	0,995 9		
0,56	0,712 3	1,26	0,896 1	1,96	0,975 0	2,66	0,996 1		
0,58	0,719 0	1,28	0,899 7	1,98	0,976 2	2,68	0,996 3		
0,60	0,725 8	1,30	0,903 2	2,00	0,977 2	2,70	0,996 5		
0,62	0,732 4	1,32	0,906 6	2,02	0,978 3	2,72	0,996 7		
0,64	0,738 9	1,34	0,909 9	2,04	0,979 3	2,74	0,996 9		
0,66	0,745 4	1,36	0,913 1	2,06	0,980 3	2,76	0,997 1		
0,68	0,751 8	1,38	0,916 2	2,08	0,981 2	2,78	0,997 3		

Annex A
(informative)

Example of application of this part of ISO 11929

A.1 Principles of unfolding using a Bayesian theory of uncertainty

There are two classes of physical quantities to be distinguished in the evaluation of measurements.

Resulting quantities (further on in the text called output quantities) Y_k ($k = 1, ..., n$) are quantities (for instance, the parameters of an unfolding procedure) which have to be determined by the evaluation of the measurement. The decision quantity X is one of them. The task is to calculate the estimates y_k of the output quantities as the results of the measurements, the standard uncertainties $u(y_k)$ associated with y_k and the covariances of the measurement uncertainties $u(y_k, y_l)$. It holds that $u^2(y_k) = u(y_k, y_k)$.

Input quantities X_i ($i = 1, ..., m$) are quantities such as ρ_i or $\rho_i \cdot t_i$ which are, for instance, derived by counting measurements. Further, they are repeatedly measured quantities, influence quantities and output quantities of previous evaluations. The estimators x_i of these input quantities and the standard uncertainties $u(x_i)$ associated with the x_i and the covariances $u(x_i, x_j)$ are either given, or have to be determined, following the procedures of the Guide to the expression of uncertainty in measurement. In counting measurements, one obtains for the quantities ρ_i, derived according to A.3 and Equation (A.9), with the counting result n_i and the counting time (or channel width) t_i: $x_i = n_i/t_i$, $u^2(x_i) = n_i/t_i^2 = x_i/t_i$, $u(x_i, x_j) = 0$ (with respect to $n_i = 0$, see A.3).

The model of the evaluation connects the output quantities mathematically with the input quantities:

$$Y_k = G_k\,(X_1, ..., X_m); (k = 1, ..., n) \tag{A.1}$$

The functions G_k do not need to be explicitly available as mathematical expressions. They may also be an algorithm, for instance, in the form of a computer code of the evaluation.

The measuring results y_k are obtained by substituting the input quantities X_i in the model equations G_k by their estimates x_i:

$$y_k = G_k(x_1, ..., x_m); (k = 1, ..., n) \tag{A.2}$$

The covariances $u(y_k, y_l)$ of their uncertainties are given by:

$$u(y_k, y_l) = \sum_{i,\,j\,=\,1}^{m} \frac{\partial G_k}{\partial X_i} \cdot \frac{\partial G_l}{\partial X_j} \cdot u\,(x_i, x_j); (k, l = 1, ..., n) \tag{A.3}$$

$u(y_k)$ is the positive square root of $u(y_k, y_k)$.

The partial derivatives need not to be explicitly calculated. This is particularly advantageous if such a calculation is difficult, or if the model equations are only available as a computer code. It is sufficient to calculate first the differential quotients:

$$\Delta_i G_k = \left\{ G_k\left[x_1, ..., x_i + \frac{1}{2}u(x_i), ..., x_m \right] - G_k\left[x_1, ..., x_i - \frac{1}{2}u(x_i), ..., x_m \right] / \, u(x_i) \right\} \tag{A.4}$$

and then

$$u(y_k, y_l) = \sum_{i,j=1}^{m} (\Delta_i G_k) \cdot (\Delta_i G_l) \cdot u(x_i, x_l); (k, l = 1, ..., n) \quad \text{(A.5)}$$

This procedure is particularly advantageous in computerized evaluation. Examples of computer codes are given, for instance, in [15] and [16] in the Bibliography. The partial derivatives may also be obtained in an analogous way experimentally by changing the input quantities by Δx_i, because one can approximate Equation (A.4) to

$$\Delta_i G_k = [G_k(x_1, ..., x_i + \Delta x_i - y_k] / \Delta x_i \quad \text{(A.6)}$$

Note that Equation (A.6) has a lower accuracy than Equation (A.4).

Let Y_1 be the decision quantity X. Then $x = y_1$ and $u(x) = u(y_1)$. In order to calculate $\tilde{u}(\xi)$ an (at least approximatively) inverse of the model shall be given for $m' \leqslant m$ quantities X_i ($i = 1, ..., m'$), the uncertainties of which depend on the true value ξ of the measurand:

$$X_i = M_i(Y_1, ..., Y_n, X_{m'+1}, ..., X_m); (i = 1, ..., m') \quad \text{(A.7)}$$

In this case, the fixed value ξ has to be substituted for $Y_1 = X$. One obtains changed estimates:

$$x'_i = M_i(\xi, y_2, ..., y_n, x_{m'+1}, ..., x_m); (i = 1, ..., m') \quad \text{(A.8)}$$

which then lead to changed covariances $u(x'_i, x'_j)$ of the uncertainties. With these changed covariances, the entire calculation according to Equations (A.1) to (A.5) has to be repeated. However, one only needs to calculate $\tilde{u}(\xi) = u(y_1)$. If a computer code operates iteratively, repetition of one iterative step is frequently sufficient.

A.2 General numerical calculation of uncertainty in measurement

In all types of unfolding, functions M_i according to Equation (A.7) are fitted to the estimates x_i of the input quantities X_i ($i = 1, ..., m' < m$). In spectrum unfolding, which is a special case of general unfolding methods, the channel counts n_i are the estimates of the input quantities $\rho_i \cdot t_i$ associated with the individual channels i.

Those input quantities X_i ($i > m'$) are parameters which are uncertain but which are not adjusted by the fitting procedure. The algorithm to solve such an unfolding problem, for instance, the method of least-squares adjustment according to [16] in the Bibliography, can be described by a model according to Equation (2). Usually, it is used in the form of a computer code. Further calculation makes use of A.1 Equation (A.7). $z_i = M_i(y_1, ..., y_n, x_{m'+1}, ..., x_m)$ ($i < m'$) are the fitted values of x_i. For the estimates $x_i = n_i/t$ of the spectral densities, $u^2(x'_i) = x'_i/t$ et $u(x'_i, x'_j) = 0$ hold.

A detailed description of the mathematical foundation of an analytical approach to Bayesian-statistical spectrum unfolding using Monte Carlo techniques is given in [8] in the Bibliography.

NOTE The measuring times associated with the individual channels may not necessarily be identical.

A.3 Examples of application of this part of ISO 11929

A.3.1 General

In general, a measurement of nuclear radiation consists, at least partially, in counting electronic pulses caused by nuclear radiation events. Such a counting measurement usually comprises several individual counting measurements. Examples are counting measurements of individual radioactive samples or blank materials, counting measurements of the natural radiation background or of another background effect, and counting of

events in the individual channels of a multi-channel spectrum or a time sequence of events under identical measuring conditions. In each counting measurement, either the counting time or maximum number of counts is preselected. On the basis of Bayesian statistics, all counting measurements are treated as described below (see [7] in the Bibliography).

An individual random variable N is assigned to the number of pulses counted in each individual counting measurement. n is the result of the measurement and t the counting time. N has the expectation $\rho \cdot t$, with ρ being the count rate, respectively the spectral density in spectrometric measurements. In the latter case, t is the channel width of the corresponding physical quantity, for example particle energy. The measurand is either ρ or $\rho \cdot t$. It is supposed that dead-time or lifetime effects, pile-up of pulses and instrumental instabilities can be neglected during the entire measurement, and that the pulses counted originate from different nuclear radiation events which are physically independent from each other. Then, the number of counts N is Poisson distributed and the number of counts obtained in individual countings are independent.

The quantity obeys a Gamma distribution, whereby ρ is considered to be a random variable. This is independent of whether n counts are registered during a preselected counting time (or a fixed channel width) t or whether the time t, is measured which is required to accumulate a preselected maximum number of counts. Then, one obtains the best estimate r of the count rate (or the spectral density) ρ and the standard uncertainty $u(r)$ associated with r:

$$r = \mathrm{E}\,\rho = n/t; u^2(r) = \mathrm{Var}(\rho) = n/t^2 \tag{A.9}$$

In the case of $n = 0$, one obtains $u(r) = 0$. This is not realistic, since one cannot be sure that $\rho = 0$ if no count has been obtained during a measurement with finite counting time. This case leads also to a division by zero if the method of least squares is applied (for example, according to DIN 1319-4) when one has to divide by $u^2(r)$. This difficulty can be avoided by replacing all counting results n by $n + 1$, or by a suitable combination of channels. Thereby, it is presumed that the counting time (or the channel width) has been chosen in such a way that at least some counts are to be expected if $\rho > 0$.

A.3.2 Spectrum unfolding in nuclear spectrometric measurement

The evaluation of a nuclear spectrometric measurement usually is an (in general non-linear) unfolding of a measured multi-channel spectrum. It can also comprise the unfolding of several measured spectra and consideration of other data. Such an evaluation is shortly called spectrum unfolding.

The input quantities X_i of the spectrum unfolding are all quantities from which measured data or other data are used in the unfolding, and which have uncertainties associated with them. These are all those quantities X_i for which a measured or estimated value x_i exists and which shall be fitted in the unfolding procedure. One of those quantities X_i is to be assigned to each individual channel of a multi-channel spectrum, that is the number of counts n_i in channel i counted during a measurement of duration t. Likewise, an input quantity X_i has to be assigned to each parameter to be determined for which an estimate is given before the evaluation. Such a parameter can be, for instance, spectrum parameters, such as the widths of spectral lines or parameters of the sensitivity matrix of the spectrometer.

Further, there are input quantities t_i for which estimates exist, but which are not subject to fit. These are, for instance, base points, calibration parameters, correction and influence quantities or other parameters which were already previously mentioned. The values ϑ_i, connected with channel i of the parameter ϑ related to the different channels in a multi-channel spectrum, are such quantities.

In principle, all quantities for which an estimate exists should be fitted. Frequently, however, this is not technically feasible, or some quantities were determined from other experiments so that it is not meaningful to fit them too. Such quantities, which are known sufficiently exact so that their uncertainties are negligible, are not treated as input quantities but as constants. If only the Poisson-statistics of the channel counts of a multi-channel spectrum shall be considered, only these quantities are input quantities. In this case, all other quantities are constants.

The output quantities Y_i of the spectrum unfolding are the parameters of the unfolding which have to be determined. The measurand for which the decision threshold, detection limit and the limits of the confidence interval have to be determined is one of them. The number of these parameters should be as small as possible. The output quantities can also be spectrum parameters, such as net areas of spectral lines or the number of background counts under a spectral line or in a particular channel or unknown parameters of the sensitivity matrix.

For spectrum unfolding, it is convenient to use matrix notation. Therefore, those quantities, values and functions being denoted by the same symbol are written as a column matrix. The same symbol is used for them, written, however, in bold face; for instance the estimates of the input quantities $\boldsymbol{x} = (x_1, x_2, ...)^T$ and $\boldsymbol{t} = (t_1, t_2, ...)^T$, the values of the output quantities $\boldsymbol{y} = (y_1, y_2, ..., y_n)^T$ and the best estimates of the input quantities $z = (z_1\ z_2\ ...)^T$. (Column matrices are written as transposed row matrices.) For convenience, the vectors $\boldsymbol{x}$ and $\boldsymbol{t}$ are combined to a vector $\boldsymbol{v} = (x_1, x_2, ..., t_1, t_2, ...)^T$.

For the unfolding, one needs the estimates $\boldsymbol{x}$ and $\boldsymbol{t}$ of the input quantities and their associated uncertainty matrix $\boldsymbol{U}_{\boldsymbol{v}} = \boldsymbol{U}(\boldsymbol{x}, \boldsymbol{t})$. This uncertainty matrix has to be calculated as a covariance matrix according to the Guide for the expression of uncertainty in measurement. The uncertainty matrix $\boldsymbol{U}(\boldsymbol{x}, \boldsymbol{t})$ is needed in the form of its functional dependence on $\boldsymbol{x}$, since $\boldsymbol{x}$ has to be adjusted if the decision threshold and detection limit are calculated while $\boldsymbol{t}$ stays constant. The uncertainty matrices $\boldsymbol{U}_{\boldsymbol{x}}$ and $\boldsymbol{U}_{\boldsymbol{t}}$ associated with $\boldsymbol{x}$ and $\boldsymbol{t}$ are partial matrices of $\boldsymbol{U}_{\boldsymbol{v}}$. The rank of $\boldsymbol{U}_{\boldsymbol{x}}$ shall not be smaller than the number of model equations. If the data for $\boldsymbol{x}$ and $\boldsymbol{t}$ originate from different independent experiments, then there is no correlation between $\boldsymbol{x}$ and $\boldsymbol{t}$, and the matrix elements of $\boldsymbol{U}_{\boldsymbol{v}}$ related to pairs x_i and t_k are zero. The diagonal element related to a quantity q is denoted $u^2(q)$. $u(q)$ is the standard uncertainty associated with q.

A more detailed description of spectrum unfolding in nuclear spectrometric measurements is given elsewhere (see [4], [7] and [8] in the Bibliography).

The model of the unfolding consists of n relationships between input and output quantities. These relationships can formally, and more generally, be written as a column matrix $\boldsymbol{M}$ of model functions M_i which depend on all these quantities:

$$\boldsymbol{M}(x, y, t) = \boldsymbol{0} \tag{A.10}$$

with **0** being the zero vector.

A model of linear unfolding can, for instance, be written with a sensitivity matrix $\boldsymbol{A}$ as:

$$\boldsymbol{M}(x, y, t) = \boldsymbol{A}(t)y - x = \boldsymbol{0} \tag{A.11}$$

In non-linear unfolding, the model is frequently of the form:

$$\boldsymbol{M}(x, y, t) = \boldsymbol{J}(t, y) - x = \boldsymbol{0} \tag{A.12}$$

If an output quantity Y_i is likewise an input quantity X_i for which an estimate x_i exists, the Equation $y_i - x_i = 0$ has to added to the model functions. If output quantities, such as for example, instance activity, particle fluence or equivalent dose, depend on other output quantities, the respective functional dependencies have also to be added to the model functions. The model functions do not need to be explicitly available as mathematical expressions. They may also be an algorithm, for instance in the form of a computer code of the evaluation.

For the unfolding of a measured multi-channel spectrum, one fits functions $\boldsymbol{M}(\boldsymbol{Y})$ according to Equation (A.7) 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 calculation of the best estimates $\boldsymbol{y}$ of the output quantities $\boldsymbol{Y}$, the uncertainty matrix $\boldsymbol{U}_{\boldsymbol{y}}$ associated with $\boldsymbol{y}$ and of the fitted values (best estimates) $\boldsymbol{z}$ of the input quantities $\boldsymbol{X}$ from the given measured and estimated

values of all input quantities $\boldsymbol{v}$ with their associated uncertainty matrix $\boldsymbol{U_v}$, is in general a non-linear fitting procedure, the solution of which has, for instance, been described in [4] in the Bibliography.

In the special case of a spectrum unfolding which is linear in the parameters $\boldsymbol{Y}$, the spectral density $H(\vartheta)$ is represented by the column matrix $\boldsymbol{X} = [H(\vartheta_i)]$. The ϑ_i are assumingly exact base points, for instance, the energies or times assigned to the individual channels. The spectral density is approximated by a system of functions $\Psi_k(\vartheta)$:

$$X_i = H(\vartheta_i) = \sum_{k=1}^{n} \Psi_k(\vartheta_i) \cdot Y_k; (i = 1, ..., m) \text{ ou } \boldsymbol{X} = \boldsymbol{M}(Y) = \boldsymbol{AY} \tag{A.13}$$

The constant sensitivity matrix $\boldsymbol{A}$ consists of the elements $A_{ik} = \Psi_k(\vartheta_i)$. The functions $\Psi_k(\vartheta_i)$ describe the shape of the individual spectral lines and of the background contributions. (For explicit examples of $\Psi_k(\vartheta_i)$ when evaluating alpha- and gamma-spectra, see A.3.4 and A.3.5.) The output quantities Y_k to be determined are, for instance, the net peak areas of the spectral lines.

The components of uncertainty of measurement comprise uncertainty matrices $\boldsymbol{U_x} = [u(x_i, x_j)]$ and $\boldsymbol{U_y} = [u(y_k, y_l)]$. $\boldsymbol{U_x}$ is the diagonal with the diagonal elements $u^2(x_i) = n_i/t^2$, i.e. $\boldsymbol{U_x} = \text{diag}\ (x_i/t)$.

Using the method of least squares one obtains:

$$\boldsymbol{y} = \boldsymbol{G}(x) = \boldsymbol{U_y}\, A^{\mathsf{T}} \boldsymbol{U}_{\boldsymbol{x}}^{-1} \boldsymbol{x}; \boldsymbol{z} = \boldsymbol{M}(y) = \boldsymbol{Ay}; \tag{A.14}$$

$$\boldsymbol{U_y} = (\boldsymbol{A}^{\mathsf{T}} \boldsymbol{U}_{\boldsymbol{x}}^{-1} \boldsymbol{A})^{-1} \tag{A.15}$$

If, for instance, Y_1 is the decision quantity X, y_1 is replaced by ξ, in order to calculate $\tilde{u}(\xi)$. This yields $\boldsymbol{y'} = (\xi,\ y_2, ...)^{\mathsf{T}}$ and $\boldsymbol{x'} = \boldsymbol{M}(\boldsymbol{y'}) = \boldsymbol{A}\ \boldsymbol{y'}$. From this, one calculates the changed uncertainty matrix $\boldsymbol{U_{x'}} = \text{diag}\ (x'_i/t)$ and inserts it into Equation (A.15). The (1,1)-element of the changed uncertainty matrix $\boldsymbol{U_y}$ is then $\tilde{u}^2(\xi)$.

A.3.3 Simple examples of linear unfolding in nuclear spectrometric measurements

The classical methods of linear or third-order polynomial background subtraction, for the determination of a net peak area of a spectral line of a spectrum, are simple examples of linear spectrum unfolding as described below.

Independent, Poisson distributed random variables N_i ($i = 1, ..., m$ and $i = g$) are assigned to selected channels of a multi-channel spectrum having the channel counts n_i. If needed, several channels of a channel area can be comprised in one channel. For the following, ϑ_i is the lower limit and ϑ_i' the upper limit of channel i; ϑ is for instance energy or time; $t_i = \vartheta'_i - \vartheta_i$ is the channel width. N_g represents the combined channel counts of the spectral line under investigation in the spectrum. The net peak area is the measurand with the true value ξ, that means the net number of counts of the channel $i = g$. A function $H(\vartheta, y_i, ..., y_m)$ with parameters y_k suitable to represent the spectral density of the background under the spectral line is adjusted so that:

$$n_i = \int_{\vartheta_i}^{\vartheta'_i} H(\vartheta, y_1, ..., y_m)\, d\vartheta; (i = 1, ..., m) \tag{A.16}$$

From this, y_k have to be calculated as functions of n_i. The background contribution to the gross peak area is then:

$$z_0 = \int_{\vartheta_i}^{\vartheta'_i} H(\vartheta, y_1, ..., y_m)\, d\vartheta \tag{A.17}$$

With a random variable Z_0 for z_0, an expression for the decision quantity is:

$$X = \rho_g t_g - Z_0 \tag{A.18}$$

With this, one obtains:

$$x = n_g - z_0; u^2(x) = n_g + u^2(z_0); u^2(z_0) = \sum_{i=1}^{m} \left(\sum_{k=1}^{m} \frac{\partial z_0}{\partial y_k} \cdot \frac{\partial y_k}{\partial n_i} \right)^2 \cdot n_i \qquad \text{(A.19)}$$

In this equation, the expression in brackets is equal to $\partial z_0/\partial n_i$. It can be calculated using Equations (A.20) and (A.1). For a given true value ξ of the net peak area in channel g, one has to replace x in Equation (A.19) by ξ. Thereby, one can eliminate n_g which is not known for a given true value ξ. On receives with $n_g = \xi + z_0$:

$$\tilde{u}^2(\xi) = \xi + z_0 + u^2(z_0) \qquad \text{(A.20)}$$

Various procedures may be used to determine net peak areas in a multi-channel spectrum. If linear background subtraction is applied (as in ISO 11929-3), the above methods are used with $m = 2$ and a linear function $H(\vartheta_i)$. If a third-order polynomial is assumed to represent the background under the spectral line, $m = 4$ and $H(\vartheta_i)$ is a third-order polynomial.

A more general expression is $H(\vartheta) = \sum_{k=1}^{m} y_k \cdot \Psi_k(\vartheta)$, which is linear in y_k and comprises given functions $\Psi_k(\vartheta)$ representing the individual shapes of the spectral lines. For this, expression (A.16) becomes a system of linear equations for the calculation of y_k. In this case, the y_k depend linearly on the channel counts n_i, but the partial derivatives in Equation (A.19) do not depend on the channel counts n_i. This latter method is, for instance, used for example in [18] in the Bibliography, in the case of alpha-spectrometric measurements (see also A.3.4).

A.3.4 Application to alpha-spectrometry

In many cases of alpha-spectrometry, an alpha-spectrum measured by a semiconductor detector or a grid ionization chamber can be considered to be a superposition of individual spectral lines, usually without a background contribution. In this case, a linear model of the type $\boldsymbol{A(t)y = x}$ can be used. The following functional representation can be used for the shape of a spectral line j (see [10] to [13] in the Bibliography):

$$L_j(E) = \int_{-\infty}^{\infty} \frac{R_j(E')}{\sqrt{2\pi\sigma_j^2}} \exp\left(-\frac{(E'-E)^2}{2\sigma_j^2} \right) \, \mathrm{d}E'$$

$$R_j(E) = \alpha_{0,j} \cdot \delta(E - E_{0,j}) + \sum_{k=1}^{3} (\alpha_{k,j}/\tau_{k,j}) \cdot \exp[(E - E_{0,j})/\tau_{k,j}] \quad \text{for} \quad (E \leqslant E_{0,j}) \qquad \text{(A.21)}$$

$R_j(E) = 0$ for $(E > E_{0,j})$:

$$\alpha_{0,j} + \alpha_{1,j} + \alpha_{2,j} + \alpha_{3,j} = 1$$

E is the energy of the alpha-particles. All other quantities are given parameters of the spectral line. These parameters of all spectral lines as well as the parameters of the energy calibration which connects the channel number i and the energy E, are input quantities $\boldsymbol{t}$ or are considered to be known constants. The elements of the sensitivity matrix are $A_{ij} = L_j(E_i)$, with E_i being the energy associated with channel i.

The spectral line at alpha-energy E_{0j} is physically characterized by the first term of $R_j(E)$, the delta function, in Equation (A.21). The three following terms describe the energy loss of the alpha-particles on their path to detection. The folding integral in the first line of Equation (A.21) considers the resolution σ of the spectrometer which usually depends on E.

The parameters y_j to be determined are the peak areas. They form the vector $\boldsymbol{y}$ of the output quantities. One of them is the quantity Y in question. For the spectrum, one hence receives the functional expression $x_i = \Sigma_j L_j \cdot y_j$ or, written as a vector with $E = E_i$ for the individual channels, $\boldsymbol{x} = \boldsymbol{A}(\boldsymbol{t})\boldsymbol{y}$.

If some of the parameters $\boldsymbol{t}$, for instance, the location of the spectral lines or parameters of the peak shape are not known but are likewise to be determined by the unfolding, then these unknown quantities are to be added to the output quantities $\boldsymbol{y}$. Then, $\boldsymbol{A}$ depends also on $\boldsymbol{y}$ and one obtains the case of a non-linear model according to Equation (A.12) with $\boldsymbol{J}(\boldsymbol{y}, \boldsymbol{t}) = \boldsymbol{A}(\boldsymbol{y}, \boldsymbol{t})\boldsymbol{y}$.

The functions $L_j(E)$ are the response functions of the spectrometer which can, for instance, be a semiconductor detector or a grid ionization 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 the set up as is required from 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, one can not only describe shapes of spectral lines. Also, the background under spectral lines can be modelled by superposition of such functions in any arbitrary way. For its application to gamma-spectrometric measurements see A.3.5.

A.3.5 Application to gamma-spectrometry

In gamma-spectrometry, background contributions generally must not be neglected. In spite of that, a linear model of the type $\boldsymbol{A}(\boldsymbol{t})\boldsymbol{y} = \boldsymbol{x}$ can be set up using the general procedure in A.3.4. 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:

$$\begin{aligned} L_1(E) &= \exp\left[-(E-E_0)^2/(2\sigma^2)\right]/\sqrt{2\pi\sigma^2} \\ L_2(E) &= \arctan\left[-(E-E_0)/a\right] \\ L_j(E) &= (E-E_0)^{j-3};\ (j = 3, 4, 5, 6) \end{aligned} \qquad \text{(A.22)}$$

The first function in Equation (A.22) describes the shape of a spectral line by a Gaussian with a spectrometer resolution σ. In actual cases, it may be necessary to use more complicated line shapes introducing, for instance, low-energy exponential tailings which increases the number of parameters of the peak shape. The second line of Equation (A.22) represents a “step function” under a spectral line, which has to be expected as a consequence of incomplete charge collection. a is a parameter characterizing the steepness of the step function and must be known beforehand. The residual functions in the third line of Equation (A.22) are used to model phenomenologically the background by a third-order polynomial. Instead of the energy E, the channel number i can also be used in the set-up of the model according to Equation (A.22).

E is the energy deposited in the detector. All other quantities are parameters of the spectral line or of the background step function. These parameters σ and a of all spectral lines, as well as of the energy calibration which connects the channel number i and the energy E, are input quantities $\boldsymbol{t}$ or are considered to be known constants. The elements of the sensitivity matrix are $A_{ij} = L_j(E_i)$, with E_i being the energy associated with channel i.

The parameters y_j to be determined are the peak areas and the background contributions. They form the vector $\boldsymbol{y}$ of the output quantities. One of the peak areas is the quantity Y in question. For the spectrum, one hence receives the functional expression $x_i = \Sigma_j L_j \cdot y_j$ or, written as a vector with $E = E_i$ for the individual channels, $\boldsymbol{A}(\boldsymbol{t})\boldsymbol{y}$.

In contrast to alpha-spectrometry, in gamma-spectrometry frequently some of the parameters $\boldsymbol{t}$ are not known exactly, for instance, the location of the spectral lines or parameters of the peak or background shape. 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 $\boldsymbol{y}$ and one obtains the case,

which is usual in complex gamma-spectrometry, of a non-linear model according to Equation (A.12) with $\boldsymbol{J}(\boldsymbol{y}, \boldsymbol{t}) = \boldsymbol{A}(\boldsymbol{y}, \boldsymbol{t})\boldsymbol{y}$.

Consequently, the model $\boldsymbol{M}(\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{t}) = \boldsymbol{J}(\boldsymbol{t}, \boldsymbol{y}) - \boldsymbol{x} = \mathbf{0}$ of a non-linear unfolding according to Equation (A.12) has to be used. The algorithm of this model can be written formally with suitable functions $\boldsymbol{F}$ and $\boldsymbol{G}$ in the form

$$\boldsymbol{y} = \boldsymbol{F}(\boldsymbol{x}, \boldsymbol{t}) = \boldsymbol{F}(\boldsymbol{v});\ \boldsymbol{z} = \boldsymbol{G}(\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{t}) = H(\boldsymbol{v}) \tag{A.23}$$

with $\boldsymbol{H}(\boldsymbol{v}) = \boldsymbol{G}[\boldsymbol{x}, \boldsymbol{F}\ (\boldsymbol{x}, \boldsymbol{t}), \boldsymbol{t}]$.

The first expression of Equation (A.23) represents formally the solution of the model Equation (7) for $\boldsymbol{y}$. The second expression yields the fitted data $\boldsymbol{z}$ as best estimates of the input quantities resulting from the fit.

The given functions or algorithms according to Equation (A.23) are formally sufficient to calculate the output quantities $\boldsymbol{y}$ and the uncertainty matrix $\boldsymbol{U}_{\boldsymbol{y}}$ associated with $\boldsymbol{y}$ and the best estimates $\boldsymbol{z}$ of the input quantities $\boldsymbol{x}$ from the given measured and estimated data of the input quantities $\boldsymbol{v}$ and the uncertainty matrix $\boldsymbol{U}_{\boldsymbol{v}}$ associated with $\boldsymbol{v}$ by non-linear unfolding.

The uncertainty matrices $\boldsymbol{U}_{\boldsymbol{y}}$ and $\boldsymbol{U}_{\boldsymbol{z}}$ associated with the quantities $\boldsymbol{y}$ and $\boldsymbol{z}$ are, according to the Guide to the Expression of Uncertainty in Measurement, given by:

$$\boldsymbol{U}_{\boldsymbol{y}} = \boldsymbol{F}_{\boldsymbol{v}}\boldsymbol{U}_{\boldsymbol{v}}\boldsymbol{F}_{\boldsymbol{v}}^{\mathsf{T}};\ \boldsymbol{U}_{\boldsymbol{z}} = \boldsymbol{H}_{\boldsymbol{v}}\boldsymbol{U}_{v}\boldsymbol{H}_{\boldsymbol{v}}^{\mathsf{T}} \tag{A.24}$$

with $\boldsymbol{F}_{\boldsymbol{v}}$ and $\boldsymbol{H}_{\boldsymbol{v}}$ denoting the matrices of the partial derivatives of the model functions:

$$\boldsymbol{F}_{\boldsymbol{v}} \equiv (\partial F_i / \partial x_k) \text{ and } \boldsymbol{H}_{\boldsymbol{v}} \equiv (\partial H_i / \partial x_k)$$

The most frequently used method for unfolding is the Method of Least Squares, for example, according to [16] in the Bibliography $\boldsymbol{y}$ and $\boldsymbol{z}$ have to be determined so that:

$$\chi^2 = (\boldsymbol{z} - \boldsymbol{x})^{\mathsf{T}}\ \boldsymbol{U}_{\boldsymbol{x}}^{-1}(\boldsymbol{z} - \boldsymbol{x}) = \min \tag{A.25}$$

under the condition:

$$\boldsymbol{M}(z, y, t) = \mathbf{0} \tag{A.26}$$

following from the substitution of $\boldsymbol{x}$ by $\boldsymbol{z}$ in Equation (7). For details of the calculational procedures and for tests whether the model used, the solution of the unfolding procedure and the input data are compatible, see [4] and [16] in the Bibliography.

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