Workshop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

5 - 16 November 2007

Uncertainty in Gamma Spectrometry.

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UNCERTAINTY IN GAMMA SPECTROMETRY

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<table>
<thead>
<tr>
<th>MODULE</th>
<th>TITLE</th>
<th>TIME</th>
<th>DURATION</th>
</tr>
</thead>
</table>
| 1      | • Metrology, measurement fundamentals  
  o Quantity of interest in **gamma spectrometry**  
  o Relative and absolute measurements  
  o Instrument calibration  
  o Efficiency transfer  
  o Nuclear data | 12/11/07 8:30-10:30 | 2:00 |
| 2      | • Uncertainty evaluation theory  
  o Fundamentals  
  o Statistical quantities  
  o Statistics applied to counting  
  o Uncertainty propagation | 12/11/07 11:00-12:30 | 1:30 |
| 3      | • Uncertainty components in **gamma spectrometry** | | |
| 4      | • Methods for uncertainty determination | | |
| 5      | • Applications, Exercises, Examples, Discussion | 12/11/07 14:00-16:45 | 2:45 |
| 6      | • Characteristic limits  
  o Theory  
  o Application to **gamma spectrometry** | 13/11/07 8:30-10:30 | 2:00 |
| 7      | • Quality control | | |
| 8      | • Discussion with participants | 13/11/07 11:00-12:30 | 2:30 |
Graphical illustration of values, error, and uncertainty

[ISO (1993), Guide to the expression of uncertainty]
Mathematical expressions

**Error propagation law**

\[ Y = f(x_1, x_2, ..., x_p) \]

\[ S_Y^2 = \sum_{i=1}^{p} \left( \frac{\partial f}{\partial x_i} \right)^2 S_{x_i}^2 + 2 \sum_{i<k} \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \text{cov}(x_i, x_j) \]

**Sample estimates**

\[ S_{x_i}^2 = \frac{1}{n-1} \sum_{k=1}^{n} (x_{ik} - \bar{x}_i)^2 \]

\[ \text{cov}(x_i, x_j) = \frac{1}{n-1} \sum_{k=1}^{n} (x_{ik} - \bar{x}_i)(x_{jk} - \bar{x}_j) \]

**Useful expressions**

\[ \begin{cases} x_i = x_i(z, ...) \\ x_j = x_j(z, ...) \end{cases} \Rightarrow \text{cov}(x_i, x_j) = \frac{\partial x_i}{\partial z} \frac{\partial x_j}{\partial z} S_z^2 \]

\[ \begin{cases} Y = \frac{x_1}{x_2} \\ Y = x_1x_2 \end{cases} \Rightarrow \left( \frac{S_Y}{Y} \right)^2 = \left( \frac{S_{x_1}}{X_1} \right)^2 + \left( \frac{S_{x_2}}{X_2} \right)^2 \]

\[ \begin{align*}
Y = \frac{x_1}{x_2} & \Rightarrow \left\{ \begin{array}{l}
\frac{1}{Y} \frac{\partial Y}{\partial x_1} = x_1 \\
\frac{1}{Y} \frac{\partial Y}{\partial x_2} = -\frac{1}{x_2}
\end{array} \right. \\
\frac{\partial f}{\partial x_i} & \approx \frac{f(x_i + \Delta x_i) - f(x_i)}{\Delta x_i} \frac{f(x_i)}{f(x_i)}
\end{align*} \]
The following quantities are required for uncertainty evaluation:

\[ Y = f(x_1, x_2, ..., x_p) \]

\[ S_Y^2 = \sum_{i=1}^{p} \left( \frac{\partial f}{\partial x_i} \right)^2 S_{x_i}^2 + 2 \sum_{i<k}^{p} \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \text{cov}(x_i, x_j) \]

Obtained by measurement, calculation, estimation, importation, derivation, …

A MODEL IS NEEDED!
References

- ISO “GUM”
- Articles on Journals
- ENEA-INMRI laboratory results
Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

UNCERTAINTY IN GAMMA SPECTROMETRY

1. Metrology, measurement fundamentals
2. Uncertainty evaluation theory
3. Uncertainty components in gamma spectrometry
4. Methods for uncertainty determination
5. Application, exercises, Examples, Discussion
6. Characteristic limits
7. Quality control
8. Discussion with participants
QUANTITY OF INTEREST IN GAMMA SPECTROMETRY
### Main quantities of interest in gamma-spectrometry

<table>
<thead>
<tr>
<th></th>
<th>Nuclide identification (photon energy measurement)</th>
<th>Nuclide assay (activity measurement)</th>
<th>Special applications</th>
</tr>
</thead>
<tbody>
<tr>
<td>Full-energy-peak location (peak centroid)</td>
<td>X</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Peak width (i.e. FWHM)</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>Full-energy-peak, single- or double-escape-peak count rate</td>
<td>X</td>
<td>X</td>
<td></td>
</tr>
<tr>
<td>Total count rate</td>
<td></td>
<td></td>
<td>X</td>
</tr>
</tbody>
</table>
Full-energy-peak location and width

- Visual location of peaks is still a good method

- Automatic routines (large number of peaks, many spectra)
  - Adjust optimum sensitivity
  - Peak deconvolution
  - Accurate (±0.1 ch) peak centroid determination (not integer values)
  - Peak width (i.e. FWHM)
Peak area

- **Methods for determination:**
  - Sum of counts in ROI and background subtraction
  - Nonlinear least-squares methods (many peak shape functions)

- **Peak area: not well-defined quantity**

- **Peak area: defined by the method used to compute it**
Total count rate

- **Methods for determination:**
  - Sum of counts in the whole pulse-height spectrum, with linear extrapolation to zero count rate
  - Integral counting above an energy threshold

- **Total count rate:** well-defined quantity (for a given threshold)

- **Corrections for background and dead time required**
Linearity conditions

- Photon energy, $E$, and peak centroid, $c$, are linearly related:

$$E = a_1 + a_2 c$$

- Source activity, $A$, proportional to the peak count rate, $R$:

$$A \propto R = \frac{N}{T}$$

where $T =$ counting time
RELATIVE AND ABSOLUTE MEASUREMENT
CALIBRATION: set of operations that establish, under specified conditions, the relationship between values of quantities indicated by a measuring instrument or measuring system, or values represented by a material measure or a reference material, and the corresponding values realised by standards.

\[ E_s = a_1 + a_2 c \]

\[ A_s \propto R = \frac{N}{T} \]
Relative and absolute measurement methods

- **RELATIVE MEASUREMENT**: measurement performed by a comparison between the value of a quantity to be measured and a known value (given by a standard) of the same physical quantity.

- **ABSOLUTE MEASUREMENT**: measurement performed with reference to the definition of the physical quantity to be measured and with no reference to other values of the same physical quantity.
Example of a relative measurement

R: Instrument reading

\[ R = \varepsilon A \]

where:

A: Physical quantity

\( \varepsilon \): Proportionality constant

Under reproducible conditions (\( \varepsilon = \text{const.} \)):

\[ \varepsilon = \frac{R_S}{A_S} = \frac{R_x}{A_x} \]

where:

x: Problem values

S: Standard values

RELATIVE MEASUREMENT: measurement performed by a comparison between the value of a quantity to be measured and a known value (given by a standard) of the same physical quantity
Example of a relative measurement of radionuclide activity (gamma emitter)

A: Source Activity ($I_\gamma = 1$)

$$R = A \varepsilon$$

where:

R: Net count rate

$\varepsilon$: Counting efficiency

Under reproducible conditions ($\varepsilon = \text{const.}$):

$$R_x = A_x \varepsilon$$

$$R_S = A_S \varepsilon$$

where:

x: Problem source

S: Standard source

if $u(R_x) = u(R_S) = u(R)$

then $u(A_x)^2 = 2u(R)^2 + u(A_S)^2$

The constant $\varepsilon$ can be experimentally determined with a standard source (during system calibration) and used at a later date to measure the activity of other sources in the same experimental conditions.

To this purpose the instrument stability must be carefully checked by measuring a reference source (not necessarily calibrated) of some long lived radionuclide (such as Ra-226 or Eu-252) whenever a measurement is made.
Example of a relative measurement method (radionuclide activity)

- Activity measurement of a radioactive source by means of a substitution method (source to be measured and standard source) under reproducible conditions, using any instrument that records either individual radiations or else measures their ionising effects.

- Within the meaning of ‘reproducible conditions’ are to be included the geometry of the equipment and the disposition and quantity of any material absorbing or scattering the radiations.

- In these respects the characteristics of the source itself are no less important than those of the measuring instrument.

- The recording device used in the substitution method can be an electroscope, a beta particle or a gamma-ray ionisation chamber, any type of alpha-beta-gamma or x-ray detector.

- The measurement method is not directly based on the definition of the physical quantity.
Definitions of some general terms used in metrology
[BIPM/ISO International Vocabulary of basic and general terms in metrology]

**METROLOGY**: science of measurement

**VALUE** (of a quantity): magnitude of a particular quantity generally expressed as a unit of measurement multiplied by a number

**TRUE VALUE** (of a quantity): value consistent with the definition of a given particular quantity

**MEASUREMENT**: set of operations having the object of determining a value of a quantity

**METHOD OF MEASUREMENT**: logical sequence of operations, described generically, used in the execution of measurements

**ACCURACY OF MEASUREMENT**: closeness of the agreement between the result of a measurement and a true value of the measurand

**REPEATABILITY** (of results of measurements): closeness of the agreement between results of successive measurements of the same measurand carried out under the same conditions of measurement

**REPRODUCIBILITY** (of results of measurements): closeness of the agreement between results of measurements of the same measurand carried out under changed conditions of measurement
INSTRUMENT CALIBRATION
ENERGY CALIBRATION

Relationship under investigation:

\[ E = a_1 + a_2 c \]
Photon energy standards

- Need of precise ($\pm 0.1$ keV) energy-calibration standards
- Current system of energy-calibration standards (from a few keV to several MeV) available since 70', based on accurate measurements of gamma-ray wavelengths (cm)
- Preference for traditional energy scale (keV)
- Continuous update

For routine gamma-spectrometry some suitable references are:
- Browne and Firestone 1986
- NCRP 1985
- IAEA-TECDOC-619 Evaluated Data
- LNHB Nucleide Gamma and Alpha Library
- BIPM Monograph 5, Table of Radionuclides, 2004 (DDEP work)
Energy and FWHM calibration

- Energy measurement usually used just for nuclide identification
- FWHM measurements needed for system check and for nuclide identification (presence of multiplets)
- Energy values not involved in quantitative calculations
- Uncertainty evaluation not always required
- Importance of effect of change in energy calibration with time, count rate and temperature (Quality Control)
- Dependence on peak type (full-energy or s.e.p., d.e.p.) and algorithm used for determination
Energy and FWHM calibration

- Accuracy of 0.01-0.03 keV (from 100 to 1000 keV) sufficient for typical applications

- Functions:
  - linear: \[ E = a_1 + a_2 c \] usually adequate
  - quadratic: \[ E = a_1 + a_2 c + a_3 c^2 \] more accurate for old systems

- At least two (or three) well chosen points are needed

- Deviations (± tenths of channel) arise mainly from amplifier-ADC non linearity

- Advisable to avoid the first and last 10% of the energy scale

- More uniform distribution of uncertainties if more than 3 peaks are used and parameters \( a_i \) determined by least-squares fits

- Caution: multiplets, 511 keV annihilation peak
Parameters affecting energy calibration

- Electronics nonlinearity
- Pulse pile-up
- Detector effects:
  - Field-increment effect, depending on angle of incidence
  - Incomplete charge-collection, depending on detector volume
  - These effects determine a small (±0.2 keV) dependence of energy calibration (in principle not affected by source geometry) on measurement geometry
  - More evident for sources in front and beside a detector
EFFICIENCY CALIBRATION

Relationship under investigation:

\[ R = kA \]
A simple counting system

- Assume a radioactive source and a detector connected to a pulse counting system.
- The net count rate, $R$, is proportional to the source Activity, $A$:

$$R = \int_g \int_s \int_d I A$$

where $I$: emission probability for the considered radiation.

- The proportionality factor is due to physical effects that can be subdivided into 3 categories:
  1. Geometric effects, $f_g$: solid angle of the detector as viewed by the source.
  2. Source effects, $f_s$: source material and source construction.
  3. Detector effects, $f_d$: detector window, intrinsic detector efficiency.
Definition of efficiency

The efficiency, $\varepsilon$, is generally defined by:

$$\varepsilon = \frac{R}{X}$$

with:
- Instrument response, $R$
- Value of physical quantity, $X$

According to the chosen instrument resp. and physical quantity:
- Full-energy-peak count rate, $R_p$
- Total count rate, $R_t$
- Photon emission rate, $I_A$
- Activity, $A$

the following definition of efficiency apply:
- Full-energy-peak (fep) efficiency, $\varepsilon_p$
- Total efficiency, $\varepsilon_t$
- Nuclide-related peak efficiency, $\varepsilon_n$
- Nuclide-related total efficiency (counting efficiency), $\varepsilon_{tn}$

where $I$, is the photon emission probability.
Application of the different efficiencies

- Full-energy-peak efficiency, $\varepsilon_p$
  - application to many problems in routine measurements
  - quite easily interpolated as function of photon energy
  - need to be corrected for coincidence summing

- Total efficiency, $\varepsilon_t$
  - not used in spectrometry (energy information is lost)
  - useful in coincidence summing corrections

- Nuclide-related peak efficiency, $\varepsilon_n$
  - useful for measurements of a same nuclide
  - no correction for coincidence summing
Considerations on the efficiency

- The efficiency is related to specific source-detector geometry and peak analysis procedure.
- Detector geometry not reproducible, then efficiency calibration is required for each individual detector (contrary to NaI detectors).
- The efficiency is given by:
  \[ \varepsilon = f_g f_s f_d \]

  but the three factors
  - \( f_g = \Omega \) = effective subtended solid angle
  - \( f_s = \varepsilon_s \) = self-absorption factor
  - \( f_d = \varepsilon_i \) = intrinsic efficiency

  are not totally independent each other and \( \Omega = \Omega(\varepsilon) \).
- Nevertheless this factorisation is very useful for calculation of correction factors (see later).
Instrument calibration

\[ R = \int_g \int_s \int_d I \, A = \varepsilon \, IA \]

- The efficiency can be determined by:
  - Calculation
    \[ \varepsilon = \int_g \int_s \int_d \]
  - Measurement (by a calibrated source)
    \[ \varepsilon = \frac{R_s}{IA_s} \]
- In both cases, \( \varepsilon = \varepsilon(E) \) is usually needed.
EFFICIENCY CALCULATION: difficult and inaccurate
Efficiency calculation

- The efficiency (fep or total) can be calculated by:
  - Analytical expressions
  - Monte Carlo codes

- Achievable uncertainties: 5-10 %

- Results are usually inaccurate due to:
  - Lack of physical meaning for the instrument response (peak shape and full-energy-peak area definition)
  - Detector geometry not sufficiently known (crystal shape and size, dead layers, etc..)
  - Statistics (MC calculations)
  - Cross section and other physical parameters (±2%)

- Nevertheless, calculations are more and more used to get relative efficiencies, especially by MC methods (see later)
Geometry effect (1/4):
the geometry factor for point sources

\[ f_g = \frac{\Omega}{4\pi} = \frac{2\pi (1 - \frac{d}{\sqrt{d^2 + R^2}})}{4d^2} \approx \frac{R^2}{4d^2} \quad (d \gg R) \]

\[ f_g = \frac{\text{number of particles entering the detector}}{\text{number of particles emitted by the source}} \]
Geometry effect (2/4): the geometry factor for plane sources

\[ f_g = \text{number of particles entering the detector} / \text{number of particles emitted by the source} \]

\[ A_s \ldots A_r \ldots dA_s \ldots dA_r : \text{total-and-elemental-areas} \]
\[ S \ldots dS : \text{total-and-elemental-emission-rates} \]

\[ \cos \theta = \frac{\vec{n} \cdot \vec{r}}{r} \]

\[ d^2 S = \frac{S}{A_s} dA_s A_r \cos \theta \frac{1}{4\pi r^2} \]

\[ f_g = \frac{1}{S} \int \int d^2 S = \frac{1}{S} \int \int \frac{S}{A_s} dA_s A_r \cos \theta \frac{1}{4\pi r^2} = \int \int \frac{1}{4\pi r^2 A_s} \frac{\vec{n} \cdot \vec{r}}{r} dA_r dA_s \]

\[ f_g = \int \int \frac{1}{4\pi r^2 A_s} \frac{\vec{n} \cdot \vec{r}}{r} dA_r dA_s \]
Geometry effect (3/4): the geometry factor for volume sources

\[ f_g = \frac{\text{number of particles entering the detector}}{\text{number of particles emitted by the source}} \]

\[ f_g = \frac{1}{\frac{1}{S_{VA}} \int \int \frac{1}{4\pi V r^2} \frac{n \cdot r}{r} dAdV} \]
Geometry effect (4/4): determination of the geometry factor

- The geometry factor can be obtained by analytical formulas in very few cases.
- Suitable approximations can be obtained by:
  - Series expansions
  - Numerical integration
  - Monte Carlo calculations
Source effects (1/3)

Main source effects are:
- Self-absorption: $f_a$
- Scattering: $f_b$

$$f_s = f_a f_b$$
Source effect (2/3): self-absorption

- $f_a = \text{self-absorption factor} = \frac{\text{number of particles emitted from the source}}{\text{number of particles produced in the source}}$
- Very important for charged particles

For $\gamma$ and $\beta$ radiations:

$$f_a = \frac{1}{S} \int_0^t S \cdot e^{-\mu(t-x)} \, dx$$

$$f_a = \frac{1}{\mu t} \left[ 1 - e^{-\mu t} \right]$$

$0 \leq f_a \leq 1$
**Source effect (3/3): Scattering**

- $f_b = \text{backscattering factor} = \frac{\text{number of particles emitted toward the detector with the support}}{\text{number of particles emitted toward the detector without the support}}$
- Very important for charged particles
Detector effects (1/2)

Main detector effects are:

- No interaction with the detector (2)
- Absorption (3) or scattering (4) in the detector window
Detector effects (2/2): intrinsic detector efficiency

- \( f_\varepsilon = \textit{intrinsic efficiency} = \frac{\text{number of pulses produced in the detector and counted}}{\text{number of particles impinging on the detector}} \)

- The detector efficiency depends upon:
  - Density and size of detector material
  - Type and energy of radiation
  - Electronics
EFFICIENCY MEASUREMENT:
easier and more accurate than calculation
## Uncertainty of the efficiency calibration

<table>
<thead>
<tr>
<th>Unc. (%)</th>
<th>Procedure</th>
<th>Needs</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt;5</td>
<td>Quick calibration with 1 or 2 multi-gamma emitters and standard interpolation procedures</td>
<td>Low experience and standard software packages</td>
</tr>
<tr>
<td>1-5</td>
<td>More sophisticated procedures with 9-10 single-gamma emitters and several corrections</td>
<td>Intermediate experience and good software packages</td>
</tr>
<tr>
<td>0.5-1</td>
<td>Application of accurate corrections</td>
<td>Considerable experience, research work in progress</td>
</tr>
</tbody>
</table>
**Multigamma/Single-gamma emitters for efficiency calibration**

### Advantages/Disadvantages

<table>
<thead>
<tr>
<th></th>
<th>Multigamma</th>
<th>Single gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{1/2}$</td>
<td>![Green]</td>
<td>![Red]</td>
</tr>
<tr>
<td>Compton background</td>
<td>![Red]</td>
<td>![Green]</td>
</tr>
<tr>
<td>Deconvolution</td>
<td>![Red]</td>
<td>![Green]</td>
</tr>
<tr>
<td>Coincidence summing</td>
<td>![Red]</td>
<td>![Green]</td>
</tr>
<tr>
<td>Covering of broad energy range</td>
<td>![Green]</td>
<td>![Red]</td>
</tr>
</tbody>
</table>
### Frequently used calibration nuclides for 60-3000 keV energy range

<table>
<thead>
<tr>
<th>Multigamma</th>
<th>Energy (keV)</th>
<th>Single gamma</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba-133</td>
<td>53-384</td>
<td>Am241</td>
<td>59</td>
</tr>
<tr>
<td>Eu-152</td>
<td>122-1408</td>
<td>Cd-109</td>
<td>88</td>
</tr>
<tr>
<td>Ra-226*</td>
<td>186-2448</td>
<td>Co-57</td>
<td>122</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>447-1562</td>
<td>Ce-139</td>
<td>166</td>
</tr>
<tr>
<td>Co-56</td>
<td>847-3451</td>
<td>Hg-203</td>
<td>279</td>
</tr>
</tbody>
</table>

* Daughters at secular equilibrium!

<table>
<thead>
<tr>
<th>Multigamma</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y-88</td>
<td>898, 1836</td>
</tr>
<tr>
<td>Co-60</td>
<td>1173, 1332</td>
</tr>
</tbody>
</table>

### Multigamma Energy

- Y-88: 898, 1836 keV
- Co-60: 1173, 1332 keV
Principal fitting functions in the 60-3000 keV energy range

- **Semiempirical**
  - \( \varepsilon(E) = a_1 \left[ \tau(E) + \sigma(E) a_2 e^{-a_3 E} \right] \)
  - \( \tau(E) \), photoelectric cross section
  - \( \sigma(E) \), Compton cross section

- **Linear relationship**
  - \( \log \varepsilon(E) = a_0 - a_1 \log(E / E_0) \)
  - \( E_0 = 1 \text{ keV} \)
  - Two joined functions: 60 < \( E < 200 \) keV, \( E > 200 \) keV

- **Polynomial functions**
  - \( \log \varepsilon(E) = \sum_{j=0}^{n} a_j \left[ \log(E / E_0) \right]^j \)
  - \( E_0 = 1 \text{ keV} \)
  - Two joined functions: 60 < \( E < 200 \) keV, \( E > 200 \) keV
  - \( n < 3 \)
Extension of the efficiency calibration range: The efficiency ratio method

- The efficiency at energy $E_i$ is obtained from the efficiency measured at energy $E_0$ and the efficiency ratio $k_i$, directly measured with an uncalibrated source (same geometry !)

- Example:
  
  - $E_0 = 122.1$ keV (Co-57)
  - $E_i = 121.8$ keV (Eu-152)
  - $E_i$ = other Eu-152 photon energies

\[
\varepsilon(E_i) = \varepsilon(E_0) k_i
\]

\[
k_i = \frac{\varepsilon(E_i)}{\varepsilon(E_0)}
\]

\[
\varepsilon(E_0) \approx \varepsilon(E_i) = \varepsilon(E_0) \frac{\varepsilon(E_i)}{\varepsilon(E_0)} \approx \varepsilon(E_0) \frac{\varepsilon(E_i)}{\varepsilon(E_1)}
\]
Total efficiency calibration

- Important in coincidence-summing corrections
- Single gamma emitters required
- Radionuclide used: Am-241, Cd-109, Ce-139, Cr-51, Sr-85, Cs-137, Mn-54
- Total count rate obtained by extrapolation to zero energy
- X-ray peaks subtracted (only full-energy-peaks !)
- For $E<60$ keV, $\varepsilon_t = \varepsilon$
Typical efficiency curves of HPGe detectors
Geometry of standard sources for efficiency calibration

- Marinelli beakers
- Liquid solutions
- Point source
- Paper filters
EFFICIENCY TRANSFER
**Relative measurement under reproducible conditions \((\varepsilon = \text{cost.})\)**

\[
R = \varepsilon A
\]

\[
\begin{align*}
R_s &= \varepsilon A_s \\
R_x &= \varepsilon A_x \\
A_x &= \frac{R_x}{R_s} A_s
\end{align*}
\]

\[
\varepsilon = \frac{R_s}{A_s} = \frac{R_x}{A_x}
\]

\[
u(A_x)^2 = u(R_x)^2 + u(R_s)^2 + u(A_s)^2
\]

**LEGEND**

<table>
<thead>
<tr>
<th></th>
<th>Standard source</th>
<th>Problem source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Instrument reading</td>
<td>(R_s)</td>
<td>(R_x)</td>
</tr>
<tr>
<td>Activity</td>
<td>(A_s)</td>
<td>(A_x)</td>
</tr>
<tr>
<td>Counting efficiency (cost.)</td>
<td>(\varepsilon)</td>
<td>(\varepsilon)</td>
</tr>
</tbody>
</table>

**RELATIVE MEASUREMENT:** measurement performed by a comparison between the **value** \((A_x)\) of a quantity to be measured and a known **value** \((A_s)\) (given by a standard) of the same physical quantity

[Ref. BIPM/ISO International Vocabulary of basic and general terms in metrology]

HP: Decadimento ed altri effetti dipendenti dal rateo di conteggio trascurabili!
Relative measurement under NON reproducible conditions 
(ε≠cost.)
“efficiency transfer“ methods

\[ R = \varepsilon A \]

\[
\begin{align*}
R_s &= \varepsilon_s A_s \\
R_x &= \varepsilon_x A_x \\
A_x &= \frac{R_x}{R_s} \varepsilon_x A_s \\
k &= \frac{\varepsilon_s}{\varepsilon_x} \\
\varepsilon_x &= \varepsilon_s \frac{1}{k}
\end{align*}
\]

\[ u(A_x)^2 = u(R_x)^2 + u(R_s)^2 + u(A_s)^2 + u(k)^2 \]

LEGEND

<table>
<thead>
<tr>
<th>Source</th>
<th>Standard Source</th>
<th>Problem Source</th>
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<tbody>
<tr>
<td>Instrument reading</td>
<td>( R_s )</td>
<td>( R_x )</td>
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<tr>
<td>Activity</td>
<td>( A_s )</td>
<td>( A_x )</td>
</tr>
<tr>
<td>Counting efficiency (cost.)</td>
<td>( \varepsilon )</td>
<td>( \varepsilon )</td>
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</table>

The constant \( k \) can be determined by::
- Measurements
- Analytic expressions
- Monte Carlo simulations
- Semi empirical models

\[ \varepsilon = f_g f_s f_d I \]

\begin{itemize}
\item \( f_g \): geometric efficiency (subtended solid angle)
\item \( f_s \): source efficiency
\item \( f_d \): detection efficiency
\item I: emission probability
\end{itemize}
UNCERTAINTY evaluation by “efficiency transfer"

\[
\begin{align*}
R_s &= \varepsilon_s A_s \\
R_x &= \varepsilon_x A_x \\
A_x &= \frac{R_x}{R_s} \frac{\varepsilon_s}{\varepsilon_x} A_s
\end{align*}
\]

\[
k = \frac{\varepsilon_s}{\varepsilon_x} \Rightarrow \varepsilon_x = \varepsilon_s \frac{1}{k}
\]

\[
\begin{align*}
\{HP : \varepsilon = \text{cost}\} &\Rightarrow \begin{cases} 
k = 1 \\
u(k) \neq 0 \end{cases} \Rightarrow A_x = \frac{R_x}{R_s} A_s \\
\{HP : \varepsilon \neq \text{cost}\} &\Rightarrow \begin{cases} 
k = \frac{\varepsilon_s}{\varepsilon_x} \neq 1 \\
u(k) \neq 0 \end{cases} \Rightarrow A_x = \frac{R_x}{R_s} kA_s
\end{align*}
\]

\[
u'(A_x)^2 = \nu'(R_x)^2 + \nu'(R_s)^2 + \nu'(A_s)^2 + \nu'(k)^2
\]

**LEGEND**

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<td>Counting efficiency (cost.)</td>
<td>(\varepsilon)</td>
<td>(\varepsilon)</td>
</tr>
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</table>
From calibration to measurement

Sample and measurement instrument

Measurement geometry

Calibration geometry

Sample treatment
- Grinding
- Homogenization
- Weighing
- Preparation of measurement geometry

Efficiency transfer
- Detector characterization
- Geometry characterization
- Calculation or measurement of efficiency transfer factor, k

\[ \varepsilon_x = \varepsilon_s \frac{1}{k} \]

\[ k = \frac{\varepsilon_s}{\varepsilon_x} \]

\[ \varepsilon_s = \frac{R_s}{A_s} \]
Example: different nuclides, detection efficiency

Under non reproducible conditions (different nuclide, same photon energy)

<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td>Instrument reading</td>
<td>( R_s )</td>
<td>( R_x )</td>
</tr>
<tr>
<td>Photon emission probability</td>
<td>( I_s )</td>
<td>( I_x )</td>
</tr>
<tr>
<td>Activity</td>
<td>( A_s )</td>
<td>( A_x )</td>
</tr>
<tr>
<td>Counting efficiency</td>
<td>( \varepsilon_s )</td>
<td>( \varepsilon_x )</td>
</tr>
<tr>
<td>Detection efficiency</td>
<td>( \varepsilon' )</td>
<td>( \varepsilon' )</td>
</tr>
</tbody>
</table>

\[
\varepsilon = I \varepsilon' \\
R = \varepsilon A = I \varepsilon' A
\]

\[
\begin{align*}
R_x &= \varepsilon_x A_x = I_x \varepsilon' A_x \\
R_S &= \varepsilon_s A_s = I_s \varepsilon' A_s
\end{align*}
\]

\[
k \equiv \frac{\varepsilon_s}{\varepsilon_x} = \frac{I_s \varepsilon'}{I_x \varepsilon'} = \frac{I_s}{I_x} \Rightarrow \left\{ A_x = \frac{R_x}{R_s} k A_s \right\}
\]
Example: extension of calibration interval (gamma spectr.), efficiency ratio method

- Efficiency at energy $E_i$ is derived from the efficiency measured at energy $E_0$ and from the ratio $k_i$, directly measured by a non calibrated source (same geometry!)

- Example:
  - $E_0 = 122.1$ keV (standard source Co-57)
  - $E_1 = 121.8$ keV (non standard source Eu-152)
  - $E_i$ = other Eu-152 photon emissions

\[
k_i = \frac{\varepsilon(E_i)}{\varepsilon(E_1)}
\]

Traceability

\[
\varepsilon(E_0) \approx \varepsilon(E_1)
\]

\[
\varepsilon(E_i) = \varepsilon(E_0) \frac{1}{k_i}
\]
Example: correction for geometry and filling volume

Calibration geometry

Reference geometry (rif)

Measurement geometry

Efficiency transfer 1 (sample holder)

Efficiency transfer 2 (sample volume)

Traceability

$F_{\text{camp}} = \frac{A_c}{R_c^{\text{camp}}}$

$C^{\text{rif}} = \frac{R_c^{\text{camp}}}{R_c^{\text{rif}}}$

$C^{\text{vol}} = \frac{R_c^{\text{rif}}}{R_c^{\text{vol}}}$

$F^{\text{rif}} = F_{\text{camp}} C^{\text{rif}}$

$F^{\text{vol}} = F^{\text{rif}} C^{\text{vol}}$

$F^{\text{vol}} = F_{\text{camp}} C^{\text{rif}} C^{\text{vol}}$
Efficiency transfer in γ-ray spec.: ETNA

ETNA (Efficiency Transfer for Nuclide Activity measurements) is a software developed by the Laboratoire National Henri Becquerel that allows calculation of:

- efficiency transfer factors
- coincidence summing corrections

Références

M.C. Lépy, M.M. Bé, F. Piton, "ETNA (Efficiency Transfer for Nuclide Activity measurements) : Logiciel pour le calcul du transfert de rendement et des corrections de coïncidences en spectrométrie gamma", Note technique LNHB/01/09/F (2001)

Detector geometry

Detector geometry is required for:

- "efficiency transfer" applications (geometry and self absorption corrections);
- total efficiency calculation (coincidence summing corrections);

This information can be:

- obtained by the detector manufacturer;
- directly measured by x-ray radiography.

NUCLEAR DATA
Decay Data Evaluation Project (DDEP) recommended data base

- Started in 1990
- Expert evaluators
- Evaluation procedure preliminary agreed
- Special attention to measurement uncertainties
- DDEP recommended (2004) by the Bureau International des Poids et Mesures (BIPM)
- DDEP on the WEB
  (http://www.nucleide.org/DDEP_WG/DDEPdata.htm)
- DDEP published also as BIPM Monographie N. 5
ICRM RECOMMENDATION
ICRM General Meeting, 9th September 2005

Given that BIPM has recommended the use of DDEP evaluated decay data in all NMI’s, the Nuclear Data Working Group recommends the adoption of DDEP data in all members institutes of the ICRM, to assure roundness and consistency in their future nuclear data studies.

International Committee for Radionuclide Metrology (ICRM)
(http://www.physics.nist.gov/icrm)
Other Nuclear data www sites

- http://www-nds.iaea.or.at/nsr/index.jsp
- http://www.nucleide.org
- http://www.radiochemistry.org/periodictable/frames/gamma_spectra.htm
- http://ie.lbl.gov/
- http://xdb.lbl.gov/
Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

**UNCERTAINTY IN GAMMA SPECTROMETRY**

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<table>
<thead>
<tr>
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<tbody>
<tr>
<td>1.</td>
<td>Metrology, measurement fundamentals</td>
</tr>
<tr>
<td>2.</td>
<td>Uncertainty evaluation theory</td>
</tr>
<tr>
<td>3.</td>
<td>Uncertainty components in gamma spectrometry</td>
</tr>
<tr>
<td>4.</td>
<td>Methods for uncertainty determination</td>
</tr>
<tr>
<td>5.</td>
<td>Application, exercises, Examples, Discussion</td>
</tr>
<tr>
<td>6.</td>
<td>Characteristic limits</td>
</tr>
<tr>
<td>7.</td>
<td>Quality control</td>
</tr>
<tr>
<td>8.</td>
<td>Discussion with participants</td>
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</tbody>
</table>
UNCERTAINTY EVALUATION
FOUNDAMENTALS
Main points

- A measurement result has little or no meaning and value unless it has an uncertainty

- “Calibration and Testing laboratories ... shall have and apply a procedure to estimate the uncertainty of measurement ...” [ISO 17025 Par. 5.4.6]
GUM (ISO 1995)

- Guide to the expression of uncertainty in measurement (GUM)

- When reporting the result of a measurement of a physical quantity, some quantitative indication of the result has to be given to assess its reliability and to allow comparisons to be made. The Guide to the expression of uncertainty in measurement establishes general rules for evaluating and expressing uncertainty in measurement that can be followed at many levels of accuracy and in many fields.

- Year of publication: 1995
Error and Uncertainty: Definitions
[BIPM/ISO Guide to the expression of uncertainty in measurements]

**ERROR** (of measurement): result of a measurement minus a true value of the measurand

**RANDOM ERROR**: result of a measurement minus the mean that would result from an infinite number of measurement of the same measurand carried out under repeatability conditions

**SYSTEMATIC ERROR**: mean that would result from an infinite number of measurements of the same measurand carried out under repeatability conditions minus a true value of the measurand

**CORRECTION**: value added algebraically to the uncorrected result of a measurement to compensate for systematic error

**CORRECTION FACTOR**: numerical factor by which the uncorrected result of a measurement is multiplied to compensate for systematic error

**UNCERTAINTY OF MEASUREMENT**: parameter, associated with the result of a measurement, that characterises the dispersion of the values that could reasonably be attributed to the measurand
Graphical illustration of values, error, and uncertainty

[ISO (1993), Guide to the expression of uncertainty]
Recommendation INC-1 (1980) (1/2)

1. The uncertainty in the result of a measurement generally consists of several components which may be grouped into categories according to the way in which their numerical value is estimated:

   A) Those which are evaluated by statistical methods
   B) Those which are evaluated by other means.

   There is not always a simple correspondence between the classification into categories A or B and the previously used classification into “random” and “systematic” uncertainties. The term “systematic uncertainty” can be misleading and should be avoided.

   Any detailed report of the uncertainty should consist of a complete list of the components, specifying for each the method used to obtain its numerical value.

2. The components in category A are characterized by the estimated variances $s^2_i$ (or the estimated “standard deviations” $s_i$) and the number of degrees of freedom $v_i$. Where appropriate, the covariances should be given.
3. The components in category B should be characterized by quantities $u_j^2$, which may be considered as approximations to the corresponding variances, the estimate of which is assumed. The quantities $u_j^2$ may be treated like variances and the quantities $u_j$ like standard deviations. Where appropriate, the covariances should be treated in a similar way.

4. The combined uncertainty should be characterized by the numerical value obtained applying the usual method for the combination of variances. The combined uncertainty and its components should be expressed in the form of “standard deviations”.

5. If, for particular applications, it is necessary to multiply the combined uncertainty by a factor to obtain an overall uncertainty, the multiplying factor used must always be stated.
Graphical illustration of evaluating the standard uncertainty of an input quantity from repeated observations

[ISO (1993), Guide to the expression of uncertainty]
Graphical illustration of evaluating the standard uncertainty of an input quantity from an a priori distribution

[ISO (1993), Guide to the expression of uncertainty]
Final recommendations

7.2.3 When reporting the result of a measurement, and when the measure of uncertainty is the expanded uncertainty \( U = k u_e(y) \), one should

a) give a full description of how the measurand \( Y \) is defined;

b) state the result of the measurement as \( Y = y \pm U \) and give the units of \( y \) and \( U \);

c) include the relative expanded uncertainty \( U/|y| \), \(|y| \neq 0\), when appropriate;

d) give the value of \( k \) used to obtain \( U \) [or, for the convenience of the user of the result, give both \( k \) and \( u_e(y) \)];

e) give the approximate level of confidence associated with the interval \( y \pm U \) and state how it was determined;

f) give the information outlined in 7.2.7 or refer to a published document that contains it.

7.2.7 In the detailed report that describes how the result of a measurement and its uncertainty were obtained, one should follow the recommendations of 7.1.4 and thus

a) give the value of each input estimate \( x_i \) and its standard uncertainty \( u(x_i) \) together with a description of how they were obtained;

b) give the estimated covariances or estimated correlation coefficients (preferably both) associated with all input estimates that are correlated, and the methods used to obtain them;

c) give the degrees of freedom for the standard uncertainty of each input estimate and how it was obtained;

d) give the functional relationship \( Y = f(X_1, X_2, \ldots, X_n) \) and, when they are deemed useful, the partial derivatives or sensitivity coefficients \( \partial f/\partial x_i \).

However, any such coefficients determined experimentally should be given.

NOTE - Since the functional relationship \( f \) may be extremely complex or may not exist explicitly but only as a computer program, it may not always be possible to give \( f \) and its derivatives. The function \( f \) may then be described in general terms or the program used may be cited by an appropriate reference. In such cases, it is important that it be clear how the estimate \( y \) of the measurand \( Y \) and its combined standard uncertainty \( u_e(y) \) were obtained.

[ISO (1993), Guide to the expression of uncertainty]
A useful Guideline

[http://physics.nist.gov]


STATISTICAL QUANTITIES
Definition of probability

- Assume that one repeats an experiment many times and observes whether or not a certain event $x$ is the outcome.

- If the experiment was performed $N$ times, and $n$ results were of type $x$, the probability $P(x)$ that any single event will be of type $x$ is equal to:

$$P(x) = \lim_{N \to \infty} \frac{n}{N}$$

- The ratio $n/N$ is called the relative frequency of occurrence of $x$ in the first $N$ trials.

- Since both $n$ and $N$ are positive numbers:

$$0 \leq \frac{n}{N} \leq 1$$

$$0 \leq P(x) \leq 1$$

- The probability is measured on a scale from 0 to 1

- If the event $x$ occurs every time the experiment is performed, then:

$$n = N$$

$$P(x) = 1$$

- If the event $x$ never occurs, then:

$$n = 0$$

$$P(x) = 0$$
Probability distribution (a)

- When an experiment is repeated many times under identical conditions, the results of the measurement will not necessarily be identical.
- A quantity $x$ that can be determined quantitatively and that in successive but similar experiments can assume different values is called a random variable.
- There are two types of random variables, discrete and continuous.
- A discrete random variable takes one of a set of discrete values.
- A continuous random variable can take any value within a certain interval.
- For every random variable $x$, one may define a function $f(x)$ as follows:
Probability distribution (b)

- **Discrete random variables:**
  
  - \( f(x_i) = \text{probability that the value of the random variables is } x_i \text{ with } i = 1, 2, \ldots, N \text{ and } N = \text{number of possible values of } x. \)
  
  - Since \( x \) takes only one value at a time, the events presented by \( f(x_i) \) are mutually exclusive:

\[
\sum_{1}^{n} f(x_i) = 1
\]
Probability distribution (c)

- **Continuous random variables:**
  - Assume that a random variable may take any value between \( a \) and \( b \):
    \[
    a \leq x \leq b
    \]
  - Then \( f(x)dx \)=probability that the values of \( x \) lies between \( x \) and \( x+dx \) so:
    \[
    \int_{a}^{b} f(x)dx = 1
    \]
  - Where \( f(x) \)=probability density function (pdf).

- The probability \( p(x_a, x_b) \) that a value of \( x \) falls into the interval between \( x_a \) and \( x_b \) is
  \[
  P(x_a, x_b) = \int_{x_a}^{x_b} f(x)dx
  \]
The Mean

- The mean, also known as the “average” or the “expectation value” of $x$, is defined by the equation:

  \[
  \bar{x} = m = \frac{1}{N} \sum_{i=1}^{N} x_i = \frac{1}{N} \sum_{i=1}^{N} f(x_i)
  \]

  \[
  \bar{g}(x) = \frac{1}{N} \sum_{i=1}^{N} g(x_i) = \frac{1}{N} \sum_{i=1}^{N} g(x_i) f(x_i)
  \]

- The mean of any function $g(x)$ is:

  \[
  \bar{g}(x) = \frac{1}{N} \sum_{i=1}^{N} g(x_i) f(x_i)
  \]

- If $a$ is a constant:

  \[
  a\bar{x} = \bar{a} + \bar{x} = a \bar{m}
  \]

  \[
  (a + x) = \bar{a} + \bar{x} = a + m
  \]

  \[
  f_1(x) + f_2(x) + \ldots + f_n(x) = \bar{f}_1(x) + \bar{f}_2(x) + \ldots + \bar{f}_n(x)
  \]
The Variance

- For practical purposes it is sufficient to know the mean together with a measure indicating how the probability density is distributed around the mean. There are several such measures called “dispersion indexes”.

- The dispersion index most commonly used is the variance $V(x)$ and its square root, called standard deviation $s$.

- The variance of a probability density function is defined as

  \[ V(x) = \sigma^2 = \int_{-\infty}^{+\infty} (x - m)^2 f(x) \, dx \]

  \[ V(x) = \sigma^2 = \sum_{i=1}^{N} (x_i - m)^2 f(x) \]

where $a$ and $b$ are constants.

- $V(a + bx) = b^2 V(x)$
Covariance and correlation coefficient

- Consider the random variables $X_1, X_2, \ldots, X_n$ with means $m_1, m_2, \ldots, m_n$ and variances $\sigma_1^2, \sigma_2^2, \ldots, \sigma_n^2$.

- The average and variance of the linear function $Q = a_1 x_1 + a_2 x_2 + \ldots + a_n x_n$ with $a_i$ constants are

  $\bar{Q} = \sum_{i=1}^{N} a_i m_i$

  $V(Q) = \sigma^2 = (Q - \bar{Q})^2 = \left[ \sum_{i=1}^{N} a_i (x_i - m_i) \right]^2 = \sum_{i=1}^{N} a_i^2 \sigma_i^2 + 2 \sum_{i,j=1}^{N} a_i \sigma_j (x_i - m_i)(x_j - m_j)$

- The “covariance” between $X_i$ and $X_j$ is defined as

  $\text{cov}(x_i, x_j) = (x_i - m_i)(x_j - m_j)$

- This equation for the covariance suffers from the serious drawback that its value change with the units used for the measurement of $X_i$ and $X_j$. To eliminate this effect, the covariance is divided by the product of the standard deviations $\sigma_i$ and $\sigma_j$ and the resulting ratio is called “covariance coefficient” $\rho(x_i, x_j)$. Thus:

  $\rho_{ij} = \rho(x_i, x_j) = \frac{\text{cov}(x_i, x_j)}{\sigma_i \sigma_j}$

- Random variable for which $\rho_{ij} = 0$ are said to be uncorrelated.
Sample estimates (a)

- If a sample of the n values $x_1, x_2, \ldots, x_n$ has been obtained from repeated measurements under identical measuring conditions (i.e., all the $x_i$ have the same uncertainty), the arithmetic mean

$$\bar{X} = \frac{1}{N} \sum_{i=1}^{N} x_i$$

is the best estimate of the expectation value $m$ of the random variable $X$.

- With increasing sample size $n$, the arithmetic mean approaches $m$.

- If the random variable $X$ is associated to a pdf $f(x)$ with variance $\sigma^2$, the quantity

$$S^2(x) = \frac{1}{N-1} \sum_{i=1}^{N} (x_i - \bar{X})^2$$

is the best estimate of $\sigma^2$. 
Sample estimates (b)

- The covariance estimated by two sets of simultaneous measurements is:

\[ \text{cov}(x_i, x_j) = \frac{1}{n-1} \sum_{i=1}^{N} (x_{ik} - \bar{x}_i)(x_{jk} - \bar{x}_j) \]

- If \( x_i \) and \( x_j \) are function of a new variable \( z \), then:

\[ \text{cov}(x_i, x_j) = \frac{\partial x_i}{\partial z} \frac{\partial x_j}{\partial z} s(z)^2 \]

- The ratio

\[ \rho(x_i, x_j) = \frac{\text{cov}(x_i, x_j)^2}{s(x_i)s(x_j)} \]

is the correlation coefficient equal to 0 if the \( x_i \) and \( x_j \) are uncorrelated and equal to 1 if they are fully uncorrelated.
STATISTICS APPLIED TO COUNTING
Counting Uncertainty

- Random error gives rise to imprecision, or irreproducibility, in any scientific experiment, but in particle counting (activity measurements, neutron flux measurements, dosimetry, ...) its influence is compounded by the fact that particle emission and detection themselves are random, i.e. stochastic, phenomenon.

- Even with the most precise instrument and observer, any two consecutive readings of particle count will probably not be the same.

- The potential for systematic error arises both from uncertainties associated with the measurements of ancillary data, such as efficiencies and dead times, needed to calculate the count rate from a simple counts of events in a given time, and from instrumental and operator bias.
Standard uncertainty of the counting rate

- The process of radioactive decay is a random sequence in time.
- From the law of radioactive decay the decay rate $\rho = dN/dt$ can be treated as a constant.
- The decay rate $\rho$ of a sample bears a known relationship, through the counting efficiency, to the “true” counting rate, $\rho'$.
- However, counting for a finite period of time $t$ can only yield an estimate $r = n/t$ of the “true” counting rate, $r'$.
- The measured $r$ is subject to statistical fluctuations. A measure for the scatter of the measured counting rate $r$ around the “true” counting rate $r'$ can be derived from the Poisson assumption. Such a measure is provided by the standard deviation.
- The standard deviation of $r$ is a function of $t$ and can be expressed as:

$$\sigma_r = \sqrt{\frac{\rho'}{t}}$$

- Because $\rho'$ is unknown, only an estimate, $s_r$, of the standard deviation, $\sigma_r$, can be computed from the measured $r$, namely:

$$s_r = \sqrt{\frac{r}{t}} = \frac{1}{t} \sqrt{n}$$

- An estimate of the standard deviation of the number of counts is:

$$s_n = \sqrt{n}$$
Performance of counting systems

- The following limits apply to representative multiplets of the standard deviation in the theoretical distribution:

<table>
<thead>
<tr>
<th>deviation</th>
<th>±0.675σ</th>
<th>±1σ</th>
<th>±2σ</th>
<th>±3σ</th>
</tr>
</thead>
<tbody>
<tr>
<td>probability that observation lies within this deviation</td>
<td>0.5</td>
<td>0.68</td>
<td>0.95</td>
<td>0.997</td>
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</table>

- The above limits allow an appraisal of the performance of a counting system to be quickly made, i.e., if a counting rate deviates more than what would be expected statistically from a previously determined rate.

- This is important when equipment is checked routinely with a performance standard (check source).

- The application of this general description can be formalised through the use of “control charts” that provide a running graphical record of the values obtained from counting a reference source or of their variability.
Counting the necessary to reach a desired degree of uncertainty

- In most instances the radiation counter exhibits a background rate that is not negligible and that has to be subtracted from the gross counting rate.
- An estimate of the statistical error due to the background must be included in the final uncertainty budget.
- On the basis that the source and background counts are additive, the standard deviation of the counting rate is:

\[
\sigma_r = \sqrt{\sigma_T^2 + \sigma_B^2} = \sqrt{\frac{\rho_T'}{t_T} + \frac{\rho_B'}{t_B}}
\]

where T refers to the counting of the sample plus background (total) and B to the counting of the background alone.

- The optimum subdivision of the available time between background and source counting has been derived by Loevinger and Berman (1951), and is given by:

\[
t_B = \frac{\rho_B'}{\sqrt{\rho_T'}}
\]

\[
t_T = \sqrt{\rho_T'}
\]

- To approximate the optimum division, estimates of \(\rho_B'\) and \(\rho_T'\) have to be obtained from a preliminary run.
UNCERTAINTY PROPAGATION
Propagating uncertainties (a)

- In most experiments the quantity of interest is not the one that can be measured.
- Instead, its value has to be derived from values of several other quantities.
- If a quantity $X$ is a function of $p$ quantities $z_1, z_2, \ldots, z_p$
  \[ X = F(z_1, z_2, \ldots, z_n) \]
  where each of the values $z_k$ of the quantities $Z_k$ has an associated uncertainty $S(z_k)$, the uncertainty of the value $x$ for the quantity $X$ is obtained by applying the “error propagation law” of Gauss which provides a variance for the value $x$ according to
  \[
  S^2(x) = \sum_{i=1}^{p} \left( \frac{\partial F}{\partial z_k} \right)_{z_k}^2 S^2(z_k)
  \]
- This equation is valid only if the quantities $Z_k$ are independent (uncorrelated), if the distribution of each $Z_k$ is a Gaussian distribution and if:
  \[ S(z_k) \ll z_k \]
- If these conditions are satisfied, the quantity $x$ tends towards a Gaussian distribution with a variance given by the $s^2(x)$. 
Propagation of uncertainties (b)

- More generally, if the individual quantities $Z_k$ are correlated the error propagation law is:

$$X = F(z_1, z_2, \ldots, z_n)$$

$$S^2(x) = \sum_{i=1}^{p} \left( \frac{\partial F}{\partial z_k} \right)_{z_k}^2 S^2(z_k) + 2 \sum_{k=1}^{p} \sum_{i<k}^{p} \left( \frac{\partial F}{\partial z_k} \right)_{z_k} \left( \frac{\partial F}{\partial z_j} \right)_{z_j} S(z_k, z_j)$$
Mathematical expressions

Error propagation law

\[ Y = f(x_1, x_2, \ldots, x_p) \]

\[ S_Y^2 = \sum_{i=1}^{p} \left( \frac{\partial f}{\partial x_i} \right)^2 S_{x_i}^2 + 2 \sum_{i<k} \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \text{cov}(x_i, x_j) \]

Sample estimates

\[ S_{x_i}^2 = \frac{1}{n-1} \sum_{k=1}^{n} (x_{ik} - \bar{x}_i)^2 \]

\[ \text{cov}(x_i, x_j) = \frac{1}{n-1} \sum_{k=1}^{n} (x_{ik} - \bar{x}_i)(x_{jk} - \bar{x}_j) \]

Useful expressions

\[ \begin{align*}
  &\{ x_i = x_i(z,\ldots) \\
  &\{ x_j = x_j(z,\ldots) \} \quad \Rightarrow \quad \text{cov}(x_i, x_j) = \frac{\partial x_i}{\partial z} \frac{\partial x_j}{\partial z} S_z^2 \\\n  &\{ Y = \frac{x_1}{x_2} \} \quad \Rightarrow \quad \left( \frac{S_Y}{Y} \right)^2 = \left( \frac{S_{x_1}}{X_1} \right)^2 + \left( \frac{S_{x_2}}{X_2} \right)^2 \\
  &\text{cov}(x_1, x_2) = 0 \quad \Rightarrow \quad Y = \frac{x_1}{x_2} \quad \Rightarrow \quad \left\{ \begin{array}{l}
  \frac{1}{Y} \frac{\partial Y}{\partial x_1} = x_1 \\
  \frac{1}{Y} \frac{\partial Y}{\partial x_2} = -\frac{1}{x_2} \\
  \frac{\partial f}{\partial x_i} \approx \frac{f(x_i + \Delta x_i) - f(x_i)}{\Delta x_i} \\
  \end{array} \right\} \\
\end{align*} \]
The following quantities are required for uncertainty evaluation:

\[ Y = f(x_1, x_2, \ldots, x_p) \]

\[ S_Y^2 = \sum_{i=1}^{p} \left( \frac{\partial f}{\partial x_i} \right)^2 S_{x_i}^2 + 2 \sum_{i<k} \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \text{cov}(x_i, x_j) \]

Obtained by measurement, calculation, estimation, importation, derivation, …

A MODEL IS NEEDED!
## Uncertainty in Gamma Spectrometry

1. Metrology, measurement fundamentals  
2. Uncertainty evaluation theory  
3. Uncertainty components in gamma spectrometry  
4. Methods for uncertainty determination  
5. Application, exercises, Examples, Discussion  
6. Characteristic limits  
7. Quality control  
8. Discussion with participants
Main sources of uncertainty in gamma-ray spectrometry

EXERCISE:
As groups, consider the following sources of uncertainty and:

a) Propose a method to estimate each uncertainty component
b) Identify type of evaluation (A or B)
c) Make an estimate of its typical value

1. Activity of standard source
2. Sampling
3. Source aliquot
4. Peak-area measurement
5. Background peak area variations
6. Variation in sample attenuation
7. Source-detector geometry
8. Dead time correction (LT/RT)
9. Pulse pile-up
10. Coincidence summing
11. Random summing
12. Radioactive decay
13. Photon emission probability
14. Full-energy peak efficiency
15. Multiplets deconvolution
Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

UNCERTAINTY IN GAMMA SPECTROMETRY

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CORRECTIONS AND ASSOCIATED UNCERTAINTIES IN GAMMA SPECTROMETRY
Efficiency transfer

\[ R = A \varepsilon \]

where:  
- \( R \): Net count rate  
- \( A \): Source Activity (\( I_\gamma = 1 \))  
- \( \varepsilon \): Counting efficiency

Under non-reproducible conditions (\( \varepsilon \) not constant):

\[
\begin{align*}
R_x &= A_x \varepsilon_x \\
R_S &= A_S \varepsilon_S \\
A_x &= \frac{R_x}{R_S} \frac{\varepsilon_S}{\varepsilon_x} \\
A_S &= \frac{R_x}{R_S} k A_S \\
\varepsilon_x &= \varepsilon_S \frac{\varepsilon_S}{\varepsilon_x} = \varepsilon_S k \\
k &= \frac{\varepsilon_S}{\varepsilon_x}
\end{align*}
\]

where:  
- \( x \): Problem source  
- \( S \): Standard source

The constant \( k \) can be experimentally or theoretically determined and used to derive the corrected efficiency.

Methods used:
- Analytical expressions
- Monte Carlo calculation
- Semiempirical models
COINCIDENCE-SUMMING
COINCIDENCE-SUMMING: occurs for radionuclides emitting two or more photons within the spectrometer resolving time

A: Single photons

B: Coincident photons
COINCIDENCE-SUMMING: spectrum deformation

a) Photoelectric-Compton: summing out

\[ n_1 = AI_{1\gamma 1} \varepsilon_1 - Ak_{12} \varepsilon_1 \varepsilon_{t2} \]

b) Photoelectric-photoelectric: summing in

\[ n_{12} = Ak'_{12} \varepsilon_1 \varepsilon_2 \]
COINCIDENCE-SUMMING:
loss of events from full-energy peak

A: Single photons
B: Coincident photons
Magnitude of coincidence-summing effect

a) p-type detectors

b) n-type detectors

HPGe

10%

50%

50%

100%

γ

X

10-100%
Coincidence-summing:  
a common source of systematic errors in  
gamma-spectrometry

Example:  
Italian gamma-spectrometry  
proficiency test (1998)
Example: coincidence summing correction in gamma spectrometry (Co-60)

\[ n_1 = A\varepsilon_1 (1 - \varepsilon_{t2}) \]

\[ n_2 = A\varepsilon_2 (1 - \varepsilon_{t1}) \]
COINCIDENCE-SUMMING CORRECTION

Example 2: Cs-134, 569 keV

\[ n_5 = AI_5 \varepsilon_5 \left( 1 - P_7 \varepsilon_{t7} - P_6 \varepsilon_{t6} \right) \]

\[ P_k = \frac{1}{1 + \alpha_t} \]
Exaple: coincidence summing correction in gamma spectrometry

Correction factor: $C_i$

\[ n_i = AI_{\gamma_i} \varepsilon_i - A \sum_j P_{tij} P_i P_j \varepsilon_j \varepsilon_i + A \sum_{k,m} P_{tkm} P_k P_m \varepsilon_k \varepsilon_m \]

\[ n_i = AI_{\gamma_i} \varepsilon_i C_i \]

[Semkov, 1990]
Methods for coincidence-summing corrections

- Analytical expression [Hoppes, 1983], [Blawn, 1993]
- Monte Carlo Methods [Sima, 1996]
- Simplified semi-empirical methods [De Felice, 2000]
UNCERTAINTY EVALUATION

The uncertainty of the total efficiency, $\varepsilon_{ti}$, is transferred to $C_i$ by the usual error propagation law.

Input uncertainties are "compressed" by partial derivatives and the output uncertainties of $C_i$ are notably reduced.

**Example:** Point source on detector window, 604.7 keV, $^{134}$Cs.

<table>
<thead>
<tr>
<th>Photon N.</th>
<th>Photon energy (keV)</th>
<th>$C_6$</th>
<th>Uncertainty component of $C_6$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>563.2</td>
<td>1.695</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>569.3</td>
<td>1.674</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>604.7</td>
<td>1.323</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>795.9</td>
<td>1.327</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>1365.2 (569+796)</td>
<td>0.696</td>
<td></td>
</tr>
</tbody>
</table>

![Graph](image)
CORRECTION PROCEDURE

\[ n_i = A I_{\gamma_i} \varepsilon_i C_i \]

\[ C_i = \left[ 1 + \frac{\sum_{k,m} P_{tkm} P_k P_m \varepsilon_k \varepsilon_m}{I_{\gamma_i} \varepsilon_i} \right] \left[ 1 - \frac{\sum_j P_{tjj} P_i P_j \varepsilon_{tj}}{I_{\gamma_i}} \right] \]

\[ R = \frac{R_\varepsilon}{R_\sigma} = KE_\gamma \]

\[ \varepsilon_{tj} = \frac{\varepsilon_j}{KR_\sigma E_\gamma} \]
DENSITY CORRECTIONS
Self-absorption effect

- Sample density from 0.4 to 3 kg/dm³
- Sample thickness from 5 to 30 mm
- Importance of chemical composition at low energy (E<100 keV)
Methods for self-absorption correction

- Empirical methods
- Analytical methods [es. Sima, 1992]
- Monte Carlo methods [Lepy, Sima]
Self absorption correction (1)
[Sima O., Health Physics 62, 445 (1992)]

\[ 
\varepsilon = \frac{\Delta S \varepsilon_p}{4\pi V} \int \frac{1}{Vr^2} e^{-\mu(r-r_m)} dV 
\]

\[ 
\varepsilon = \frac{\Delta S \varepsilon_p}{4\pi \eta V} \int \left[ 1 - e^{-\mu} \right] d\Omega 
\]

\[ 
\varepsilon = \frac{\Delta S \varepsilon_p}{4\pi \eta V} \frac{\Delta \Omega}{\Delta \Omega} \left[ 1 - e^{-\mu t} \right] 
\]

\[ 
\frac{\varepsilon_s}{\varepsilon_{ref}} = \frac{1 - e^{-\mu_s t}}{1 - e^{-\mu_{ref} t}} \frac{\mu_{ref}}{\mu_s} 
\]

\( N_0 \): photon emission rate per unit volume
\( \mu \): linear attenuation coefficient
\( \varepsilon_p \): intrinsic full-energy-peak efficiency
\( \varepsilon \): total efficiency
\( R \): full-energy-peak count rate
\( \Delta S \): detector area (supposed spherical)
\( t \): equivalent thickness
GEOMETRY CORRECTIONS
Efficiency variation with distance

- **Basic approach**
  - $\varepsilon(d_0)$ measured
  - $\varepsilon(d)$ obtained by calculation
  - HP: $\varepsilon$ geometry independent
  - Not very good approximation

- **Extension**
  - New concept
  - Fictitious point detector
  - Located at effective detector centre

\[ \Omega(d) = 2\pi \left(1 - \left(1 + \frac{R^2}{d^2}\right)^{-1/2}\right) \]

\[ \varepsilon_d = \varepsilon_{d_0} \frac{\Omega(d)}{\Omega(d_0)} \]

\[ n(d) \propto \frac{1}{(d + d_e)^2} \]

\[ \frac{1}{\sqrt{n(d)}} \propto (d + d_e) = 0 \]

\[ \Rightarrow d = -d_e \]
Area sources (1/2)

- [Alfassi, 1975]
  - a: area source
  - p: point source
  - $\varepsilon_p(r,d)$ experimentally determined
  - accuracy $\pm 2-5\%$

\[
\varepsilon_p(r,d) = \varepsilon_p(0,d)e^{-c(d)r^2}
\]

\[
\varepsilon_a(d) = \frac{2}{R_s^2} \int_0^{R_s} \varepsilon_p(r,d)rdr \Rightarrow
\]

\[
\varepsilon_a(d) = \varepsilon_p(0,d) \frac{1-e^{-cR_s^2}}{cR_s^2}
\]
Area sources (2/2)

- **Basic approach**
  - a: area source
  - p: point source
  - accuracy ±2% for R/d < 0.5

\[
\varepsilon_a(d) = \varepsilon_p(d) \frac{\Omega_a(d)}{\Omega_p(d)}
\]

\[
\Omega_a(d) = \frac{2}{R_s^2} \int_0^{R_s} \Omega_p(r, d) r dr \approx \frac{1}{2} \left[ 1 - \left( 1 + \frac{R^2}{d^2} \right)^{-1/2} - \frac{3}{8} R_s^2 \frac{R^2}{d^4} \left( 1 + \frac{R^2}{d^2} \right)^{-5/2} + \ldots \right]
\]
### Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

**UNCERTAINTY IN GAMMA SPECTROMETRY**

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Example N. 1
Variability of activity values of a set of sources
Excel file from ENEA-INMRI
Example N. 2
Uncertainty of the counting efficiency

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
<th>Standard Uncertainty</th>
<th>Relative Standard Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{1/2}$</td>
<td>Half-life</td>
<td>462.6</td>
<td>d</td>
<td>0.4</td>
<td>0.086</td>
</tr>
<tr>
<td>$I_\gamma$</td>
<td>Photon emission probability</td>
<td>3.65</td>
<td>%</td>
<td>0.06</td>
<td>1.64</td>
</tr>
<tr>
<td>$A_c$</td>
<td>Activity at reference date</td>
<td>20130</td>
<td>Bq</td>
<td>50</td>
<td>0.25</td>
</tr>
<tr>
<td>$T$</td>
<td>Counting Time</td>
<td>80000</td>
<td>s</td>
<td>160</td>
<td>0.2</td>
</tr>
<tr>
<td>$C_T$</td>
<td>Gross counts</td>
<td>163133</td>
<td>-</td>
<td>403</td>
<td>0.25</td>
</tr>
<tr>
<td>$C_B$</td>
<td>Background counts</td>
<td>14013</td>
<td>-</td>
<td>118</td>
<td>0.84</td>
</tr>
<tr>
<td>$C_N$</td>
<td>Net counts</td>
<td>149129</td>
<td>-</td>
<td>420</td>
<td>0.28</td>
</tr>
<tr>
<td>$\Delta T$</td>
<td>Decay time</td>
<td>1095.75</td>
<td>d</td>
<td>negligible</td>
<td>negligible</td>
</tr>
<tr>
<td>$DF$</td>
<td>Decay factor</td>
<td>0.19362</td>
<td>-</td>
<td>0.00027</td>
<td>0.14</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Counting efficiency</td>
<td>1.31</td>
<td>%</td>
<td>0.02</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Expressions

\[ u(C_T) = \sqrt{C_T} \]
\[ u(C_B) = \sqrt{C_B} \]
\[ C_N = C_T - C_B \]
\[ u(C_N) = \sqrt{u(C_T)^2 + u(C_B)^2} \]
\[ DF = \exp(-\ln(2)\frac{\Delta T}{T_{1/2}}) \]
\[ u(DF) = \frac{DF \ln(2)\Delta T}{T_{1/2}^2} \]
\[ \varepsilon = \frac{C_N}{T_{1/2} A_\gamma DF} \]
\[ u(\varepsilon) = \sqrt{\sum \left(\frac{u(x_i)}{x_i}\right)^2} \]

Counting efficiency: Uncertainty budget

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Expression</th>
<th>Value (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Counting statistics</td>
<td>$u(C_N)/C_N$</td>
<td>0.28</td>
</tr>
<tr>
<td>Timing</td>
<td>$u(T)/T$</td>
<td>0.2</td>
</tr>
<tr>
<td>Emission probability</td>
<td>$u(I_\gamma)/I_\gamma$</td>
<td>1.64</td>
</tr>
<tr>
<td>Standard source activity</td>
<td>$u(A_c)/A_c$</td>
<td>0.25</td>
</tr>
<tr>
<td>Decay</td>
<td>$u(DF)/DF$</td>
<td>0.14</td>
</tr>
<tr>
<td>Combined standard uncertainty</td>
<td>quadratic sum</td>
<td>1.7</td>
</tr>
</tbody>
</table>
Example N. 3
Uncertainty of the difference of net count rates

Background: source of correlation between net count rates

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
<th>Standard Uncertainty</th>
<th>Relative Standard Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>b</td>
<td>background count rate</td>
<td>200</td>
<td>1/s</td>
<td>14.0</td>
<td>7.0</td>
</tr>
<tr>
<td>n₁</td>
<td>gross count rate sample 1</td>
<td>230</td>
<td>1/s</td>
<td>13.8</td>
<td>6.0</td>
</tr>
<tr>
<td>n₂</td>
<td>gross count rate sample 2</td>
<td>300</td>
<td>1/s</td>
<td>15.0</td>
<td>5.0</td>
</tr>
<tr>
<td>A₁</td>
<td>net count rate sample 1</td>
<td>30</td>
<td>1/s</td>
<td>19.66</td>
<td>65.5</td>
</tr>
<tr>
<td>A₂</td>
<td>net count rate sample 2</td>
<td>100</td>
<td>1/s</td>
<td>20.52</td>
<td>20.5</td>
</tr>
<tr>
<td>D</td>
<td>Difference of net count rates</td>
<td>70</td>
<td>1/s</td>
<td>20.38</td>
<td>29.1</td>
</tr>
</tbody>
</table>

**Expressions**

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_1 = n_1 - b$</td>
<td>$u(A_1) = \sqrt{u(n_1)^2 + u(b)^2}$</td>
</tr>
<tr>
<td>$A_2 = n_2 - b$</td>
<td>$u(A_2) = \sqrt{u(n_2)^2 + u(b)^2}$</td>
</tr>
<tr>
<td>$D = A_2 - A_1 = n_2 - n_1$</td>
<td>$u(D) = \sqrt{u(n_1)^2 + u(n_2)^2}$</td>
</tr>
<tr>
<td>$\text{cov}(A_1, A_2)$</td>
<td>$-1)(-1)u(s_b)^2 = u(s_b)^2$</td>
</tr>
</tbody>
</table>

**D: Uncertainty budget**

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Expression</th>
<th>Value</th>
<th>Value (%)</th>
<th></th>
<th>Value</th>
<th>Value (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A₁ or n₁</td>
<td>$\frac{\partial D}{\partial A_1} u(A_1) = u(A_1)$</td>
<td>-19.66 s⁻¹</td>
<td>28.1</td>
<td>$\frac{\partial D}{\partial n_1} u(n_1) = u(n_1)$</td>
<td>-13.8 s⁻¹</td>
<td>19.7</td>
</tr>
<tr>
<td>A₂ or n₂</td>
<td>$\frac{\partial D}{\partial A_2} u(A_2) = u(A_2)$</td>
<td>+20.52 s⁻¹</td>
<td>29.3</td>
<td>$\frac{\partial D}{\partial n_2} u(n_2) = -u(n_2)$</td>
<td>-15.0 s⁻¹</td>
<td>21.4</td>
</tr>
<tr>
<td><strong>SUB TOTAL</strong></td>
<td></td>
<td>28.42 s⁻¹</td>
<td>40.6</td>
<td></td>
<td>20.38 s⁻¹</td>
<td>29.1</td>
</tr>
<tr>
<td>Correlations between A₁ and A₂</td>
<td>$2 \frac{\partial D}{\partial A_1} \frac{\partial D}{\partial A_2} \text{cov}(A_1, A_2) = -2u(s_b)^2$</td>
<td>-392 s⁻²</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Combined standard uncertainty</strong></td>
<td></td>
<td>20.38 s⁻¹</td>
<td>29.1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Example N. 4
Uncertainty of the ratio of activity values

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
<th>Standard Uncertainty</th>
<th>Relative Standard Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( n_1 )</td>
<td>net count rate lab 1</td>
<td>10.0</td>
<td>1/s</td>
<td>0.10</td>
<td>1</td>
</tr>
<tr>
<td>( n_2 )</td>
<td>net count rate lab 2</td>
<td>12.1</td>
<td>1/s</td>
<td>0.12</td>
<td>1</td>
</tr>
<tr>
<td>( \varepsilon_1 )</td>
<td>full-energy peak efficiency lab 1</td>
<td>1.01</td>
<td>%</td>
<td>0.01</td>
<td>1</td>
</tr>
<tr>
<td>( \varepsilon_2 )</td>
<td>full-energy peak efficiency lab 2</td>
<td>0.82</td>
<td>%</td>
<td>0.0082</td>
<td>1</td>
</tr>
<tr>
<td>( \Gamma )</td>
<td>photon emission probability</td>
<td>0.1</td>
<td>-</td>
<td>0.05</td>
<td>50</td>
</tr>
<tr>
<td>( A_1 )</td>
<td>sample activity lab 1</td>
<td>9.90</td>
<td>kBq</td>
<td>4.95</td>
<td>50.02</td>
</tr>
<tr>
<td>( A_2 )</td>
<td>sample activity lab 2</td>
<td>14.76</td>
<td>kBq</td>
<td>7.41</td>
<td>50.02</td>
</tr>
<tr>
<td>( R )</td>
<td>Ratio ( A_1 / A_2 )</td>
<td>0.6707</td>
<td>-</td>
<td>0.013</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Expressions

\[
A_i = \frac{n_i}{\varepsilon_i I_i} \\
A_i = \frac{n_i}{\varepsilon_i I_i} \\
HP: \text{cov}(\varepsilon_1, \varepsilon_2) = 0 \\
R = A_1 / A_2 \\
\text{cov}(A_1, A_2) = A_1 A_2 \text{cov}(I_1, I_2)
\]

R: Uncertainty budget

Source of uncertainty

<table>
<thead>
<tr>
<th>Expression</th>
<th>Value</th>
<th>Rel. Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{1}{R} \frac{\partial R}{\partial A_1} u(A_1) = u'(A_1) )</td>
<td>0.5002</td>
<td>0.01</td>
</tr>
<tr>
<td>( \frac{1}{R} \frac{\partial R}{\partial A_2} u(A_2) = u'(A_2) )</td>
<td>0.5002</td>
<td>0.01</td>
</tr>
<tr>
<td>( \sqrt{u'(A_1)^2 + u'(A_2)^2} )</td>
<td>0.70739</td>
<td>0.01</td>
</tr>
<tr>
<td>( 2 \frac{\partial R}{R^2 \partial A_1 \partial A_2} \text{cov}(A_1, A_2) = -2u'(I_1)^2 )</td>
<td>-0.5000</td>
<td>0.01</td>
</tr>
<tr>
<td>( \sqrt{u'(A_1)^2 + u'(A_2)^2 + \frac{2}{R^2} \frac{\partial R}{\partial A_1} \frac{\partial R}{\partial A_2} \text{cov}(A_1, A_2)} )</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>( \frac{\partial R}{R \partial \varepsilon_1} u(\varepsilon_1) = u'(\varepsilon_1) )</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>( \frac{\partial R}{R \partial \varepsilon_2} u(\varepsilon_2) = u'(\varepsilon_2) )</td>
<td>0.01</td>
<td></td>
</tr>
</tbody>
</table>

Combined standard uncertainty

The combined standard uncertainty of the ratio is calculated as:

\[
\sqrt{u'(A_1)^2 + u'(A_2)^2 + 2 \frac{\partial R}{R^2 \partial A_1 \partial A_2} \text{cov}(A_1, A_2)}
\]
Example N. 5
Uncertainty of the activity value (Co-60)

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
<th>Standard Uncertainty</th>
<th>Relative Standard Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>c1</td>
<td>net count rate (P1) calibr. source</td>
<td>10</td>
<td></td>
<td>0.1</td>
<td>1.0</td>
</tr>
<tr>
<td>c2</td>
<td>net count rate (P2) calibr. source</td>
<td>9.8</td>
<td></td>
<td>0.098</td>
<td>1.0</td>
</tr>
<tr>
<td>A&lt;sub&gt;c&lt;/sub&gt;</td>
<td>Activity calibration source</td>
<td>1000</td>
<td></td>
<td>30</td>
<td>3.0</td>
</tr>
<tr>
<td>n1</td>
<td>net count rate (P1) probl. source</td>
<td>11.1</td>
<td></td>
<td>0.111</td>
<td>1.0</td>
</tr>
<tr>
<td>n2</td>
<td>net count rate (P2) probl. source</td>
<td>10.3</td>
<td></td>
<td>0.103</td>
<td>1.0</td>
</tr>
<tr>
<td>e1</td>
<td>FEP efficiency (P1)</td>
<td>0.01</td>
<td></td>
<td>0.000316228</td>
<td>3.16</td>
</tr>
<tr>
<td>e2</td>
<td>FEP efficiency (P2)</td>
<td>0.0098</td>
<td></td>
<td>0.000309903</td>
<td>3.16</td>
</tr>
<tr>
<td>a&lt;sub&gt;1&lt;/sub&gt;</td>
<td>-</td>
<td>1.11</td>
<td></td>
<td>0.015697771</td>
<td>1.41</td>
</tr>
<tr>
<td>a&lt;sub&gt;2&lt;/sub&gt;</td>
<td>-</td>
<td>1.051</td>
<td></td>
<td>0.014863673</td>
<td>1.41</td>
</tr>
<tr>
<td>X&lt;sub&gt;1&lt;/sub&gt;</td>
<td>Activity problem source (P1)</td>
<td>1110</td>
<td></td>
<td>36.81433517</td>
<td>3.32</td>
</tr>
</tbody>
</table>

Expressions

\[
\varepsilon_i = \frac{c_i}{A_c} \quad u'(\varepsilon_i) = \sqrt{u'(c_i)^2 + u'(A_c)^2}
\]

\[
X_i = \frac{n_i}{\varepsilon_i} = \frac{n_i A_c}{c_i} \quad u'(X_i) = \sqrt{u'(n_i)^2 + u'(c_i)^2 + u'(A_c)^2}
\]

\[
a_i = \frac{n_i}{c_i} \quad u'(a_i) = \sqrt{u'(n_i)^2 + u'(c_i)^2}
\]

\[
A_x = \frac{X_1 + X_2}{2} = \frac{a_1 + a_2}{2} \frac{A_c}{\varepsilon_c}
\]

\[
cov(X_1, X_2) = 0.5 a_1 a_2 \frac{u(A_c)^2}{\varepsilon_c}
\]

A<sub>x</sub>: Uncertainty budget

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Expression</th>
<th>Value</th>
<th>Rel. Value</th>
<th>Expression</th>
<th>Value</th>
<th>Rel. Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>X&lt;sub&gt;1&lt;/sub&gt;</td>
<td>(\frac{\partial A}{\partial X_1} u(X_1) = \frac{1}{2} u(X_1))</td>
<td>18.407</td>
<td></td>
<td>(\frac{\partial A}{\partial a_1} u(a_1) = \frac{A_c}{2} u(a_1))</td>
<td>7.849</td>
<td></td>
</tr>
<tr>
<td>X&lt;sub&gt;2&lt;/sub&gt;</td>
<td>(\frac{\partial A}{\partial X_2} u(X_2) = \frac{1}{2} u(X_2))</td>
<td>17.429</td>
<td></td>
<td>(\frac{\partial A}{\partial a_2} u(a_2) = \frac{A_c}{2} u(a_2))</td>
<td>7.432</td>
<td></td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
<td>(\frac{\partial A}{\partial A_c} u(A_c) = \frac{a_1 + a_2}{2} u(A_c))</td>
<td>32.415</td>
<td></td>
</tr>
<tr>
<td><strong>SUB TOTAL</strong></td>
<td>(\sqrt{\left(\frac{\partial A}{\partial X_1} u(X_1)^2\right) + \left(\frac{\partial A}{\partial X_2} u(X_2)^2\right) + 2 \frac{\partial A}{\partial X_1} \frac{\partial A}{\partial X_2} \text{cov}(X_1, X_2)})</td>
<td>25.35</td>
<td></td>
<td>-</td>
<td>34.17</td>
<td></td>
</tr>
<tr>
<td>Correlations between A&lt;sub&gt;1&lt;/sub&gt; and A&lt;sub&gt;2&lt;/sub&gt;</td>
<td>(\frac{\partial A}{\partial X_1} \frac{\partial A}{\partial X_2} \text{cov}(X_1, X_2) = \frac{1}{2} a_1 a_2 u(A_c)^2)</td>
<td>525.0</td>
<td></td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Combined standard uncertainty</td>
<td>(\sqrt{\left(\frac{\partial A}{\partial X_1} u(X_1)^2\right) + \left(\frac{\partial A}{\partial X_2} u(X_2)^2\right) + 2 \frac{\partial A}{\partial X_1} \frac{\partial A}{\partial X_2} \text{cov}(X_1, X_2)})</td>
<td>34.17</td>
<td></td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>
Example N. 6
Uncertainty of the activity value (Cd-109)

Nuclear data: source of correlation between results [Rif. Exampl. 2]

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Unit</th>
<th>Standard Uncertainty</th>
<th>Relative Standard Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C_{TX}</td>
<td>Gross counts</td>
<td>102415</td>
<td>-</td>
<td>320</td>
<td>0.31</td>
</tr>
<tr>
<td>C_{BX}</td>
<td>Background counts</td>
<td>8841</td>
<td>-</td>
<td>94</td>
<td>1.06</td>
</tr>
<tr>
<td>C_{NX}</td>
<td>Net counts</td>
<td>93574</td>
<td>-</td>
<td>333</td>
<td>0.36</td>
</tr>
<tr>
<td>TX</td>
<td>Counting Time</td>
<td>80000</td>
<td>s</td>
<td>160</td>
<td>0.2</td>
</tr>
<tr>
<td>ε</td>
<td>Counting efficiency</td>
<td>1.31</td>
<td>%</td>
<td>0.02</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Expressions

\[ \varepsilon = \frac{C_N}{T \gamma A_s DF} \]

\[ A_x = \frac{C_{N,x}}{T_{X,s} I_{x,s} e} = \frac{C_{N,x}}{T_{X,s} I_{x,s} C_N} = \frac{C_{N,s} T_{A,s} DF}{T_{X,s} C_N} \]

\[ A_x = \frac{C_{N,s} T_{A,s} DF}{T_{X,s} C_N} \]

Source of uncertainty budget

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Expression</th>
<th>Value</th>
<th>Rel. Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>C_{NX}</td>
<td>( u'(C_{N,x}) )</td>
<td>0.356</td>
<td>0.28</td>
</tr>
<tr>
<td>TX</td>
<td>( u'(I_x) )</td>
<td>0.200</td>
<td>0.2</td>
</tr>
<tr>
<td>( I_x )</td>
<td>( u'(I_x) )</td>
<td>-1.6438</td>
<td>0.25</td>
</tr>
<tr>
<td>ε</td>
<td>( u'(\varepsilon) )</td>
<td>1.704</td>
<td>0.14</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>0.200</td>
<td>0.356</td>
</tr>
<tr>
<td><strong>SUB TOTAL</strong></td>
<td></td>
<td>2.403</td>
<td>0.606</td>
</tr>
</tbody>
</table>

Correlation between \( I_x \) and \( \varepsilon \)

\[ \frac{1}{A_x^2} \left( \frac{\partial A_x}{\partial I_x} \right) \frac{\partial A_x}{\partial \varepsilon} \left( \frac{\partial I_x}{\partial \varepsilon} \right) = -2u'(I_x) u'(\varepsilon) = -5.38 \times 10^{-3} \]

Combined standard uncertainty

\[ 0.607 \]

\[ \frac{1}{A_x^2} \left( \frac{\partial A_x}{\partial I_x} \right) \frac{\partial A_x}{\partial \varepsilon} \left( \frac{\partial I_x}{\partial \varepsilon} \right) = 2 \left( \frac{1}{A_x} \frac{\partial A_x}{\partial \varepsilon} \right) \left( \frac{1}{A_x} \frac{\partial A_x}{\partial I_x} \right) \left( \frac{\partial I_x}{\partial \varepsilon} \right) = 2 \left( \frac{1}{A_x} \frac{\partial A_x}{\partial \varepsilon} \right) \left( \frac{1}{A_x} \frac{\partial A_x}{\partial I_x} \right) \left( \frac{\partial I_x}{\partial \varepsilon} \right) = 2 \left( \frac{1}{I_x} \right) \left( \frac{1}{\varepsilon} \right) \left( \frac{\partial I_x}{\partial \varepsilon} \right) = 2u'(I_x) u'(\varepsilon) = -2u'(I_x) \]
Example N. 7a
Reproducibility of counting geometry

\[ \varepsilon_a(d, h) = \varepsilon_p(d) \frac{\Omega_a(d, h)}{\Omega_p(d)} \]

\[ \Omega_a(d, h) = 1 - \frac{1}{h} \left( \sqrt{d^2 + h^2}^2 + R^2 - \sqrt{d^2 + R^2} \right) \]

This expression was obtained by integration over \( h \) of the fractional solid angle for point source:

\[ f_g = \frac{\Omega}{4\pi} = 2\pi \left( 1 - \frac{d}{\sqrt{d^2 + R^2}} \right) \approx \frac{R^2}{4d^2} (d \gg R) \]
Example N. 7b
(Reproducibility of counting geometry

[M. Makarewics, 2005]
Example of uncertainty evaluation

According to ISO (GUM)

Example: components of the standard uncertainty (%) of the activity concentration

<table>
<thead>
<tr>
<th>Radionuclide and corresponding photon energy (keV)</th>
<th>$^{40}$K</th>
<th>$^{228}$Ac</th>
<th>$^{212}$Pb</th>
<th>$^{208}$Tl</th>
<th>$^{214}$Pb</th>
<th>$^{214}$Bi</th>
<th>$^{208}$Tl</th>
<th>$^{228}$Ac</th>
<th>$^{214}$Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source of uncertainty</td>
<td>dead time</td>
<td>counting statistics</td>
<td>background</td>
<td>counting efficiency</td>
<td>photon emission probability</td>
<td>spectral deconvolution</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1461</td>
<td>911</td>
<td>238</td>
<td>2614</td>
<td>351</td>
<td>609</td>
<td>253</td>
<td>830</td>
<td>839</td>
</tr>
<tr>
<td>dead time</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.8</td>
<td>0.6</td>
<td>0.6</td>
<td>1.2</td>
<td>1.1</td>
<td>1.3</td>
</tr>
<tr>
<td>counting statistics</td>
<td>0.9</td>
<td>2.0</td>
<td>1.1</td>
<td>2.9</td>
<td>1.8</td>
<td>1.2</td>
<td>1.15</td>
<td>1.14</td>
<td>0.73</td>
</tr>
<tr>
<td>background</td>
<td>0.7</td>
<td>1.0</td>
<td>1.4</td>
<td>1.7</td>
<td>4.9</td>
<td>5.4</td>
<td>92</td>
<td>109</td>
<td>78</td>
</tr>
<tr>
<td>counting efficiency</td>
<td>1.3</td>
<td>1.3</td>
<td>1.3</td>
<td>13</td>
<td>13</td>
<td>1.3</td>
<td>1.3</td>
<td>1.3</td>
<td>1.3</td>
</tr>
<tr>
<td>photon emission probability</td>
<td>0.6</td>
<td>0.001</td>
<td>0.9</td>
<td>0.2</td>
<td>0.9</td>
<td>1.4</td>
<td>9.8</td>
<td>3.3</td>
<td></td>
</tr>
<tr>
<td>spectral deconvolution</td>
<td>1.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1.5</td>
<td>-</td>
<td>2.0</td>
<td>2.7</td>
<td>3.0</td>
</tr>
<tr>
<td>Combined standard uncertainty (%)</td>
<td>2.1</td>
<td>2.6</td>
<td>2.4</td>
<td>3.7</td>
<td>6.0</td>
<td>5.8</td>
<td>147</td>
<td>181</td>
<td>107</td>
</tr>
</tbody>
</table>
Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

UNCERTAINTY IN GAMMA SPECTROMETRY

1. Metrology, measurement fundamentals
2. Uncertainty evaluation theory
3. Uncertainty components in gamma spectrometry
4. Methods for uncertainty determination
5. Application, exercises, Examples, Discussion
6. Characteristic limits
7. Quality control
8. Discussion with participants
THEORY


Detection Limit and Decision threshold: Definitions

**DECISION QUANTITY**: random variable for the decision whether the physical effect to be measured is present or not

**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

**DETECTION LIMIT**: smallest value of the measurand which is detectable by the measuring method

**CONFIDENCE INTERVAL**: 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 a given probability
Three simple questions in particle counting

\( N_0 \) : BACKGROUND counts

\( N_s \) : SAMPLE counts

**QUESTION N°1**: Which is the value of the net count that, when exceeded by the result of an actual measurement, one decides that there is a real contribution from the sample?

**QUESTION N°2**: Which is the smallest value of the sample contribution that is detectable by the measuring system?

**QUESTION N°3**: If such a contribution has been detected, which is the interval that includes the true value with a given probability?
Possible answer to question N. 1

- Compare $N_s$ with $N_0$, considering the statistical fluctuation of $N_0$:
  - $\text{Var}(N_0) = N_0$ (Poisson)
  - $L = 3 \times (N_0)^{1/2}$ (3σ Criteria, ICRU 22, 1979)

**PROBLEMS:**

1. arbitrary choice of the factor 3;

2. $N_s > L$: can be a background fluctuation?

3. $N_s < N_0$: there is no contribution from the sample?

**A MORE RIGOROUS ANSWER IS NEEDED:**
Answer to question N. 1 (Decision threshold)

**DECISION THRESHOLD:**

\[ N_n^* = k_\alpha \sigma_n \]  
(Currie, 1968)

THE DECISION THRESHOLD is the critical value for the statistical test for the decision between the hypothesis that the sample 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 shall be designed such that the probability of wrongly rejecting the hypothesis (error of the first kind) is equal to a given value \( \alpha \).

### Table: Counts, Expectation Value, and Standard Deviation

<table>
<thead>
<tr>
<th></th>
<th>COUNTS</th>
<th>EXPECTATION VALUE</th>
<th>STANDARD DEVIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>BACKGROUND</td>
<td>( N_0 )</td>
<td>( \nu_0 )</td>
<td>( \sigma_0 = \sqrt{\nu_0} )</td>
</tr>
<tr>
<td>SAMPLE</td>
<td>( N_s )</td>
<td>( \nu_s )</td>
<td>( \sigma_s = \sqrt{\nu_s} )</td>
</tr>
<tr>
<td>NET</td>
<td>( N_n = N_s - N_0 )</td>
<td>( \nu_n = \nu_s - \nu_0 )</td>
<td>( \sigma_n = \sqrt{\nu_s + \nu_0} )</td>
</tr>
</tbody>
</table>

**HP:** \( \nu_n = 0 \) \( (\nu_s = \nu_0) \) \( \sigma_n = (2\nu_0)^{1/2} \) **NO contribution from sample**

\[ \alpha \quad K_\alpha \]

<table>
<thead>
<tr>
<th>( \alpha )</th>
<th>( K_\alpha )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>1.28</td>
</tr>
<tr>
<td>0.05</td>
<td>1.64</td>
</tr>
<tr>
<td>0.025</td>
<td>1.96</td>
</tr>
<tr>
<td>0.001</td>
<td>3.09</td>
</tr>
</tbody>
</table>

\[ N_n^* = k_\alpha \sigma_n \quad \sigma_n = \sqrt{2\nu_0} \]

\[ \alpha = 0.05 \Rightarrow N_n^* = 2.32\sqrt{\nu_0} \]
Answer to question N. 2 (Detection limit)

DECISION THRESHOLD:

\[
\nu_n^* = N_n + K_\alpha \sigma_n = (K_\alpha + K_\beta) \sigma_n \quad \text{(Currie, 1968)}
\]

THE DETECTION LIMIT is the smallest true value of the measurand which is associated with the statistical test and hypothesis (made for the decision threshold) 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) shall be at most equal to a given value \(b\).

\[
\sigma_n = (2\nu_0)^{1/2}
\]

<table>
<thead>
<tr>
<th>COUNTS</th>
<th>EXPECTATION VALUE</th>
<th>STANDARD DEVIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>BACKGROUND</td>
<td>(N_0)</td>
<td>(\nu_0)</td>
</tr>
<tr>
<td>SAMPLE</td>
<td>(N_s)</td>
<td>(\nu_s)</td>
</tr>
<tr>
<td>NET</td>
<td>(N_n = N_s - N_0)</td>
<td>(\nu_n = \nu_s - \nu_0)</td>
</tr>
</tbody>
</table>

HP: \(\nu_n \neq 0 \quad (\nu_s > \nu_0) \quad \sigma_n = (2\nu_0)^{1/2} \quad \text{YES contribution from sample}\)
Generalization (a): Test of Hypothesis

**HYPOTHESIS H₀:** No sample contribution to the count.

<table>
<thead>
<tr>
<th></th>
<th>H₀ accepted</th>
<th>H₀ rejected</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>H₀ true</strong></td>
<td>OK P=1-α</td>
<td>Type I error P=α</td>
</tr>
<tr>
<td><strong>H₀ false</strong></td>
<td>Type II error P=β</td>
<td>OK P=1-β</td>
</tr>
</tbody>
</table>

α: Probability of **rejecting** the hypothesis H₀ when, in reality, it is true

β: Probability of **accepting** the hypothesis H₀ when, in reality, it is false

Analogue considerations can be made in case of counting with preset count condition
### Generalization (b): Definitions

<table>
<thead>
<tr>
<th></th>
<th>COUNTS</th>
<th>COUNTING TIME</th>
<th>COUNT RATE</th>
<th>EXPECTATION VALUE</th>
<th>STANDARD DEVIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>BACKGROUND</td>
<td>$N_0$</td>
<td>$t_0$</td>
<td>$R_0$</td>
<td>$\rho_0$</td>
<td>$\sigma_0$</td>
</tr>
<tr>
<td>SAMPLE</td>
<td>$N_s$</td>
<td>$t_s$</td>
<td>$R_s$</td>
<td>$\rho_s$</td>
<td>$\sigma_s$</td>
</tr>
<tr>
<td>NET</td>
<td>-</td>
<td>-</td>
<td>$R_n = R_s - R_0$</td>
<td>$\rho_n = \rho_s - \rho_0$</td>
<td>$\sigma_n$</td>
</tr>
</tbody>
</table>

**DECISION THRESHOLD:** Critical value $R_n^*$ of the statistical test for the decision between the alternative hypothesis:

A) $\rho_s = \rho_0$

B) $\rho_s > \rho_0$

with given probability $\alpha$ of type I error:

$$R_n^* = k_\alpha \sigma_n$$

**DETECTION LIMIT:** Smallest expectation value $\rho_0^*$, associated to the statistical test between the hypothesis A and B above, which determines a type II error with given probability $\beta$.

$$\rho_n^* = (k_{\alpha} + k_{\beta})\rho_n$$
Generalization (c): Use of $R_n^*$ and $\rho_n^*$

- $R_n^*$ should be compared with measurement results to assess whether a sample contribution has been detected (a-posteriori criteria):
  - $R_n > R_n^*$ \(\Rightarrow\) sample contribution detected;
  - $R_n < R_n^*$ \(\Rightarrow\) sample contribution not detected.

- $\rho_0^*$ should be used to check whether a measuring procedure is suitable for the purpose of the measurement. It should be compared with a specific guideline value $S(\beta)$ as specific requirements on the sensitivity of the measuring procedure for scientific, legal or other reasons (a-priori criteria):
  - $\rho_0 > \rho_0^*$ \(\Rightarrow\) the sample contribution will be detected with probability greater than $1-\beta$;
  - $\rho_0 < \rho_0^*$ \(\Rightarrow\) the sample contribution will be detected with probability less than $1-\beta$.
  - $\rho_0 < S(\beta)$ \(\Rightarrow\) measurement procedure is not adequate for the intended purpose.

- When reporting decision threshold and detection limits it is important to give the values of $\alpha$ and $\beta$ used.

- “All knowledge is divided into two categories: a priori and a posteriori knowledge”, I. Kant, Critique of Pure Reason (1791).
Estimation of background repeatability

a) Assume Poisson (or other) statistics and use the uncertainty propagation law (spectrometric measurements)

b) Measure the background variability if sources of fluctuation else than counting statistics are envisaged (sample treatment, counting system instability …)
Detection Limit and Decision Threshold: Bibliography

- Altschuler B. and Pasternak B.: Health Physics 9, 293-298 (1963)
- DIN 25482, Limit of detection and limit of decision for nuclear radiation measurements – Part 1-10
APPLICATION TO GAMMA SPECTROMETRY
Example: Decision Threshold and Detection Limit in gamma-ray spectrometry

<table>
<thead>
<tr>
<th></th>
<th>Counts</th>
<th>Count rate</th>
<th>Expectation value</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Background</td>
<td>(N_0)</td>
<td>(t)</td>
<td>(R_0)</td>
<td>(\nu_0)</td>
</tr>
<tr>
<td>Gross peak area</td>
<td>(N_s)</td>
<td>(t)</td>
<td>(R_s)</td>
<td>(\nu_s)</td>
</tr>
<tr>
<td>Net peak area</td>
<td>(N_n = N_s - N_0)</td>
<td>(t)</td>
<td>(R_n = R_s - R_0)</td>
<td>(\nu_n = \nu_s - \nu_0)</td>
</tr>
</tbody>
</table>

\[N_n = N_s - N_0\]

\[N_0 = (N_1 - N_2) \frac{b}{2l}\]

\[b \geq 4\text{ channels}\]

\[b \leq 2l \leq 10b\]

\[N_n = N_s - (N_1 + N_2) \frac{b}{2l}\]

\[\text{var}(N_n) = N_s + \frac{b}{2l} N_0\]

\[\text{var}(\rho_n)_{\rho_n = 0} = \frac{N_0}{t^2} (1 + \frac{b}{2l})\]

\[R_n = k_{1-\alpha} \sqrt{\frac{R_0}{t} (1 + \frac{b}{2l})}\]

\[\rho_n = (k_{1-\alpha} + k_{1-b}) \sqrt{\frac{R_0}{t} (1 + \frac{b}{2l})}\]

\[1 < 1 + \frac{b}{2l} < 2\]
Example of application of a decision threshold

without threshold

with threshold
Worksop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

UNCERTAINTY IN GAMMA SPECTROMETRY

1. Metrology, measurement fundamentals
2. Uncertainty evaluation theory
3. Uncertainty components in gamma spectrometry
4. Methods for uncertainty determination
5. Application, exercises, Examples, Discussion
6. Characteristic limits
7. Quality control
8. Discussion with participants
Objectives of Quality Control

- Continuous control of measurement quality
- Control of measurement system characteristics (stability, drifts, etc...)
- Possibility to start preventive actions
Internal Quality Control (QC)

- QC at system installation (short term)
- QC during system use (long term)
QC at system installation

- Start a "system book - log book"
- Follow manufacturer instruction
- Accurate definition of working conditions
- Measurement of initial system parameters
- Full documentation (spectra, analysis reports, measurement results, etc… ) regard this initial phase
Accurate definition of working conditions

- pre-regulations
- application of bias voltage to detector
- amplifier DC level
- ADC conversion gain and zero
- amplifier gain
- shaping time
- pole-zero
- amplifier "Busy" threshold
- ADC lower discrimination level
- spectrum stabiliser
Initial system performance
(to be compared with those quoted by manufacturer and with successive measurements for long term stability studies)

- optimal HV (polarisation)
- energy resolution and peak shape
- peak-to-Compton ratio
- relative (to NaI) efficiency
- electronic noise equivalent energy
- detector geometrical parameters
- window and dead layer thickness
- dead time correction accuracy
- background
Example of HV setting (1/3)

HPGe (N)

FWHM GMX 60min
40 keV

122 keV

FWHM GMX 60min
122 keV

344 keV

FWHM GMX 60min
344 keV

FWHM GMX 60min
778 keV

1112 keV

FWHM GMX 60min
1112 keV

1408 keV

FWHM GMX 60min
1408 keV

FWHM GMX 60min
40 keV

600 1100 1600 2100

HV (Volt)

FWHM (ch.)
Example of HV setting (2/3) HPGe (N)

Rateo GMX 60min 40 keV

Rateo GMX 60min 122 keV

Rateo GMX 60min 344 keV

Rateo GMX 60min 778 keV

Rateo GMX 60min 1112 keV

Rateo GMX 60min 1408 keV
Example of HV setting (3/3)
HPGe (N)
Energy resolution, FWHM (1)

- For a correct measurement:
  - total count rate < 1000 s\(^{-1}\)
  - shaping time between 2 and 8 µs
  - counts in fep > 20000
  - FWHM > 4 channels
Energy resolution, FWHM (2)

- The FWHM is measured in channels (ch) and converted in keV by the relation:
  \[ \text{FWHM}_{\text{keV}} = \text{FWHM}_{\text{ch}} \frac{(E_2-E_1)}{(C_2-C_1)} \]
  where C1 and C2 are the centroids of the full-energy-peaks corresponding to E1 and E2 respectively.

- Photon emissions used:
  - Co-60 \( E_1 = 1173.2 \text{ keV} \quad E_2 = 1332.5 \text{ keV} \)
  - Fe-55 \( E_1 = 5.9 \text{ keV} \quad E_2 = 6.49 \text{ keV} \)
  - Co-57 \( E_1 = 14.4 \text{ keV} \quad E_2 = 122.1 \text{ keV} \)
Peak-to-Compton ratio

- It is obtained from a pulse-height spectrum recorded with a Co-60 source.

- The peak-to-Compton ratio, also measured in accordance with ANSI/IEEE 325–1996, is the key indicator of a detector’s ability to distinguish low-energy peaks in the presence of high-energy sources.

- The peak-to-Compton ratio is obtained by dividing the height of the 1.33 MeV peak by the average Compton plateau between 1.040 and 1.096 MeV.
Efficiency relative to NaI, $\varepsilon_r$

- $\varepsilon_r = \frac{N}{T A} \varepsilon_{\text{NaI}}$

where:
- $A$, activity of a Co-60 point source,
- $N$, number of counts in 1332 keV fep,
- distance of 25 cm from detector window
- $T$, measurement time
- $\varepsilon_{\text{NaI}} = 1.2 \times 10^{-3}$.

- $A < 500$ kBq (to keep pile-up low);
- $N > 10000$ (counting statistics < 1%).
Noise equivalent energy

- The electronic noise, $N$, is a component of energy resolution.

- $\text{FWHM} = (N^2 + k E)^{1/2}$

- $N$ can be obtained by a measurement of the FWHM of a "artificial" peak generated by a pulser.
Detector geometry

- Detector geometry is required for:
  - "efficiency transfer" applications (geometry and self absorption corrections);
  - total efficiency calculation (coincidence-summing corrections);

- This information can be:
  - obtained by the detector manufacturer;
  - directly measured by x-ray radiography.
X-ray radiographies of HPGe (P) detectors

Window and Ge dead layer thickness

- Needed for several applications
- Thickness of the materials in front of the crystal must be given by the manufacturer
- Dead layer can be determined by a Se-75 source
Ge dead layer measurement

• Se-75 x-ray emissions: a) 10.5 keV, b) 1.7 keV

• Massic attenuation coefficient for Ge changes drastically around 11.1 keV.

• Dead layer thickness, \( t \), is obtained by the relation:

\[
\frac{N_a}{N_b} = \frac{p_a \exp(-\mu_a t)}{p_b \exp(-\mu_b t)}, \quad \text{where:}
\]

- \( N_a \) e \( N_b \) : fep counts
- \( p_a = 0.498, \ p_b = 0.076, \ \mu_a = 32.5 \text{ cm}^2 \text{ g}^{-1}, \ \mu_b = 170 \text{ cm}^2 \text{ g}^{-1} \),

• Then: \( \frac{N_a}{N_b} = 6.55 \exp(0.073 \ t) \).
Accuracy of dead time correction

The pulser peak area count rate is studied as a function of the total count rate.

\[ y = -51,066x + 50,074 \]

\[ R^2 = 1.000 \]

\[ y = -1,450x + 50,084 \]

\[ R^2 = 0.958 \]
QC during system use (1)

- Periodical check of system characteristics
- Frequency: daily, weekly or monthly
- Needed after:
  - voltage supply failure
  - cooling periods
  - repair
  - changes in electronics
  - replacement of parts
QC during system use (2)

- Main controls are:
  - energy resolution at different photon energies;
  - full-energy-peak efficiencies at different photon energies;
  - full-energy-peak position;
  - dead time correction
  - total background count rate
Example of a source support for QC
Source-detector geometry for QC
Periodical check of FWHM and full-energy-peak efficiency

- Eu-152 point source
- Energy interval: 40-1408 keV.
- Reproducible geometry
- Analysis software routinely used
- Energy resolution and decay corrected count rates for the main full-energy-peaks
- Use of control charts
- Importance of source stability
Example of QC (1)
Eu-152 spectrum
HPGe (P)

SP-308 152Eu Q.A. (sp. 01011386.chn)

Conteggi

Energia fotonica (keV)

Real Time : 10000 s
Live Time : 9858.78 s
Example of QC (2)
Energy resolution HPGe (N)
Example of QC (3)

Full-energy-peak efficiency HPGe (P)

Q.A. COAX con Eu152 (122-245-344 KeV)

Data di misura

Q.A. COAX Eu152 (778-964-1408 KeV)

Data di misura
Example of QC (4)

Full-energy-peak efficiency HPGe (N)

Q.A. GM X Eu152 (122-245-345 KeV)

Q.A. GM X Eu152 (778-964-1408 KeV)

Data di misura
Example of QC (5)
Full-energy-peak efficiency HPGe (N) 40 keV

Q.A. GMX con Eu152 (39.9 KeV)

\[ y = -7.13 \times 10^{-7} x + 3.59 \times 10^{-2} \]

Data di misura
Example of QC (6)
Dead time HPGe (P)
Example of QC (7) Live Time HPGe (N)

![Graph showing live time data](image-url)
Background control (1)

- Background must be checked after installation and periodically

- Possible causes for changes in background:
  - contamination of detector or shielding
  - changes of indoor gamma background
  - changes in human presence in room
  - changes in air ventilation
  - radioactive sources in proximity of detector
  - electronic noise
Background control (2)

- Background control particularly needed for low-level measurements
- Control of total count rate (threshold at $\approx 20$ keV)
- Spectrometry for deeper investigation
Example of QC (8)
Background spectrum HPGe (P)
Example of QC (9)
Background fep HPGe (P)
### Example of QC (10) HPGe (P)

Count rates of the main full-energy-peaks observed in the background spectrum

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Energy (keV)</th>
<th>Average count rate (1/s)</th>
<th>Std. unc. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{212}$Pb</td>
<td>238.6</td>
<td>0.00395</td>
<td>15%</td>
</tr>
<tr>
<td>$^{214}$Pb</td>
<td>241.5</td>
<td>0.00181</td>
<td>49%</td>
</tr>
<tr>
<td>$^{212}$Pb</td>
<td>265.2</td>
<td>0.00321</td>
<td>46%</td>
</tr>
<tr>
<td>$^{214}$Pb</td>
<td>351.5</td>
<td>0.00524</td>
<td>43%</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>609.3</td>
<td>0.00376</td>
<td>47%</td>
</tr>
<tr>
<td>$^{228}$Tl</td>
<td>911.1</td>
<td>0.000999</td>
<td>14%</td>
</tr>
<tr>
<td>$^{228}$Ac</td>
<td>969.9</td>
<td>0.00055</td>
<td>17%</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>1122.3</td>
<td>0.00389</td>
<td>31%</td>
</tr>
<tr>
<td>$^{228}$Ac</td>
<td>1227.9</td>
<td>0.00011</td>
<td>44%</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>1764.5</td>
<td>0.00117</td>
<td>25%</td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>2614.6</td>
<td>0.00285</td>
<td>10%</td>
</tr>
</tbody>
</table>
Proficiency test and Quality Assurance
Example: the Italian QA programme

- The main Quality Assurance program conducted by ENEA-INMRI in the field of radioactivity measurements regarded the national network for environmental radioactivity surveillance.

- The program is based on periodical calibration and intercomparison campaigns carried out by ENEA-INMRI under request of the National Agency for Environmental Protection. This program started more than 15 years ago. Beta counting and g-ray spectrometry in environmental samples are the main objects of the program.

- This QA program was effective in reducing to about 10% the maximum deviation of the results among the network laboratories. A new national intercomparison campaign was carried out for g-ray spectrometry measurements on spiked simulated filters. To this purpose about 60 sources were prepared and distributed to the participating laboratories. Results of the intercomparison are under evaluation by the ENEA-INMRI.
Preparation of standard sources for the national radioactivity surveillance network 2004-2005 intercomparison

Co-57 (# 1116) Eu-152 (# 1503) Cs-134 (# 201) Cs-137 (# 1574) Co-60 (# 40)

HCl 1N + carriers

MRL-1837

MIX

SF-1838

SF-1918

SF-1919

SF-1917

n° 80 sorgenti su disco (geom. filtro) depositate per volumetria

n° 2 sorgenti su disco (geom. filtro) depositate per gravimetria

n° 3 sorgenti su disco (geom. puntiforme) depositate per gravimetria
Quality Systems
MAIN TOPICS OF A QUALITY MANUAL

- Quality Policy
- Responsibilities
- Modalities to manage the QS (general procedures and criteria)
- Records
Outline of a typical Quality System

with reference to the following elements of the QM

1. Document control (# 4.3, ISO IEC EN 17025)
2. Review of request, tenders and contract (# 4.4, ISO IEC EN 17025)
3. Subcontracting (# 4.5, ISO IEC EN 17025)
4. Service to the client (# 4.7, ISO IEC EN 17025)
5. Corrective action (# 4.10, ISO IEC EN 17025)
6. Audit (# 4.13, ISO IEC EN 17025)
7. Management review (# 4.14, ISO IEC EN 17025)
8. Test and calibration method - method validation (# 5.4, ISO IEC EN 17025)
9. Equipment (# 5.5, ISO IEC EN 17025)
10. Assuring the quality of test and calibration results (# 5.9, ISO IEC EN 17025)
Workshop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

UNCERTAINTY IN GAMMA SPECTROMETRY

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TRACEABILITY
TRACEABILITY: definition
[Ref. BIPM/ISO International Vocabulary of basic and general terms in metrology]

- **TRACEABILITY**: property of the result of a measurement or the value of a standard whereby it can be related to stated references, usually national or international standards, through an unbroken chain (traceability chain) of comparisons all having stated uncertainties.
Standards: Definitions
[BIPM/ISO International Vocabulary
of basic and general terms in metrology]

- **MEASUREMENT STANDARD**: material measure, measuring instrument, reference material or measuring instrument intended to define, realise, conserve or reproduce a unit or one or more values of a quantity to serve as a reference.

- **INTERNATIONAL STANDARD**: standard recognised by an international agreement to serve internationally as the basis for assigning values to other standards of the quantity concerned.

- **NATIONAL STANDARD**: standard recognised by a national decision to serve, in a country, as the basis for assigning values to other standards of the quantity concerned.

- **PRIMARY STANDARD**: standard that is designated or widely acknowledged as having the highest metrological qualities and whose value is accepted without reference to other standards of the same quantity.

- **SECONDARY STANDARD**: standard whose value is assigned by comparison with a primary standard of the same quantity.

- **REFERENCE STANDARD**: standard, generally having the highest metrological quality available at a given location or in a given organisation, from which measurements made there are derived.

- **WORKING STANDARD**: standard that is used routinely to calibrate or check material measures, measuring instruments or reference materials.

- **TRANSFER STANDARD**: standard used as an intermediary to compare standards.

- **TRAVELLING STANDARD**: standard, sometimes of special construction, intended for transport between different locations.
Reference Materials: definitions
[Ref. BIPM/ISO International Vocabulary of basic and general terms in metrology]

- **REFERENCE MATERIAL**: material or substance one or more of whose property values are sufficiently homogeneous and well established to be used for the Calibration of an apparatus, the assessment of a measurement method, or for assigning values to materials.

- **CERTIFIED REFERENCE MATERIALS**: reference material, accompanied by a certificate, one or more of whose property values are certified by a procedure which establishes traceability to an accurate realisation of the unit in which the property values are expressed, and for which each certified value is accompanied by an uncertainty at a stated level of confidence.
The role of standards
[Ref. NCRP]

- The primary role of any standard is to enable the result of a measurement to be communicated as a quantity, in terms of a number and a unit, to users or to other workers in a field.

- The standard must be accompanied by a certificate giving the value of the quantity concerned to within stated limits of uncertainty and characterising all other important parameters, for example, chemical composition, ambient conditions...

- Such standards can then be used to calibrate instruments in order to use these instruments to quantify the value of the physical quantity concerned in suitable measuring conditions.
International traceability and the role of standards

The figure illustrates how the national and international measuring systems can be envisaged as being traceable to and consistent with each other. The National Metrology Institutes (NMI) maintain traceability links to the Bureau International des Poids et Mesures (BIPM) and with each other, while each of the national laboratories will presumably seek to maintain traceability with other measurement laboratories in its own jurisdiction.
International traceability and equivalence are established by:

- **International comparisons:**
  - organised by the “Bureau International des Poids et Mesures” (BIPM)
  - International reference system (SIR-BIPM)
  - Regional Metrology Organisations (EUROMET,…)

- **Bilateral comparisons between NMI’s**
The task of the BIPM is to ensure world-wide uniformity of measurements and their traceability to the International System of Units (SI).

It does this with the authority of the Convention of the Metre, a diplomatic treaty between fifty-one nations, and it operates through a series of Consultative Committees, whose members are the national metrology laboratories of the Member States of the Convention, and through its own laboratory work.

The BIPM carries out measurement-related research. It takes part in, and organizes, international comparisons of national measurement standards, and it carries out calibrations for Member States.

[http://www.bipm.org]
The MRA was drawn up by the International Committee of Weights and Measures (CIPM), under the authority given to it in the Metre Convention, for signature by directors of the NMIs of Member States of the Convention and Associates of the CGPM.

**Objectives**

- to establish the degree of equivalence of national measurement standards maintained by NMIs;
- to provide for the mutual recognition of calibration and measurement certificates issued by NMIs;
- thereby to provide governments and other parties with a secure technical foundation for wider agreements related to international trade, commerce and regulatory affairs.

**Process**

- international comparisons of measurements, to be known as key comparisons;
- supplementary international comparisons of measurements;
- quality systems and demonstrations of competence by NMIs.

**Outcome:** statements of the measurement capabilities of each NMI in a database maintained by the BIPM and publicly available on the Web.
Thank you