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Uncertainties in gamma-ray spectrometry



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Definition of uncertainty

- “parameter, associated with the result of a measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measurand” GUM 2.2.3
- Uncertainty of measurement is the **doubt** that exists about the result of any measurement. NPL-11: 2.1
It tells us something about the quality of a measurement.
- Measurand = measurement result \pm uncertainty

**There is no valid measurement result without
associated uncertainty!**

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Definition of uncertainty (2)

- Measurand = measurement result \pm uncertainty
- Example:

$$A(^{137}\text{Cs}) = (772 \pm 32) \text{ Bq}\cdot\text{kg}^{-1}$$

where the activity concentration A is the *measurand*



On belief and probability

Not possible to state how well the true value of a measurand is known, but only how well it is *believed* to be known.

Measurement uncertainty as measure of how well one believes to know ...

The *degrees* of belief are quantified in terms of *probabilities*

On belief/doubt and probability (2) **OR** Expressing uncertainty of measurement

- There is always a margin of doubt about a measurement. Questions to ask: 'How big is that margin?' and 'How bad is the doubt?'
- Therefore, two numbers are needed in order to quantify an uncertainty: One is the width of the margin, or *interval*. The other is a **confidence level**, and states how sure we are that the 'true value' is within that margin.

$$A(^{137}\text{Cs}) = (772 \pm 32) \text{ Bq}\cdot\text{kg}^{-1}$$

What is missing here?

Determine the Basic Equation

The simplified Basic Equation for gamma-ray spectrometry

$$C = AP_{\gamma}t\varepsilon$$

$$A = \frac{C}{t\varepsilon P_{\gamma}}$$

Peak Count (points to C)
 Gamma-ray emission probability (points to P_{γ})
 FEP (Full Energy Peak) efficiency (points to ε)
 Measurement time (live time) (points to t)



Way to combine uncertainties

The simplified Basic Equation for gamma-ray spectrometry

Method: Partial differentiation and summation in quadrature

$$y = f(x_1, x_2, \dots, x_n)$$

$$dy^2 = \sum \left(\frac{\partial y}{\partial x_i} \right)^2 dx_i^2 \quad \text{alternative notation: } u_c^2(y) = \sum c_i^2 \cdot u^2(x_i)$$

sensitivity coefficient, $c_i = \partial y / \partial x_i$

Special cases:

(i) $y = x_1 + x_2$ or $y = x_1 - x_2 \Rightarrow u_c^2(y) = u^2(x_1) + u^2(x_2)$

(ii) $y = x_1 \cdot x_2$ or $y = x_1 / x_2 \Rightarrow u_{c,rel}^2(y) = u_{rel}^2(x_1) + u_{rel}^2(x_2)$



Way to determine uncertainties

- **Sensitivity analysis** – Vary a parameter with one sigma up and down and see the effect on the activity (useful in Monte Carlo calculations)
- **Calculation** – Based on theoretical consideration, such as knowing that the decay process is a Poisson process
- **Tabulated data** – such as uncertainty of emission probability (P_γ) and half-life ($T_{1/2}$)
- **Experience and educated estimates** – often used to exclude uncertainties knowing that they have negligible influence, e.g. time measurements (not dead-time though)





$$A = \frac{C}{t\varepsilon P_\gamma}$$

Partial
derivatives

$$\frac{\partial A}{\partial C} = ?$$

$$\frac{\partial A}{\partial t} = ?$$

$$\frac{\partial A}{\partial \varepsilon} = ?$$

$$\frac{\partial A}{\partial P_\gamma} = ?$$

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$$A = \frac{C}{t\varepsilon P_\gamma}$$

Partial
derivatives

$$\frac{\partial A}{\partial C} = \frac{1}{t\varepsilon P_\gamma}$$

$$\frac{\partial A}{\partial t} = -\frac{C}{t^2\varepsilon P_\gamma}$$

$$\frac{\partial A}{\partial \varepsilon} = -\frac{C}{t\varepsilon^2 P_\gamma}$$

$$\frac{\partial A}{\partial P_\gamma} = -\frac{C}{t\varepsilon P_\gamma^2}$$

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$$A = \frac{C}{t \varepsilon P_\gamma}$$

			method 1	method 2
			rel.uncert. (dx _i /x _i)	(Sensitivity coefficient) ∂A/∂x _i
				(∂A/∂x _i) ² · dx _i ²
	value x _i	uncertainty dx _i		
C	350 counts	18.7 counts		
t	43200 s	10 s		
ε(FEP)	0.05	0.003		
P _γ	0.8	0.012		
A = _____ Bq			dA/A = _____ %	dA = (equation?)
			dA = _____ Bq	dA = _____ Bq

The same results with both methods ?



$$A = \frac{C}{t \varepsilon P_\gamma}$$

			method 1	method 2
			rel.uncert. (dx _i /x _i)	(Sensitivity coefficient) ∂A/∂x _i
				(∂A/∂x _i) ² · dx _i ²
	value x _i	uncertainty dx _i		
C	350 counts	18.7 counts	5.34 %	0.5787 · 10 ⁻³
t	43200 s	10 s	0.02 %	4.689 · 10 ⁻⁶
ε(FEP)	0.05	0.003	6.0 %	4.051 · 10 ⁰
P _γ	0.8	0.012	1.5 %	0.2532 · 10 ⁰
A = 0.203 Bq			dA/A = 8.17 %	dA = √ Σ (∂A/∂x _i) ² · dx _i ²
			dA = 0.0166 Bq	dA = 0.0166 Bq

The same results with both methods*: A = (203 ± 17) mBq

*Since it is in fact the same method

The (almost) complete basic equation for gamma-ray spectrometry

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$$A = \frac{C_{TOT} - C_{Peak}^{Bkg} - C_{Continuum}}{\epsilon_{REF}^{Exp} \frac{\epsilon_{Sample}^{MC}}{\epsilon_{REF}^{MC}} P_{\gamma}} e^{\lambda t_d} \frac{\lambda}{(1 - e^{-\lambda t_m})} K_1 K_2 K_3$$

Measured
(Exp)
Reference
sample (Ref)

Correction factor from
calculation or Monte
Carlo code (MC)

K_1 = summing correction

K_2 = Branching correction

K_3 = Equilibrium correction

t_d = decay time (to a reference date)

t_m = measurement live time

Also deadtime correction!

Combine activities from several gamma-rays from one radionuclide

Combine activities from several daughters into one activity for the mother (like for ^{226}Ra and the ^{222}Rn -daughters)

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Activity calculations & efficiency

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$$A = \frac{C_{TOT} - C_{Peak}^{Bkg} - C_{Continuum}}{\epsilon_{REF}^{Exp} \frac{\epsilon_{Sample}^{MC}}{\epsilon_{REF}^{MC}} P_{\gamma}} e^{\lambda t_d} \frac{\lambda}{(1 - e^{-\lambda t_m})} K_1 K_2 K_3$$

Reference
sample similar
to the
measured

Correction factor

K_1 = summing correction

K_2 = Branching correction

K_3 = Equilibrium correction

t_d = decay time (to a reference date)

t_m = measurement live time

Also deadtime correction!

Each parameter carries an uncertainty!!

Correlated efficiency values, only counting stats. reduce u_c

Combined activities from several gamma-rays and from several daughters to activity for one radionuclide

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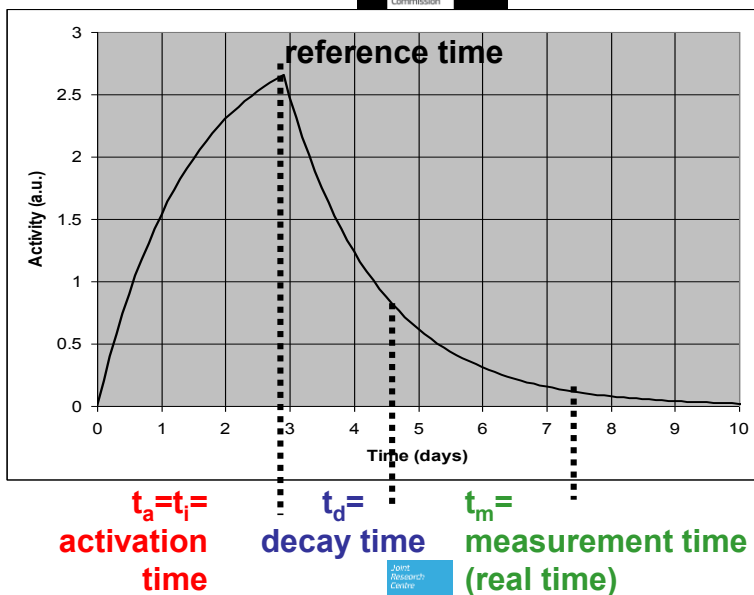
Example: Decay corrections

Correcting for decay from reference date to measurement date

$$e^{\lambda t_d}$$

where t_d is the decay time

Definitions – Time corrections



Uncertainty of decay corrections

$$\lambda = \frac{\ln(2)}{T} \Rightarrow \frac{d\lambda}{dT} = -\frac{\ln(2)}{T^2} \Rightarrow d\lambda = -dT \frac{\ln(2)}{T^2}$$

$$A = Ke^{-\lambda t_d} \Rightarrow \frac{dA}{d\lambda} = -t_d Ke^{-\lambda t_d} \Rightarrow dA = -d\lambda t_d Ke^{-\lambda t_d} = -d\lambda t_d A$$



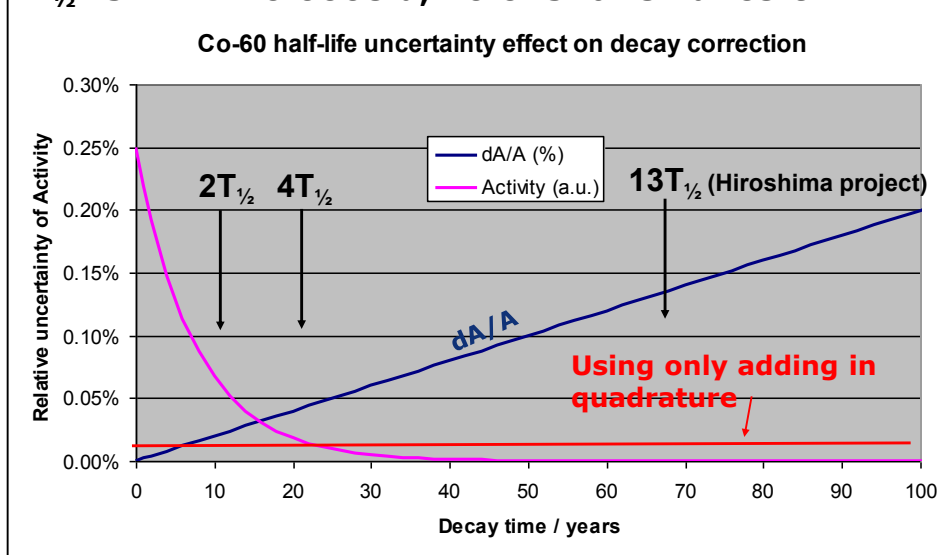
$$\frac{dA}{A} = -d\lambda t_d = t_d \ln(2) \frac{dT}{T^2}$$

... is proportional to decay time

→ adding in quadrature is incorrect

Example

$T_{1/2} = 5.2711 \pm 0.0008 \text{ a}$, 0.015% rel. uncert.



Example

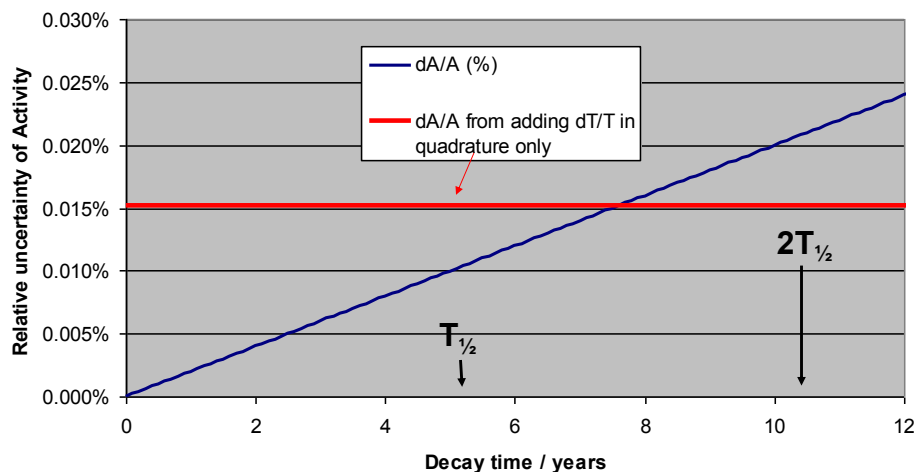
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$T_{1/2} = 5.2711 \pm 0.0008$ a, 0.015% rel. uncert.

Co-60 half-life uncertainty effect on decay correction



Example

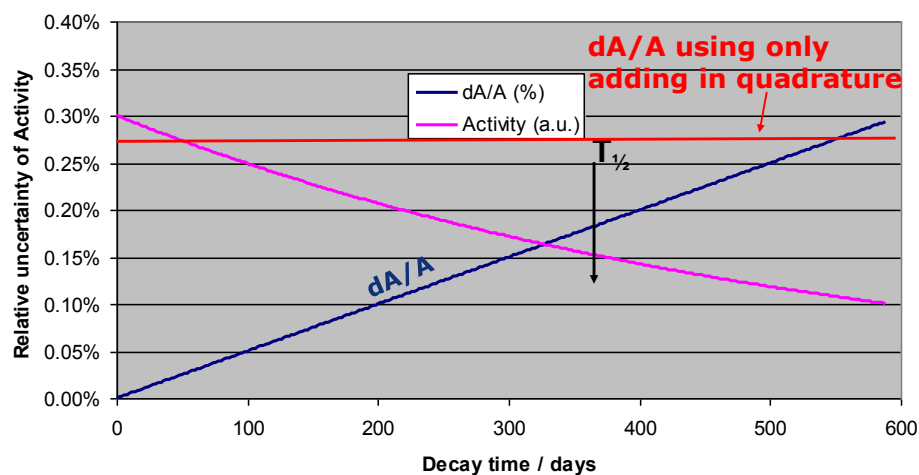
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$T_{1/2} = 372.6 \pm 1.0$ d, 0.27% rel. uncert.

Ru106 half-life uncertainty effect on decay correction



Basic Equation – Time corrections

Decay during measurement

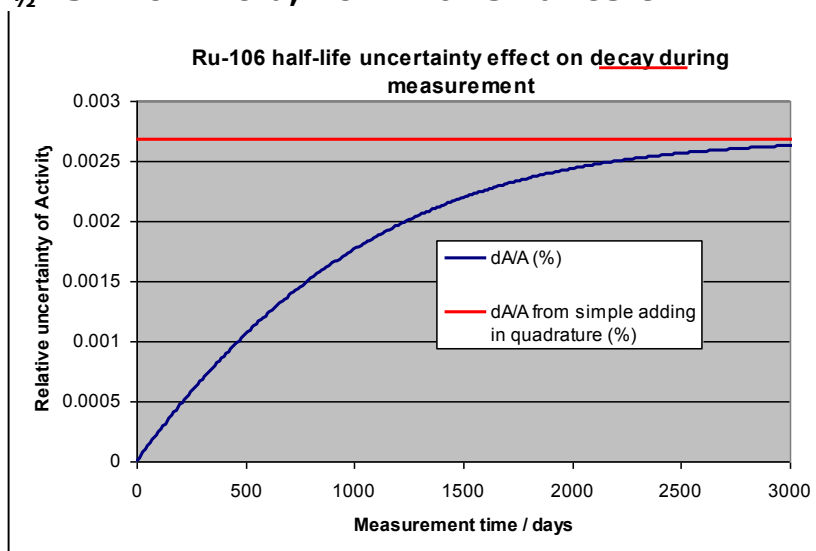
$$\frac{1}{t_m} \longrightarrow \frac{\lambda}{(1 - e^{-\lambda t_m})}$$

Uncertainty of decay during measurement

Gives more complicated expression which is not linear with measurement time

Example

$T_{1/2} = 372.6 \pm 1.0$ d, 0.27% rel. uncert.

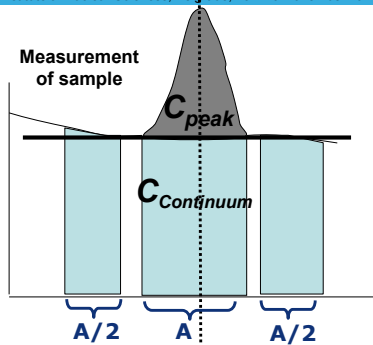


Basic Equation – Peak Counts determined using RoI method (particularly useful < 100 peak counts)

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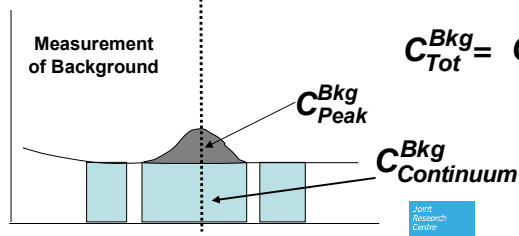


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$$C_{\text{NET}} = C_{\text{Peak}} - C_{\text{Peak}}^{\text{Bkg}}$$

$$C_{\text{TOT}} = C_{\text{Continuum}} + C_{\text{Peak}}$$



$$C_{\text{Tot}}^{\text{Bkg}} = C_{\text{Continuum}}^{\text{Bkg}} + C_{\text{Peak}}^{\text{Bkg}}$$



Peak Counts determined using RoI method

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$$C_{\text{NET}} = C_{\text{Peak}} - C_{\text{Peak}}^{\text{Bkg}} = C_{\text{TOT}} - C_{\text{Continuum}} - C_{\text{Tot}}^{\text{Bkg}} + C_{\text{Continuum}}^{\text{Bkg}}$$

$$(u_{C_{\text{NET}}})^2 = (u_{C_{\text{TOT}}})^2 + (u_{C_{\text{Continuum}}})^2 + (u_{C_{\text{Tot}}^{\text{Bkg}}})^2 + (u_{C_{\text{Continuum}}^{\text{Bkg}}})^2$$



Basic Equation – Peak Counts determined using Rol method (useful < 100 net counts)

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Example 1 (assuming same measurement time of sample and background)

$$C_{Tot} = 169 \pm 13$$

$$C_{Continuum} = 121 \pm 11$$

$$C_{Tot}^{Bkg} = 49 \pm 7$$

$$C_{Continuum}^{Bkg} = 25 \pm 5$$

$$C_{Net} = 169 - 121 - 49 + 25 \text{ counts}$$

$$u_{C_{NET}} = \sqrt{169 + 121 + 49 + 25}$$

$$\longrightarrow C_{Net} = 24 \pm 19 \text{ counts} \quad u_{C_{NET}}^{rel.} = 80 \%$$

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Basic Equation – Peak Counts determined using Rol method (useful < 100 net counts)

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Example 2 (assuming same measurement time of sample and background)

$$C_{Tot} = 169 \pm 13$$

$$C_{Continuum} = 121 \pm 11$$

$$\left. \begin{array}{l} C_{Tot}^{Bkg} = 0 \pm 0 \\ C_{Continuum}^{Bkg} = 0 \pm 0 \end{array} \right\}$$

When do you “know” that the background is 0? – you may have to assign a one-sided uncertainty even if you detect no counts.

$$C_{Net} = 48 \pm 17 \text{ counts}$$

$$u_{C_{NET}}^{rel.} = 35 \%$$

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Peak Counts determined using RoI method (useful < 100 net counts)

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Example 3 (assuming same measurement time of sample and background)

$$C_{Tot} = 169 \pm 13$$

$$C_{Continuum} = 0 \pm 0 \quad \leftarrow$$

Tricky case. You have no counts in the two background regions. Use educated estimates of uncertainty or approximation

$$C_{Tot}^{Bkg} = 0 \pm 0$$

$$C_{Continuum}^{Bkg} = 0 \pm 0$$

$$C_{Net} = 169 \pm 13 \text{ counts} \quad u_{C_{NET}}^{rel.} = 7 \%$$



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***Detection limits with zero background**

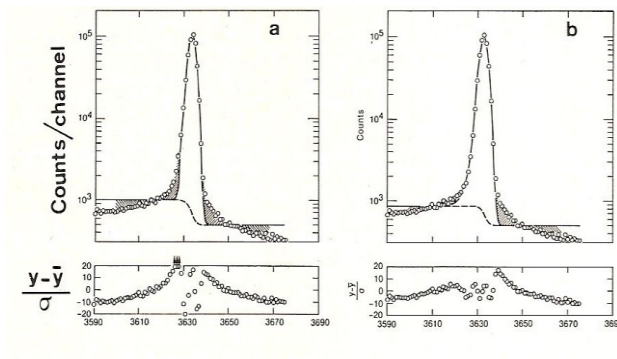
See e.g. Hurtgen, Jerome and Woods, Applied Radiation and Isotopes 53 (2000) 45-50.





A Golden Rule

- **Don't trust an automatic fit. Use visual inspection whenever possible!**



Studying residuals can help to estimate uncertainty



Other sources of peak area uncertainties (from fitting)

- Interfering peaks – *due to (i) non-identified radionuclides, (ii) coincidence summing, (iii) random summing, (iv) escape peaks....*
- Non-Gaussian peak shape (X-rays, poor counting statistics, nuclear recoil or a bad detector)
- Bad decay data (mainly emission probabilities) when using library-driven fitting
- Change in background count rate – *due to (i) change in cosmic ray flux, (ii) variations in radon-concentration, (iii) non-stable electronics, (iv) contaminated sample container,*



Uncertainties from decay data

- P_γ (gamma-ray emission probability) and λ (decay constant) can vary significantly between different sources of data - in normal gamma-ray spectrometry situations, the P_γ is more crucial than λ as the activity is directly proportional to the emission probability

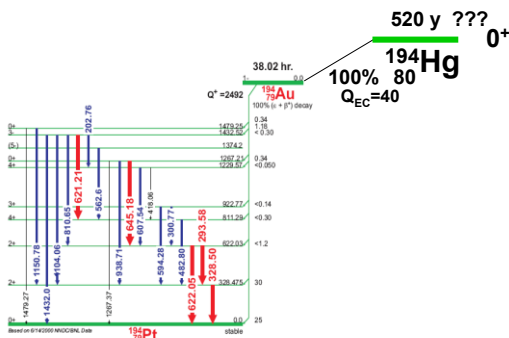


Uncertainties from decay data

- Library data may not be up to date and not from DDEP.
- Pay attention to equilibrium between mother – daughter, which decay constant (half-life) to use. Not always correct in commercial software!

EXAMPLE-1 of bad decay data ($T_{1/2}$)

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Secular equilibrium between ^{194}Au and ^{194}Hg

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Half-life of ^{194}Hg

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- **444** \pm 77 years as tabulated in the Nuclear Data Sheets [NuDat, 2007].
- This value is the mean of two measurements of **367** \pm 55 y (Probst, Alderliesten and Jahn, 1979) and **520** \pm 20 y (Hornshøj et al., 1981)
- Before that the “best” value was **260** y (Probst, Alderliesten and Jahn, 1976)
- Before that “best” value was **> 15** y (Nucl. Chem. Lett. 9, 1973, p.611)
- Before that “best” value was **1.3** y (Nucl. Data B7, 1972, p.95)
- First detected by Brunner et al. in 1955: $T_{1/2}$ **130** d

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Published half-lives of ^{194}Hg

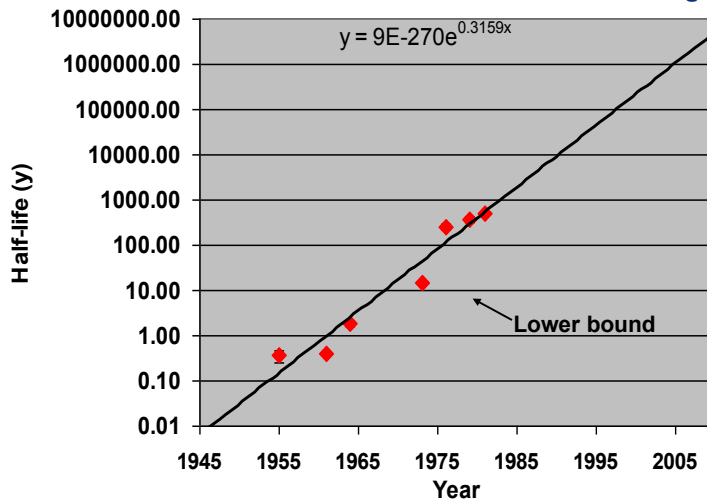
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=> Issue for radioactive waste management!

Half-life development of Hg-194



Bad data, example-2: U-238 (P_γ of Th^{234})

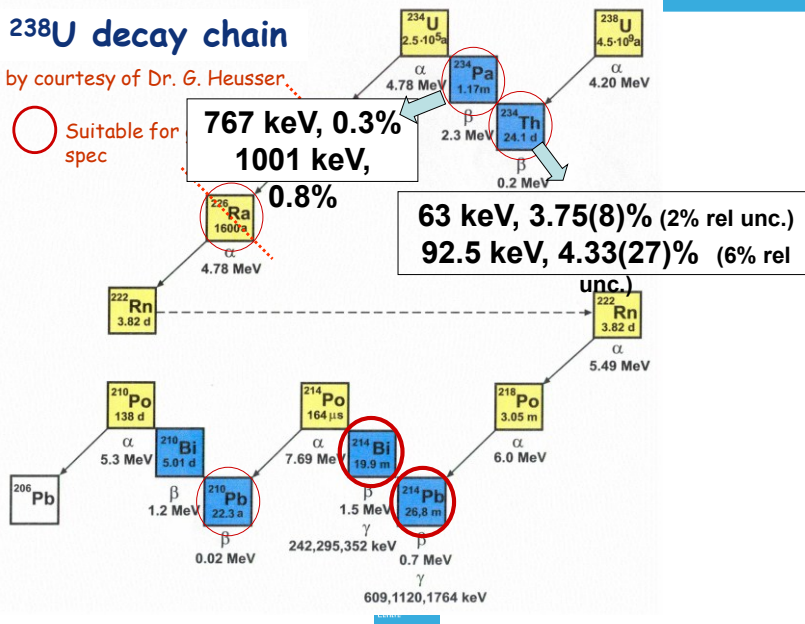
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^{238}U decay chain

by courtesy of Dr. G. Heusser

Suitable for spec





U-238 is difficult to measure accurately using gamma-ray spectrometry.

Still it is widely done since GS is used anyhow for determining other radionuclides



Decay data – well known?

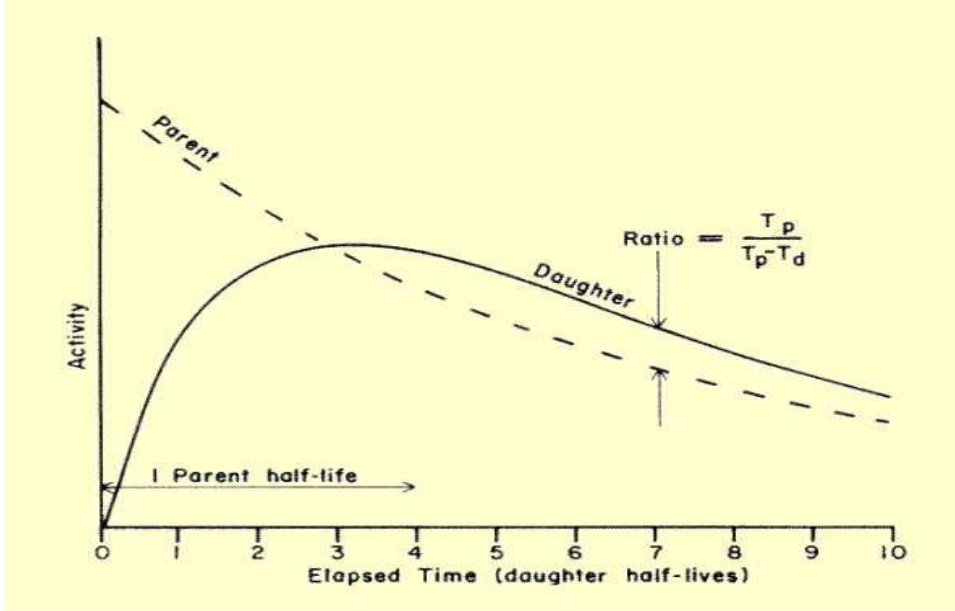


P_{γ} (63 keV):

Reported value	Reference
4.8 (6)%	Nucléide - 2000
4.80%	Mini Table de Radionucléides, 2007
4.49%	Genie-2000
4.1 (7)%	$\alpha\beta\gamma$ -Table, Wahl
4.1 (7)%	PTB-bericht 1998
4.00 (6)%	Nuclides2000
3.75 (8)%	DDEP - 2009
3.7 (2)%	The Radiochemical Manual (1988)
3.7 (4)%	NNDC
3.69 (7)%	NDS - 2007
3.6 (1)%	PTB-Ra-16/3, 1989

Std.dev: 0.45; Rel Std. dev. 11 %; (Max-min)/mean: 30 %





Basic Equation – Efficiency

1. Measure FEP efficiency in a calibration source with several gamma-rays
2. Fit an empirical (or semiempirical) function to the data
3. If the “real sample” is different from the calibration source in shape, mass or composition – calculate efficiency correction using an efficiency transfer code

And... Note that decay corrections are usually needed !!!

$$\varepsilon = \frac{C}{tAP_{\gamma}}$$

BUT: Beware of coincidence summing corrections!!!

Basic Equation – Efficiency Transfer

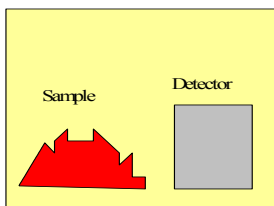
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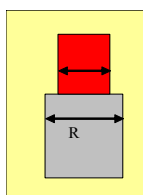
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From calibration to measurement

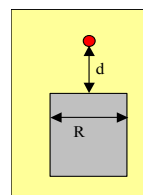
Sample and measurement instrument



Measurement geometry



Calibration geometry



Sample treatment

- Grinding
- Homogenization
- Weighing
- Preparation of measurement geometry

Efficiency transfer

- Detector characterization
- Sample characterization
- Measurement of a standard
- Calculation of the efficiency transfer factor

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Basic Equation – Efficiency Transfer

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Several codes for calculating efficiency transfer exist:
EFFTRAN, GESPECOR, DETEFF, ETNA

Efficiency corrections can be calculated using general
purpose Monte Carlo codes like: GEANT, PENELOPE,
EGS4 and MCNP

$$\varepsilon = \varepsilon_{REF}^{Exp} \frac{\varepsilon_{Sample}^{MC}}{\varepsilon_{REF}^{MC}}$$

Efficiency correction factor

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Uncertainty contributions to FEP efficiency

1. Uncertainty of calibration source
2. Uncertainties from the measurement of the calibration source
3. Uncertainty of coincidence summing corrections (and decay corrections) of calibration source
4. Uncertainty of fitting function
5. Geometrical and physical uncertainties of the sample, detector and shield – used in calculating the efficiency transfer –

5 bis. Is there e.g. a homogeneous distribution of activity in the sample?? Think of radon-daughters escaping a powder sample or activation products in steel. The uncertainty of this can be determined with a sensitivity analysis using Monte Carlo simulations.



Estimating uncertainty contributions to FEP efficiency

Often difficult to estimate a complete set of uncertainty contributions to the efficiency. Some possibilities are:

1. Participate in proficiency testing regularly. After some time one can see that the efficiencies are within a certain range and in many laboratories the uncertainty of the efficiency is around 5% if a carefully calibrated setup is used. *Being very meticulous, this might be reduced to 1% or 2%.*
2. Do a sensitivity analysis. See what are realistic uncertainties of e.g. the deadlayer thickness of a HPGe-detector. Calculate the efficiency after changing the deadlayer thickness by $\pm 1 \text{ u}$.



Estimating uncertainty contributions to FEP efficiency using only Monte Carlo simulation

In cases of new types of samples when one does not have the possibility of using a reference standard, one often has to rely on the absolute value of ε , calculated using a Monte Carlo simulation.

This approach is somewhat dangerous and requires a very good model of the *detector and sample* as well as experience. Uncertainties around 20% are usually reached. Uncertainties of 5% might be reached when great efforts have been made to establish an even better model.

Blunders – not uncertainty



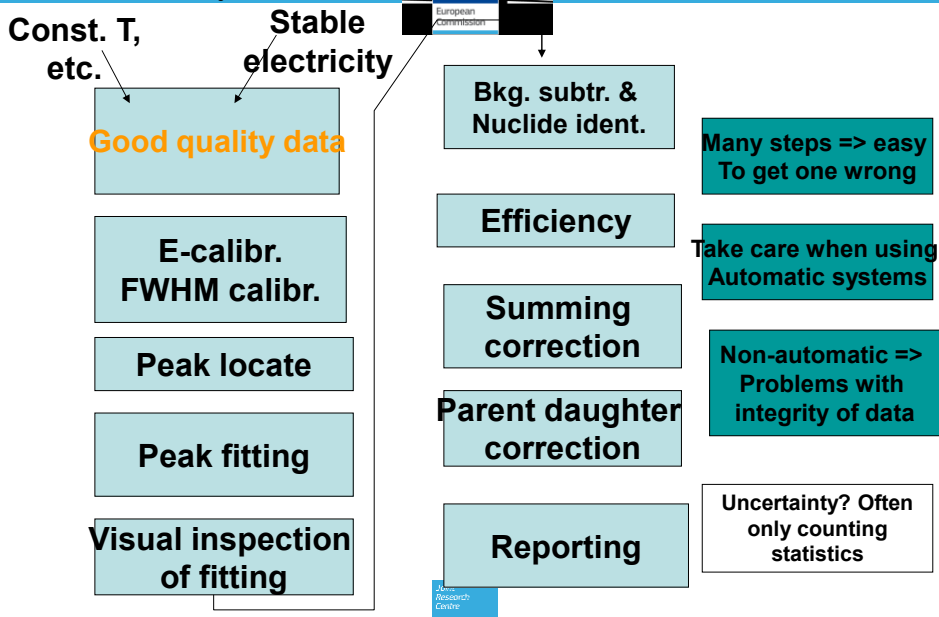
Calculations – where blunders are made, i.e. unrecognized errors, missing as uncertainty

1. Making full use of commercial software
 - Most suited for routine analysis (analysis of many samples of the same type - matrix, mass, radionuclides)
 - Low risk of blunders
 - Drawbacks: User has not full control of all calculations.
2. Home-made specially designed software
 - Takes long to implement
 - Debugging is time-consuming, but absolutely needed
3. Making tailor-made calculation sheets in general purpose codes like MS Excel
 - Can get “messy” unless care is taken to “programme cleanly”
 - Copy-paste errors

Spectrum analysis and calculations in commercial software

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$$A = \frac{C_{TOT} - C_{Peak}^{Bkg} - C_{Continuum}}{\epsilon_{REF}^{Exp} \frac{\epsilon_{Sample}^{MC}}{\epsilon_{REF}^{MC}} P_{\gamma}} e^{\lambda t_d} \frac{\lambda}{(1 - e^{-\lambda t_m})} K_1 K_2 K_3$$

K_1 = summing correction

K_2 = Branching correction

K_3 = Equilibrium correction

Combine activity from several gamma-rays to activity for one radionuclide

Combine activity from several daughters to one parent

Ex.: ^{226}Ra from ^{214}Bi and ^{214}Pb



Combining several gamma-rays per radionuclide (and several daughters per mother nuclide)

1. Calculating weighted means

- Identify correlated and uncorrelated quantities – **add only uncorrelated quantities like counting statistics in quadrature.** “The uncertainty of the efficiency does not reduce when you make many measurements”
- Most parameters are correlated, like decay data and efficiency
- Possibly exclude outliers if there are good reasons (e.g. outside calibrated energy range or only poor decay data available)

2. Using software for “full spectrum analysis”

- One obtains one value per radionuclide
- Uncertainties in the derived activity concentrations are influenced by the covariances between the standard spectra



COMPLETE UNCERTAINTY BUDGET

- Specialised software like “GUM-Workbench”
- Often it is “good enough” to add the main relative uncertainties in quadrature (*but one must be sure of that!*)
- For “non-linear” factor (e.g. decay constant)
 - Either: calculate the “maximum” uncertainty, which often is very small and can therefore be neglected (needs to be shown)
 - Or: make a sensitivity analysis – calculate the activity when adjusting the half-life by $\pm 1 \text{ } u \text{ } (T_{1/2})$

TO SUMMARISE:

The main uncertainty components in gamma-ray spectrometry are

- **FEP efficiency** (including a full analysis of calibration sources, fitting of efficiency curve and uncertainty of geometry)
- **Corrections for true coincidence summing**
- **Counting statistics** (both sample and background) - particularly for low activity samples of course

Rounding of values (results/uncertainties)

How many digits are significant?

Does this make sense?

$$d = 50 \text{ m}$$

$$d = 50 \text{ m} \pm 0.3 \text{ m}$$

$$d = 50.123456 \text{ m} \pm 0.12 \text{ m}$$

$$d = 50 \text{ m} \pm 10 \text{ cm}$$

$$d = 50.123456 \text{ m} \pm 0.123456 \text{ m}$$

Concept: Uncertainty of uncertainty

Rounding of uncertainty

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Rounding: Rule of thumb

Assuming (at least) 10% uncertainty on the uncertainty, mention only 1 digit, or maxium 2 digits for small values

$\left\{ \begin{array}{l} 10, 11, 12, \dots 19 \\ 20, 21, 22, \dots 29 \dots 32 \end{array} \right\}$	}	→ 1 or 2 digits
$\left\{ \begin{array}{l} 3 \\ 4 \\ \dots \\ 9 \end{array} \right\}$	}	→ 1 digit

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Rounding of values

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Rounding should be performed only at the very end of the process (to avoid rounding errors).
The final **uncertainty** is only rounded up.

Examples

$d = 12.3456 \text{ m} \pm 0.03923 \text{ m}$	→	$12.35 (4) \text{ m}$
$t = 0.023447 \text{ s} \pm 1.622 \cdot 10^{-3} \text{ s}$	→	$2.34 (17) \cdot 10^{-2} \text{ s}$ $0.0234 (17) \text{ s}$ $23.4 (17) \text{ ms}$
$m = 7.35 \cdot 10^{22} \text{ kg} \pm 2.7 \cdot 10^{20} \text{ kg}$	→	$7.35 (3) \cdot 10^{22} \text{ kg}$

THE END

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Bibliography to uncertainty estimation

- GUM, Evaluation of measurement data – Guide to the expression of uncertainty in measurement (GUM), JCGM 100:2008; <http://www.bipm.org/en/publications/guides/>
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Rounding of values



Uncertainty of uncertainty

An experiment is repeated n times. The data are normally distributed with a standard deviation σ . The corresponding variance of a sample standard deviation is $\sigma^2/2n$.

Calculate the relative variation of the sample standard deviation; this is the ratio of its standard error to its mean:

$$\sqrt{\frac{\sigma^2}{2n}} / \sigma = \sqrt{\frac{1}{2n}}$$

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Rounding

Use this equation to **decide on the number of significant digits** in the following examples and round the result properly:

$$\sqrt{\frac{\sigma^2}{2n}} / \sigma = \sqrt{\frac{1}{2n}}$$

Mean	st dev mean	n	unc. on unc.	unc	result
1.1934	0.0245	8	25%	0.03	1.19
1100.4	123.12	50	10%	0.12 E3	1.10 E3
523.45	9.9123	5000	1%	10.0	523.5

