Nuclear data measurement of Lanthanides

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Introduction

- Radionuclides are intensively used worldwide in many fields
  - Nuclear medicine, nuclear decommissioning, environment monitoring…
- Increase interest for new isotopes in nuclear medicine
  - There is a need for a precise knowledge of nuclear data (half-life, energy, emission probability…) and precise quantification of activity to optimize isotopes production and therapy
- National metrology Institutes (NMIs) are developing primary measurement techniques to measure the activity with an accuracy of around 0.5%, also used to measure nuclear data.
  - Activity measurement results are traceable to an international measuring system, the SIR

SIR
Système International de Référence

- In practice, the unit (Bq for activity) is maintained in each country by a NMI or DI (a Designated Institute) and are traceable to the SIR.
- To ensure good measurements in each country, a program of comparison is defined to regularly demonstrate the capability of each NMI to properly measure the **activity** of a given sample isotope.
  - **The specificity of the radioactivity (each isotope has different properties - emission type, half-life, emission probabilities…) implies that the activity of each isotope has to be measured independently.**
- This capability is evaluated with results of comparison between NMIs as well as publications from these NMIs
  - The comparison results provide also the mean **to compare and validate the different measurement techniques** used in the different NMIs

Isotope standardization

As primary standardization of activity is based on the detection and quantization of the emitted radiation, it involves different techniques and approaches from one radionuclide to another.

The best primary standardization methods are designed as:

- Their calibration based on basic physical principles, not on other radioactivity measurements.
- The ideal method is under statistical control and the total uncertainty on the result preferably reduced to a minimum.
- Their result is independent of the nuclear decay data (and associated uncertainties)

In general, the primary standardization methods are based on the counting of individual decays, via the emitted particles.

We distinguish:

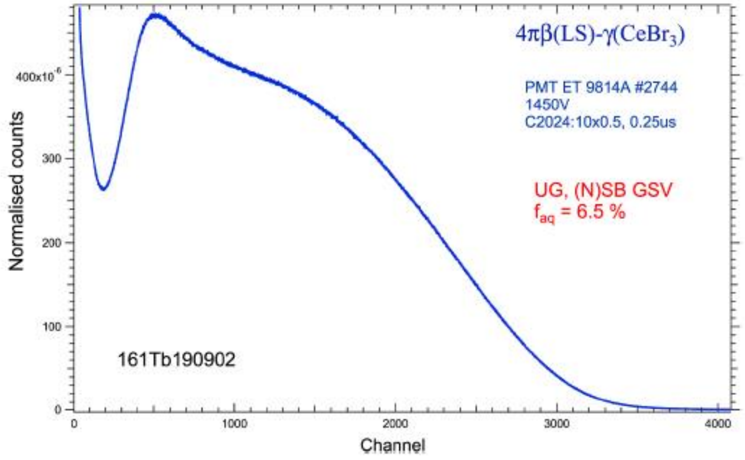
- **high-geometry (4π or 2π) methods**, with a 100% or 50% geometrical efficiency
- **defined solid-angle counting methods**
- **coincidence counting methods.**

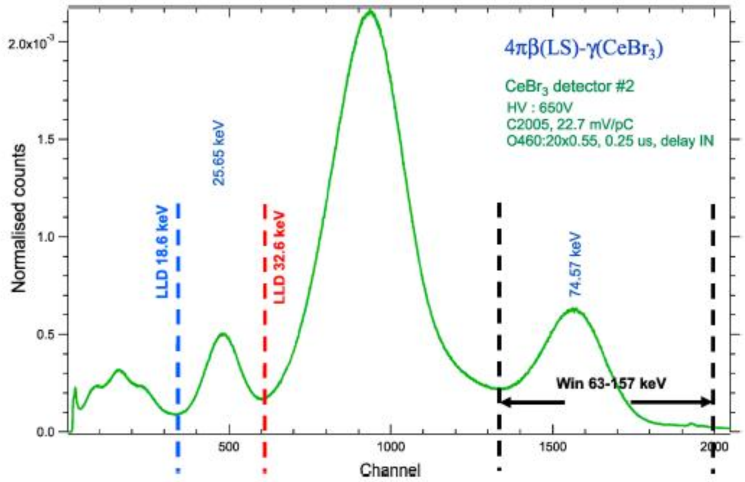
Primary standardization techniques

Primary techniques depending on the isotopes to be measured

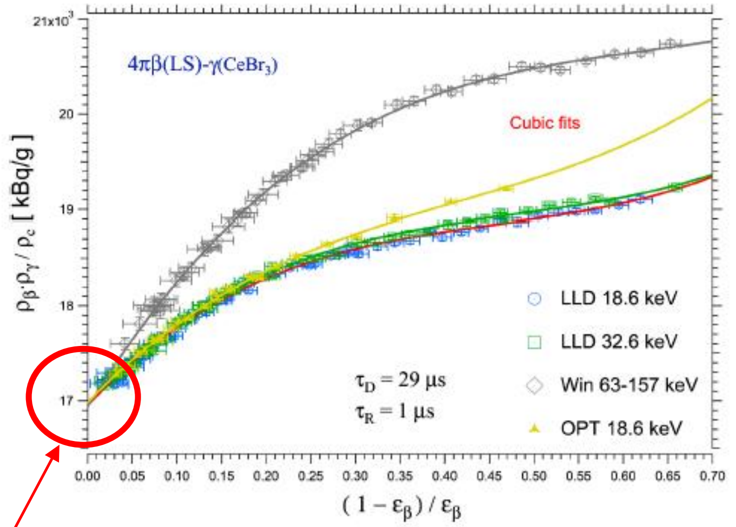
- 4π-γ counting
  - scintillation detector under a quasi-full solid angle
  - Relatively cheap and accurate method. But requires a detailed efficiency calculation
- Coincidence counting
  - Set-up consists of two detectors, β-channel and γ- channel, measuring the β-decay followed a γ-emission due to the de-excitation of the daughter nuclide.
  - An additional counting channel records the number of coincident events in both detectors.
  - The source activity can be directly derived from the count rates in the three channels.
- Liquid scintillation counting (TDCR or CIEMAT-NIST)
  - Useful for low energy beta, mixing the radioactive material with liquid scintillation cocktail
- Defined solid angle
  - Precise measurement for α emitters
- γ - γ coincidence, 4π or 2π proportional counter…

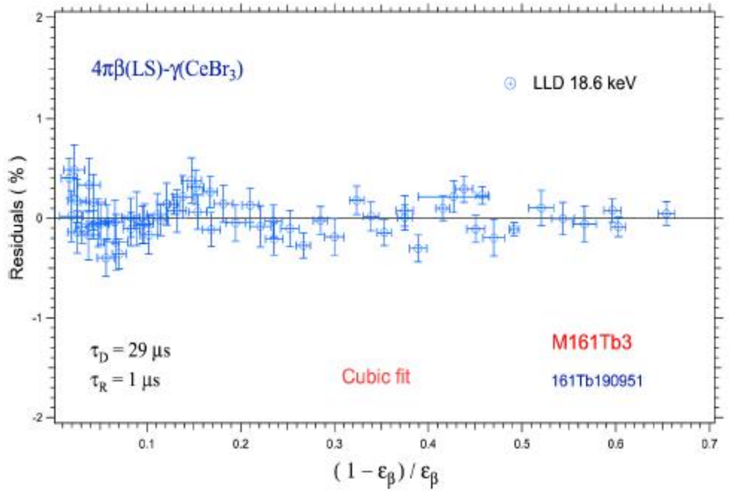
Tb-161 standardization by coincidence



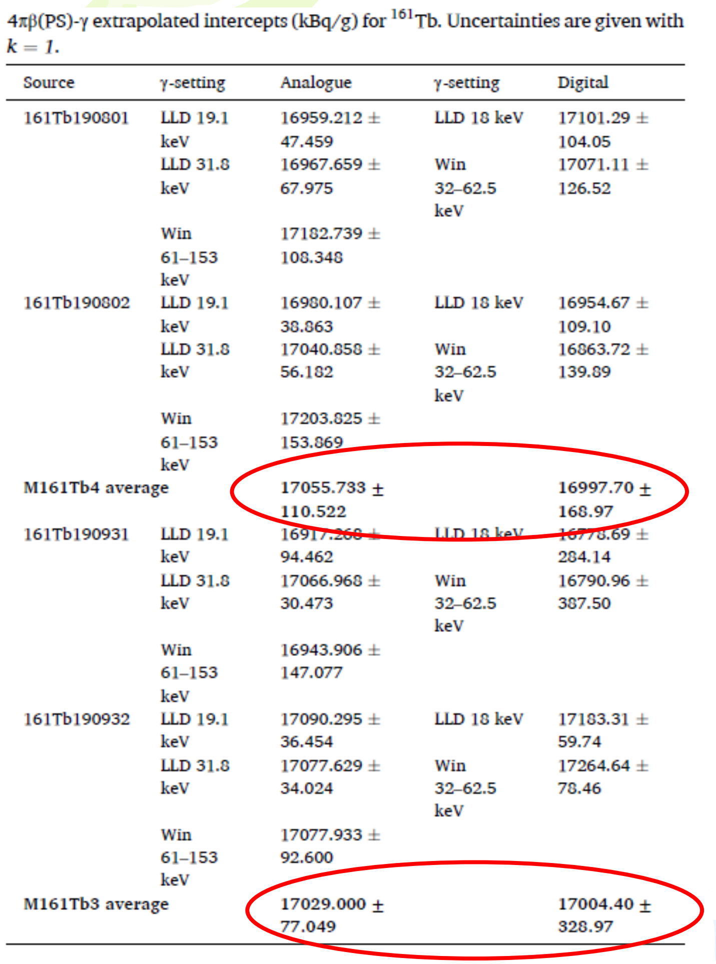


- The variation of the β efficiency channel is obtained by changing the low-level energy threshold or by optical filtering
- 3 gamma channels setting are used for the extrapolation curves
- Intercep gives the activity



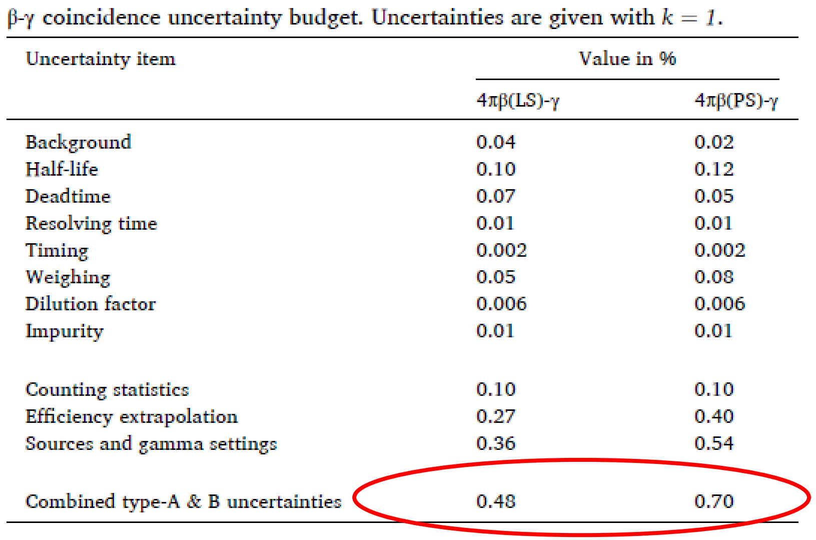


Tb-161 standardization



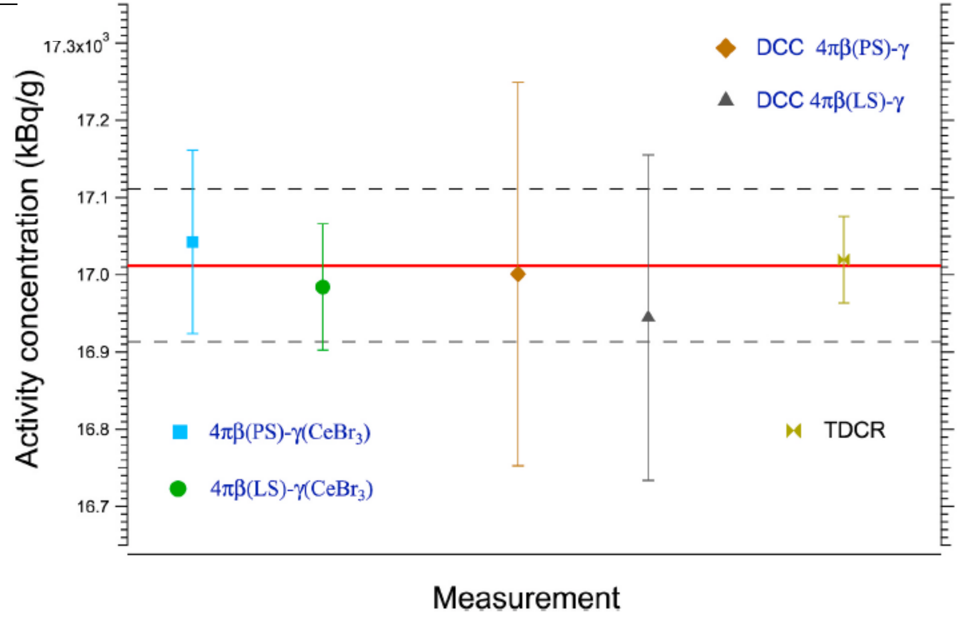
$4\pi\beta$(PS)-γ extrapolated intercepts (kBq/g) for ^{161}Tb. Uncertainties are given with *k = 1*.

Source	γ-setting	Analogue	γ-setting	Digital
161Tb190801	LLD 19.1 keV	16959.212 ± 47.459	LLD 18 keV	17101.29 ± 104.05
	LLD 31.8 keV	16967.659 ± 67.975	Win 32–62.5 keV	17071.11 ± 126.52
	Win 61–153 keV	17182.739 ± 108.348		
161Tb190802	LLD 19.1 keV	16980.107 ± 38.863	LLD 18 keV	16954.67 ± 109.10
	LLD 31.8 keV	17040.858 ± 56.182	Win 32–62.5 keV	16863.72 ± 139.89
	Win 61–153 keV	17203.825 ± 153.869		
M161Tb4 average		**17055.733 ± 110.522**		**16997.70 ± 168.97**
161Tb190931	LLD 19.1 keV	16917.268 ± 94.462	LLD 18 keV	16778.69 ± 284.14
	LLD 31.8 keV	17066.968 ± 30.473	Win 32–62.5 keV	16790.96 ± 387.50
	Win 61–153 keV	16943.906 ± 147.077		
161Tb190932	LLD 19.1 keV	17090.295 ± 36.454	LLD 18 keV	17183.31 ± 59.74
	LLD 31.8 keV	17077.629 ± 34.024	Win 32–62.5 keV	17264.64 ± 78.46
	Win 61–153 keV	17077.933 ± 92.600		
M161Tb3 average		**17029.000 ± 77.049**		**17004.40 ± 328.97**



β-γ coincidence uncertainty budget. Uncertainties are given with *k = 1*.

Uncertainty item	Value in %	
	4πβ(LS)-γ	4πβ(PS)-γ
Background	0.04	0.02
Half-life	0.10	0.12
Deadtime	0.07	0.05
Resolving time	0.01	0.01
Timing	0.002	0.002
Weighing	0.05	0.08
Dilution factor	0.006	0.006
Impurity	0.01	0.01
Counting statistics	0.10	0.10
Efficiency extrapolation	0.27	0.40
Sources and gamma settings	0.36	0.54
Combined type-A & B uncertainties	0.48	0.70

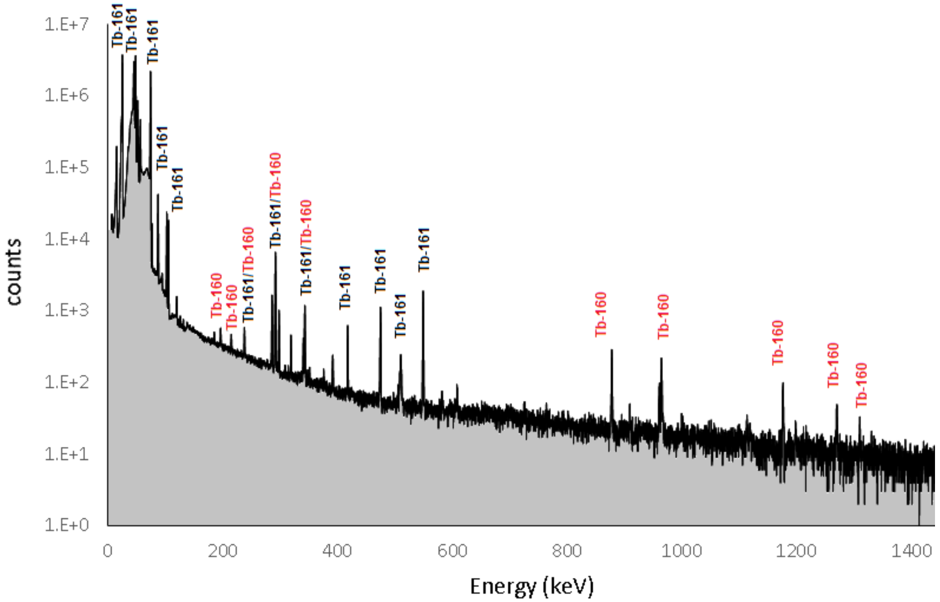


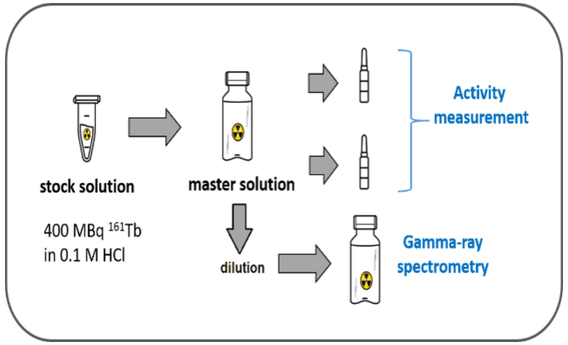
Tb-161 gamma emission

The γ-ray spectrometry was performed using a high purity germanium (HPGe) cylindrical detector. The detector is surrounded by a cylindrical shielding with wall thicknesses of 10 cm of lead and 2 mm of copper.

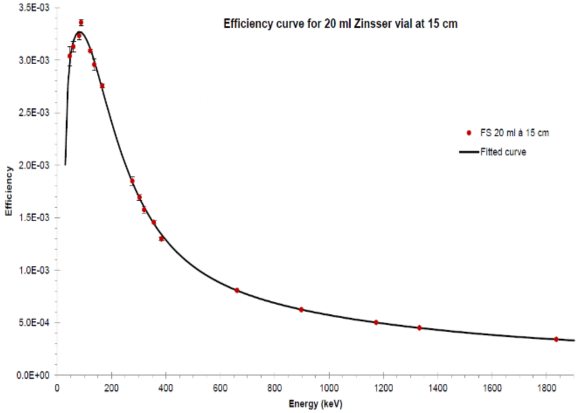
The measured activity concentration of the sample is given by standardization measurement.

$$P_\gamma = \frac{N}{t.A.\varepsilon_\gamma}.C_{dec.}.C_{meas.}C_{sum.}$$

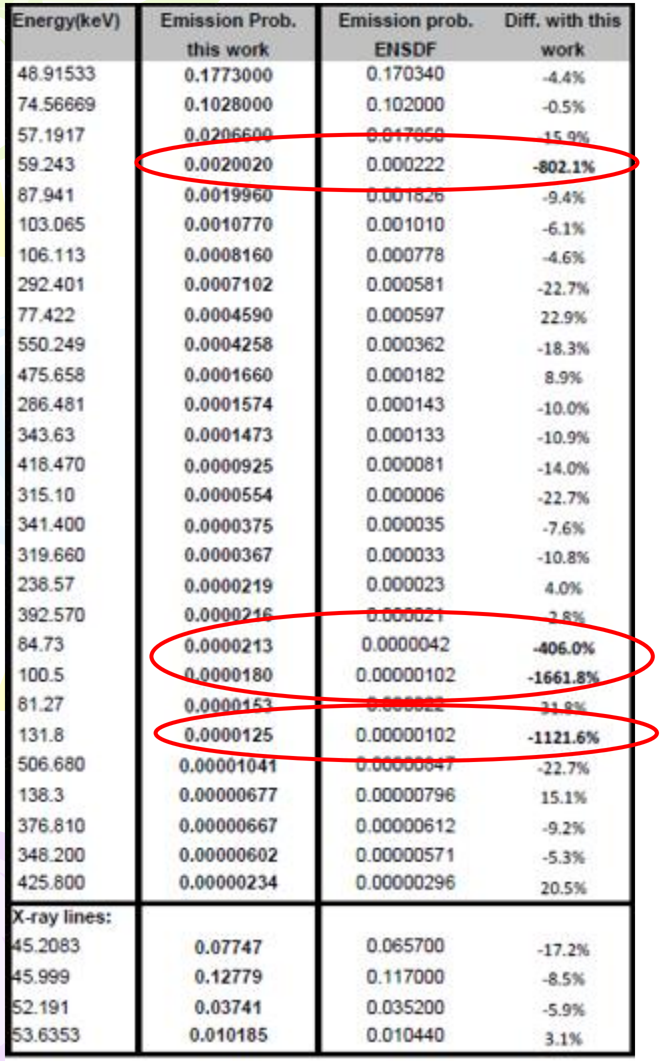




Known with primary measurement



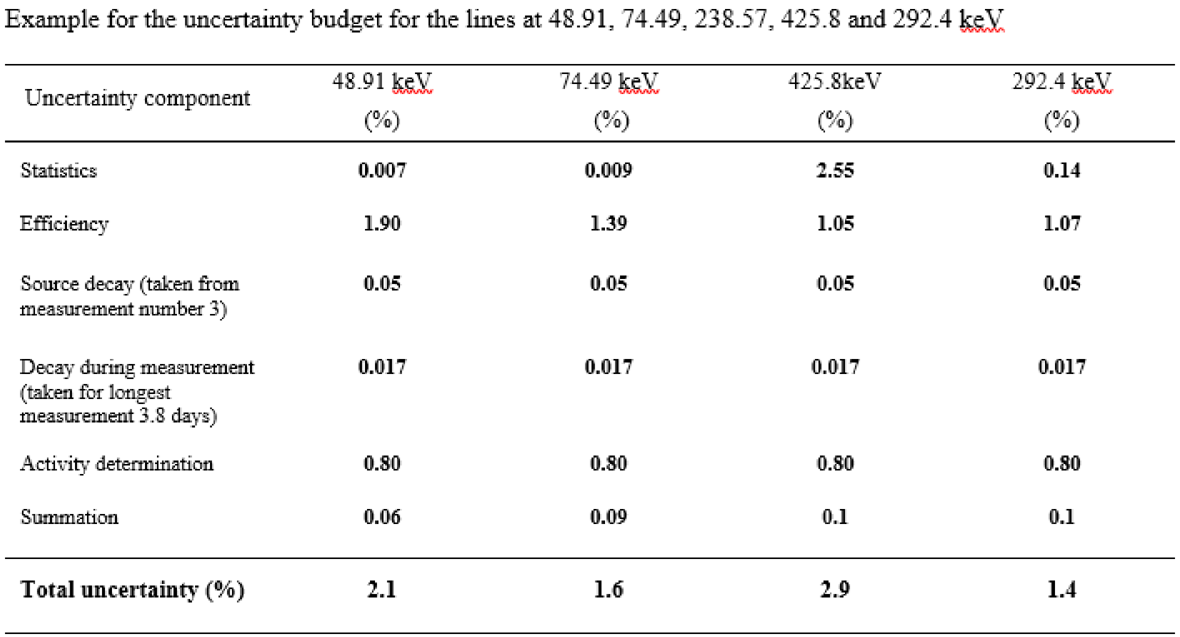
Tb-161 gamma emission



Energy(keV)	Emission Prob. this work	Emission prob. ENSDF	Diff. with this work
48.91533	0.1773000	0.170340	-4.4%
74.56669	0.1028000	0.102000	-0.5%
57.1917	0.0206600	0.017850	-15.9%
59.243	0.0020020	0.000222	**-802.1%**
87.941	0.0019960	0.001826	-9.4%
103.065	0.0010770	0.001010	-6.1%
106.113	0.0008160	0.000778	-4.6%
292.401	0.0007102	0.000581	-22.7%
77.422	0.0004590	0.000597	22.9%
550.249	0.0004258	0.000362	-18.3%
475.658	0.0001660	0.000182	8.9%
286.481	0.0001574	0.000143	-10.0%
343.63	0.0001473	0.000133	-10.9%
418.470	0.0000925	0.000081	-14.0%
315.10	0.0000554	0.000006	-22.7%
341.400	0.0000375	0.000035	-7.6%
319.660	0.0000367	0.000033	-10.8%
238.57	0.0000219	0.000023	4.0%
392.570	0.0000216	0.000021	-2.8%
84.73	0.0000213	0.0000042	**-406.0%**
100.5	0.0000180	0.00000102	**-1661.8%**
81.27	0.0000153	0.000022	31.8%
131.8	0.0000125	0.00000102	**-1121.6%**
506.680	0.00001041	0.00000847	-22.7%
138.3	0.00000677	0.00000796	15.1%
376.810	0.00000667	0.00000612	-9.2%
348.200	0.00000602	0.00000571	-5.3%
425.800	0.00000234	0.00000296	20.5%
X-ray lines:			
45.2083	0.07747	0.065700	-17.2%
45.999	0.12779	0.117000	-8.5%
52.191	0.03741	0.035200	-5.9%
53.6353	0.010185	0.010440	3.1%

Most of the measured intensities agree with previous measurements. But 4 are in total disagreement.

In addition, our work allows to reduce considerably the uncertainty value to around 2% instead of around 10%.

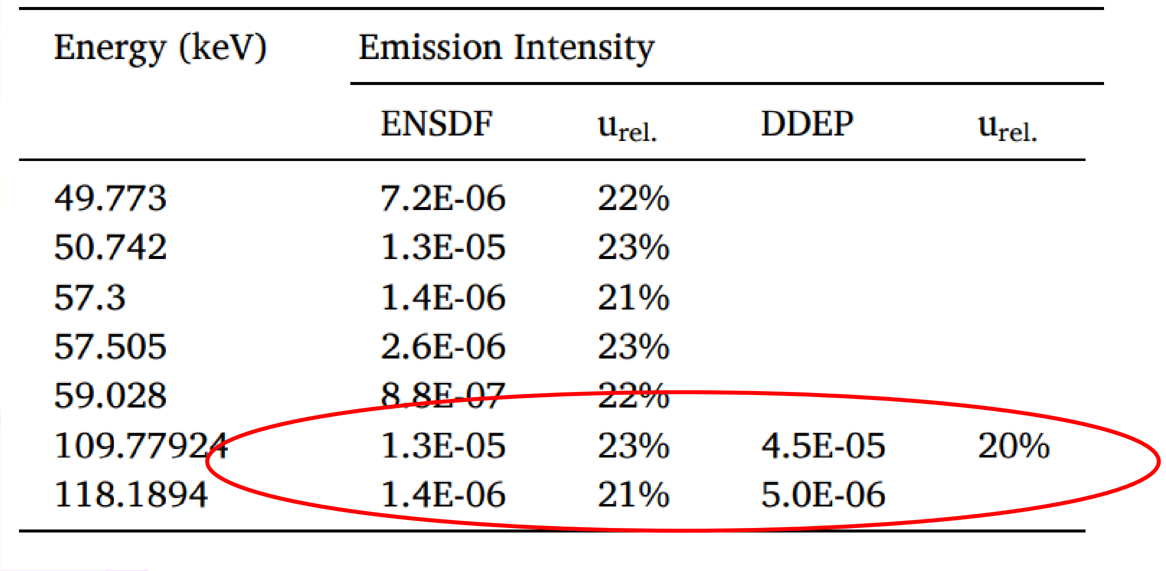


Example for the uncertainty budget for the lines at 48.91, 74.49, 238.57, 425.8 and 292.4 keV

Uncertainty component	48.91 keV (%)	74.49 keV (%)	425.8keV (%)	292.4 keV (%)
Statistics	**0.007**	**0.009**	**2.55**	**0.14**
Efficiency	**1.90**	**1.39**	**1.05**	**1.07**
Source decay (taken from measurement number 3)	**0.05**	**0.05**	**0.05**	**0.05**
Decay during measurement (taken for longest measurement 3.8 days)	**0.017**	**0.017**	**0.017**	**0.017**
Activity determination	**0.80**	**0.80**	**0.80**	**0.80**
Summation	**0.06**	**0.09**	**0.1**	**0.1**
Total uncertainty (%)	**2.1**	**1.6**	**2.9**	**1.4**

Er-169 gamma emission

- 2 gamma lines at 109 and 118 keV low intensities and large uncertainties (measurements before 70s!)
- But disagreement between different database by a large factor ~3.5

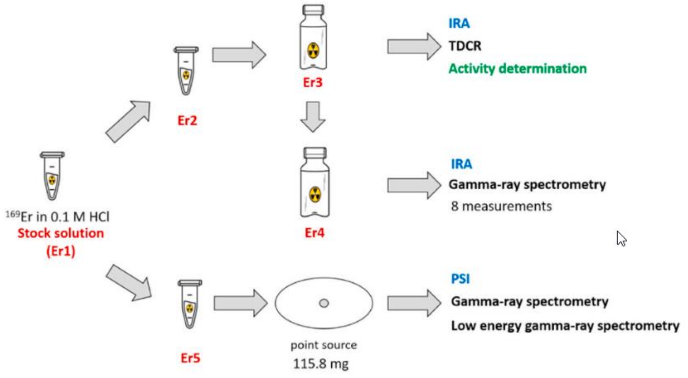


Energy (keV)	Emission Intensity			
	ENSDF	$u_{rel.}$	DDEP	$u_{rel.}$
49.773	7.2E-06	22%		
50.742	1.3E-05	23%		
57.3	1.4E-06	21%		
57.505	2.6E-06	23%		
59.028	8.8E-07	22%		
109.77924	1.3E-05	23%	4.5E-05	20%
118.1894	1.4E-06	21%	5.0E-06	

Clear need to have better data!

Er-169 gamma emission

- Gamma emission measured at IRA and PSI with HPGe detectors
- Activity samples measured using TDCR method giving an uncertainty of 0.4%



^{169}Er in 0.1 M HCl
Stock solution
(Er1)

Er2

Er3

IRA
TDCR
Activity determination

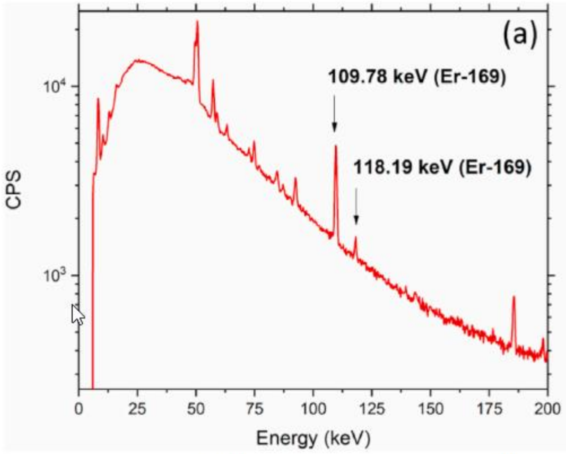
Er4

IRA
Gamma-ray spectrometry
8 measurements

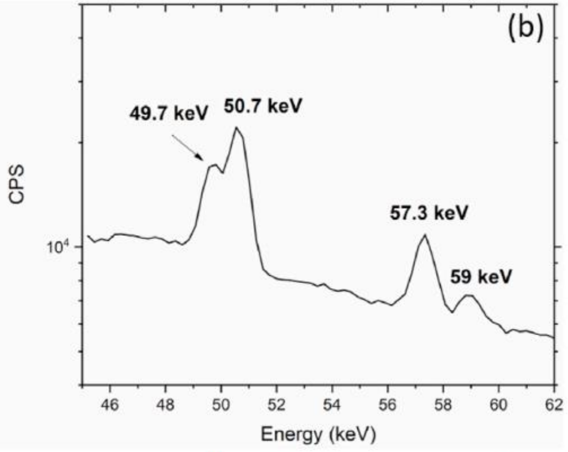
Er5

point source
115.8 mg

PSI
Gamma-ray spectrometry
Low energy gamma-ray spectrometry



Gamma spectrum of ^{169}Er measured over a period of 5.9 days



X-ray lines of ^{169}Er at 49.67, 50.64, 57.34, 58.98 keV

	Intensity 109 keV	Rel. uncertainty (%)	Intensity 118 keV	Rel. uncertainty (%)
IRA Gamma-ray spectrometry	1.4080E-05	1.04	1.515E-06	2.35
PSI Gamma-ray spectrometry	1.4430E-05	1.52	1.533E-06	6.00
ENSDF value	1.3(3)E-05	23	1.4(3)E-06	21
DDEP value	4.5(9)E-05	20	5.0E-06	–

- Results confirm the value from ENSDF database (reject DDEP)
- Large improvements on the uncertainties from 20% to around 1-2%.

Half-life measurement

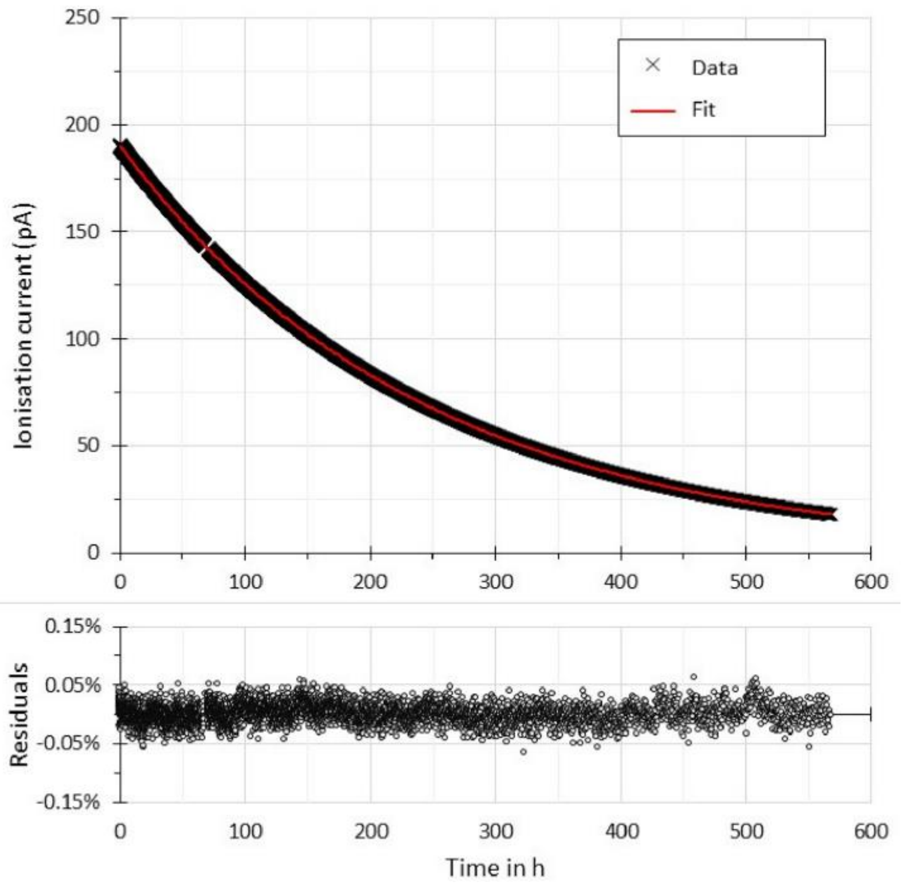
- For short half-lifes (several minutes to few months), the decay can be used by measuring the signal in a dedicated set-up (ionization chamber, gamma spectrometer…)

- Longer half-lifes, use the relation $A = \lambda . N$
  - $\lambda = \ln(2)/T_{1/2}$
  - A is measured with primary techniques
  - N (number of atoms) is measured using mass spectrometry

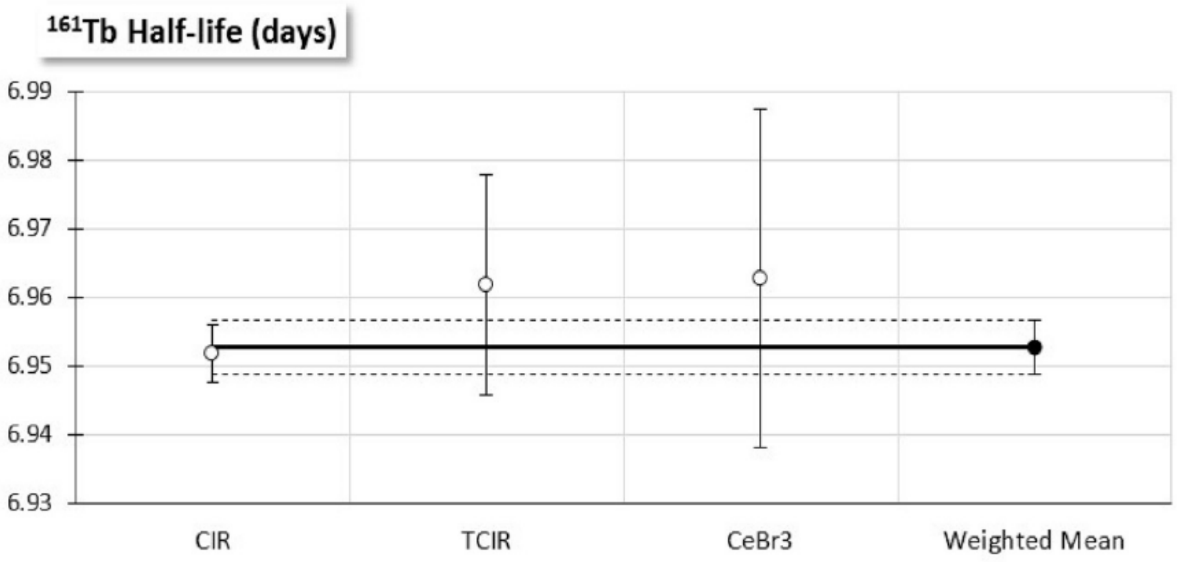
Tb-161 half-life

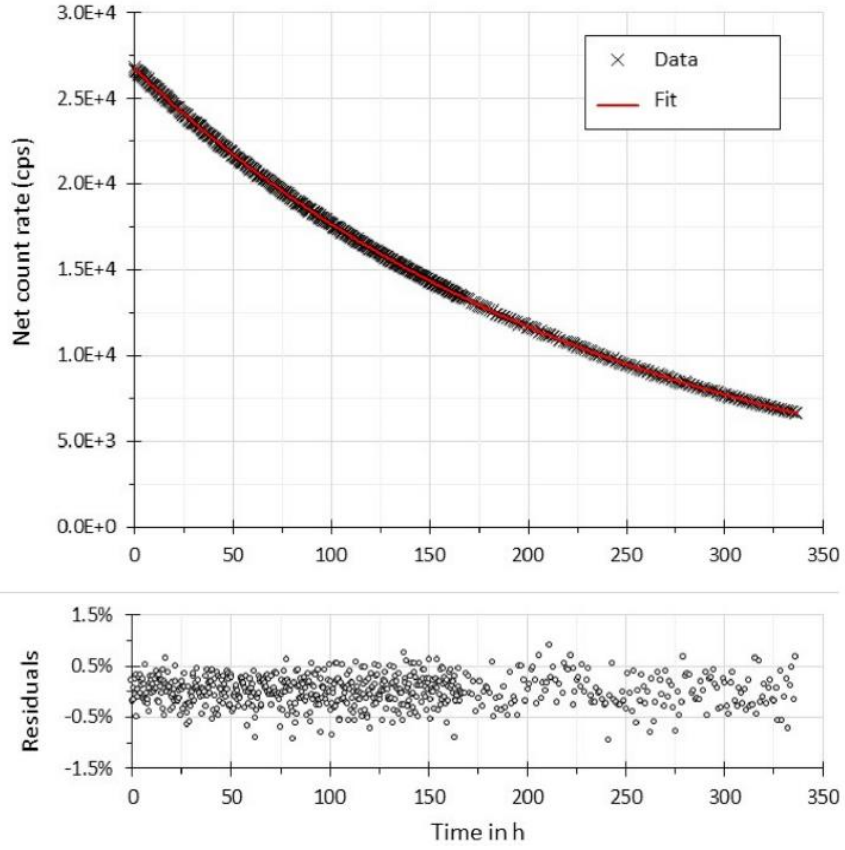
- As the half-life is small (6.9 days) it can be measured with a fit on the decay curve

- 3 independent systems are used
  - CIR (reference ionization chamber)
    - Current is precisely measured (<0.05%)
    - 23 days of measurements (6564 points)
  - TCIR (transportable ionization chamber)
    - 16 days of measurement, 2579 points
  - γ spectrum measurement using a $4\pi\gamma$ counting system.
    - Events counting above a threshold
    - 14 days of measurements, 672 points



Tb-161 half-life





<u>Final value :</u> 6.953(2) days
More precise values than previous measurement
Uncertainty is 0.03% (10 times less)

This method was also used to determine the half-life of Yb-175, Lu-177, Tc-99m, Cu-61…

Ho-166m half-life measurement

- Half-life ~1200 years with 15% uncertainty! (measured in 1963)
- New measurement using the number of atoms counting method, A = λ.N
  - Activity was measured using $4\pi\beta-\gamma$ coincidence technique and ionisation chamber, uncertainty 0.24%
  - Number of atoms using multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS), uncertainty 0.25%

- Half-life value is: **1132.6(39) years, uncertainty 0.34%!**
  - 5.6% shorter than database value and a reduction of the uncertainty from 15% to 0.34%

Lanthanides measured at IRA

Isotopes	$T_{1/2}$	rel. unc. (%)	Meas. technique
Ho-166m	1132.6(39) years	0.34	Atoms number
Tb-161	6.953(2) days	0.03	Decay
Yb-175	4.1615(30) days	0.07	Decay
Lu-177	6.6429(70) days	0.11	Decay
Tb-149	4.1615 h (preliminary)		Decay
Tm-167, Tm-168	Under progress		Decay

Additional isotopes are also studied (Ac-225, Cu-61, Ba-128…)

Gamma intensities	Tb-161, Er-169	Tm-167 under progress

Additional isotopes are also studied (Ra-223, Ac-225…)

Beta spectrum	Shape, E_{max} : Tm-171	Previously: Cl-36 Under progress: Y-90 (positron)

Summary

- As metrology institute, IRA is maintaining the unit of the activity, the Bq, for Switzerland.
  - Use precise primary techniques to measure activity for many isotopes. Measurements are validated by international comparison through the SIR

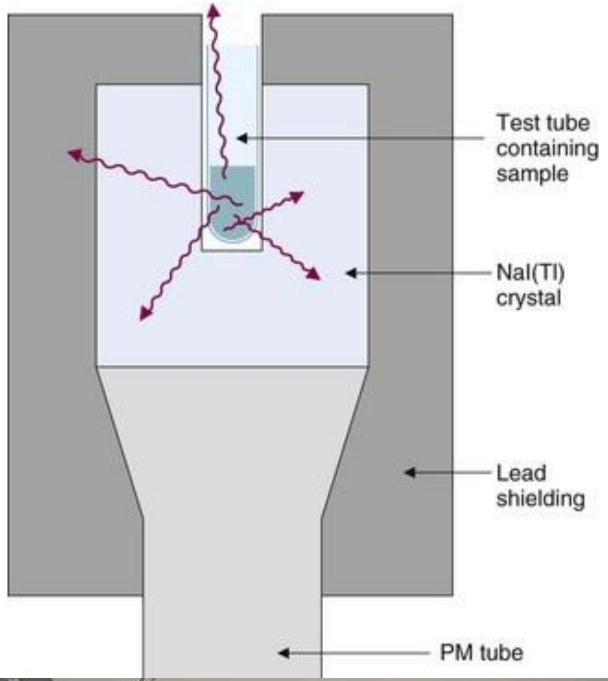
- The measurement techniques are also used to measure Nuclear Data ($T_{1/2}$, gamma intensities, beta spectrum)
  - Lanthanides: Tb-161, Yb-175, Tb-149, Tm-167…

- Improve the data precision and reduce the uncertainties

- We are interested and ready to measure new isotopes

Primary activity measurements with 4πγNaI(Tl) counting

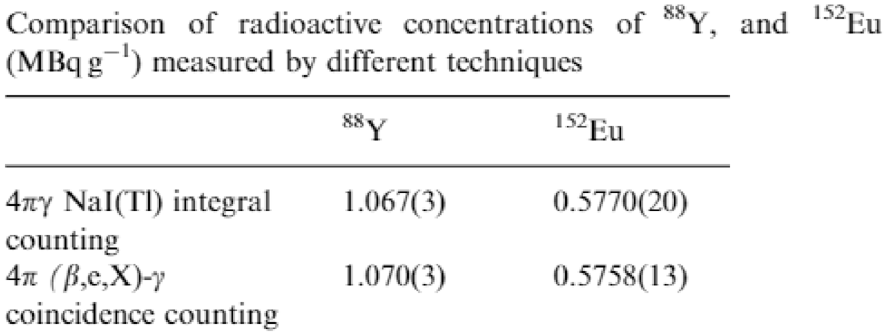
- NaI(Tl) scintillation detector under a quasi-full solid angle (99.1% of 4π for IRA set-up)
- Its appeal lies in the rather simple electronic circuit to process and count the pulses above some threshold and its equal suitability for point, surface and volume sources.
- Well-type detector with a 5”x5” NaI(Tl) crystal.
  - Sensitive volume of 1560 cm^3, measurement chamber 43.5 cm^3.
  - The crystal and photomultiplier are housed at the bottom of a 50 cm diameter and 5 cm thick cylindrical shielding covered with a sliding square 6 cm thick armoured plate



Primary activity measurements with 4πγNaI(Tl) counting

- Principle:
  - Count the number of events above a threshold
  - Activity concentration $C_A = \frac{(Rate-Background)}{\varepsilon_{tot}}\frac{1}{m}$

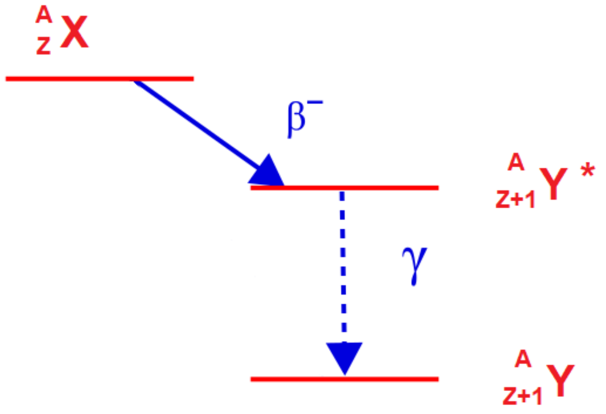
    where *m* is the masse of the measured sample and ε_{tot} the total efficiency
  - Integral counting from zero energy is unreliable (poor resolution, electronic noise, Bremsstrahlung radiations…), counting threshold depends on the nuclide, but it is usually set at 22.6 keV (K X-ray peak of ^{109}Cd)
  - Use Monte Carlo simulations for efficiency calculation which can produce the detection efficiency for any given discrimination threshold.
  - Different sources geometry can be used (point-like, ampoules, vials…)
- **4$\pi\gamma$ counting is a relatively cheap and accurate method. <u>However, it requires a detailed calculation of efficiency</u>**



Comparison of radioactive concentrations of ^{88}Y, and ^{152}Eu (MBq g^{-1}) measured by different techniques

	^{88}Y	^{152}Eu
4πγ NaI(Tl) integral counting	1.067(3)	0.5770(20)
4π (β,e,X)-γ coincidence counting	1.070(3)	0.5758(13)

$4\pi\beta-\gamma$ coincidence counting principle

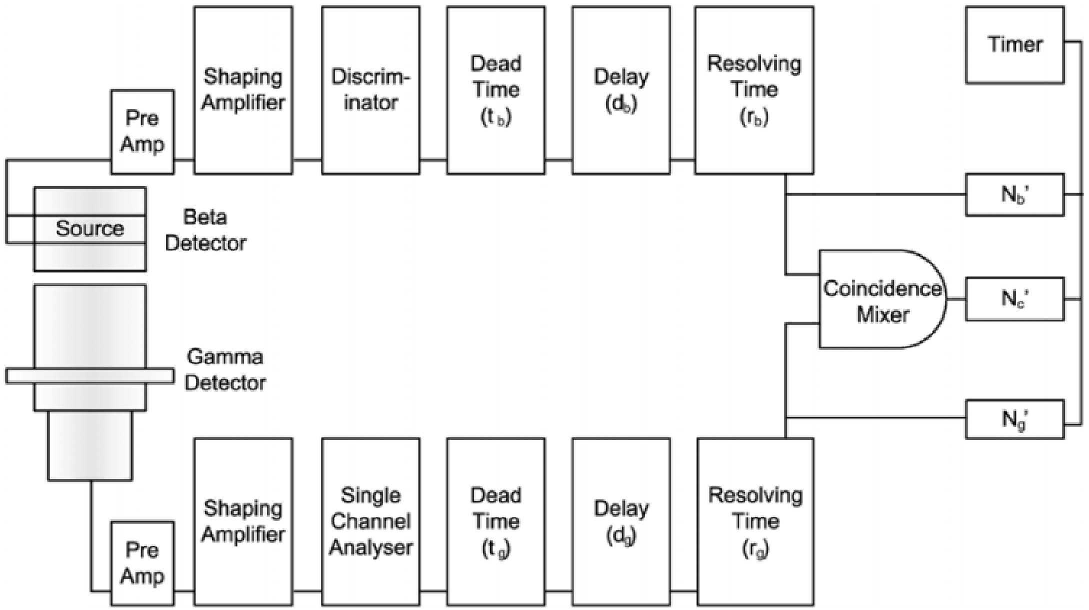


$$N_\beta = N_0\,\varepsilon_\beta$$

$$N_\gamma = N_0\,\varepsilon_\gamma$$

$$N_c = N_0\,\varepsilon_\beta\,\varepsilon_\gamma$$

A typical traditional coincidence counting system.



Source activity $A = N_0 = \dfrac{N_\beta\,N_\gamma}{N_c}$

$4\pi\beta-\gamma$ coincidence counting principle

Even for a simple decay-scheme radionuclide, the $4\pi\beta$ counter is generally affected by additional counts due to its sensitivity to the γ –transition.
The counting rates have to be modified to take into account the interaction of γ -photons and conversion electrons.

$$N_\beta = A \cdot \left[\varepsilon_\beta + (1-\varepsilon_\beta) \left\{ \frac{1}{1+\alpha_T} (\alpha_T \cdot \varepsilon_{ce} + \varepsilon_{\beta\gamma}) \right\} \right]$$

$$N_\gamma = A \cdot \varepsilon_\gamma / (1+\alpha_T)$$

$$N_c = A \cdot \left[\frac{\varepsilon_\beta \cdot \varepsilon_\gamma}{1+\alpha_T} + (1-\varepsilon_\beta) \cdot \varepsilon_c \right]$$

α_T is the total internal conversion coefficient

ε_{ce} is the detection efficiency for conversion electrons of the β-detector

$\varepsilon_{\beta\gamma}$ is the detection efficiency for γ-photons in the β-detector

ε_c represents the probability of observing additional coincidences

$4\pi\beta\!-\!\gamma$ coincidence counting principle

In case of complex decay-scheme with n different β branches with intensities a_r, the count rates can be expressed as:

$$N_\beta = A \sum_{r=1}^{n} a_r \left(\varepsilon_{\beta_r} + (1 - \varepsilon_{\beta_r})(\frac{\alpha\varepsilon_{ce} + \varepsilon_{\beta\gamma}}{1+\alpha})_r \right)$$

$$N_\gamma = A \sum_{r=1}^{n} a_r \varepsilon_{\gamma_r}$$

$$N_c = A \sum_{r=1}^{n} a_r \left(\varepsilon_{\beta_r} \varepsilon_{\gamma_r} + (1 - \varepsilon_{\beta_r}) \varepsilon_{c_r} \right)$$

The activity cannot be determined directly from the counting rates as they are depending on the emissions probabilities.

4πβ–γ coincidence counting principle

One can demonstrate that :

$$N_\beta = N_0\left[(1-K) + K\frac{N_c}{N_\gamma}\right] \longrightarrow N_0 \quad \text{when} \quad \frac{N_c}{N_\gamma} \longrightarrow 1$$

where $K = \sum_{r=1}^{n} \frac{a_r C_r}{k}\left(1 - \frac{\alpha\varepsilon_{ce}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_r$ is a constant

When $\varepsilon_\gamma, \varepsilon_c, \varepsilon_{ce}, \varepsilon_{\beta\gamma}$ remain fixed

K and **N_0 can be determined graphically varying $\frac{N_c}{N_\gamma}$ without precise knowledge of the nuclear data (decay branching ratio, emission probabilities, conversion coefficients…)**

$4\pi\beta-\gamma$ coincidence counting principle

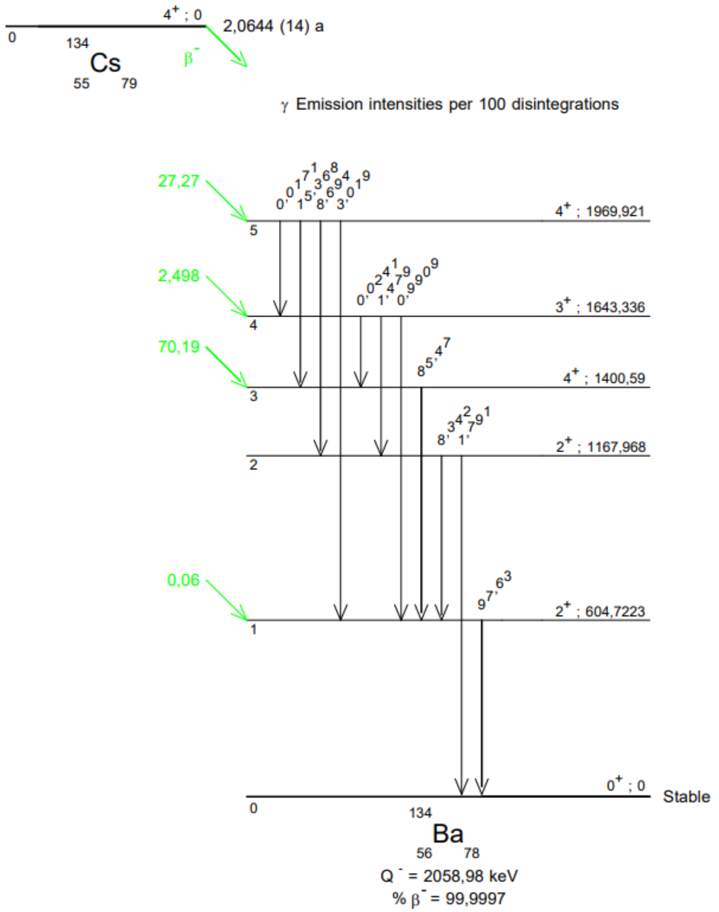
- In practice we use:

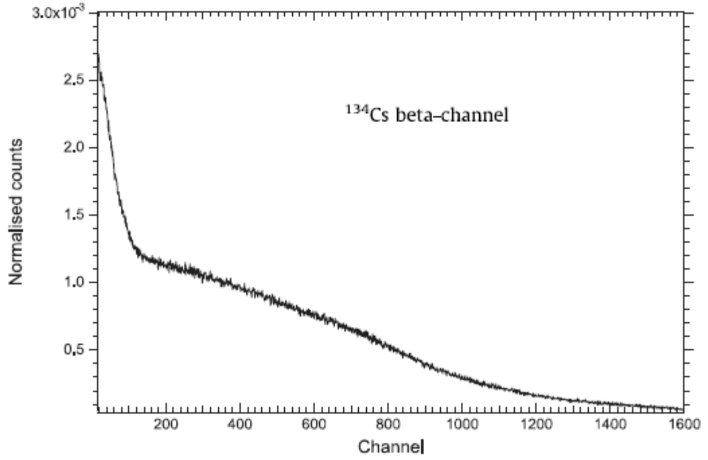
$$N_\beta = N_0[(1-K) + K\frac{N_c}{N_\gamma}] \quad \text{or} \quad \frac{N_\beta N_\gamma}{N_c} = N_0[1 + (1-K)(\frac{1-{}^{N_c}/_{N_\gamma}}{{}^{N_c}/_{N_\gamma}})$$

A plot of $\frac{N_\beta N_\gamma}{N_c}$ vs $\frac{N_c}{N_\gamma}$ or $\frac{1-{}^{N_c}/_{N_\gamma}}{{}^{N_c}/_{N_\gamma}}$ yields a straight line with slope $(1-K)N_0$ and intercept N_0 for $\frac{N_c}{N_\gamma} = 1$

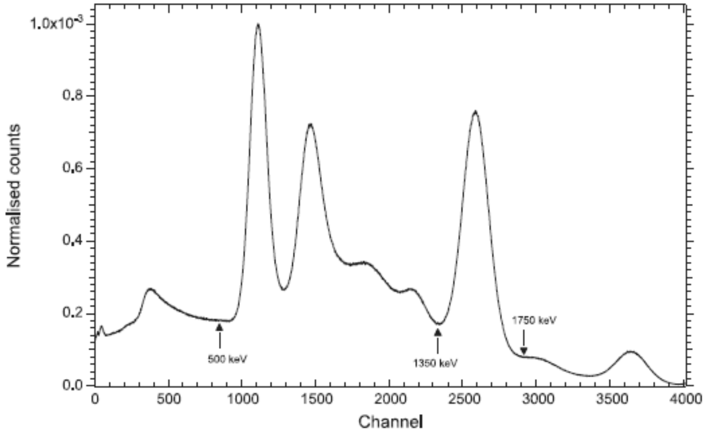
In practice we keep $\varepsilon_\gamma, \varepsilon_c$ fixed and we modified ε_β either by adding plastic films with different densities in front of the detector, by changing the detector energy threshold level or changing the geometry (distance between source-detector)

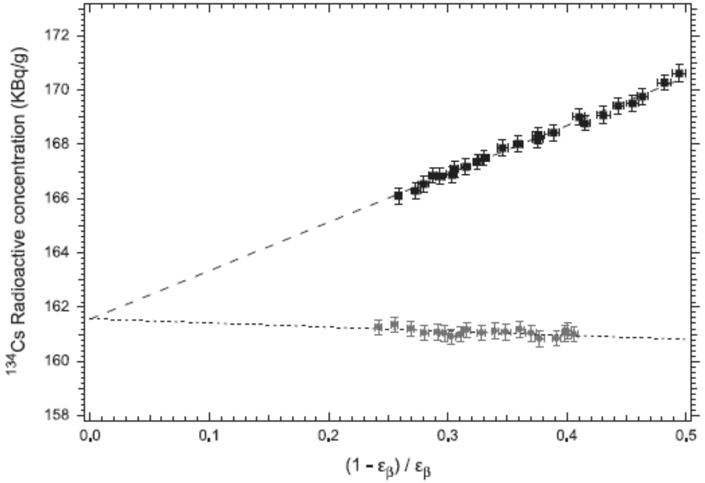
Example of Cs-134



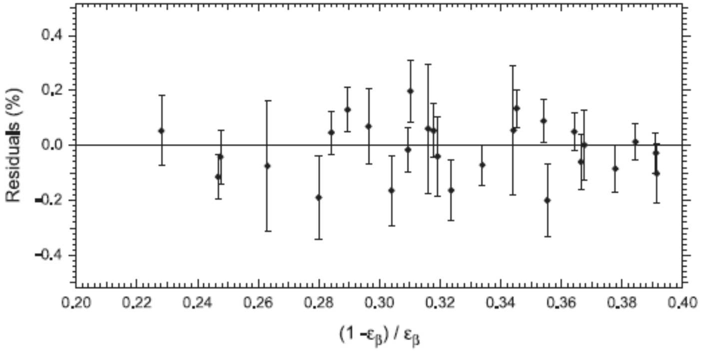


Well-type NaI(Tl) gamma energy spectrum for ^{134}Cs.





Efficiency extrapolation for the 1350–1750 keV window (■) and the 500– 1800 keV window (●) for ^{134}Cs.



^{134}Cs relative residuals of the least-square fit of the linear efficiency function for the 500–1800 keV window in the gamma channel.