Experimental efficiency calibration for a HPGe gamma-ray detector

NKS – GammaSpec 2017 Seminar

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Environmental Radiation Surveillance and Emergency Preparedness

Outline

1. Introduction (STUK's γ-ray lab)
2. The problem … and how to overcome it
3. Peak efficiency
4. Total efficiency
5. Methodology of the uncertainty determination
6. Final result, comparison between uncertainties and conclusions

1. Introduction (STUK's γ-ray lab)

- 16 HPGe spectrometers (7 Ortec, 9 Canberra).
- 4 electrically cooled, others are LN2-cooled (5 Möbius, 7 Cryo-Cycle).
- Digital MCAs (different DSPEC generations).
- 3000 – 4000 analyses per year.

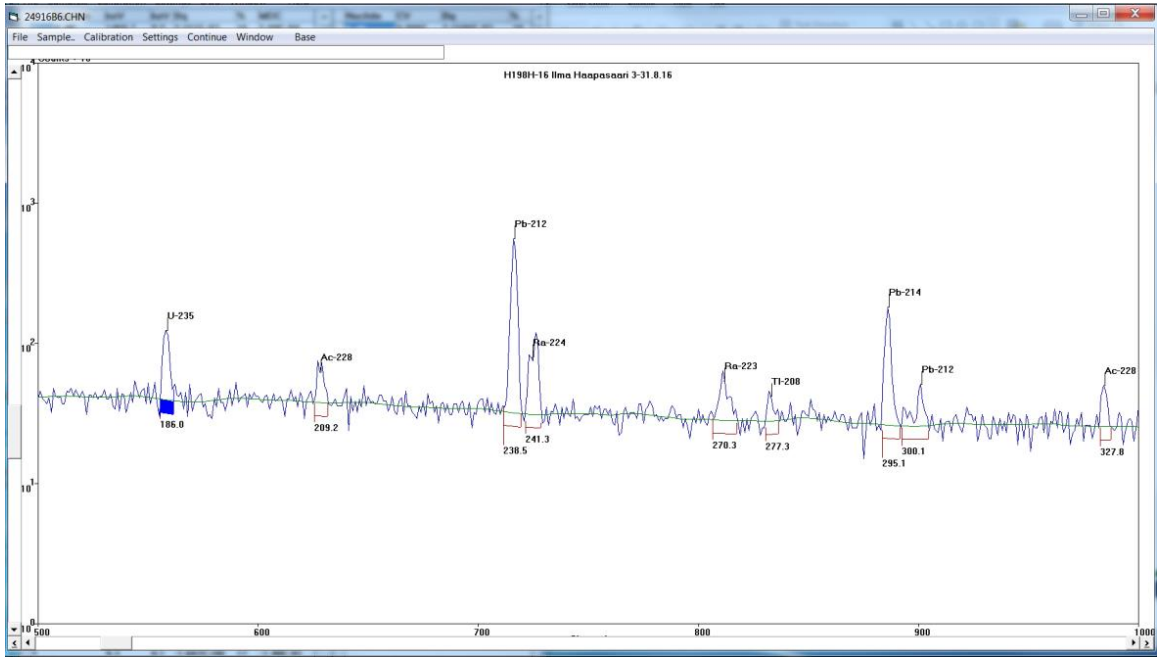
FINAS
Finnish Accreditation Service
T167 (EN ISO/IEC 17025)


γ1

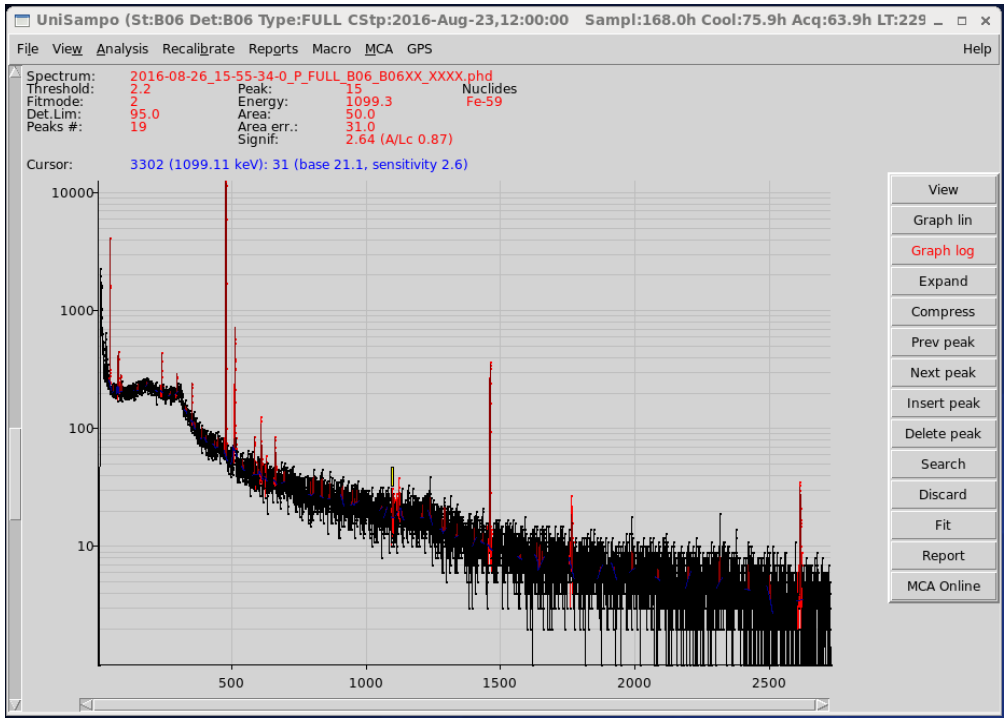
γ2

Spectrum analysis

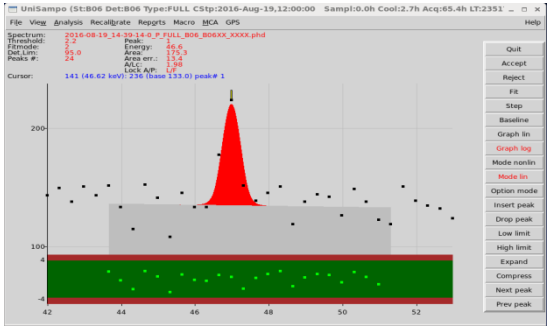
- GAMMA-99 software developed by STUK
  - cascade summing correction since 1983 !
  - sample height and density corrections
  - automatic and interactive operation modes



- UniSampo/Shaman
  - UniSampo: peak search, baseline & peak fitting, peak areas
  - Shaman: rule-based peak identification and activity calculation, mimics human analyst



Interactive fitting



Measurement geometries

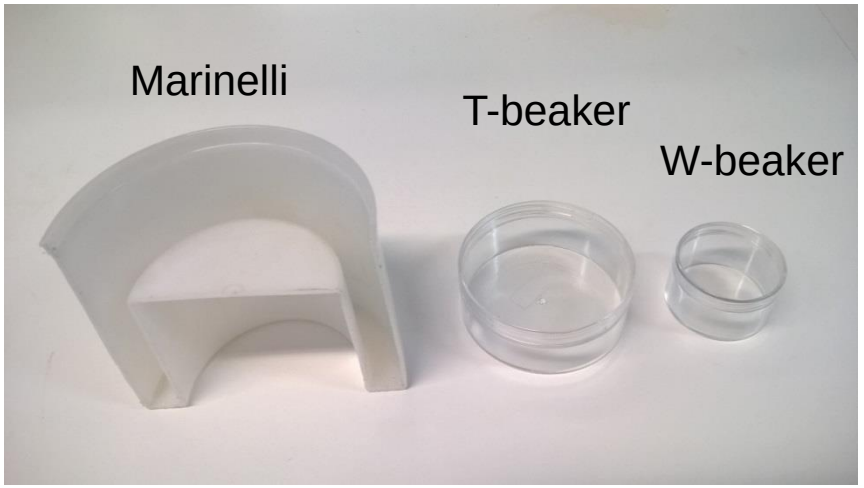
Simple cylindrical:

- 0-30 mL, free sample height
- 0-100 mL, free sample height

Marinelli:

- 0.5 L, fixed sample height

Some special geometries, too



Calibrations & the NAMIT software

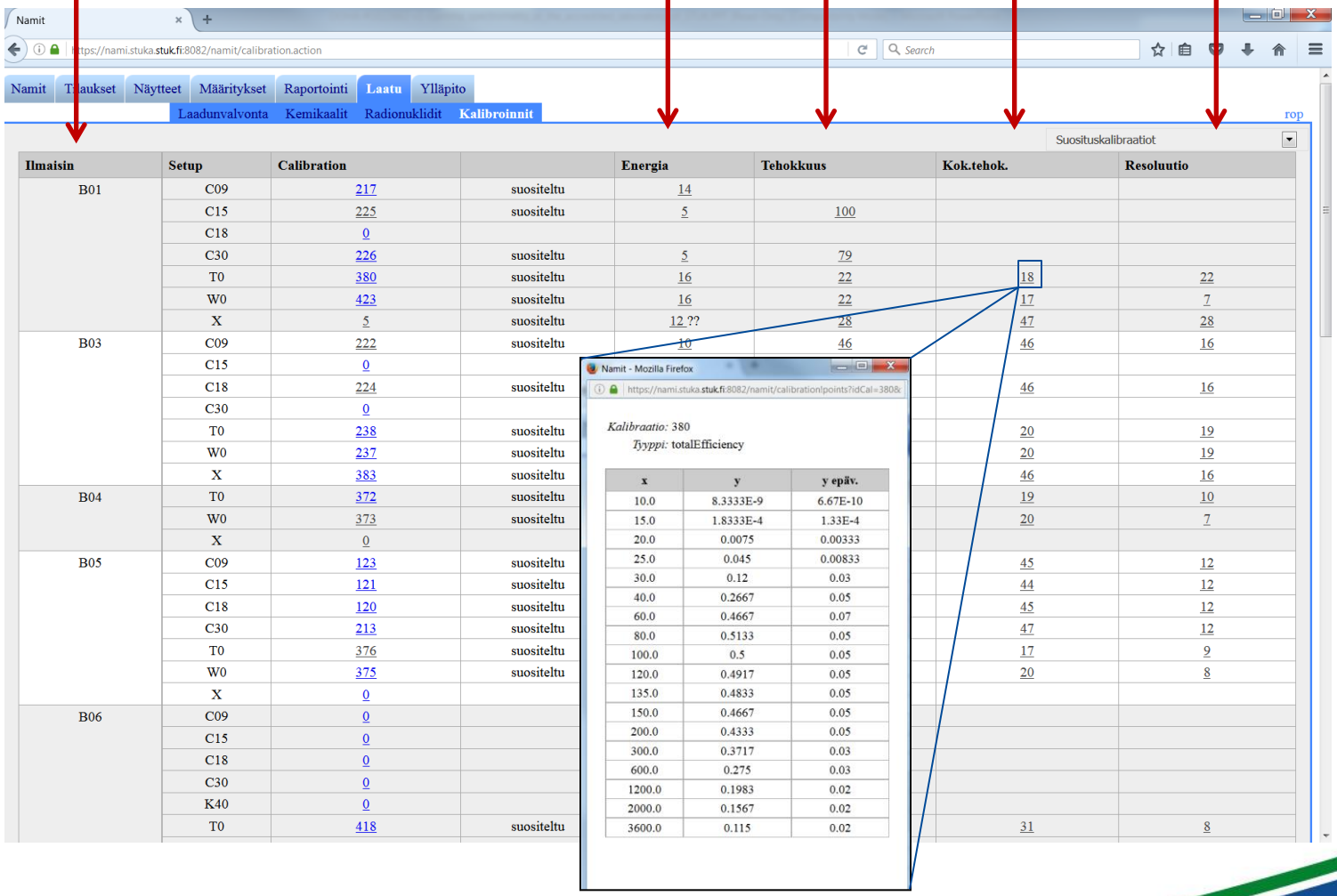
Detectors (here 5 different)

Energy

Peak Eff.

Tot. Eff.

Resolution

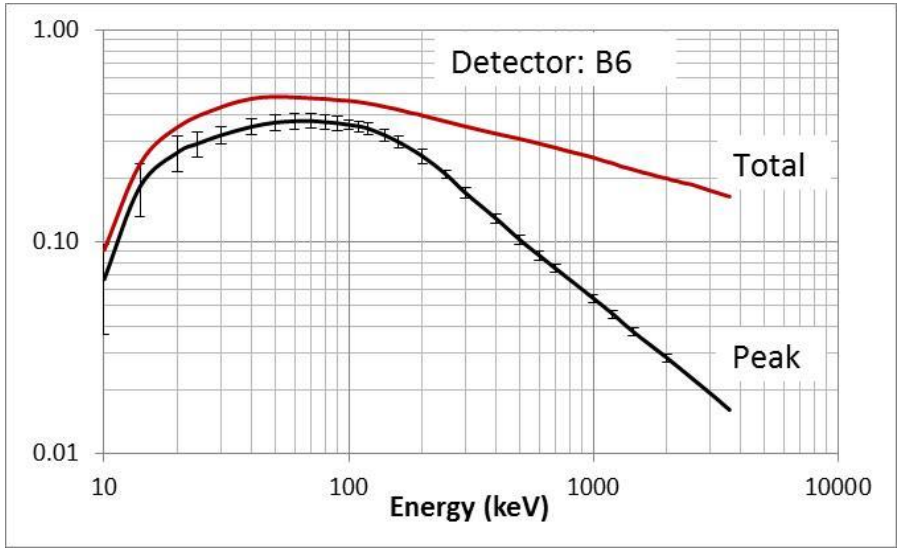


Ilmaisin	Setup	Calibration		Energia	Tehokkuus	Kok.tehok.	Resoluutio
B01	C09	217	suositeltu	14			
	C15	225	suositeltu	5	100		
	C18	0					
	C30	226	suositeltu	5	79		
	T0	380	suositeltu	16	22	18	22
	W0	423	suositeltu	16	22	17	7
	X	5	suositeltu	12 ??	28	47	28
B03	C09	222	suositeltu	10	46	46	16
	C15	0					
	C18	224	suositeltu			46	16
	C30	0					
	T0	238	suositeltu			20	19
	W0	237	suositeltu			20	19
	X	383	suositeltu			46	16
B04	T0	372	suositeltu			19	10
	W0	373	suositeltu			20	7
	X	0					
B05	C09	123	suositeltu			45	12
	C15	121	suositeltu			44	12
	C18	120	suositeltu			45	12
	C30	213	suositeltu			47	12
	T0	376	suositeltu			17	9
	W0	375	suositeltu			20	8
	X	0					
B06	C09	0					
	C15	0					
	C18	0					
	C30	0					
	K40	0					
	T0	418	suositeltu			31	8

x	y	y epäv.
10.0	8.3333E-9	6.67E-10
15.0	1.8333E-4	1.33E-4
20.0	0.0075	0.00333
25.0	0.045	0.00833
30.0	0.12	0.03
40.0	0.2667	0.05
60.0	0.4667	0.07
80.0	0.5133	0.05
100.0	0.5	0.05
120.0	0.4917	0.05
135.0	0.4833	0.05
150.0	0.4667	0.05
200.0	0.4333	0.05
300.0	0.3717	0.03
600.0	0.275	0.03
1200.0	0.1983	0.02
2000.0	0.1567	0.02
3600.0	0.115	0.02

2. The problem …

- Present efficiency calibrations are not adequately traceable.
- In general, the calibrations are ok, but some deficiencies have been detected (for example: in some cases there have been shortcomings in small energies).
- Unclear and non-traceable uncertainty calculation.



Different measurement geometries and different detector characteristics

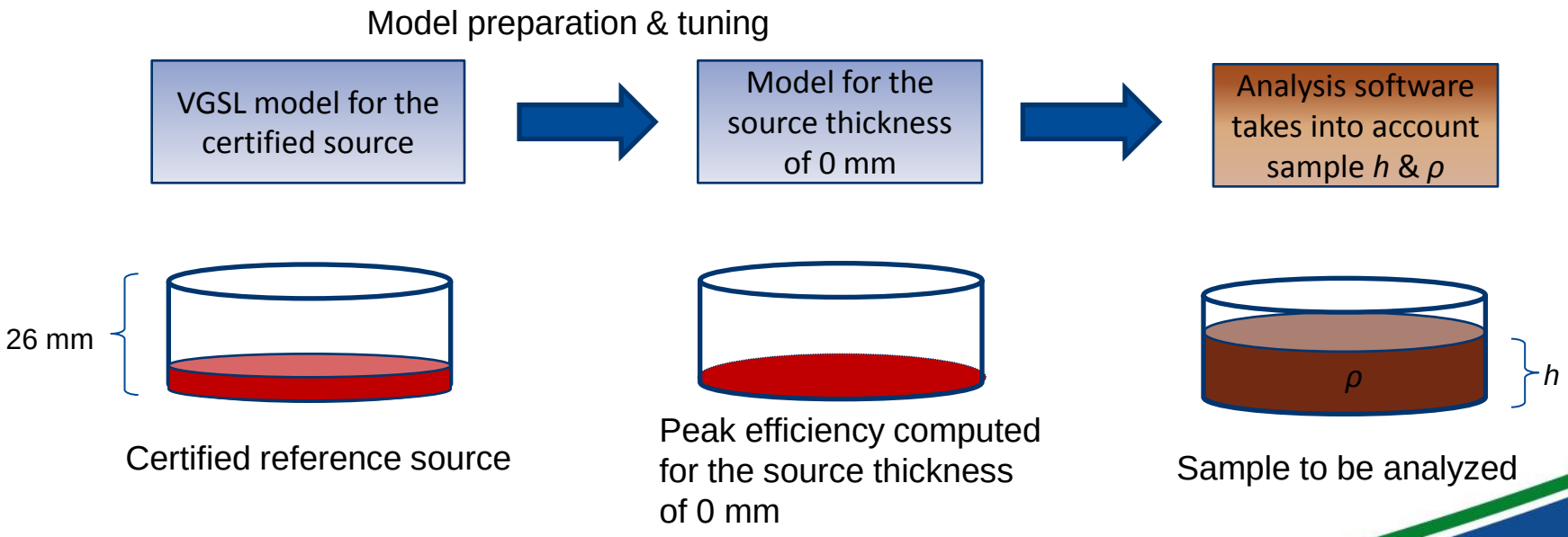
- Typically 2–5 measurement geometries/detector
- 16 detectors
- 1 FEP efficiency curve + 1 Tot. efficiency curve per geometry

➡ Total number of the efficiency curves: ∼3 × 16 × 2 ≈ **100 !**

➡ The ways of performing efficiency determination properly must be designed carefully.

- Different type of samples must be measured:
  - Air filters. Not a very complex case (from the efficiency calibration point of view).
  - General environmental samples. Measurements done using predetermined geometries. Challenge: various sample mass, density and elemental composition.
  - Other samples. Other than ”normal” geometries possible. Efficiency calculated by computational means (DECCA, EFFTRAN, VGSL, GESPECOR,…).

- Existing calibrations have previously been done as follows:
  - Certified cylindrical source of X mm in thickness (typically X = 5 mm) was measured first.
  - Using an appropriate detector model the calibrations were computed by VGSL for the source thickness of 0 mm.
  - Real sample density and height are corrected by the spectrum analysis software.



... and how to overcome it

- Certified sources of thickness 0 mm were purchased → no need to recalculate/correct the efficiencies back to the thickness of 0 mm.
- Minimizing the use of MC-methods or other similar type of computations (but their use cannot be avoided).
- ∼ 100 efficiency curves → an individual specialist cannot process all these → the pain of making efficiency calibrations must be shared between the analysts → advantage: rising the expertise.
- Special attention to the uncertainty calculation.

3. Peak efficiency

Peak efficiency, ε_i , for the gammas of energy i

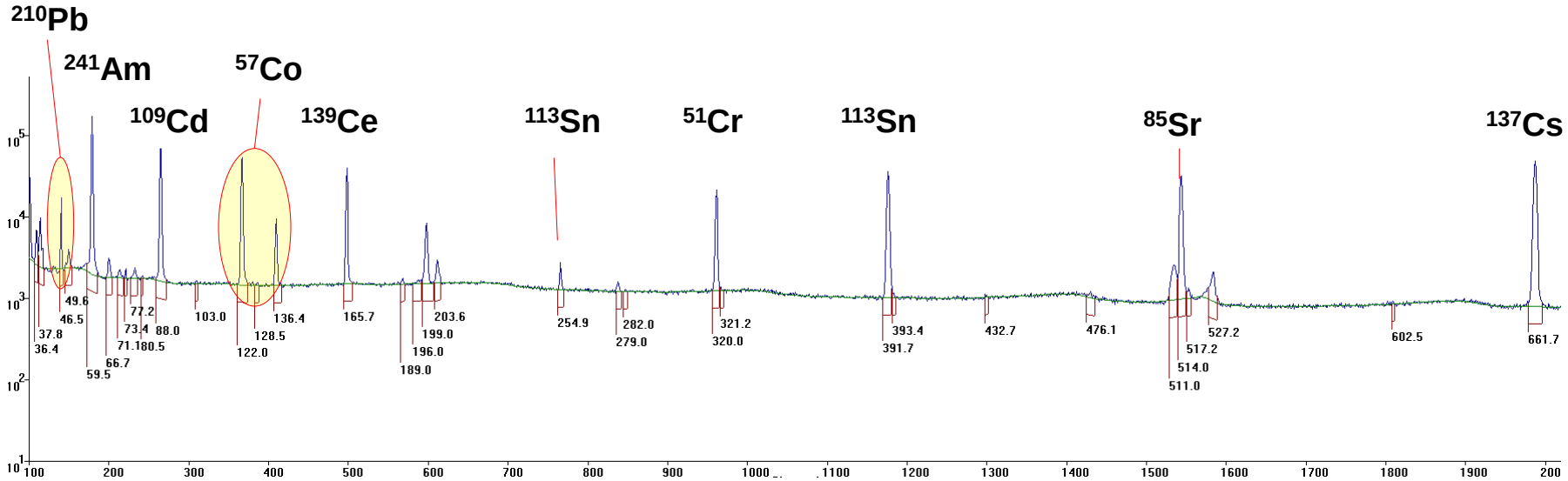
$$\varepsilon_i = \frac{\textit{Number of detected gammas per unit time}}{\textit{Number of emitted gammas per unit time}}$$

$$\varepsilon_i = \frac{n_i \cdot C_i}{A_i \cdot I_\gamma}$$

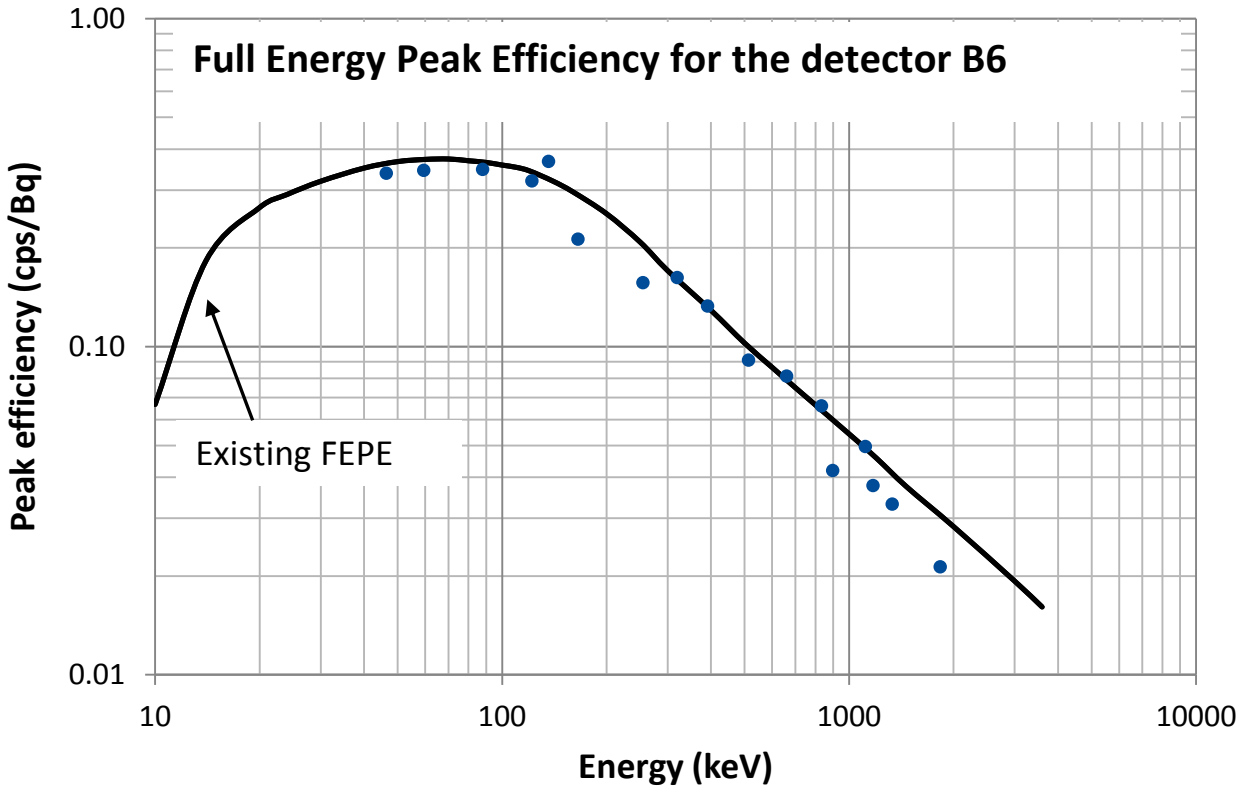
This is obtained by measurements (with appropriate corrections)

This is reported in the source certificate

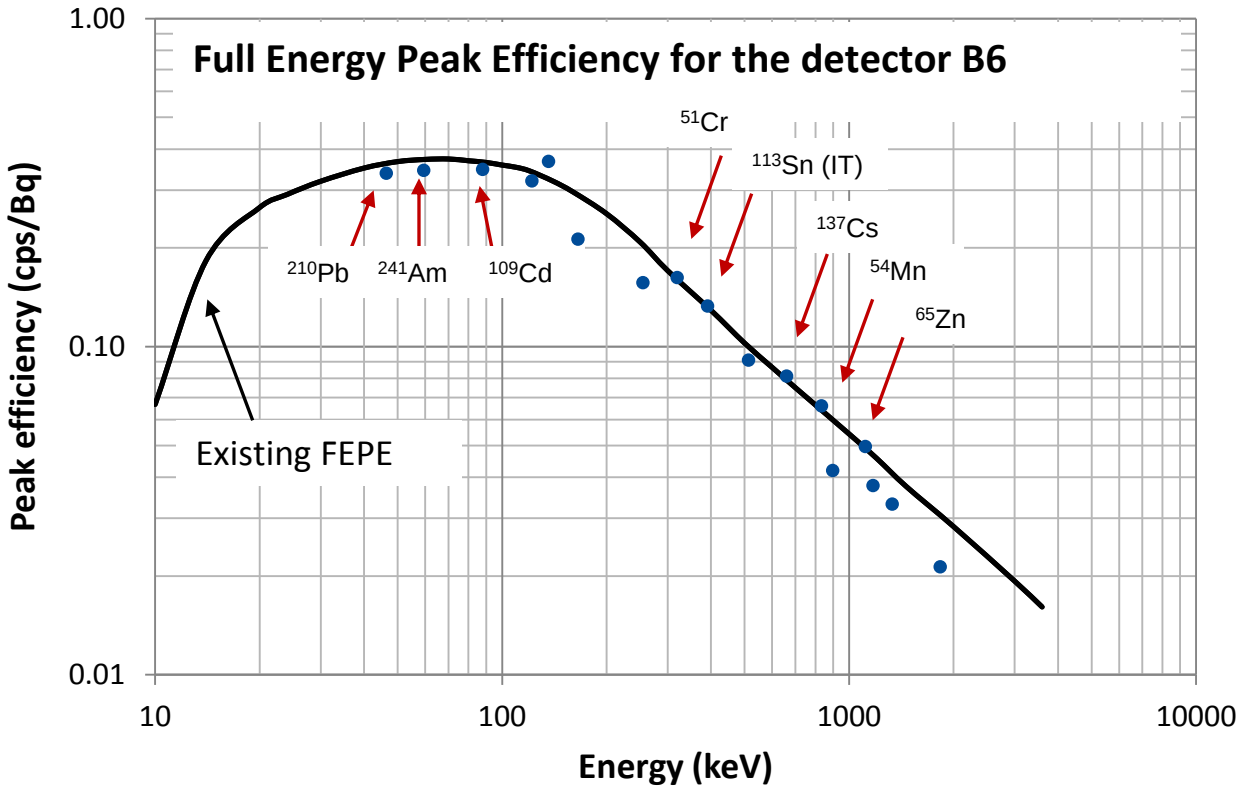
- In the following, a BeGe detector labelled as B6 and a W-beaker with close measurement geometry is considered.
- A certified reference source of thickness ∼ 0 was measured (dead time 3.6% → no significant random coincidences).
- The source contained nuclides ^{210}Pb … ^{88}Y (46 … 1836 keV).



- The very first result

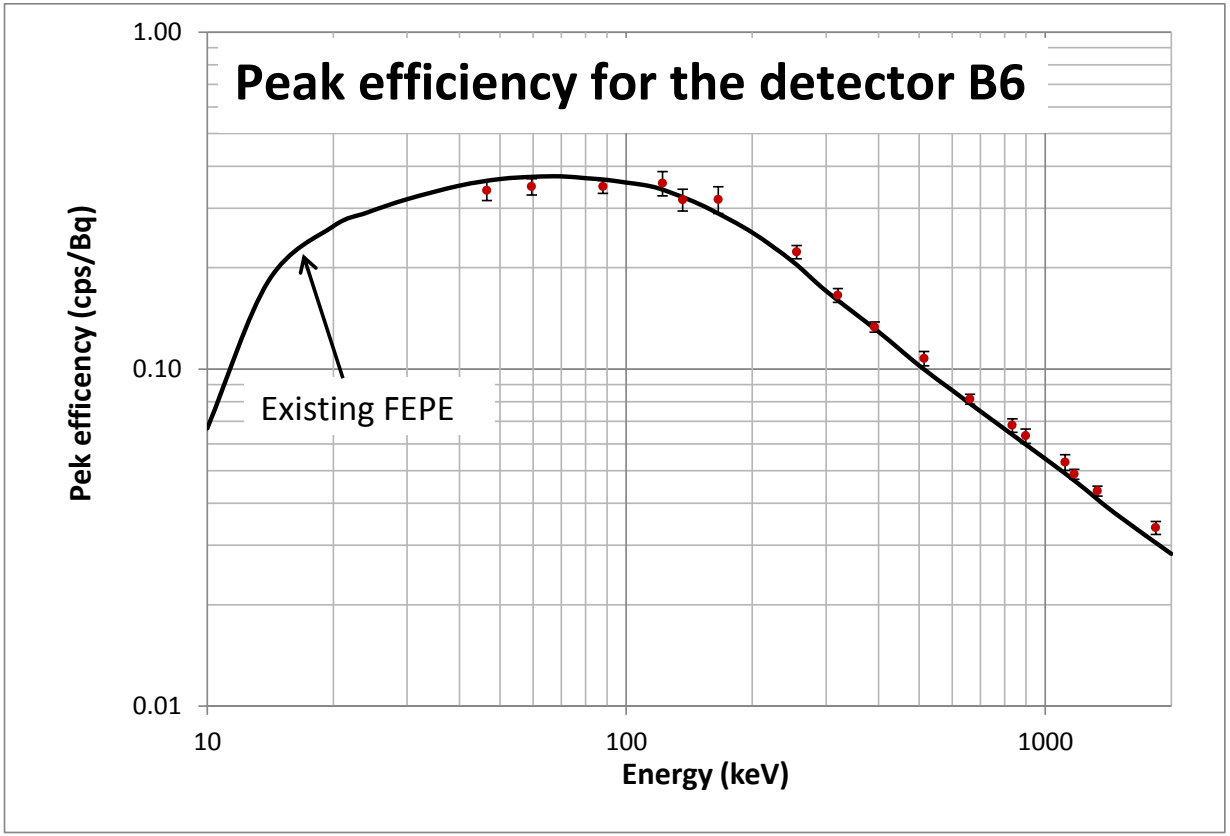


- The very first result



Nuclide for which the CC is 1 (or ∼1)

- Secondly, Unisampo-Shaman true coincidence correction factors were taken into account (+ preliminary uncertainties).



Much better result, but still some deficiencies. Why?

- I contacted with the developer of the Unisampo-Shaman software → several calibration nuclides have γ - X-ray true coincidences, but there were deficiencies in the decay scheme data library (X-ray cutoff 15 keV, X-ray energies lumped).

Gammas from ^{88}Y (106.65 d 4)

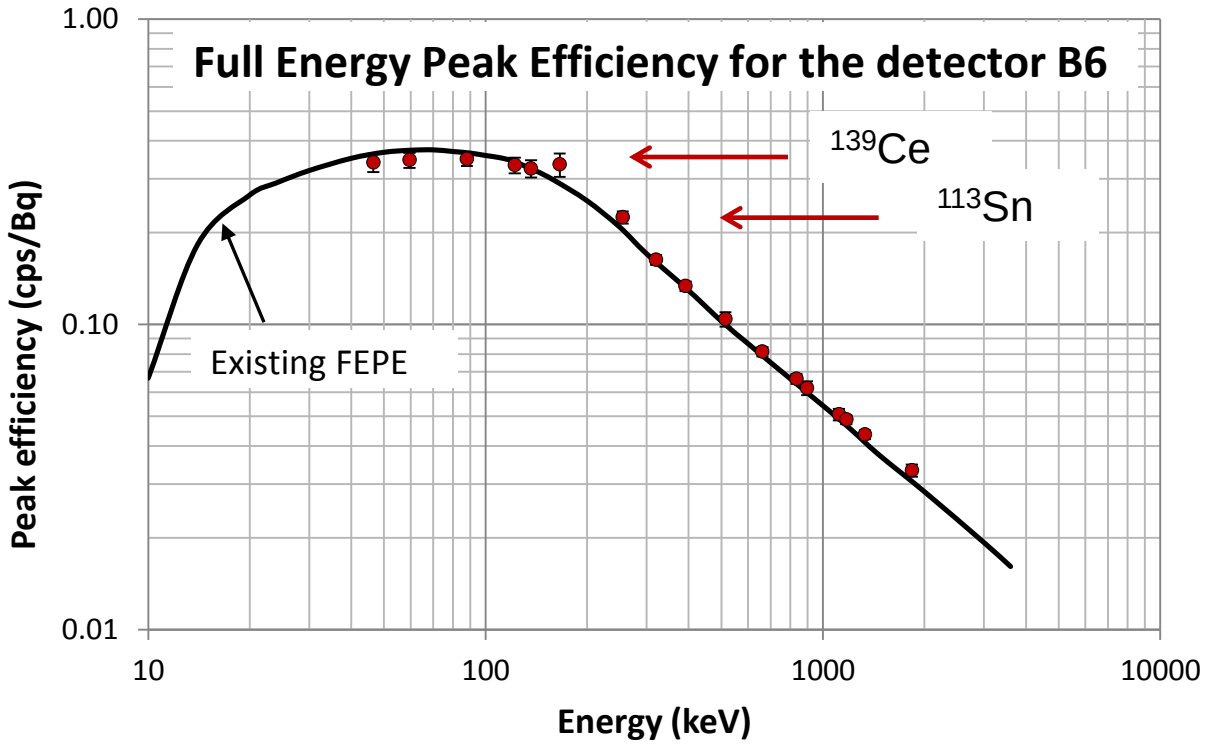
Eγ (keV)	Iγ (%)	Decay mode
850.647 *24*	0.065 *13*	ε+β$^+$
898.042 *3*	93.7 *3*	ε+β$^+$
1382.406 *26*	0.021 *6*	ε+β$^+$
1836.063 *12*	99.2 *3*	ε+β$^+$
2734.086 *13*	0.71 *7*	ε+β$^+$
3218.48 *4*	0.007 *2*	ε+β$^+$

X-rays from ^{88}Y (106.65 d 4)

E (keV)	I (%)	Assignment
1,582	0.067 *15*	Sr L_l
1,649	0.038 *10*	Sr L_η
1,805	0.15 *3*	Sr $L_{\alpha 2}$
1,806	1.4 *3*	Sr $L_{\alpha 1}$
1,872	0.77 *19*	Sr $L_{\beta 1}$
1,902	0.0076 *16*	Sr $L_{\beta 6}$
1,937	0.038 *11*	Sr $L_{\beta 4}$
1,947	0.055 *17*	Sr $L_{\beta 3}$
2,196	0.0046 *14*	Sr $L_{\gamma 2}$
2,196	0.010 *3*	Sr $L_{\gamma 3}$
13,888	1.96E-04 *9*	Sr $K_{\alpha 3}$
14,098	17.8 *7*	Sr $K_{\alpha 2}$
14,165	34.3 *14*	Sr $K_{\alpha 1}$
15,825	2.55 *11*	Sr $K_{\beta 3}$
15,836	4.94 *20*	Sr $K_{\beta 1}$
15,971	0.0189 *10*	Sr $K_{\beta 5}$
16,085	0.88 *4*	Sr $K_{\beta 2}$
16,105	0.150 *8*	Sr $K_{\beta 4}$

Note: In the case of the BeGe detectors the FEPE values are not negligible for low energies.

- Almost perfect FEPE values! However, there are still points at the energy of 166 keV (^{139}Ce) and 255 keV (^{113}Sn) that are out of the FEPE curve. In addition, low-energy (<46 keV) values are missing.



X-rays from ^{139}Ce (137.640 d *23*)

E (keV)	I (%)	Assignment
4,121	0.183 *25*	La L_l
4,529	0.079 *8*	La L_η
4,634	0.50 *6*	La $L_{\alpha 2}$
4,651	4.5 *5*	La $L_{\alpha 1}$
5,042	2.8 *3*	La $L_{\beta 1}$
5,062	0.43 *9*	La $L_{\beta 4}$
5,143	0.72 *15*	La $L_{\beta 3}$
5,212	0.038 *4*	La $L_{\beta 6}$
5,384	0.94 *11*	La $L_{\beta 2}$
5,483	0.0095 *11*	La $L_{\beta 5}$
5,792	0.42 *5*	La $L_{\gamma 1}$
5,891	0.0045 *6*	La $L_{\gamma 6}$
6,061	0.12 *3*	La $L_{\gamma 2}$
6,075	0.17 *4*	La $L_{\gamma 3}$
32,658	0.00340 *12*	La $K_{\alpha 3}$
33,034	23.8 *7*	La $K_{\alpha 2}$
33,442	43.6 *12*	La $K_{\alpha 1}$
37,720	4.11 *11*	La $K_{\beta 3}$
37,801	7.93 *22*	La $K_{\beta 1}$
38,084	0.096 *4*	La $K_{\beta 5}$
38,726	2.57 *7*	La $K_{\beta 2}$
38,826	0.62 *3*	La $K_{\beta 4}$

Total efficiencies must be checked!

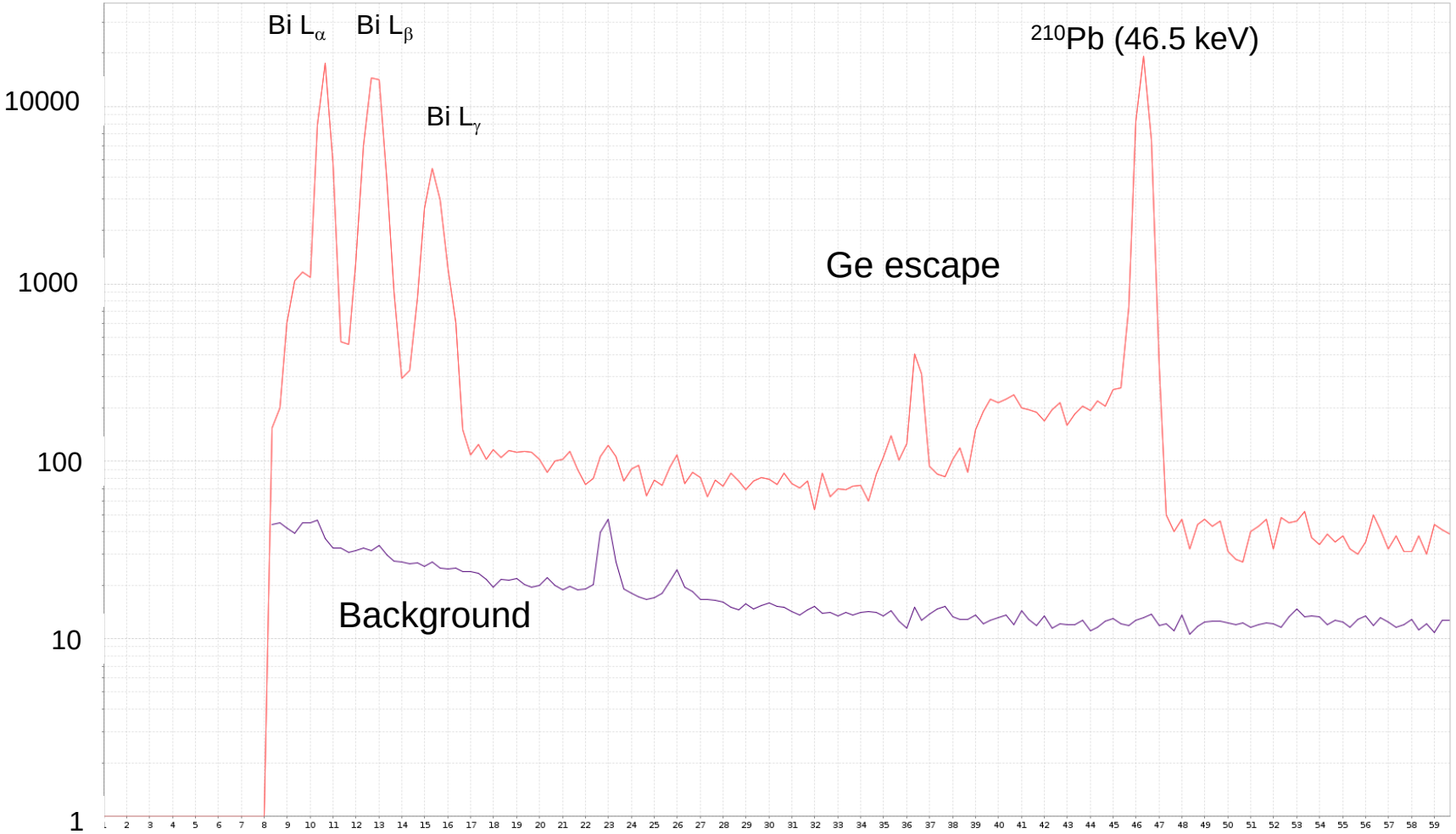
- Problem in the case of low-energy γ's: appropriate nuclides (and having no CC) are difficult to find → let's try to use X-rays!
- Another problem: In terms of energy the X-rays are located close to each other → the peaks are lumped → Gamma99 software was used for the lumped peak areas!
- Example: ^{210}Pb.

Gammas from ^{210}Pb (22.3 y 2)

Eγ (keV)	Iγ (%)	Decay mode
46.539 *1*	4.25 *4*	β^-

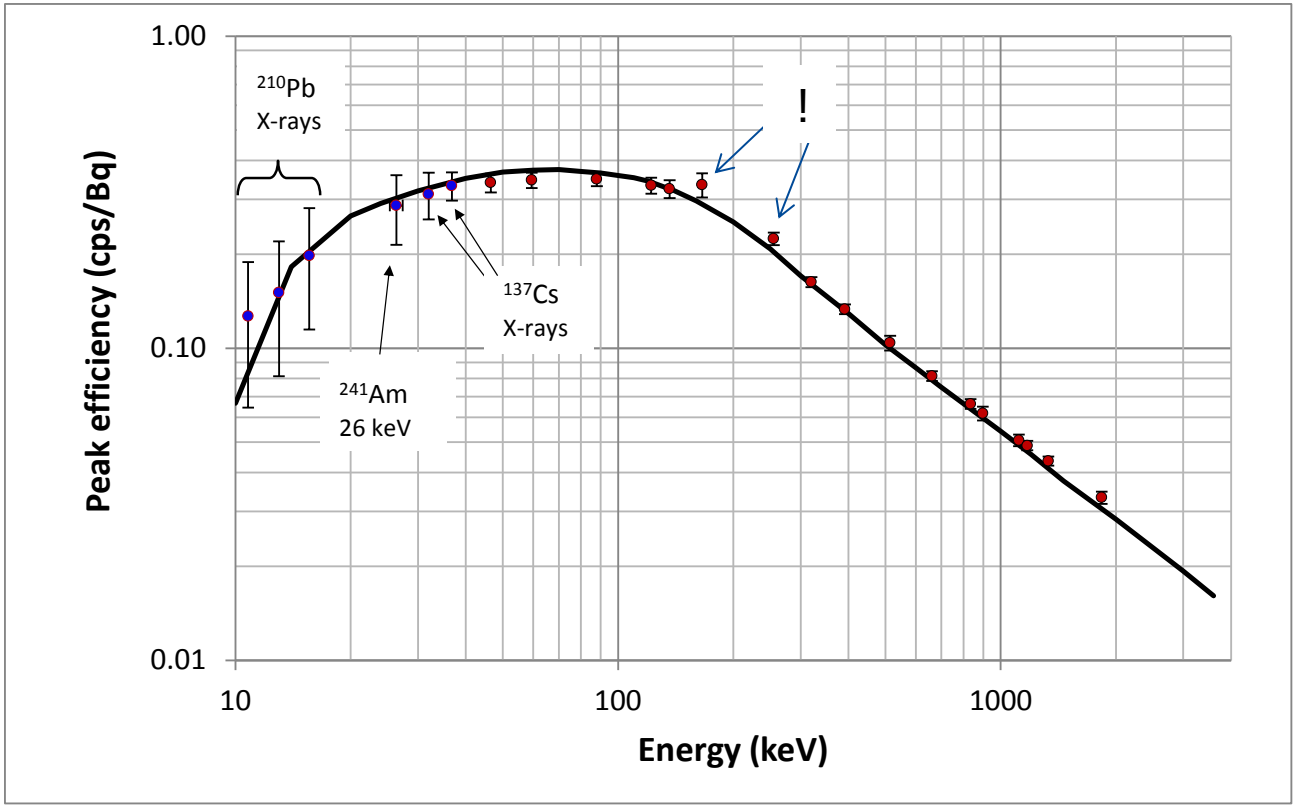
X-rays from ^{210}Pb (22.3 y 2)

	E (keV)	I (%)	Assignment
	9,420	0.49 *5*	Bi L_l
9.6%	10,731	0.97 *6*	Bi $L_{\alpha 2}$
9.6%	10,839	8.6 *5*	Bi $L_{\alpha 1}$
	11,712	0.083 *5*	Bi L_η
11%	12,480	0.137 *9*	Bi $L_{\beta 6}$
11%	12,691	2.2 *3*	Bi $L_{\beta 4}$
11%	12,967	2.15 *14*	Bi $L_{\beta 2}$
11%	13,023	3.53 *23*	Bi $L_{\beta 1}$
11%	13,211	2.6 *4*	Bi $L_{\beta 3}$
11%	13,393	0.299 *19*	Bi $L_{\beta 5}$
2.6%	15,247	0.75 *5*	Bi $L_{\gamma 1}$
2.6%	15,582	0.76 *12*	Bi $L_{\gamma 2}$
2.6%	15,685	0.137 *13*	Bi $L_{\gamma 6}$
2.6%	15,709	0.94 *15*	Bi $L_{\gamma 3}$



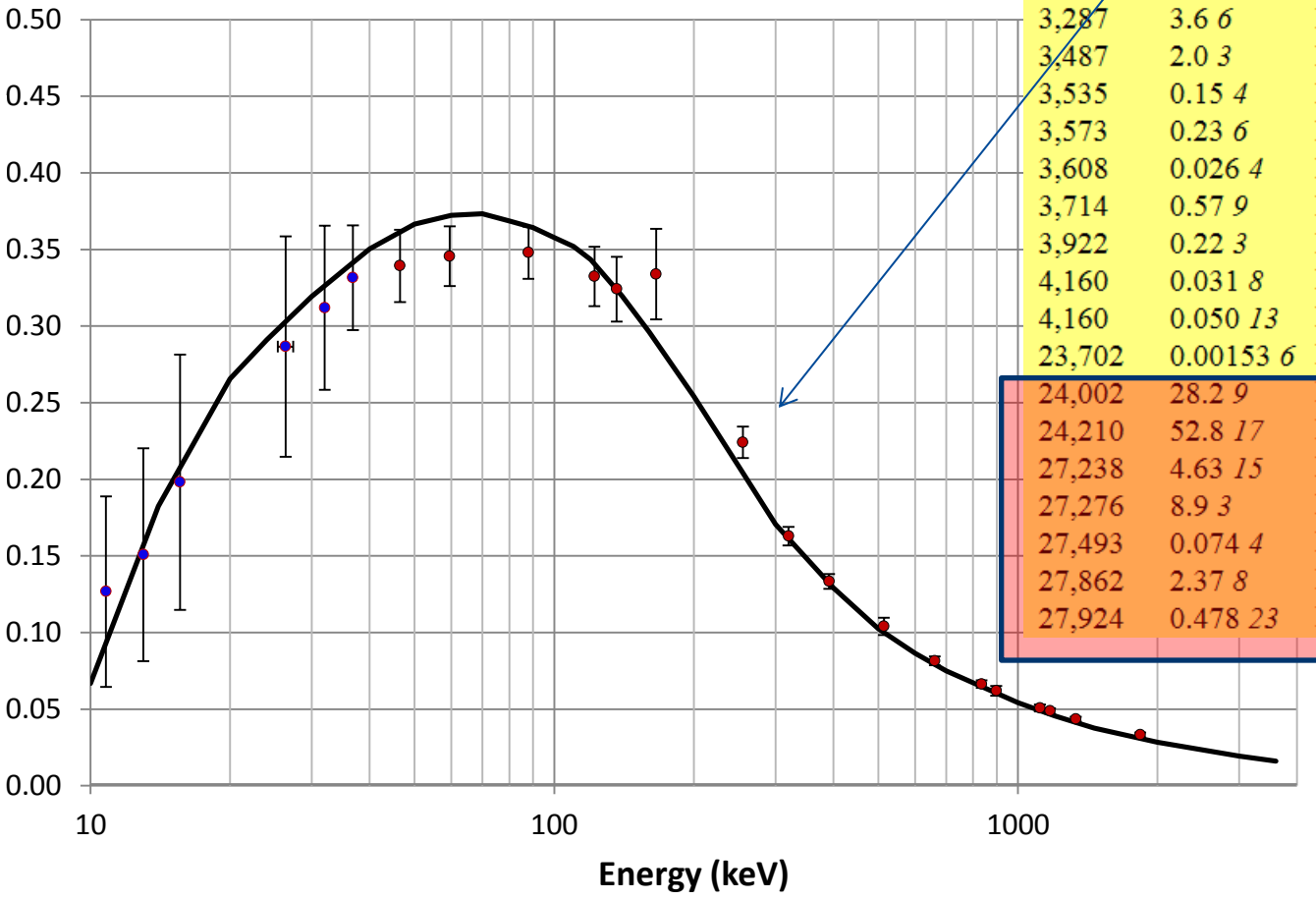
Rough efficiency estimate from the X-ray peaks can be obtained.

- ^{137}Cs (Ba) x-rays (32 and 37 keV) and ^{241}Am 26-keV gamma were also taken into account.



Quite nice! But still: ^{139}Ce and ^{113}Sn 255 keV should fit better.

Linear y-axis:

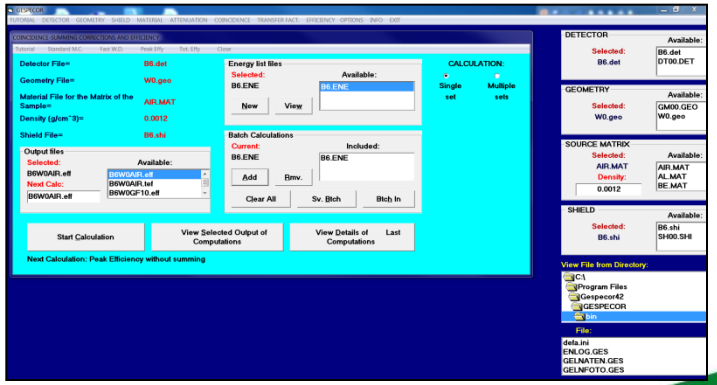


X-rays from ^{113}Sn (115.09 d 4)

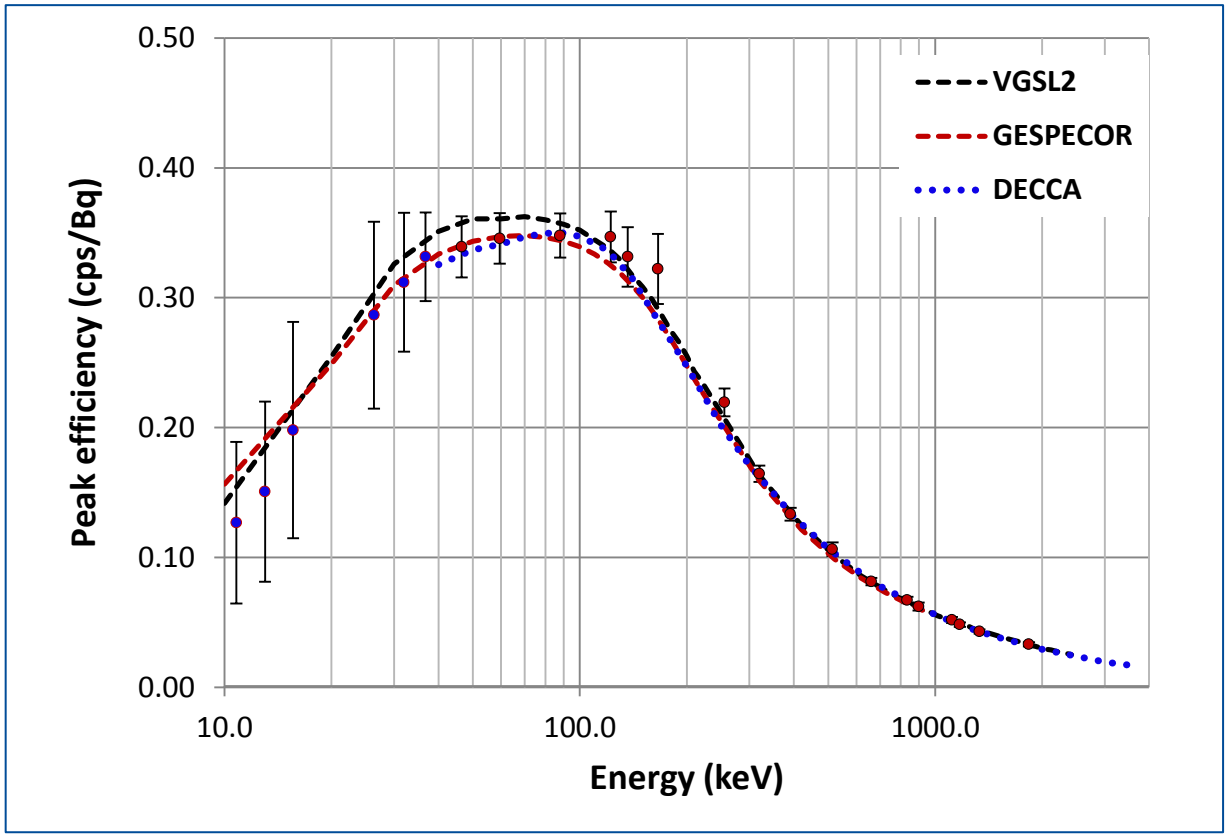
E (keV)	I (%)	Assignment
2,905	0.15 *3*	In L_l
3,112	0.069 *10*	In L_η
3,279	0.41 *6*	In $L_{\alpha 2}$
3,287	3.6 *6*	In $L_{\alpha 1}$
3,487	2.0 *3*	In $L_{\beta 1}$
3,535	0.15 *4*	In $L_{\beta 4}$
3,573	0.23 *6*	In $L_{\beta 3}$
3,608	0.026 *4*	In $L_{\beta 6}$
3,714	0.57 *9*	In $L_{\beta 2}$
3,922	0.22 *3*	In $L_{\gamma 1}$
4,160	0.031 *8*	In $L_{\gamma 2}$
4,160	0.050 *13*	In $L_{\gamma 3}$
23,702	0.00153 *6*	In $K_{\alpha 3}$
24,002	28.2 *9*	In $K_{\alpha 2}$
24,210	52.8 *17*	In $K_{\alpha 1}$
27,238	4.63 *15*	In $K_{\beta 3}$
27,276	8.9 *3*	In $K_{\beta 1}$
27,493	0.074 *4*	In $K_{\beta 5}$
27,862	2.37 *8*	In $K_{\beta 2}$
27,924	0.478 *23*	In $K_{\beta 4}$

4. Total efficiency

- ε_{tot} is used for the determination of the true coincidence correction.
- ε_{tot} must be redetermined in order to get better ”fit” for the peak efficiency of ^{139}Ce (166 keV) and ^{113}Sn (255 keV).
- VGSL2 and GESPECOR calculations were performed to fix the input data → same input models were used for simulating ε_{tot} → new coincidence correction values for the determination of the peak efficiency.

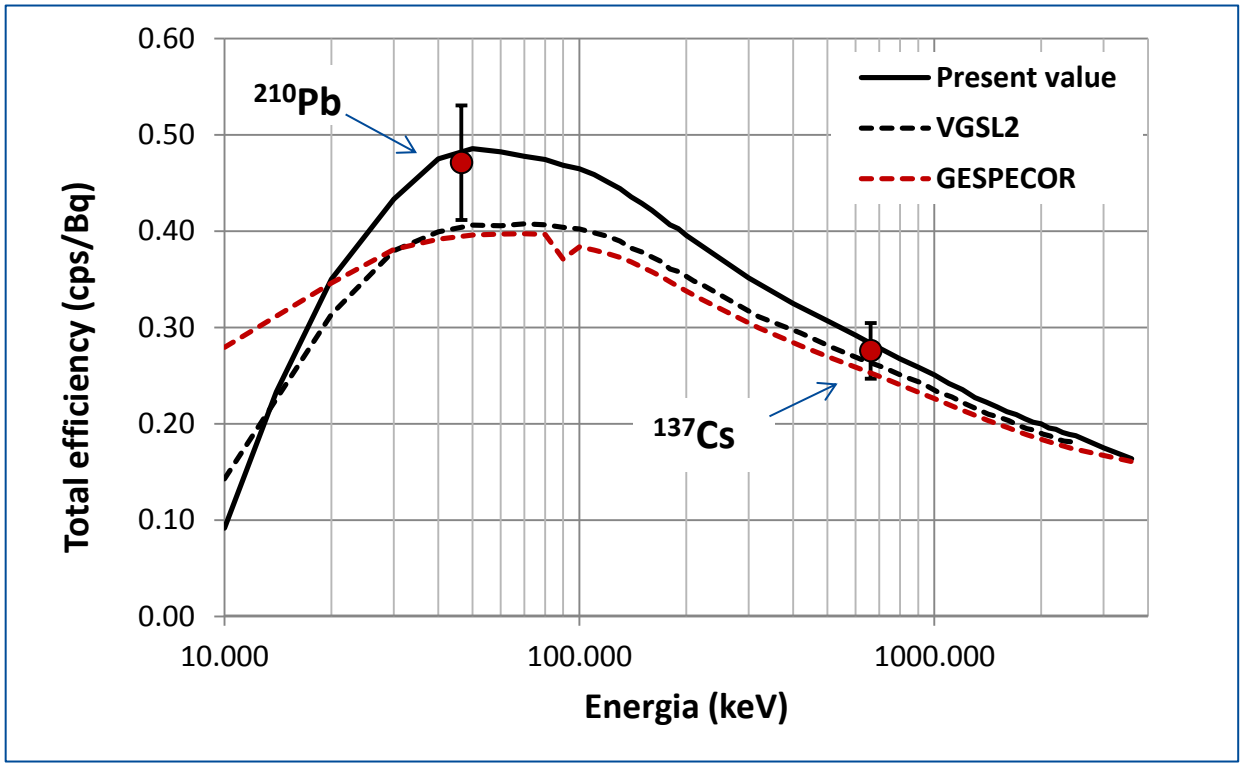


MC-simulated peak efficiencies (and the DECCA result)



Not a bad result!

Present and MC-simulated total efficiencies



Notable differences between the present and simulated total efficiencies! Average value of VGSL and GESPECOR were selected (except for the energies of 10 keV and 3.6 MeV)

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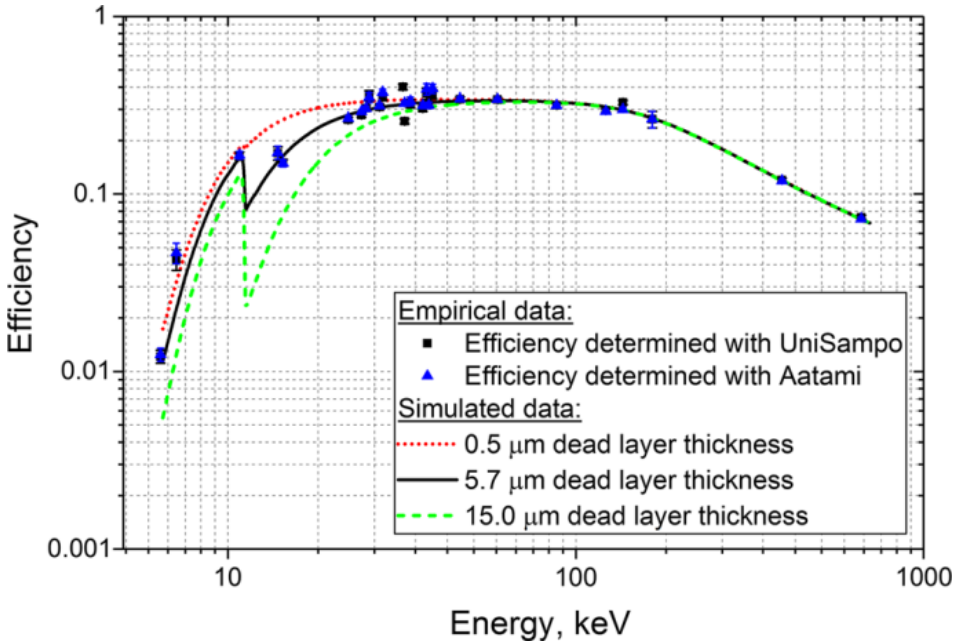
Accurate efficiency characterization of a BEGe detector in the low-energy range using empirical and Monte Carlo simulation approaches

Pawel Mekarski[1] · Weihua Zhang[1] · Chuanlei Liu[1] · Kurt Ungar[1]



5. Methodology of the uncertainty determination

- Efficiency values are out of relevance if there is no proper uncertainty estimation → efficiency uncertainties are part of the full uncertainty budget when “real” samples are under investigation.
- Components affecting the uncertainty
  - must be identified and
  - must be quantified
- Components not included in the uncertainty calculation must be identified (here e.g. inhomogeneity of the ”real” samples and gamma-ray attenuation).

Note! Present practice/convention in the uncertainty estimation of the efficiencies:

Uncertainties associated with the samples under investigation are accounted for in the peak efficiency uncertainty determination !!!???

The uncertainties are not only associated with n_i, A_i or I_γ.
The corrections also have uncertainties:

- Decay correction prior to the counting period
- Decay correction during the counting period
- Random coincidence summing correction
- True coincidence summing correction
- Correction for the self attenuation
- Correction for the source inhomogeneity
- Correction for other source characteristics
- …

$$\varepsilon_i = \frac{n_i \cdot C_i}{A_i \cdot I_\gamma}$$

Standard uncertainty propagation formula used here

At least following points/items must be accounted for to determine efficiency uncertainties:

A. Equipment used in the measurements
B. Measurement conditions (environment)
C. Characteristics of the reference source
D. Measurement geometry
E. Data acquisition
F. Spectrum analysis
G. Other points influencing the uncertainties

A. Equipment used in the measurements

- Stability especially when long data acquisition times are used (variation of the resolution, energy and efficiency calibration in time.
- MCA clock and settings (conversion gain, pole zero, pulse shaping time,…).

→ Not estimated but most probably their contribution to the uncertainty << 1%

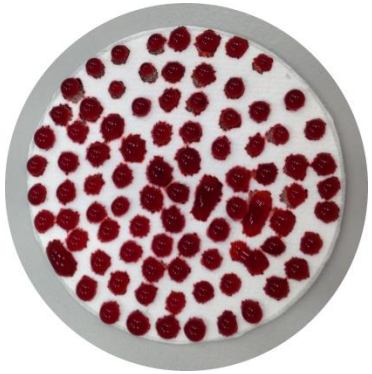
B. Measurement conditions (environment)

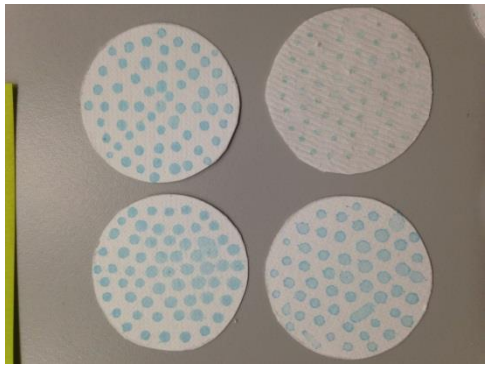
- Variation of the background as a function of time (routine background measurements and routine Rn monitoring).
- Means to prevent contamination.
- Variation of the air temperature, pressure and moisture (air conditioning).

→ Not quantified but certainly << 1%

C. Characteristics of the reference sources

- Testing the source (filter material) homogeneity in NPL





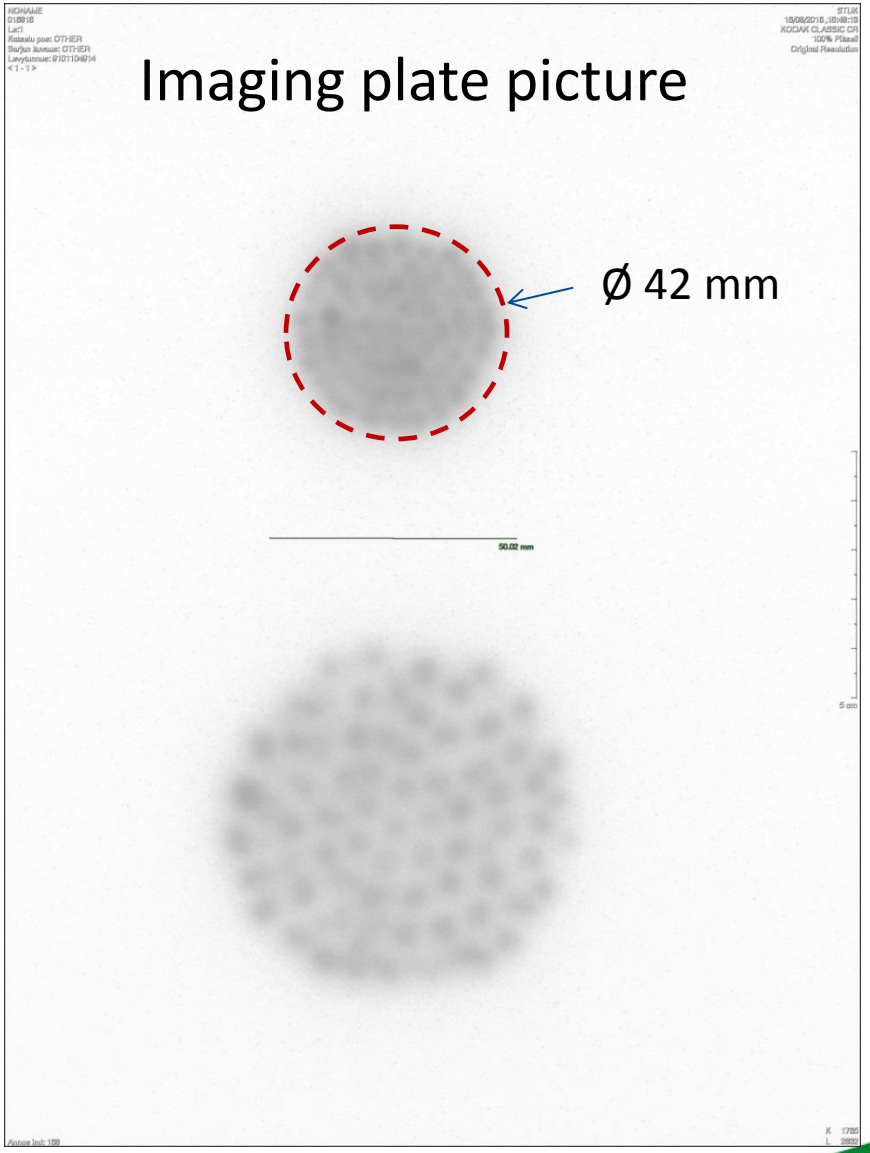
Text in the source certificate:

While efforts are made to ensure the source is as uniform as practicable, the distribution of activity may not perfectly reflect that of a real source. The effect this will have on the derived detector efficiency will depend upon the measurement geometry, detector construction and any prior treatment of the source such as crushing. In the absence of locally-determined correction factors, NPL suggests adding an uncertainty of ± 5 % to cover such effects.

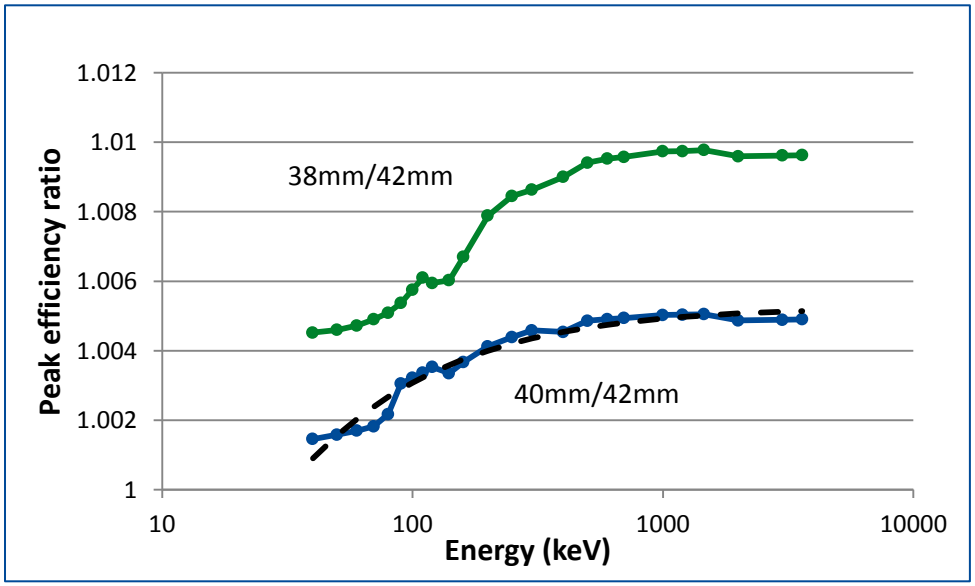
(*k*=2)

Does inhomogenous radionuclide distribution have an effect?

Beaker inner diameter 42 mm



- Correction for the source diameter (42 mm → 40 mm):
  - Without correction, smaller reference source diameter lead to slightly higher peak efficiency (but probably a small effect).
  - DECCA-calculations of the efficiency was performed by assuming smaller diameters.
  - Fitting the DECCA-results using a ”smooth” function allows applying the correction to the energies of the reference nuclides.



→ Effect < 1%

No other programs (such as MCNP) were used to quantify the effect of inhomogeneous radionuclide distribution!

- Correction for the thickness of the reference source:
  - Whatmann GF 10 filter paper of thickness 0.35 mm and density 0.2 g cm^{-3}.
  - Without correction, reference source thickness lead to slightly lower peak efficiency.
  - DECCA program and fitting → less than 0.5% effect.

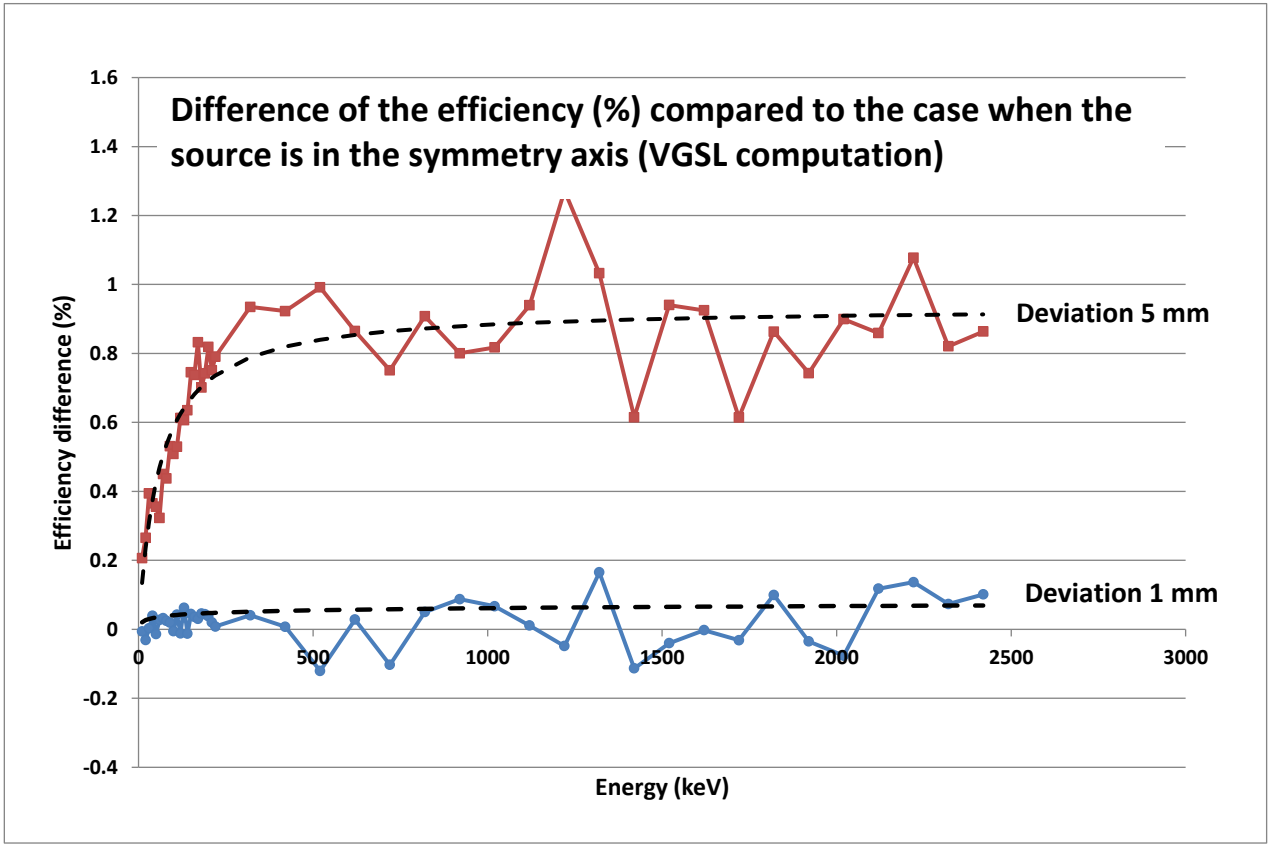
Note! the effects of true diameter and thickness of the reference source were accounted for as corrections (having no uncertainty). Additional uncertainty of 1% (not 2.5% as suggested by NPL) was used for the peak efficiencies.

D. Measurement geometry

At least following points must be quantified:

- Accuracy of the positioning of the reference source with respect to the symmetry axis of the detector (± 1 mm assumed, no sample guide used).
- In the case of ”real” samples the uncertainty of the positioning might be up to ± 5 mm.
- Dimension, thickness, density and composition variation of the beaker (container walls and bottom)

Example: The effect of the source horizontal displacement



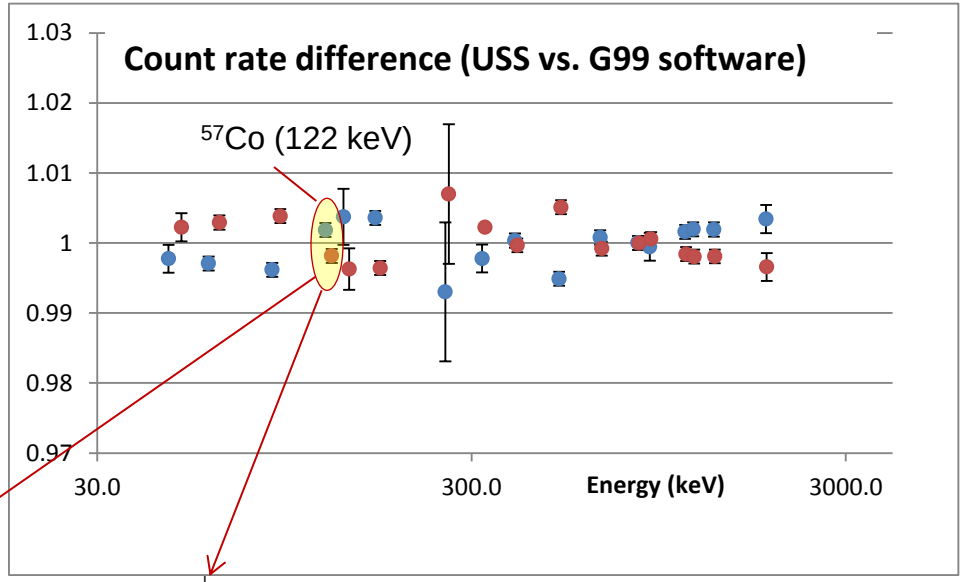
Effect < 1%

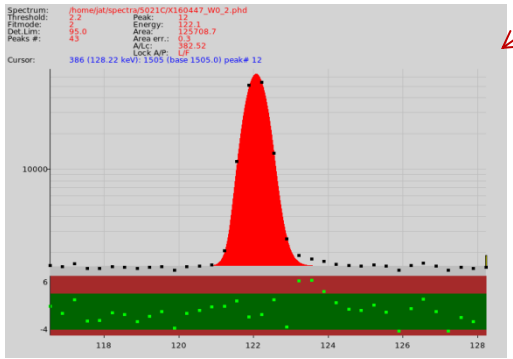
E. Data acquisition

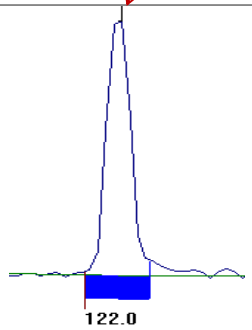
- Dead time in the reference source measurements < 5% → no random coincidences.
- Data acquisition time selected so that the areas of the main peaks ≈ 10^5 → statistical uncertainty < 1%.

F. Spectrum analysis

UniSampo-Shaman and Gamma99 software used in the analysis.







Additional uncertainty of 0.5% added to the peak areas

> 10^5 counts!

Coincidence correction (and its uncertainty) is of relevance!

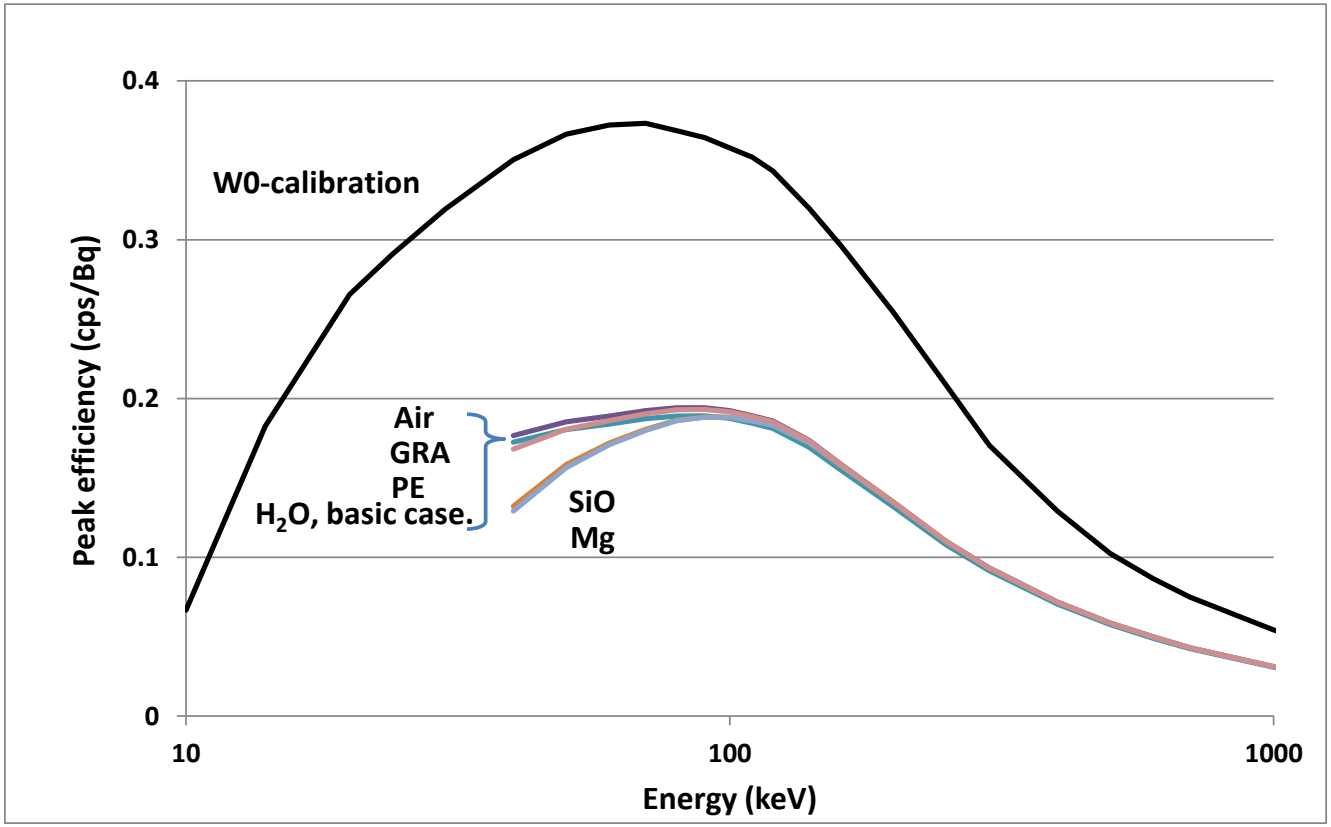
Nuclide	$t_{1/2}$	E_γ (keV)	COINCIDENCE CORRECTION cc_{G99}	cc_{USS}	$cc_{EFFTRAN}$	$cc_{GESPECOR}$	cc_{Final}	Δ_{cc}
Pb-210	22.23(12) a	46.5	1	1	1	1	1.000	0.000
Am-241	432.6(6) a	59.5	1	1	1.022	1	1.000	0.011
Cd-109	461.9(4) d	88.0	1	1	1	1	1.000	0.000
Co-57	271.80(5) d	122.1	1.02	1.058	1.11	1.1013	1.039	0.042
Co-57	271.80(5) d	136.4	0.86	0.87	0.816	0.9129	0.865	0.040
Ce-139	37.641(20) d	165.9	1.43	1.441	1.355	1.3227	1.387	0.058
Sn-113EC	115.09(3) d	255.2	1.35	1.347	1.401	1.352	1.349	0.026
Cr-51	27.704(4) d	320.1	0.98	1.009	1	1.0086	0.995	0.014
Sn-113IT	115.09(3) d	391.7	1	1	1	1	1.000	0.000
Sr-85	64.850(7) d	514.0	1.14	1.12	1.223	1.097	1.130	0.055
Cs-137	30.05(8) a	661.7	1	1	1	1	1.000	0.000
Mn-54	312.19(3) d	834.8	0.99	1.012	1.024	1.0155	1.001	0.014
Y-88	106.63(5) d	898.0	1.43	1.403	1.569	1.4652	1.508	0.073
Zn-65	244.01(9) d	1115.5	1.02	1.034	1.067	1.0866	1.027	0.030
Co-60	5.2711(8) a	1173.2	1.27	1.269	1.293	1.2656	1.270	0.013
Co-60	5.2711(8) a	1332.5	1.28	1.283	1.307	1.2776	1.282	0.014
Y-88	106.63(5) d	1836.1	1.49	1.457	1.634	1.5213	1.474	0.077

~5% unc.

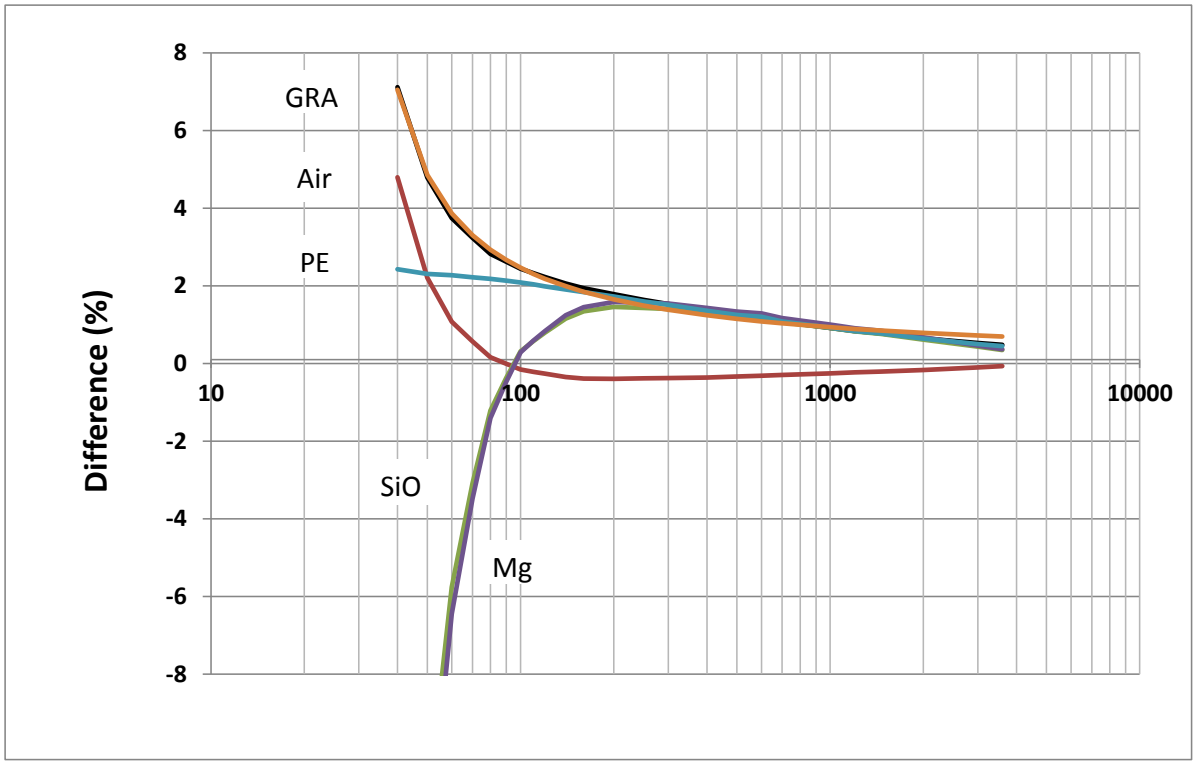
G. Other points influencing the uncertainties

- Elemental composition of the sample to be analyzed.
- Uncertainties associated with the measurement of the sample height (± 1 mm assumed) and density (mass determined with hich accuracy) determination.
- Variation of the beaker wall thickness (± 0.1 mm assumed); because of geometrical reasons this is not relevant for the reference source of thickness ∼0 mm.
- Horizontal displacement of the sample (± 5 mm assumed).

DECCA computation: The incluence of the elemental composition to the peak efficiency, sample height 26 mm.

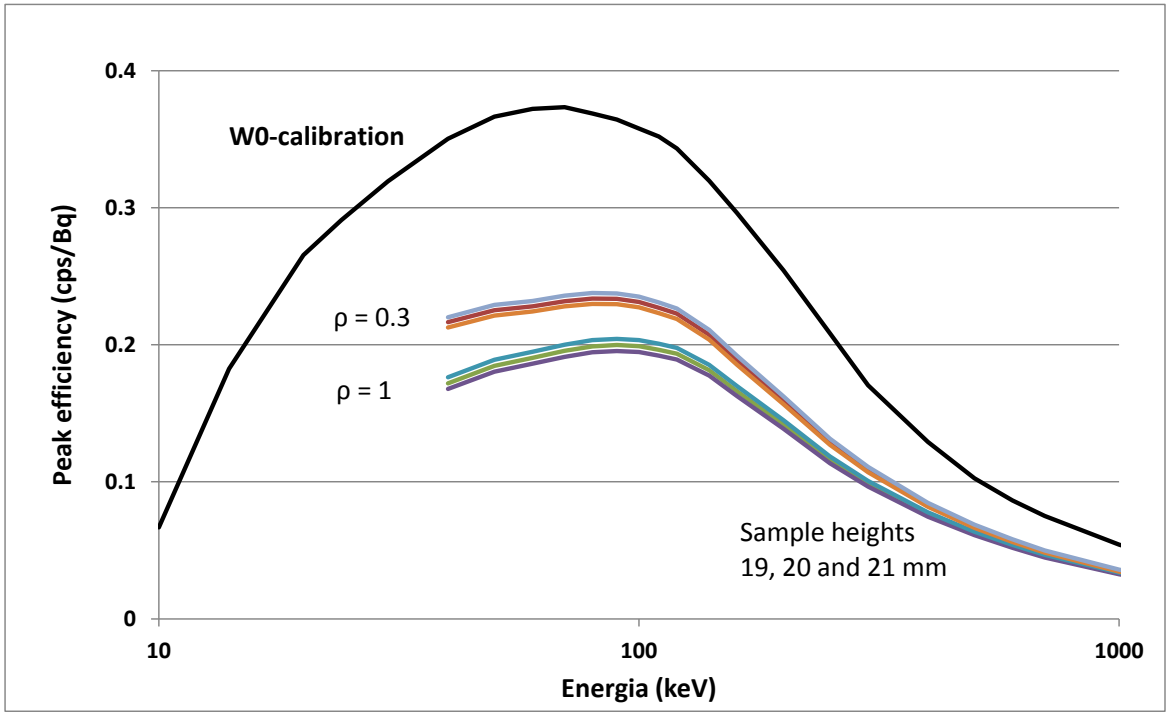


DECCA computation: The incluence of the elemental composition to the peak efficiency compared with the case of H_2O (density = 1 for all cases). Sample height 26 mm.



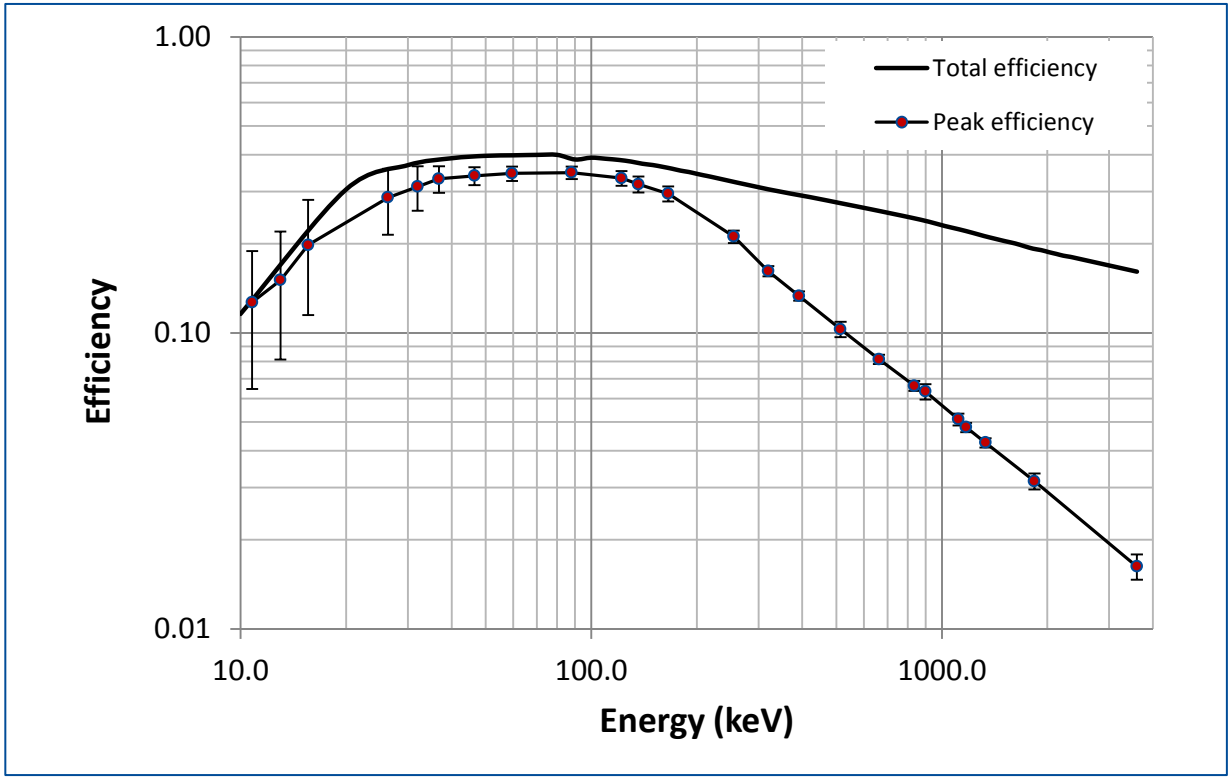
Conservative (?) selection for the uncertainty estimation: GRA

DECCA computation: The incluence of the sample height uncertainty (here ± 1 mm assumed for a sample of 20 mm in thickness; sample densities 1 and 0.3).



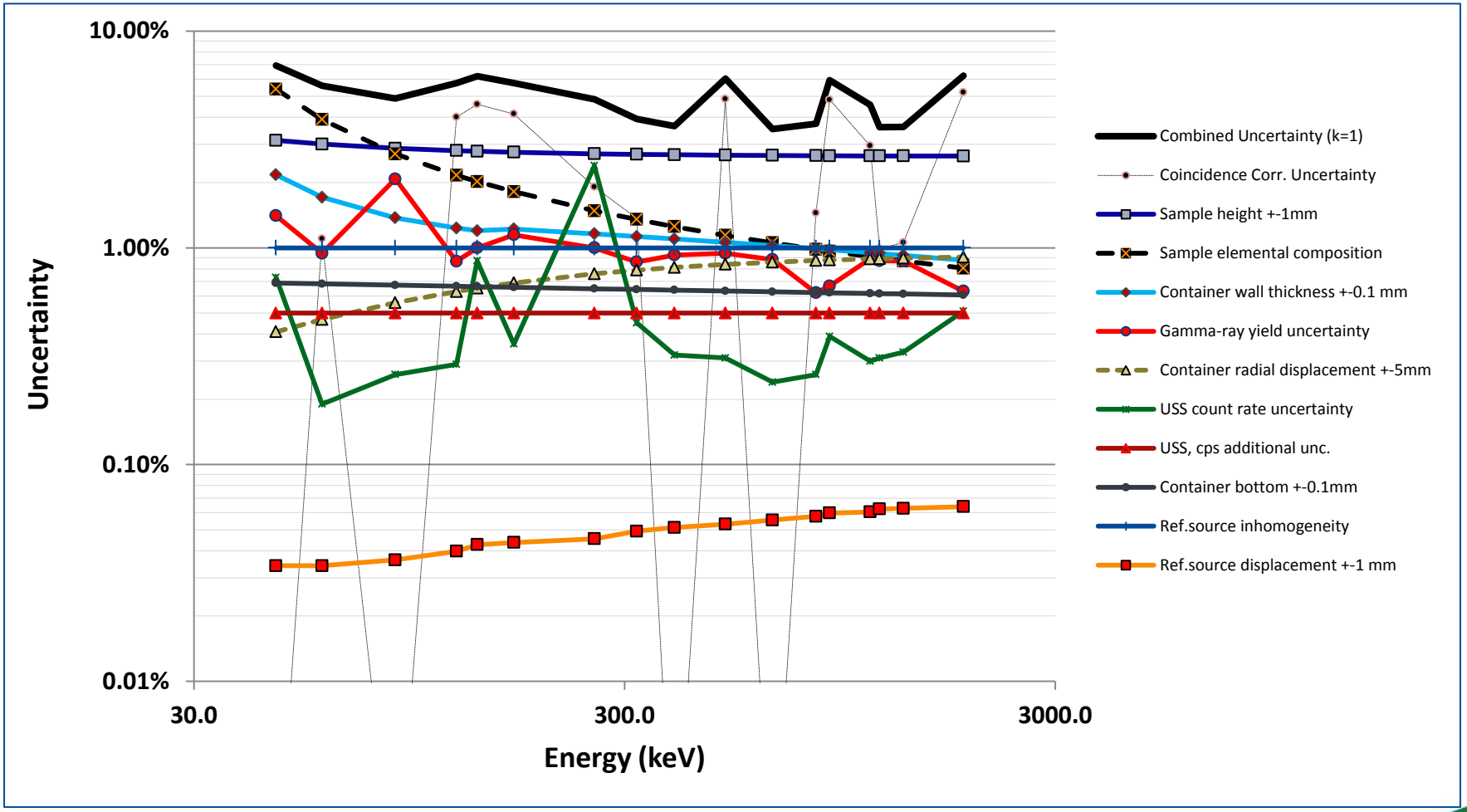
The effect is a bit larger in the case of a 5 mm thick source. This is accounted for in the uncertainty estimation.

5. Final result, comparison between the uncertainties and conclusions

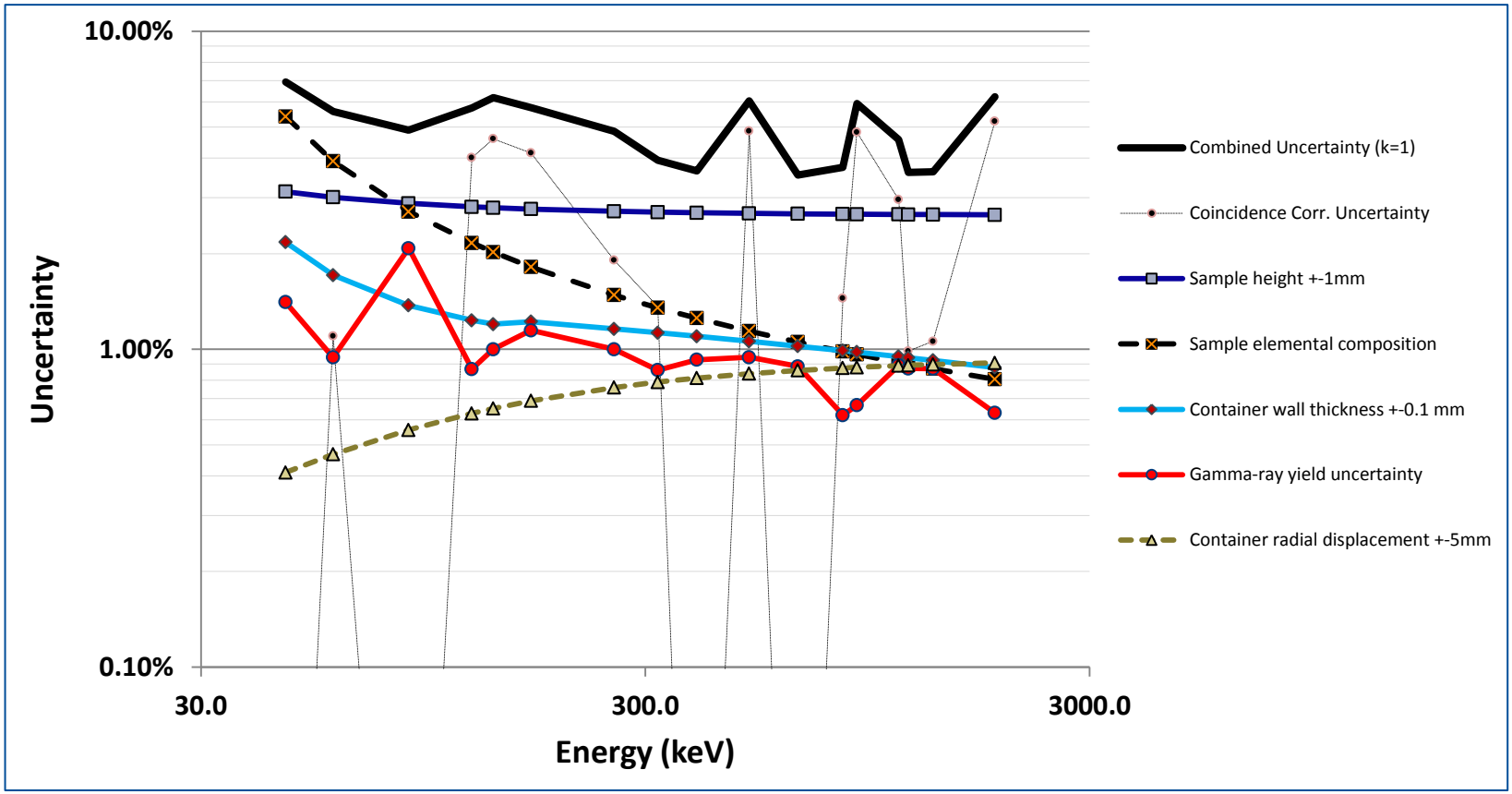


E (keV)	*ε* (cps/Bq)	Unc. (%)
10.8	0.127	49.1
13.0	0.151	46.1
15.6	0.198	42.1
26.3	0.287	25.1
32.0	0.312	17.2
36.8	0.332	10.3
46.5	0.339	6.9
59.5	0.346	5.6
88.0	0.348	4.9
122.1	0.333	5.7
136.4	0.318	6.2
165.9	0.295	5.8
255.2	0.212	4.9
320.1	0.162	3.9
391.7	0.133	3.6
514.0	0.103	6.0
661.7	0.081	3.5
834.8	0.066	3.7
898.0	0.063	5.9
1115.5	0.051	4.6
1173.2	0.048	3.6
1332.5	0.043	3.6
1836.1	0.032	6.2
3600.0	0.016	9.9

Uncertainty components as a function of energy (46 keV – 1836 keV)



Main uncertainty components: Coincidence correction, elemental composition and sample height of the ”real” sample.



A full and traceable efficiency determination with thorough uncertainty estimation is a tedious process including measurements and simulations/computations.

→ How to apply the methodology for a large number of detectors of different characteristics?

Main uncertainties are attached to the sample under investigation

→ Uncertainties associated with the efficiency calibration and those of the samples must be separated.

The results must also be compared in the case of other certified reference sources.