Analysis of Radioactive Disequilibrium in Natural Decay Chains due to Processing

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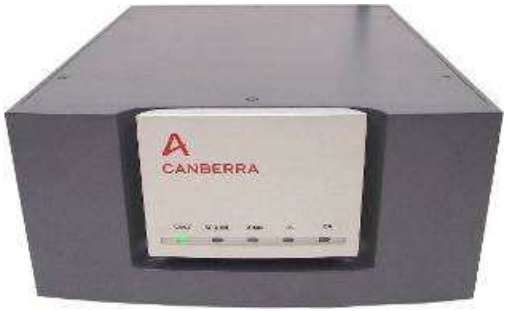
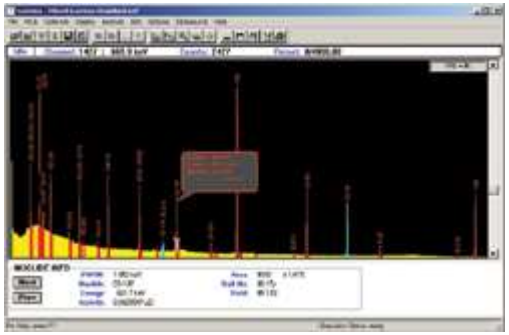
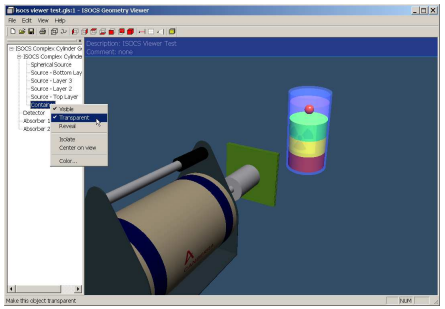
Fundamental basics

What do we need for accurate activity measurements of Natural Occuring Radioactive Material?







Fundamental basics

Information about nuclear decays (nuclear data)

nuclide library information

Detector efficiency as a function of detector, geometry, matrix, ……

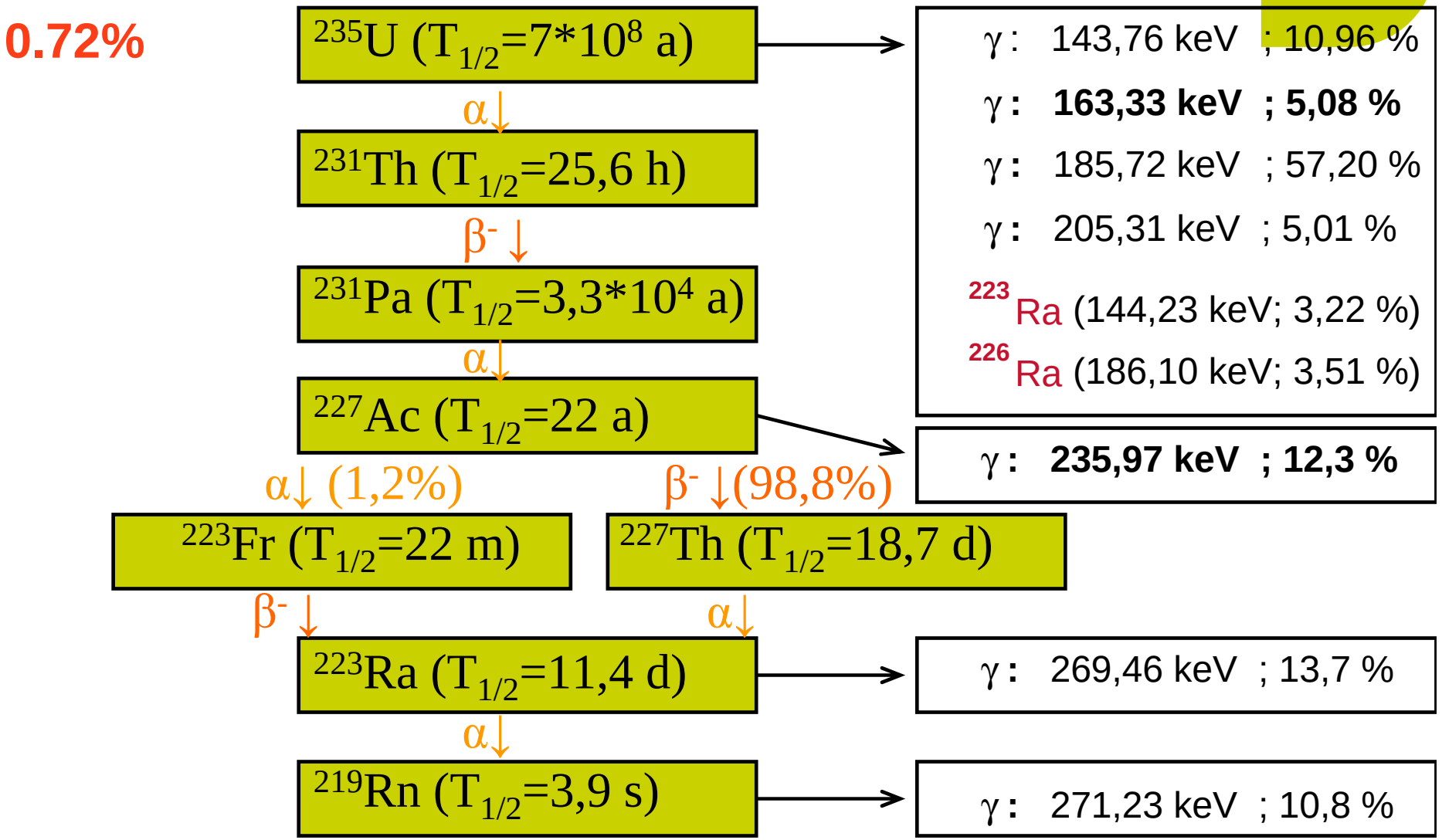
Improvement of background by passive and active shielding, flushing with nitrogen

Adjustment of peak fit algorithm. Interactive check!

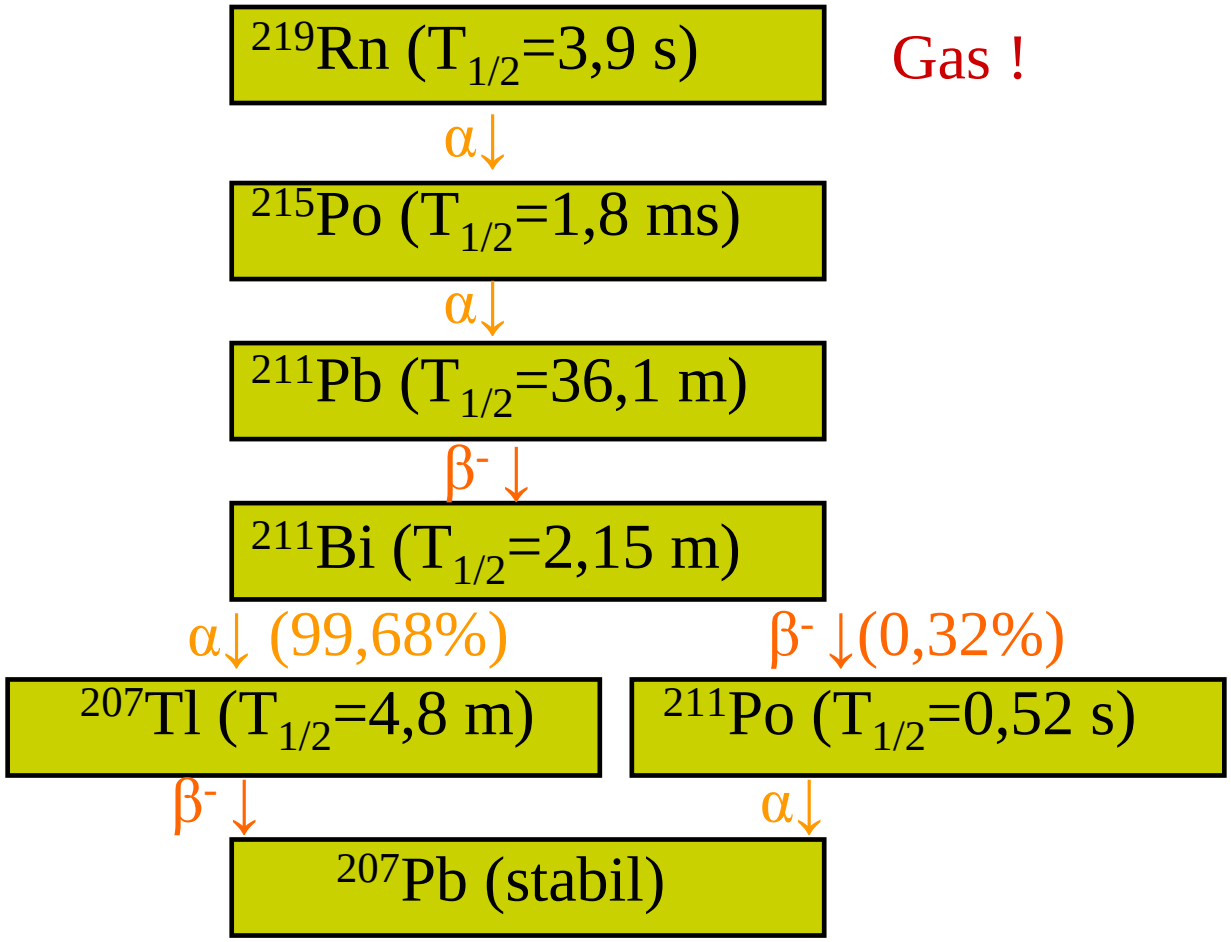
Information about processing of material prior to measurement

True Coincidence effects on special energies for certain nuclides

Measurement of ^{235}U and daughters

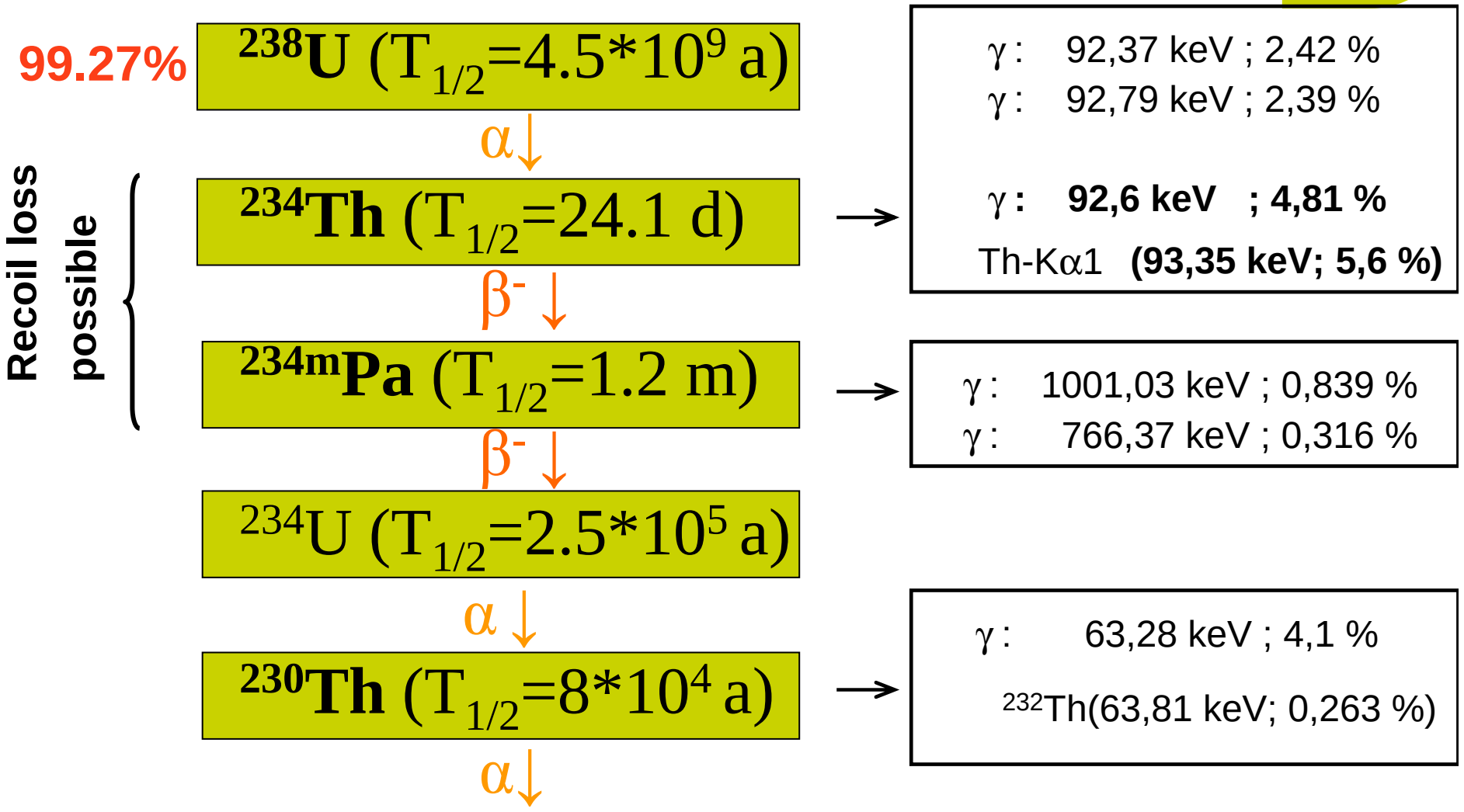


Measurement of ^{235}U and daughters

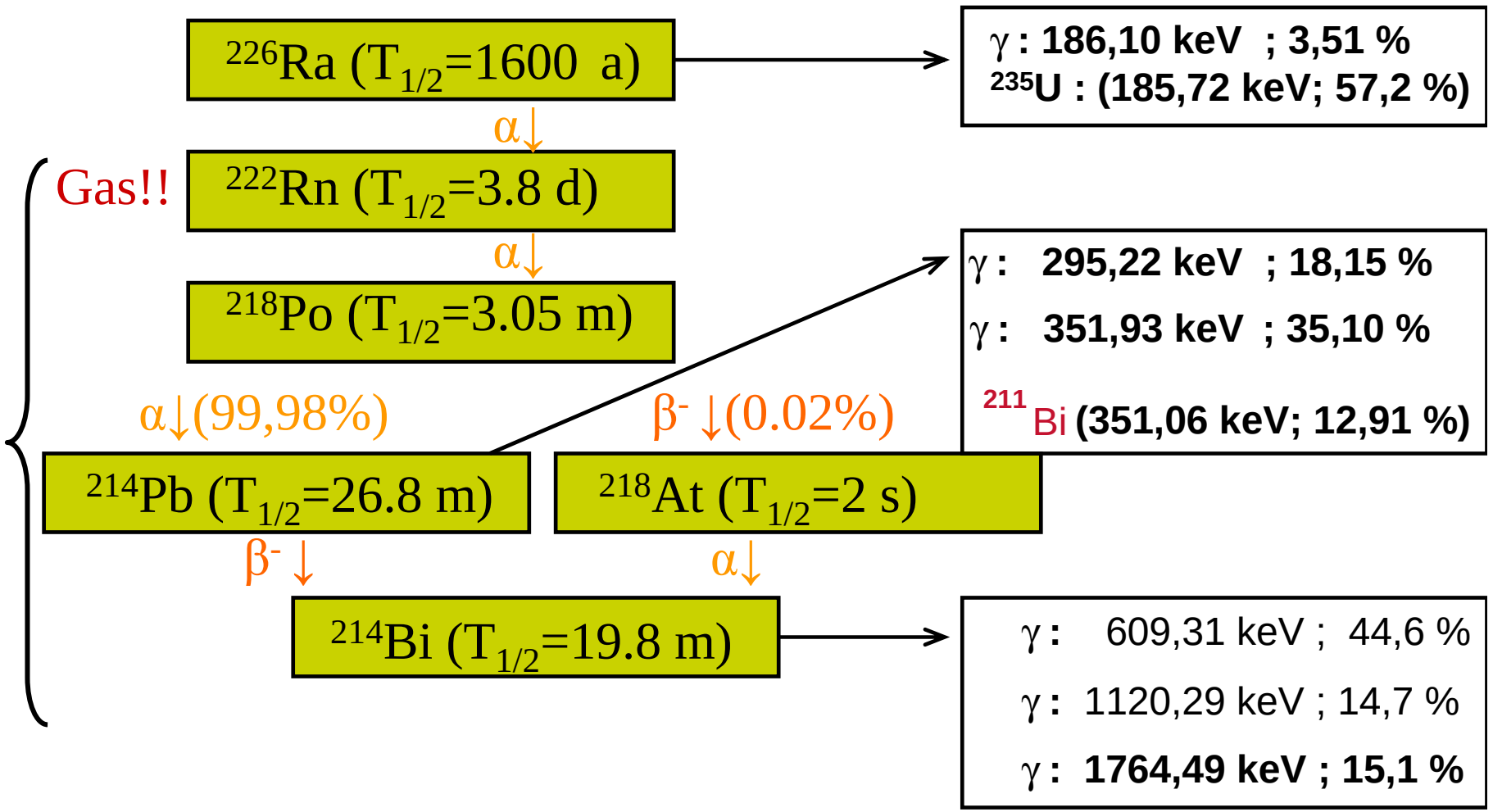


Additionally ^{219}At, ^{215}Bi und ^{215}At are produced!

Measurement of ^{238}U and daughters



Measurement of ^{238}U, ^{226}Ra and daughters



Measurement of ^{238}U, ^{226}Ra and daughters

- Measurement of the 186,10 keV-Linie ; 3,51 %

- But interfering with 185,72 keV; 57,2 % of ^{235}U

1- Use of ^{235}U-activity from other energy lines
and correct the ^{226}Ra-activity
2- Calculate ^{238}U-activity from daughter nuclides.
Use of natural ration for ^{238}U/ ^{235}U=21,7 for ^{235}U-activity

=> For 2 equillibrium is essential!

Measurement of ^{238}U, ^{226}Ra and daughters

- **Measurement of daughters ^{214}Pb and ^{214}Bi**

- **But ^{222}Rn gaseous => losses**

^{214}Pb

γ : 295,22 keV ; 18,15 %

γ : 351,93 keV ; 35,10 %

Interferenz ^{211}Bi **(351,06 keV; 12,91 %)**

^{214}Bi

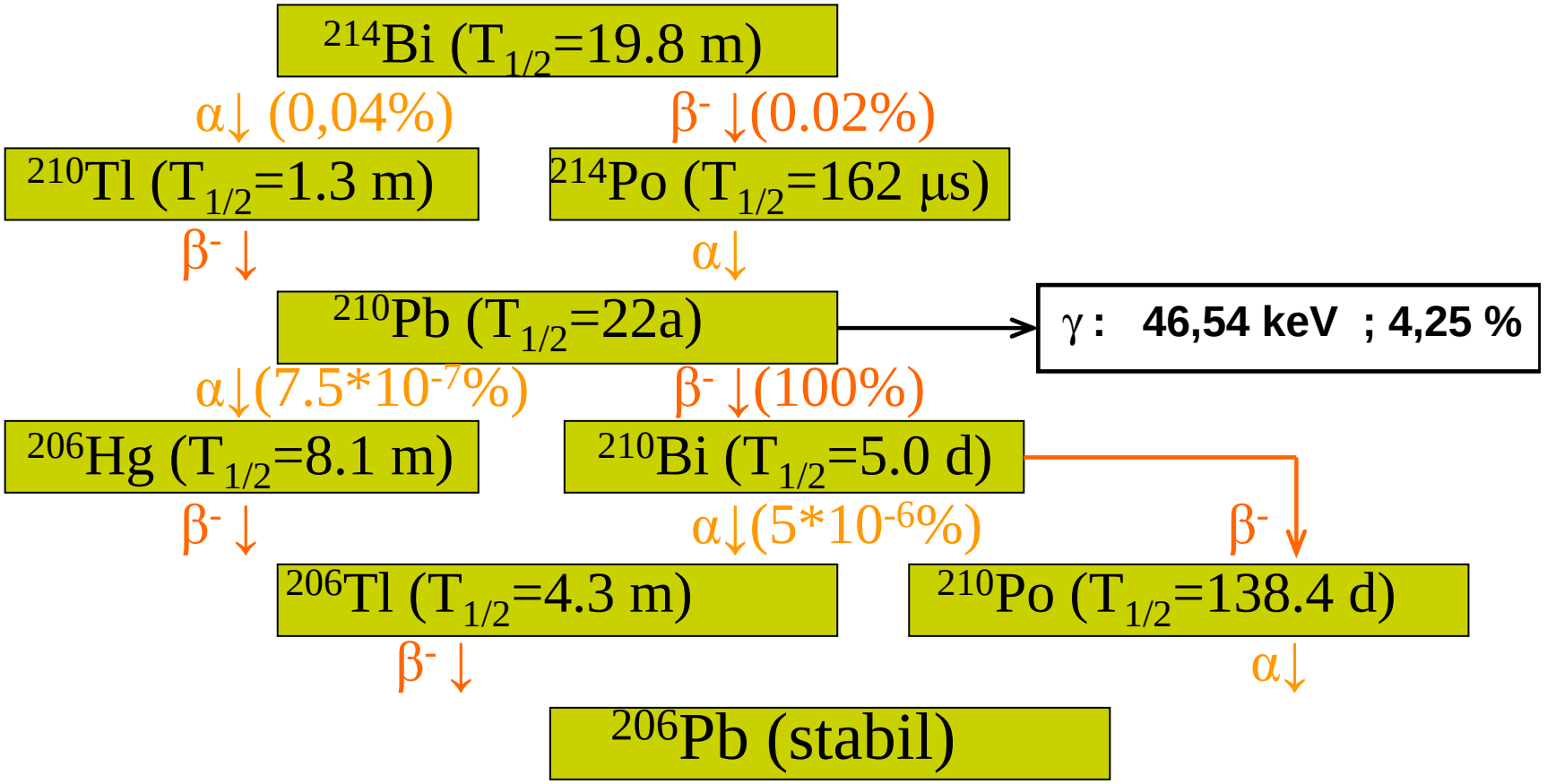
γ : 609,31 keV ; 44,6 %

γ : 1120,29 keV ; 14,7 %

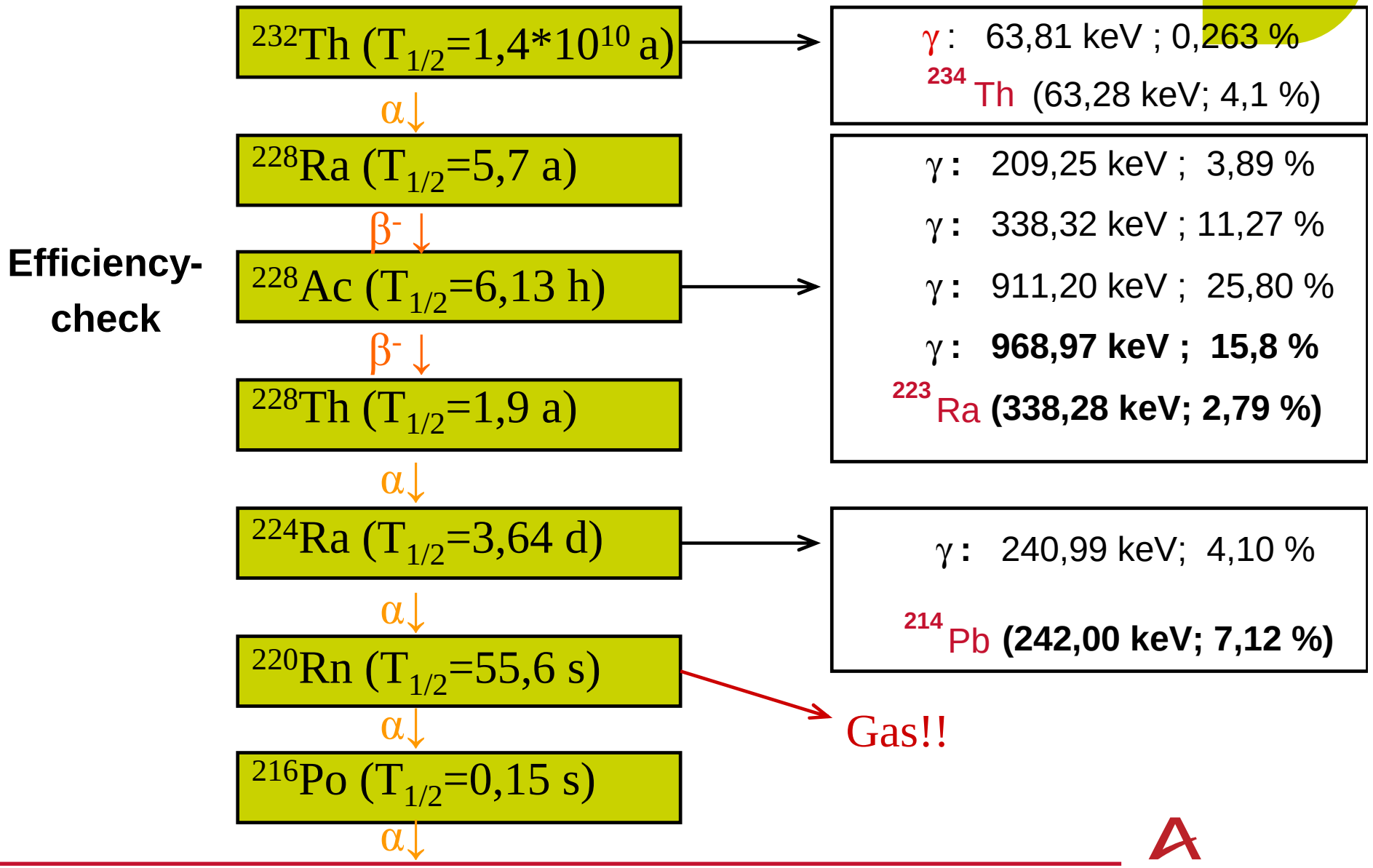
γ : 1764,49 keV ; 15,1 %

Gastight container for measurement and storage needed!

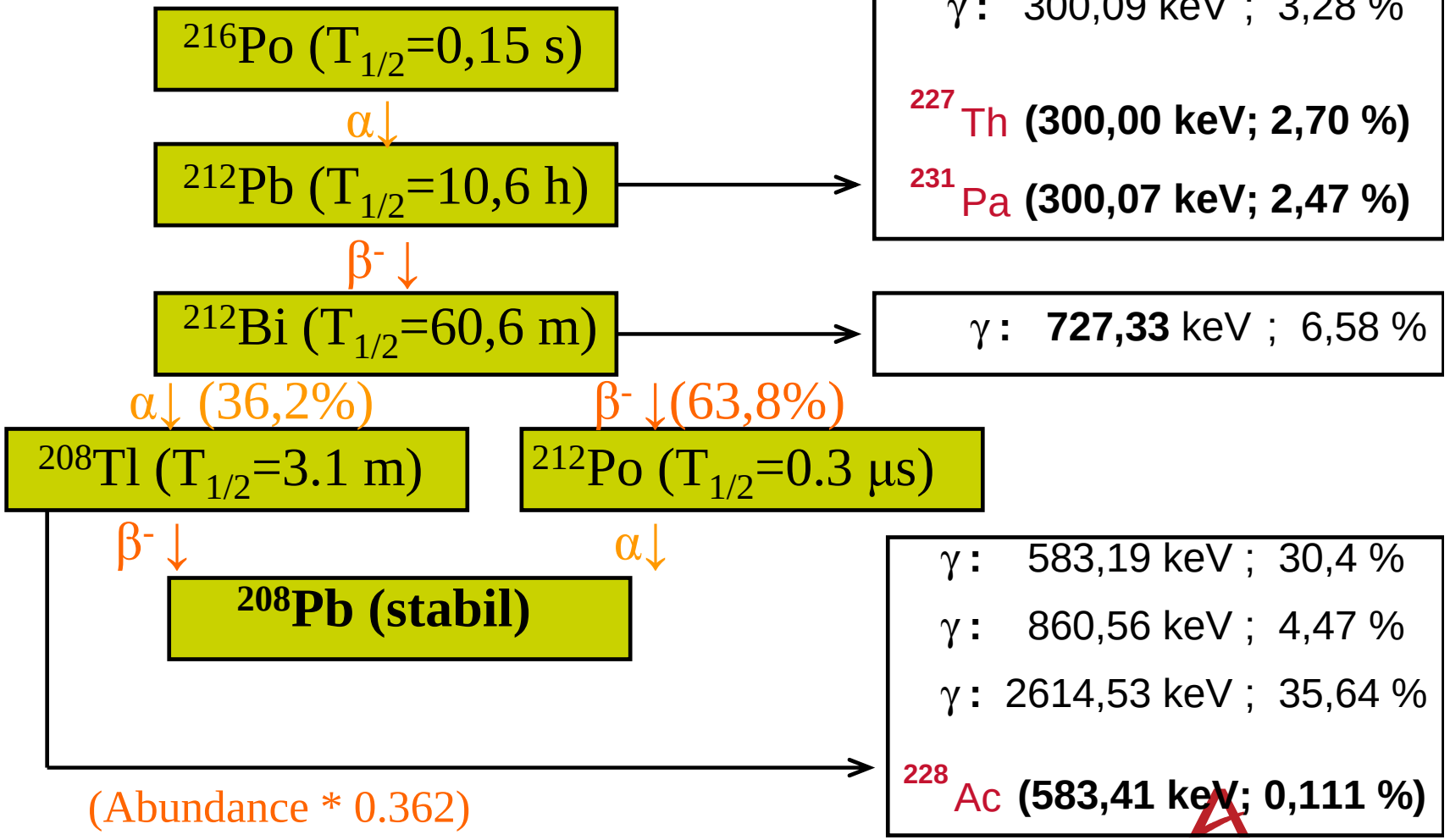
Measurement of ^{238}U, ^{210}Pb and daughters



Measurement of ^{232}Th and daughters



Measurement of ^{232}Th and daughters



Measurement of natural decay chains

Step 1: **Is there a consistent analysis with all 3 decay chains in equilibrium?**

Step 2: **Is there a constant factor between the ^{238}U-chain and the ^{235}U-chain of 21.7**

=> easy interpretation

Nuclide library with all lines correlated to the progenitor!

^{235}U, ^{238}U or ^{232}Th

Abundances and halflives!

Measurement of natural decay chains

Step 3: What are the nuclides that caused the disequilibrium?

Does the disequilibrium comes from:

1- Geochemical processes

2- Effects from processing

3- Gas losses from measurement container

Step 4: How do these disequilibria influence radiation protection

Examples: 1- $^{238}U/^{234}U$-activity Recoil followed by solution in water

2- ^{214}Pb-^{214}Bi different to ^{226}Ra due to emanation

3- ^{210}Pb activity increased by filtering of aerosols or electrostatic effects

=> Exact analysis of peak areas important

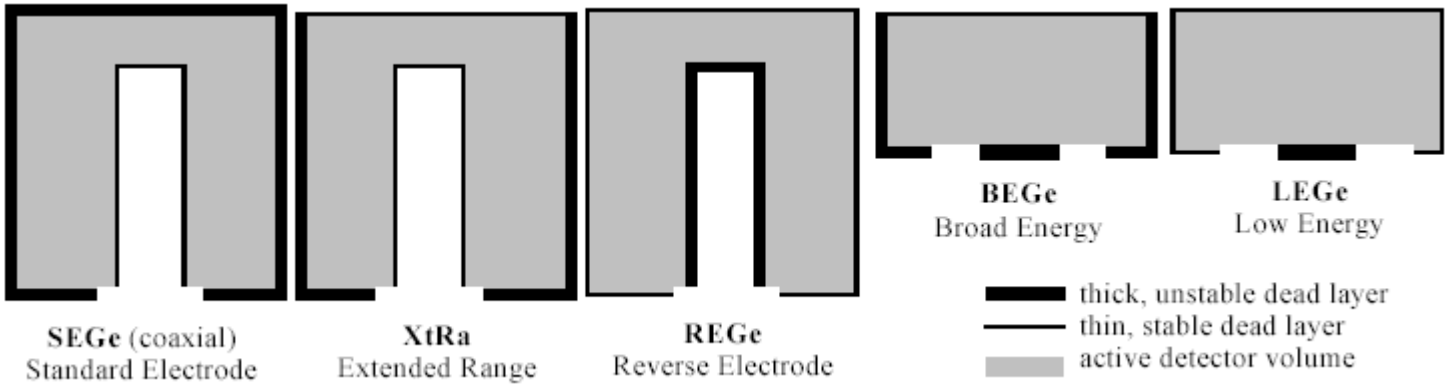
Steps to improve accuracy of net peak areas

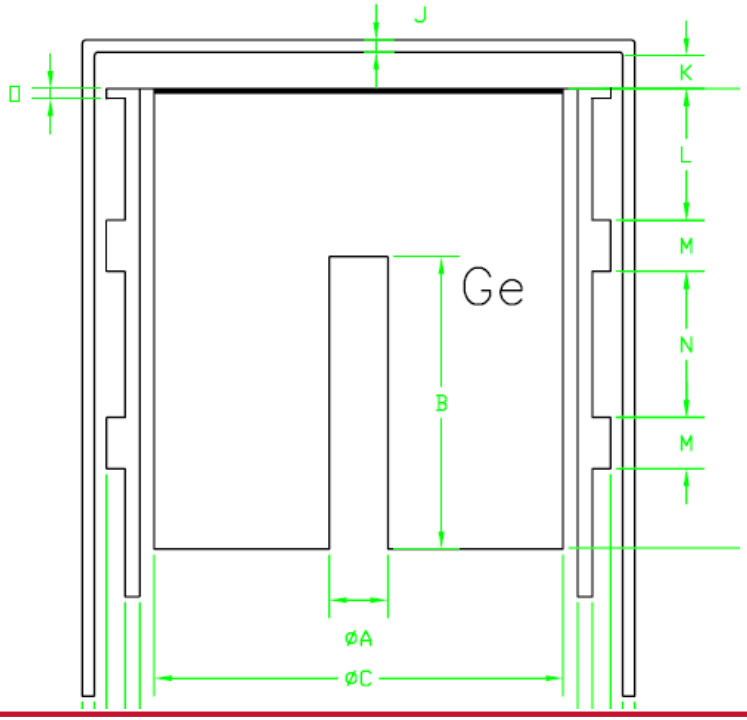
$$A_{N,\,DMin} = 3{,}3 \bullet K_G \bullet \sqrt{R_0 \bullet \left(\frac{1}{t_0} + \frac{1}{t_b}\right)}$$

$$A^{\#} = A^{*} + k_{1-\beta} \cdot u\left(A^{\#}\right)$$

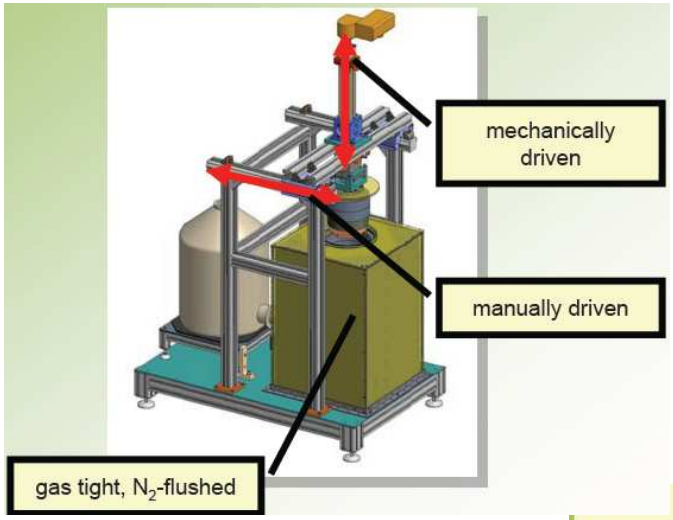
- Background reduction (passive/active shielding)
- Improved efficiency functions and check of results
- Peak fit algorithm

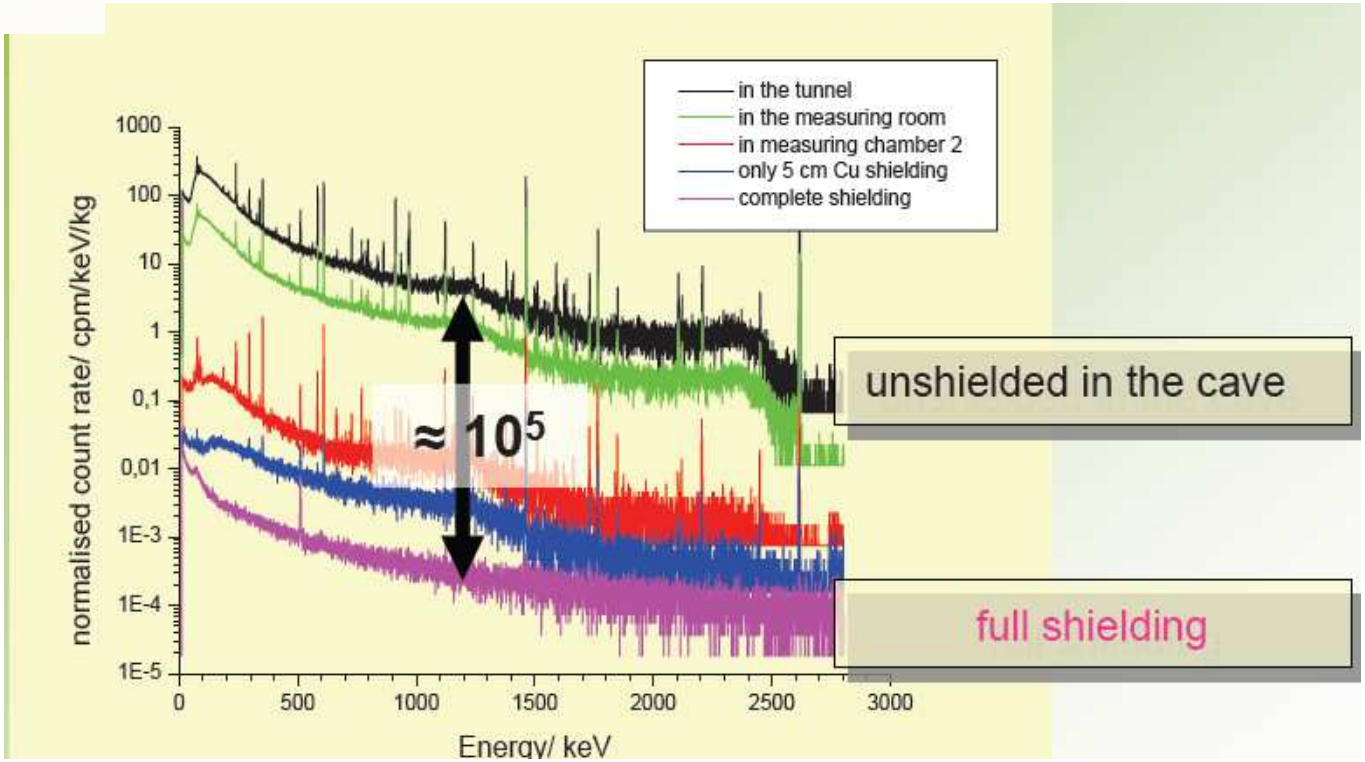
Choice of detector (Calibration factor)





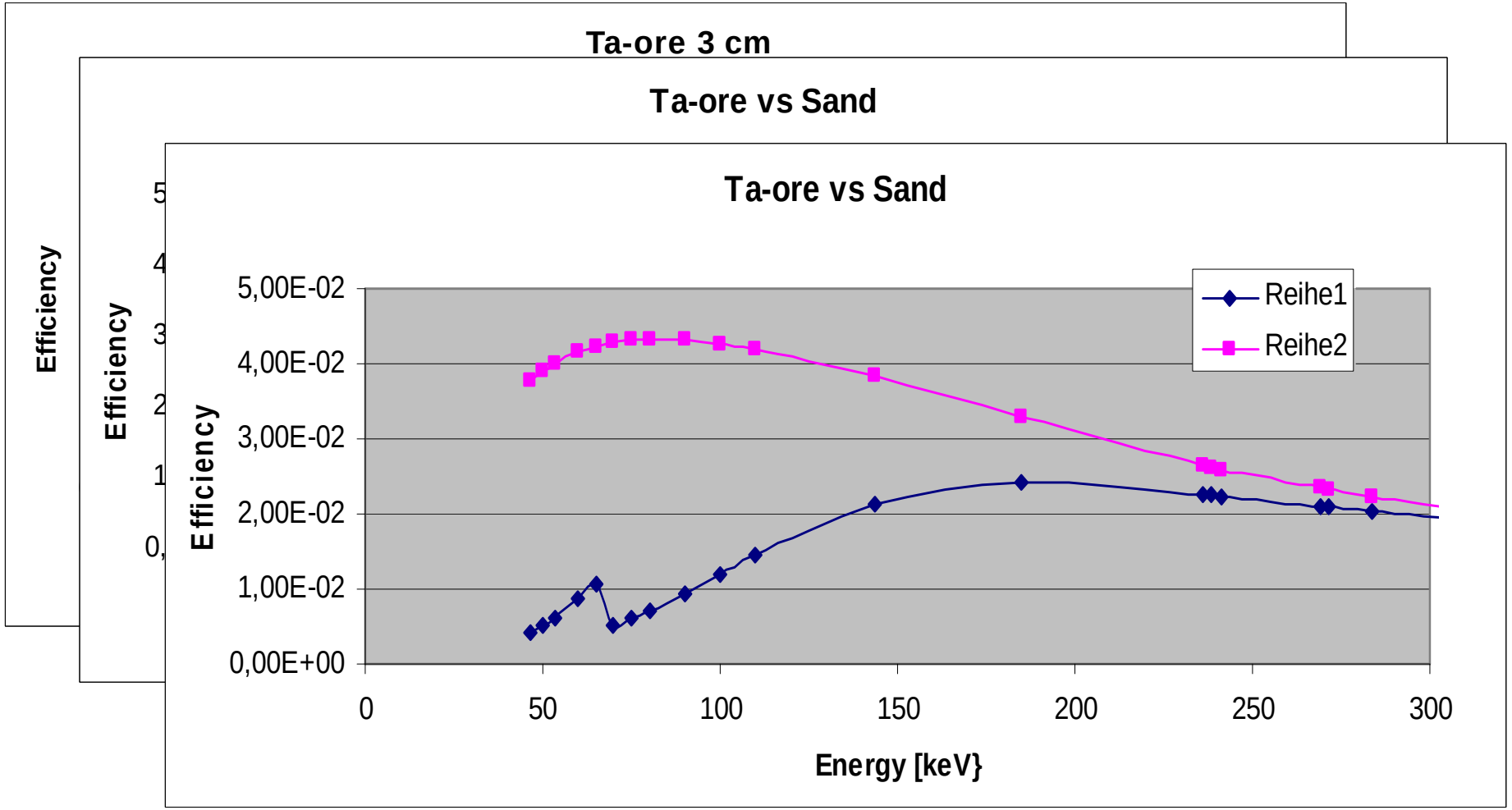
Ultra-Low-Level measurements
Cellar Collaboration





Source: Cellar booklett and priv. Communication M. Köhler

Efficiencies for different samples

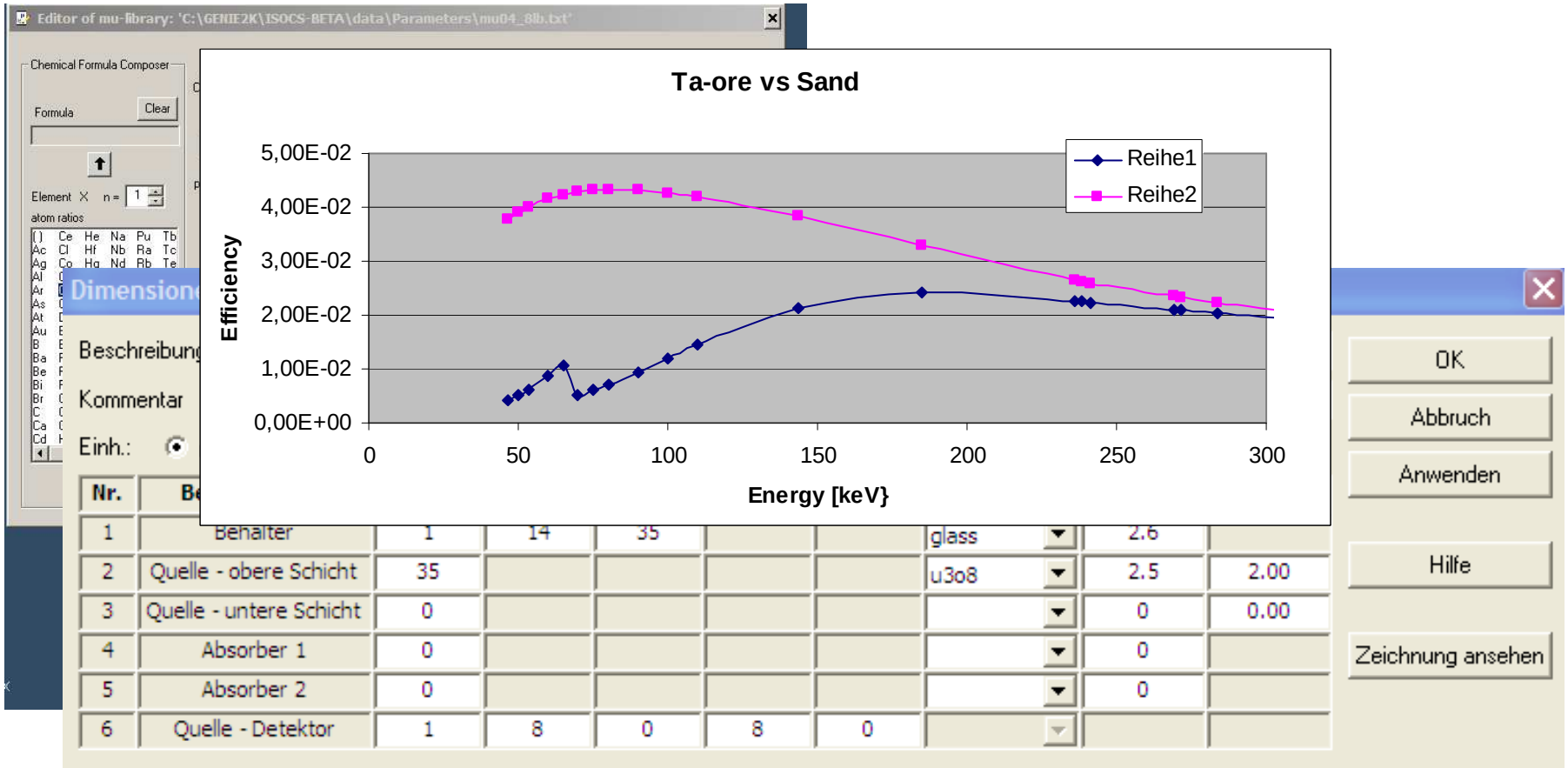


Fundamental basics
x-ray energies of elements

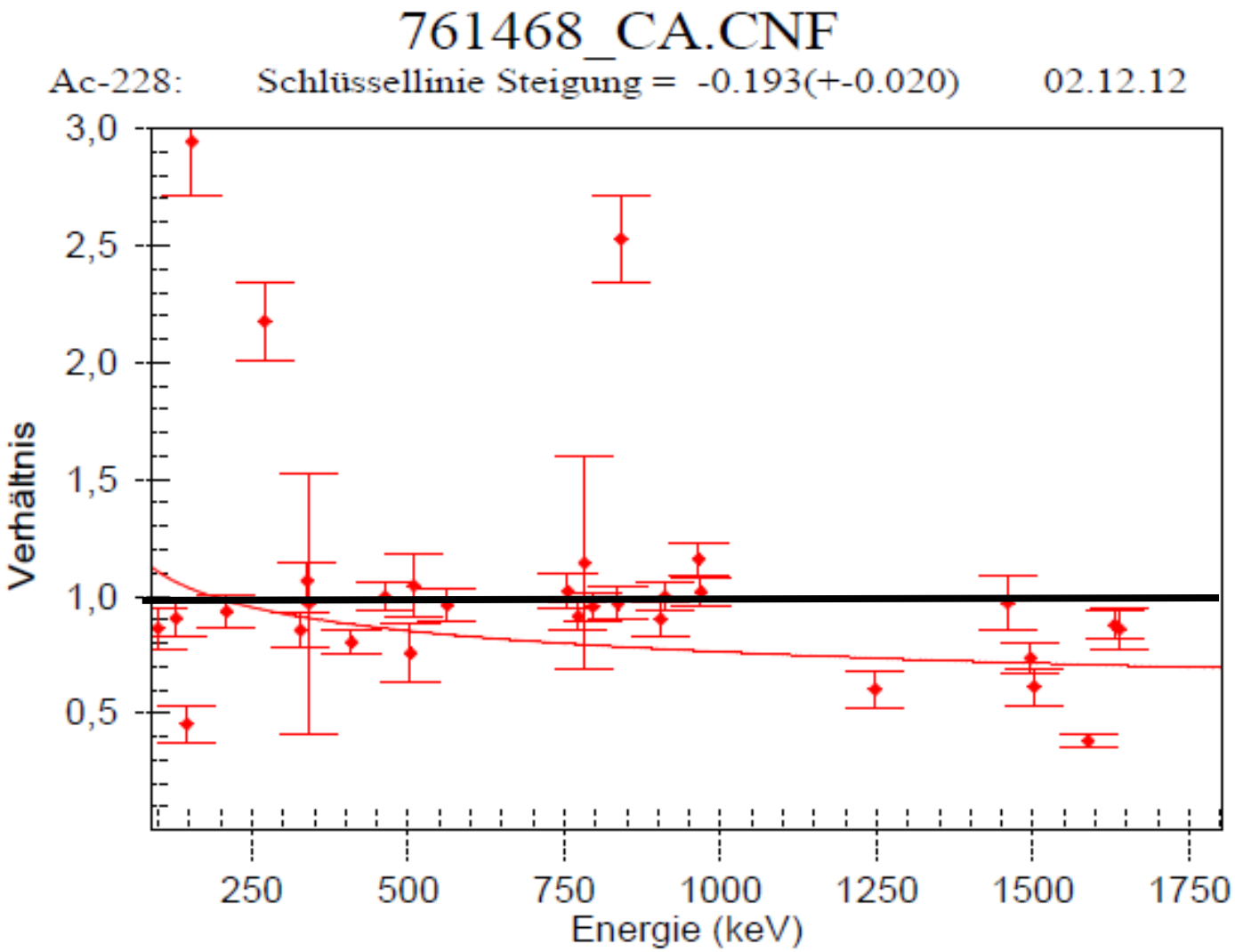
Element	kα1 [eV]	kα2 [eV]	kβ1 [eV]
O	277		
Fe	6.403	6.391	7.058
Ge	9.886	9.855	10.982
Zr	15.775	15.690	17.668
Nb	16.615	16.521	18.622
I	28.612	28.317	32.295
Ta	57.532	56.277	65.223
Hg	70.819	68.895	80.253
Pb	74.969	72.804	84.936

X-ray fluorescence analysis

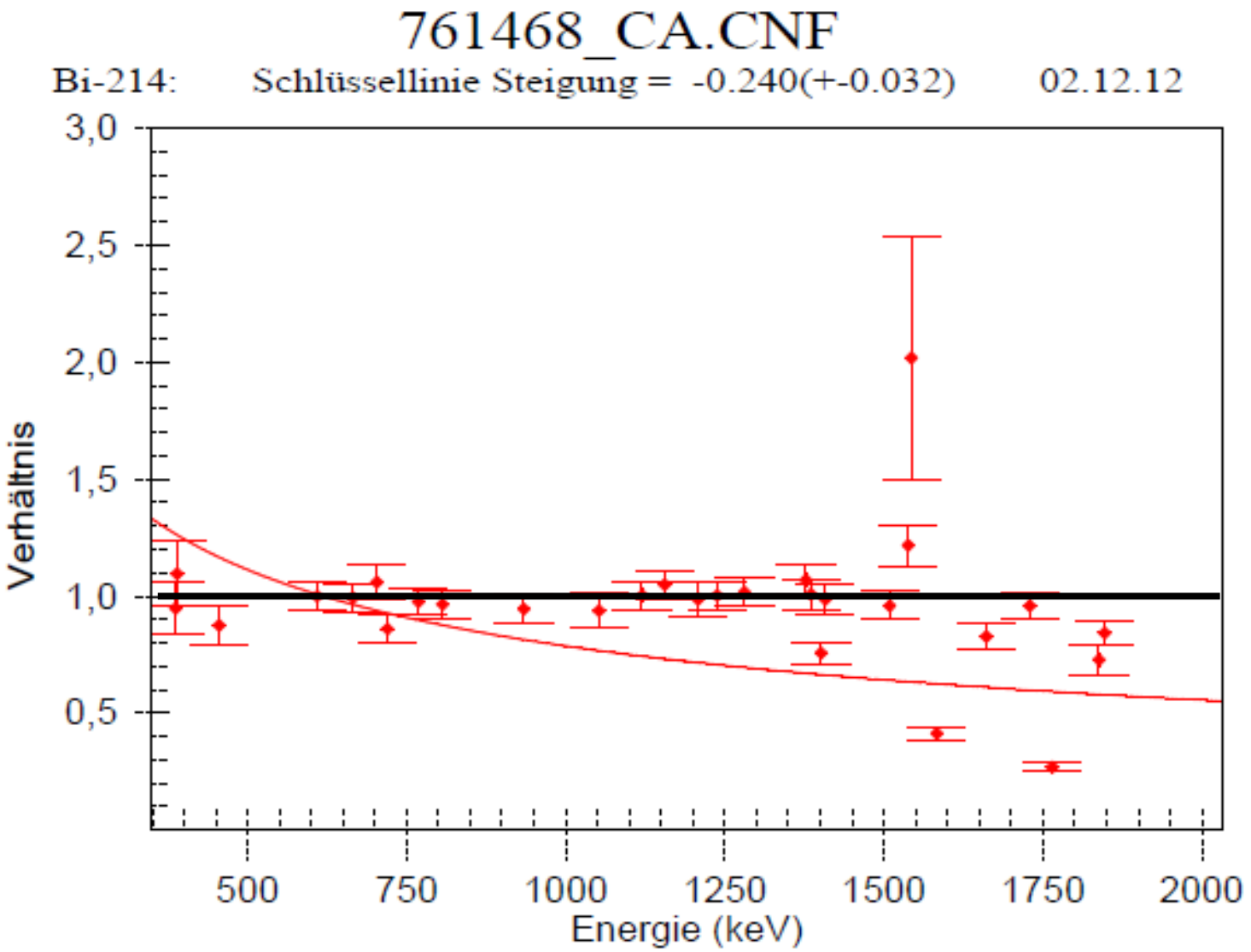
Output of x-ray fluorescence analysis can be used as input for Monte-Carlo modelling of efficiency function



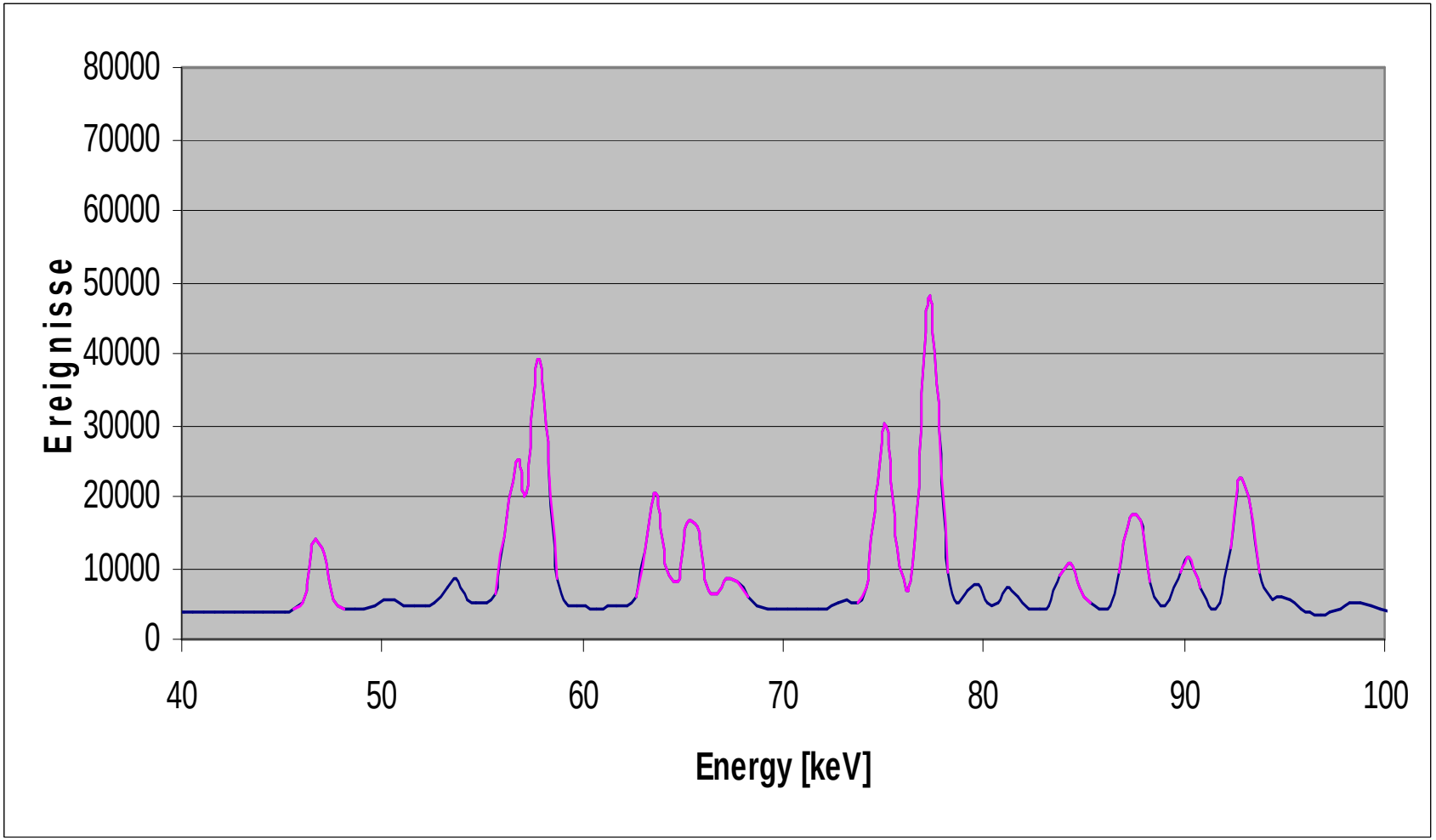
Validation of efficiency function



Validation of efficiency function



Validation of peak fit results



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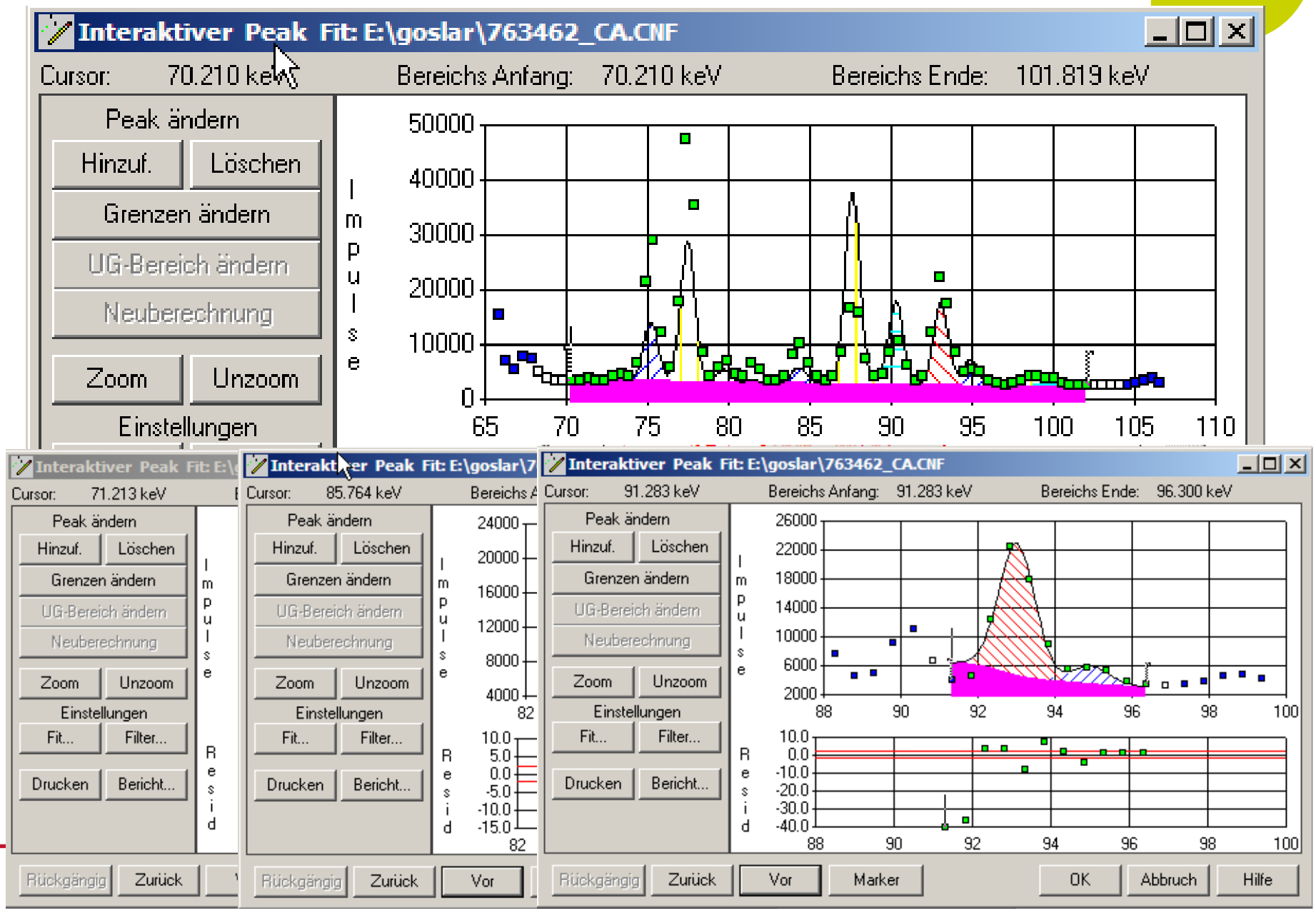

Validation of peak fit results

Energy [keV]	FWHM	Events	Energy [keV]	FWHM	Events	
63.59	0.99	3.31E+004	63.59	1.00	3.37E+004	1
65.44	0.99	2.95E+004	65.45	1.01	3.02E+004	1
67.42	1.00	1.08E+004	67.42	1.01	1.15E+004	1
73.18	1.03	1.38E+003	73.20	0.98	2.48E+003	2
75.10	1.03	2.33E+004	75.10	0.98	5.28E+004	2,2
77.37	1.04	5.57E+004	77.37	0.99	8.48E+004	1,6
79.58	1.04	4.97E+003	79.58	0.99	6.48E+003	1,4
81.36	1.05	9.43E+002	81.35	1.00	6.40E+003	7
84.20	1.05	6.13E+003	84.19	1.00	1.49E+004	2,1
87.48	1.06	7.77E+004	87.48	1.04	3.13E+004	0,5
90.15	1.06	3.39E+004	90.16	1.04	1.41E+004	0,4
92.93	1.07	3.35E+004	92.98	1.10	4.19E+004	1,3
94.75	1.08	1.07E+004	94.90	1.10	5.53E+003	0,5
98.50	1.08	5.11E+003	98.54	1.12	3.95E+003	0,8
99.67	1.08	2.53E+003	99.72	1.12	2.04E+003	0,8

Time evolution of activity

Example of Ra extraction from geothermal facilities

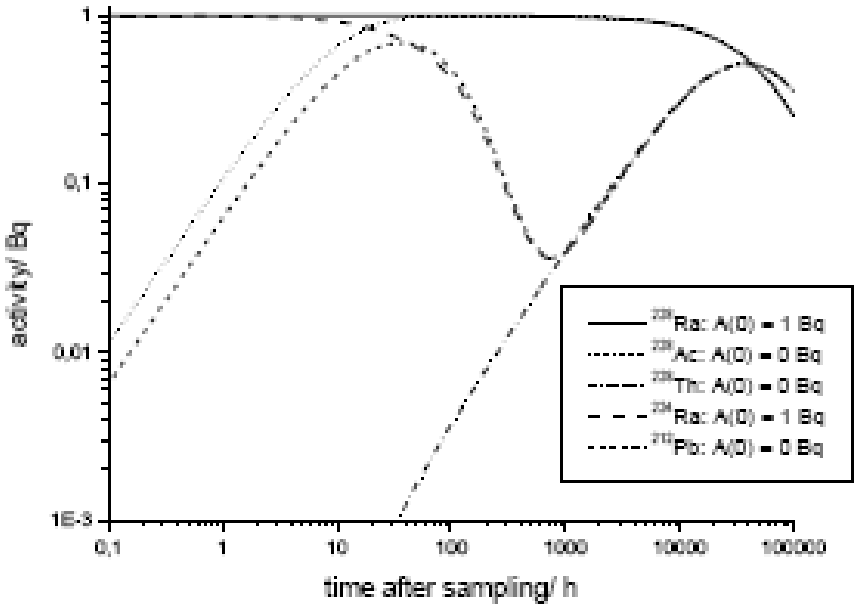

Fig. 2 Simulated time evolution of the system ^{228}Ra ...^{212}Pb from the ^{232}Th decay series containing only unsupported Ra at sampling. An initial activity of 1 Bq was assumed for both Ra isotopes. Note that the scales are logarithmic.

- **Batemann, H. 1910, Solution of a System of Differential Equations Occuring in the Theory of Radioactive Transformations, Proc. Cambridge Philos. Soc. 15, 423-427.**
- **Decay Engine, http://www.nucleonica.net/Application/FullDecay.aspx**
- **Degering, D., Köhler, M., SAAGAS meeting, Sept. 2010**

D. Degering: Private communication from SAAGAS meeting september, 2010

More:..

- True Coincidence
  - γ- γ-coincidence
  - X-ray- γ-coincidence
  - γ-β-coincidence
- Inhomogenities
- …
- Input: treatment of material and sample