Calibration (including TCS) and use of commercial gamma spectrometric software – lessons learned

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Situation

- Home made software in use for routine measurements since 1993, a basic *BASIC* program, orginally written to help the author to understand something in gamma spectrometry.
- Tailored for simple matrices and Cs-137
- Asks the user what is needed, makes basic corrections (for density, variable filling heights, radioactive decay) and writes a summary into a CSV file

Challenges

- The BASIC software ("Greina" = Analyse) cannot cope with many nuclides, complex spectra (e.g. TCS) or matrices
- Working towards accreditation, a commercial solution preferable to a home made one.
- Do not have resources (funding, time) for time consuming calibration work

Solution

- Use GammaVision (which we have for routine acquisition of spectra) and utilise the TCS correction in the program.
- Interesting feature of the GammaVision approach:
  - Instead of requiring many (single gammaline) standards for each geometry, only one is needed.
  - The required standard must contain a mixture of single line radionuclides and multi line with TCS effects.

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Cascade summing in gamma-ray spectrometry in marinelli-beaker geometries: the third efficiency curve

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Abstract

Radionuclides emitting multiple gamma-rays in cascade give rise to summing effects that may be a source of error in the efficiency-curve based interpretation of gamma-ray spectra obtained in highly efficient counting geometries. Correction methods for sources that are small enough for the detector efficiency to be constant over the source volume are well-known. However, in geometries where the detector efficiency is not constant throughout the sample volume, such as Marinelli-beaker geometries, appreciable underestimation of the source activity may still occur if the variation of the efficiencies over the source volume is not accounted for. By introducing a third efficiency curve that accounts for the variation of the detector efficiency over the source volume, we have developed a practical, easy-to-use method that allows for determination of all three efficiency curves from a single, high-resolution gamma-ray spectrum, as well as for accurate correction for cascade summing effects.

Customer: Gammadata Instruments AB / Icelandic Radiation Protection Institute
P.O. No.: 303901, Item 1
Calibration Date: 01-Mar-2008 12:00 EST **Grams of Master Source:** 0.09974

This standard radionuclide source was prepared using aliquots measured gravimetrically from master radionuclide solutions. Calibration and purity were checked using a germanium gamma spectrometer system. At the time of calibration no interfering gamma-ray emitting impurities were detected. The gamma-ray emission rates for the most intense gamma-ray lines are given. Analytics maintains traceability to the National Institute of Standards and Technology through a Measurements Assurance Program as described in USNRC Regulatory Guide 4.15, Revision 1, February, 1979, and compliance with ANSI N42.22-1995, "Traceability of Radioactive Sources to NIST."

Nuclide	Gamma-Ray Energy (keV)	Half-Life, Days	Activity, Bq	Branching Ratio	γps	Uncertainty, % Type u_A	u_B	U
Am-241	59.5	157860	1.093E+04	0.3590	3.923E+03	0.57	1.70	3.59
Cd-109	88.0	462.60	9.909E+04	0.0363	3.597E+03	0.57	1.70	3.59
Co-57	122.1	271.79	2.570E+03	0.8556	2.199E+03	0.34	1.30	2.69
Ce-139	165.9	137.6	3.494E+03	0.7989	2.792E+03	0.35	1.10	2.31
Hg-203	279.2	46.61	8.442E+03	0.8146	6.877E+03	0.40	1.10	2.34
Sn-113	391.7	115.1	5.639E+03	0.6497	3.664E+03	0.42	1.10	2.35
Cs-134	604.7	754.2	1.237E+04	0.9762	1.208E+04	0.70	1.20	2.78
Cs-137	661.7	10983	2.927E+03	0.8510	2.491E+03	0.70	1.20	2.78
Cs-134	795.9	754.2	1.237E+04	0.8553	1.058E+04	0.70	1.20	2.78
Mn-54	834.9	312.1	6.989E+03	0.9998	6.988E+03	0.70	1.20	2.78
Y-88	898.0	106.6	1.292E+04	0.9370	1.211E+04	0.50	1.10	2.42
Zn-65	1115.6	244.3	1.872E+04	0.5060	9.474E+03	0.60	1.10	2.51
Y-88	1836.1	106.6	1.292E+04	0.9920	1.282E+04	0.90	1.10	2.84

* Master Source refers to Analytics' TCC mixture which is calibrated semi-annually.

The Nordic co-operation

- Question to Stefan@GammaData: Which colleagues in the Nordic countries are using the TCS correction in GammaVision?
- Answer: **None** that he was aware of!

- Conclusion:
  - Give the calibration a try,
  - test the results by participating in the 2008 NPL intercomparison (lab # 110 in report)
  - Present results to Nordic colleagues at Gammasem

Results

- The NPL mixture measured was the “GL”; a ‘low-level’ mixture of γ-emitting radionuclides
- Of the 78 laboratories that participated in all the tests, we were amongst the 12 that passed as having all results in agreement.
- It should be noted, however, that although this exercise tests the ability to deal with TCS and some other corrections, it does not test the ability to deal with attenuation and geometrical corrections.

HOW WERE THESE RESULTS OBTAINED?

Ortec references used

- The GammaVision user manual
- Ortec instructions from the Web:
  - GammaVision-32 Version 5.2 How-To Guide: Make Gamma-Ray Measurements Today
    - http://www.ortec-online.com/application-notes/gv_start.pdf
  - A Closer Look at the Calibration Wizard
    - http://www.ortec-online.com/countlab/calwizard.htm
  - Using GammaVision - Creating Libraries with **Nuclide Navigator III** — Simple and Quick!
    - http://www.ortec-online.com/countlab/nn3library.htm

TCC and libraries

The True Coincidence Correction

The true coincidence correction (TCC) is the correction necessary to account for all of the pulses removed from the full-energy peak due to cascade summing. This correction is a simple divisor of the net peak area, that is the net peak area is increased by the correction factor. The correction factor is detector and sample geometry dependent. The correction factor depends on the full-energy efficiency, that is the ability of the detector to detect the total energy of the gamma ray, and the total efficiency, that is the ability of the detector to detect any part of the gamma ray energy. The full-peak efficiency is determined in the efficiency calibration and the total efficiency is determined in the TCC part of **Calibration Wizard.**

Creating Libraries for TCC

Libraries for true coincidence corrections are accomplished using Nuclide Navigator Version III. Creating libraries for a TCC calibration is essentially the same as for any other purpose (the user creates a library that includes the certified nuclides of the calibration source). However, for coincidence summing corrections, the user must include not only information about the certified nuclides, but also comprehensive information about the daughter nuclides of the decay series and the decay path and intensity values of the parent/daughter relationships. Nuclide Navigator Version III simplifies this to a mouse click with the Copy Daughter Nuclides option. Simply choose the certified nuclides and check the Copy Daughter Nuclides option and Nuclide Navigator does the rest! The library can then be used by the Calibration Wizard in GammaVision to correct for cascade summing effects.

Important to note that the **library must be created using the Microsoft Access format**.

Libraries in this format contain far more information on the cascade processes than is in the ordinary basic GV libraries.

For thought:
How can the Nuclide Navigator III library be updated?

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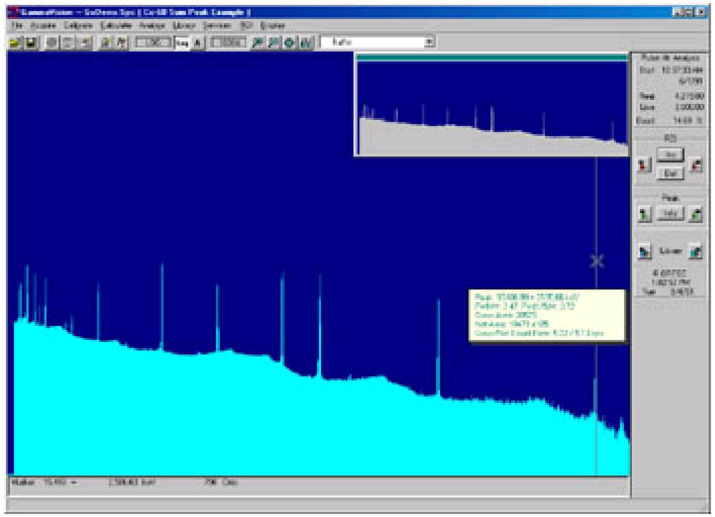
Using GammaVision
Creating Libraries with Nuclide Navigator III — Simple and Quick!

GammaVision reads libraries of gamma-ray energies and yields created by Nuclide Navigator III in Microsoft Access format. The feature simplifies the creation of libraries, including parent-daughter nuclide relationships for True Coincidence Summing (TCS) corrections. The simple steps needed to create comprehensive parent-daughter libraries quickly for TCS calibrations and measurements are illustrated here. First a note about the True Coincidence Summing problem.

True Coincidence Summing



With radioactive decay, atomic nuclei transition from an excited state to a lower energy state. In many instances, gamma-radiation is released during this transition. The release of gamma rays results in a familiar gamma-ray spectrum. The transition is often a single gamma ray. But two or more gamma ray transitions could be involved[1]. These multiple gamma rays from a single nuclide are said to be in cascade if they are emitted at roughly the same time — and are thereby detected at the same time. In a typical gamma ray spectrum, the result is the gamma rays appear as a gamma peak of energy equal to the sum of the two energies. This phenomenon is known as **True Coincident Summing (TCS) or Cascade Summing (CS)**. The summation of

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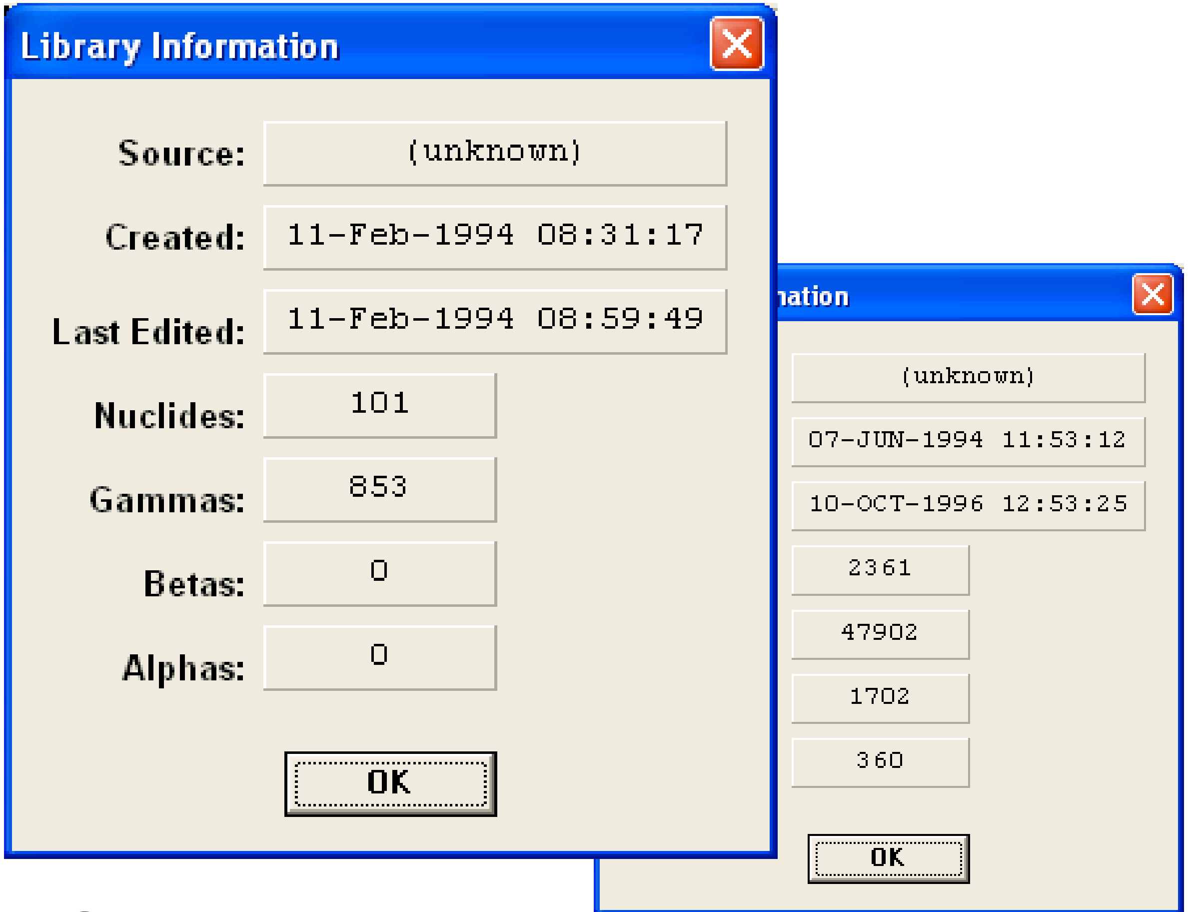
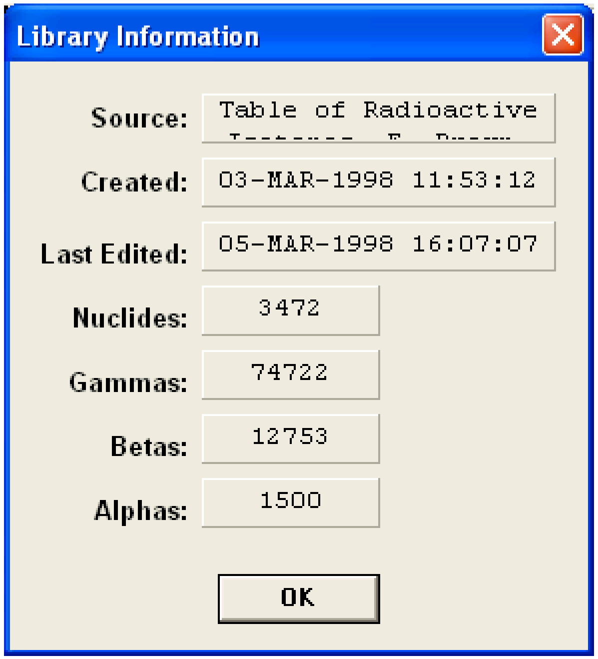
Well known nuclide libraries included



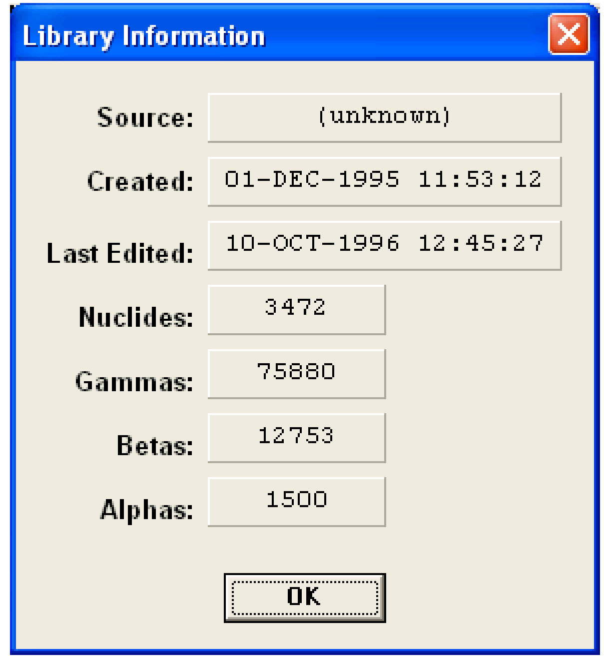
Suspect

Master library

Larger libraries are also included

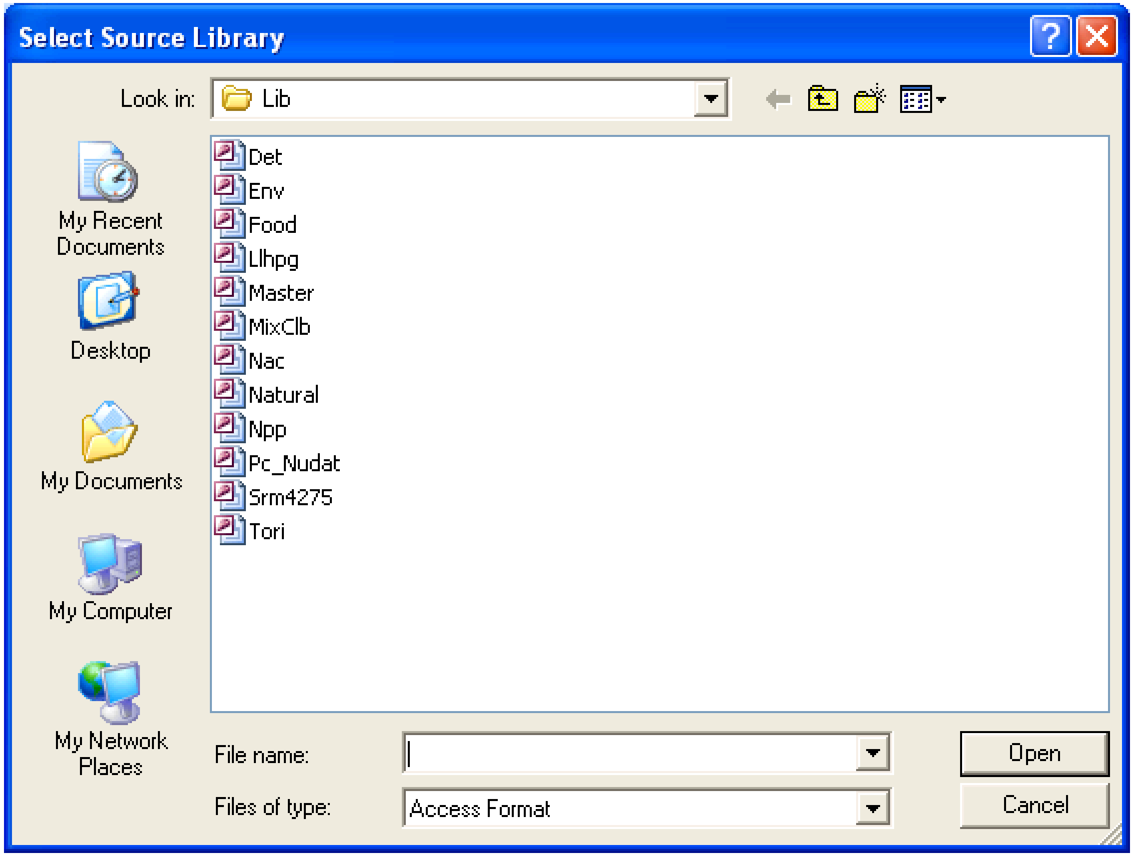


Tori



PC-NuDat

In fact many special libraries can be found



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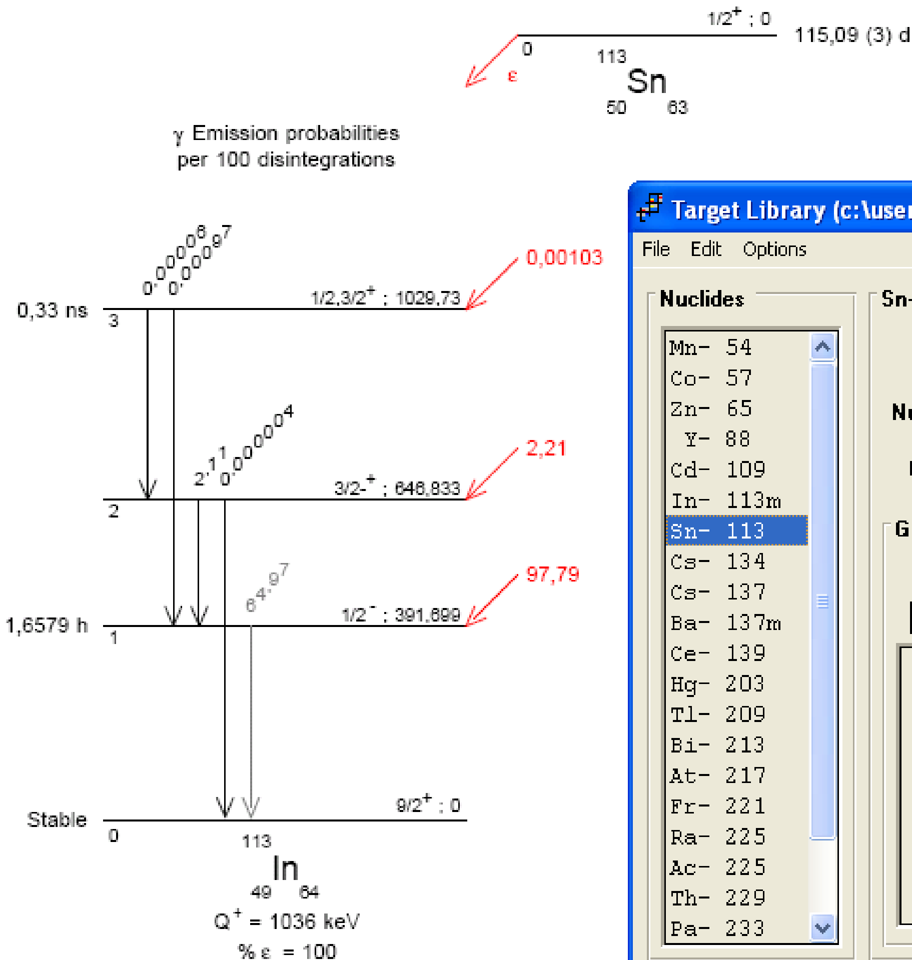
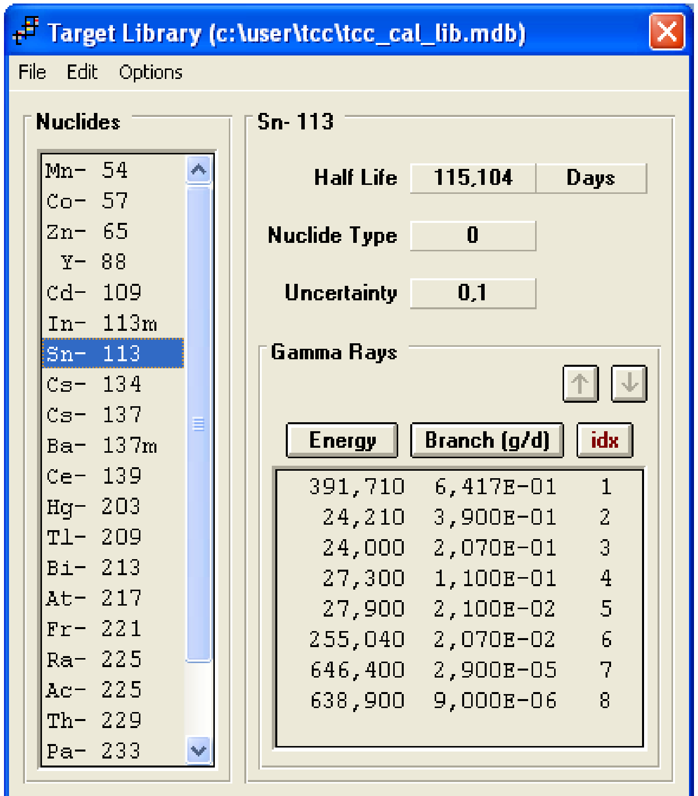
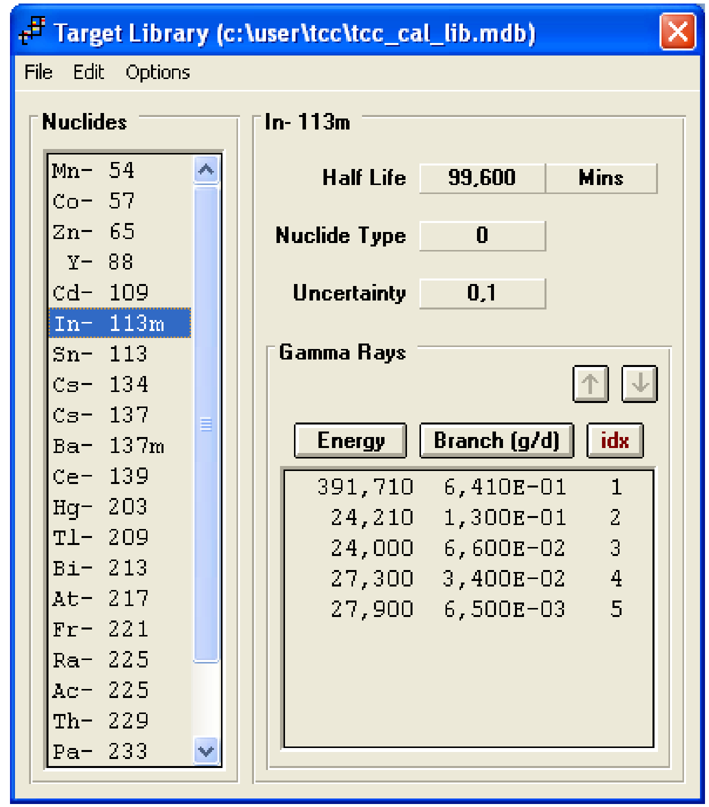
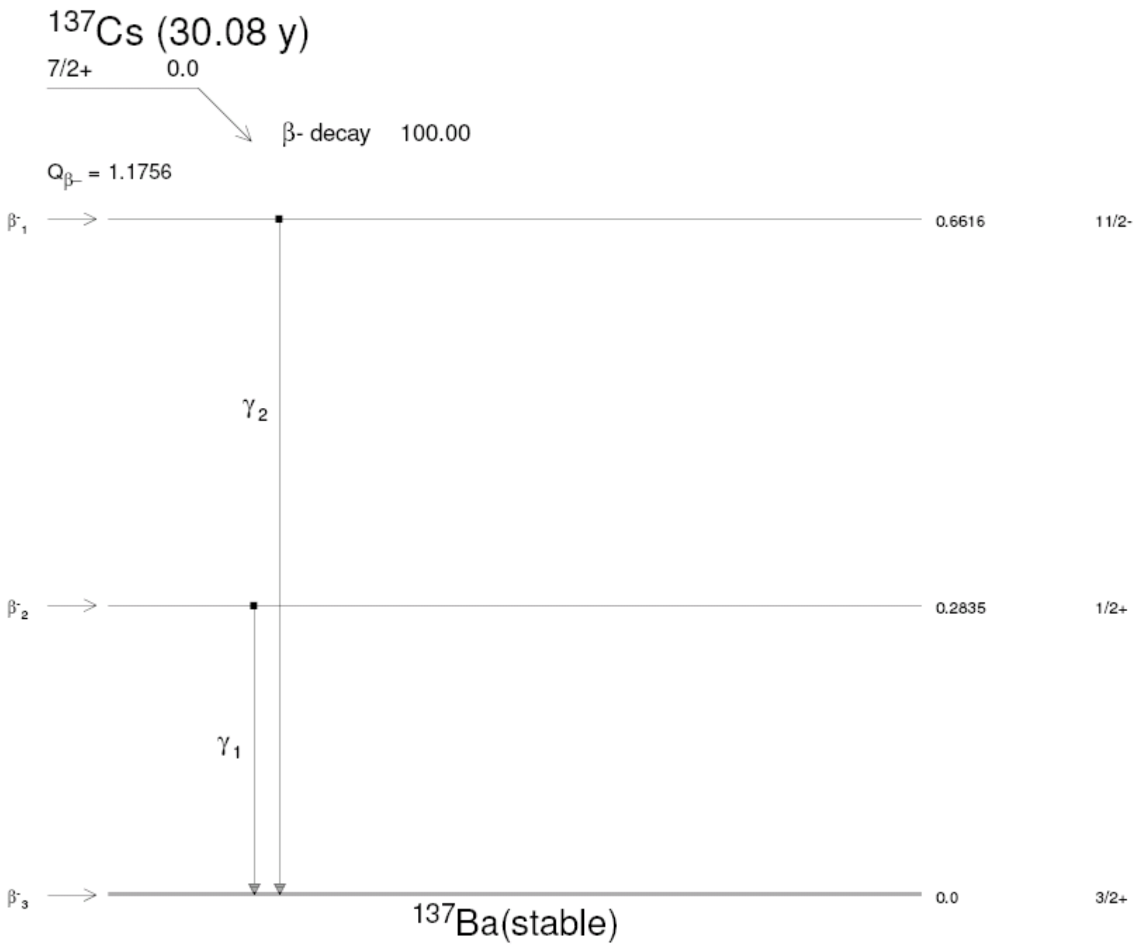
Note: Gamma lines of meta stable daughters often also listed under parent radionuclide







Measuring Cs-137 or Ba-137m ?



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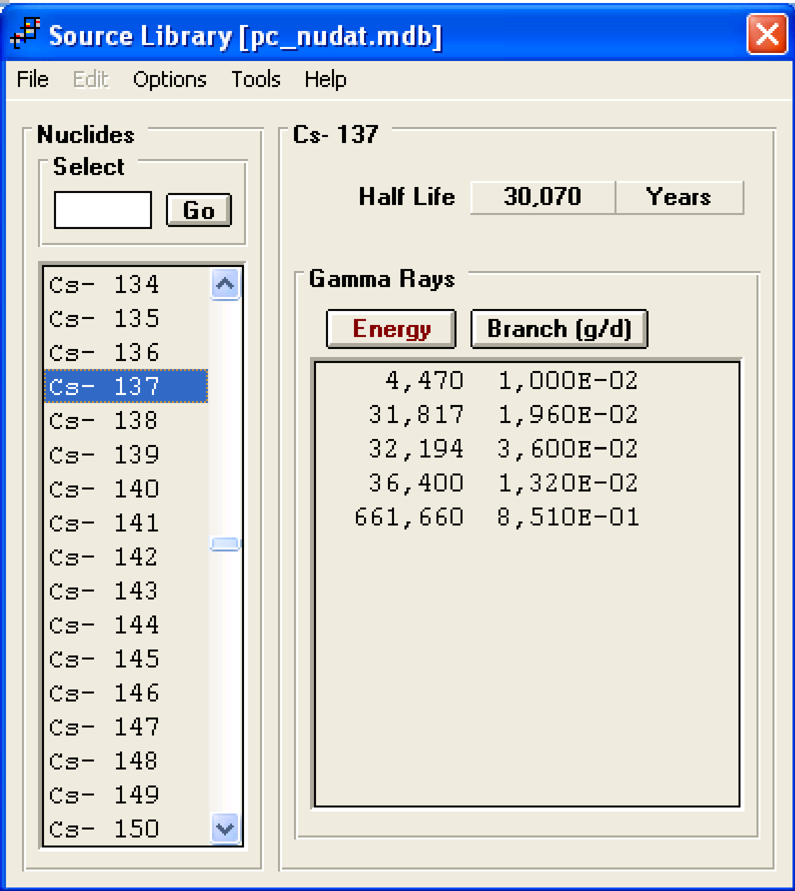
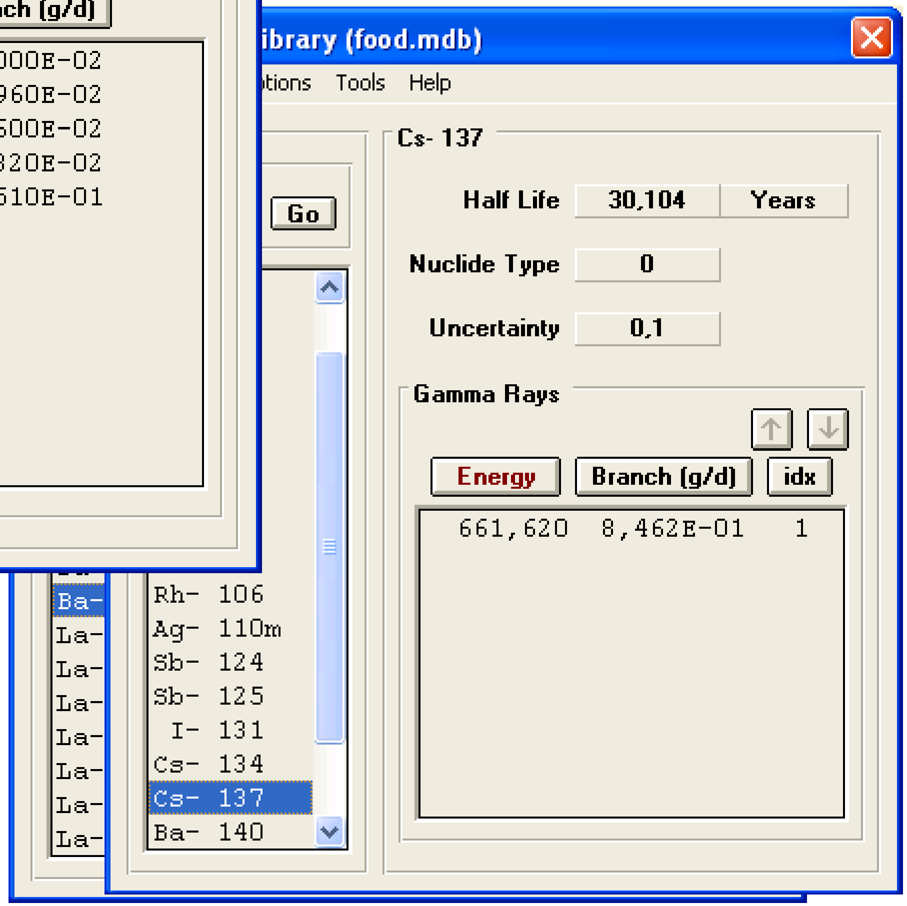
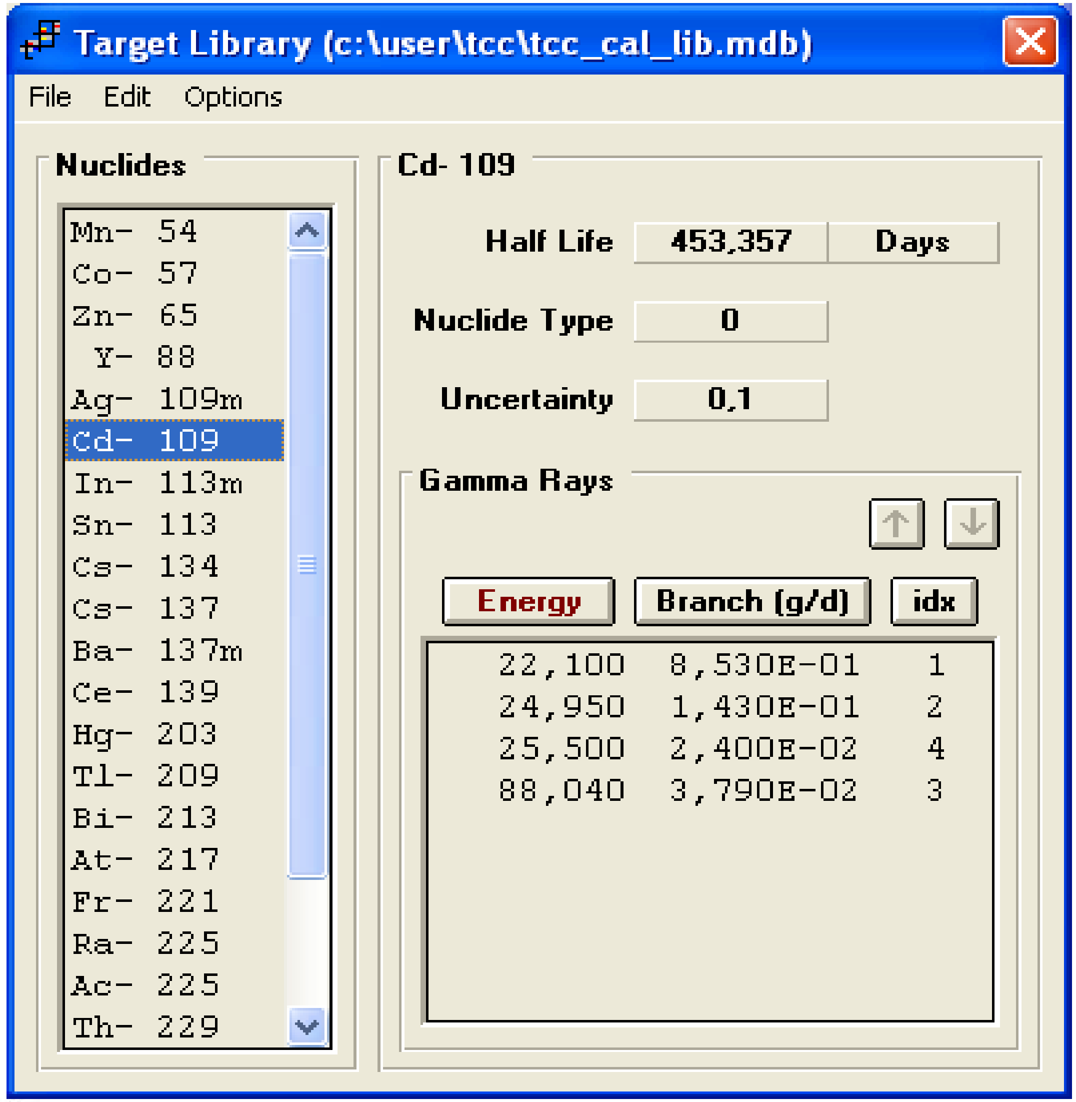
Numbers of lines listed can vary between libraries

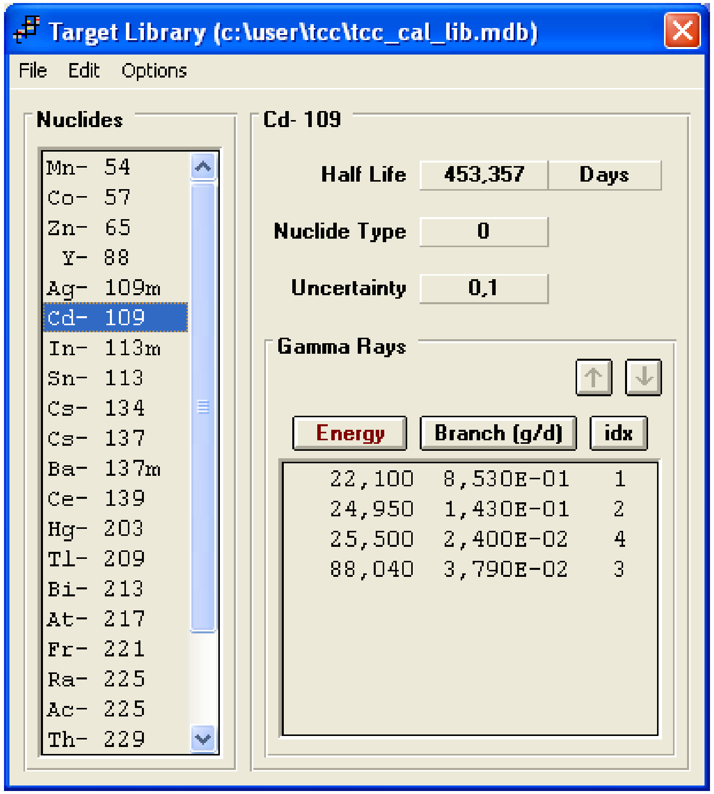


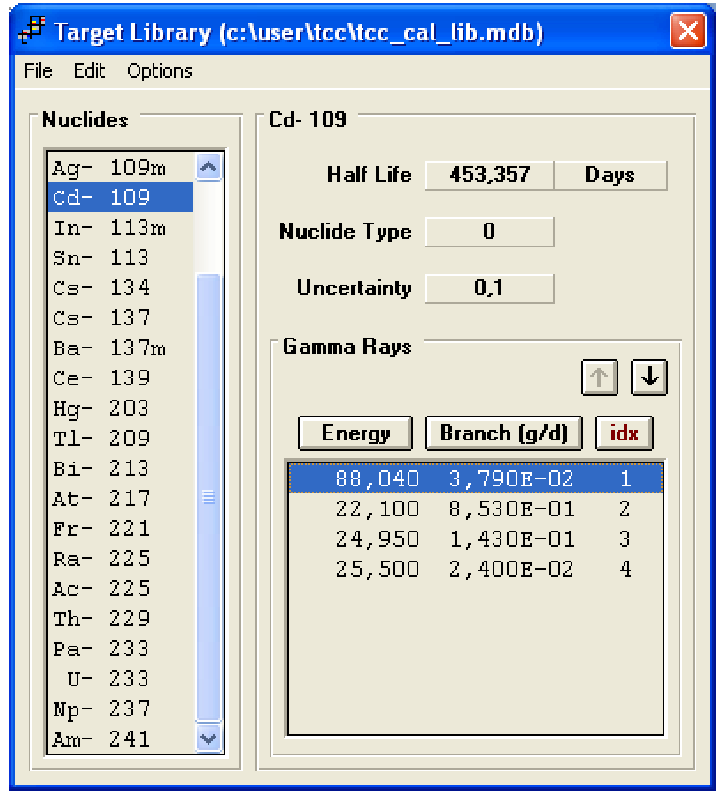


Originally Cd-109 was not found



Order of line is important
GV uses the first for identification





An invisible low energy line in the 1st place in a library will result in that the radionuclide will not be identified, no matter how strong the other lines may be.

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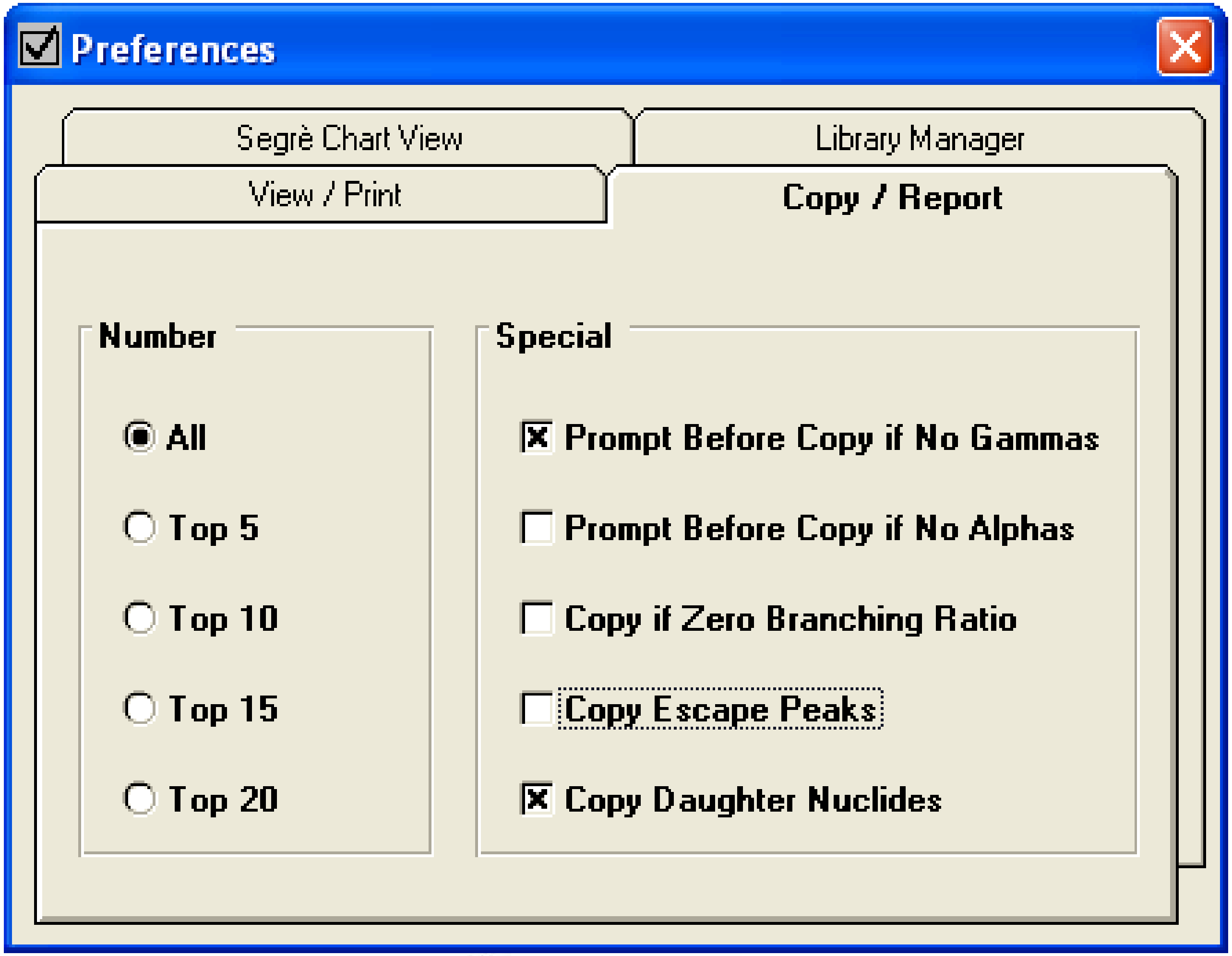
Recommend procedure for creating a library



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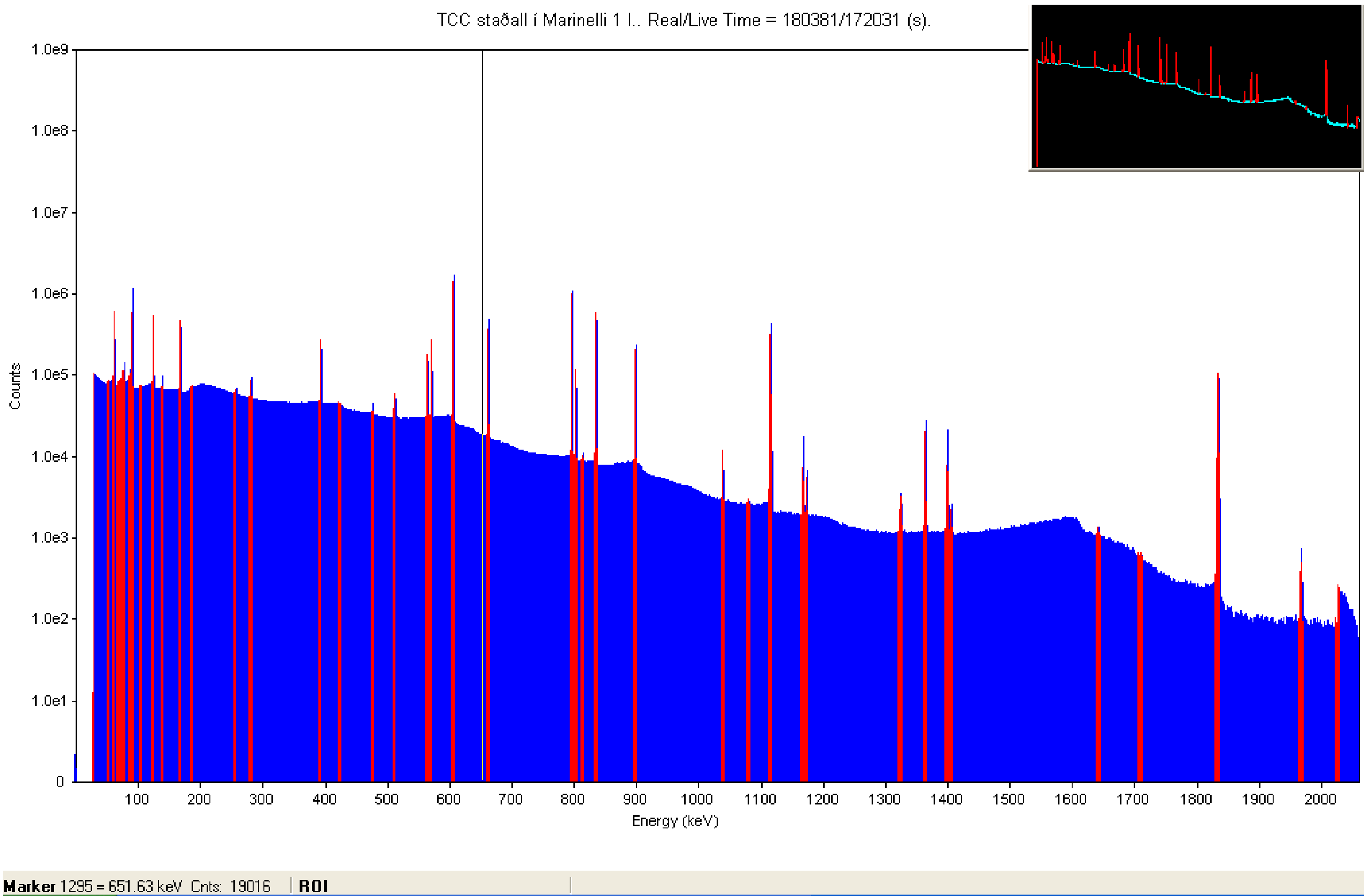
- Metastable daugher nuclides removed
- For each radionuclide lines removed with:
  - energy below Am-241 (ca. 60 keV)
  - lines with low probability (< 0.1%)
- At-217 (with no gamma listed) deleted.
- Np-237 removed (caused interference with Cd-109 at 88 keV)

TCC calibration standard



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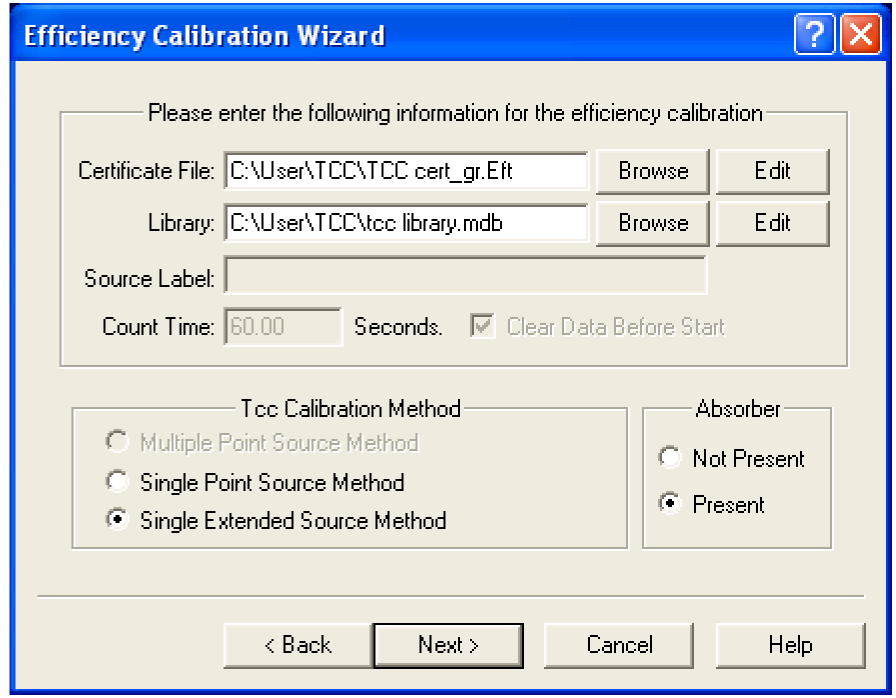
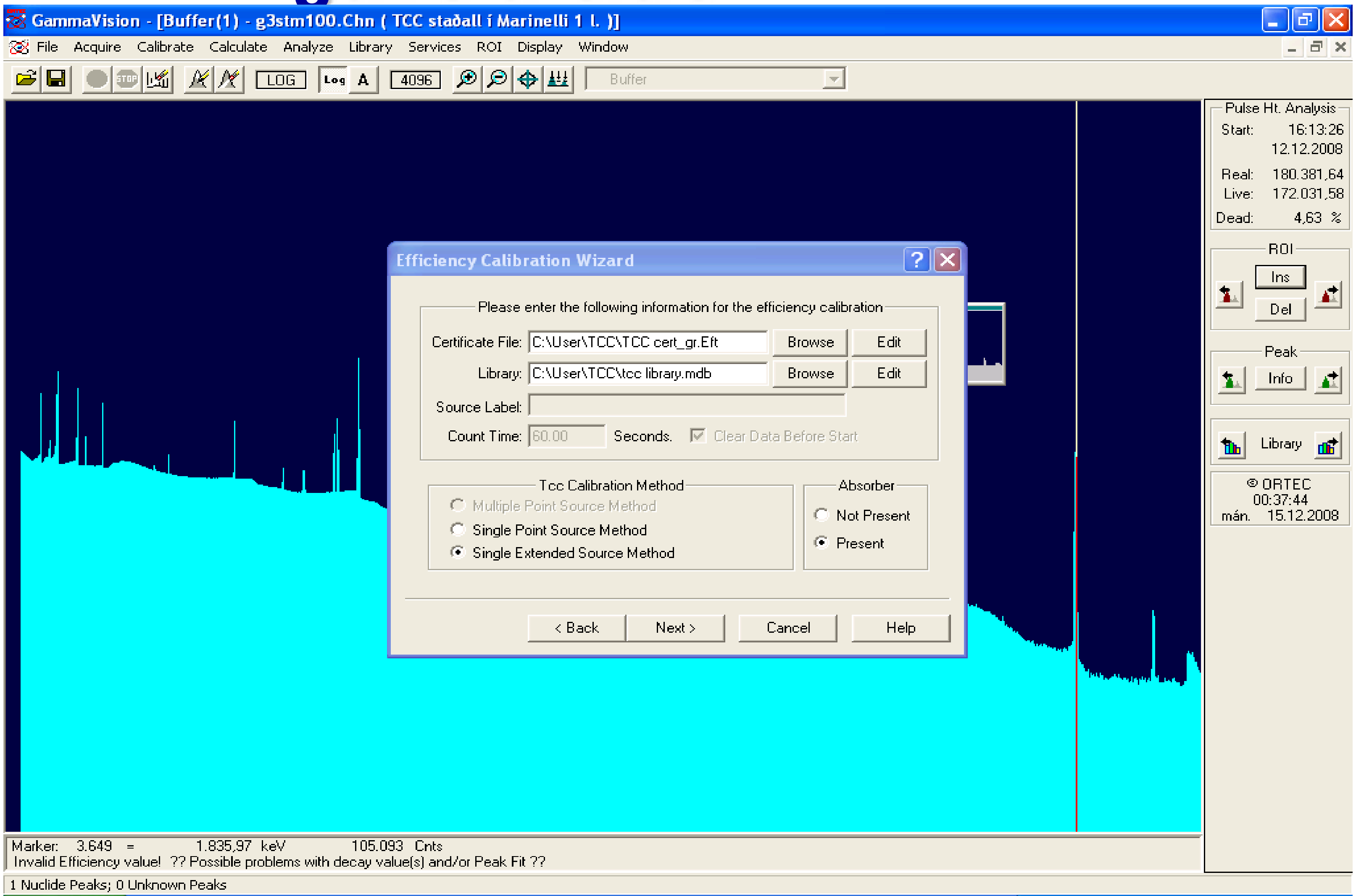
Calibration came to a polite but sudden end, even though all instructions had been followed



Strange behaviour but no crash if “Absorber Not Present”

The last screenshot before the crash gave a clue



Something wrong with decay calculations or treatment of time?

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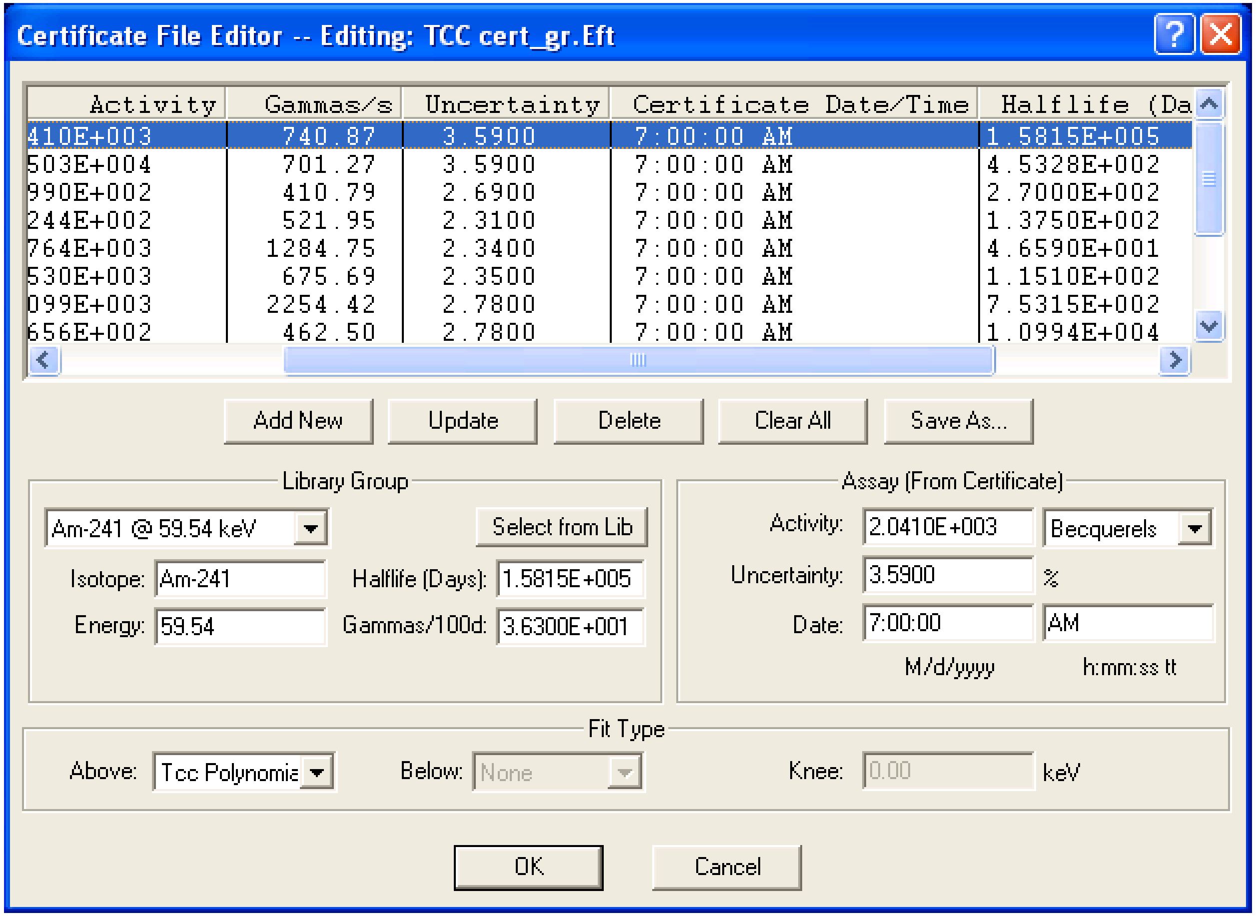
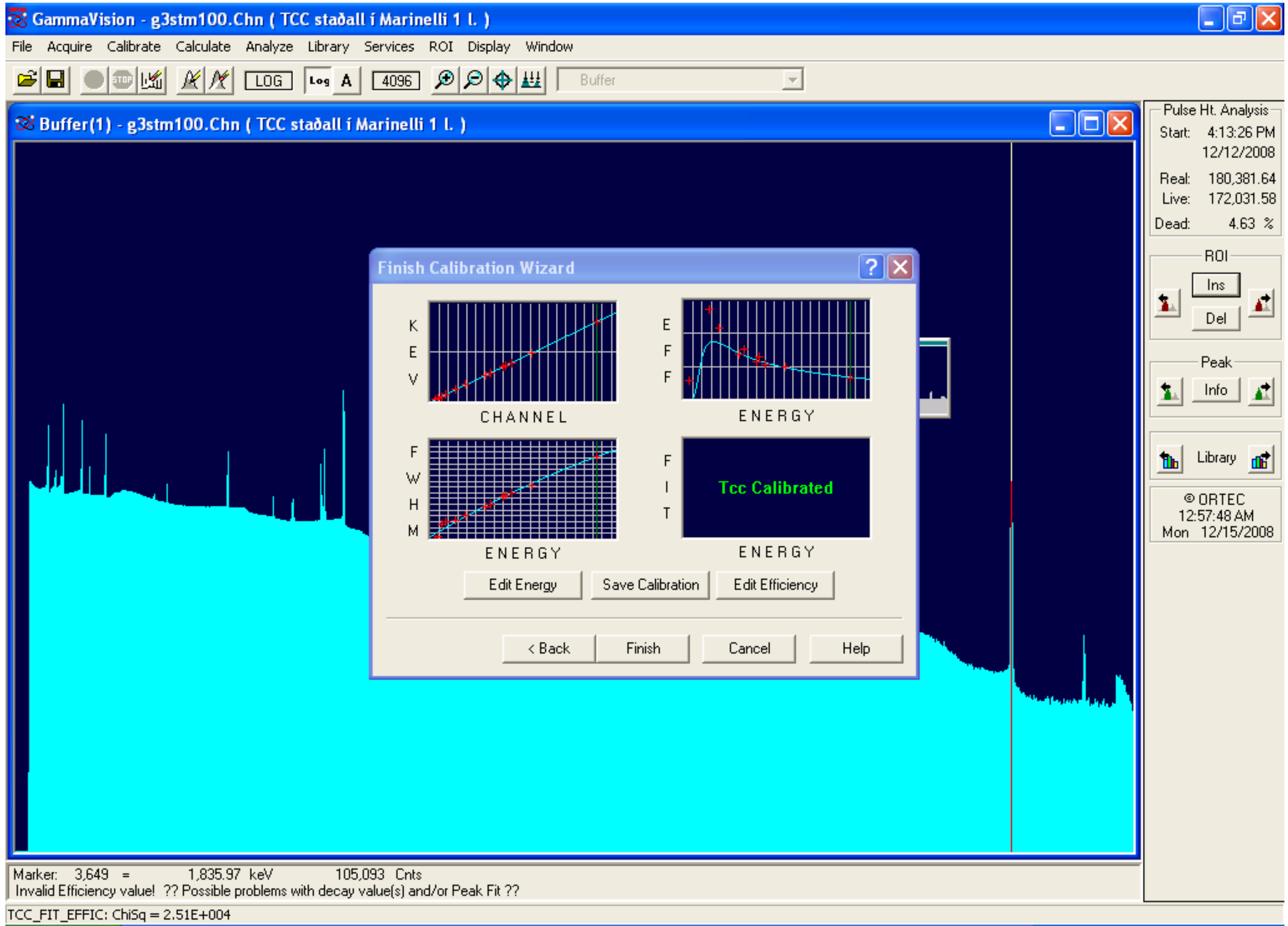


Calibration AFTER configuring the computer as a US computer
Time calculations in correct format

Using the “**Absorber present**” option as recommended did now not cause a crash, but a poor efficiency fit and very high Chi sq value at bottom of screen



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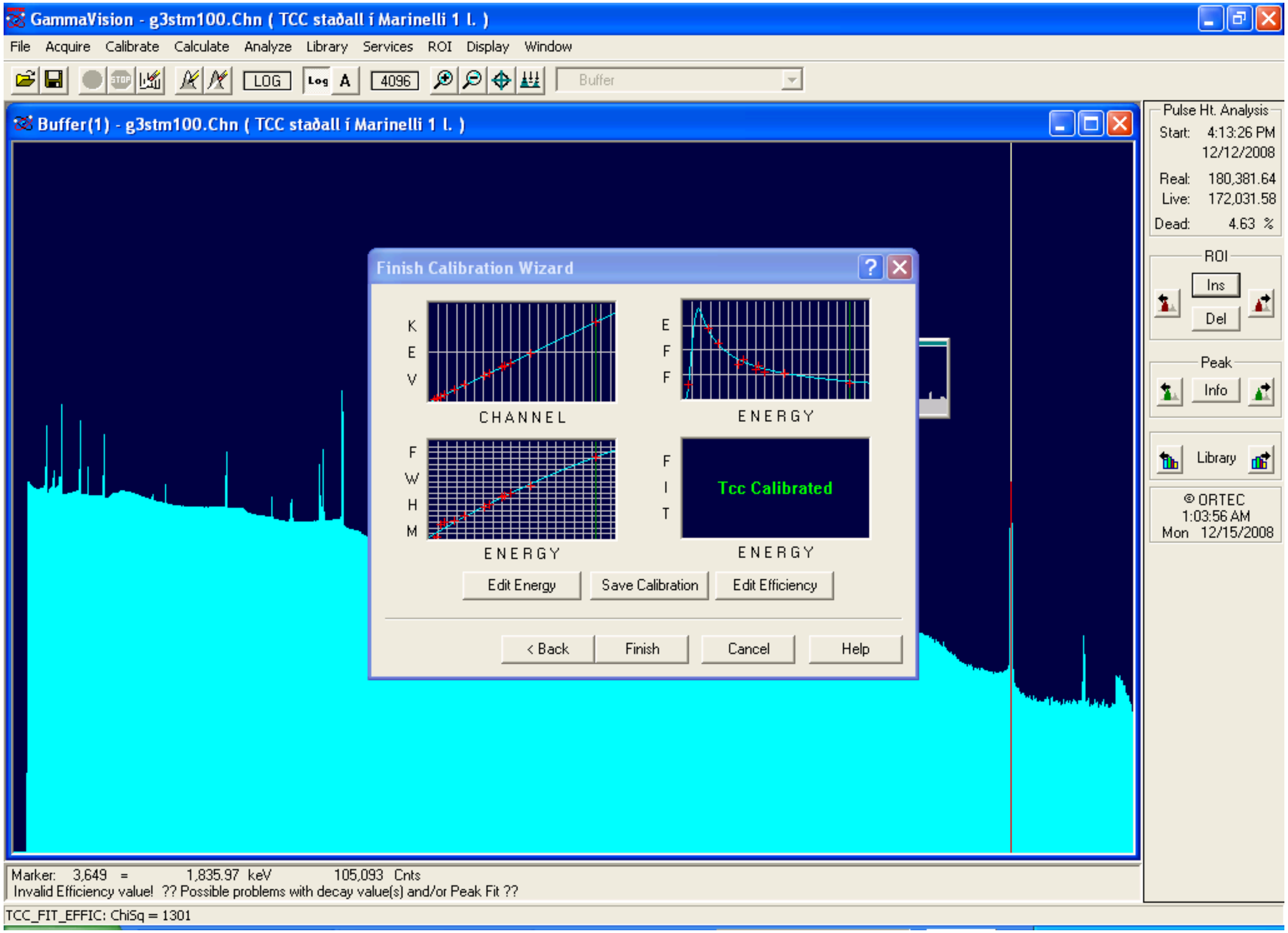
Rerun with "**Absorber Not Present**", much better fit and a lower Chi Sq. value



Note on the engines in GammaVision

- It is not consistent if decay correction is applied or not in intermediate results, for some engines it is applied, for others not.

MEASUREMENT OF NPL COMPARISON SAMPLE

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The NPL comparison exericse

The Gamma Low (GL) sample was best suited to our needs

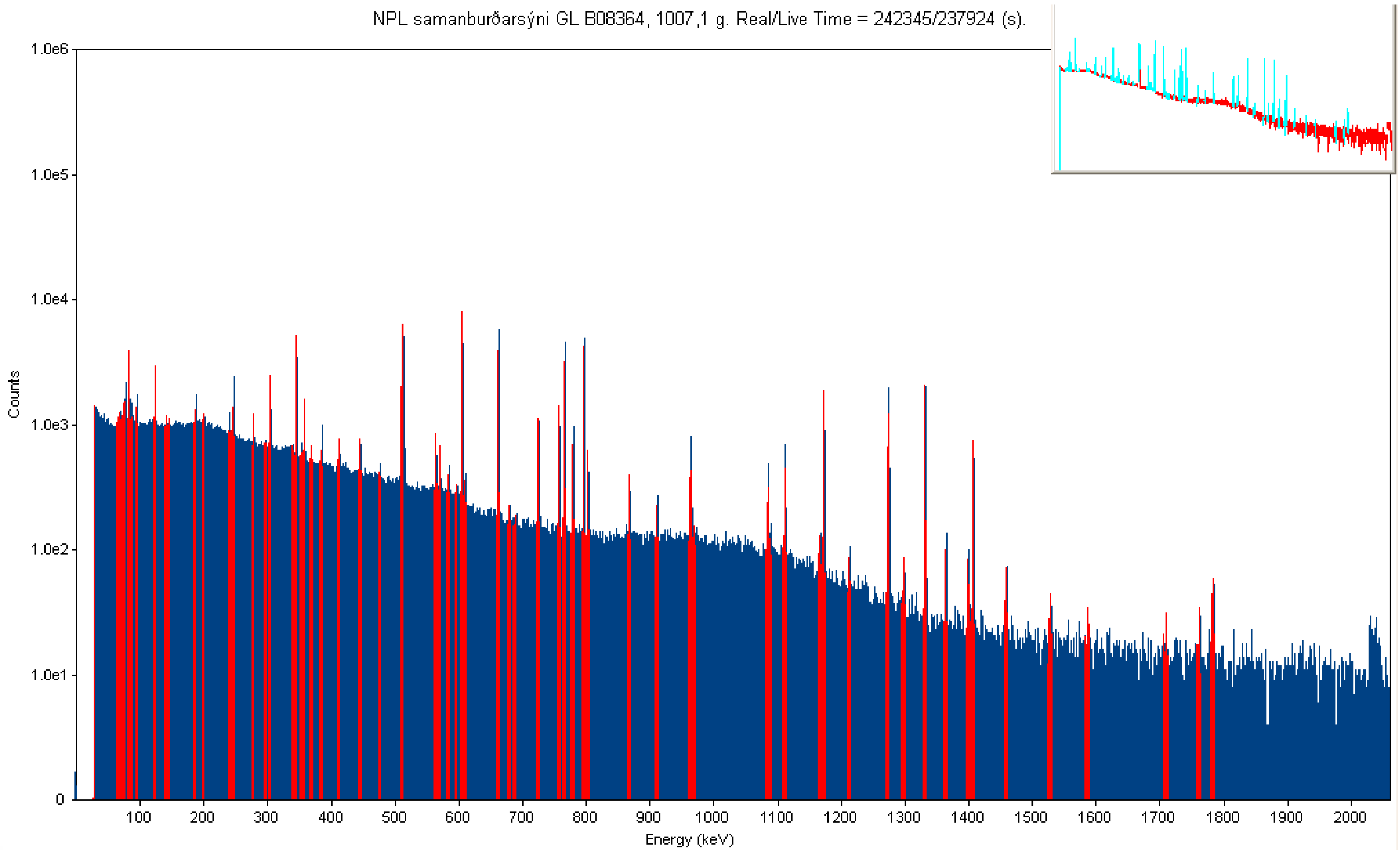
Gamma High (GH):	100 g aqueous sample in HDPE bottle, nuclides 1-20 Bq/g
Gamma Low (GL):	500 g aqueous sample in HDPE bottle, nuclides 1-20 Bq/kg

Mixture of at least eight γ-emitting radionuclides from the following candidate list:

^{7}Be, ^{22}Na, ^{40}K, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{56}Co, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{85}Sr, ^{88}Y, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{109}Cd, ^{110m}Ag, ^{111}Ag, ^{113}Sn, ^{123m}Te, ^{124}Sb, ^{125}Sb, ^{125}I, ^{129}I, ^{134}Cs, ^{137}Cs, ^{133}Ba, ^{140}Ba, ^{139}Ce, ^{141}Ce, ^{144}Ce, ^{147}Nd, ^{152}Eu, ^{154}Eu, ^{155}Eu, ^{153}Gd, ^{160}Tb, ^{166m}Ho, ^{170}Tm, ^{192}Ir, ^{203}Hg and ^{207}Bi

46 possible nuclides (+ daughters?)

Spectrum of NPL GL sample



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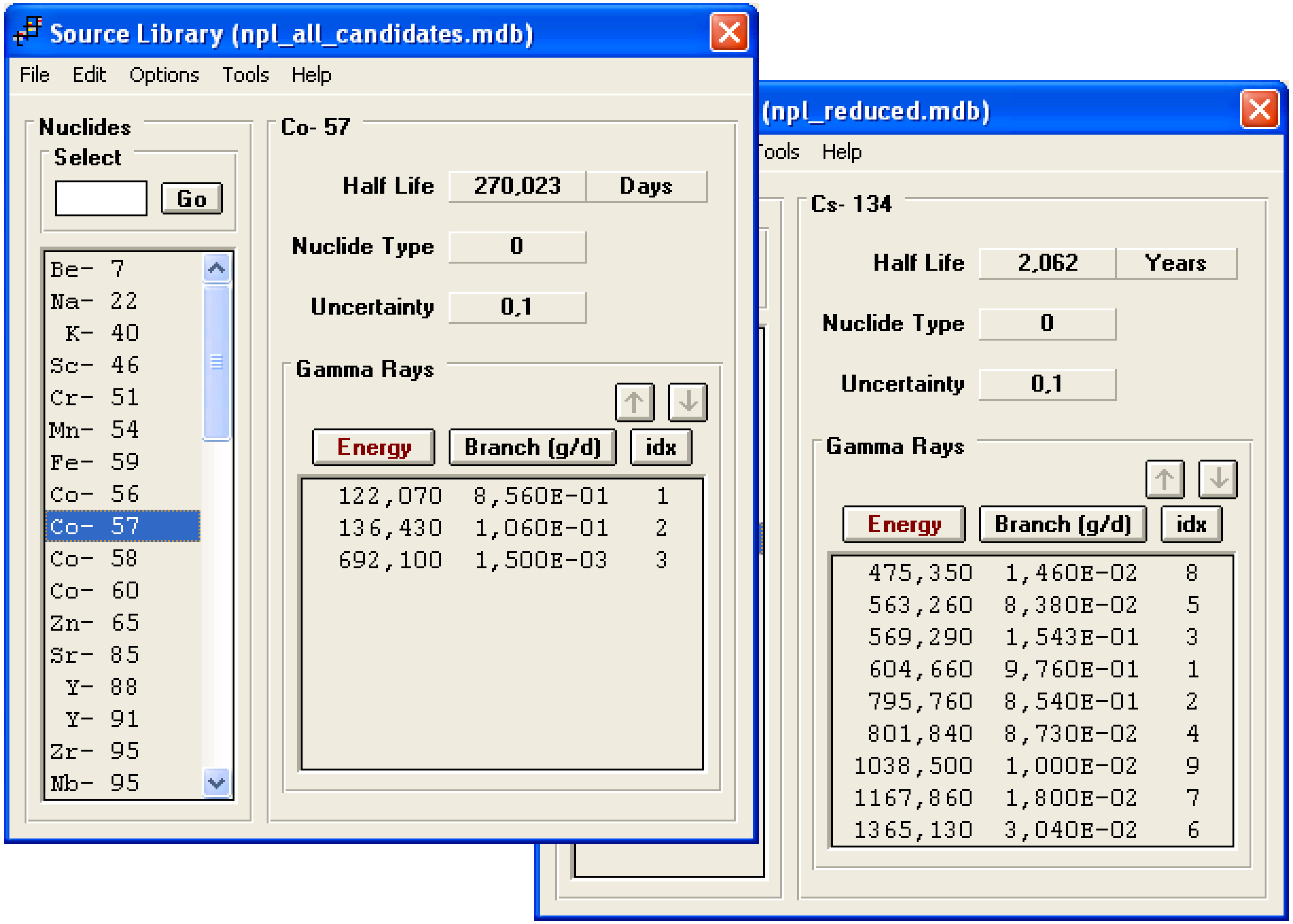
NPL sample - identification

- Create a library with these 46 nuclides + possible short lived daughters.
- Remove gammalines that will not be used
- Measure a standard before and after measuring the NPL sample to have as exact energy (+ FWHM) calibration as possible
- Analyse the sample, use peaked background subtraction
- Check which nuclides are seen in sample, remember each should be at least 1 Bq. It should now be possible to explain all lines.
- Watch out for short-lived radionuclides!

NPL sample: estimation of amount

- 8 radionuclides seem to be in the sample:
  - Create a new reduced library with these 8 nuclides
- Reanalyse the sample, use decay correction.
- Check for possible daughter – parent radionuclide effects:
  - Nb-95.
  - Zr-95 decays to Nb-95 (and partly to Nb-95m), which decays to Mo-95.
  - One needs to make corrections based on the concentrations of the radionuclides at the time of measurement

Reduction of library file



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Corrections applied

Corrections	Status	Comments
Decay correct to date:	YES	01-Oct-2008 12:00:00
Decay during acquisition:	NO	
Decay during collection:	NO	
True coincidence correction:	YES	
Peaked background correction:	YES	npl_all_candidates.Pbc 06-Jan-2009 00:35:48
Absorption (Internal):	NO	
Geometry correction:	NO	
Random summing:	NO	

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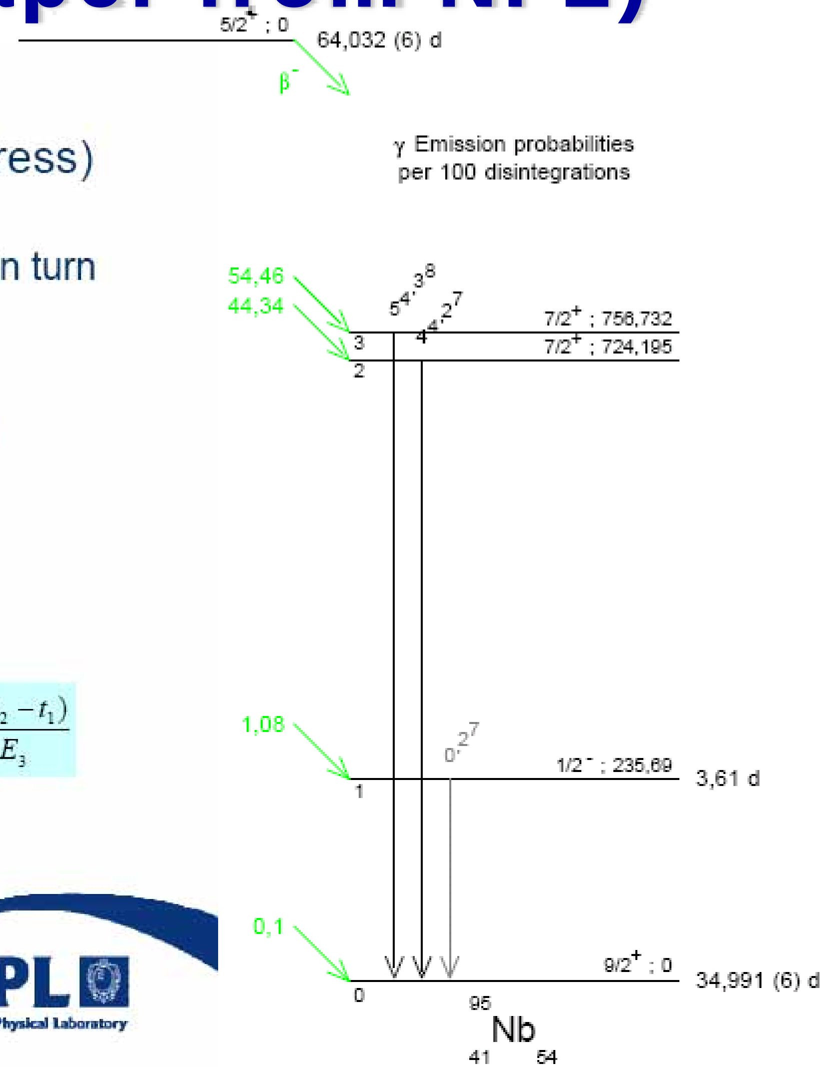
Correction for Nb-95
(see finar report / paper from NPL)



Decay of ^{95}Nb (Applied Radiation and Isotopes, in press)

^{95}Zr (1) decays to ^{95}Nb (3) and ^{95m}Nb (2) (p = 0.0112), which in turn decays to ^{95}Nb (3) (q = 0.975) and stable ^{95}Mo

Decay/ingrowth during measurement combined with a decay correction to the reference time of the exercise

$$A_3(t_{ref}) = F_{31}(t_1,t_2,t_{ref})\,C_1 + F_{32}(t_1,t_2,t_{ref})\,C_2 + F_{33}(t_1,t_2,t_{ref})\,C_3$$

$$F_{31}(t_1,t_2,t_{ref}) = \left[\frac{((1-p)(\lambda_2-\lambda_1)+q\,p\,\lambda_2)(\lambda_1 E_3-\lambda_3 E_1)}{(\lambda_3-\lambda_1)(\lambda_2-\lambda_1)E_1} + \frac{q\,p\,\lambda_2(\lambda_3 E_2-\lambda_2 E_3)}{(\lambda_3-\lambda_2)(\lambda_2-\lambda_1)E_2}\right]\frac{\lambda_3(t_2-t_1)}{E_3}$$

$$F_{32}(t_1,t_2,t_{ref}) = \frac{q\,\lambda_3(t_2-t_1)}{(\lambda_3-\lambda_2)}\left[\frac{\lambda_2}{E_2}-\frac{\lambda_3}{E_3}\right]$$

$$F_{33}(t_1,t_2,t_{ref}) = \frac{\lambda_3\,(t_2-t_1)}{E_3}$$

$$E_i = e^{-\lambda_i(t_1-t_{ref})} - e^{-\lambda_i(t_2-t_{ref})}$$

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THANK YOU!

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