Evaluation and validation of national standard geometries for gamma spectrometry

Assessment of national standard geometries for gamma spectrometry, aimed for the emergency preparedness in Sweden, 2007

GammaSem 090917

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Steps in the project gGEO and gGEO2

Selection of common standard geometries for gamma spectrometry to be used nationally (spring 2005)

Purchase of standard sources (autumn 2005)

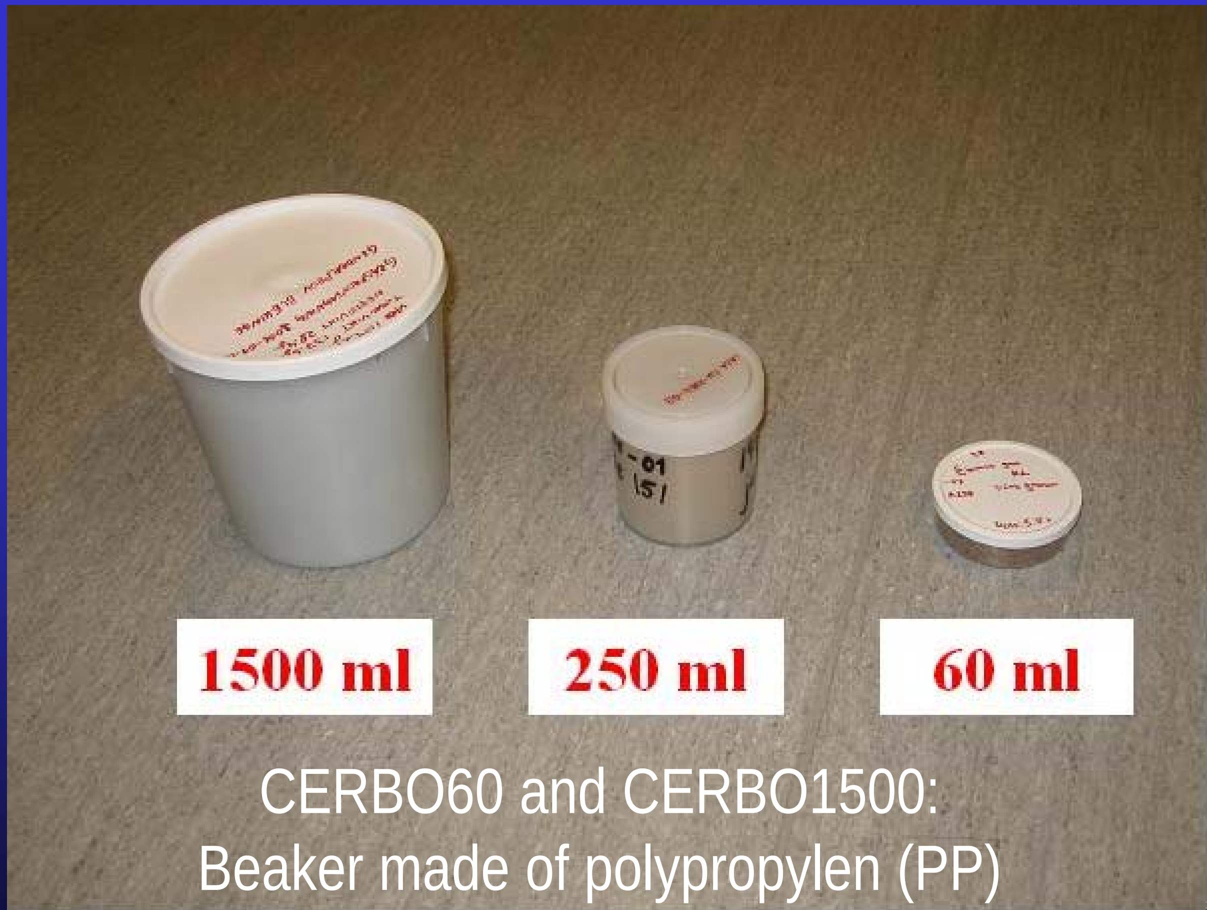
Circulation of sources to allow all participating laboratories to calibrate their HPGe-systems (spring 2006)

Selection and purchase of validation sources (autumn 2007)

Submission of report sheet and validations samples in an open intercomparison, i.e. the certificate values were known to participants (ht 2007)

Compilation and assessment of reported data – report to SSM (summer 2008)

Standard geometries selected for the Swedish national emergency preparedness



CERBO60 and CERBO1500:
Beaker made of polypropylen (PP)
Lids made of white polyethylene LD (PELD)

Standard geometries selected for the Swedish national emergency preparedness

Beaker size	Target density	Principal targetted matrices
60 ml	1.0	Sediment, dried vegetation
250 ml	1.0	Liquids (urine, milk, water) Soil
1500 ml	0.2	Fresh vegetation
	1.0	Urine/feces, milk, water

Standard calibrations sources purchased by SSM from ISOTRAK

60 ml gross
60 ml active volume

250 ml gross
200 ml active volume

1500 ml gross
1300 ml active volume

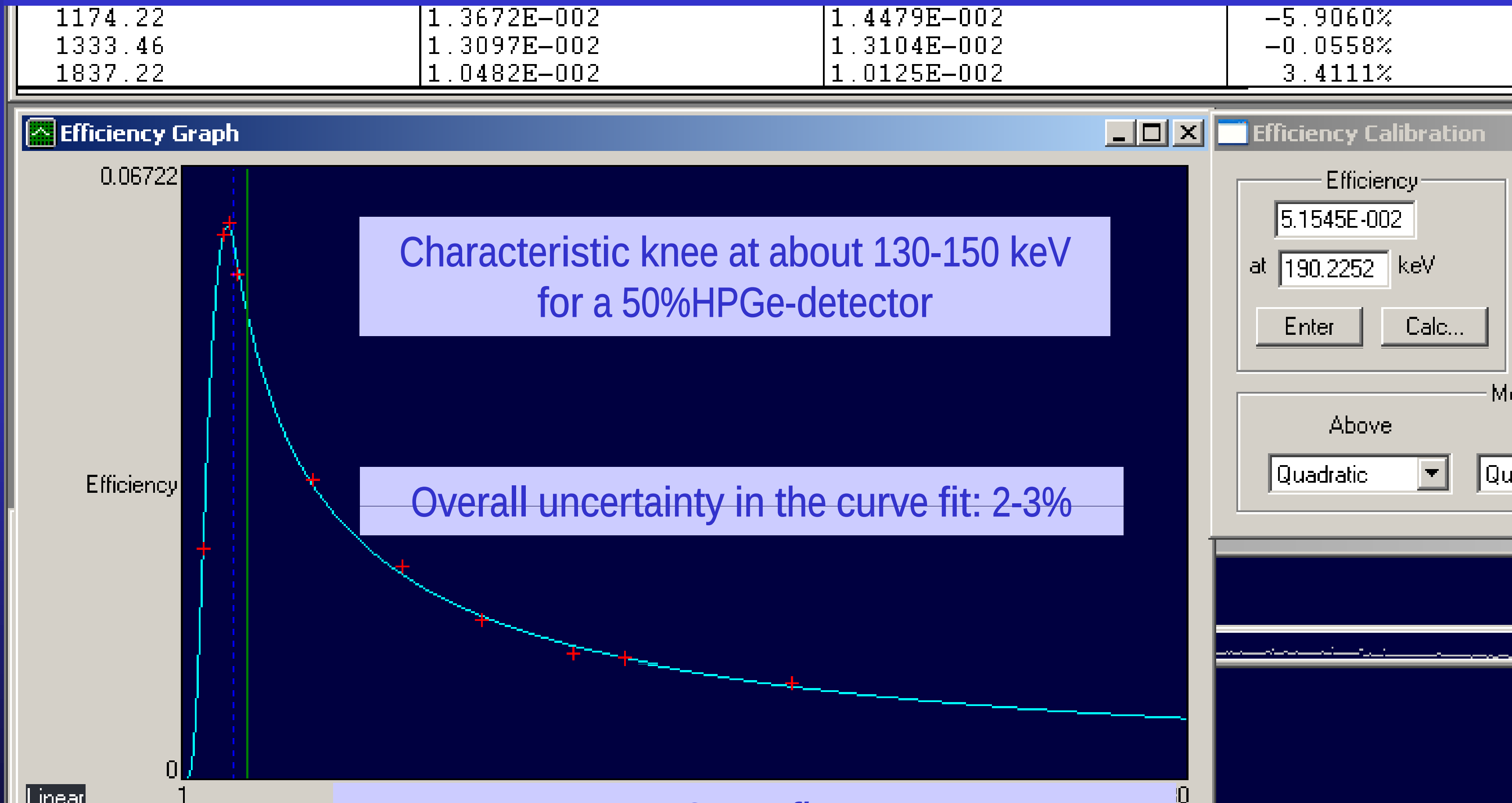
Cocktail of: Cs-137, Cd-109, Co-60, Co-57, Y-88, Sr-85, Am-241. Sn-113, Hg-203 AND Cs-134

Reference date 2005-11-01

Specified uncertainty: ±1,5% (1 SD)

Price: ~2 kEuro

HPGe efficiency curve



Characteristic knee at about 130-150 keV for a 50%HPGe-detector

Overall uncertainty in the curve fit: 2-3%

Curve fit:

$$\varepsilon(E_\gamma)=Exp(a_1*E_\gamma+a_2*E_\gamma^{0}+a_3*E_\gamma^{-1}+a_4*E_\gamma^{-2}+a_5*E_\gamma^{-3}+\ldots)$$

Validation samples used

Sample ID	Reference date	Origin	Certified accuracy (1 SD)
IAEA_154	1987-08-31	Dried wheat	±2,2% (^{40}K) ±1,9% (^{137}Cs) ±2,3% (^{134}Cs)
IAEA_375	1991-12-31	Dried soil	±0,45% (^{40}K) ±0,5% (^{134}Cs) ±0,4% (^{137}Cs) ±2,5% (^{226}Ra)
NIST_4350B	1981-09-09	River sediment	±2,1% (^{137}Cs) ±3,3% (^{226}Ra) ±7% (^{241}Am)
NIST_4357	1994-02-16	Coastal water sediment	±1,1% (^{40}K) ±0,8% (^{137}Cs) ±1,3% (^{232}Th)

Reference material purchased from IAEA (Vienna) and NIST (USA).

Validation samples used

Reference materials were re-packaged into the three types of standard beakers

Sample ID	Origin	Density g/cm³	Active volume ml
250ml standardburk (NU652)		*1,0*	*200*
GGEO250_1	IAEA_154	0,696	200
GGEO250_2	IAEA_154	0,704	250
GGEO250_3	IAEA_375	1,451	200
1500 ml standardburk (NU653 och NU654)		*1,0 resp. 0,2*	*1300*
GGEO1500_1	IAEA_154	0,713	1300
GGEO1500_2	IAEA_154	0,684	1450
GGEO1500_3	IAEA_375	1,488	1300
60 ml standardburk (NU651)		*1,0*	*60*
GGEO60_1	IAEA_154	0,712	60
GGEO60_2	IAEA_154	0,723	60
GGEO60_3	IAEA_154	0,745	60
GGEO60_4	IAEA_154	0,703	60
GGEO60_5	IAEA_375	1,508	60
GGEO60_6	IAEA_375	1,467	60
GGEO60_7	NIST 4357	1,055	60
GGEO60_8	NIST 4350b	0,943	60

N.B. that we were unable to find matrices that precisely match the densities of the standard sources, densities ranging from 0.71-1.51.

Particpants in the validation

Laboratory	Detector systems calibrated
Medicinsk strålningsfysik, Linköping	42% HPGe
Medicinsk strålningsfysik, Lund	22% HPGe 50% HPGe 100% HPGe
Medicinsk strålningsfysik, Malmö	56% HPGe
Statens strålskyddsinstitut, Stockholm	16% HPGe, 40% HPGe och 60% HPGe
Studsvik Nuclear, Nyköping	30% HPGe (Berylliumfönster) 30% HPGe (HP) 30% HPGe(ULB)
Sveriges lantbruksuniversitet, SLU; Uppsala	42% HPGe
Totalförsvarets forskningsinstitut, FOI, Kista	77% HPGe (remote and close geometry), 79% HPGe (remote and close geometry)
Totalförsvarets forskningsinstitut, FOI, Umeå	50% HPGe 80% HPGe

Reported measurand

The deviation normalised to the total estimaed unceratanty was used as a measurand of the accordance/consistency between the particpating laboratories.

$$z = \frac{(m_{labb} - m_{cert})}{\sqrt{(s_{labb}{}^{2} + s_{cert}{}^{2})}}$$

m_{labb}= reported activity concentration of a given radionuclide for a given validation sample [Bq/kg]

m_{cert}= certified value of the activity concentration of a given radionuclide for a given validation sample [Bq/kg]

s_{labb}=reported standard deviation (1 sigma) of the reported value of m_{labb} [Bq/kg]

s_{cert}= uncertainty (1 sigma) of m_{cert} as stated by the supplier of the validation samples [Bq/kg]

The significance of z?

$$z = \frac{(m_{labb} - m_{cert})}{\sqrt{(s_{labb}{}^{2} + s_{cert}{}^{2})}}$$

Z>0: Reporting laboratory OVER-estimates the activity

Z<0: Reporting laboratory UNDER-estimates the activity

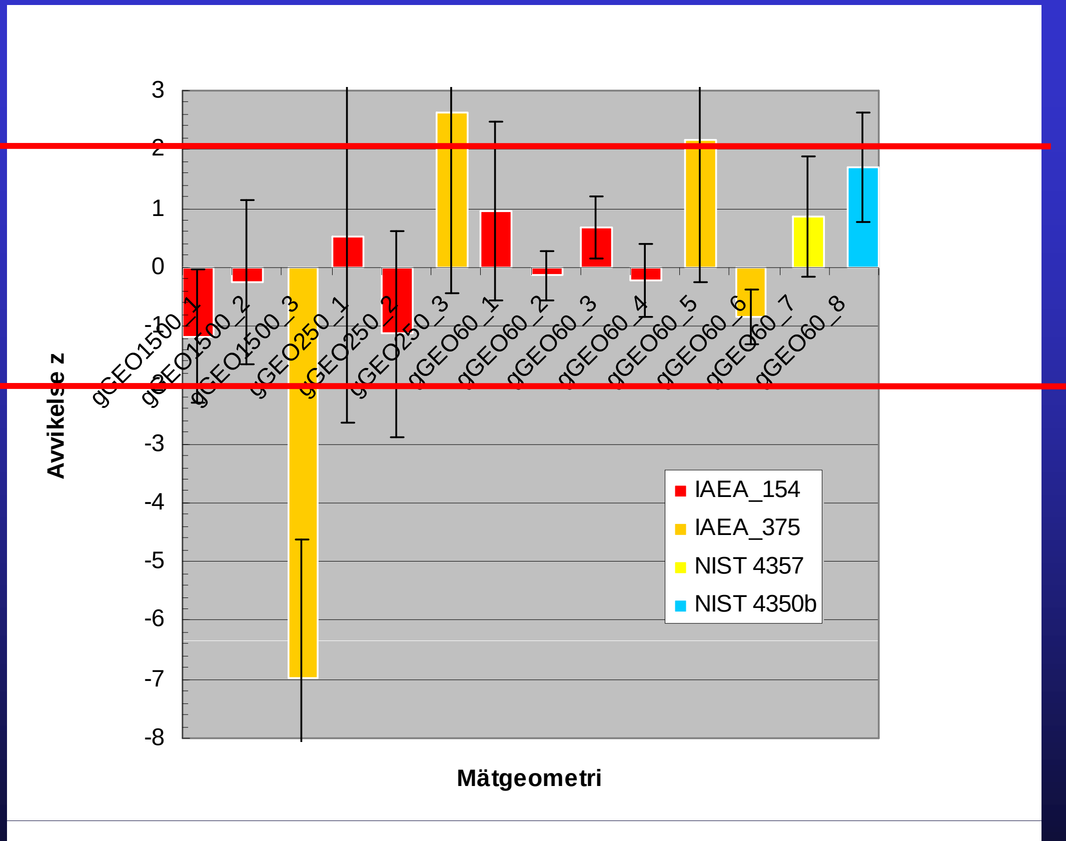
IZI<2: Less than 5% probability that the deviation is caused by chance – the reported value is considered to NOT SIGNIFICANTLY deviate from the certified value

IZI>2: At least 5% probability that the deviation is caused by chance – the reported value is considered to SIGNIFICANTLY deviate from the certified value

Disclaimer: We never clearly defined what the reporting laboratories should include in the uncertainty estimation, s_{labb}, hence the possibility of different confidence levels of the m_{labb}.

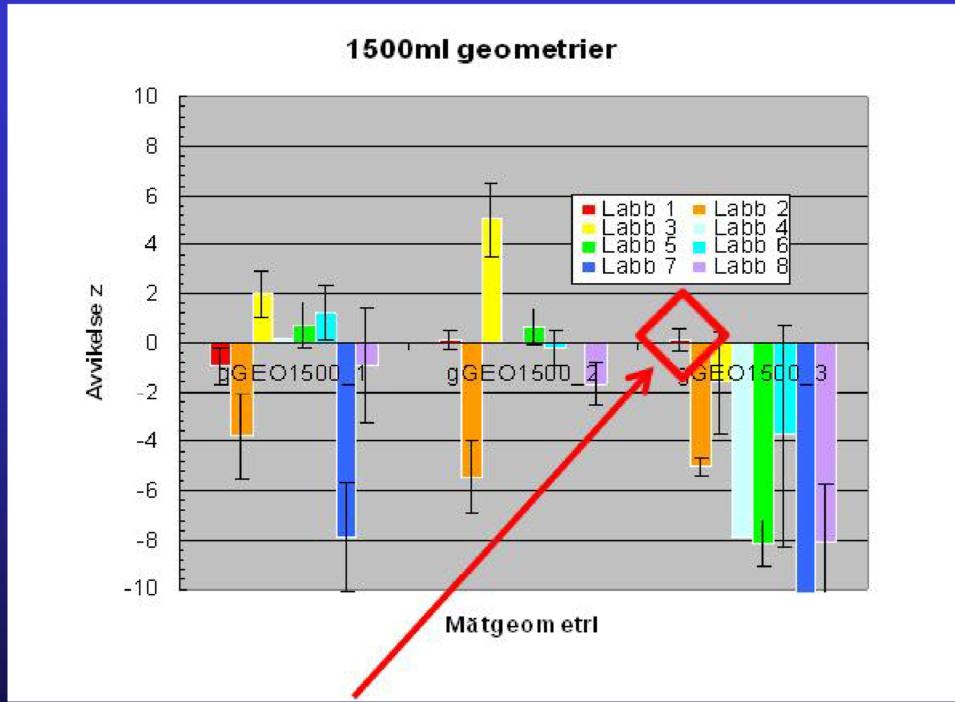
Results

Standardised deviation from certified values, z, for the 13 validation samples. Averaged over all radionuclides and all participating laboratories



Resultat (cont'd)

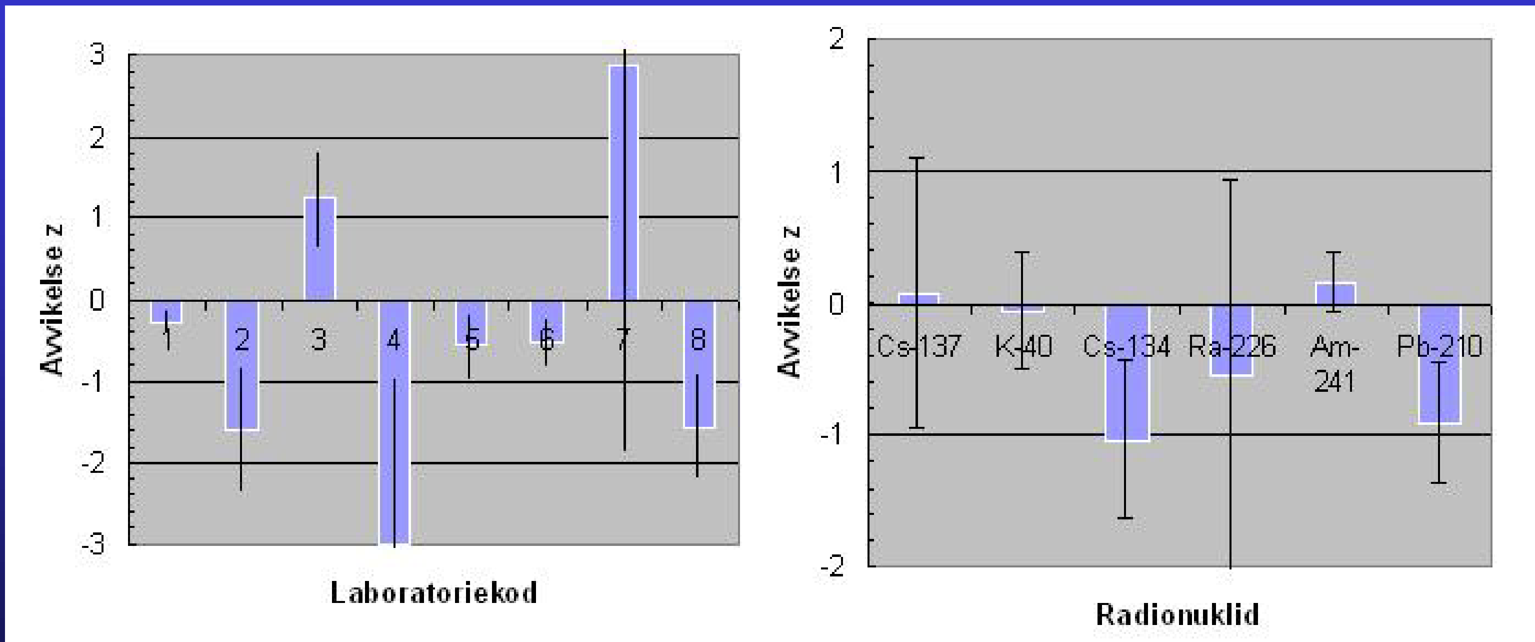
1500ml-geometries; gGEO1500_1, gGEO1500_2 och gGEO1500_3.



Highest z-value found for gGEO1500_3; a soil sample with density 1.45. All participants except two reported significant underestimates on the activity contents.

Results (contn'd)

Normalised deviation, z, as averaged over participating labporatories and radionuclides, respectively

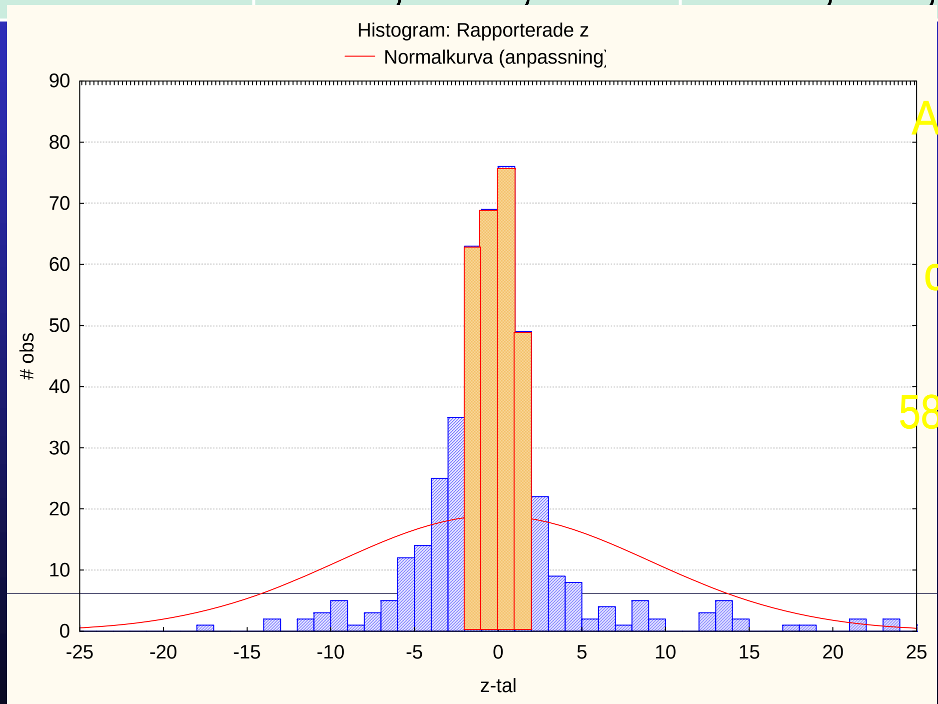


Certain laboratories exhibit, on average, large deviances (Labb 4 och Labb 7), whereas some display very insignificant deviations (Labb 1, Labb 5 och Labb 6).

^{134}Cs and ^{210}Pb tend to be underestimated

Results (contn´d)

Factor	<z> ±1 SE	<|z|> ±1 SE
Labb	2,34±0,58	5,1±0,96
Geometry	0,09±0,62	3,2±0,52
Nuclide	-0,28±0,23	3,2±0,85



A certain fraction of the reported values deviate extremely from the certified valuies, however, in no particular sense.

58 % of all reported data were confined within :

-2<z<2.

Conclusions

- *The quality in the gamma spectrometric analysis depends still more on who's carrying out the analysis, than on soft-ware or the use of calibration standards.*
- The large variance in the reported z-values may origin from incomplete uncertainty assessment of the measurements. REMEDY: Clarify the various components of uncertainty and make realistic uncertainty estimations of these components.
- Some of the large z-values may arise from erroneous procedures in obtaining the efficiency curves, e.g. merging calibration points from coincidence prone nuclides (^{134}Cs) with the rest of the gamma nuclides in the calibration sources.
- Density correction procedures makes a difference: Correction procedures carried out appear to satisfactory compensate for the loss of counts to yield correct activity values.

The future

The accuracy and inter-laboratory agreement can be further improved in Sweden if…

…a common approach for the Swedish gamma spectrometry laboratories to carry out the uncertainty analysis and find a transparent algorithm for assessing the uncertainty propagation in the various steps of their sample assessment.

A possible option to extend the harmonisation of gamma geometries to include also other Nordic countries?

Lessons learned for another validation

- The normalised deviation does not adequately describe the quality of a measurement; it needs to be combined with some absolute values of agreement, regardless of the expanded uncertainties in the certificate and measurement.
- Validation samples need to have similar densities as the original standard sources on which the calibrations is based.
- Validation samples need to have a wider variety of gamma lines to properly test all parts of the efficiency curves/calibration of the HPGe-systems at the laboratory.