STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

Efficiency transfer for low-energy (30-100 keV) gamma-ray spectrometry analyses

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Efficiency transfer for low-energy (30-100 keV) gamma-ray spectrometry analyses

Michel Bruggeman, Leen Verheyen, Tim Vidmar

Low Level Radioactivity Measurements

michel.bruggeman@sckcen.be

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  - Proposal procedure

Efficiency transfer for routine gamma-ray analyses

- Efficiency transfer is required for unbiased results
  - Filling height (if no 100% filling is used)
  - Apparent sample density
  - Sample composition (30-100 keV)
  - Any other sample parameter that is different from the reference

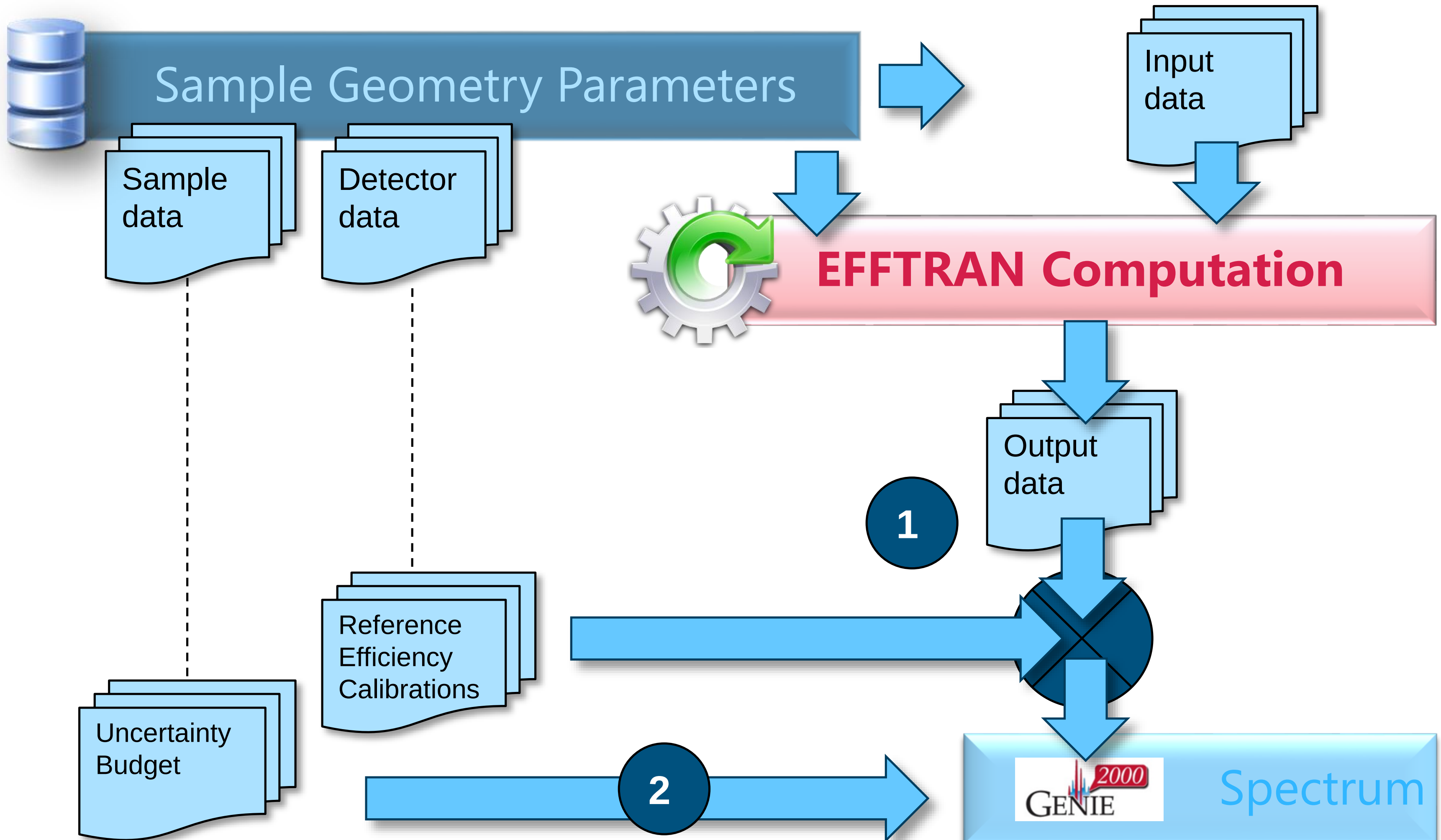
- Other corrections that may depend on geometry
  - Corrections for ambient background
    - Filling height, density,... may modify background radiation that enters the detector
    - Generally neglected
  - True summing corrections
    - unbiased or small bias when the actual sample parameters are used

Organization at SCK-CEN

- Database with sample data
  - Sample container parameters (dimensions, materials)
  - Sample net weight
  - Sample volume (filling height)
  - Sample composition → material list → attenuation data via XCOM[1]
  - All sample parameters configured according to EFFTRAN
- Database with detector model data (according to EFFTRAN)
- Reference efficiency calibrations
  - Multi gamma sources + additional nuclides
  - Water matrix (acidified)
  - Fixed volume/filling height (per geometry)
  - Stored as Genie 2000 (cal files)
  - Referenced in database (by geometry ID and detector ID)

[1] Photon Cross Sections Database, NIST

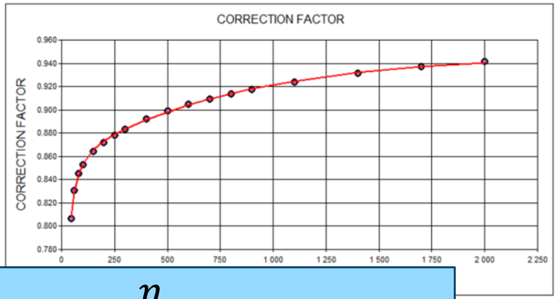
Efficiency Transfer and Uncertainty Budget



Efficiency Transfer data model
Correction function

EFFTRAN Computation

Actual Sample ↔ Reference Sample



$E_1 \rightarrow CF(E_1)$

$E_2 \rightarrow CF(E_2)$

$E_3 \rightarrow CF(E_3)$

$E_4 \rightarrow CF(E_4)$

…

$E_n \rightarrow CF(E_n)$

$$\ln\left(\mathbf{CF}(\ln(E)\right) = \sum_{k=0}^{n} \boldsymbol{C_k} \ln(E)^k$$

Spectrum (cam file data)

$$\ln \varepsilon_{ref}\,(E) = \sum_{k=0}^{n} A_k \ln(E)^k$$

$$\ln \varepsilon_{Corr}\,(E) = \sum_{k=0}^{n} (A_k + \boldsymbol{C_k}) \ln(E)^k$$

$$\boldsymbol{\varepsilon_{Sam} = \varepsilon_{ref}}\ \mathbf{CF}$$

Applied to low and high-energy dual efficiency curve

Uncertainty Budget (function of gamma-ray energy)

Sample type → Uncertainty Polynomial

$$Err\,(\ln(E)) = \sum_{k=0}^{n} E_k \ln(E)^k$$

Spectrum (cam file data)

Covariance matrix of polynomial fitting + source uncertainty

$$\sigma^2(\ln \varepsilon\,(\ln(E))) = \sum_{j=0}^{n}\sum_{i=0}^{n} M_{ij} \ln(E)^{i+j}$$

$$\sigma^2(\ln(\varepsilon)) = \frac{\sigma^2(\varepsilon)}{\varepsilon^2}$$

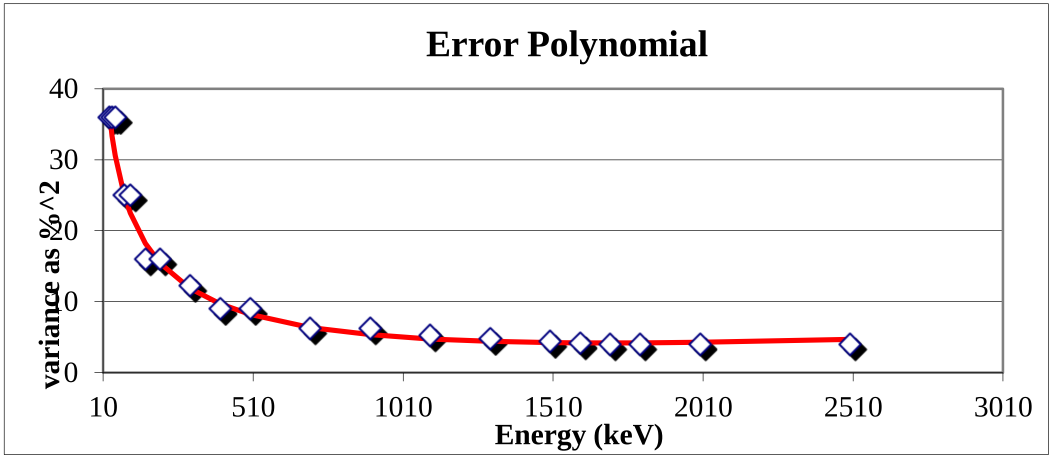
Efficiency function for standard uncertainty used by Genie

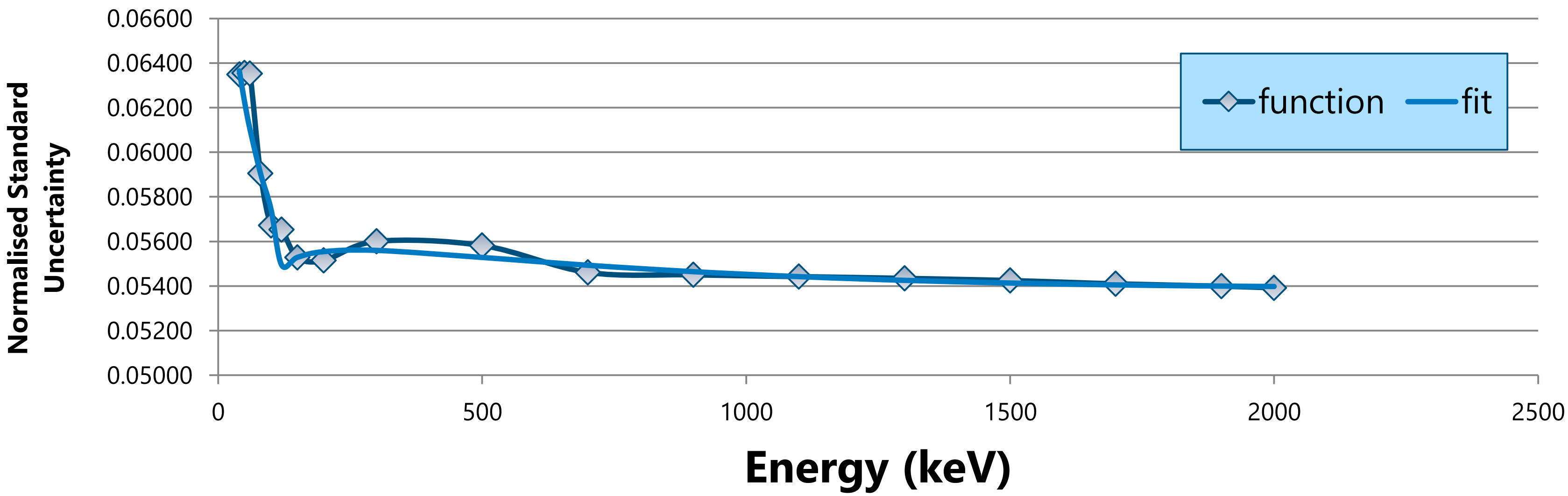
Applied to low and high-energy data

Determination of energy depended uncertainty budget

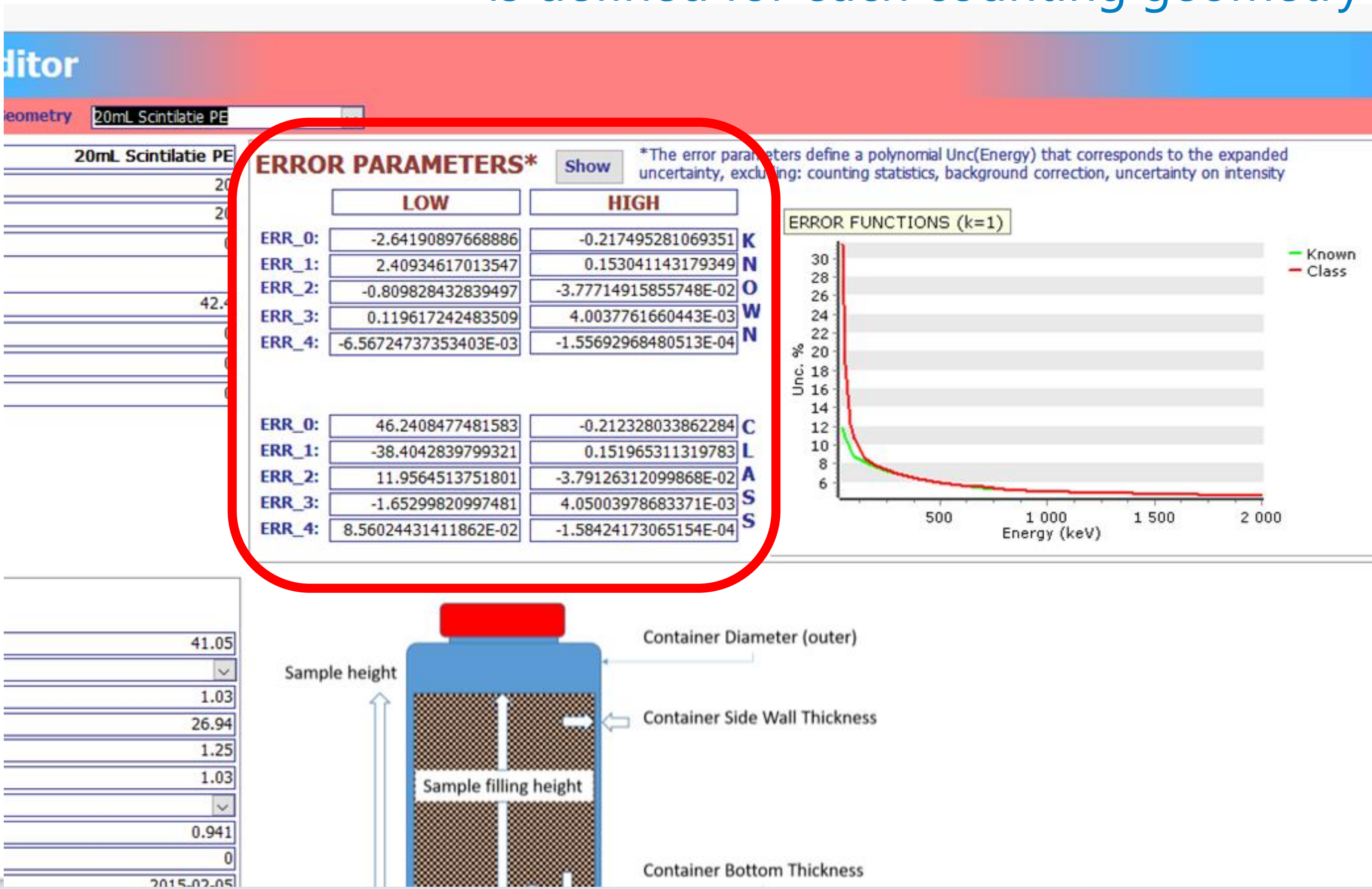
	REF waarde ENERGY(keV)	VULHOOGTE			DIAMETER			BODEM DIKTE			SAMPLE DENSITY			Sample COMPOSITION			POSITIONERING STAAL	CURVE FITTING	REF KALIBRATIEBRON	SYSTEEM DRIFT	GAMMA INTENSITEIT	SUMMING CORRECTION	TEL VERLIEZEN	ENERGY (keV)	STANDARD COMBINED UNCERTAINTY	VARIANCE
		-3%	3%	MAX	-0.2%	0.2%	MAX	-2%	2%	MAX	-3%	3%	MAX	A	B	MAX	MAX		0.02 k=1	0.01	0 bib	0.02	0.01			Variance
1	45	-0.0537	0.0489	0.031	-0.0016	0.0009	0.0009	-0.0018	0.0011	0.001	-0.0239	0.0177	0.0138			0.4092	0.0680	0.02	0.02	0.01	0	0.02	0.01	45	0.419	0.1754
2	60	-0.0534	0.0481	0.031	-0.0008	0.0016	0.0009	-0.0016	0.0016	0.001	-0.0213	0.0162	0.0123			0.2333	0.0650	0.02	0.02	0.01	0	0.02	0.01	60	0.249	0.0619
3	80	-0.0526	0.0474	0.030	-0.0010	0.0015	0.0009	-0.0015	0.0015	0.001	-0.0201	0.0153	0.0116			0.1261	0.0610	0.02	0.02	0.01	0	0.02	0.01	80	0.151	0.0228
4	100	-0.0521	0.0474	0.030	-0.0009	0.0013	0.0007	-0.0017	0.0017	0.001	-0.0188	0.0147	0.0108			0.1248	0.0590	0.02	0.02	0.01	0	0.02	0.01	100	0.149	0.0221
5	150	-0.0510	0.0467	0.029	-0.0012	0.0012	0.0007	-0.0020	0.0012	0.001	-0.0170	0.0139	0.0099			0.0419	0.0540	0.02	0.02	0.01	0	0.02	0.01	150	0.087	0.0076
6	200	-0.0501	0.0458	0.029	-0.0008	0.0012	0.0007	-0.0016	0.0016	0.001	-0.0164	0.0133	0.0095			0.0307	0.0500	0.02	0.02	0.01	0	0.02	0.01	200	0.080	0.0063
5	150	-0.0510	0.0467	0.029	-0.0012	0.0012	0.0007	-0.0020	0.0012	0.001	-0.0170	0.0139	0.0099			0.0419	0.0540	0.02	0.02	0.01	0	0.02	0.01	150	0.080	0.0063
6	200	-0.0501	0.0458	0.029	-0.0008	0.0012	0.0007	-0.0016	0.0016	0.001	-0.0164	0.0133	0.0095			0.0307	0.0500	0.02	0.02	0.01	0	0.02	0.01	200	0.080	0.0063
7	250	-0.0492	0.0448	0.028	-0.0008	0.0012	0.0007	-0.0016	0.0016	0.001	-0.0159	0.0122	0.0092			0.0294	0.0480	0.03	0.01	0.01	0	0.02	0.01	250	0.084	0.0070
8	300	-0.0484	0.0443	0.028	-0.0004	0.0012	0.0007	-0.0017	0.0012	0.001	-0.0154	0.0117	0.0089			0.0286	0.0460	0.02	0.01	0.01	0	0.02	0.01	300	0.076	0.0057
9	400	-0.0474	0.0430	0.027	-0.0004	0.0013	0.0007	-0.0017	0.0013	0.001	-0.0138	0.0117	0.0080			0.0266	0.0420	0.02	0.02	0.01	0	0.02	0.01	400	0.072	0.0052
10	500	-0.0463	0.0424	0.027	-0.0009	0.0009	0.0005	-0.0017	0.0009	0.001	-0.0133	0.0106	0.0077			0.0261	0.0390	0.02	0.02	0.01	0	0.02	0.01	500	0.070	0.0049
11	600	-0.0452	0.0416	0.026	-0.0004	0.0013	0.0008	-0.0013	0.0013	0.001	-0.0122	0.0101	0.0071			0.0247	0.0370	0.01	0.01	0.01	0	0.02	0.01	600	0.063	0.0040
12	700	-0.0447	0.0411	0.026	-0.0004	0.0013	0.0008	-0.0013	0.0013	0.001	-0.0117	0.0096	0.0068			0.0232	0.0350	0.01	0.01	0.01	0	0.02	0.01	700	0.062	0.0038
13	800	-0.0443	0.0403	0.026	-0.0004	0.0013	0.0008	-0.0013	0.0013	0.001	-0.0112	0.0096	0.0065			0.0226	0.0340	0.01	0.01	0.01	0	0.02	0.01	800	0.061	0.0037
14	900	-0.0435	0.0401	0.025	-0.0009	0.0009	0.0005	-0.0018	0.0009	0.001	-0.0107	0.0091	0.0062			0.0216	0.0320	0.01	0.01	0.01	0	0.02	0.01	900	0.059	0.0035
15	1100	-0.0426	0.0394	0.025	-0.0005	0.0009	0.0005	-0.0014	0.0009	0.001	-0.0103	0.0082	0.0060			0.0199	0.0300	0.01	0.01	0.01	0	0.02	0.01	1100	0.057	0.0033
16	1400	-0.0418	0.0382	0.024	-0.0005	0.0010	0.0006	-0.0014	0.0010	0.001	-0.0089	0.0078	0.0051			0.0173	0.0270	0.01	0.01	0.01	0	0.02	0.01	1400	0.054	0.0030
17	1700	-0.0409	0.0375	0.024	-0.0005	0.0010	0.0006	-0.0010	0.0010	0.001	-0.0079	0.0074	0.0046			0.0153	0.0250	0.01	0.01	0.01	0	0.02	0.01	1700	0.053	0.0028
18	2000	-0.0399	0.0371	0.023	-0.0005	0.0010	0.0006	-0.0010	0.0010	0.001	-0.0081	0.0063	0.0047			0.0137	0.0230	0.01	0.01	0.01	0	0.02	0.01	2000	0.051	0.0026

Uncertainty function and corresponding polynomial





An uncertainty budget function is defined for each counting geometry



Generic sample compositions for sample matrices

- well defined materials (water, PE, metals…)
- material class
  - Organic matter
  - Soil/dirt

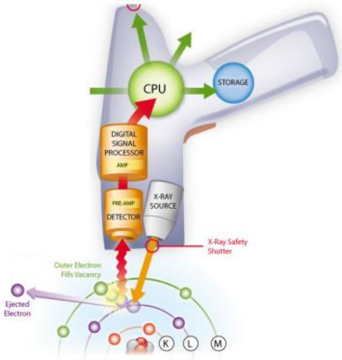
Uncertainty budget does not assume variability of the sample composition

Uncertainty budget **accounts for variability of the sample composition** and its impact on detection efficiency

Uncertainty gets unrealistically large at low energy !

Means to determine sample attenuation

- Generic compositions
- By comparison with representative standards
  - (not very practical for general use)
- XRF-handheld monitor, AOS-MS,…
  - Specify the sample material for the EFFTRAN sample model
    - Elements from Mg → U, other elements O and H ?
  - Apply a standard efficiency transfer with EFFTRAN
    - Attenuation data from XCOM

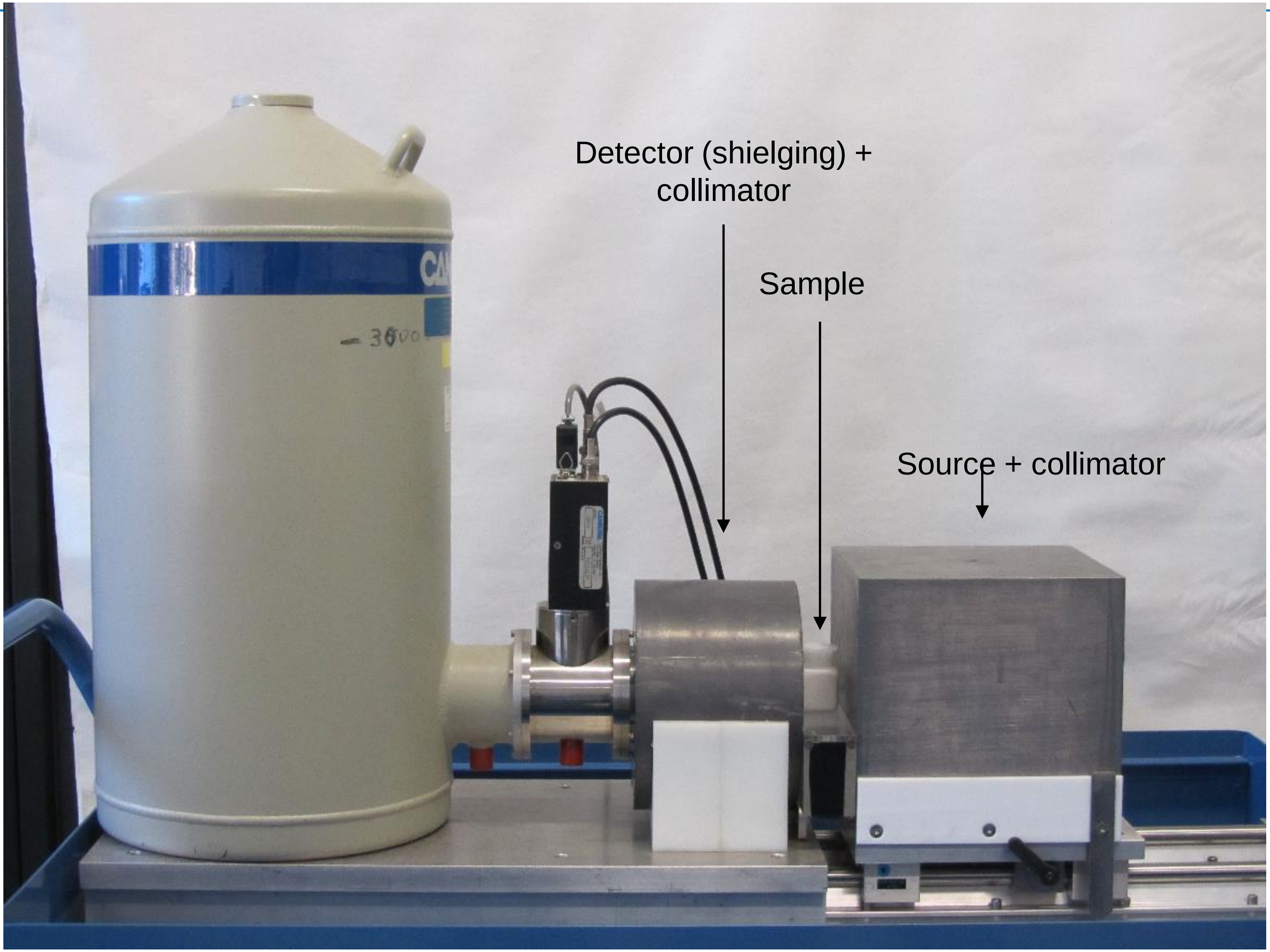


- Multi Energy gamma-ray transmission measurement through sample material

$$T(E) = \exp(-\mu_l(E)\,x)$$

Only valid for parallel beams and well defined geometry ?

Transmission setup at SCK-CEN



Experimental determination of linear attenuation coefficient

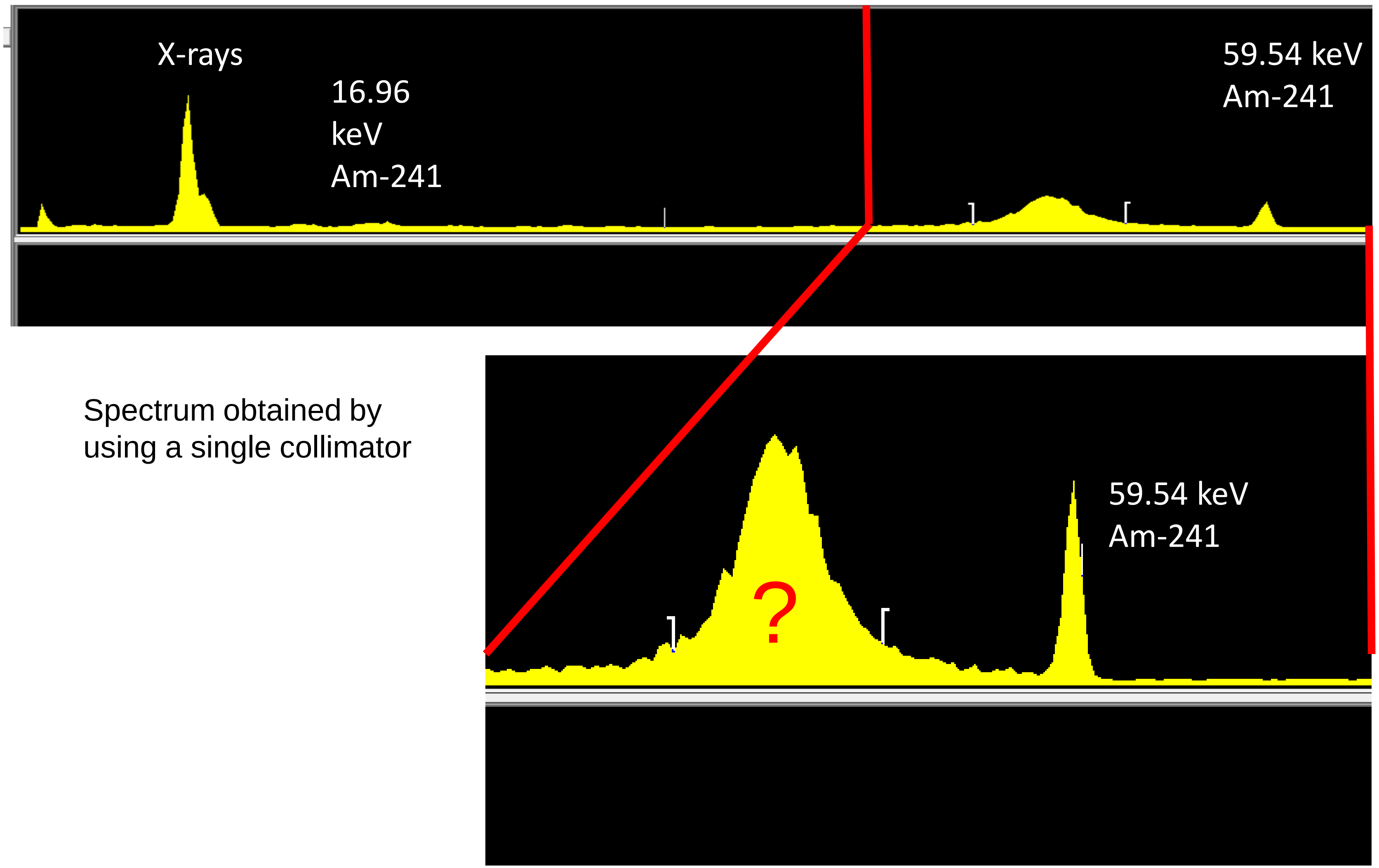
- Well collimated beam is required

$$R(x) = R_0 \exp(-\mu_l x)$$

$$\mu_l = -\frac{\ln\left(\frac{R_0}{R}\right)}{x}$$

- High intensity sources required
- At low energy – small angle Compton scattering contributions may bias the results
- Time consuming

Effects of low-angle scattering may complicate multi energy transmission



Spectrum obtained by
using a single collimator

Attenuation Correction Procedures based on Transmission

- NUREG/CR-5550 (LA-UR-90-732) “Passive Nondestructive Assay of Nuclear Materials”, 1991
  - Chapter 6: “Attenuation Correction Procedures” by J.L. Parker and references therein
  - **Far-field** form for self-attenuation correction factor for a box-shaped sample

$$CF(AT) = \frac{\mu_l x}{1 - \exp(-\mu_l x)}$$

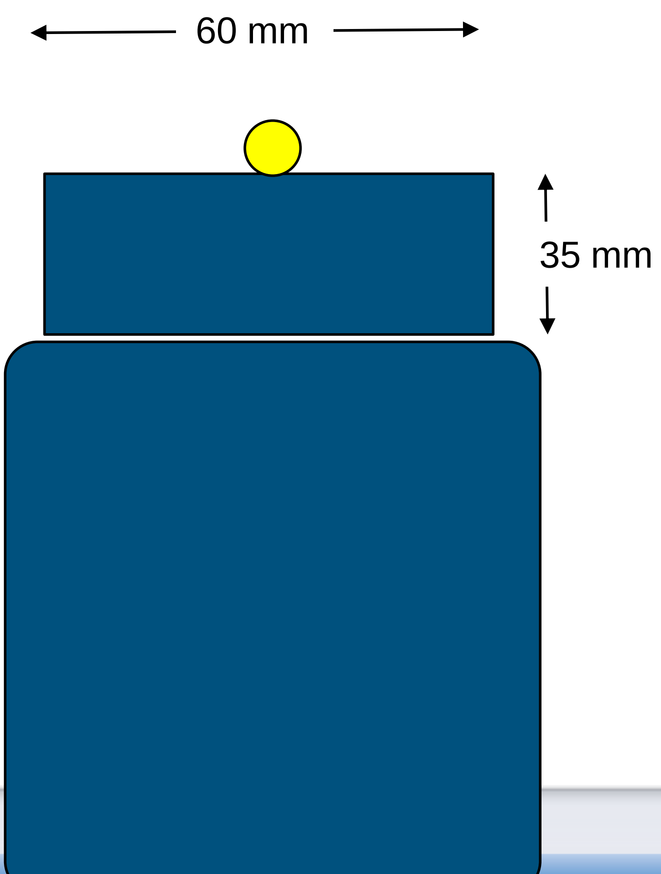
$$CF(AT) = \frac{-\ln(T)}{1 - T}$$

  - These equations are only valid if the gamma-rays travel in a perpendicular direction from the sample towards the detector
  - This is generally not the case, especially not when the sample is measured close to the detector

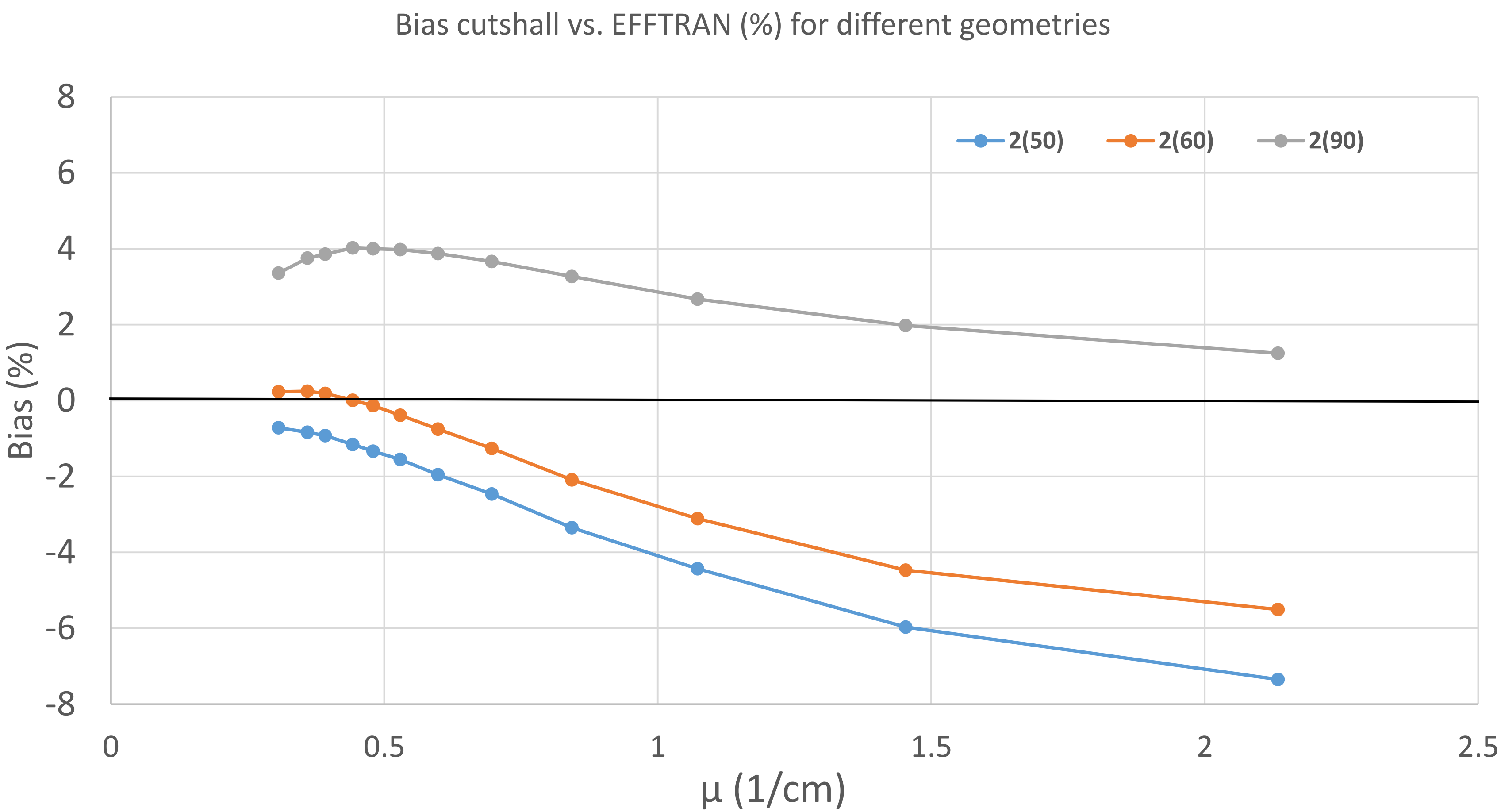
Attenuation Correction Procedures

- N.H. Cutshall, correction for Pb^{210} in sediments (1981)
- **Near field (wide angle)**

$$CF(AT) = \frac{-\ln(T)}{1 - T}$$

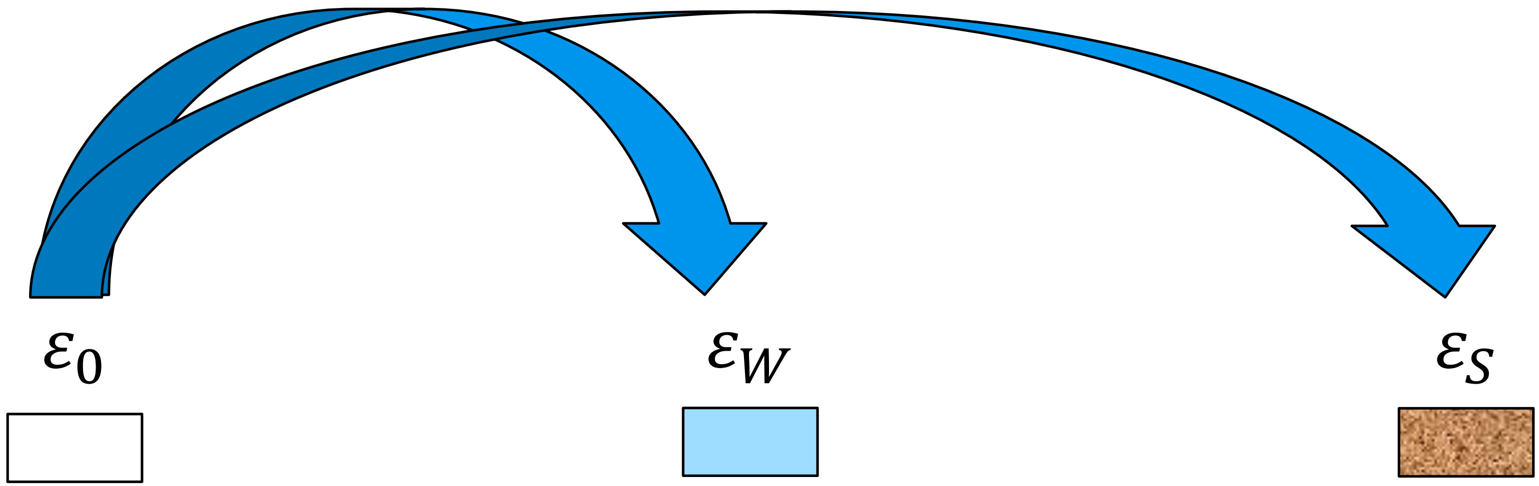


Cutshall correction (relative to air) compared to EFFTRAN



Sediment sample in pillbox (20mm heigh at different diameters, 50mm, 60mm and 90mm)
Cutshall relative to air
On a 60mm x 60mm detector

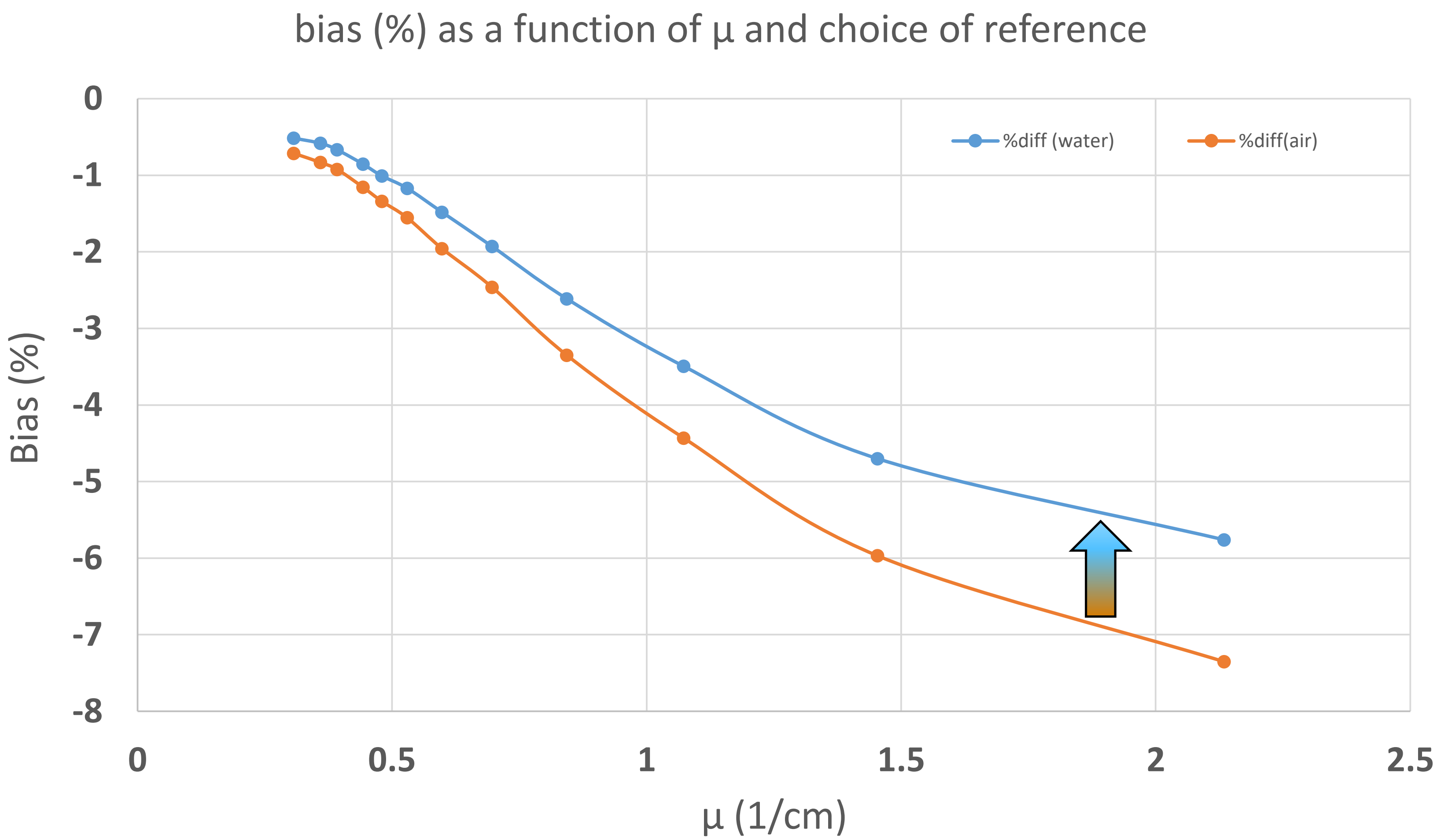
Cutshall relative to another reference sample material



$$\varepsilon_W = \frac{-\ln(T_W)}{(1 - T_W)} \varepsilon_0 \qquad \varepsilon_S = \frac{-\ln(T_S)}{(1 - T_S)} \varepsilon_0$$

$$\varepsilon_S = \frac{\ln(T_S)(1 - T_W)}{\ln(T_W)(1 - T_S)} \varepsilon_W$$

Transmission relative to reference closer to the sample improves accuracy



What to do to get unbiased results ?

- Cutshall method may give acceptable results (few % bias)
  - Specially for not too high attenuation
  - Well selected geometry
  - Live with the bias and include it in the standard uncertainty
- Generally Cutshall is biased
  - Requires additional corrections
- Better to use full simulation to fix the relation between transmission data and sample material (μ-values) for a specified geometry
  - General purpose Monte Carlo codes like MCNP,...
  - Gespecor

  } Not easy to include in automation or simple working procedure

  - TEFFTRAN (by Tim Vidmar)

Single energy versus energy range corrections

- **Single Energy Correction**
  - Correct the required energy e.g. 46.54 keV for Pb-210
  - Use a transmission source that exactly emits this energy

- **Corrections in an Energy Range**
  - Multi energy gamma-ray source
  - Low energy peaks suffer bad counting statistics when measured with a multi-energy gamma-ray source
  - Only the low energy window 30 keV – 120 keV requires element specific corrections, for the higher energy a correction based on density only is appropriate
  - What is an appropriate multi energy source ?
    - I-129 – Am-241
  - Interpolation between transmission/attenuation data
  - How to deal with K,L…-edges

Inter/extra-polation of attenuation data

- 1) Via polynomial fitting of the attenuation data as a function of energy (= a strongly varying function of energy at low energy)
- 2) Via fitting with an appropriate material composition

$$\mu(E) \approx \sum_{i=1}^{N} w_i \, \mu_i(E)$$

With $\mu_i(E)$ the mass attenuation coefficient for element i from XCOM

With w_i the relative contribution of element i and $\sum w_i = 1$

With N the number of elements in a well defined set of elements

Inter/extra-polation of attenuation data

- General least squares fitting minimizing F by changing w_i

$$F(E_j, w_i) = \left(\mu_{exp}(E_j) - \sum_{i=1}^{N} \boldsymbol{w_i} \, \mu_i(E_j) \right)^2 = 0$$

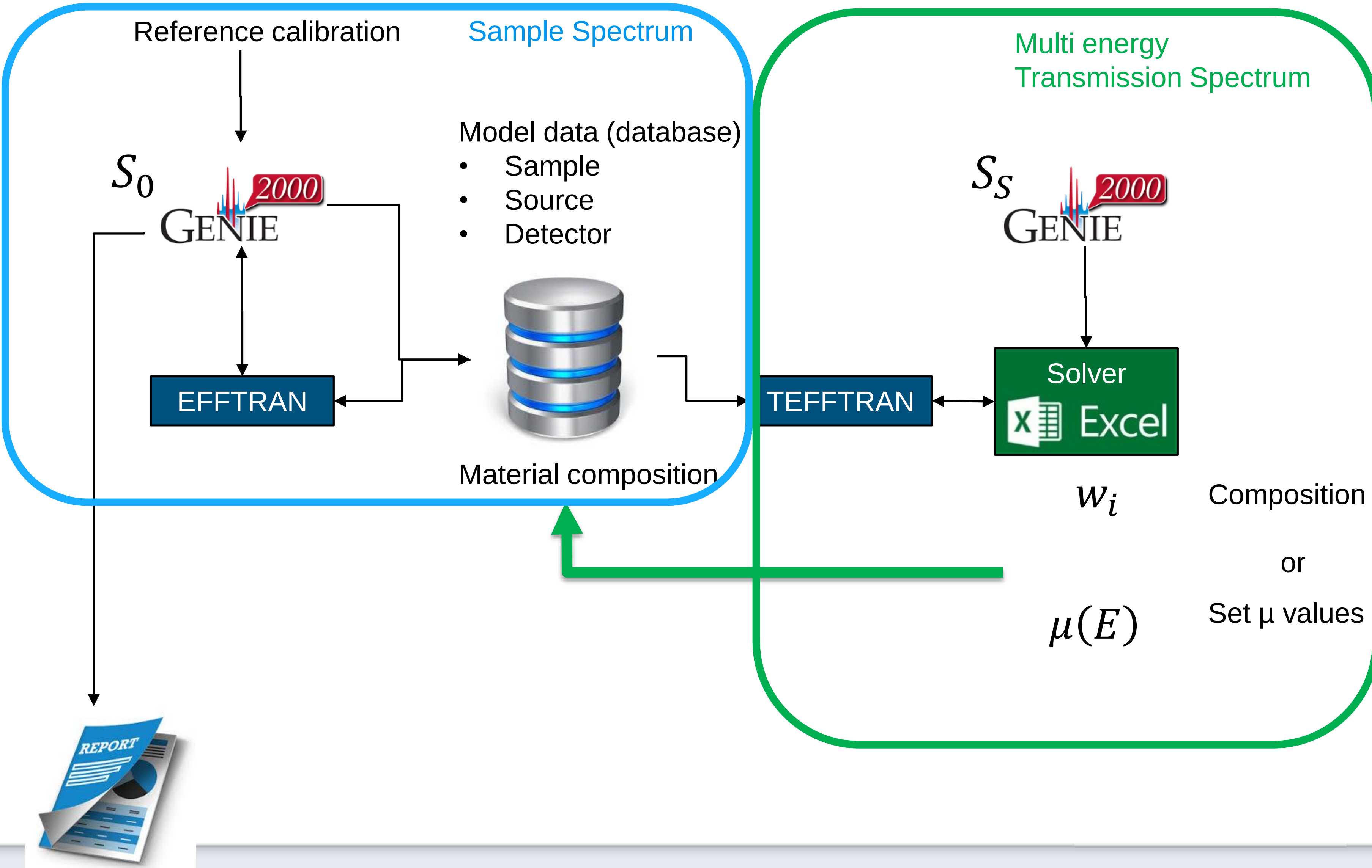
XCOM

- The set of elements should be well selected
  - Not complete periodic table
  - Element sets can be defined for specific materials (weak point)
- Least squares problem can be solved in e.g. Excel (Solver add-in)
- Outcome is not a polynomial but a sample composition
  - Inter & extrapolation (by XCOM)
  - K-edges are included (if elements data set is correct)
- The computed sample composition is not necessary the correct composition but yields an equivalent composition

TEFFTRAN: the way to µ-values from uncollimated experiments

- Uses computational procedures like in EFFTRAN
- Considers a cylindrical source (dimensions)
- Source is on top of sample (cutshall geometry)
- Sample is a cylinder (all walls same thickness, composition and density are to specify
- Detectormodel as in EFFTRAN
- No absolute response but a ratio of two computations at different density and composition is to be compared with the experimental ratio
- TEFFTRAN can be run from a batch command once the input files are defined
  - In an iteration process the µ-value can be obtained from the simple transmission measurement

Procedure for low-energy attenuation correction based on TEFFTRAN and EFFTRAN



Combining transmission data and element composition

- To fill the gap of missing elements in the element composition (H-Na, Mg-S)
- Use the measured heavy elements to compose a basic composition
  - Refine the composition with missing elements, H, O, C,...
  - Compare the measured μ-data with the computed μ-data
- Make efficiency transfer with the optimized composition
- Uncertainty component $\Delta\mu \rightarrow \Delta CF$

Conclusion

- Gamma-ray attenuation data can be obtained from a transmission experiment without collimator using an appropriate modeling software of TEFFTRAN
- Multi Energy Transmission data only do not give full details on actual gamma-attenuation
  - No means to account for K-edges
  - Determination of an equivalent composition with an appropriate set of elements may (partially) cope with K-edges
  - A combination of transmission + the measurement of the element composition of the sample material (X-ray,…) may result in the most complete information
- A procedure for efficiency transfer relaying on EFFTRAN and TEFFTRAN was proposed

Thank you for your attention