SCK CEN

Belgian Nuclear Research Centre

# Measuring mass attenuation coefficients for materials with unknown composition by performing transmission measurements with a HPGe detector for X-rays and low-energy gamma rays

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Laboratory for gammaspectrometry

Sample types - Gamma lims – efftran – Genie2K

Materials and vials

Calibration curves with transmission measurements

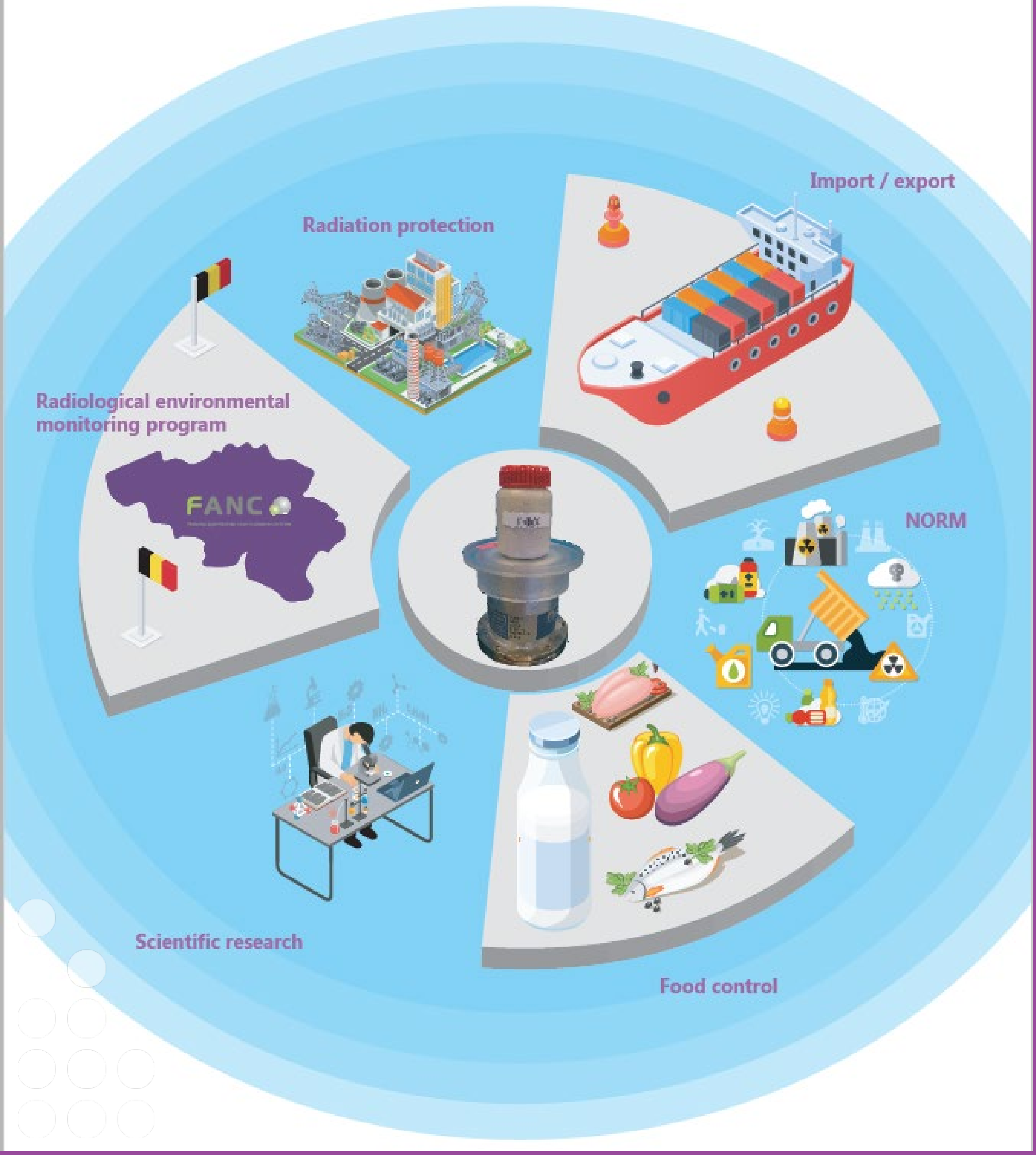
Implementation in routine lab

Conclusion

# Laboratory for gammaspectrometry

## 20 HPGe det >> 5 Low Energy, 1 Well typen, LEGE and 12 coaxial

- Home made gamma lims
- Automatically N2 filling system
- Weighing balance with online registration
- Digital signal analyzer LYNX (Mirion)

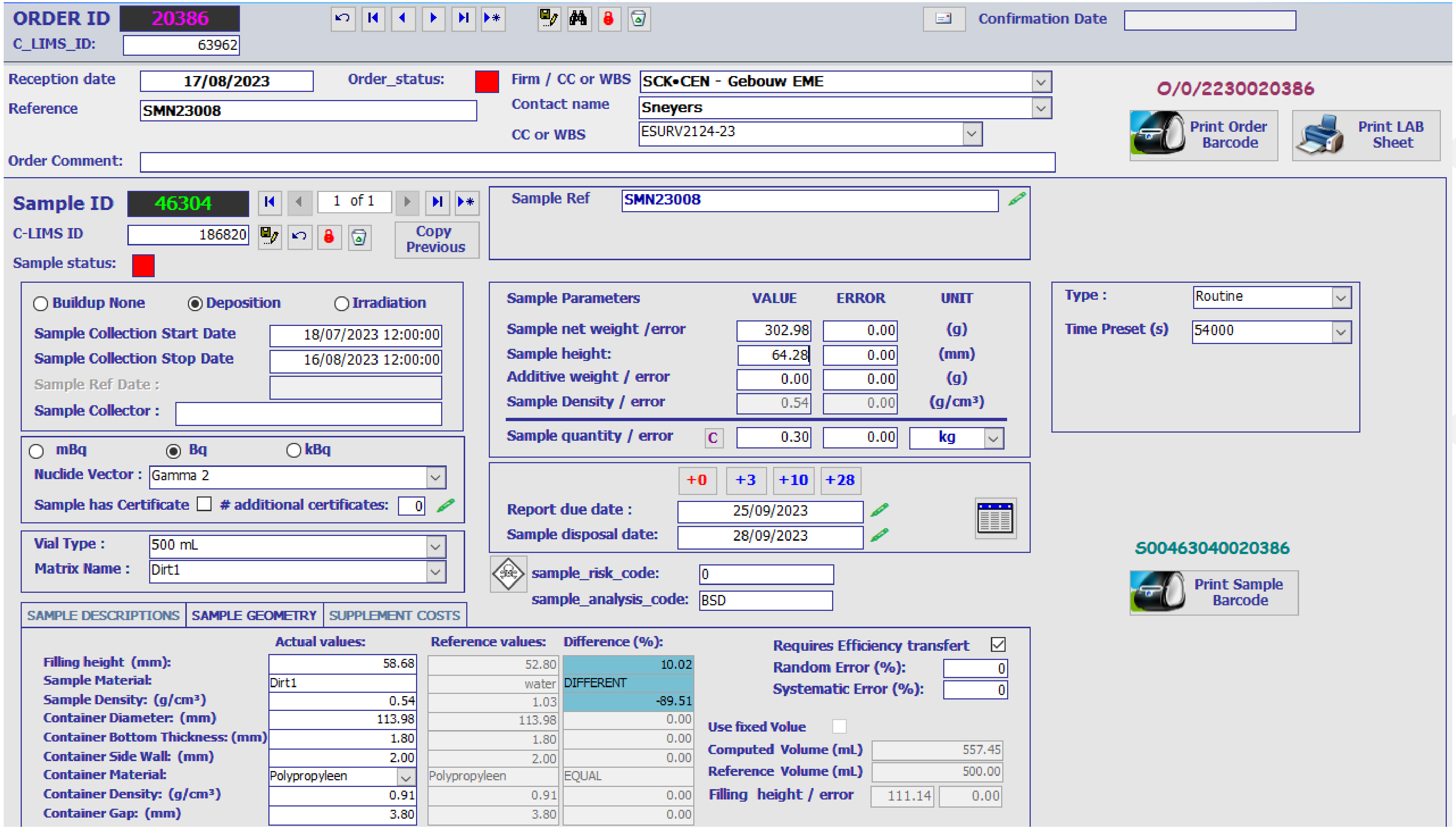



# Samples

## Different geometries

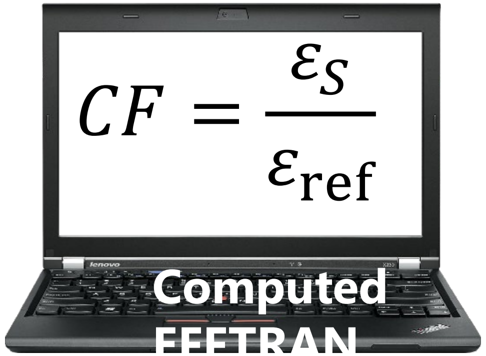
- +/- 3500 samples per year
- Weekly QA and monthly Background
- Measurement in different geometries
- Volumes from 4 - 2400 ml
- Filters
- Cartridges
- …

# Gamma lims


ORDER ID 20386
C_LIMS_ID: 63962
Confirmation Date
Reception date 17/08/2023
Order_status:
Firm / CC or WBS SCK•CEN - Gebouw EME
Reference SMN23008
Contact name Sneyers
CC or WBS ESURV2124-23
Order Comment:
O/0/2230020386
Print Order Barcode
Print LAB Sheet
Sample ID 46304
1 of 1
C-LIMS ID 186820
Copy Previous
Sample status:
Sample Ref SMN23008
Buildup None
Deposition
Irradiation
Sample Collection Start Date 18/07/2023 12:00:00
Sample Collection Stop Date 16/08/2023 12:00:00
Sample Ref Date :
Sample Collector :
Sample Parameters
VALUE
ERROR
UNIT
Sample net weight /error 302.98 0.00 (g)
Sample height: 64.28 0.00 (mm)
Additive weight / error 0.00 0.00 (g)
Sample Density / error 0.54 0.00 (g/cm³)
Sample quantity / error C 0.30 0.00 kg
Type : Routine
Time Preset (s) 54000
mBq
Bq
kBq
Nuclide Vector : Gamma 2
Sample has Certificate
# additional certificates: 0
+0 +3 +10 +28
Report due date : 25/09/2023
Sample disposal date: 28/09/2023
S00463040020386
Print Sample Barcode
Vial Type : 500 mL
Matrix Name : Dirt1
sample_risk_code: 0
sample_analysis_code: BSD
SAMPLE DESCRIPTIONS
SAMPLE GEOMETRY
SUPPLEMENT COSTS
Actual values:
Reference values:
Difference (%):
Filling height (mm): 58.68 52.80 10.02
Sample Material: Dirt1 water DIFFERENT
Sample Density: (g/cm³) 0.54 1.03 -89.51
Container Diameter: (mm) 113.98 113.98 0.00
Container Bottom Thickness: (mm) 1.80 1.80 0.00
Container Side Wall: (mm) 2.00 2.00 0.00
Container Material: Polypropyleen Polypropyleen EQUAL
Container Density: (g/cm³) 0.91 0.91 0.00
Container Gap: (mm) 3.80 3.80 0.00
Requires Efficiency transfert
Random Error (%): 0
Systematic Error (%): 0
Use fixed Value
Computed Volume (mL) 557.45
Reference Volume (mL) 500.00
Filling height / error 111.14 0.00

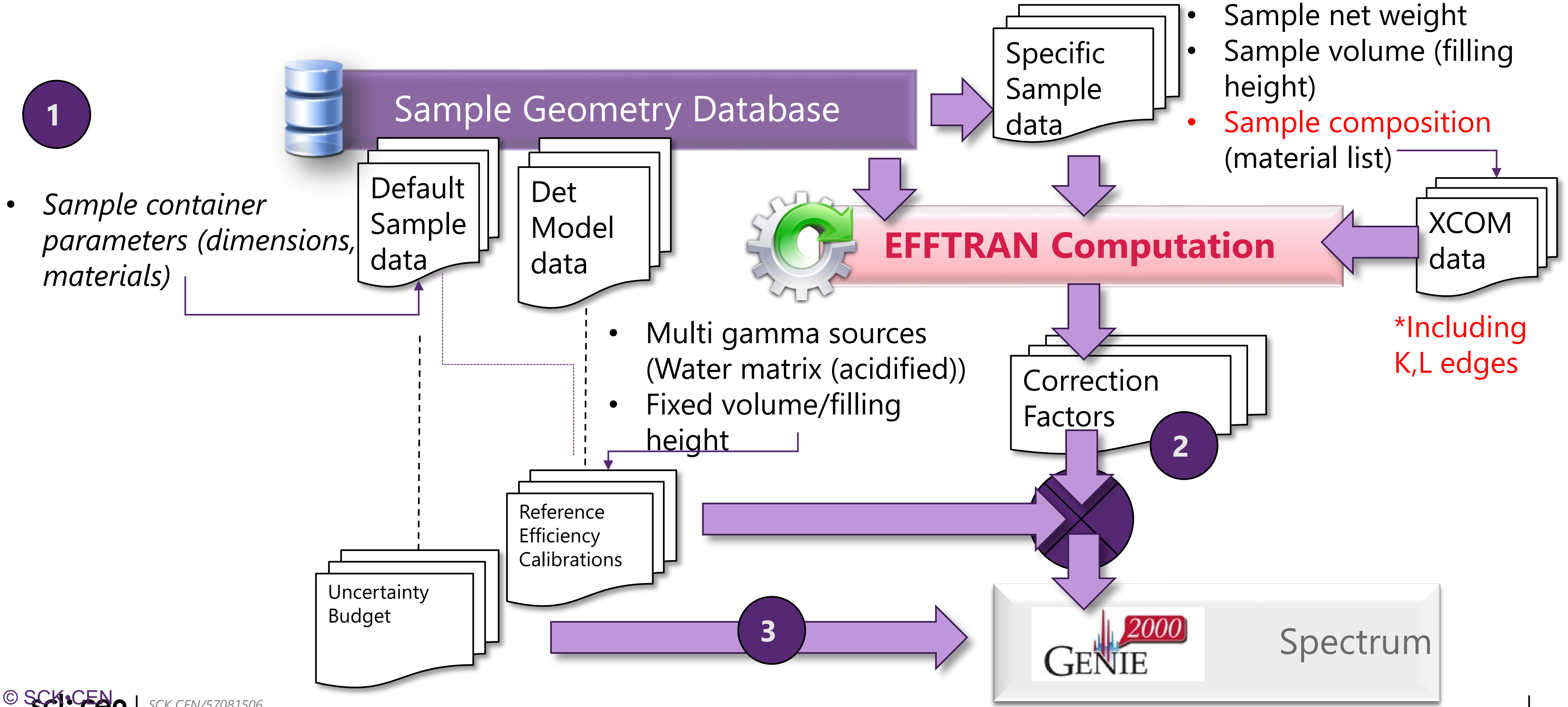
# Efficiency transfer for routine gamma-ray analyses

$$\varepsilon_S(E) = CF(E)\varepsilon_{\text{ref}}(E)$$




- Efficiency transfer → systematic corrections for various counting parameters deviating from reference calibration (EFFTRAN)
- Systematically in each analysis
  - Filling height (if no 100% filling is used)
  - Apparent sample density
  - Sample material
    - **chosen from list** for well known materials
    - Requires specific element composition when gamma-ray energy **(30-100 keV)**
  - Any other sample parameter that is different from the reference

# Efficiency Transfer and Uncertainty Budget *imported in Genie 2000*



# Uncertainty polynomial

## *Known material versus material class*

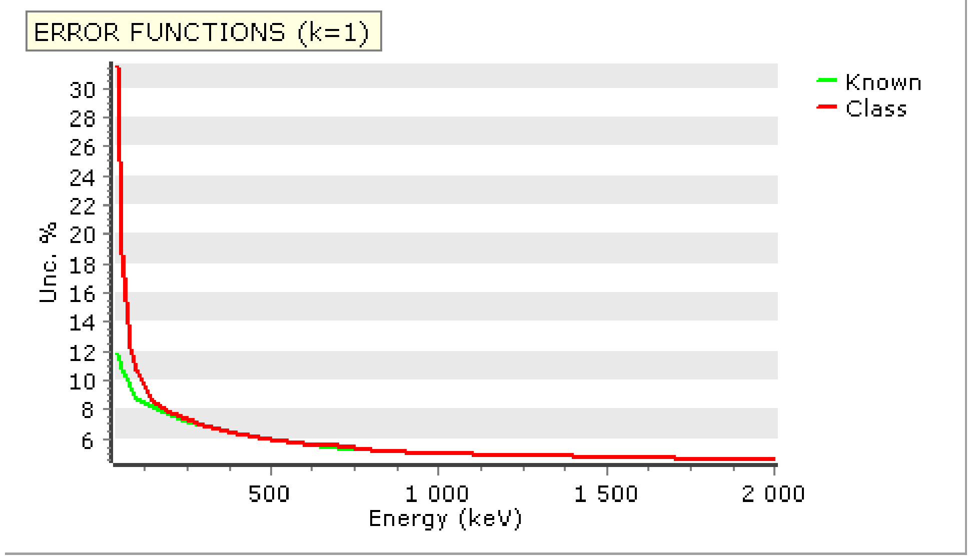
- Known
  - Water
  - PE
  - metals, …
- Class
  - Organic matter
  - Soil, dirt

**ERROR PARAMETERS*** Show

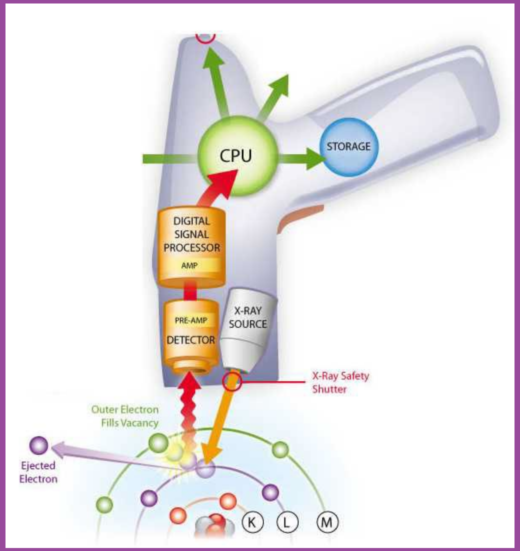
*The error parameters define a polynomial Unc(Energy) that corresponds to the expanded uncertainty, excluding: counting statistics, background correction, uncertainty on intensity

| | LOW | HIGH | |
|---|---|---|---|
| ERR_0: | -2.64190897668886 | -0.217495281069351 | K |
| ERR_1: | 2.40934617013547 | 0.153041143179349 | N |
| ERR_2: | -0.809828432839497 | -3.77714915855748E-02 | O |
| ERR_3: | 0.119617242483509 | 4.0037761660443E-03 | W |
| ERR_4: | -6.56724737353403E-03 | -1.55692968480513E-04 | N |

| | LOW | HIGH | |
|---|---|---|---|
| ERR_0: | 46.2408477481583 | -0.212328033862284 | C |
| ERR_1: | -38.4042839799321 | 0.151965311319783 | L |
| ERR_2: | 11.9564513751801 | -3.79126312099868E-02 | A |
| ERR_3: | -1.65299820997481 | 4.05003978683371E-03 | S |
| ERR_4: | 8.56024431411862E-02 | -1.58424173065154E-04 | S |

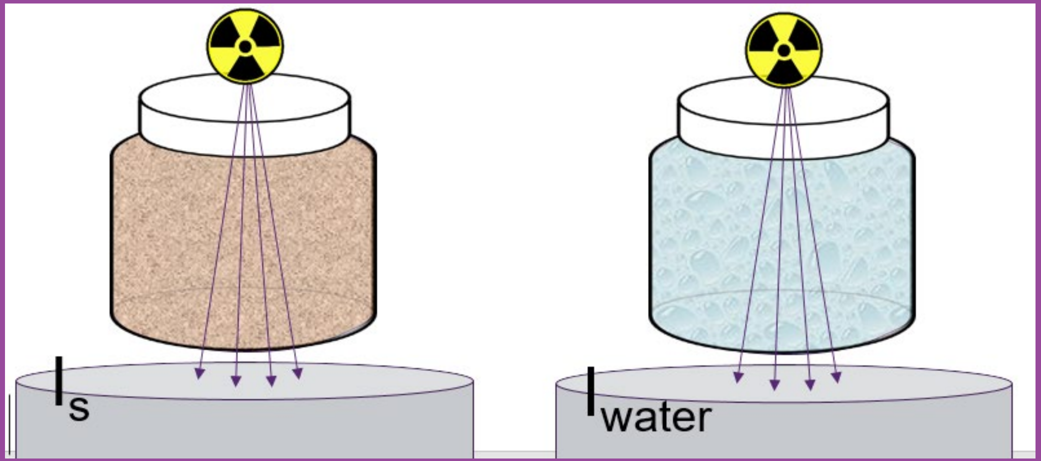



- Error curve for each counting geometry
- Filling height, diameter, bodem thickness, sample density, positioning, curve fitting, calibration source, system drift, gamma intensity, counting loses, …

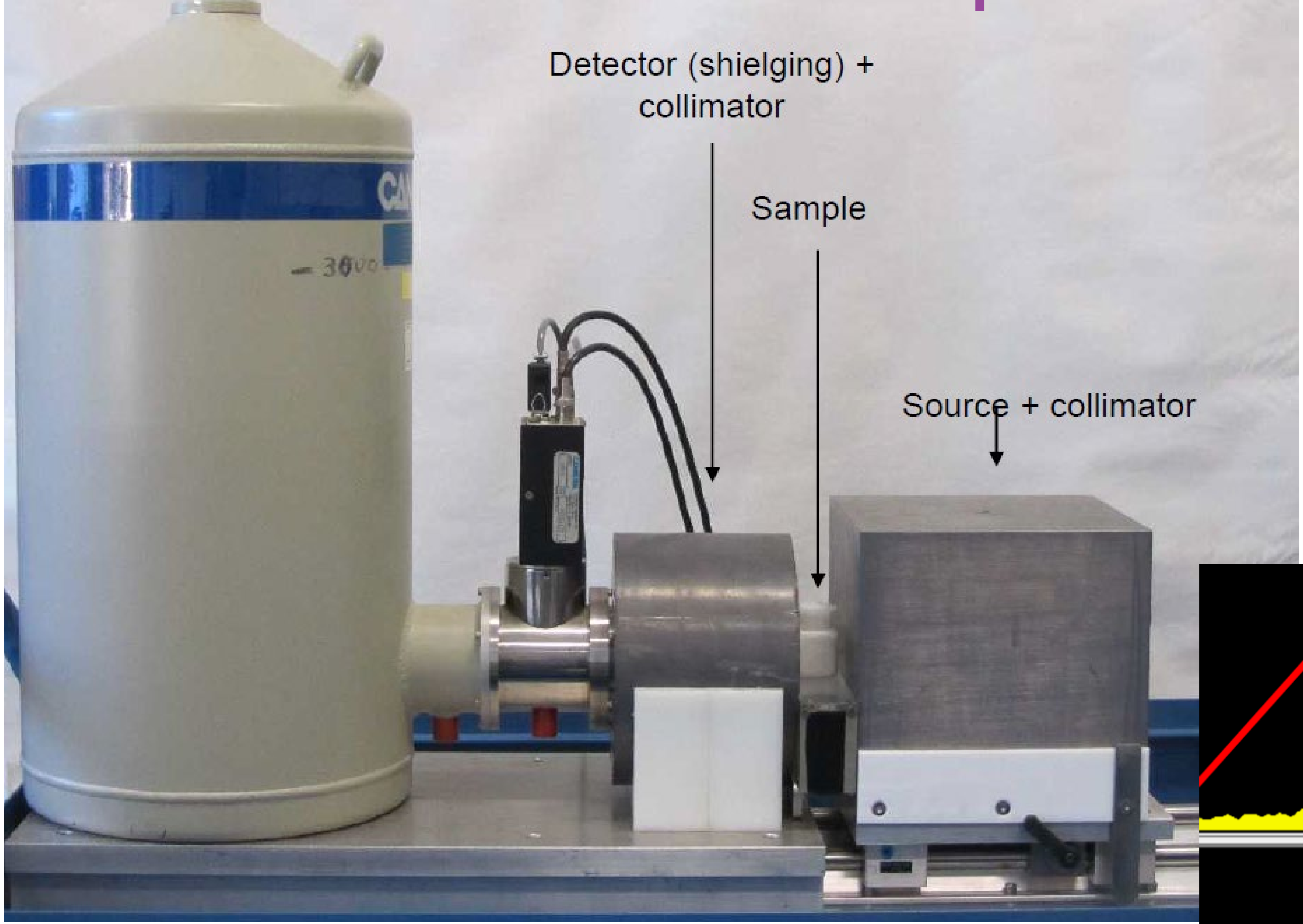


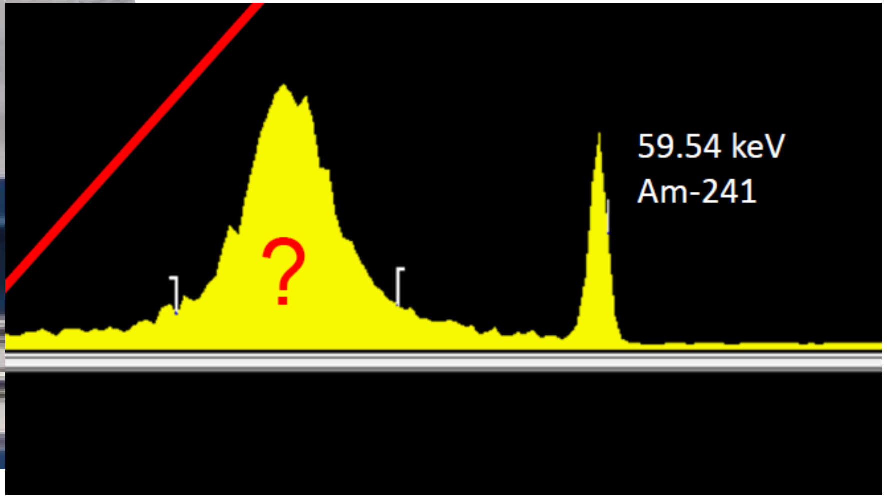

# Means to determine sample attenuation

- Generic compositon
- Comparison with representative standards
  - Not very practical
- XRF-handheld monitor, ICP-MS, …
  - Specify sample material for the EFFTRAN model >> elements from Mg → U >> ? O, H
- Multi Energy gamma-ray transmission
  - Absolute measurement using collimated beam
  - Relative attenuation measurement using no collimation



# Previous transmission setup

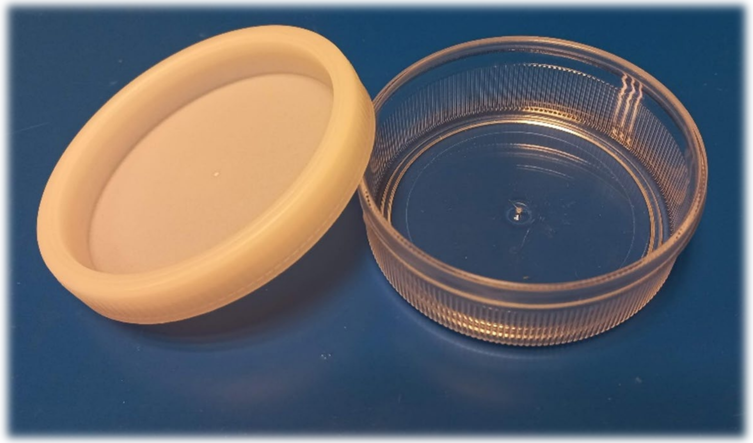


# Calibration curves with transmission measurements

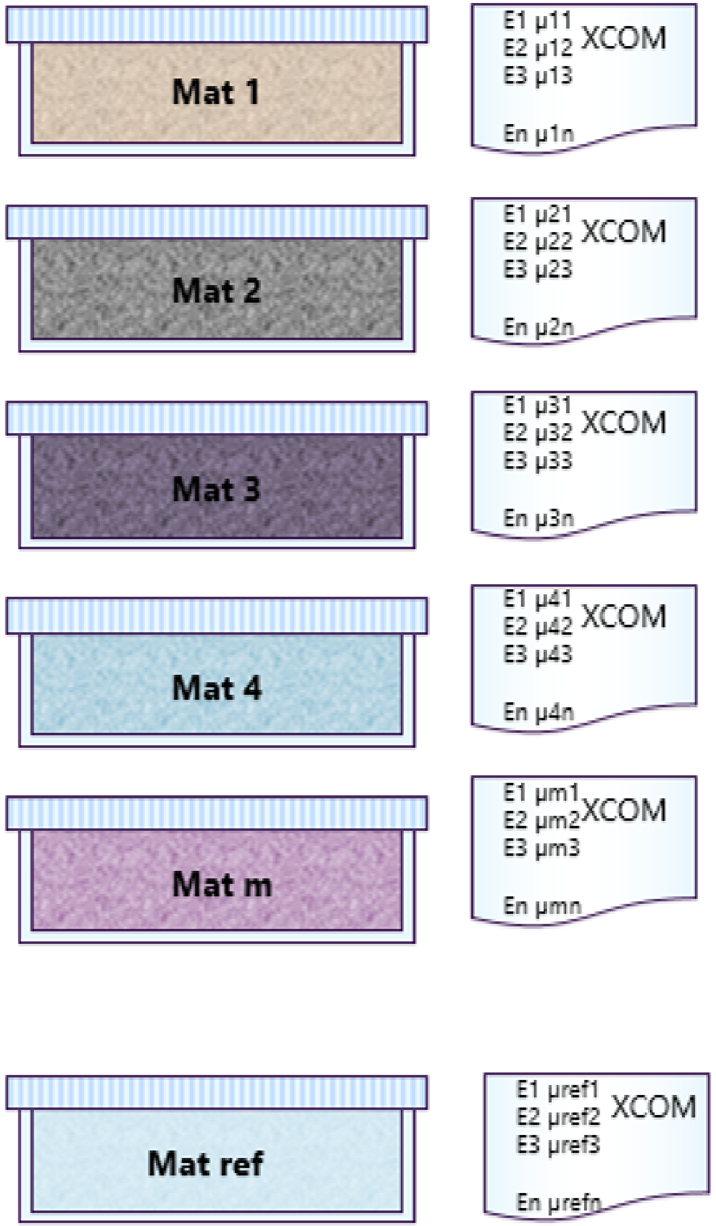
## Ba-133 and Am-241

- X- and $\gamma$-rays in the range (21.2 - 303 keV)
- on a set of well known sample materials for which the mass attenuation coefficients are derived from XCOM
- Vial: thickness 10 mm

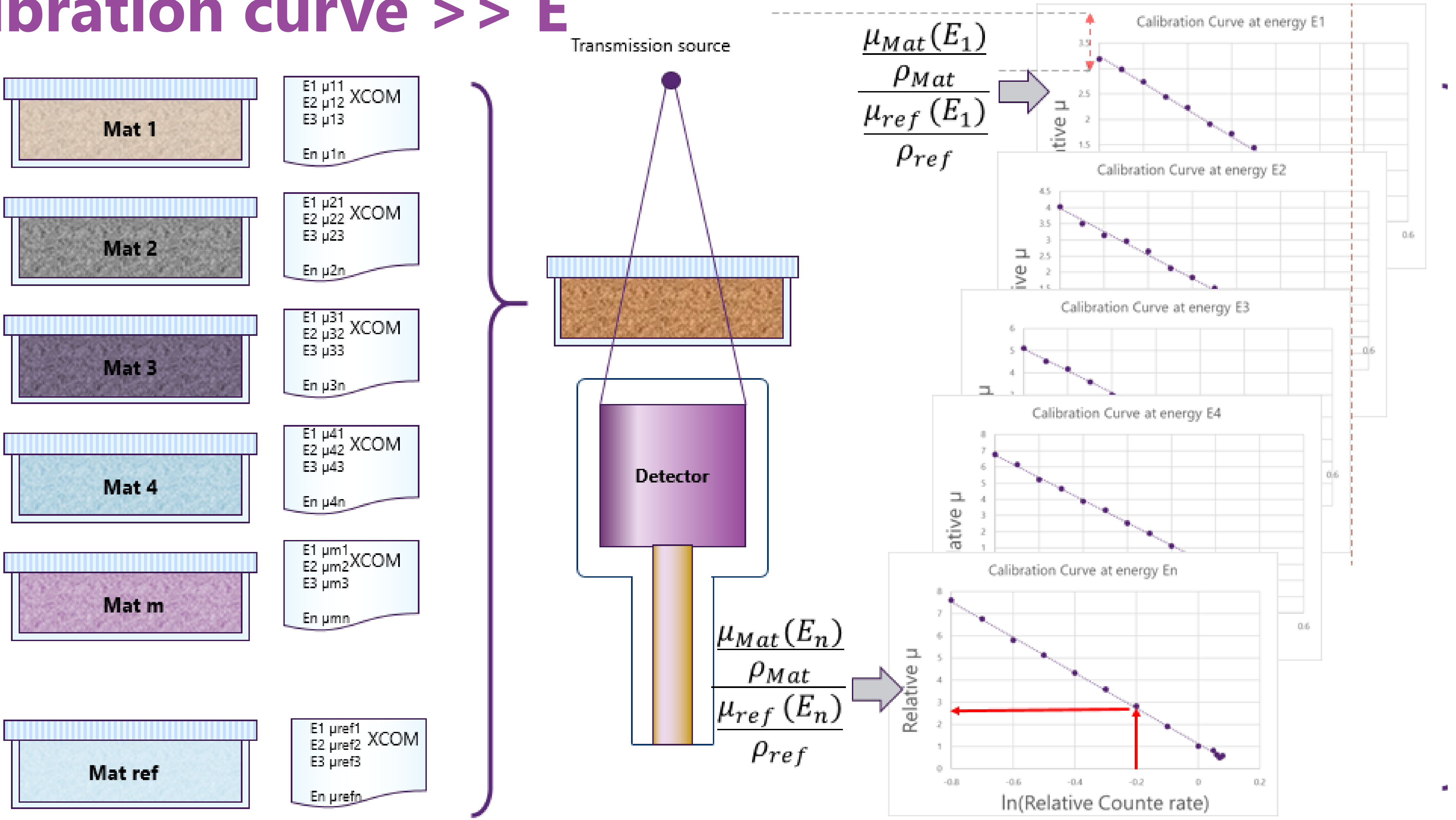


# Materials

- *Set of materials with well-known chemical composition for which the attenuation data at the transmission energies are computed with XCOM.*
- *Salts, plastics, metals, …*

# Calibration curve >> E



# Calibration curve >> mass att. file

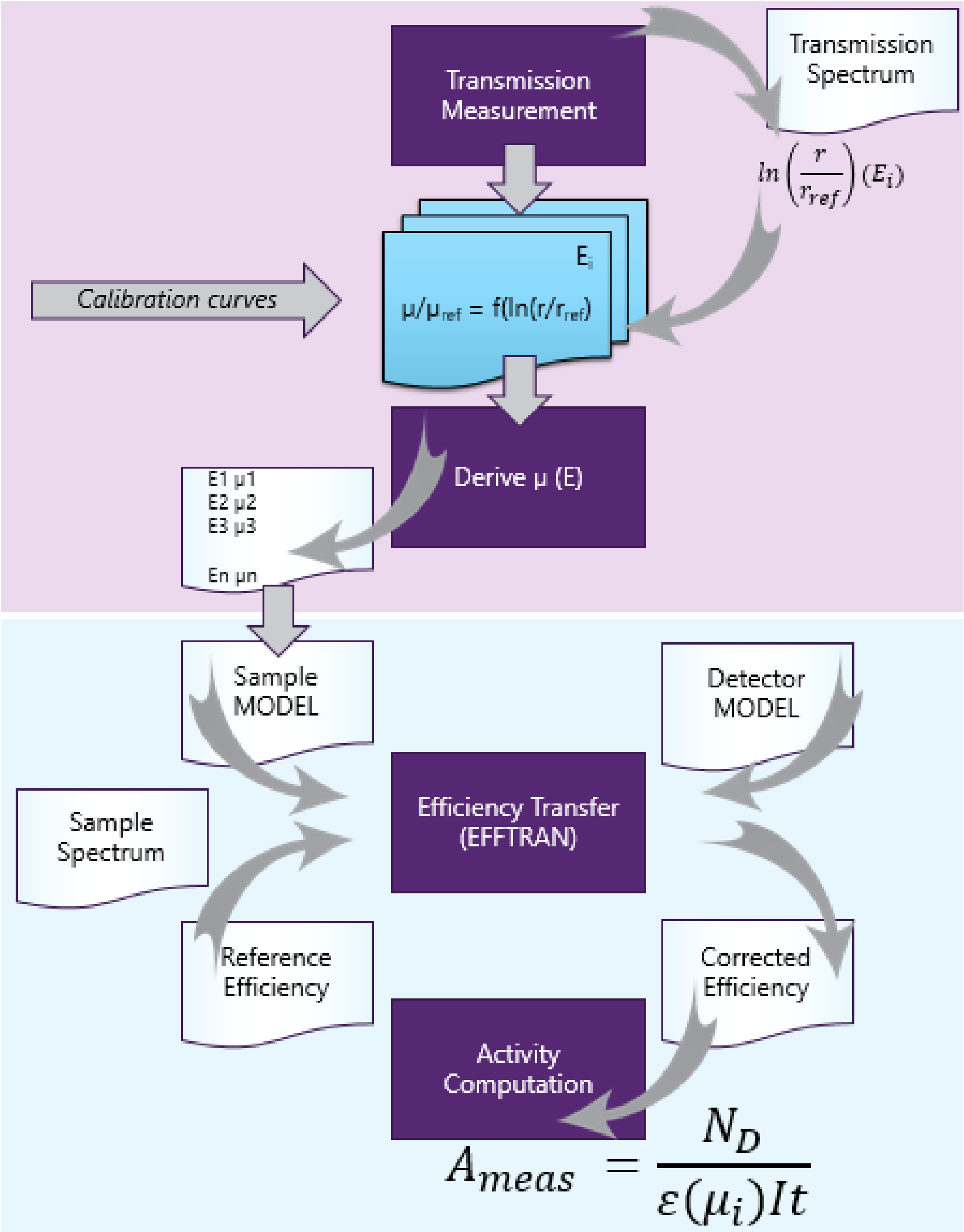
- calibration curves

$$\mu_E\left(ln\left(\frac{r}{r_{ref}}\right)\right)_{rel} = a_E ln\left(\frac{r}{r_{ref}}\right) + b_E$$

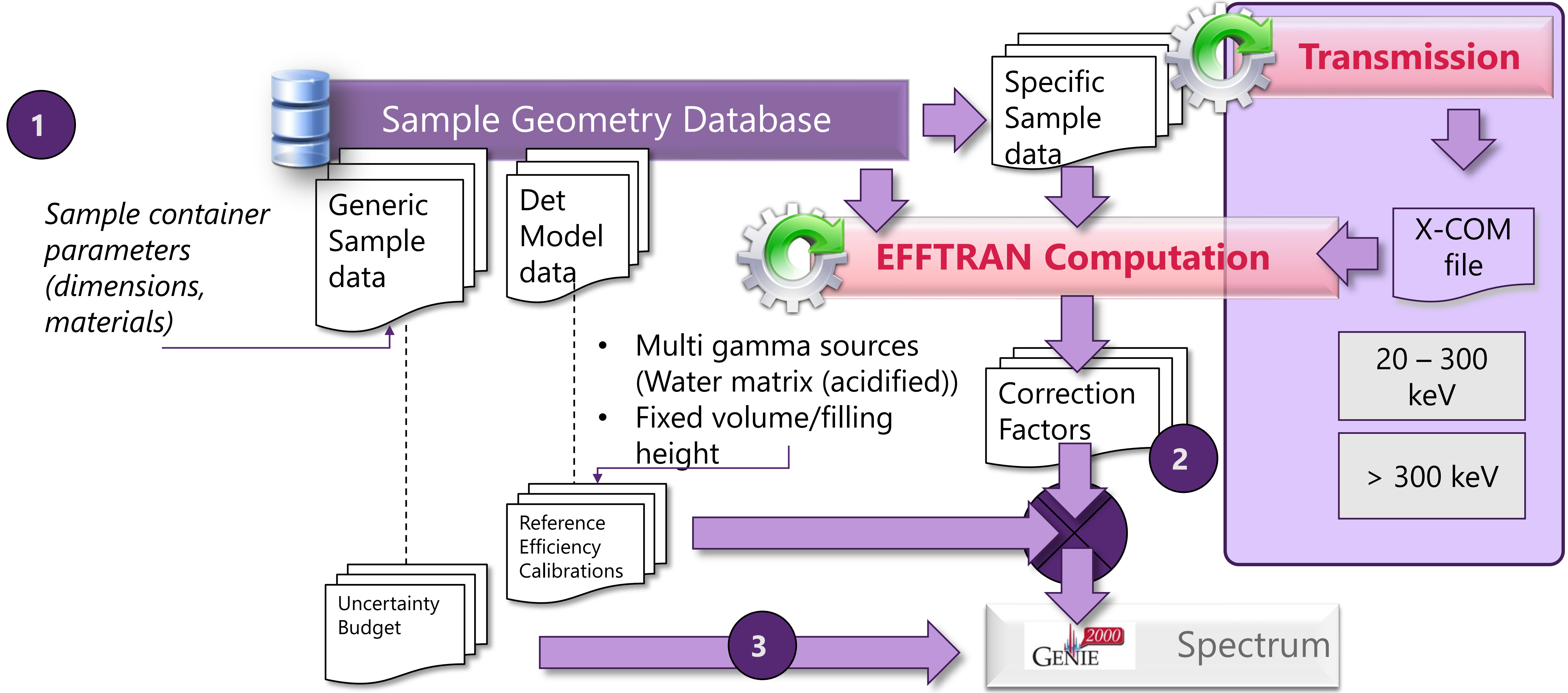
- $\frac{r}{r_{ref}}(E)$ >> transmission trough a material relative to the count rate for transmission trough the reference material (water in our case)
- Unknwon material X >> mass attenuation coefficients can then be determined from the calibration curves as

$$\mu_X = \mu_0 \frac{\rho_{ref}}{\rho_X}\left(a_E(E)\ln\left(\frac{r_X}{r_{ref}}\right) + b_E\right)$$

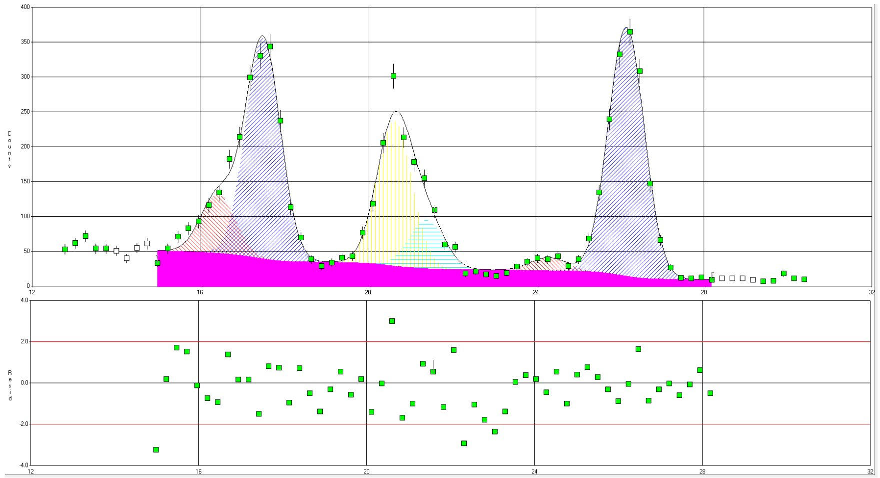
→ a mass attenuation file can be set up for an unknown material

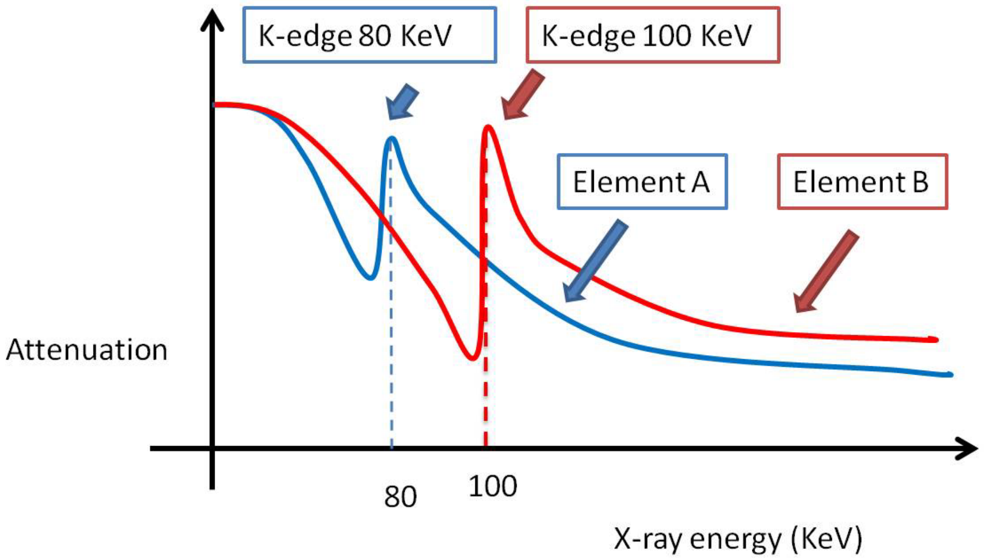


# Efficiency Transfer procedure with measured µ-data

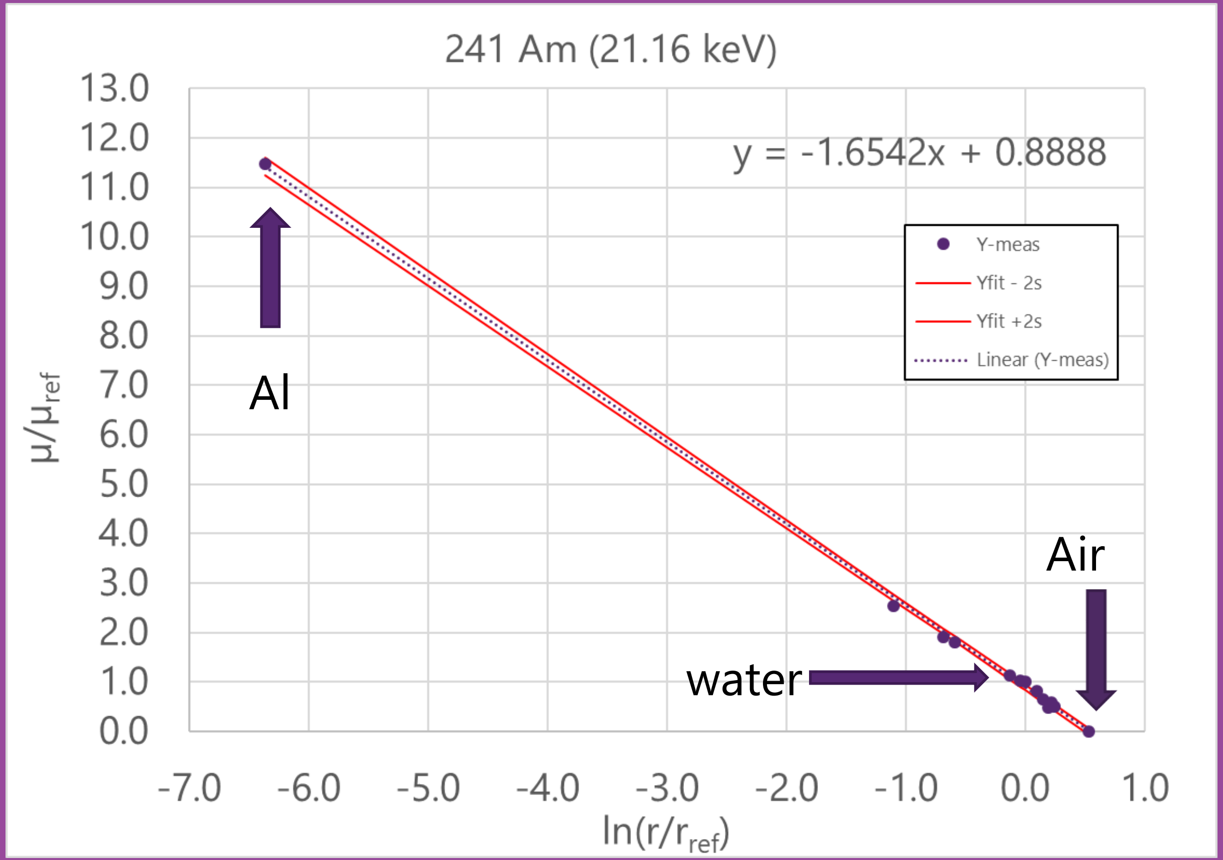


# Transmission spectrum






- *20 keV X-ray complex of Am-241 through a PTFE sample ($\rho$ = 2.2 g/cm²)*
- *Attention for a good fit!*
- *K-edges, a source of discontinuities in gamma attenuation*




*Calibration curve for determining $\mu_{rel}$ at 21.6 keV from transmission relative to water. The materials air, water and aluminium are indicated. $\mu/\mu_{rel} = 1$ means $\mu$ is equal to the mass attenuation of water.*

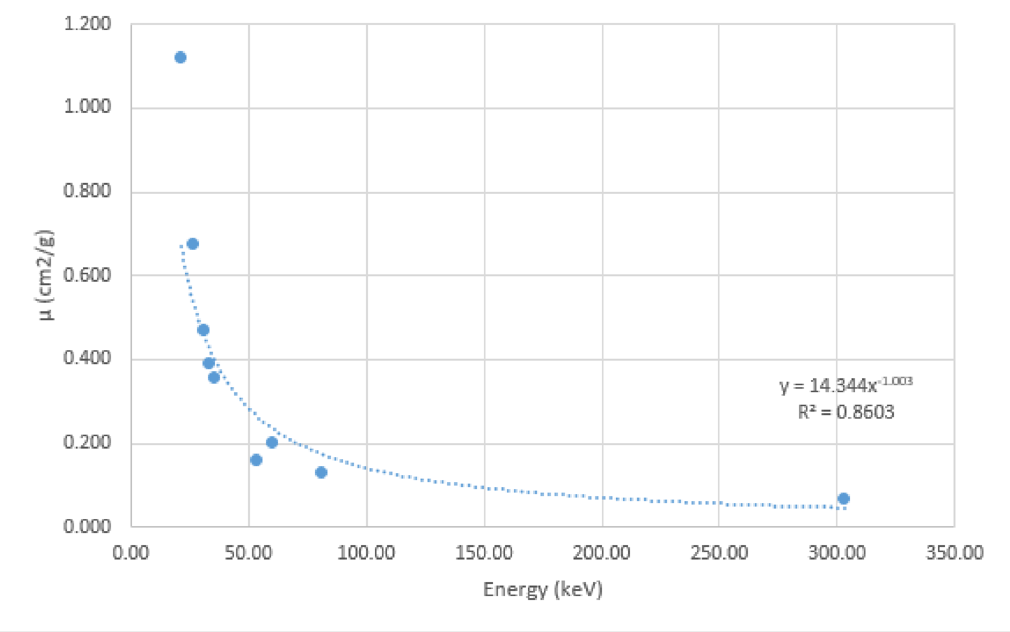
# Implementation routine

2 transmission measurements
Peak surface
Tranfer to mass attenuation file > .txt

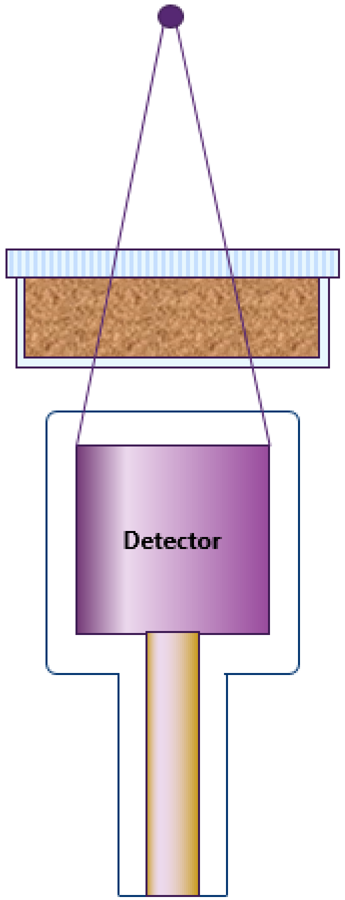
**TRANSMISSION CALCULATIONS**

| Parameters | |
|---|---|
| Reference | Na2CO3 |
| Date | 20/05/2022 |
| Source | Am + Ba |
| Density (g/cm3) | 1.202 |
| Live time Am241 (s) | 4201.9 |
| Live time Ba133 (s) | 4201.9 |

| Source | Energy (keV) | Peak surface (cts) | µ (cm2/g) | Relative µ uncertainty |
|---|---|---|---|---|
| Am241 | 21.16 | 9.30E+04 | 1.120 | 0.028 |
| Am241 | 26.34 | 1.10E+05 | 0.678 | 0.272 |
| Ba133 | 30.97 | 1.30E+06 | 0.469 | 0.919 |
| Am241 | 33.20 | 8.10E+03 | 0.389 | 0.258 |
| Ba133 | 35.05 | 3.54E+05 | 0.356 | 1.045 |
| Ba133 | 53.16 | 3.68E+04 | 0.161 | 0.484 |
| Am241 | 59.54 | 3.21E+06 | 0.203 | 0.064 |
| Ba133 | 81.00 | 6.89E+05 | 0.133 | 0.178 |
| Ba133 | 303.00 | 1.77E+05 | 0.069 | 1.180 |



# Conclusion




- The transmission source should be at a distance of at least 20 cm from detector to avoid true coincidence summing effects and measurements are best made with Ba-131 and Am-241 sources sequentially;
- Attenuation parameters can be determined with a precision of a few percent depending on the quality of the calibration curve and the complexity of the spectra;
- No K or L edges in the attenuation of the sample material can be determined with this method;
- The method may fail if spectra are dominated by X-ray fluorescence peaks of the sample material.

# Thank you for your attention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Belgian Nuclear Research Centre

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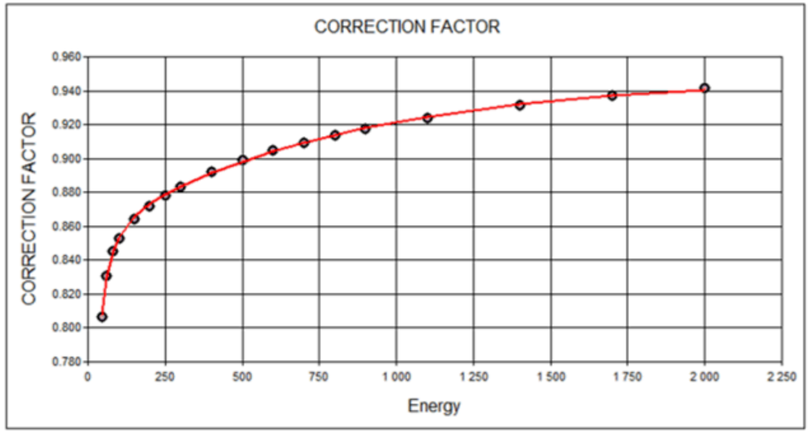
# Efftran Transfer data model >> correction function

**EFFTRAN Computation**
**Actual Sample ↔ Reference Sample**




E1 → CF(E1)
E2 → CF(E2)
E3 → CF(E3)
E4 → CF(E4)
....
En → CF(En)

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

$\varepsilon_{sam} = \varepsilon_{ref}$ **CF**

Spectrum (cam file data)

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

$$\text{Ln } \varepsilon_{\text{corr}}(E) = \sum_{k=0}^{n} (A_k + C_k) \ln(E)^k$$

Applied to low and high-energy dual efficiency curve

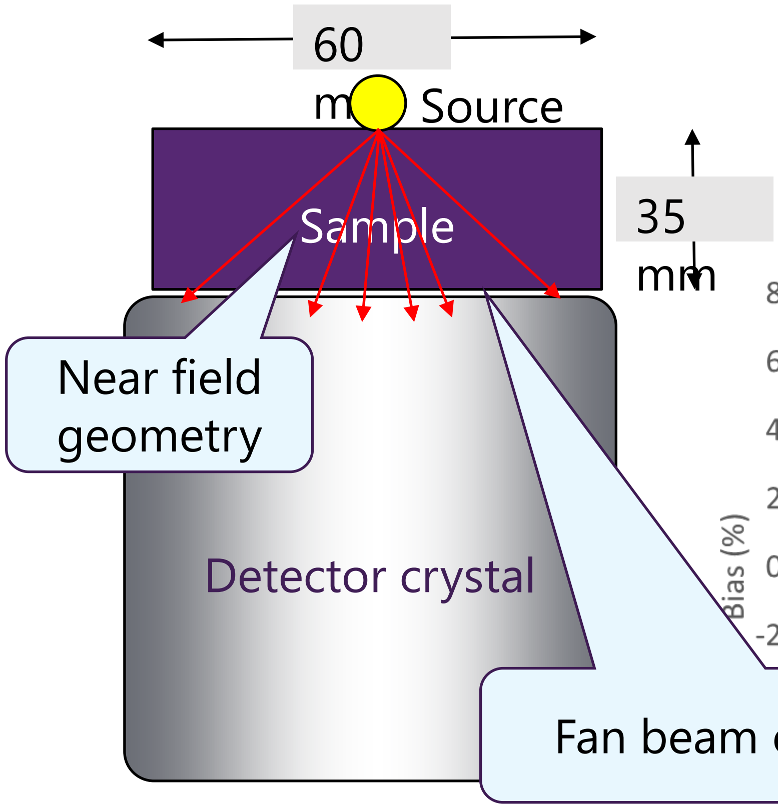
# Cutshall Attenuation Correction Procedures (two times wrong = right ?)

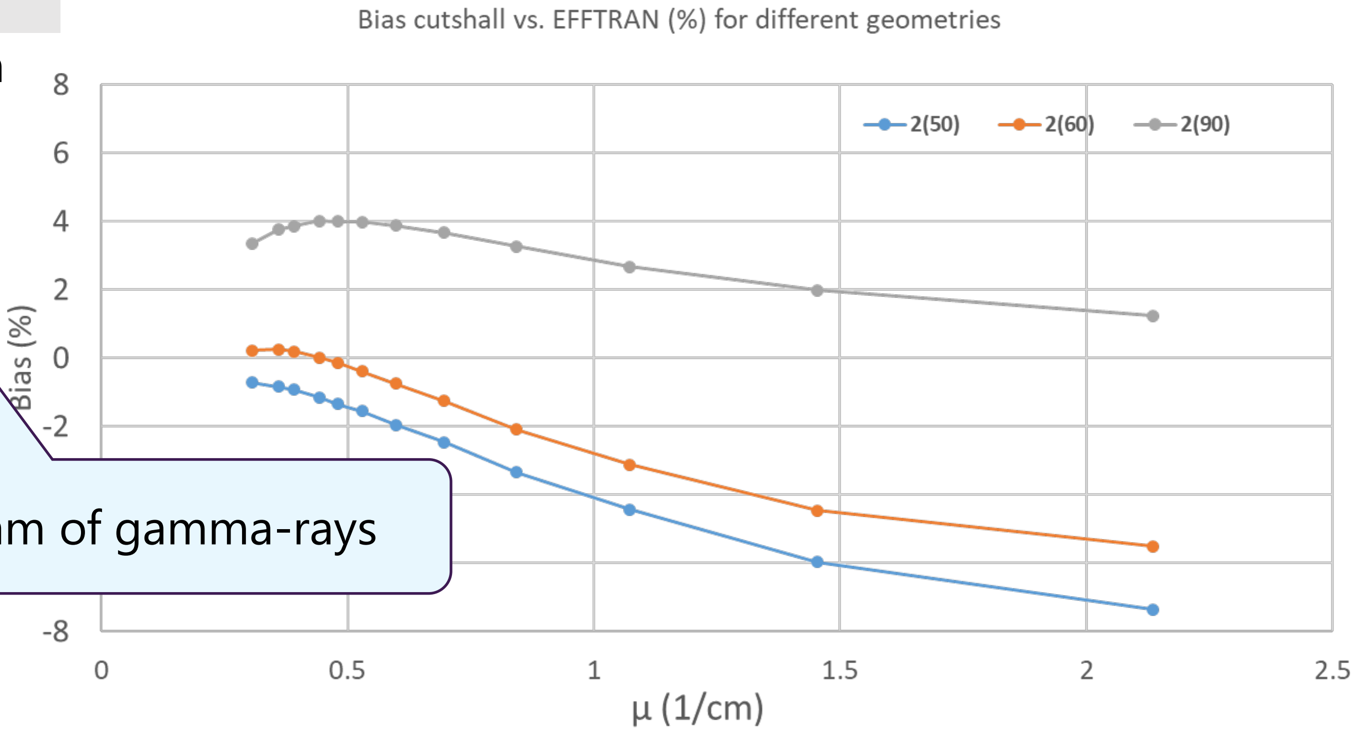
- N.H. Cutshall, correction for $^{210}Pb$ in sediments (1981)
- **Near field (wide angle)**

Needs parallel beam of gamma-rays !

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

Correction only valid for far field geometry !




Sediment sample in pillbox (20mm heigh at different diameters; 50mm, 60mm, 90mm)
Cutshall relative to air
On a 60mm x 60mm detector
But bias is limited!