International Atomic Energy Agency Technical Visit on

Coincidence summing and geometry correction in gamma spectrometry

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Session 5 – Self-attenuation corrections: Theory

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Layout

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   - Self-attenuation effects, self-attenuation correction factor

   - Applications

2. Sample composition and density

   – linear attenuation coefficient - XCOM

3. Simple analytical formulae for specific cases

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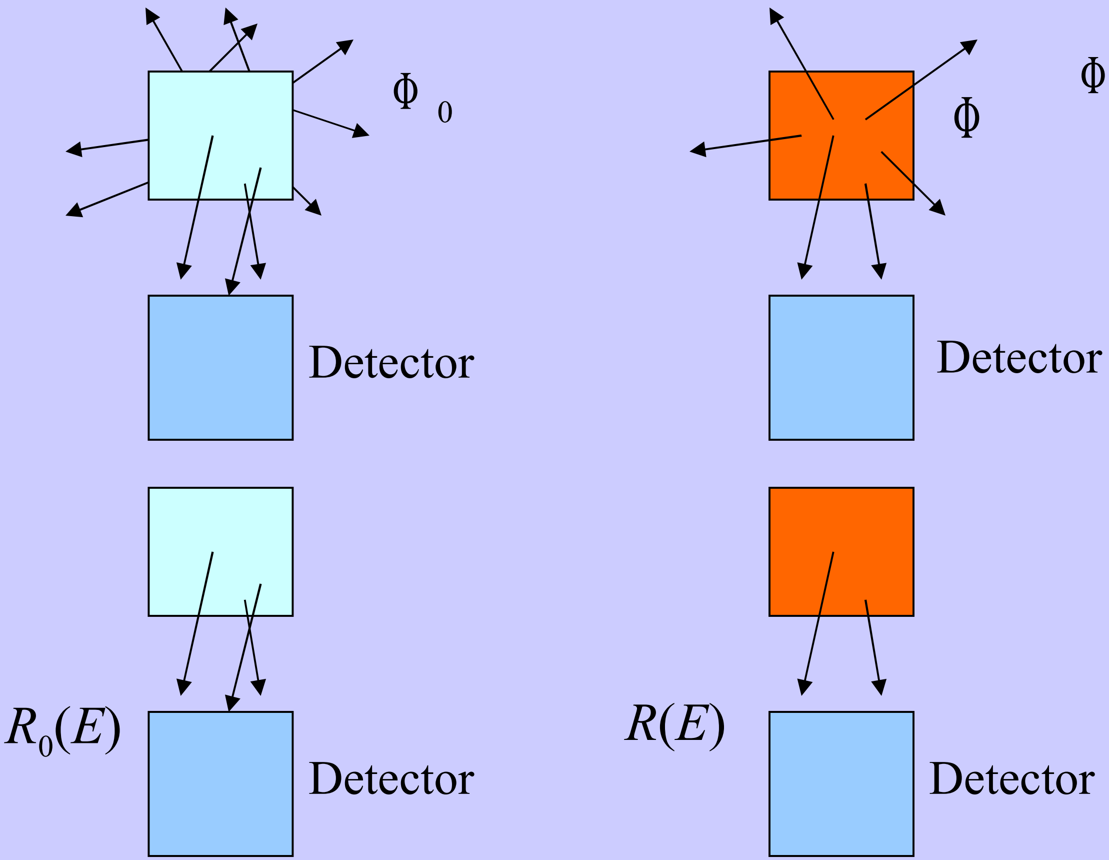
7. Summary

1. Introduction

Self-attenuation effects: the dependence of the peak count rate on photon interactions in the sample

$$F_a = \frac{R(E)}{R_0(E)}, \quad F_a \neq \frac{\Phi}{\Phi_0}$$



Φ , Φ_0 = Photon fluxes escaping from the samples without interactions

$R(E)$, $R_0(E)$ = Peak count rates for the sample and for a completely transparent sample (0)

Self-attenuation correction factor for a sample of matrix *m* with respect to a standard of matrix *s*:

$F_a(E; m; s) = \varepsilon(E; m)/\varepsilon(E; s)$

Useful for the computation of the efficiency for sample with matrix *m* on the basis of measured efficiency for sample *s* and of the computed value of the self-attenuation correction factor

$\varepsilon(E; m) = F_a(E; m; s)\, \varepsilon(E; s)$

Absolute self-attenuation factor – self-attenuation correction factor with respect to a vacuum sample

$F_{a0}(E; m; 0) = \varepsilon(E; m)/\varepsilon(E; 0)$ 0 for vacuum sample

-Depends essentially on:

- -photon linear attenuation coefficient in the sample μ
- -Dimensions of the sample

Applications

=> Computation of the efficiency for a sample with matrix m on the basis of a standard with a different matrix s:

$\varepsilon(E; m) = F_a(E; m; s)\ \varepsilon(E; s)$

=> Compatibility test of reference sources with the same geometry but different matrices m_1, m_2, .., m_k

$\varepsilon_{(1)}(E; 0) = F_a(E; 0; m_1)\ \varepsilon(E; m_1)$

$\varepsilon_{(2)}(E; 0) = F_a(E; 0; m_2)\ \varepsilon(E; m_2)$

The values $\varepsilon_{(1)}(E; 0)$, $\varepsilon_{(2)}(E; 0_2)$ … should be compatible.

The best value of $\varepsilon(E; 0)$ is their weighted average if all are compatible.

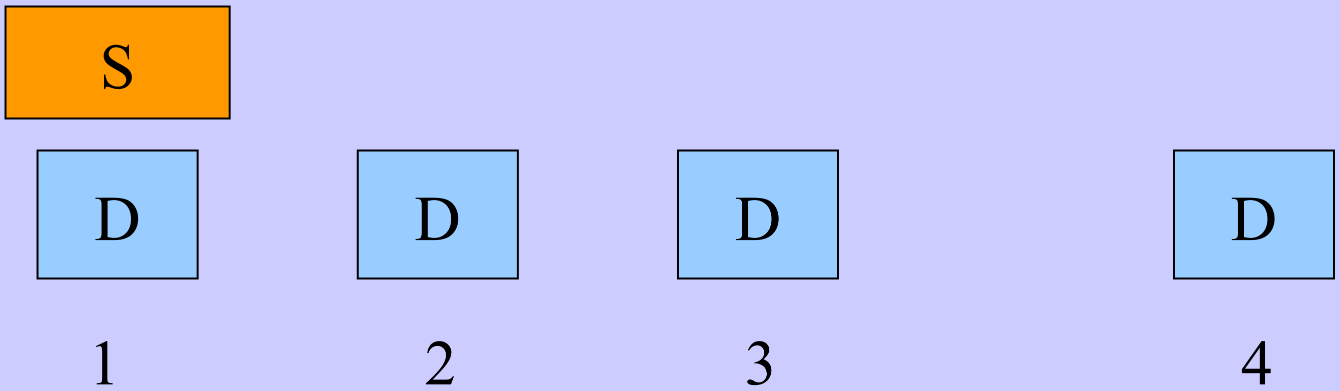
This best value should be used for the computation of the efficiency for other matrices

=> Estimation of the efficiency for a bulk sample with a volume higher than the volume of available certified reference material (CRM)

Sources bigger than the CRM available

In the case of vacuum sources the count-rate (CR) for the big source satisfies:

$$CR_4(3S) = CR_1(S) + CR_2(S) + CR_3(S)$$



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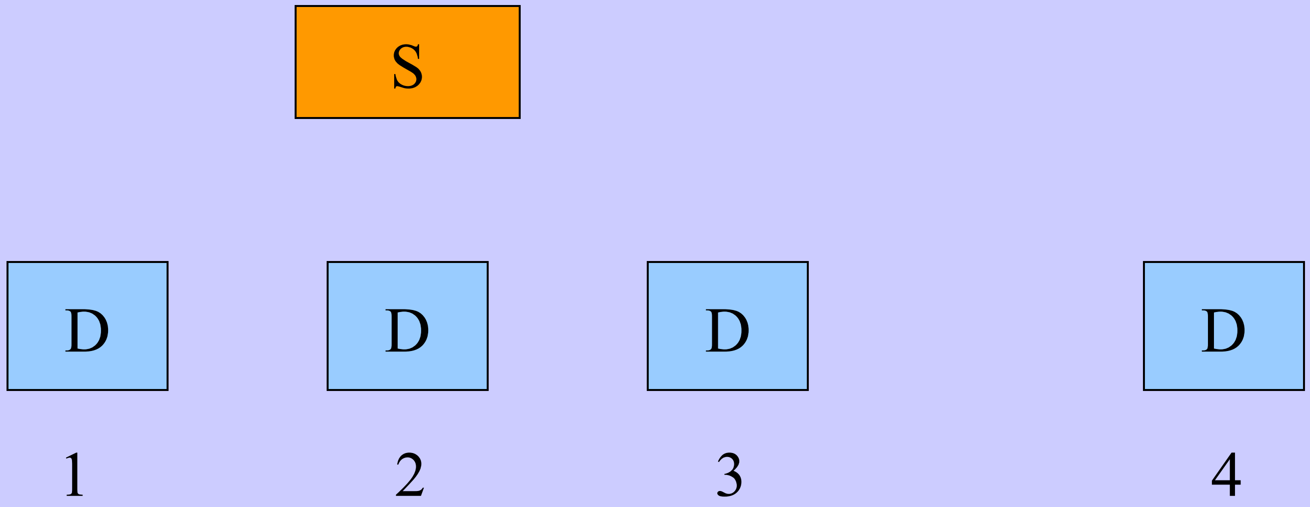

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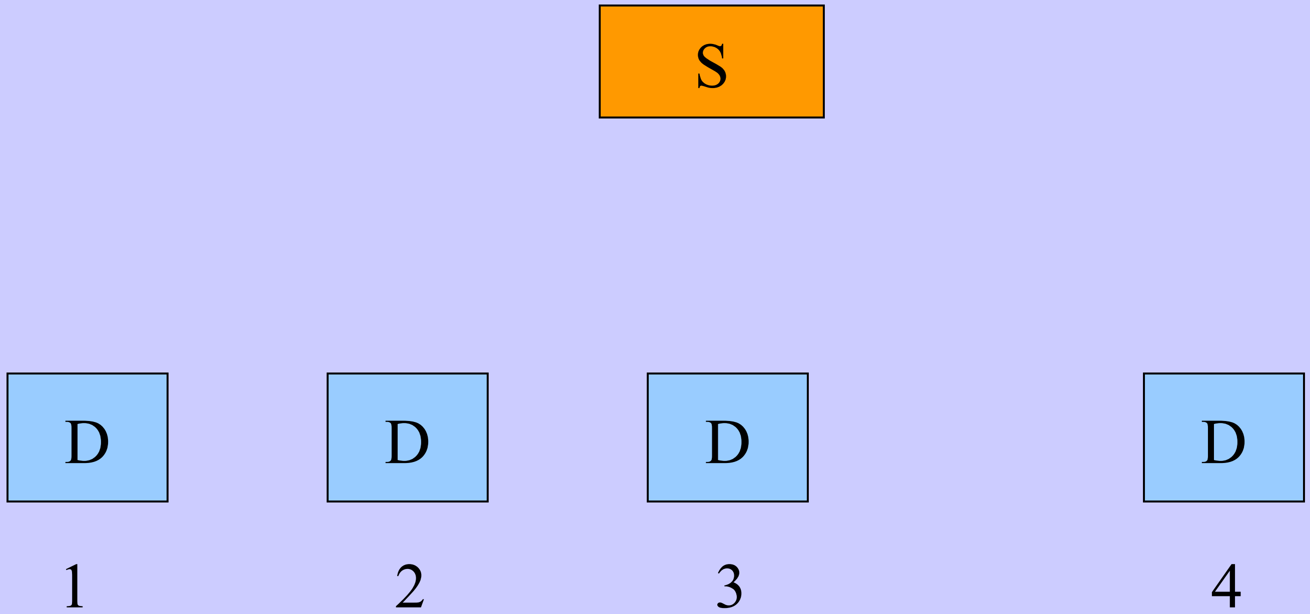

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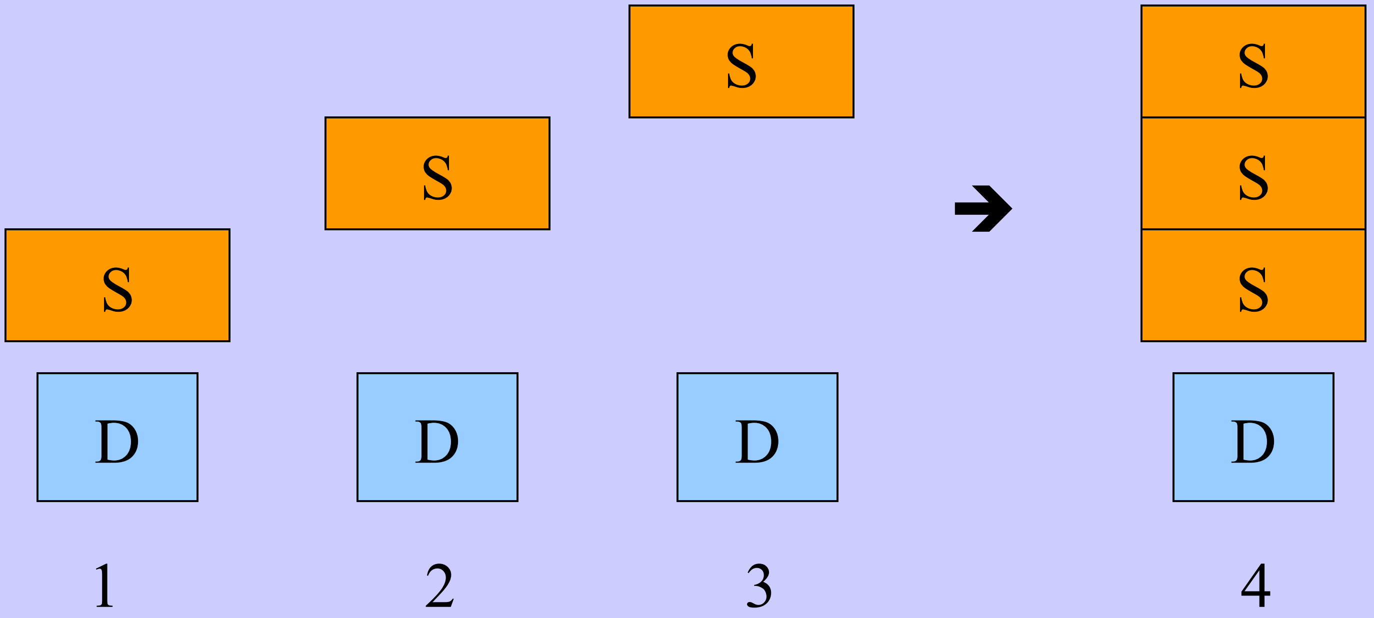


⇒Correct the effects of self-attenuation: $\varepsilon_i(E; 0) = F_{ai}(E; 0; m)\, \varepsilon_i(E; m)$

⇒ Linear relations between the values of efficiency in geometry 4 and the efficiencies in geometries 1, 2 and 3 with reliable coefficients

$\varepsilon_V(E; 0) = [\varepsilon_1(E; 0)\, V_1 + \varepsilon_2(E; 0)\, V_2 + \varepsilon_3(E; 0)\, V_3]/V$, with $V=V_1+V_2+V_3$

$\varepsilon_V(E; m) = F_{aV}(E; m; 0)\, \varepsilon_V(E; 0)$

Sima and Dovlete, JRNCL 200 (1995) 191

2. Sample density and composition

-Linear attenuation coefficient depends on:

- -Sample composition
- -Sample density
- -Photon energy

Computation of interaction coefficients if the composition is known:

-Tabulated values

- -Usually mass attenuation coefficients μ_m
- -Linear attenuation coefficients: $\mu = \mu_m \rho$, ρ = density

Software and database with values of the photon interaction coefficients:

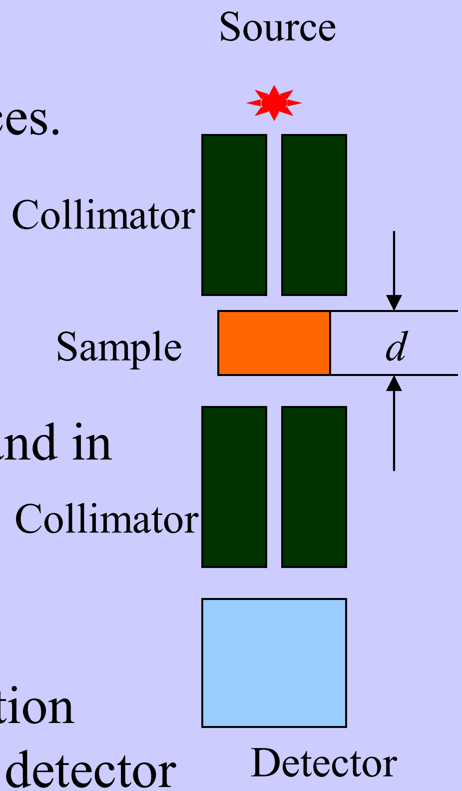
XCOM (M.J.Berger et al., NIST)

Software for visualization of the dependence of interaction coefficients on element and energy – EPICSHOW (NEA databank)

Experimental determination of linear attenuation coefficient for samples with unknown composition

1. Collimated beam transmission experiments

μ: transmission measurements with collimated point sources.



$R(d) = R_0 \exp(-\mu d) \Rightarrow \mu = \ln(R_0 / R) / d$

d = the length of photon path through the material

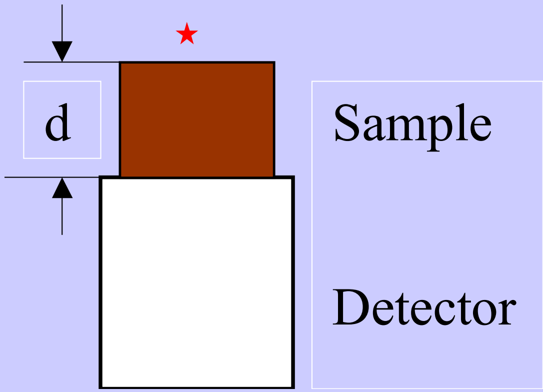
R and R_0 = the count rate in the peak, in the presence (R) and in the absence (R_0) of the material

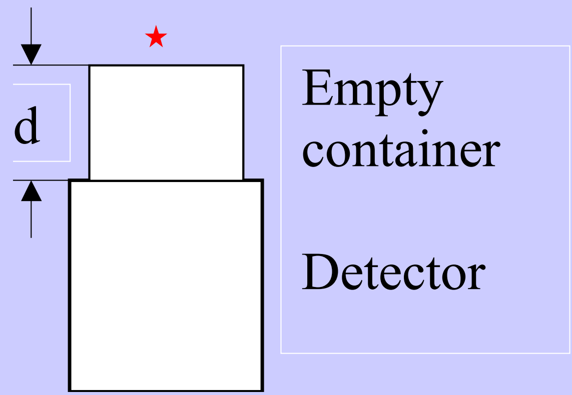
Problems:

- High intensity sources required
- At low energy – small angle Compton scattering contribution
  => the collimated source and the sample far from detector
- If possible single line gamma emitters should be used
- Time consuming, problems with the storage of high activity sources

2. Uncollimated beam transmission experiments

- Point source placed directly above the sample
- Measure count rate with the sample and with an identical empty container





-Advantage: low activity sources can be used

-Disadvantages:

- -The path length through the sample are not constant
- -Each path has a different probability to contribute to peak count rate
- -Low angle Compton scattering

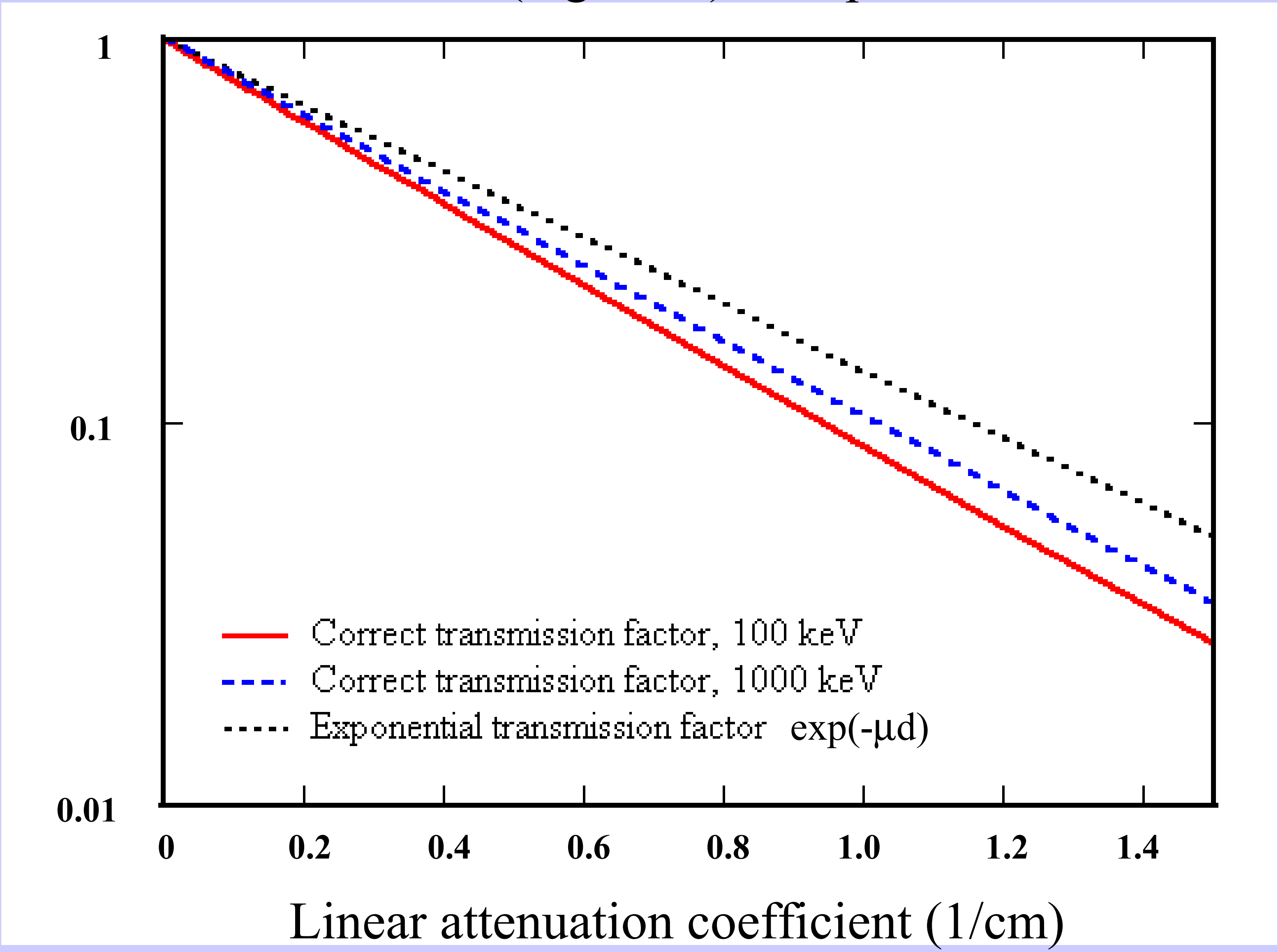
-Coincidence summing effects can seriously distort the results

- -Single gamma emitting nuclides should be used

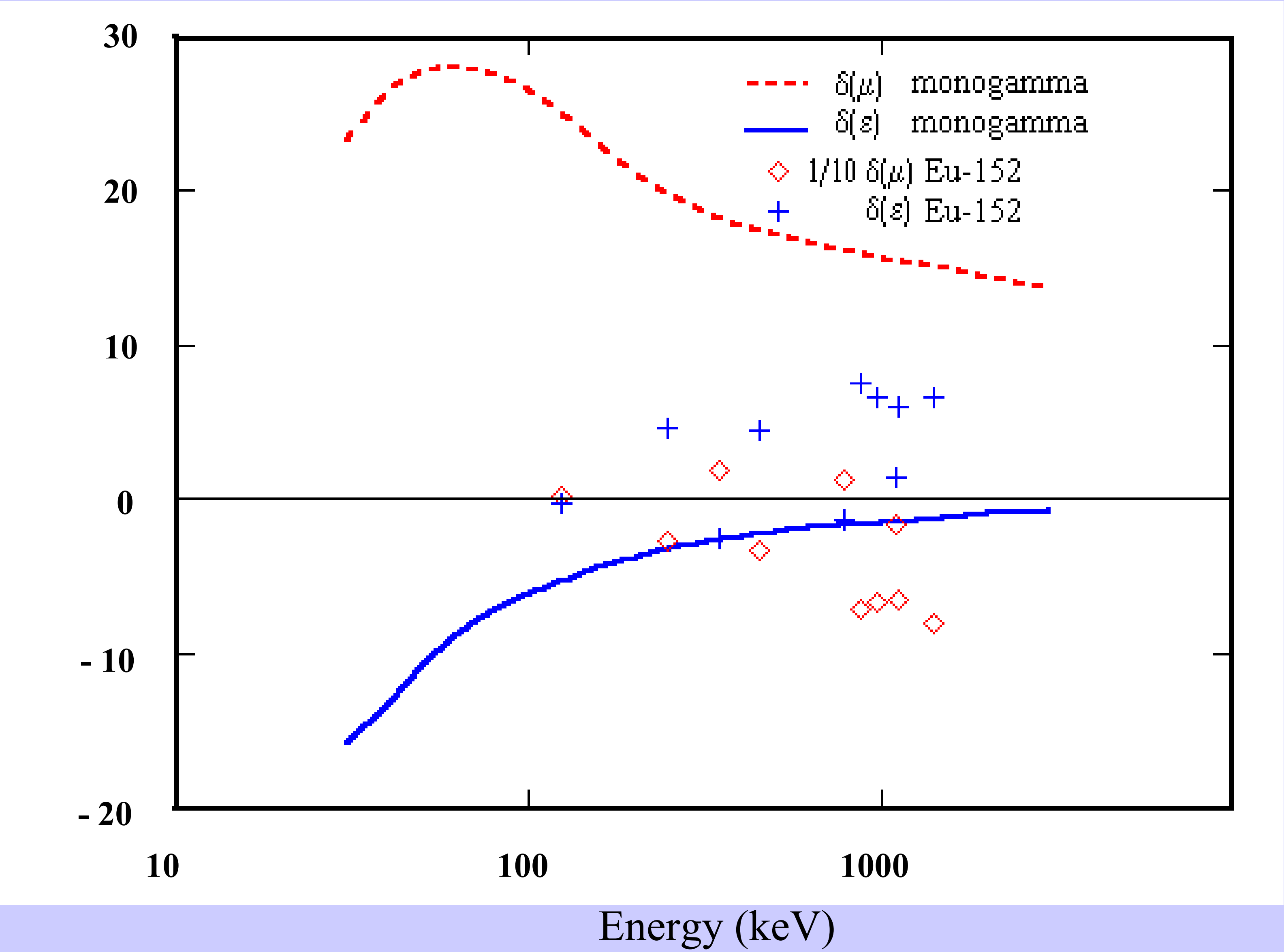
-Correct results: realistic simulation of the experiment

-Transmission factor computed by Monte Carlo [$\neq \exp(-\mu d)$]

Transmission factors (log scale). Sample: R=3.5, H=2 cm



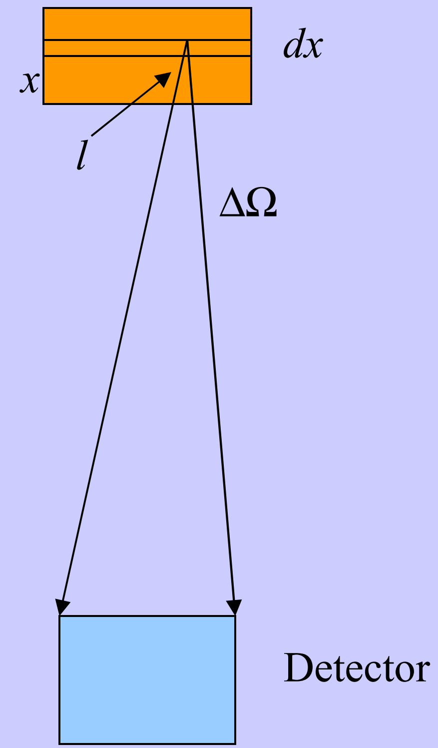
Relative error δ (%). Soil sample, R=3.5 cm, H=2 cm.



Sima and Arnold, 56 (2002) 71

Simplified analytical relations

Cylindrical sample



Cutshall et al. NIM 206 (1983) 309

The number of photons emitted in a given direction from a section of the sample that escape without interactions:

$$\frac{A}{V} P_\gamma \, dS \cdot dx \cdot \frac{d\Omega}{4\pi} \cdot \exp[-\mu \cdot l(\Omega)]$$

A, V = activity and volume of the source

P_γ = photon emission probability

$dS\, dx$ = emission volume element

$d\Omega$ = elementary solid angle within the solid angle $\Delta\Omega$ of the detector

Approximations:

-small solid angle, $l(\Omega)=x$, independent of the direction of the photon and of the position of the emission point within dx;

-$\Delta\Omega$ independent on the emission point.

Integration over dS and $d\Omega$ => replacement of dS with ΔS (surface of the sample) and of $d\Omega$ with ΔΩ

Integration over x from 0 to d (thickness of the sample) gives the number of photons emitted from the sample that hit the detector without any interaction in the sample:

$$\frac{A}{V} P_{\gamma} \Delta S \cdot \frac{\Delta \Omega}{4\pi} \cdot \frac{[1 - \exp(-\mu \cdot d)]}{\mu}$$

The same quantity in the case when attenuation in the sample is negligible is:

$$\frac{A}{V} P_{\gamma} \Delta S \cdot \frac{\Delta \Omega}{4\pi} \cdot d$$

Assuming that the probability of a count in the peak for each photon that hits the detector without having any interaction in the sample is the same, the ratio of the count rate R in the peak for the sample of interest to the count rate R_0 for the sample with negligible attenuation is:

$$\frac{R}{R_0} = \frac{1 - \exp(-\mu \cdot d)}{\mu \cdot d} = \frac{\varepsilon}{\varepsilon_0}$$

ε the efficiency in the presence of attenuation

ε_0 the efficiency in the absence of attenuation

Evaluation of the linear attenuation coefficient: transmission measurements

$$\frac{I}{I_0} = \exp(-\mu \cdot d), \quad \mu \cdot d = \ln\left(\frac{I_0}{I}\right)$$

$$=> \qquad \frac{\varepsilon}{\varepsilon_0} = \frac{1 - \dfrac{I}{I_0}}{\ln\left(\dfrac{I_0}{I}\right)}$$

Approximations:

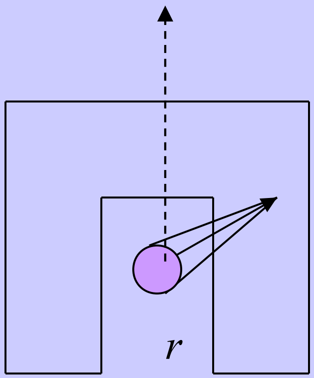
-each photon trajectory is perpendicular on detector surface

-all the photons incident on the detector have equal probability to be recorded in the peak

Miller, NIMA 258 (1987) 281 and Galloway, NIMA 300 (1991) 367

Marinelli beaker, small uniformly efficient detector

Sima, Health Phys. 62 (1992) 445



The number of photons emitted from a volume element dV located at (r, θ, ϕ) that hit the detector:

$$\frac{A}{V} \cdot P_\gamma \cdot dV \cdot \frac{S}{4\pi \cdot r^2} \cdot \exp\{-\mu \cdot [r - r_m(\theta)]\}$$

A, V = activity and volume of the source

P_γ = photon emission probability

S = detector surface

r_{m} and r_{M} = distances from the center of the detector to the entrance and exit from beaker.

Using $dV = r^2 \cdot d\Omega$ the radial integral can be evaluated

$$\frac{A}{V} \cdot P_\gamma \cdot \frac{S}{4\pi\mu} \cdot \{1 - \exp[-\mu \cdot l(\theta,\phi)]\} \cdot d\Omega \quad \text{with} \quad l(\theta,\phi) = r_M - r_m$$

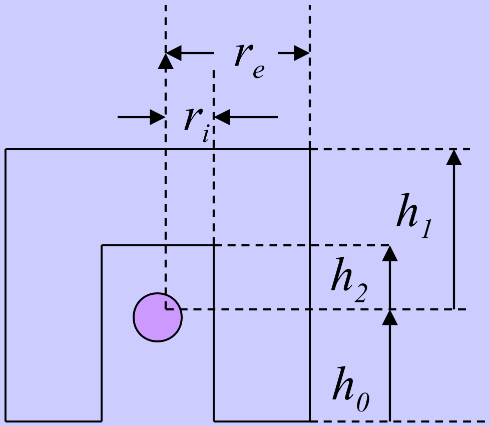
Then the angle integral can be evaluated as

$$\frac{A}{V} \cdot P_\gamma \cdot \frac{S}{4\pi\mu} \cdot \Delta\Omega \cdot \{1 - \exp[-\mu \cdot t]\} \quad \text{with} \quad t = \frac{1}{\Delta\Omega} \int_{\Delta\Omega} l(\theta,\phi)\, d\Omega$$

For a sample with negligible attenuation

$$\frac{A}{V} \cdot P_\gamma \cdot \frac{S}{4\pi} \cdot \Delta\Omega \cdot t$$

$$=> \quad \frac{\varepsilon}{\varepsilon_0} = \frac{1 - \exp(-\mu \cdot t)}{\mu \cdot t}$$



t can be evaluated analytically:

$$t = \frac{2\pi}{\Delta\Omega}\left[F(r_e, h_1) + F(r_e, h_0) - F(r_i, h_2) - F(r_i, h_0)\right]$$

$$F(r,h) = r \cdot arctg(h/r) + \frac{h}{2} \cdot \ln[(r/h)^2 + 1]$$

$$\Delta\Omega = 2\pi\left(1 + \frac{h_0}{\sqrt{h_0^2 + r_i^2}}\right)$$

Dryak et al., 135 (1989) 281; Sima, Health Phys. 62 (1992) 445

4. General formalism

Analytical expressions: approximate, based on simplifying assumptions

- -Cutshall: parallel trajectories, same probability to be recorded in the peak;
- -Sima: small uniformly efficient detector, narrow distribution of $l(\theta, \phi)$

Exact expression for the absolute value of the self-attenuation factor (homogeneous source):

$$F_a(E;m,0) = \frac{\int_V dV \int_\Omega \exp[-\mu_m(E) \cdot l_m(\vec{r},\vec{n})] \cdot T(E;\vec{r},\vec{n}) \cdot p_i(E;\vec{r},\vec{n}) \cdot d\Omega}{\int_V dV \int_\Omega T(E;\vec{r},\vec{n}) \cdot p_i(E;\vec{r},\vec{n}) \cdot d\Omega}$$

$\vec{r},\vec{n}$ coordinates and direction of the emission in the sample of volume V

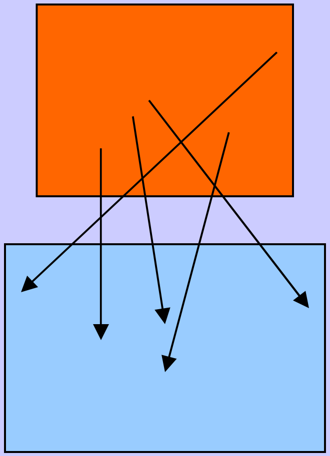
μ_m, l_m linear attenuation coefficient and length of trajectory through the sample

$T(E;\vec{r},\vec{n})$ transmission factor through the walls of the container, end cap etc.

$p_i(E;\vec{r},\vec{n})$ probability to record in the peak the photon that enters in the detector

=> the effects of sample matrix, of the geometry and of the intrinsic efficiency can not be described independently of each other (Moens et al., NIM 187 (1981) 451)

***F_a* depends mainly on linear attenuation coefficient μ and on the geometry of the sample**



Is it a property of the sample and of the matrix?
-Slight dependence on the detector:
    -Dimensions

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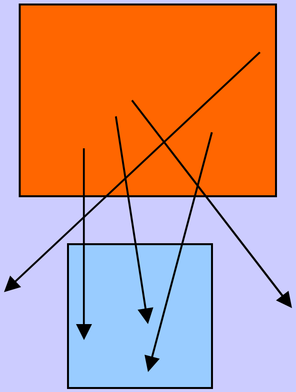

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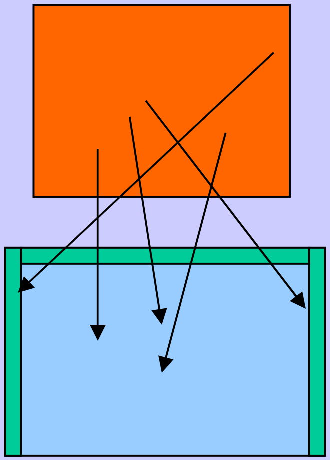
Is it a property of the sample and of the matrix?
-Slight dependence on the detector:
- -Dimensions
- -Type (p, n)

p-type



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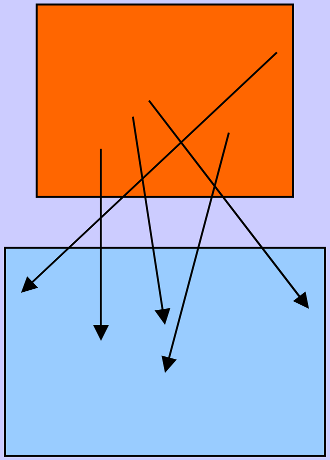

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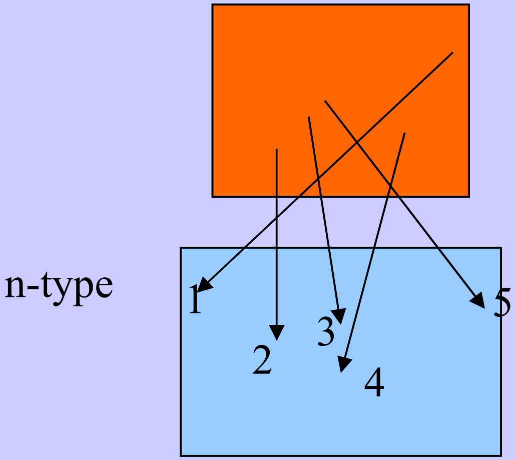

If the value of μ is fixed, does F_a depend on E ?

- n-type detector –low energy trajectories 1 and 5 contribute, at high energy not

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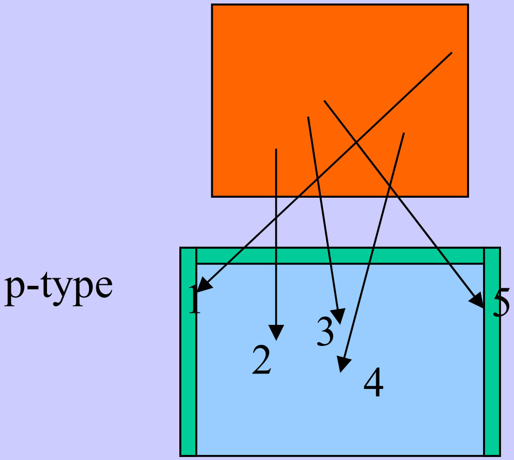

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- p-type detector – more complex, higher probability of complete absorption in the peak versus higher absorption in the dead layer at low photon energies

Sima, Progr. Nucl. Energy 24 (1990) 327

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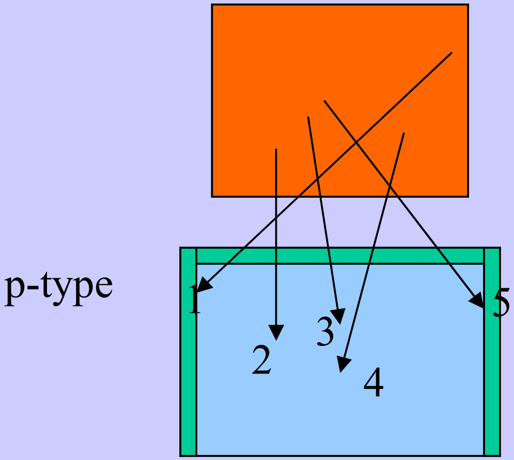

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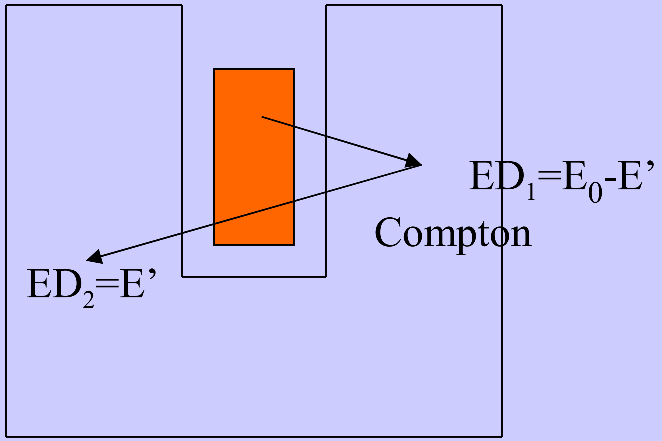
Sima, Progr. Nucl. Energy 24 (1990) 327

For the same sample and detector F_a depends on the distance between the sample and the detector

If two matrices have $\mu_1(E_0) = \mu_2(E_0) = \mu_0$ and are in identical containers, measured with the same detector in the same configuration, is $F_{a1}(E_0; m_1, 0)$ equal to $F_{a2}(E_0; m_2, 0)$?

- closed end coaxial detectors: yes
- well-type detectors: not



If $ED_1+ED_2 = E_0$ => signal in the peak of energy E_0

- the probability of traversing the sample at energy E' depends on μ(E')

=> Rigorously in the case of well-type detectors F_a depends on the complete curve μ(E) for $E<E_0$ and not only on the value μ_0 of μ(E) for $E= E_0$

=> In current conditions self-attenuation effects are small in the case of well type detector – the dependence of F_a on the complete curve μ(E) is very weak

Sima and Arnold, ARI 47 (1996) 889

Observation:

-In case of high attenuation only a thin layer of the sample located close to the detector is important; e.g. for μ=10 cm^{-1} only a layer of a few mm is important

⇒If that layer is not representative for the complete sample (non-homogeneity of matrix or of the radionuclide distribution) then wrong values are computed for the efficiency of the sample on the basis of the measured efficiency for the standard and of the computed values of F_a.

-In case of grains, at very high attenuation the distribution of activity inside the grains is very important

Example: Forster and Umbarger, NIM 117 (1974) 597 – metallic spheres containing Pu

5. Monte Carlo calculation

First realistic, direct, computations of F_a:

- Nakamura and Suzuki, NIM 205 (1983) 211

Realistic computations with a very fast algorithm:

- Sima, Prog. Nucl. Energy 24 (1990) 327; Sima and Dovlete ARI 48 (1997) 59

The absolute self-attenuation factor

$$F_a(E;m,0) = \frac{\int_V dV \int_\Omega \exp[-\mu_m(E)\cdot l_m(\vec{r},\vec{n})]\cdot T(E;\vec{r},\vec{n})\cdot p_i(E;\vec{r},\vec{n})\cdot d\Omega}{\int_V dV \int_\Omega T(E;\vec{r},\vec{n})\cdot p_i(E;\vec{r},\vec{n})\cdot d\Omega}$$

Usually computed by independent Monte Carlo runs for each case of interest (matrix effects interdependent with geometry and intrinsic efficiency effects)

However:

=> The dependence of F_a on sample matrix is only through the value of the linear attenuation coefficient $\mu=\mu_m(E)$ at the energy E of the peak (exception: well-type detectors)

=> Optimized procedure

⇒Power series expansion of the exponential:

$$\exp[-\mu \cdot l_m(\vec{r},\vec{n})] = 1 - \frac{1}{1!}\cdot[\mu \cdot l_m(\vec{r},\vec{n})] + \frac{1}{2!}\cdot[\mu \cdot l_m(\vec{r},\vec{n})]^2 - \frac{1}{3!}\cdot[\mu \cdot l_m(\vec{r},\vec{n})]^3 \ldots$$

=> Monte Carlo evaluation of the moments of the length of the trajectories:

$$a_k = (-1)^k \cdot \frac{1}{k!} \cdot \frac{\int_V dV \int_\Omega [l_m(\vec{r},\vec{n})]^k \cdot T(E;\vec{r},\vec{n}) \cdot p_i(E;\vec{r},\vec{n}) \cdot d\Omega}{\int_V dV \int_\Omega T(E;\vec{r},\vec{n}) \cdot p_i(E;\vec{r},\vec{n}) \cdot d\Omega}$$

a_k depend on sample geometry, detector dimensions, energy, but not on the matrix of the sample

-all a_k coefficients evaluated in a single Monte Carlo run and saved

-a single Monte Carlo simulation is required for any combination (geometry of the sample and detector)

F_a becomes a polynomial function of μ:

$$F_a(\mu) = \sum_k a_k \cdot \mu^k$$

⇒Very fast calculation of F_a for any linear attenuation coefficient if the coefficients a_k have been already evaluated

Procedure implemented in GESPECOR

6. GESPECOR

Realistic computation of the absolute and of the relative self-attenuation correction factors for cylindrical and Marinelli beaker samples with any matrix and density, for coaxial HPGe detectors and well-type detectors

Typical computations:

-Fast procedure based on Monte Carlo evaluation of the moments of the length of the photon trajectory through the sample

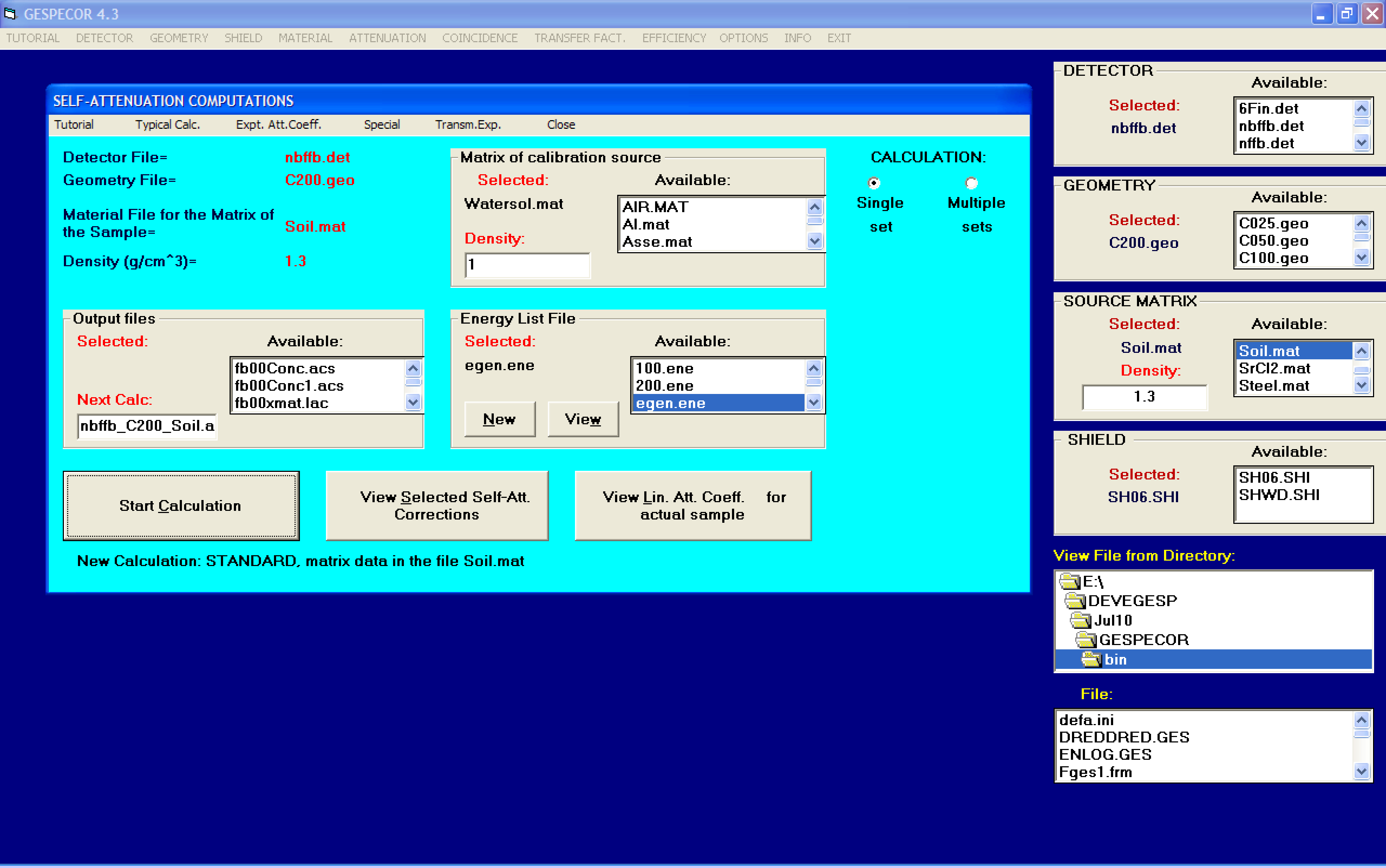
-Linear attenuation coefficient in the sample:

- -Based on sample composition and density using XCOM
- -Measured values of the linear attenuation coefficient
    - -Collimated beam transmission measurements
    - -Uncollimated point source transmission measurements, evaluated rigorously using Monte Carlo simulation

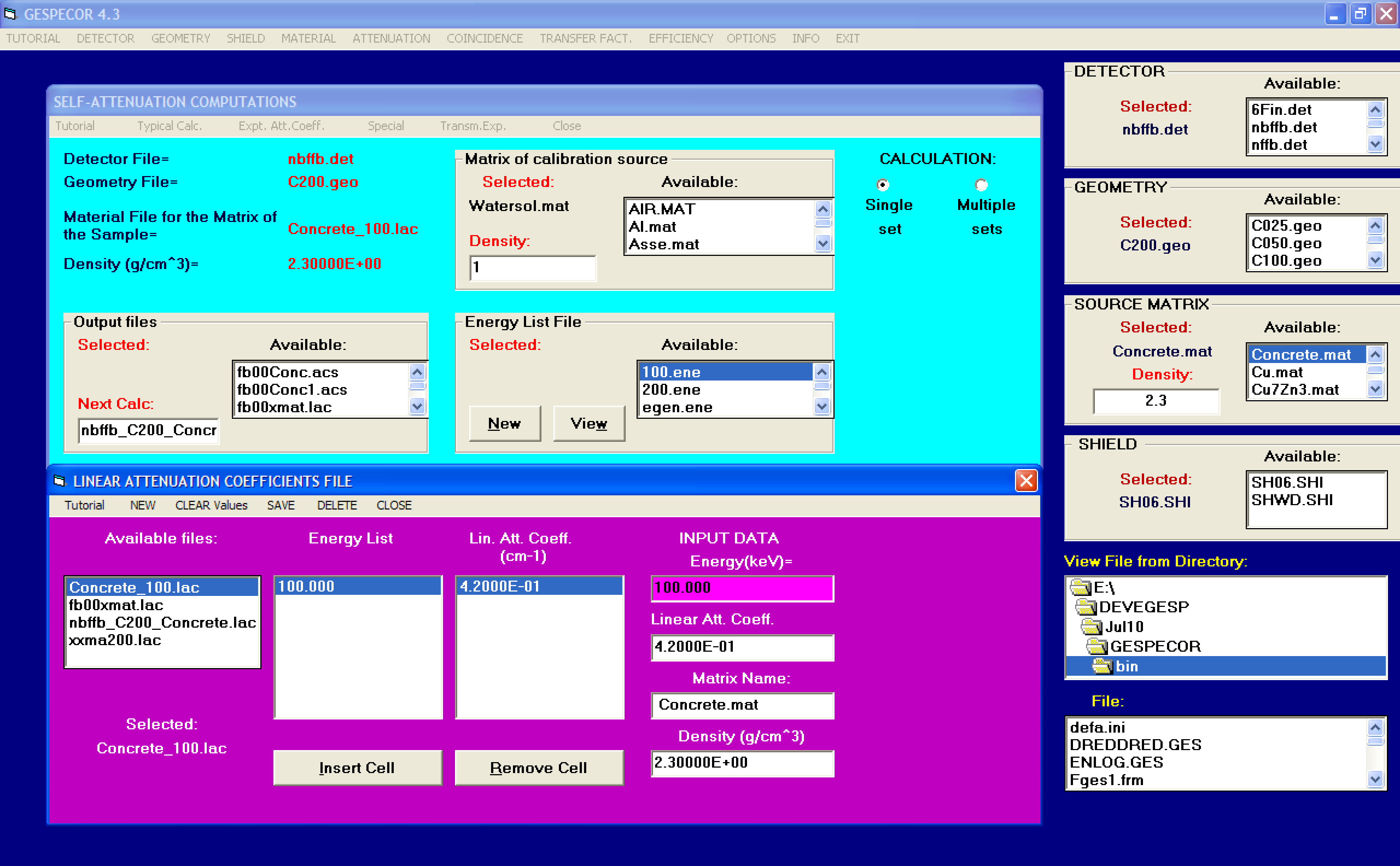
Special computations:

-Independent Monte Carlo computation for each case

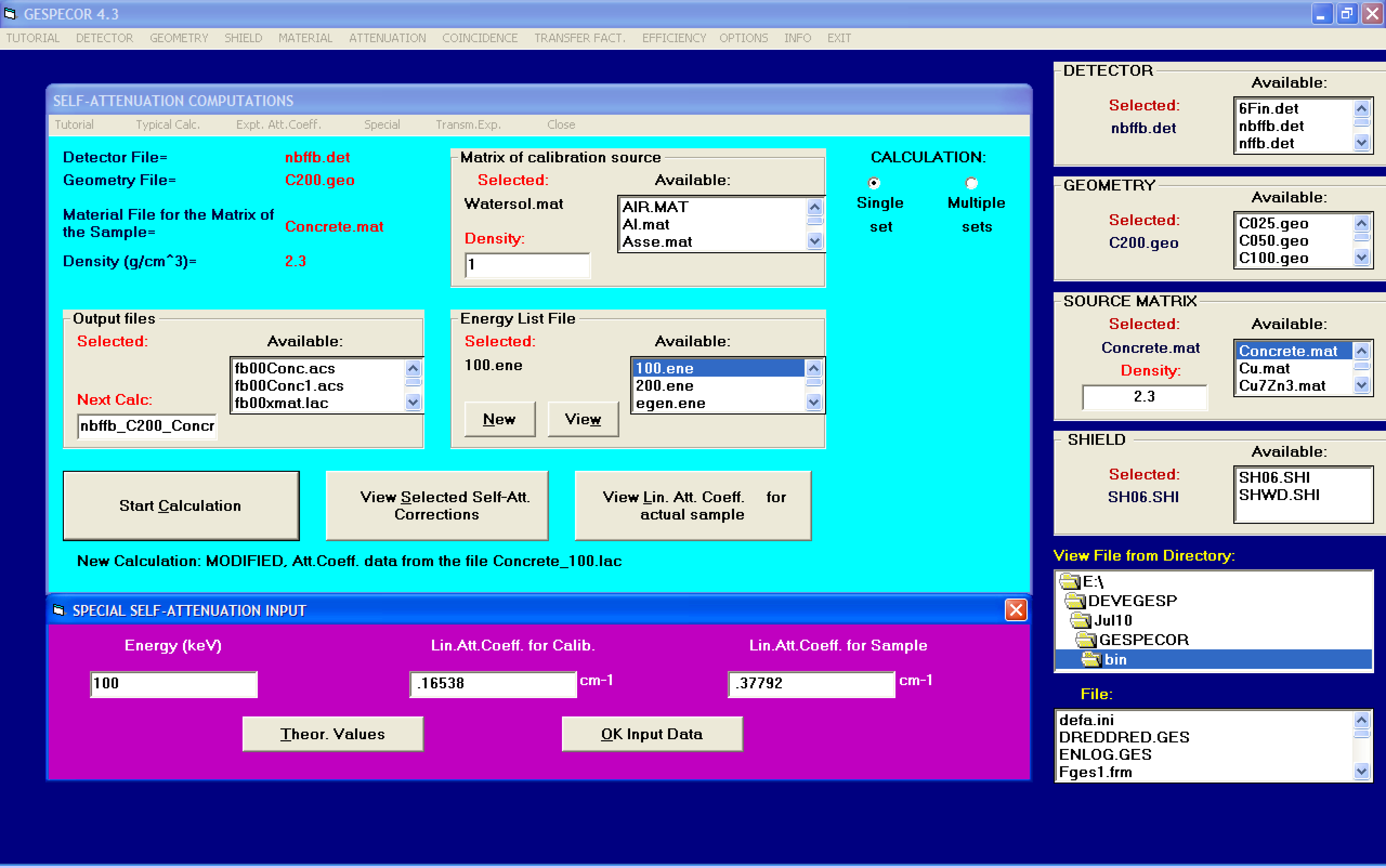
-Linear attenuation coefficient in the sample – as above



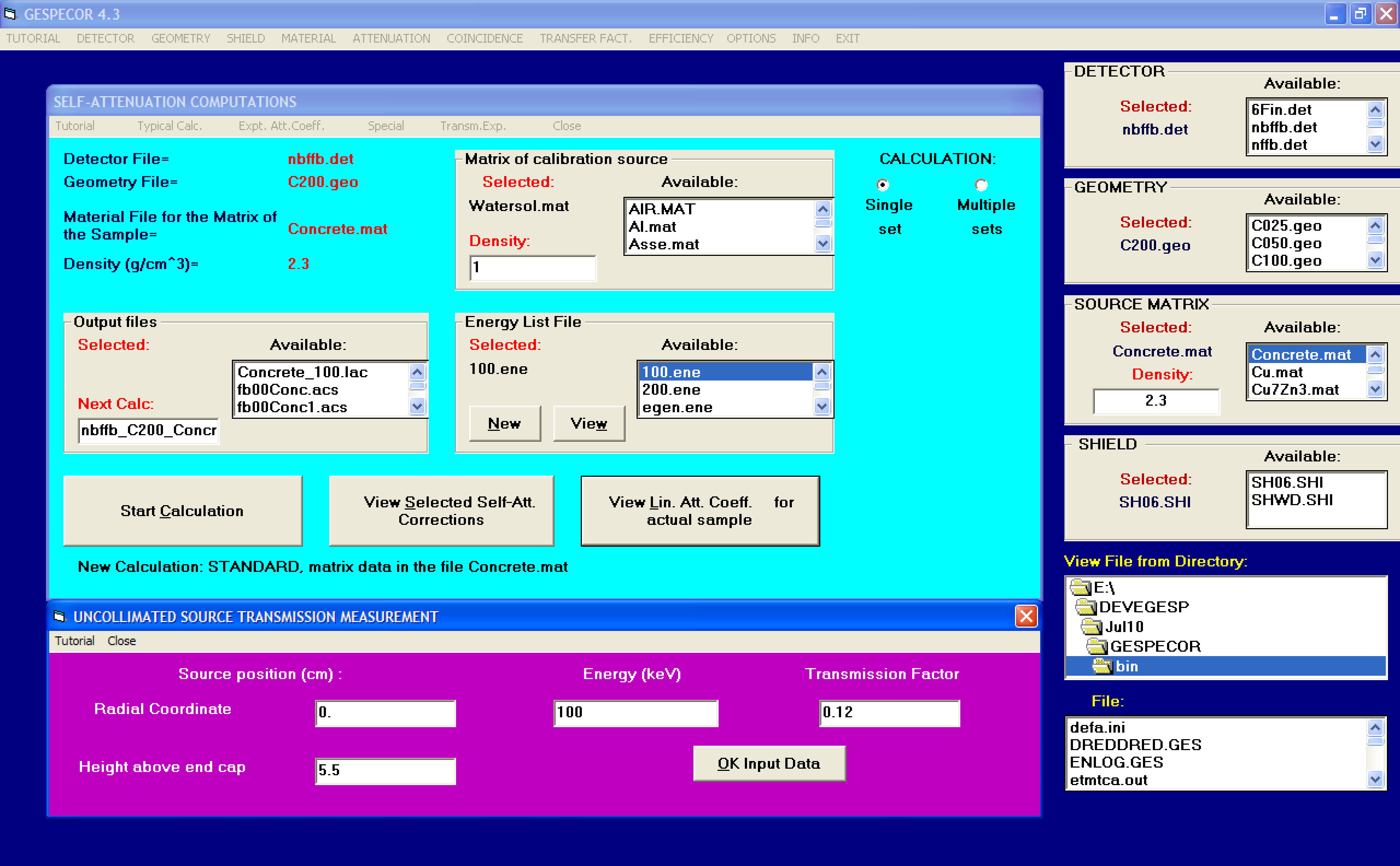
GESPECOR: Standard computation of self-attenuation corrections for a soil sample with respect to a water standard using the composition of the matrices



GESPECOR: Standard computation of self-attenuation corrections for a concrete sample with respect to a water standard using experimental values of μ for concrete



GESPECOR: Special computation of self-attenuation corrections for a concrete sample with respect to a water standard using values of μ given by the user



GESPECOR: Input data for the computation of the linear attenuation coefficient at 100 keV based on uncollimated point source transmission measurements

7. Summary

Realistic and fast procedures are available for accurate evaluation of matrix effects (self-attenuation corrections) in gamma-ray spectrometry of volume samples.

Linear attenuation coefficients can be computed on the basis of sample composition and density or using collimated or uncollimated transmission measurements

Sample homogeneity very important especially at high attenuation

Self-attenuation correction factors are most useful for computing the efficiency for a sample with a specific matrix on the basis of a standard with a different matrix.

Also useful for checking compatibility of standards with the same geometry but different matrices and for the evaluation of the efficiency for a sample of volume higher than the available certified reference material on the basis of measured efficiencies.