Special problems in self-attenuation

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Outline

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2. Sources with intrinsic inhomogeneity
3. Marinelli beaker samples
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1. Self-attenuation dependence on experimental conditions

Self-attenuation effects depend mainly on:

- linear attenuation coefficient μ
  - sample composition and density, photon energy E
  - evaluation: theoretical values (known composition) or experimental values (transmission experiments)
- sample geometry

Much weaker dependence of self-attenuation effects on:

- detector (dimensions, type)
- sample position
- photon energy (direct dependence, distinct from the implicit dependence through the value of μ(E))

Problems related to the contribution to the peak area of small angle Compton scattering

Absolute self-attenuation correction factor for a bulk sample with matrix m:

$$F_a(E;m)=\varepsilon(E;m)/\varepsilon_0(E)$$

$\varepsilon(E;m)$ = efficiency for the sample with matrix m

$\varepsilon_0(E)$ = efficiency for a sample with negligible self-attenuation

Both samples measured in the same configuration (identical sample geometry, same detector)

Relative self-attenuation correction factor for a sample with matrix m_1 with respect to a sample with matrix m_2:

$$F_a(E;m_1;m_2)=\varepsilon(E;m_1)/\varepsilon(E;m_2)=F_a(E;m_1)/F_a(E;m_2)$$

Note:

- F_a can be computed with a much lower uncertainty that ε

⇒ Best procedure to obtain $\varepsilon(E;m)$ for a matrix m in the absence of a standard with that matrix:

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

$\varepsilon(E,s)$ = experimental efficiency for a standard with matrix s

$F_a(E;m;s)$ = computed self-attenuation correction factor

Evaluation of the linear attenuation coefficient

If the composition is known:

-Tabulated values

Example of high attenuation cases:

- Efficiency reduction by 10 – 20 times due to self-attenuation

Experimental values: D. Arnold (PTB), E_γ = 46.54 keV (^{210}Pb)

- Lead with known activity of ^{210}Pb
- Theoretical values: GESPECOR

Matrix	Density (g cm^{-3})	Geometry	F_{ca} (th.)	F_{ca} (exp.)	Ratio
$Pb(NO_3)_2$	2.25	25 ml Cyl.	0.105	0.108	0.97
Zircon sand	3.01	1 l Marinelli	0.043	0.048	0.91
$Pb(NO_3)_2$	2.57	1.155 ml Cyl.	0.120	0.126	0.96
$PbSO_4$	2.79	1.155 ml Cyl.	0.101	0.101	1.00

Source: Sima and Arnold, ARI 47 (1996) 889; Sima, ARI 47 (1996) 919

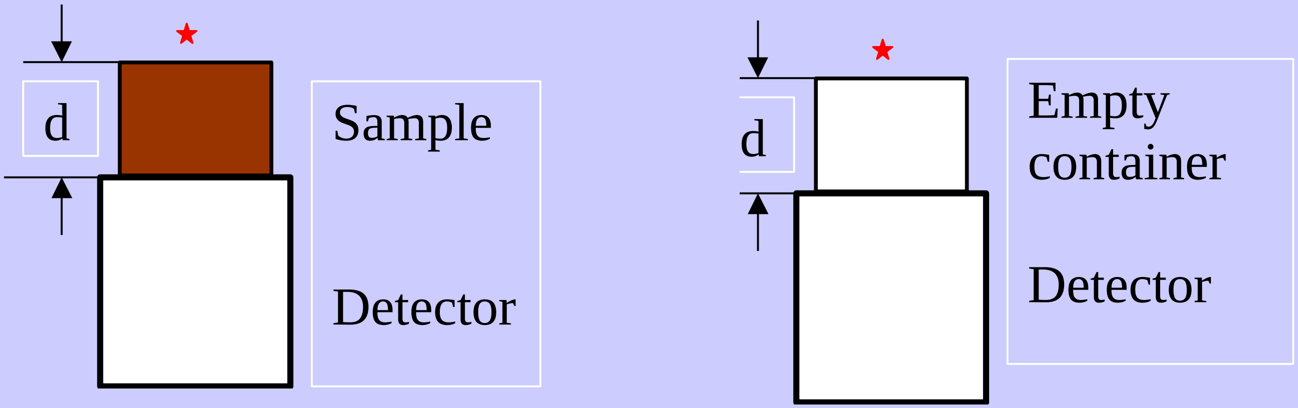
If the composition is not known:

-Transmission measurements with collimated sources: $T=R/R_0=\exp[-(\mu-\mu_0)\cdot d] \Rightarrow \mu$

-Uncollimated point source transmission measurements – Cutshall method

Uncollimated beam transmission experiments

- Point source placed directly above the sample
- Measure count rate with the sample R and with an identical empty container R_0

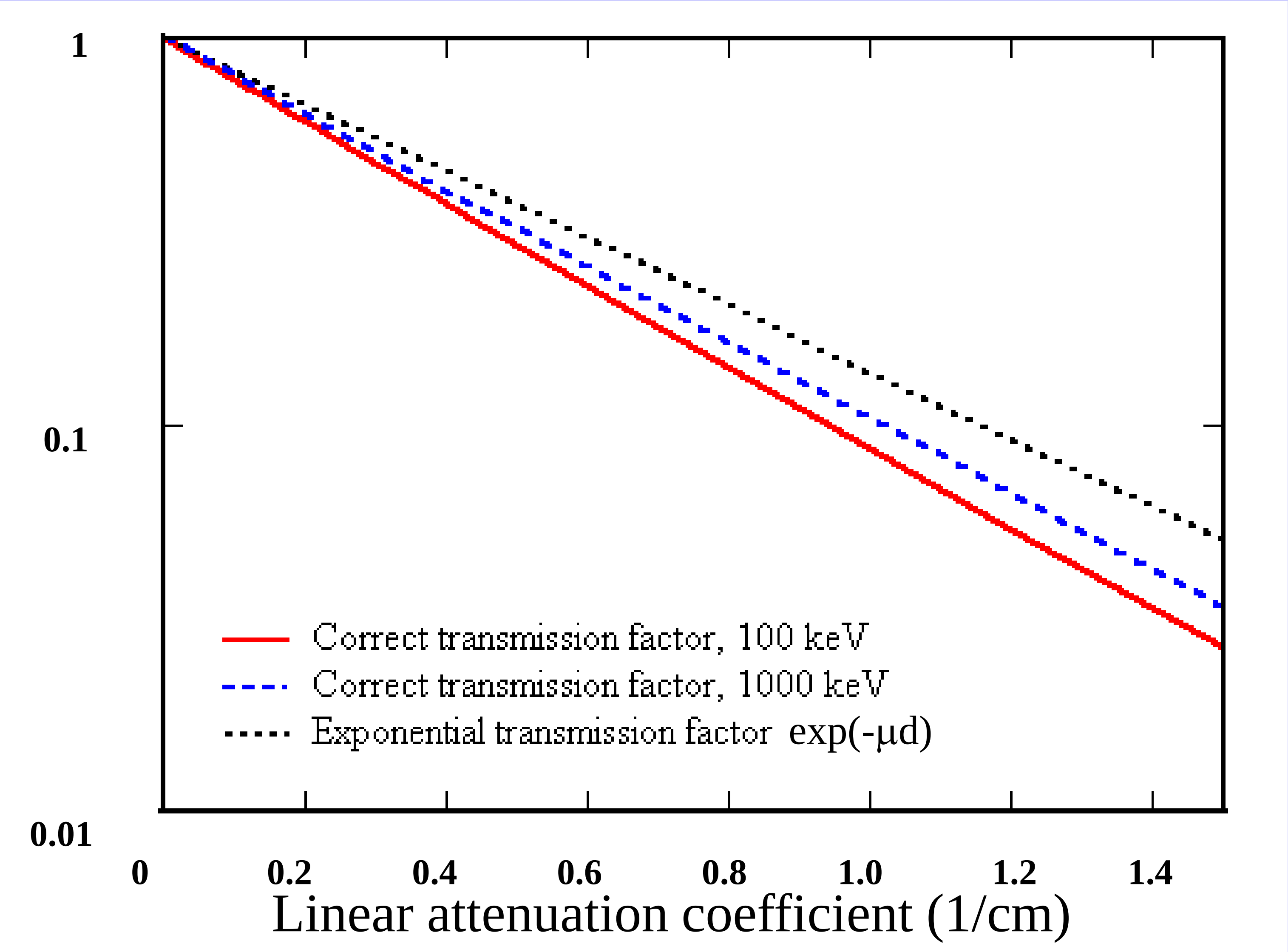


- -Advantage: low activity sources can be used
- -Disadvantages:
  - -The path lengths through the sample are not constant
  - -Each path has a different probability to contribute to peak count rate
  - -Low angle Compton scattering
- -Coincidence summing effects can seriously distort the results
  - -Single gamma emitting nuclides should be used

-Correct results: realistic simulation of the experiment is required

-Transmission factor computed by Monte Carlo [≠ Cutshall approximation = $\exp(-\mu d)$]

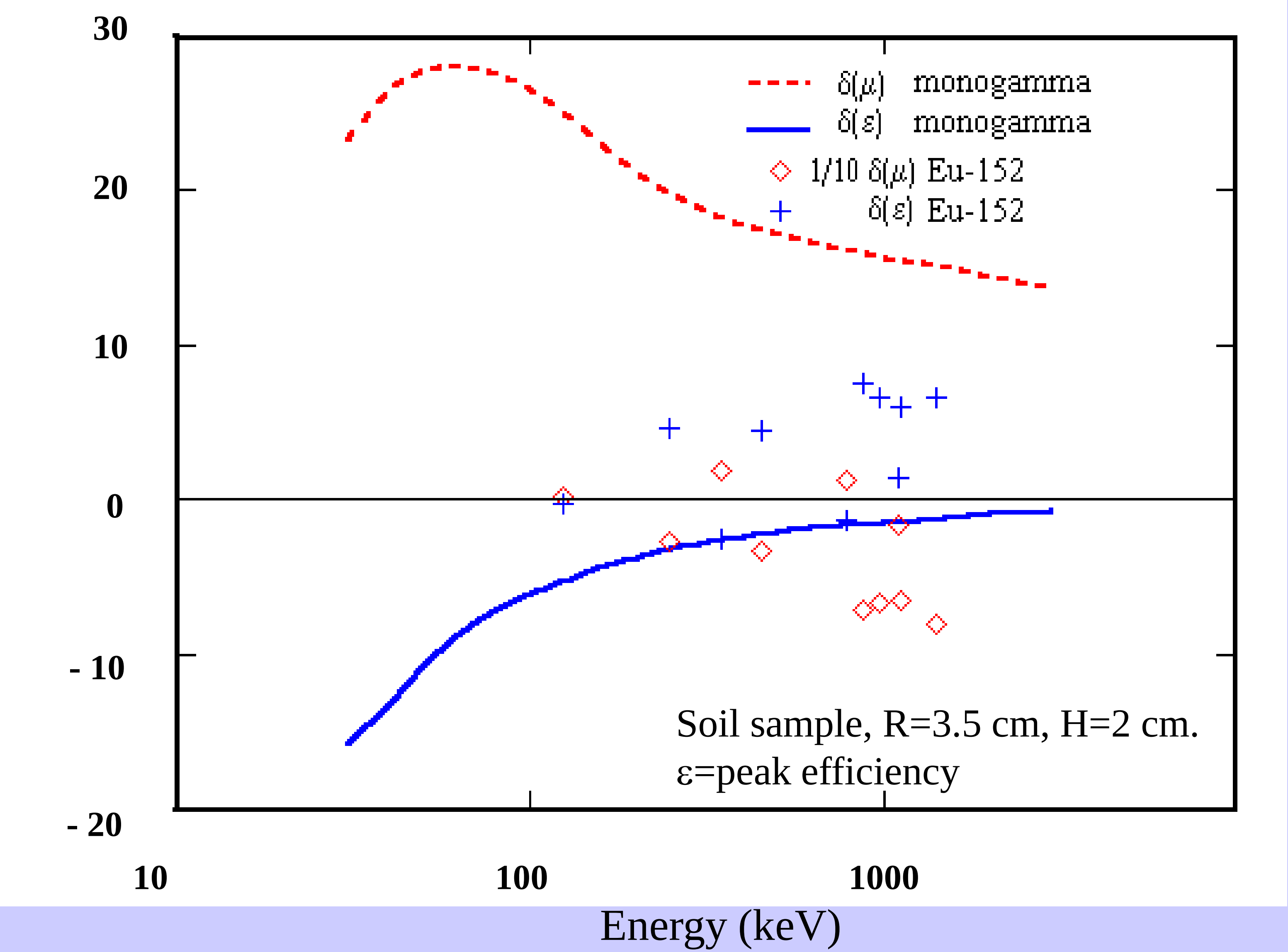
Transmission factors (logarithmic scale). Sample: R=3.5, H=2 cm



Correct value of T – smaller than exp(–μd) (inclined trajectories result in higher attenuation)

In the presence of coincidence summing effects, very difficult to get correct results

δ (%) = rel. difference between the correct value and the value based on T=exp(−μd)



Sima and Arnold, Appl. Radiat. Isot. 56 (2002) 71

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

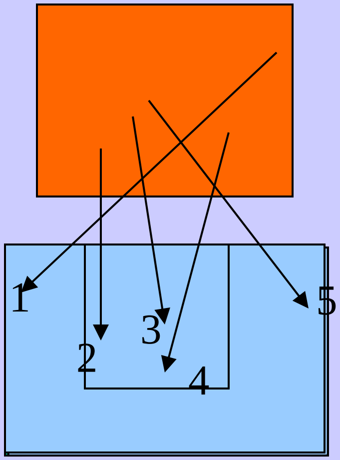
Is it a property of the sample and of the matrix?

-Slight dependence on the detector:

-Dimensions: paths 2,3,4 contribute both for a small and for a big detector; 1,5 only for big

-Type (p, n) – inclined trajectories – dead layer

p-type



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

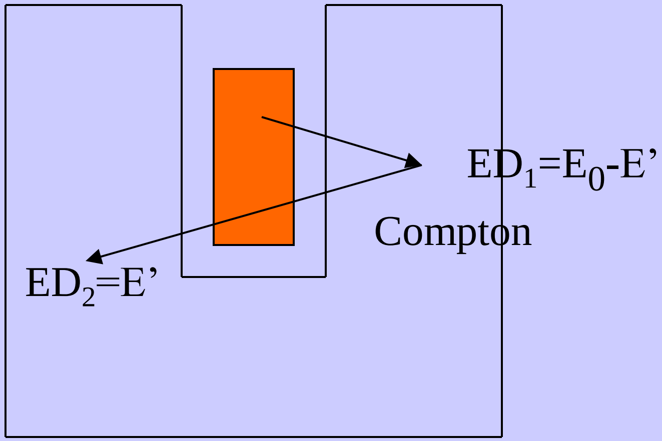
- p-type detector – more complex, higher probability of complete absorption in the peak versus higher absorption in the dead layer at low photon energies

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

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 $F_{a1}(E_0; m_1)$ equal to $F_{a2}(E_0; m_2)$?

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



If $ED_1+ED_2 = E_0$ => signal in the peak of energy E_0 (ED=energy deposited in detector)

- 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, Appl. Radiat. Isot. 47 (1996) 889

Observation:

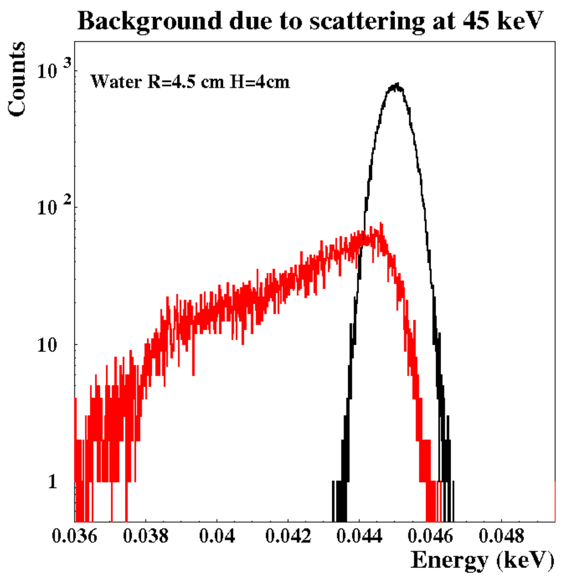
-In case of high attenuation only a thin layer of the sample located close to the detector is important; e.g. for μ=10 cm^{-1} only a layer of a few mm is important

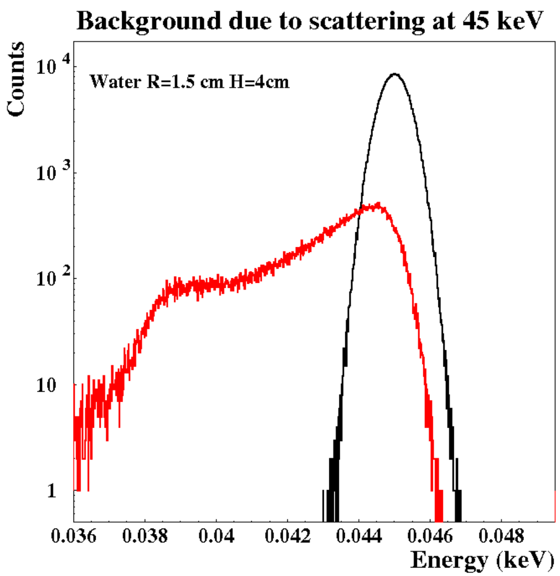
⇒If that layer is not representative for the complete sample (non-homogeneity of matrix or of the radionuclide distribution) then wrong values are computed for the efficiency of the sample on the basis of the measured efficiency for the standard and of the computed values of F_a .

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

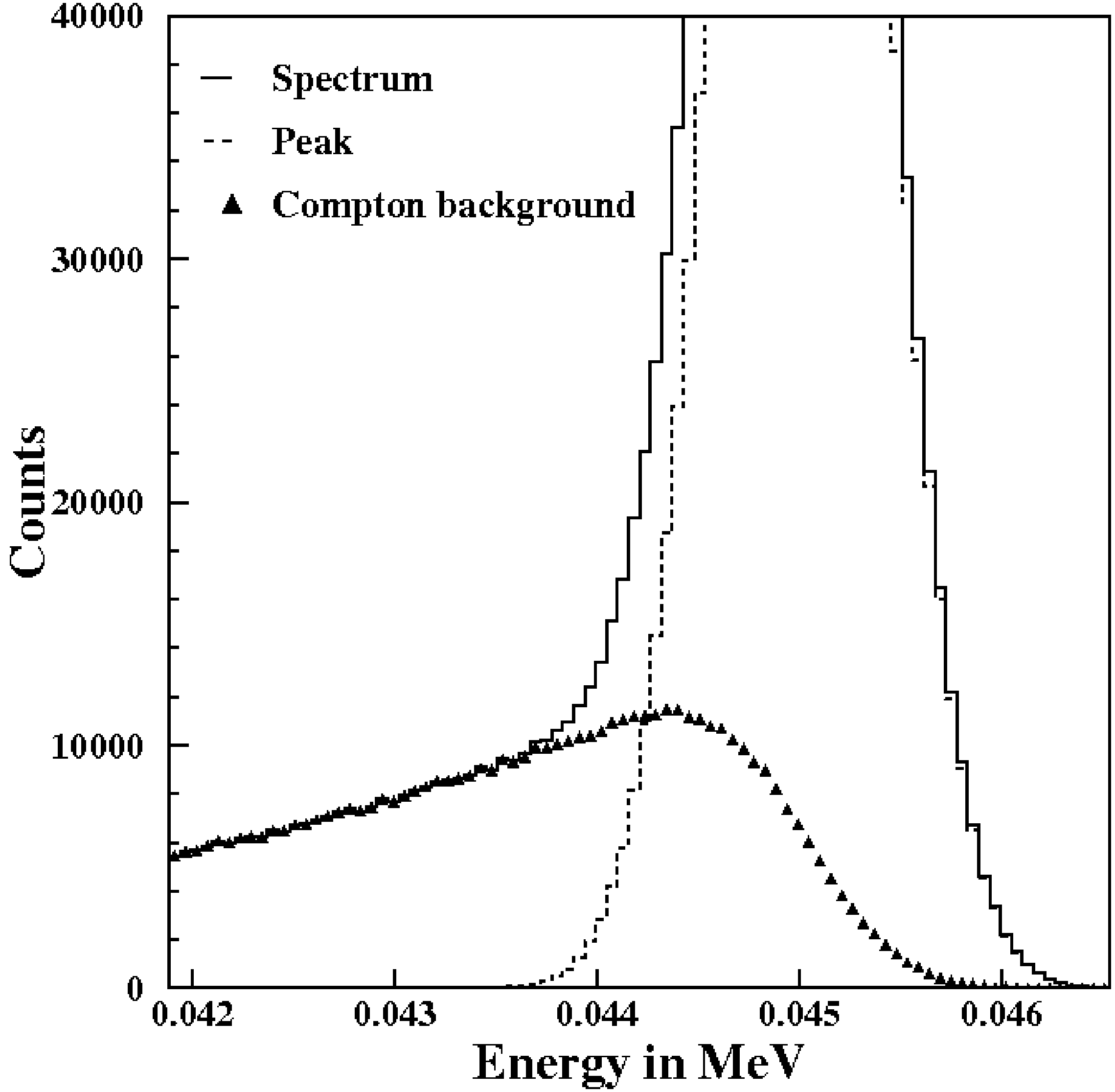
Example: Forster and Umbarger, NIM 117 (1974) 597 – metallic spheres containing Pu

Problems related to small angle Compton scattering





Contribution of Compton scattered photons to the peak area:
13% (sample with R=4.5 cm), 10% (sample with R=1.5 cm)
=> depends on sample and on detector resolution



Water sample
R=4.5 cm, H=4 cm

Compton contribution under the peak of 45 keV: 13%

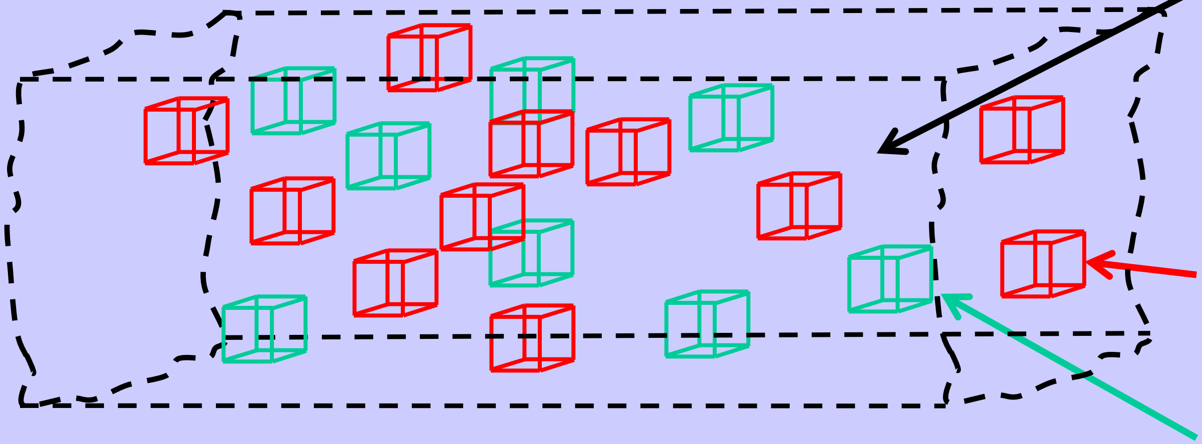
The linear or the step approximation for the background do not remove completely this contribution

Sima and Arnold, ARI 67 (2009) 701

2. Sources with intrinsic inhomogeneity

Sima, ARI 126 (2017) 146; Sima, ARI 134 (2018) 137

Environmental medium

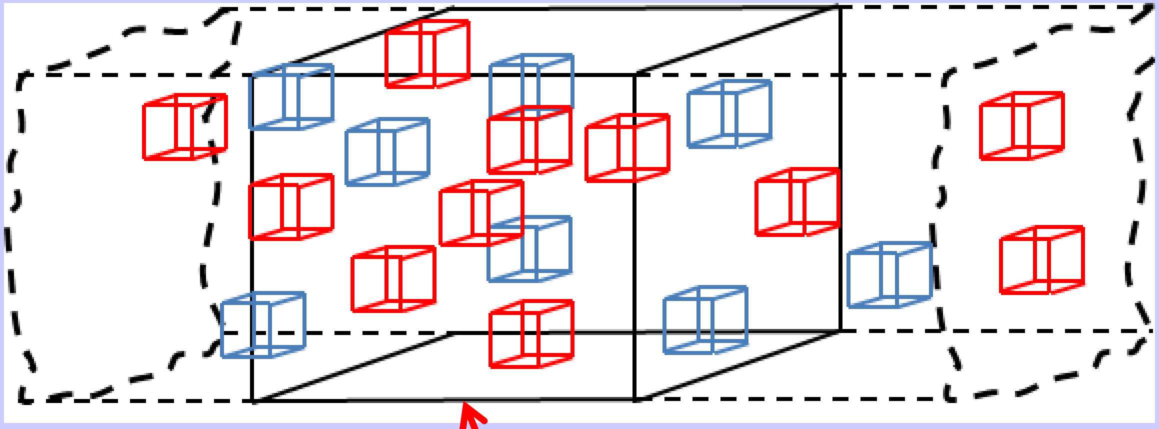


Class 1 Component (main matrix; blocks not shown)

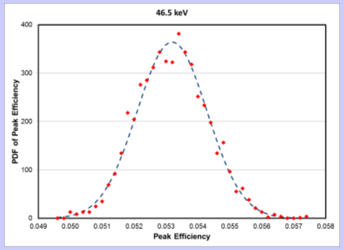
Embeded: Class 2 Component

Class 3 Component

Sample of environmental medium



Parallelepiped Sample



Random effects (number and position of blocks of each type) =>uncertainty

Systematic effects (activity segregation in different blocks) => distortion of efficiency

Monte Carlo simulation of the distribution of efficiency values

Step 1:

- Preparation of the input data characterizing a sample:
  - For blocks j=1 to N_t sample the class, according to the probabilities p_i
  - Save block class and position

Step 2:

- Simulation of the efficiency for the given sample
  - Randomly select the emission point, according to activity distribution
  - Transport the photon through the source taking into account the matrix distribution
  - Evaluate the efficiency using the procedures available in the standard version of GESPECOR (Sima, Arnold and Dovlete, JRNC 248 (2001) 359)

Repeat Step 1 and Step 2 many times

Step 3

- Construct and summarize the distribution of the values of the peak efficiency
  - Best estimate of the efficiency
  - Standard uncertainty

Implemented as an extension of GESPECOR

Examples

- Case 1: relatively weak inhomogeneity
- Soil ref.1 from Kaminsky et al. (ARI 94 (2014) 306)
  - classes: specific minerals, organic matter and air. Fraction by weight and density (g cm^{-3}) given below:

SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	MnO	P_2O_5	Org	
66.9	13.8	4.1	1.7	2.9	2.2	2.8	0.5	0.1	0.2	5.0	W
2.65	4.0	5.25	3.58	3.34	2.27	2.30	4.0	5.43	2.39	1.0	ρ

- One additional class: air, fraction by volume (NTP) 0.2

- Case 2: high inhomogeneity
- Soil including pitchblende grains

- In both cases, interest for nuclides from U-Ra decay chain: 46.5 keV (^{210}Pb), 92.4 keV (^{234}Th), 186.2keV (^{226}Ra), 1001.44 (^{234m}Pa)

- Sample: 5x5x2 cm, detector: 47% efficiency n-type HPGe

Case 1

- Construction of the equivalent homogeneous matrix, containing the same total mass of each component, uniformly distributed in the same total volume (index 0 for quantities evaluated for the homogeneous matrix)
- Monte Carlo calculation of the efficiency ε_0 for the homogeneous matrix
- Definition of the scale of the inhomogeneity (dimension of the blocks): two values, d=0.0167 cm and d=0.1 cm
- Activity distribution between classes:
  - Several scenarios, from extreme cases when the activity is completely imbedded in one component, to more homogeneous (same activity in each component)
  - Monte Carlo simulation of the efficiency ε_i for each scenario
- Inhomogeneity effects higher for the photons with E=46.5 keV

Activity of the elementary block											d=0.0167		d=0.1	
SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	MnO	P_2O_5	Org	$\varepsilon/\varepsilon_0$	R/R_0	$\varepsilon/\varepsilon_0$	R/R_0
1	1	1	1	1	1	1	1	1	1	1	1.01	1.01	1.04	1.03
2	1	1	1	1	1	1	1	1	1	1	1.01	1.01	1.04	1.03
10	1	1	1	1	1	1	1	1	1	1	1.01	1.01	1.04	1.04
50	1	1	1	1	1	1	1	1	1	1	1.01	1.01	1.04	1.04
1	0	0	0	0	0	0	0	0	0	0	1.01	1.01	1.04	1.04
1	1	1	1	1	1	1	1	1	1	2	1.01	1.01	1.04	1.04
1	1	1	1	1	1	1	1	1	1	10	1.01	1.01	1.05	1.04
1	1	1	1	1	1	1	1	1	1	50	1.01	1.01	1.07	1.05
0	0	0	0	0	0	0	0	0	0	1	1.01	1.01	1.07	1.05
0	0	1	0	0	0	0	0	0	0	0	0.95	0.96	0.76	0.80

$\varepsilon/\varepsilon_0$ for 46 keV

$R=\varepsilon(46)/\varepsilon(92)$

- Results: generally ε_i differs from ε_0 by about 1% in the case of d=0.0167 cm, and by <5% for d=0.1 cm; exceptions: 5% (d=0.0167 cm) and 25% (d=0.1 cm) in the case when the activity is completely imbedded in Fe_2O_3.

- Activity distributed on the surface of the blocks (except for air blocks): efficiency differs from the efficiency for the homogeneous matrix and uniform activity by less than 3% (scenario Fe_2O_3, d=0.1 cm)

Conclusion:
Generally the inhomogeneity effects are not important
For accurate evaluation of the efficiency, information on activity distribution among components is needed

In the absence of information on activity distribution among components, uncertainty evaluation should include a contribution resulting from the lack of specific knowledge on activity distribution, besides uncertainty resulting from the block distributions

Case 2 (high inhomogeneity)

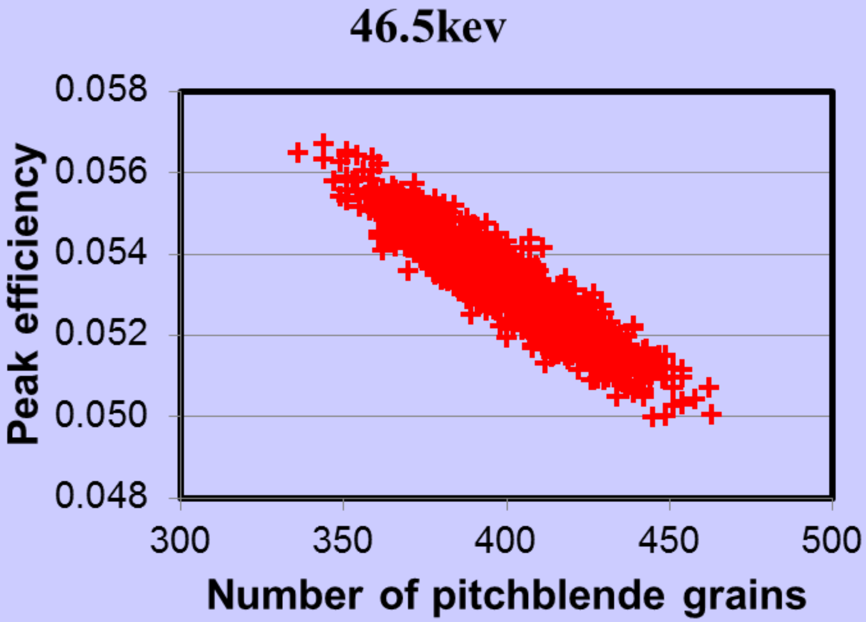
- Soil including pitchblende grains
  - Classes:
    - typical soil, density 1.2 g·cm^{-3}, probability of blocks p_1
    - Pitchblende (UO_2), density 10.8 g·cm^{-3}, probability p_2 (denoted by P)
    - Air, density 0.0012 g·cm^{-3}, probability p_3

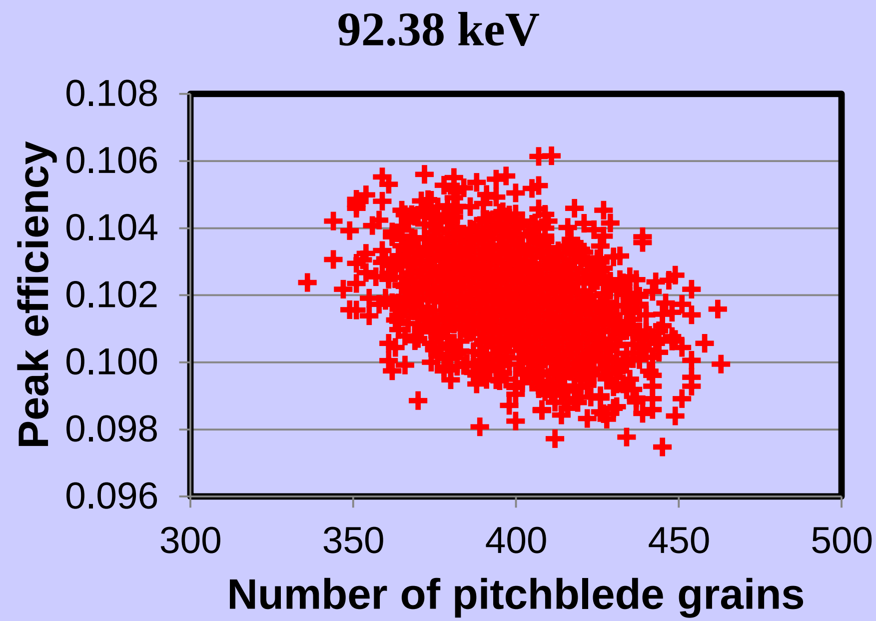
- Interest for nuclides from U-Ra decay chain: 46.5 keV (^{210}Pb), 92.4 keV (^{234}Th), 186.2 keV (^{226}Ra), 1001.44 (^{234m}Pa)
- Sample: 5x5x2 cm, detector: 47% efficiency n-type HPGe

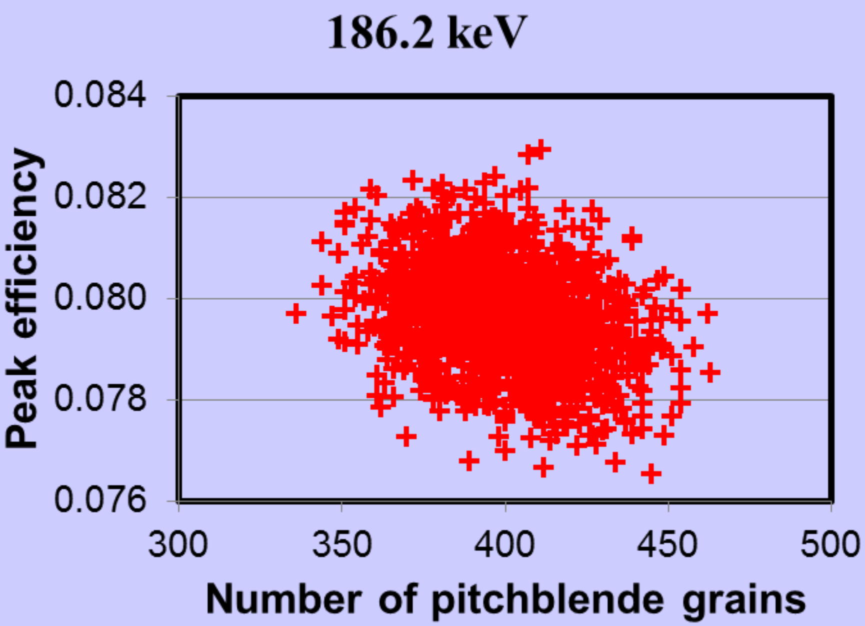
Simulations parameters:

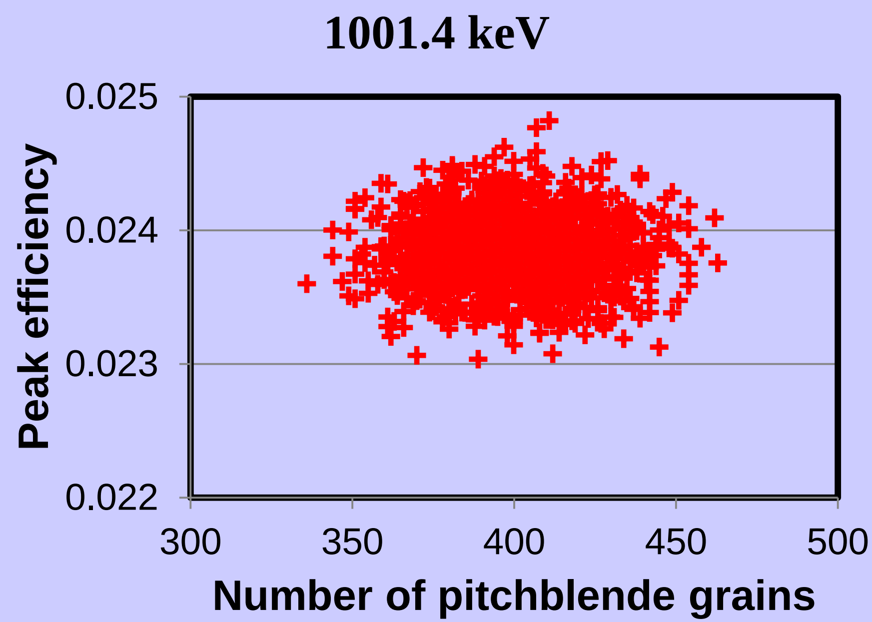
- d $\in$ (0.0167, 0.25) cm
- Air blocks probability = 0.2, Pitchblende blocks probability P $\in$ (0.00005, 0.01); specific simulations made also for P=0.025, 0.05, 0.1, 0.25, 0.50.
- Air blocks activity = 0; ratio between the activity of pitchblende component and the total activity $A_P/A_T \in (0, 1)$.

- For each P the equivalent homogeneous matrix was constructed, ε_0 was evaluated
- For each (d, P, A_P/A_T) the efficiency ε was evaluated





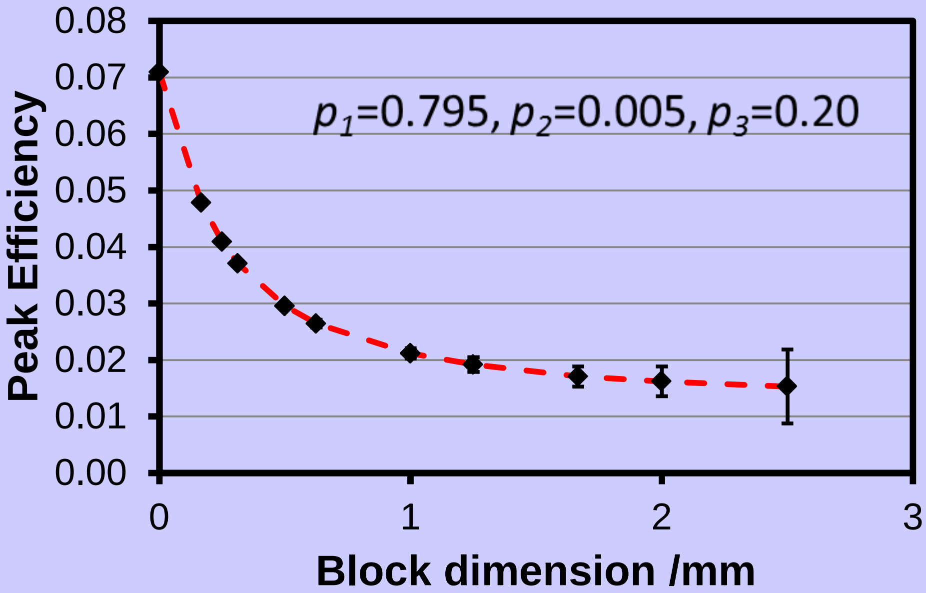




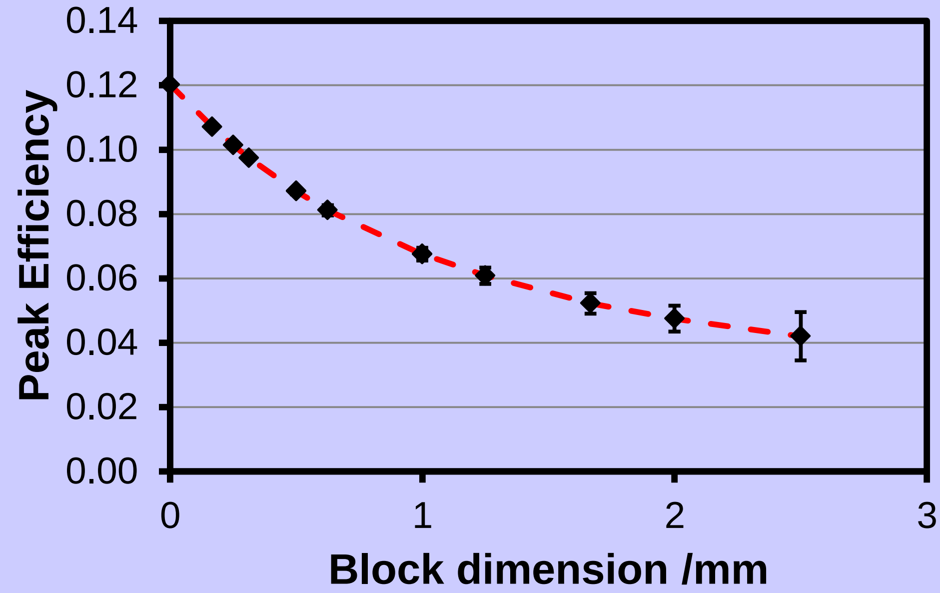
Size of the blocks: d=0.5 mm; N_t=400000; p_1=0.799, p_2=0.001, p_3=0.20

Efficiency decrease with N_2: higher attenuation if more photons emitted from pitchblende

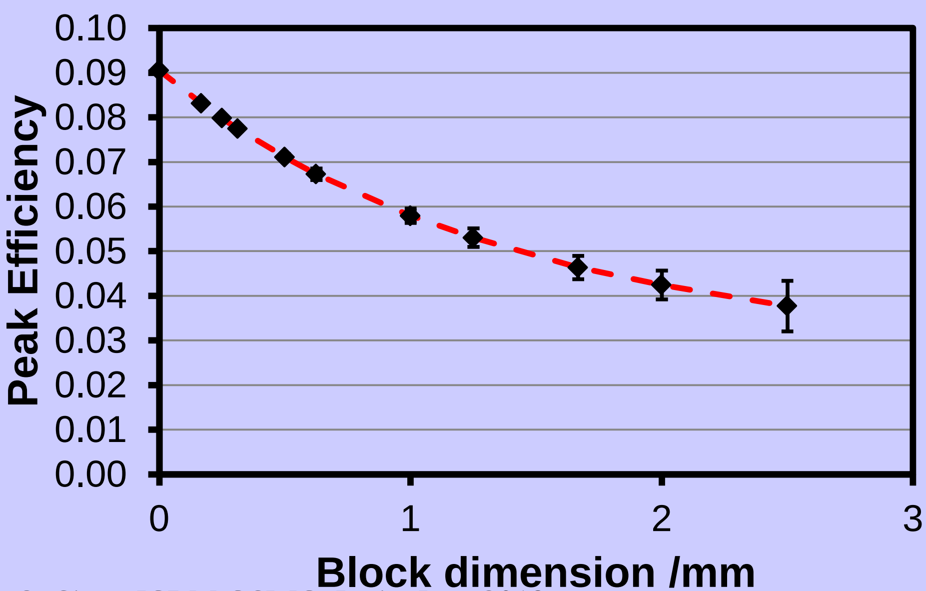
46.5 keV



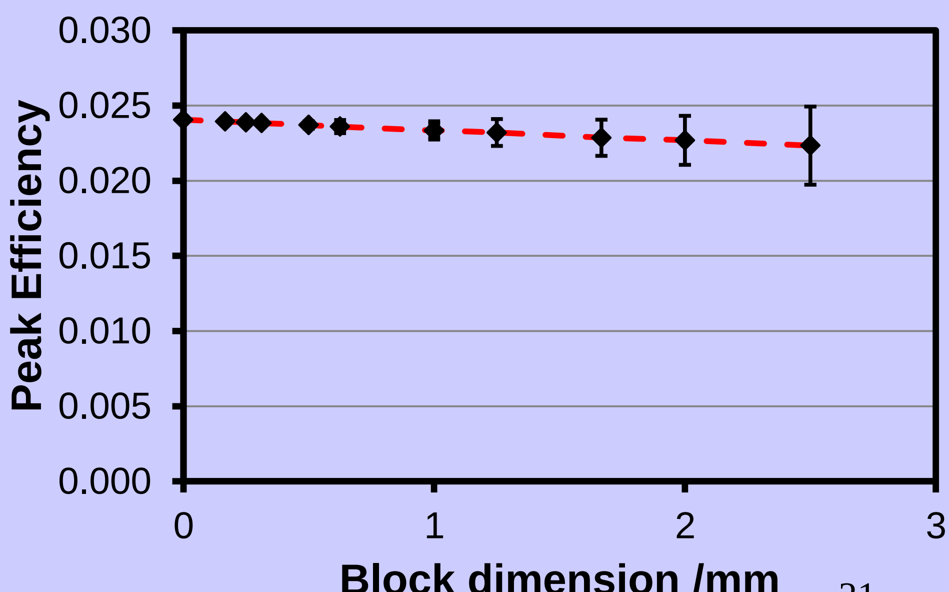
92 keV



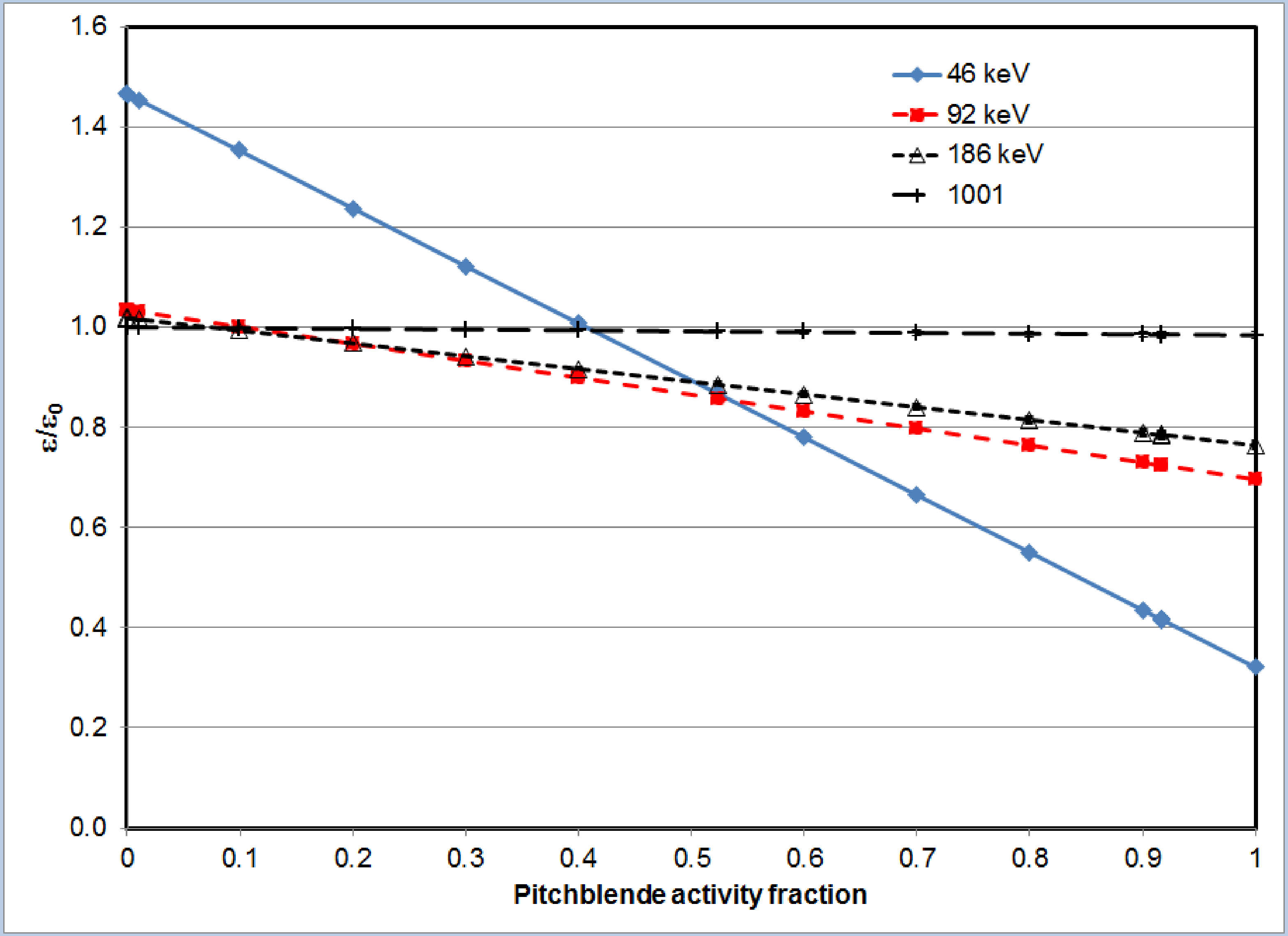
186 keV



1001 keV



Efficiency ratio as a function of activity distribution. P=0.005, d=0.05 cm



- The best estimate of the efficiency and the standard deviation depend on the parameters of the model of inhomogeneity, thus in principle differ from the results obtained assuming the homogeneity of the sample

- The deviations are higher in the case of low energy, large elementary blocks and high inhomogeneity (e. g. one highly attenuating component) than in the opposite cases

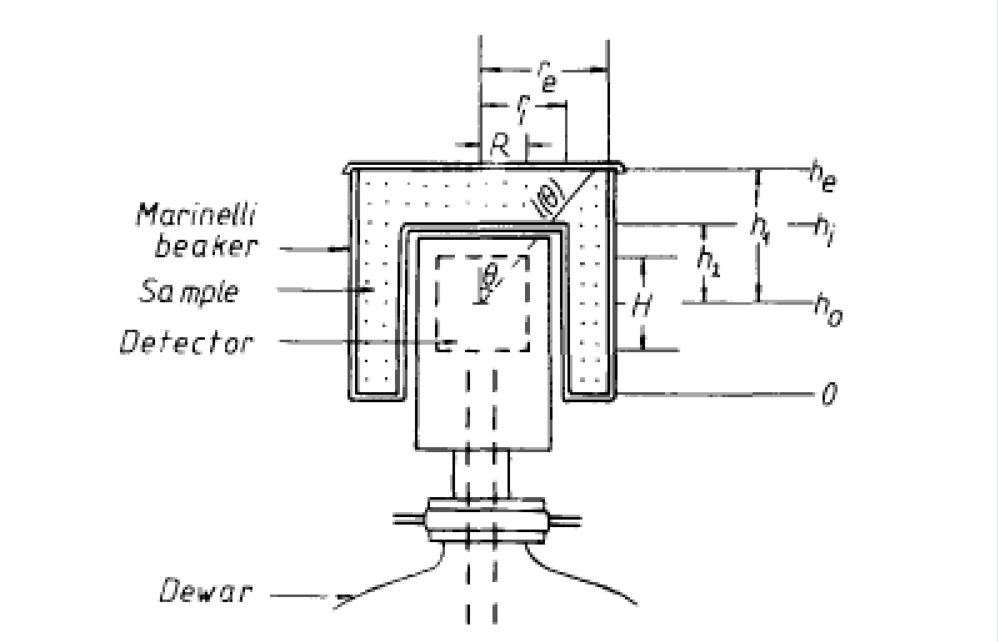
- In the case of weak inhomogeneity the efficiency calibration using homogeneous standards or spiked samples gives usually acceptable results for environmental assessment
- In the case of high inhomogeneity, it is recommended to evaluate the efficiency for the particular composition and granulometry of the sample by Monte Carlo simulation instead of using the homogeneity approximation

- In these cases additional information concerning activity distribution among components is required for the evaluation of the efficiency with low uncertainty

- In absence of such information, a realistic evaluation of the uncertainty can be obtained by simulations using reasonable scenarios of activity distribution among the components.

3. Marinelli beaker samples

- Advantage: highest efficiency for high volume samples
- Self-attenuation computations:
  - Monte Carlo (e.g. GESPECOR, MEFTRAN)
  - Simplified analytical formula



Dryak et al JRNCL 135 (1989) 281

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

Sima, Health Phys. 62 (1992) 445

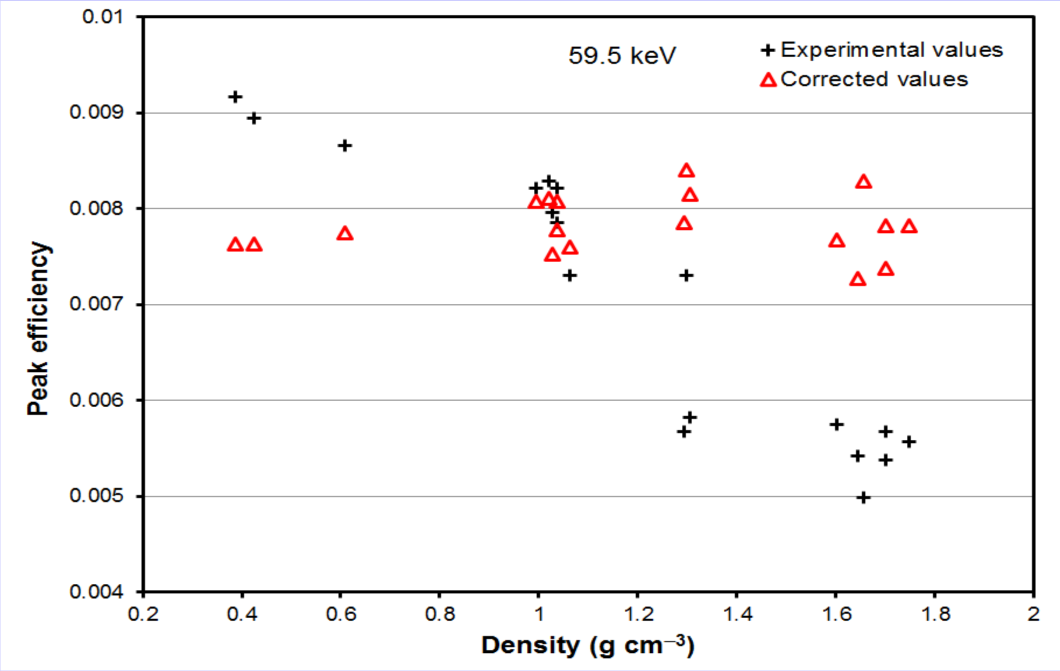
$$t = \frac{2\pi}{\Delta\Omega}\left[F(r_e,h_1)+F(r_e,h_0)-F(r_i,h_2)-F(r_i,h_0)\right]$$

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

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

Example – Marinelli beaker of 1000 cm^3

- spiked reference materials prepared from different matrices (Jerome et al., Nucl. Instrum. Meth. A 339 (1994) 55)
- Experimental efficiencies determined for each sample
- Self-attenuation correction factor computed by the formula, using μ based on sample composition
- Correction factor applied to experimental efficiencies for computing the expected efficiency for a gel sample with density 1 g/cm^3 – P. De Felice et al.



P. De Felice et al.

=> Equivalent peak efficiency for the gel matrix almost independent of the matrix of the reference material

4. Applications of self-attenuation computations

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

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

  Both general purpose programs (GEANT, MCNP) and specific purpose programs have been applied, especially for environmental samples

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

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

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

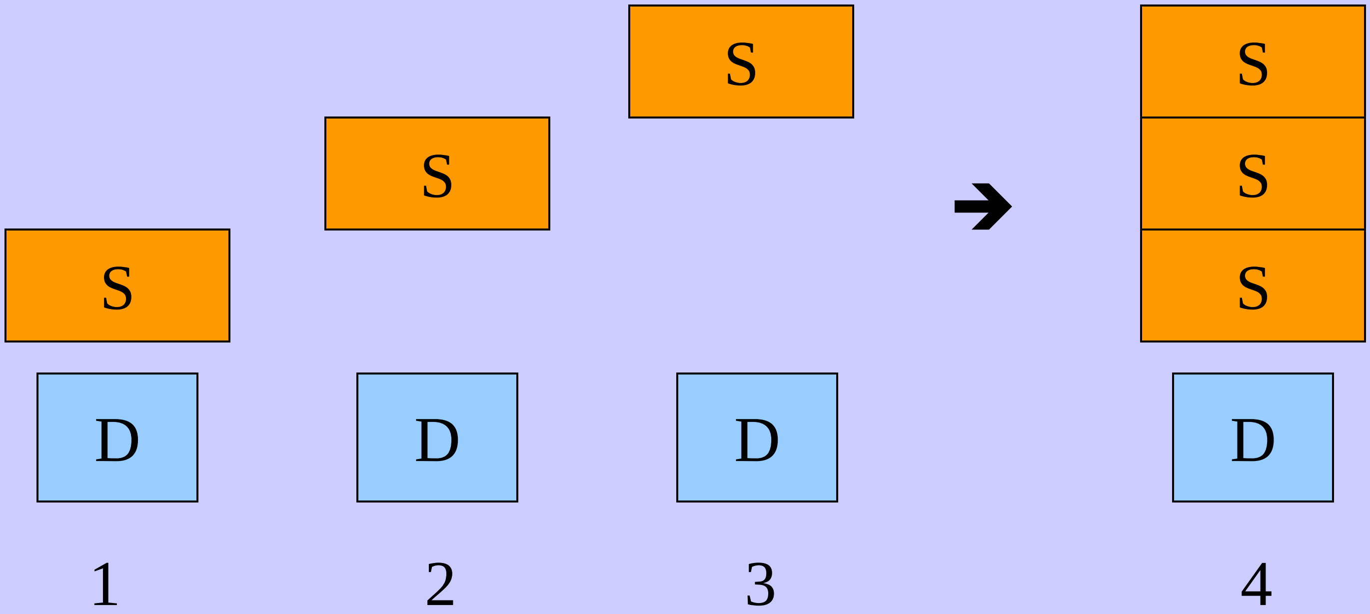
  The values $\varepsilon_{(1)}(E; 0)$, $\varepsilon_{(2)}(E; 0_2)$ … should be compatible.
  The best value of $\varepsilon(E; 0)$ is their weighted average if all are compatible.
  This best value should be used for the computation of the efficiency for other matrices

- 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:

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



⇒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 $V=V_1+V_2+V_3$

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

(Sima and Dovlete, JRNCL 200 (1995) 191)

5. Summary and conclusions

- Self-attenuation effects are very important in the measurement of volume samples
  - Depend mainly on linear attenuation coefficient $\mu(E)$ and sample geometry
  - More important at low energies
  - Slight dependence of self-attenuation on experimental details
- The linear attenuation coefficient can be obtained using
  - Sample composition and density, or
  - Transmission measurements – caution in the case of uncollimated beam!
- Reliable values of the self-attenuation correction can be computed for homogeneous samples
  - Problems in the case of samples with high intrinsic inhomogeneity

Applications:

- Efficiency calibration for samples with any matrix using the computed self-attenuation corrections and the measured efficiency for a standard matrix with the same geometry
- Consistency check of the efficiency curve for different matrices
- Estimate efficiency for samples of larger volumes than the volume of the standard, using properties of efficiency for samples with negligible attenuation