Special problems in self-attenuation

Octavian Sima
University of Bucharest, Romania
National Institute for Physics and Nuclear Engineering
“Horia Hulubei”
Magurele-Bucharest, Romania
Outline

1. Self-attenuation dependence on experimental conditions
2. Sources with intrinsic inhomogeneity
3. Marinelli beaker samples
4. Applications
5. Summary and conclusions
1. Self-attenuation dependence on experimental conditions

Self-attenuation effects depend mainly on:
- linear attenuation coefficient $\mu$
  - 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 $\mu(E)$)

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 $\varepsilon$

$\Rightarrow$ 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), $E_\gamma = 46.54$ keV ($^{210}$Pb)
- Lead with known activity of $^{210}$Pb
- Theoretical values: GESPECOR

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Density (g cm$^{-3}$)</th>
<th>Geometry</th>
<th>$F_{ca}$ (th.)</th>
<th>$F_{ca}$ (exp.)</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb(NO$_3$)$_2$</td>
<td>2.25</td>
<td>25 ml Cyl.</td>
<td>0.105</td>
<td>0.108</td>
<td>0.97</td>
</tr>
<tr>
<td>Zircon sand</td>
<td>3.01</td>
<td>1 l Marinelli</td>
<td>0.043</td>
<td>0.048</td>
<td>0.91</td>
</tr>
<tr>
<td>Pb(NO$_3$)$_2$</td>
<td>2.57</td>
<td>1.155 ml Cyl.</td>
<td>0.120</td>
<td>0.126</td>
<td>0.96</td>
</tr>
<tr>
<td>PbSO$_4$</td>
<td>2.79</td>
<td>1.155 ml Cyl.</td>
<td>0.101</td>
<td>0.101</td>
<td>1.00</td>
</tr>
</tbody>
</table>


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 $\neq$ Cutshall approximation $= \exp(-\mu d)$

O. Sima, ICRM GSWG, Paris, June 2018
Transmission factors (logarithmic scale). Sample: $R=3.5$, $H=2$ cm

Correct value of $T$ – smaller than $\exp(-\mu d)$ (inclined trajectories result in higher attenuation)
In the presence of coincidence summing effects, very difficult to get correct results

\[ \delta \text{ (\%)} = \text{rel. difference between the correct value and the value based on } T=\exp(-\mu d) \]

Soil sample, R=3.5 cm, H=2 cm.

\( \varepsilon \) = peak efficiency

$F_a$ depends mainly on linear attenuation coefficient $\mu$ 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

If the value of $\mu$ 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


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 $ED_1 + ED_2 = E_0 \Rightarrow$ signal in the peak of energy $E_0$ ($ED$=energy deposited in detector)
- the probability of traversing the sample at energy $E'$ depends on $\mu(E')$

$\Rightarrow$ Rigorously in the case of well-type detectors $F_a$ depends on the complete curve $\mu(E)$ for $E < E_0$ and not only on the value $\mu_0$ of $\mu(E)$ for $E = E_0$

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

Observation:
- In case of high attenuation only a thin layer of the sample located close to the detector is important; e.g. for $\mu=10 \text{ cm}^{-1}$ only a layer of a few mm is important
  $\Rightarrow$ 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)

\( \Rightarrow \) depends on sample and on detector resolution

O. Sima, ICRM GSWG, Paris, June 2018
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

Random effects (number and position of blocks of each type) => uncertainty
Systematic effects (activity segregation in different blocks) => distortion of efficiency

O. Sima, ICRM GSWG, Paris, June 2018
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
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:

<table>
<thead>
<tr>
<th></th>
<th>SiO$_2$</th>
<th>Al$_2$O$_3$</th>
<th>Fe$_2$O$_3$</th>
<th>MgO</th>
<th>CaO</th>
<th>Na$_2$O</th>
<th>K$_2$O</th>
<th>TiO$_2$</th>
<th>MnO</th>
<th>P$_2$O$_5$</th>
<th>Org</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight</td>
<td>66.9</td>
<td>13.8</td>
<td>4.1</td>
<td>1.7</td>
<td>2.9</td>
<td>2.2</td>
<td>2.8</td>
<td>0.5</td>
<td>0.1</td>
<td>0.2</td>
<td>5.0</td>
</tr>
<tr>
<td>Density</td>
<td>2.65</td>
<td>4.0</td>
<td>5.25</td>
<td>3.58</td>
<td>3.34</td>
<td>2.27</td>
<td>2.30</td>
<td>4.0</td>
<td>5.43</td>
<td>2.39</td>
<td>1.0</td>
</tr>
</tbody>
</table>

- 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), 186.2 keV ($^{226}$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 $\varepsilon_0$ 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 $\varepsilon_i$ for each scenario
- Inhomogeneity effects higher for the photons with $E=46.5$ keV

<table>
<thead>
<tr>
<th>Activity of the elementary block</th>
<th>d=0.0167</th>
<th>d=0.1</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon/\varepsilon_0$</td>
<td>$R/R_0$</td>
<td>$\varepsilon/\varepsilon_0$</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>Al$_2$O$_3$</td>
<td>Fe$_2$O$_3$</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>10</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>50</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

$\varepsilon_0/\varepsilon_0$ for 46 keV
$R=\varepsilon(46)/\varepsilon(92)$
- Results: generally $\varepsilon_i$ differs from $\varepsilon_0$ 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$_2$O$_3$.

- 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$_2$O$_3$, $d=0.1$ 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, \(\varepsilon_0\) was evaluated
- For each \((d, P, A_P/A_T)\) the efficiency \(\varepsilon\) was evaluated
Size of the blocks: $d=0.5 \text{ mm; } N_t=400000; p_1=0.799, p_2=0.001, p_3=0.20$

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

O. Sima, ICRM GSWG, Paris, June 2018
$p_1=0.795, p_2=0.005, p_3=0.20$
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

\[
\varepsilon = 1 - \exp(-\mu \cdot t)
\]

\[
\frac{\varepsilon_0}{\mu \cdot t}
\]

Dryak et al JRNCL 135 (1989) 281


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

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

\[
\Delta\Omega = 2\pi \left( 1 + \frac{h_0}{\sqrt{h_0^2 + r_i^2}} \right)
\]
Example – Marinelli beaker of 1000 cm$^3$

- Experimental efficiencies determined for each sample
- Self-attenuation correction factor computed by the formula, using $\mu$ 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.

$\Rightarrow$ Equivalent peak efficiency for the gel matrix almost independent of the matrix of the reference material

P. De Felice et al.
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 \( V=V_1+V_2+V_3 \)

\[ \Rightarrow \varepsilon_V(E; m) = F_{av}(E; m; 0) \varepsilon_V(E; 0) \]

(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