Self-absorption correction in gamma-ray spectrometry of environmental samples – an overview of methods and correction values obtained for the selected geometries

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Abstract Of major concern are the self-absorption correction factors C_s in precise gamma-ray spectrometry of environmental samples. The overview of C_s determination methods (experimental, Monte Carlo calculations, analytical function application) is presented. Among several available methods, the author chose the one proposed by Debertin where the uncertainty level of C_s correction factor is 1÷2%. The point-like detector model is assumed whilst its efficiency is taken to be proportional to the weighted sum (integral) of the number of photons coming from sample volume elements. Computer program was developed to support the computation of C_s by the Debertin's method for samples in cylindrical geometries and Marinelli beakers. The input data are sample dimensions, density and the mass attenuation coefficient. The C_s uncertainty due to an arbitrarily chosen position of a point-like detector would not exceed 1% for the considered photon energies and the sample density range. Utilising this computer program, the relationships $C_s(E, \rho)$ were obtained for the applied geometries and a SiO_2 matrix to support routine measurements. The SiO_2 matrix is widely encountered in environmental spectrometry. This relationship was derived by fitting the computed data with the function $C_s(E, \rho)$ in the form proposed by Bolivar. It was shown that while handling this function to samples with the H_2O matrix, the error involved in estimations would not exceed 3%.

Key words gamma-ray spectrometry • self-attenuation • self-absorption • correction factor

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Introduction

The activity of samples in gamma-ray spectrometry is derived from the efficiency curve $\varepsilon(E)$ determined for a calibration source (standard). In the case of volume sources the method gives excellent results when the sample and the standard are handled in the same geometric setup as they have the same chemical composition and density and hence the same self-absorption.

In practical applications, it is not always so as the sample and source materials might be entirely different. Environmental samples may vastly differ in their chemical composition, their density ranging from near 0 to about 2.0 g/cm³. That is why the self-absorption correction factor C_s is required to account for the differences in self-absorption between the source and the standard.

For a given geometric setup, the correction factor is expressed as the ratio of efficiency of standard to that of the sample:

$$(1) \qquad C_s(E) = \frac{\varepsilon(E, \text{standard})}{\varepsilon(E, \text{sample})}$$

No matter which method is selected in the self-absorption correction determination, the procedure is to be applied, whereby C_s is obtained for various densities ρ and photon energies E and then data

collected for each photon energy are fitted to an appropriate function $C_s(\rho)$. In widespread use is the form of the function $C_s(\rho)$ written as:

$$(2) \qquad C_s(\rho) = a\,\exp\cdot(-b\rho)$$

where a, b are the adjustable parameters.

Finally, the procedure yields a family of curves $C_s(\rho)$ corresponding to various radiation energies (see Fig. 2). When the self-absorption correction is sought for the energy for which the relationship $C_s(\rho)$ is not given, the desired values are obtained by interpolation of the available data.

Some authors [2, 17] tend to fit the experimental data with the function $C_s(E,\rho)$ written as:

$$(3) \qquad C_s(E,\rho) = a(E)\exp\cdot(-b(E)\cdot\rho)$$

Thus, the fitted parameters a, b become the functions of energy E, whilst the desired values of C_s are derived straightforwardly from this relationship, for all energies.

While applying the formulas $C_s(\rho)$ and $C_s(E,\rho)$, one has to bear in mind that for low energies of radiation (below about 100 keV) these formulas are applicable only for materials of similar composition on account of the relationship between the mass attenuation coefficient and the atomic number [9].

Self-absorption correction can be determined:
– experimentally,
– using the Monte Carlo computation techniques,
– using analytical methods.

Self-absorption correction for a given geometric setup is obtained experimentally through measurements and comparison of the efficiency ε (or a number of counts in a photopeak) for samples with different densities ρ [3, 4, 9]. The formula (1) is then applied to yield C_s. The experimental method, however, is time-consuming and inconvenient, and it requires that the curves be fitted to a small number of measurement data, which gives rise to uncertainty.

In practical applications, self-absorption corrections are calculated using computing techniques, which prove to be faster and more universal than experimental methods. Of major importance are widely applied analytical methods allowing quick estimation of corrections in laboratories. Monte Carlo techniques are not so widely adopted in the laboratory conditions, as they require considerable skill and experience in computer simulations.

While compared to techniques of efficiency ε computing, the methods involving computations of C_s are less biased by simplifications and uncertainties due to detector dimensions. The self-absorption correction factor is expressed as the ratio of efficiency of standard to that of the sample and, as such, ought to be treated as a relative term. As a consequence, the errors in efficiency computations will be partially cancelled.

In Monte Carlo simulations, the history of each photon is tracked, starting from the moment it is emitted from the source till its energy is wholly dissipated or till it leaves the analysed space. Two methods of computing the correction factor C_s are used. In the first approach, the efficiencies of the standard and the sample are computed and their ratio is obtained in accordance with Eq. (1) [11, 18, 21]. In the latter method, the photon path distribution in the sample is analysed and the correction factors derived from the distribution parameters, in the simplest case, on the basis of the mean path [10, 13, 20].

The exact analytical description of self-absorption is a complex task, that is why simplified models are adopted instead. In the first place, it is assumed that the self-absorption correction is proportional to the term $\exp(-\mu x)$ or $\exp(-\mu_m \rho x)$, where μ, μ_m – linear and mass attenuation coefficient, respectively, x – sample thickness (or effective thickness) [10, 16].

Another widely applied analytical formula [2, 12, 14] providing a simplified description of self-absorption in cylindrical samples is known as the self-absorption equation involving the summation (integration) of photons of the specified energy coming from subsequent sample layers (normally for layer surface) and reaching the detector. Absorption in the sample layers below is taken into account. Accordingly

$$(4) \qquad C_s^a = \frac{\mu x}{1-\exp(-\mu x)}$$

where: C_s^a – self-absorption correction for the sample with reference to air (matrix $\mu \approx 0$).

In order to apply this formula, it is required that self-absorption be determined for the standard (index c) and the currently measured sample (index s) with reference to air. Accordingly:

$$(5) \qquad C_s(E) = \frac{C_{s,s}^a(E)}{C_{s,c}^a(E)} = \frac{\left[1-\exp\left(-\mu_c(E)x\right)\cdot\mu_s(E)\right]}{\left[1-\exp\left(-\mu_s(E)x\right)\cdot\mu_c(E)\right]}$$

Dryák *et al*. [8] proposed a new interpretation of the self-absorption equation (valid for cylindrical samples), so that it would be applicable to Marinelli geometries as well. The sample thickness in Eq. (4) is replaced by effective thickness. The formula governing the effective thickness in the Marinelli geometry was proposed by Sima [19], who highlighted the underlying assumptions as well as the major constraints.

Another analytical method of determining the self-absorption correction was proposed by Debertin *et al*. [6, 7]. It is outlined in more detail in the following section.

In order to apply C_s computing techniques, it is required that attenuation coefficients of the standard and sample materials be known beforehand. For high radiation energies (in excess of 100 keV or 200 keV), the mass attenuation coefficient is obtained on the basis of the sample approximate chemical composition, assuming that for a given energy the value of C_s depends on the sample density exclusively. It is a well-established fact that for this energy range mass attenuation coefficient μ_m is almost constant for all materials to be found in the natural environment.

For low energies (below about 100 keV), the mass attenuation coefficient in different materials may vary significantly, hence the influence of chemical composi-

tion on the sample absorption features appears to be decisive and the attenuation coefficient has to be determined experimentally. The transmission method developed by Cutshall [5] links the transmission measurement of the attenuation coefficient with the self-absorption Eq. (4) for cylindrical samples. In transmission measurements, a point-like radioactive source is positioned above the sample located on the detector and the number of counts (originated from the source) in the full energy peak is measured. The sample self-absorption correction factor C_s with reference to the standard is given as [12]:

$$(6) \qquad C_s = \frac{\ln\left(I_c / I_s\right)}{1 - I_s / I_c}$$

where: I_c, I_s – transmission experiment results (detector count rates) for the standard and sample, respectively.

Methods

In the present study, the self-absorption correction factor C_s is derived utilising computer program based on the method proposed by Debertin *et al.* [6, 7]. In Debertin's method the point-like detector model is adopted and detector efficiency for the specified geometry is assumed to be proportional to the weighted sum (integral) of photons coming from individual volume elements dV, into which the sample may be divided (Fig. 1). The weight of each element of the volume dV is determined by:
- the corresponding solid angle – proportional to the inverse of the squared distance z between the element dV and a point-like detector,
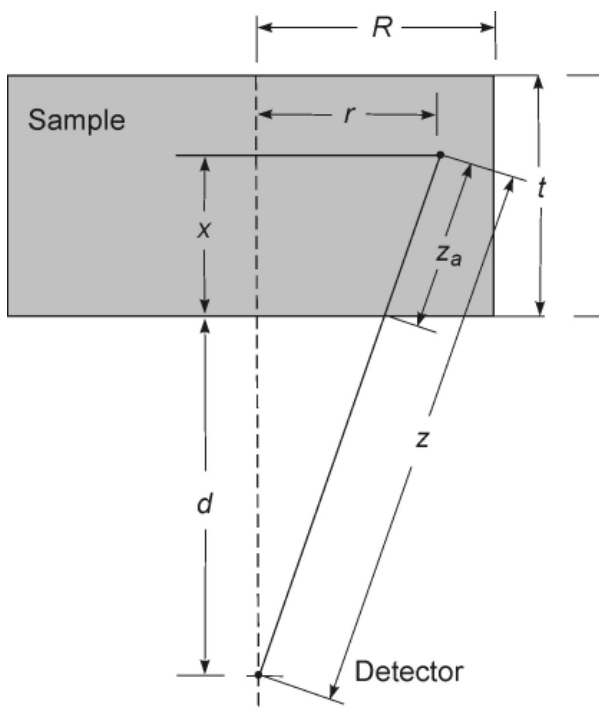
Fig. 1. Cylindrical sample positioned on a point-like detector – quantities used in Eq. (9).

- self-absorption in the sample layer z_a through which radiation can pass – proportional to $\exp[-\mu(E)z_a]$.

The overall efficiency for the whole sample is the integral of efficiency contributions of all elements dV, over the sample volume:

$$(7) \quad \varepsilon(E) = \frac{1}{V} \int_V \varepsilon_{dV}(E) dV = \frac{1}{V} \cdot \int_V \frac{c(E)\exp\left(-\mu(E)z_a\right)}{z^2}$$
$$\cdot \, dV = \frac{c(E)}{V} I(\mu)$$

where: $c(E)$ – proportionality factor; $I(\mu)$ – term associated with self-absorption with reference to air, dependent on μ and easy to compute.

Accordingly, we get:

$$(8) \qquad C_s(E) = \frac{\varepsilon_c}{\varepsilon_s} = \frac{c(E)/V}{c(E)/V} \frac{I\left(\mu_c\right)}{I\left(\mu_s\right)} = \frac{I\left(\mu_c\right)}{I\left(\mu_s\right)}$$

It is worthwhile to mention that the terms $I(\mu_s)$ and $I(\mu_c)$ do not represent the efficiency or the solid angle for the given geometric setup, whilst their ratio C_s has a specific physical interpretation.

For cylindrical geometries (see Fig. 1), the formula yielding $I(\mu)$ assumes the form [6]:

$$(9) \qquad I(\mu) = \int_0^R \int_0^t \frac{\exp\left(-\mu(E)z_a\right)}{z^2} \cdot dx \cdot dr$$

where $z_a = x\sqrt{r^2 + (x + d)^2}/(x + d)$.

Debertin and Ren [7] introduced a formula yielding $I(\mu)$ for samples in Marinelli beakers, analogous to Eq. (9). For clarity, the authors divide the sample in three parts and for each part a simple formula is derived and the integration procedure applied.

Debertin's method, widely adopted in many laboratories, gives good results, the uncertainty level approaching 1–2% [7, 15].

The author developed a computer program to compute the correction factor C_s, following the Debertin method. This program is written in FORTRAN and allows the computation of correction factors for samples in cylindrical geometry and for samples in Marinelli beakers.

This program utilises the subroutine DGAUSS from the CERN library to compute $I(\mu)$, applying the integration procedure (Eq. (9)) proposed by Debertin.

The input data are:
- sample dimensions (e.g. for cylindrical samples: diameter and height),
- distance between the point-like detector and the sample lower surface positioned over the detector,
- linear attenuation coefficient or mass attenuation coefficient and sample density.

When cylindrical geometries are handled, Eq. (9) can be applied directly. In the case of Marinelli beakers, however, the computation procedure becomes more complex as the point-like detector is positioned in the sample cavity. In that case the source, is to be divided

into two cylindrical parts and the integration procedure is performed for these two sections separately. The first cylinder is positioned above the point-like detector, the other is below. In other words, the sample is divided by a plane parallel to the detector window and passing through the point-like detector. In such sample partitioning, the integration is performed over the cavity, too. Integrand values for the points in the cavity are assumed to be 0.

Furthermore, an assumption is made that the point-like detector is positioned in the geometric centre of a real detector. It appears that the computed value of C_s depends on the point-like detector position in a minor degree only, which is borne out by data provided by Debertin and Ren [7] and by the author. The author considered a HPGe detector with 40% efficiency (a cylinder 6.1 cm in diameter and 6 cm in height), the samples being positioned directly on the detector. The distance between the point-like detector and the sample lower surface was equal to 3.91 cm. The difference between the values C_s for various point-like detector location on real detector axis would not exceed 2.5% for the considered parameters (i.e. for energies in excess of 150 keV and the density range 0÷2.4 g/cm^3). Under the assumption that the point-like detector does not lie on the crystal upper surface but deeper inside, the corresponding correction factor C_s uncertainty estimation approaches 1% and for typical values of C_s (0.85÷1.15) this uncertainty goes down to 0.5%. For lower energies (60÷150 keV), the uncertainty level would rise to 3% (1% for typical values of C_s).

Results and discussion

Utilising the developed computer program, the correction factor C_s was computed for two widely applied sample geometries: a cylindrical geometry (beaker) (diameter: 70 mm, height: 31.5 mm) and a Marinelli beaker (volume: 710 ml, external diameter: 125 mm, layer thickness: 19 mm). It is assumed that the sample is positioned directly on the detector. C_s values were computed for energy 150÷2600 keV and in the sample density range from 0.001293 g/cm^3 (air) to 2.40 g/cm^3.

It was assumed that the sample and standard materials display the same absorption properties as silica (SiO$_2$); in other words, the mass attenuation coefficient of silica was applied. The following self-absorption correction factors were calculated:
– C_s^a – with reference to air. This quantity represents the level of self-absorption in the sample and allows the value of C_s to be computed for the silica standard with any density (see Eq. (5)),
– C_s – with reference to the currently employed standard (silica with the density 1.5 g/cm^3).
The calculated self-absorption correction with respect to the standard does not exceed 19% for the Marinelli beaker and 25% for cylindrical geometries (see Fig. 2).

Routine measurements need not be supported each time by the computer program to compute C_s. The functional relationship $C_s(E,\rho)$ is employed. This relationship for SiO$_2$ is obtained through fitting the computed C_s values with the function proposed by

Bolivar *et al.* [1], derived from the self-absorption equation:

$$(10) \quad C_s(E,\rho_s) = \exp\left[c_1 \exp\left[c_2 (\ln E)^2\right](\rho_s - \rho_c)\right]$$

where c_1, c_2 are the adjustable parameters.

The correction factor derived by the author is governed by the functions:
– for Marinelli beakers

$$(11) \quad C_s^a(E,\rho_s) = \exp\left[0.32017 \exp\left[-0.033624 (\ln E)^2\right]\rho_s\right]$$

$$(12) \quad C_s(E,\rho_s) = \exp\left[0.31267 \cdot \exp\left[-0.033247 (\ln E)^2\right](\rho_s - \rho_c)\right]$$

– for cylindrical geometries

$$(13) \quad C_s^a(E,\rho_s) = \exp\left[0.41837 \exp\left[-0.032860 (\ln E)^2\right]\rho_s\right]$$

$$(14) \quad C_s(E,\rho_s) = \exp\left[0.40116 \cdot \exp\left[-0.032194 (\ln E)^2\right](\rho_s - \rho_c)\right]$$

The differences between the values of the fitting function and the computed values of C_s and C_s^a do not exceed 1% for the cylindrical geometry (0.6% when $0.85 < C_s < 1.15$) and 0.5% for Marinelli beakers. Figure 2 shows the plot of $C_s(E,\rho)$ for the relevant geometries.

When samples of any other material are considered, thus C_s values computed for silica must be regarded with caution as the absorption properties of the sample and the standard material are assumed to be the same as of silica. For example, when the sample material
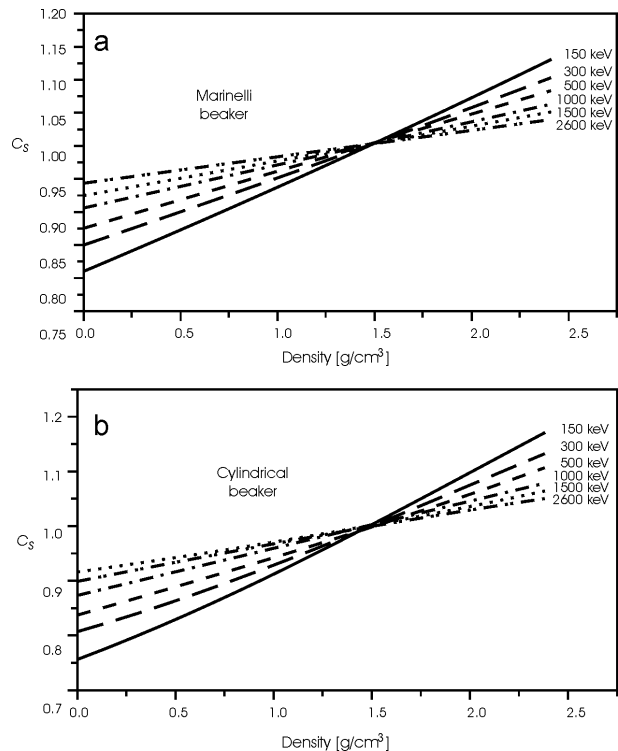
Fig. 2. $C_s(E,\rho)$ with reference to the standard (a) for Marinelli beakers (b) for cylindrical beakers. This relationship is derived from data obtained using the Debertin method. It is assumed that the sample and the standard display absorption properties of the silica, the standard density being 1.5 g/cm^3.

displays the absorption properties of water (μ_m values for water and silica differ significantly), the error involved in estimation of C_s goes up to 3%. In precise measurements, when the sample chemical composition is known, and particularly when the mass attenuation coefficient of the sample vastly differs from that for silica, the C_s values computed for silica need not be applied, instead C_s values ought to be estimated individually for each sample, e.g. utilising the computer program based on the Debertin method.

Conclusions

On the basis of an overview of available methods, the Debertin method for the determination of self-absorption correction factor C_s was chosen. Utilising the computer program based on this method the relationships $C_s(E,\rho)$ were determined for the applied geometries and the SiO_2 matrix.
–　In the case of routine measurements of typical materials, C_s values can be calculated using the relationships $C_s(E,\rho)$ determined for SiO_2 matrix.
–　In the case of precise measurements C_s values ought to be calculated individually for each sample, e.g. using the Debertin method.

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