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Determination of gamma-ray self-attenuation correction in environmental samples by combining transmission measurements and Monte Carlo simulations

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- Gamma-ray spectrometry.
- Self-attenuation correction.
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- Linear attenuation coefficient.

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We develop a simple and widely applicable method for determining the self-attenuation correction in gamma-ray spectrometry on environmental samples. The method relies on measurements of the transmission of photons over the matrices of a calibration standard and an analysed sample. Results of this experiment are used in subsequent Monte Carlo simulations in which we first determine the linear attenuation coefficients (μ) of the two matrices and then the self-attenuation correction for the analysed sample. The method is validated by reproducing, over a wide energy range, the literature data for the μ of water. We demonstrate the use of the method on a sample of sand, for which we find that the correction is considerable below ~ 400 keV, where many naturally occurring radionuclides emit gamma rays. At the lowest inspected energy (~ 60 keV), one measures an activity that is by a factor of ~ 1.8 smaller than its true value.

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1. Introduction

In order to obtain a proper analytical accuracy in gamma-ray spectrometry, one must carry out an efficiency calibration for every particular measurement setup. This, however, does not lead straightforwardly to accounting for every aspect of the mentioned requirement.

In particular, determining the self-attenuation of gamma rays within an environmental sample may often pose a considerable problem, which calls for versatile solutions that can be easily applied in standard gamma spectrometric measurements.

In most measurement setups in gamma-ray spectrometry on environmental samples, radionuclides are homogeneously

distributed in a certain material that is confined in a container, and such a sample is brought in contact with a detector of high-energy photons. The chemical composition and density – that is, characteristic self-attenuation properties – of the material define the matrix (of a sample). The shape and volume of the container, together with its position and orientation relative to the detector, is termed the geometry (of a measurement setup). If a sample contains radionuclides of known activities, distributed in a certain matrix, we can use it to perform an efficiency calibration for the given geometry; we term such a sample the calibration sample (sample C). This calibration sets a foundation for the determination of unknown activities in a measured sample (sample M) which comprises its own matrix but is measured in the same geometry. The condition of the same geometry for samples C and M guarantees that both samples emit the same percentage of photons in the direction of the detector. However, only if the matrices of samples C and M are the same, their self-attenuation properties are equal and the same portions of photons reach the

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detector in both cases. Though this situation can be realised experimentally (McMahon et al., 2004), it is not common to have a (spiked) sample C for every matrix. Moreover, the composition of every matrix material is not always known, and even if it is known, matrix packing density (affects self-attenuation properties) may vary from sample to sample due to partial reproducibility in filling a container. For instance, samples of soil may vary both in composition and density considerably (Bonczyk, 2013).

A number of published papers address methods for solving the above problem. Some of these papers offer solutions via transmission measurements with collimated sources, the results of which are then used in calculating the corresponding self-attenuation corrections (Cutshall et al., 1982; Khater and Ebaid, 2007; Misiak et al., 2011; Al-Masri et al., 2013). A disadvantage of this method is that it requires equipment for collimation of gamma rays, which could often introduce a necessity for substantial modifications of a measurement setup. There are approaches that focus on using Monte Carlo (MC) simulations for the calculation of a self-attenuation correction (Sima, 1996; Sima and Arnold, 1996; Vargas et al., 2002). In this method, the linear gamma-ray attenuation coefficient μ [accounting for the photon transmission probability over a distance L being $\exp(-\mu L)$] of a measured sample is a critical input parameter for an MC calculation. It is, therefore, essential to know *a priori* the value of μ , which is the main downside of the method since μ depends not only on (sometimes unknown) matrix composition but also on its density when packed in a container. There are also other computational approaches, often in the form of a software, which either require μ as an input to the model (Korun, 1999; Sima and Arnold, 2002; Hurtado et al., 2004), or use self-attenuation factors which include only density dependent data from transmission measurements (Bolivar et al., 1997; McMahon et al., 2004).

A common shortcoming of all the methods outlined is the lack of an experimental insight into the μ of a sample, which one often deals with by using the most reasonable approximation – knowing the type of a matrix – from the literature. Furthermore, while the mentioned approaches may be useful in certain situations, they are of limited applicability in the most common situation when sample M is just placed on the top of a detector and measured after the setup has been calibrated with sample C. In this paper, we elaborate a method for finding the self-attenuation correction factor in this case, using a combination of a simple transmission experiment and MC simulations. In our approach, no modification of a measurement setup is required and μ is determined on the basis of measurement results for every particular sample. The method is based on three transmission measurements involving an absorption matrix between a source of gamma rays and a photon detector. The absorption matrices are air, the matrix of sample C (without gamma emitters), and the matrix of sample M. Results of these measurements are used as inputs to the MC simulations which lead to obtaining μ for the matrices of samples C and M. Finally, the results for μ are processed in subsequent MC simulations for finding the self-attenuation correction to the activity of sample M. We demonstrate the whole procedure in a stepwise manner by using an aqueous solution of radionuclides as sample C, and sand as sample M. The method is validated by reproducing the literature data for the μ of water over a wide energy range. Above ~ 400 keV, the self-attenuation correction is close to unity and the measured activity of sand is close to the true activity. However, the correction becomes appreciable at low energies, being as high as ~ 1.8 at ~ 60 keV, which means that low-energy activities are underestimated considerably if the self-attenuation phenomena are disregarded.

2. Description of the method

Self-attenuation poses a nontrivial problem in gamma-ray spectrometry on environmental samples, especially if a sample is thick and/or contains a matrix with a high μ . We develop a method which offers a practical solution to this problem, having in mind that such a method must be simple and widely applicable while preserving scientific rigour. Furthermore, it should include not only numerical simulations but also experimental indicators, as only in this way one can take into account departures from idealised (i.e., fully theoretical) measurement conditions. Thus, our approach is based on combining an undemanding gamma spectrometric transmission experiment and appropriate MC simulations. In this section, we formulate the problem and explain the experimental design, whereas the MC simulations are presented in Section 3. In Section 4, we exemplify the use of the method, step by step, by turning to the self-attenuation in a realistic sample.

2.1. Calibration and measurement in gamma-ray spectrometry

We start by addressing arguably the most common measurement setup in gamma-ray spectrometry on environmental samples, where the unknown activities of radionuclides in a measured sample are quantified on the basis of an appropriate efficiency calibration. In Fig. 1, we show a typical setup for both an efficiency calibration (with sample C) and a subsequent measurement of sample M. Geometrical relations between the samples and the detector are in both cases identical, i.e., the size and shape of the sample containers (in this case, cylindrical boxes of the same radii r_b and heights h_b), as well as their positions and orientations relative to the detector (a cylinder of a radius r_d), are the same. The calibration explicates how many photons – emitted in random directions from random positions – will be detected by the detector, which also includes the situation where an emitted photon is absorbed within the matrix before it leaves sample C. Only if the matrices of samples C and M are the same – or at least have very similar self-attenuation properties – they absorb the same (or very similar) number of photons and the calibration with sample C is sufficient for determining the activity of sample M. If this is not the case, a self-attenuation correction for sample M should be found by additional considerations.

The general equation which relates the activity A_s of a sample at a given energy to the rate $R_{m;g}$ of counts recorded by a detector is

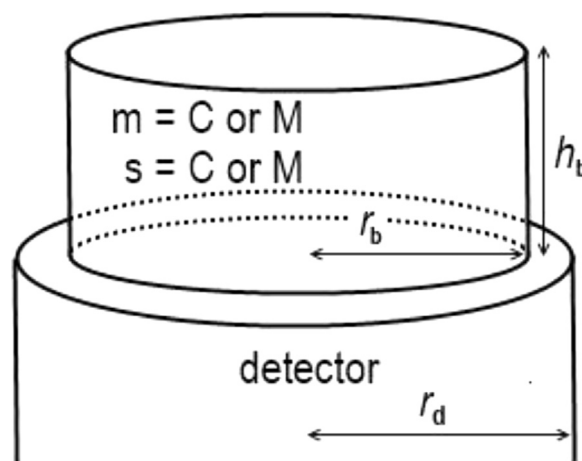


Fig. 1. A common setup for calibration and measurement in gamma-ray spectrometry on environmental samples. We designate this geometry as γ , the source of gamma rays is denoted by s , and the sample matrix by m (both being either C or M).

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