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Uncertainty of gamma-ray spectrometry measurement of environmental samples due to uncertainties in matrix composition, density and sample geometry

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HIGHLIGHTS

- Uncertainties of gamma-ray spectrometry measurements were assessed.
- Efficiencies were calculated for a wide range of environmental matrices.
- The effect of matrix compositions and density on efficiency was studied.
- The effect of geometry parameters on efficiency was considered.

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ABSTRACT

This paper is intended to identify the uncertainties of activities in environmental samples measured with gamma-ray spectrometry that result from uncertainties in matrix composition, density and geometrical dimensions of the sample. For that purpose efficiencies were calculated for a wide range of environmental matrices such as fresh and ashed food samples, water samples and soil samples. Compositions were mainly taken from literature. Densities and geometry parameters were varied in a range occurring in practice. Considered energies cover a range from 46.5 keV to 2000 keV. Finally, a couple of recommendations in respect to gamma-ray spectrometric measurements of environmental samples are given.

1. Introduction

Gamma-ray spectrometry is widely used to determine activities of a variety of radionuclides in environmental samples.

For the quantification, an efficiency calibration needs to be done that allows the connection between the measured signal and the activity in the sample. Efficiency in gamma-ray spectrometry does not only depend on the measurement system, but also on the density and composition of the sample, the sample-detector geometrical arrangement and the photon energy. Efficiency calibrations can be done in different ways. On the one hand efficiency calibration can be carried out using calibration sources which equal the sample to be measured in geometry (container, filling height of the sample and sample-detector arrangement), matrix and density. As a result, for different samples different calibration sources need to be prepared which may be very difficult to realize for all samples occurring in an environmental laboratory.

Another possibility is to prepare a smaller number of calibration sources, e.g. aqueous solutions in different containers, maybe also with various filling heights, and subsequently, if the sample and calibration source differ with respect to matrix and density, make a correction for the different self-attenuation of calibration source and sample. Software packages as for example GESPECOR (Sima et al., 2001; Sima and Arnold, 2002) are available for that purpose. Efficiency transfer software is also usable to transfer efficiencies measured for one geometry to another (Lépy et al., 2001).

A third procedure, which can be applied in the case when the detector has been characterized by factory measurements, is to perform mathematical efficiency calibrations. Without the need for radioactive sources, the user can compute the efficiencies directly, for example with the mathematical efficiency calibration software LabSOCS. The combination of detector characterization produced with the MCNP modeling code, mathematical geometry

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templates, and physical sample parameters allows the user to perform efficiency calibrations for various samples without the need for radioactive sources (Bronson, 2003; Venkataraman et al., 2005).

In any case the composition of the sample should be known. The composition can be determined using XRF- or mass-spectrometry, but in most radioactivity laboratories these methods are not available. Large variation of matrices and densities may also occur within one single sample type. Carrazana González et al. (2010) varied in a Monte Carlo-based hypothetical proficiency test the chemical composition of soil samples for the quantification of low-energy gamma-ray emitters keeping the density constant. At e.g. 46.5 keV efficiency ratios of four soils of different compositions related to a reference soil composition ranging from 0.696 to 1.357 were determined. There is one possibility to determine self-attenuation correction factors for samples with unknown compositions by making gamma-ray transmission measurements (Cutshall et al. (1983); McMahon et al. (2004)). But facing the variety of samples, measured by environmental monitoring laboratories, this time-consuming method will only be used in special cases when very small uncertainties are needed. So usually estimations of the composition of the sample need to be made.

Also small differences in the geometry of the sample to be measured and the calibration source lead to uncertainties in the efficiency determination. Reading of the filling level of the sample is done with uncertainties. Wall thicknesses of sample containers may change and also have a statistical distribution, either as a result of manufacturers tendency to reduce material costs, or as a specific tolerance in the manufacturing process.

For the assessment of these uncertainties, which lead to uncertainties in the efficiency determination, energy-dependent efficiencies were calculated for a wide range of environmental matrices as fresh and ashed food samples, water samples and soil samples. Also density and geometry parameters were varied focusing on values which occur in the practice of an environmental monitoring laboratory. Finally, some recommendations concerning gamma-ray spectrometric measurements of environmental samples are given.

2. Methodology

Tested matrices were food (ashed and fresh samples), water samples and soil samples. The compositions were mainly taken from data in literature; the other parameters (geometry, density) were fixed on values that are realistic for the particular sample type. To study the density effect, a density range was chosen, that represents the densities that usually occur in the measurement of environmental samples. Geometry parameters that were considered are the container wall thickness and the sample filling height. For all investigations, only one parameter was varied and the others were fixed to study the influence of the regarded parameter.

2.1. Efficiency calibration

The commercially available calibration software LabSOCS based on Monte Carlo simulation and factory measurements (Venkataraman et al., 2005) was applied to establish mathematical efficiency calibrations. The LabSOCS software uses cross section data of nuclides, derived from the 2002 MCPLIB04 library. Efficiencies were computed for 10 photon energies (46.5, 59.5, 81.0, 105.3, 122.1, 136.5, 165.9, 238.6, 364.5, 583.2, 661.7, 911.1, 1173.2, 1460.8, and 2000 keV) and for a Broad Energy HPGe detector (p-type crystal; crystal diameter: 70 mm; crystal thickness: 31 mm; extra thin, around 0.3 μm N+ contact on the front window side; thickness of the carbon epoxy window: 0.5 mm). Uncertainties of mathematical efficiency calibrations amount to values between 4% for higher energies and 10% for low energies. These values were taken from the LabSOCS user manual. However, the uncertainties of the calculated relative deviations of the efficiency due to the variation of one input parameter are much smaller, as uncertainties deriving from an inexact detector characterization are almost completely canceled out.

2.2. Sample containers

The following sample containers were used for the investigations: 1 L Kautex bottle (inner diameter=93 mm) filled with 1 L, 500 mL, and 100 mL sample volume, corresponding filling heights are 158 mm, 78 mm, and 17 mm, respectively; 100 mL Kautex bottle (inner diameter=47 mm) filled with 100 mL, 50 mL, and 10 mL sample volumes, corresponding filling heights are 61 mm, 30 mm, and 7.5 mm, respectively; 1 L Marinelli beaker (inner diameter at top=159 mm, filling height=70 mm below and 52 mm above the top of the endcap) filled with 1 L sample volume. The filling heights are given from the deepest inner point of the container.

2.3. Tested environmental matrices

2.3.1. Food samples

Food samples are usually ashed before measurement to reduce their volume and to get a homogeneous sample. Their compositions can be found in food composition and nutrition tables. For this study, average values were taken from Souci et al. (1994) and are listed in Table 1. A review of databases in the internet showed

Table 1
Composition of the 15 examined food ashes (average values in % of mass) taken from Souci et al. (1994). Effective atomic numbers (mass-weighted average atomic numbers) are given in the last column.

Ash	Zn	Fe	Mn	Ca	K	Cl	P	Si	Al	Mg	Na	O	NO_3	B	Effective atomic number
Milk (raw milk)				16.2	21.2	13.8	12.4			1.6	6.5	28.3			14.7
Beef muscle	0.3	0.2		0.5	28.9	4.2	15.8			1.9	4.6	43.6			13.0
Pork muscle	0.2	0.1		0.3	39.8		19.4			2.6	5.3	32.3			14.1
Wheat	0.2	0.2	0.2	2.1	21.2	3.1	18.9	0.4		7.1	0.5	46.1			12.6
Rye	0.2	0.3	0.2	3.4	26.8	1.1	17.7	0.5		6.3	0.2	43.3			13.1
Lettuce				2.8	23.9	7.9	3.1	0.3	0.1	1.2	1.0	29.3	30.4		11.9
Potato				0.6	40.3	4.4	4.9			2.0	0.3	46.6	0.9		13.3
Asparagus		0.1		4.2	32.7	8.6	7.4			2.9	0.7	32.7	10.7		13.5
Carrot		0.2		4.8	33.7	7.1	4.1			2.1	7.0	35.2	5.8		13.7
Apple		0.2		2.2	45.0	0.7	3.8	0.2		2.0	0.9	44.1	0.9		13.7
White cabbage				7.8	35.3	6.3	4.7			3.9	2.0	33.2	6.8	0.1	13.9
Spinach		0.3		8.3	41.9	3.6	3.6			3.8	4.3	23.2	11.0		14.5
Trout				0.9	31.3		18.6			2.0	4.8	42.4			13.1
Hering				2.3	24.5	9.9	17.0			2.1	8.0	36.2			13.4
Cod				2.0	29.4	13.1	15.2			2.1	6.0	32.2			14.0