

Applied Radiation and

Applied Radiation and Isotopes

journal homepage: <www.elsevier.com/locate/apradiso>

Improving the assessment of activity in samples with non-uniform distribution using the sum peak count rate

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HIGHLIGHTS

- \bullet A ⁶⁰Co point source placed in several positions within a soil sample was measured.
- The ratio of the count rate R (2505 keV)/R (1332 KeV) is strongly correlated with the apparent efficiency for 1173 keV.
- The correlation is also present in the measurement with a Compton-suppressed spectrometer.
- A method for the computation of the activity valid when a hot particle is present in the sample was proposed.
- The method reduces the uncertainty of the computed activity which is due to the uncertainty of the source distribution.

article info

Available online 22 March 2013

Keywords: Gamma-ray spectrometry Hot particle $^{60}\mathrm{Co}$ Coincidence summing effects Monte Carlo simulation

ABSTRACT

In this work a method for the evaluation of the activity when a point source containing 60 Co is located in an unknown position within a sample is developed. The method can be applied if the count rate in the 2505 keV sum peak has an acceptable uncertainty. It is based on the correlation between the apparent efficiency for the 1173 keV peak and the ratio of the count rate in the sum peak of 2505 keV and in the 1332 keV peak. The correlation was observed in the measurements of a 60 Co point source located in various positions in a soil sample. The measurements were done with a 47% efficiency n-type HPGe detector. The correlation is also observed in the measurements and simulations done with a Comptonsuppressed spectrometer having a 100% n-type HPGe detector. The results obtained with the proposed method are less affected by the uncertainty of the position of the point source than the results obtained using the standard methods of activity evaluation.

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1. Introduction

In gamma-ray spectrometry, frequently volume samples are assessed by assuming homogeneous distribution of radioactive nuclei. In specific cases this hypothesis is false. If the distribution is known, then appropriate efficiency values can be computed and the correct activity can be reported (see e.g. [Sima, 1996; Carconi](#page--1-0) [et al., 2012](#page--1-0) for the case of radon distribution). If the distribution is not known, but some degree of inhomogeneity is suggested by specific tests [\(Pauwels et al., 1998; Suvaila et al., 2012](#page--1-0)), then an additional variance of the computed activity should be added to take into account the effects of radioactivity distribution.

A particular case of inhomogeneous distribution is observed in environmental samples containing hot particles, i.e. high (compared with the average) activity concentrations in a small volume

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of the complete sample. If the sample is analyzed assuming uniform distribution of activity, then the computed activity is biased and also the activity concentration is biased and not representative for the particular environmental factor. The bias in the value of the activity depends on the position of the hot particle within the measured sample.

In this paper we show that in the case of hot particles containing nuclides like 60 Co, 134 Cs, measured with high efficiency detectors, information about the presence of the hot particle and about its location can be inferred using the count rate from the sum peaks if they are observed in the spectra. Furthermore, the bias of the computed activity can be substantially reduced using the additional information provided by the sum peaks.

2. Experimental method

Most of the measurements reported in this work were done in the gamma-spectrometry laboratory of the Physics Department,

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University of Bucharest, Romania, using an n-type coaxial HPGe detector with 47% relative efficiency (spectrometer 1). In addition, several measurements were done with the Compton-suppressed spectrometer located in the underground laboratory of the IAEA's Environment Laboratories, Monaco (spectrometer 2). This spectrometer is based on a 100% relative efficiency n-type HPGe detector and has an anti-Compton shield comprising an annular NaI(Tl) detector housing the HPGe detector and a top NaI(Tl) module placed above the sample inside the channel of the annular detector.

In the measurements done with spectrometer 1 a point source of ⁶⁰Co with (5230 +/−50) Bq activity (1 σ) was used. The source active material is a very thin layer with a radius less than 2 mm placed between two polyethylene foils welded together over the whole area and encased in a circular aluminum frame with the thickness of 2 mm. For the present measurements the assembly was further covered with a thin aluminum foil. The ⁶⁰Co source was placed successively in different positions within a soil sample ([Suvaila, 2011](#page--1-0)). In order to place the source in a given position, a layer of soil was put into the container (inner radius 3.7 cm, inner height 3.7 cm.) and pressed gently; the operation was repeated until the thickness of the soil had the desired value. The target density of all the soil samples was 1.34 g cm⁻³; the actual density was in the interval (1.27–1.36) g cm^{−3}. Then the ⁶⁰Co source was positioned over the layer of soil, at a radial distance R from the axis of the container (always centered on the end cap of the detector). Finally layers of soil were added until the container was completely filled. A first set of measurements was done with the source on the axis ($R=0$) at different heights $h=0.0, 0.7, 1.3, 2.15, 2.75$ and 3.45 cm; h denotes the ordinate of the lower plane of the source assembly, and so the ordinate of the radioactive material is $h+1$ mm. In a second set the source was placed in three planes $(h=0.0, 1.5$ and 3.4 cm) and in each plane at three radial coordinates $R=0$, 0.95 and 1.9 cm. In each case the uncertainty of both h and R was less than 1 mm. Two additional measurements were done on the axis of the container, one with the source placed on the bottom of the empty container, the other with the source placed on the top of the empty container.

In the measurements carried out with spectrometer 2 a 60 Co source of (20.3 +/−0.2) Bq (1 σ), prepared in house gravimetrically from a standard solution, was used. The source was measured in two positions inside of a container used in typical measurements with this system. The container has inner radius of 4 cm and inner height of 2.6 cm. In the first measurement the source was fixed on the center of the bottom of the container using adhesive band; in the second measurement the container was turned upside down, so that the source was on the inner side of the top of the container. With spectrometer 2 the spectra were acquired both in the suppressed mode and in the unsuppressed mode.

The spectra collected in all the measurements were analyzed and the count rates in the 1173 and 1332 keV peaks, as well as in the 2505 keV pure sum peak of the two photons, were obtained.

Both spectrometers were previously calibrated by combining experimental data with Monte Carlo simulation done using GESPECOR [\(Sima et al., 2001](#page--1-0)); the parameters of the GESPECOR models of the detectors developed in previous studies ([Suvaila](#page--1-0) [et al., 2012; Sima and Osvath, 2013\)](#page--1-0) were kept unchanged. In order to check the quality of the detector models and of the simulations, the experimental apparent efficiencies at the energies of the three peaks, evaluated by:

$$
\varepsilon^{app}(E; \overrightarrow{r}) = \frac{R(E; \overrightarrow{r})}{A \cdot I(E)}
$$
\n(1)

were compared with the values computed using GESPECOR. In Eq. (1) $\varepsilon^{app}(E; \overrightarrow{r})$ is the apparent efficiency [\(Sima and Arnold, 2000\)](#page--1-0) for the source located at \overrightarrow{r} , $R(E; \overrightarrow{r})$ the count rate in the peak of

energy E , A the activity and $I(E)$ the photon emission probability, with the convention that for the sum peak $I(2505)=1$. The experimental values of the ratio of the count rate in the 2505 keV peak to the count rate in the 1332 keV peak were also compared with the values obtained by GESPECOR simulations; this ratio is independent of the activity of the source, but depends on its position. In Fig. 1 the comparison of the apparent efficiency and of the ratio of the count rate in the two peaks is presented for the set of measurements carried out with spectrometer 1 with the source located on the axis of the container.

3. Results and discussion

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In order to test the quality of the assessment of the activity when the position of the source is known and when it is not known, the measurements of the point source placed in different positions inside the soil sample were analyzed using different methods.

3.1. Methods of activity assessment applied when the position of the source is known

The first method uses the peak count rate and the efficiency for the specific position of the source,

$$
A = \frac{R(E; \vec{\tau})}{\varepsilon^{app}(E; \vec{\tau}) \cdot I(E)}
$$
 (2)

The values of the apparent efficiency applied in Eq. (2) were computed using GESPECOR for the particular position of the source by the equation:

$$
\varepsilon^{app}(E; \overrightarrow{r}) = F_C(E; \overrightarrow{r}) \cdot \varepsilon(E; \overrightarrow{r})
$$
\n(3)

In this equation $F_C(E; \vec{r})$ is the coincidence summing correction factor and $\varepsilon(E; \vec{\tau})$ the ideal efficiency.

The second method is the sum peak method [\(Debertin and](#page--1-0) [Helmer, 1988; Vidmar et al., 2009](#page--1-0)). It is based on the count rate equations for the normal peaks at 1173 keV and 1332 keV (both affected by coincidence summing effects) and for the pure sum

> ε**app(1332) exp** ε**app(1332) MC N(2505)/N(1332) exp**

Fig. 1. Comparison between experimental and Monte Carlo values of the apparent efficiency at 1332 keV and of the ratio of the count rate in the 2505 keV and 1332 keV peaks for the source located on the axis of the container at several distances (h) from the bottom.

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