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A New Method for Gamma-efficiency Calibration of Voluminal Samples in Cylindrical Geometry

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ABSTRACT

In this paper, a new method for the gamma-efficiency calibration of voluminal sea-sediment samples in cylindrical geometry is proposed. The efficiency function can be written as the product of a water-matrix-sample efficiency in the same geometry and a correction factor, improved with regard to those used to date, dependent on the energy and the apparent density. The method has advantages such as its simplicity and speed and has been validated successfully in the energy range of 100–1500 keV. © 1997 Elsevier Science Ltd

INTRODUCTION

One of the main problems in gamma-ray spectrometry is the accurate determination of the photopeak-efficiency curve for a given sample matrix. This problem is especially important in voluminal samples, where self-absorption effects are significant for a wide range of energies (Khiem *et al.*, 1995).

The most accurate method for calculating the gamma efficiency of uncharacterized voluminal samples is to use the same composition and density samples in the same geometry, spiked with the same radionuclide for measurement (method of the standard addition). However, this is not always possible, above all when we want to measure environmental samples.

In our case, we have been working with environmental voluminal sea-sediment samples. The compositions of the sediments were not very different, but this is not true for the apparent density (density of the sediment in the measuring geometry, after being dried, powdered and sieved). This implies a different efficiency for every sample, as self-absorption depends on the apparent density. The traditional approach to quantify self-absorption, and avoiding calculation of a different efficiency curve for every sample, consists of using different density samples inside the experimental range of densities. This allows a curve that is dependent on the density and energy to be obtained. However, preparing samples of several densities, checking the homogeneity, counting them in a gamma-ray-spectrometry system and calculating the efficiencies is time-consuming. In this paper, a fast and simple method for gamma-efficiency calibration is proposed. It is based on The Generalized Transmission Method [Bolívar *et al.* (1994), generalization of Cutshall *et al.* (1983) and Kitto (1991)], which allows the efficiency function of a sea-sediment matrix sample to be written as the product of a standard sediment-matrix-sample reference efficiency in the same geometry and a correction factor, *f*, dependent on the apparent density and the energy. Here, it will be shown that by using a slightly different correction factor, it is possible to utilize a standard water-matrix sample for calculating the reference efficiency without losing any accuracy. Consequently, two important successes have been achieved: improvement of the correction factor and utilization of a water matrix sample for the reference efficiency, which has advantages such as easy homogenization and preparation, and being less time-consuming.

THEORETICAL FUNDAMENTALS

The use of radioactive sea-sediment matrix samples ranging the experimental density; it is a very difficult and time-consuming task. The first approach to avoid this, was the Gamma ray Transmission Method developed by Cutshall *et al.* (1983) who wrote the self-absorption equation, depending on the energy, as a function of the transmission coefficient, *T*:

$$\mathbf{T(E)} = \frac{\mathbf{I}}{\mathbf{I_0}} = \mathbf{e}^{-\mu\rho x}, \qquad (1)$$

where I/I_0 is the ratio, for every energy, between the intensities detected when a radioactive source is placed above the container with sea-sediments and above the empty container. Consequently, Cutshall writes the self-absorption factor as

$$\mathbf{F(E)} = \frac{\mathbf{T(E)} - \mathbf{1}}{\ln(\mathbf{T(E)})}. \tag{2}$$

We have supposed that efficiency can be expressed as a product of a factor dependent on the self-absorption and another one dependent on the geometry. If we divide the efficiency of the sea-sediment matrix (ε_s) by the efficiency of the water matrix (ε_W) for samples in the same geometry, we obtain a factor only dependent on the ratio between self-absorption factors:

$$\varepsilon_S = \mathbf{f} \cdot \varepsilon_W, \tag{3}$$

so that

$$\frac{\varepsilon_S}{\varepsilon_W} = f = \frac{\frac{T_s(E)-1}{\ln(T_s(E))}}{\frac{T_w(E)-1}{\ln(T_w(E))}}, \tag{4}$$

where T_s is the ratio between the intensities counted in the detector, when a point source is placed above the container with sea-sediments, and the empty container, and in a similar way, T_w is the ratio between the intensities counted when we place the same point source above the container with water and the empty container.

Until now, the correction factors employed (Bolívar *et al.*, 1994; Kitto, 1991) were slightly different, as they standardize the transmissions to water and a reference sea-sediment, respectively, directly using Eq. (2) instead of Eq. (4), with I_0 referred to a water matrix or a sea-sediment matrix.

We will show here that it is possible to use a water matrix as a reference sample with the correction factor that we propose. Eq. (4) can be applied directly for every sea-sediment matrix sample and every energy, calculating f experimentally. However, this is an impractical and time-consuming method. Therefore, we have decided to find a functional dependence of our correction factor on the apparent sea-sediment density and energy, which is possible as there are no important differences in our sediment compositions.

EXPERIMENTAL PROCEDURES

The measurements were performed with an integral preamplifier-Canberra GC2020-7500SL p-type coaxial HPGe detector system. The useful energy range of this detector was from 50 keV to more than 10 MeV. The Peak/Compton ratio was 46 for the 1333-keV ^{60}Co photon. The detector had a diameter of 50·5 mm and a length of 46·5 mm. The relative efficiency for the

7·62 cm×7·62 cm INa (TI) detector was 20%, and resolutions at 122 and 1332 keV of 1·1 and 2 keV, respectively, were obtained. The spectrometer was shielded by a 10-cm cubic geometry box of low-background lead (smelted more than 500 years ago), with an inner 1-mm Cd + 1-mm Cu layer.

The detector was connected to a standard set-up: Canberra Model 2020 Amplifier and a PC-based 8 K multichannel analyzer.

The spectrometer was situated in a laboratory where temperature and humidity were maintained at constant levels, using the Airwell SLM 7/9 air-conditioning system. The lab temperature and humidity were (22±2)°C and (60±5)% respectively, and were verified with a maximum–minimum thermometer and a psychrometer.

Environmental samples of sea-sediments were collected from zones at the Bay of Cádiz where there is thought to be a strong anthropogenic influence. The sediments were dried, powdered, and sieved (less than 0·5 mm) in order to reach homogeneity, the apparent densities were determined, and the samples were stored in polyethylene cylindrical containers (64-mm inner diameter), filled to a height of 5·5 cm. Spectrometric measurements were carried out with the bottom of the cylindrical containers at a distance of 116 mm from the window detector to avoid any summing-corrections (Quintana and Fernández (1995)). Water and sea-sediment matrix samples, spiked with ^{152}Eu, were prepared. The water matrix sample was used to calculate ε_W and the sea-sediment matrix one in order to ratify the method.

RESULTS

The water matrix sample was used to calculate the efficiency ε_W. It is a very well-known fact that the efficiency curve in the range 200–2500 keV is well described by a second-order polynomial in a log–log display (Bolívar *et al.*, 1994). Therefore, the experimental values were fitted to:

$$\ln(\varepsilon_w) = \sum_{i=0}^{i=2} \mathbf{a_i} \ln \left(\frac{\mathbf{E}_\gamma}{\mathbf{E}_0}\right)^{i} . \tag{5}$$

We tried several other curves in the real display. The best one was:

$$\varepsilon_w A + \frac{B}{(E_\gamma/E_0)} + \frac{C}{(E_\gamma/E_0)^2}, \tag{6}$$

and this was found to be even better than Eq. (5) because it describes the low energy range better (less deviations with regard to the experimental values); both curves have very similar deviations at high energies. Thus, the water matrix sample efficiency can be expressed by Eq. (6) with: $A = 3{\cdot}46\ 10^{-5}$,

$B=0{\cdot}76879$, and $C=-42{\cdot}73982$. The factor $E_0=1\,\text{keV}$ is introduced by dimensional reasons, E_γ is given in keV and ε_w is less than 1. The curve correlation is 0·9997. Higher precision curves need more parameters (Jäeckel *et al.*, 1987; Sánchez-Reyes *et al.*, 1987) and do not imply an improvement for us (environmental samples where the statistical error clearly dominates the total uncertainty of the measured activity).

Then, the correction factors given by Eq. (4) are calculated, using a ^{152}Eu point source. The results are presented in Table 1.

Finally, we look for a functional dependence on f of energy and apparent density.

The function f has been chosen according to the physical meaning of the correction factor. This can be expressed as:

$$f=\frac{\dfrac{1-e^{-\mu_s\rho \mathrm{l}}}{\mu_s\rho l}}{\dfrac{1-e^{\mu_w\rho_w \mathrm{l}}}{\mu_w\rho_w l}}, \tag{7}$$

an expression derived from Eq. (1) and Eq. (4), and therefore, in the approximation for small x, $(1-e^{-x})/x=e^{-x/2}$, the cited factor can be written as:

$$f=e^{\mu_w\rho_w \mathrm{l}/2}e^{-\mu_s\rho \mathrm{l}/2} \tag{8}$$

where the dependence on the apparent density of the sediment, ρ, is clearly an exponential function $(Ae^{-B\rho})$, and the dependence on the energy occurs through the water and sediment attenuation coefficients, μ_w and μ_s, respectively. Thus, the correction factor can be expressed as:

TABLE 1

Correction Factors for Some Energies and Apparent Densities

E	ρ							
	1·7076	*1·6426*	*1·5928*	*1·3105*	*1·2778*	*1·2186*	*1·2112*	*1·1734*
121·78	0·7951	0·8093	0·8330	0·9078	0·9142	0·9316	0·9300	0·9353
244·7	0·8399	0·8543	0·8751	0·9380	0·9490	0·9546	0·9590	0·9671
344·28	0·8618	0·8760	0·8950	0·9497	0·9606	0·9678	0·9738	0·9770
443·97	0·8777	0·8838	0·9053	0·9477	0·9593	0·9719	0·9710	0·9911
778·92	0·8937	0·9069	0·9251	0·9602	0·9728	0·9770	0·9804	0·9802
867·39	0·9032	0·9165	0·9246	0·9609	0·9769	0·9810	0·9794	0·9782
964·06	0·9085	0·9190	0·9305	0·9648	0·9758	0·9825	0·9869	0·9882
1112·09	0·9176	0·9167	0·9395	0·9652	0·9789	0·9812	0·9891	0·9907
1299·15	0·9131	0·9248	0·9319	0·9566	0·9739	0·9874	0·9783	0·9929
1408·02	0·9224	0·9287	0·9419	0·9658	0·9801	0·9811	0·9845	0·9875

The first row shows densities (g cm^{-3}) from the chosen sediments inside the experimental range 1·10–1·71 g cm^{-3}, and the first column shows the energies (keV).

$$\mathbf{f} = \mathbf{A}\mathbf{e}^{-\mathbf{B}\rho} \tag{9}$$

where

$$A = d'e^{f(E)}, \tag{10}$$

$$B = f'(E). \tag{11}$$

The functions $f(E)$ and $f'(E)$ can be expressed as a second or first order polynomial in ln$E$ for the energy range (100–3000) keV, as the attenuation coefficient can be fitted in this way.

The correction factor is shown in Fig. 1 for some energies as a function of apparent density and was fitted to a curve given by Eq. (9), where A and B are parameters dependent on the energy; some of the values for these are shown in Table 2.

In order to validate the correction factor obtained inside the proposed model, we determine its value in two extreme cases: when the sea-sediment density goes to zero and when the energy is very high.

In the first case, and because of

$$\lim_{\rho\to\mathbf{0}} \frac{\mathbf{T}_{\mathrm{S}} - \mathbf{1}}{\ln(\mathbf{T}_{\mathrm{S}})} = \mathbf{1}, \tag{12}$$

the extrapolation of Eq. (4) to zero density in the sea-sediment implies:

$$\mathbf{f}(\rho = \mathbf{0}, \mathbf{E}) = \left(\frac{\mathbf{T}_{\mathrm{W}} - \mathbf{1}}{\ln(\mathbf{T}_{\mathrm{W}})}\right)^{-1}, \tag{13}$$

a value that can be determined experimentally. Such experimental values together with those derived from Eq. (9) in this hypothesis ($f = A$) are shown in Table 3, in which it can be seen that in the worst case (E = 121·78 keV), the deviation is no greater than 6%.

However, for high energies, self-absorption is negligible (f = 1), so in the proposed function, the parameters A and B must tend towards 1 and 0, respectively (see Table 2).

The results allow the supposition that the proposed correction factor function is valid in a larger range of densities and energies, not just in the experimental one.

From Fig. 1, it can be seen that the exponential curve can be approximated by a straight line. Both fits (exponential and linear) have similar correlations (0·997 for nearly all energies). However, the extrapolation to zero density is much better for the exponential fit (13% of maximum deviation for the straight line against 6% for the exponential fit).

The parameters A and B depend on the energy, as shown in Fig. 2. We have fitted them to the following expressions based on Eqs (10) and (11):

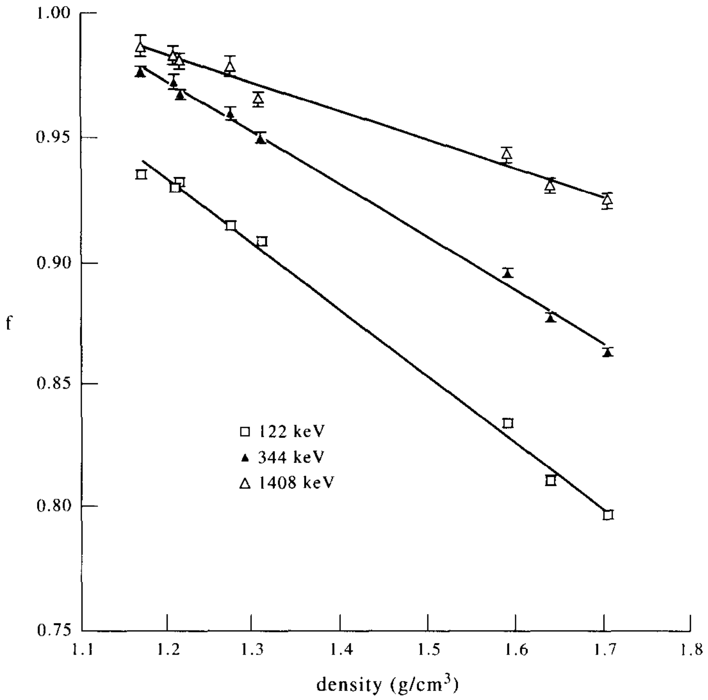

Fig. 1. Correction factor vs. apparent density, for some energies.

TABLE 2
Values of the Parameters *A* and *B* for Some Energies

Energy (keV)	A	B
121·78	1·360	0·313
244·7	1·319	0·262
344·28	1·291	0·235
443·97	1·261	0·213
778·92	1·205	0·172
867·39	1·183	0·156
964·06	1·190	0·157
1112·09	1·177	0·147
1299·15	1·170	0·145
1408·02	1·146	0·126

$$\mathbf{B} = \mathbf{a} + \mathbf{b} \cdot \ln \mathbf{E}, \tag{14}$$

$$\mathbf{A} = \mathbf{a}' \cdot \mathbf{e}^{\ln \mathbf{E} - \mathbf{b}')^2/\mathbf{c}'}, \tag{15}$$

where the constants *a*, *b*, *a′*, *b′*, and *c′* with the correlation coefficients are given in Table 4.

TABLE 3
Theoretical and Experimental Correction Factors Inside the Proposed Model in the Extrapolation for Zero Density

Energy (keV)	A	$\left(\frac{T_w(E)-1}{\ln(T_w(E))}\right)^{-1}$
121·78	1·360	1·449
244·7	1·319	1·346
344·28	1·291	1·309
443·97	1·261	1·279
778·92	1·205	1·209
867·39	1·183	1·203
964·06	1·190	1·187
1112·09	1·177	1·171
1299·15	1·170	1·158
1408·02	1·146	1·155

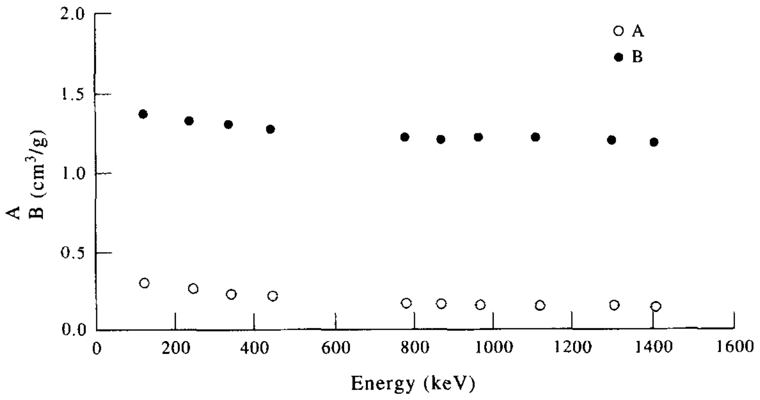

Fig. 2. Parameter A and B vs. energy.

TABLE 4
Constants and Correlation Coefficients After the Experimental Fit of Eqs (14) and (15)

Constants	*Values*	r
a′	1·471	0·994
b′	1·793	
c′	−120·842	
a	0·672	0·997
b	−0·075	

Thus, we propose as correction factor the function:

$$\mathbf{f} = \mathbf{a}' \cdot \mathbf{e}^{(\ln \mathbf{E} - \mathbf{b}')^2/\mathbf{c}'} \cdot \mathbf{e}^{-(\mathbf{a}+\mathbf{b}\ln \mathbf{E})\rho}, \qquad (16)$$

whose deviations with regard to the experimental values are below 2%.

The efficiency curve for our environmental sea-sediment samples in cylindrical geometry is a product of Eqs (6) and (16), and has been validated using a sea-sediment matrix sample spiked with ^{152}Eu (Table 5). The experimental efficiency of this sediment sample (ε_s) deviates by a maximum of 11% from the values obtained with the curve, which is inside the experimental uncertainty (±10% error for a confidence level of 95%).

From Table 5, it can be seen that ε_s is, for all energies, less than the values obtained with our curve ($f\varepsilon_w$). This is because our correction factor is overestimated as we have employed a model for the self-absorption factor (the Cutshall Model) which can be improved (Galloway, 1991). However, for our purposes and working with uncertainties of less than 10%, this model was good enough.

In our model, the correction factor is a ratio between the Cutshall self-absorption factors for sediment and water, respectively. Galloway showed that the real self-absorption is bigger than the Cutshall one and that the Cutshall approximation is better when we have low-thickness samples. It should also be mentioned that the photons detected from a highly attenuating sample come predominantly from the front face of the sample so that the 'effective sample' is both thinner and has its centre

TABLE 5

Validation of the Efficiency Curve[a]

Energy (keV)	ε_s	f	ε_w	$f\varepsilon_w$	$(\varepsilon_s - f\varepsilon_w) \times 100/\varepsilon_s$
121·78	0·00259 (7)	0·818 (16)	0·0035 (2)	0·0028 (2)	8%
244·7	0·00198 (6)	0·858 (17)	0·00246 (17)	0·00211 (17)	6%
344·28	0·00152 (4)	0·875 (17)	0·00191 (13)	0·00167 (13)	8%
443·97	0·00122 (5)	0·887 (18)	0·00155 (11)	0·00137 (11)	11%
778·92	0·00079 (2)	0·911 (18)	0·00095 (7)	0·00087 (7)	9%
867·39	0·00070 (2)	0·915 (18)	0·00086 (6)	0·00079 (6)	10%
964·06	0·00068 (2)	0·919 (18)	0·00079 (6)	0·00072 (6)	6%
1112·09	0·000588 (17)	0·924 (18)	0·00069 (5)	0·00064 (5)	8%
1299·15	0·000546 (2)	0·929 (19)	0·00061 (4)	0·00056 (4)	2%
1408·02	0·000505 (14)	0·931 (19)	0·00056 (4)	0·00052 (4)	3%

[a] The following are compared: efficiency of a sea-sediment matrix sample of apparent density 1·64 g cm^{-3}, ε_s, and the product of the correction factor, f, and the efficiency of a water matrix sample, ε_w, for every energy.

closer to the detector than a sample of only slightly attenuating material. Thus, the Cutshall self-absorption for sediment will be more similar to the real one than for water (underestimated) as the effective thickness for a gamma photon in the sediment is less. This led to an overestimated correction factor, which will be less important for high energies (more penetrating radiation).

CONCLUSIONS

A new fast and simple method for gamma-efficiency calibration of voluminal sea-sediment samples is proposed in this paper. The efficiency can be expressed as a factor dependent on the sample density and the gamma ray energy by the efficiency of a water matrix sample in the same geometry. This method has been validated successfully using a sea-sediment matrix sample spiked with ^{152}Eu, and as main goals, we can mention its speed and simplicity. The water matrix sample spiked with ^{152}Eu can be prepared very easily, and the experimental correction factor can also be easily calculated with a point source above the sample, and far enough to obey the normal incidence approximation of the photons through the sample (approximation employed in the self-absorption equation).

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