

A semi-empirical approach for determination of low-energy gamma-emitters in sediment samples with coaxial Ge-detectors

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Abstract

A simple method for the determination in sediment samples of low-energy gamma-emitters with coaxial Ge detectors and using a cylindrical sample geometry is outlined. This method allows an easy estimation of self-absorption effects by knowing the apparent densities and the composition of the investigated samples. As calibration matrixes solid samples enriched in known amounts of natural radionuclides emitting low-energy gamma rays were used. In this way, the difficult homogenisation steps needed when liquid spikes are added to a solid blank for manufacturing the calibration sample are avoided.

The method has been carefully checked and validated by applying it to sediment samples with known activities of some low-energy gamma-ray emitters. Additionally, these sediment measurements have allowed to evaluate the magnitude and importance of the self-absorption corrections.

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1. Introduction

Low-energy gamma emitters are used in environmental studies in a wide range of applications: for example, ²¹⁰Pb (46.5 keV) and ²⁴¹Am (59.5 keV) are widely determined in lacustrine and marine sediment layers for dating purposes (Appleby et al., 1991; Battarbee et al., 2002), while ²³⁴Th (63.3, 92.4 and 92.8 keV) is extensively used as a natural tracer to study biological mixing and particle scavenging processes in recent sediments (Fuller et al., 1999).

The main advantage of the determination of these low-energy gamma emitters in environmental samples by gamma-ray spectrometry is that its quantification can be achieved without the application of skilled and tedious sample preparation. However, the precise determination of the activity of each radionuclide by gamma-ray spectrometry requires the knowledge of the

full energy peak efficiency at each energy of interest for a given geometry. And, in the energy range below 150 keV, these full energy peak efficiencies are affected by the high self-absorption of the gamma rays emitted, which strictly depend on the energy of the gamma-ray considered as well as on the composition and apparent density of the analysed sample (Hasan et al., 2002; Jurado Vargas et al., 2002).

In this work, we propose a method to estimate for each sample the full energy peak efficiencies in the energy range 40–180 keV with a standard coaxial gamma-spectrometric system. This method, which is applied in our case to sediment samples in a cylindrical geometry, takes into account the self-absorption of the low-energy γ -rays, considering that the full energy peak efficiency, at a fixed geometry and energy, depends on the composition and on the density of the samples.

The proposed method is based in the determination for a calibration sample of the full energy peak energy efficiencies in the energy range indicated, and the application of corrections to take into account the

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different influence of the self-absorption effects in the calibration and investigated samples.

As calibration matrix was used a solid environmental sample highly enriched in known amounts of several low-energy gamma emitters, avoiding in this way the homogenisation processes needed when liquid spikes are added to a solid blank for manufacturing the calibration sample.

2. Material and methods

In this work, we have used different solid samples for calibrating or validating purposes: phosphate rocks and sediments. In these samples, the low-energy gamma emitters were measured by using the gamma spectrometric system described below.

^{210}Po and ^{238}U concentrations were also determined in most of the samples by alpha-spectrometry in order to validate through an alternative technique the activities of some radionuclides that are low-energy gamma emitters (^{234}Th and ^{210}Pb).

2.1. Gamma-ray spectrometric system

Gamma-ray spectrometry measurements were performed by using a Canberra Extended Range Ge detector (XtRa), with 38% of relative efficiency and FWHM of 0.95 keV at 122 keV. The detector works coupled to a conventional electronic chain, and is shielded with Fe 15 cm thick containing a certified ^{210}Pb activity lower than 10 Bq kg^{-1} .

Before the gamma-ray measurement, each sample was dried at 60°C , homogenised and grind. Then, the samples were introduced in polyethylene containers of cylindrical geometry (diameter 6.5 cm) and sealed. In each sediment sample measurement, the statistical uncertainties from the net counts under each photopeak of interest were kept below 5% by maintaining a minimum of 24 h counting time, while in the calibration matrix the same counting time led to uncertainties of 1%.

2.2. Alpha-particle spectrometry determination

For ^{210}Po and ^{238}U determination by alpha spectrometry, we have applied a well-established sequential solvent extraction method (Holm and Fukai, 1977) using ^{209}Po and ^{232}U as radiochemical recovery tracers. Recovery yields of 60–70% for Po and 60–80% for U were obtained.

The Po and U sources were measured by using an EG&Ortec supplied alpha spectrometry system equipped with ion-implanted silicon detectors, while the activity quantifications were performed by the application of the isotope dilution technique. Counting

times ranged from 2 to 7 days, being reached minimum detectable activities (MDA) lower than 1 mBq for ^{210}Po and ^{238}U .

The method used for ^{210}Po and ^{238}U determination in sediment and solid samples by alpha-particle spectrometry has been periodically validated through the successful participation of our laboratory in several inter-comparison exercises over the last 10 years. In addition, the goodness of the ^{238}U alpha-spectrometric determinations (phosphate rock and sediments) has been verified in each sample by the independent determination of this element by ICP- mass spectrometry.

2.3. Elemental composition analysis

The elemental composition of the different samples (phosphate and sediment samples) was determined by applying the TTPIXE technique in the 3 MV Van de Graaff accelerator of the “Instituto Tecnológico e Nuclear” (ITN), Sacavém (Portugal) and by following the method described in Martin et al. (1996, 1999).

3. Full energy peak efficiency determinations in the calibration sample

The full energy peak efficiency calibration of the gamma-ray system in the energy interval 40–180 keV was carried out using a phosphate rock sample with a high and known content in ^{238}U and daughters. In this way, we were able to use some gamma emissions of these daughters in the low-energy range for efficiency determinations. This calibration sample has an apparent density of 1.60 g cm^{-3} , and their ^{238}U , ^{226}Ra and ^{210}Po specific activities ($^{238}\text{U} = 1187 \pm 52 \text{ Bq kg}^{-1}$; $^{226}\text{Ra} = 1152 \pm 54 \text{ Bq kg}^{-1}$; $^{210}\text{Po} = 1103 \pm 26 \text{ Bq kg}^{-1}$) were determined as the average values of the results obtained in the analysis of several aliquots by alpha-spectrometry. Details about its elemental composition are given in Table 1.

Table 1

Elemental composition (mass fraction in percentage) of the phosphate rock used in this work as calibration sample

Calibration sample: phosphate rock, $\rho = 1.60 \text{ g cm}^{-3}$			
Ca (%)	38.25	Al (%)	0.63
O (%)	36.60	S (%)	0.13
P (%)	17.71	Zn (%)	0.067
F (%)	3.61	Ti (%)	0.026
Si (%)	2.18	Cu (%)	0.006
Fe (%)	0.83	Pb (%)	0.003

This elemental composition was determined by the application of the TTPIXE technique and following the method proposed in Martin et al. (1996).

The full energy peak efficiency calibration was performed at different sample heights (i.e. masses), because the amount of dry material in the sediments that need to be analysed may vary from one sample to another, with the result that it is not always possible to fix a constant sample height in the cylindrical containers. For this reason we have determined, by taking spectra of the calibration sample with different heights (from 1 to 5 cm with intervals of 0.5 cm), the experimental values of the full energy peak efficiency, ε_C , at different energies and heights. For these calculations, the gamma-emission probabilities P_γ of the gamma-rays considered were taken from tabulated data (Kocher, 1981), while secular equilibrium between ^{238}U and ^{234}Th and between ^{210}Pb and ^{210}Po was considered (the sample is more than 2 years old). In Table 2 the full energy peak efficiencies at different heights for the different emissions considered in the energy range of interest (uncertainties are expressed at one sigma level) are compiled.

In order to express empirically at each of the considered energies the dependence of the full energy efficiency peak, ε_C , on sample height, h , or mass, m_c , (h and m_c are related variables because $m_c = \pi r^2 \delta_c h = kh$, where m_c (g) is the mass of calibration sample, δ_c (g cm^{-3}) its apparent density and r (cm) the radius of cylindrical container), we have fitted at each energy (weighted least-square fitting) the values of ε_C to the function:

$$\ln \varepsilon_C = A + Bm_c. \quad (1)$$

This exponential relation between ε_C and m_C was elected because the predicted values showed a good agreement with the experimental ones (see values of the correlation coefficients, Table 3). We have tested other polynomial functions, adding to Eq. (1) quadratic and cubic terms, but the parameters obtained (order two and higher) were not statistically significant at 5% level.

The values of the parameters A and B obtained from these fittings are also shown in Table 3. As can be seen from this table, the parameter A is nearly constant (with a mean value of -1.99 ± 0.15), while B depends on

energy. This different behaviour can be understood if we take into account the following relationship derived from Eq. (1):

$$\varepsilon_C = e^A e^{Bm_c} = \varepsilon_{oc} e^{Bm_c}. \quad (2)$$

As can be deduced from Eq. (2), $\varepsilon_{oc} = e^A$ represents the full energy peak efficiency of a hypothetical sample with null thickness, the same diameter than our container and sited at the same distance bottom container-detector window. In these hypothetical conditions (no self-absorption effects), it is well known the constancy of the full energy peak efficiencies at low energies in extended range Ge detectors. On the other hand, the dependence of parameter B on energy is also not surprising because B means the relative variation or change of ε_C per unit of mass(height) increase.

Once the relations $\varepsilon_C - m_C$ were determined at each of the selected energies, we were able to obtain the dependence of the parameters A and B on the energy. The objective was to find a general function giving the full energy peak efficiency, at different heights and energies, valid for the energy range 40–180 keV and for any sample height in the interval 1–5 cm. The fitting of the values of A and B on the energy gave us the

Table 3
Parameters obtained from the fittings of ε_C against sample mass for different energies

E (keV)	$-A$	$-B(10^{-3} \text{g}^{-1})$	R^2
46.5	2.24 ± 0.07	6.2 ± 0.4	0.97
63	1.97 ± 0.08	5.1 ± 0.4	0.983
67	1.88 ± 0.09	5.0 ± 0.5	0.963
77	2.02 ± 0.04	4.0 ± 0.2	0.98
93	1.90 ± 0.03	4.5 ± 0.2	0.991
144	1.75 ± 0.10	3.9 ± 0.6	0.95
163	2.04 ± 0.14	4.5 ± 0.8	0.973
186	2.12 ± 0.04	4.0 ± 0.2	0.987

Uncertainties are expressed at one sigma level. The correlation coefficients associated to the fittings are also shown.

Table 2

Experimental full energy peak efficiency values (%), at different heights, for the gamma-emission energies considered from the calibration sample

E (keV)	Radionuclide P_γ (%)	1 cm	1.5 cm	2 cm	2.5 cm	3 cm	3.5 cm	4 cm	4.5 cm	5 cm
46.5	^{210}Pb 4.05(3)	8.8 (3)	6.6 (2)	5.2 (2)	4.4 (2)	3.7 (1)	3.3 (1)	2.8 (1)	2.5 (1)	2.3 (1)
63	^{230}Th 3.8(3)	11.3 (10)	9.0 (8)	7.5 (7)	6.8 (6)	7.0 (6)	5.1 (5)	4.5 (4)	4.1 (4)	3.7 (3)
67	^{230}Th 1.64(11)	12.1 (14)	11.0 (12)	8.0 (9)	7.9 (7)	6.2 (7)	6.6 (7)	5.6 (6)	4.6 (5)	4.1 (4)
77	^{214}Pb 10.5(4) + ^{212}Pb 18.0(7)	11.5 (6)	9.4 (5)	8.9 (5)	7.2 (4)	7.1 (4)	6.5 (3)	5.7 (3)	5.2 (3)	4.7 (2)
93	^{230}Th 5.41(28)	12.6 (7)	10.6 (7)	9.1 (6)	8.2 (5)	7.1 (5)	6.5 (4)	5.7 (3)	5.3 (3)	4.7 (2)
144	^{235}U 10.5(8)	14 (2)	13 (2)	12 (1)	10.2 (12)	8.1 (10)	9.0 (11)	7.2 (8)	7.3 (8)	6.3 (7)
163	^{235}U 4.7(4)	10 (2)	9.5 (16)	8 (1)	7.1 (10)	5.8 (10)	5.8 (10)	5.5 (8)	4.1 (7)	3.9 (6)
186	^{226}Ra 3.28(3) + ^{235}U 54(1)	9.7 (5)	8.6 (4)	8.4 (4)	6.9 (3)	6.2 (3)	5.7 (3)	5.0 (2)	4.7 (2)	4.2 (2)

Uncertainties are expressed at one sigma level.

following results:

$$A = -1.99 \pm 0.15, \quad (3)$$

$$\text{Ln}(-B \times 10^3) = (2.7 \pm 0.4) - (0.27 \pm 0.09) \text{Ln } E_\gamma. \quad (4)$$

The addition of quadratic and cubic terms in the function expressing the dependence of parameter B with the energy does not produce any improvements in the fitting, being the parameters associated to the added terms not significant at 5% level. The uncertainties predicted by Eq. (4) for B , considering the covariance terms, are in the 2–4% range.

Finally, by replacing parameters A and B (Eqs. (3) and (4)) in Eq. (1) and relating the mass of the calibration sample (m_c) with its density ($\delta_c = 1.60 \text{ g cm}^{-3}$), the radius of the cylindrical container ($r = 3.25 \text{ cm}$) and its sample height (h), the following general relation (valid for any sample with the same apparent density and composition than the calibration one) is obtained:

$$\text{Ln } \varepsilon_C = (-1.99 \pm 0.15) - (0.8 \pm 0.3) E_\gamma^{-0.27 \pm 0.09} h, \quad (5)$$

where E_γ is expressed in keV and h in cm.

The uncertainties in ε_C predicted by this equation has been evaluated as lower than 9% in all the energy range covered, taking into account that the associated parameters are not independent quantities.

4. Full energy peak efficiency determinations in sediment samples

Once the general full energy peak efficiency function has been obtained for the calibration sample, the following step is to determine the full energy peak efficiency for a sample with different apparent density and composition than the calibration one. For this purpose it is necessary to introduce self-absorption corrections.

The evaluation of these corrections have been carried out using a semi-empirical method developed previously for ^{210}Pb determinations by gamma spectrometry (San Miguel et al., 2002) and that can be highlighted for its simplicity and goodness of the provided results. Through this approach, based on the fundamentals of the transmission method, it is concluded that the self-absorption correction factors can be simply determined by knowing the apparent densities and the composition of the samples analysed (and of the calibration sample). Indeed, to obtain the full energy peak efficiency (at a given energy, E_γ) for a sediment sample, ε , we have to correct the value of ε_C at this energy by a factor f , that, as detailed in San Miguel et al. (2002), can be written as

$$f = \left(\frac{1 - e^{-\mu\rho h}}{\mu\rho h} \right) \left(\frac{\mu_c \rho_c h}{1 - e^{-\mu_c \rho_c h}} \right). \quad (6)$$

In Eq. (6), ρ and ρ_c are the apparent densities of the sediment and calibration samples respectively, μ and μ_c are their corresponding mass attenuation coefficients at the selected energy, and h is the height of the sample.

The sample mass attenuation coefficients depend on their elemental composition. In fact, they can be determined at each selected energy by knowing the mass attenuation coefficient and the mass fraction of each component present in the sample through the application of the well known Bragg's formula. The elemental mass attenuation coefficients can be determined at the energies of interest by interpolation from tabulated data (Hubbell and Seltzer, 1995).

5. Validation exercise

The proposed method has been validated through its application to sediment samples of the Huelva estuary (South-West of Spain). These samples are characterised by the following facts: (a) they are enriched in radionuclides from uranium series due to anthropogenic releases affecting the area (Bolivar et al., 2000), (b) their elemental composition are known (see Table 4), because it was determined by applying the TPIXE technique (Martin et al., 1996), (c) their ^{210}Po and ^{238}U concentrations have been determined by α spectrometry, and (d) the radionuclides ^{210}Pb and ^{238}U are in the samples in secular equilibrium with their corresponding daughters ^{210}Po and ^{234}Th because they are sediments more than 2 years old.

In Table 5, ^{210}Pb and ^{234}Th activities determined by gamma spectrometry are reported for sediment samples. These determinations have been carried out: (a) by considering the efficiency of the calibration sample and (b) by considering the efficiency corrected using the method proposed in this paper. The objective was to analyse the importance of the self-absorption corrections in order to measure accurate ^{210}Pb and ^{234}Th activities. In fact, the magnitude of these corrections are highlighted in Table 5, by showing the relative differences (Δ) between corrected (A_{corr}) and uncorrected (A_{un}) values. These relative differences are in the range 70–90% for ^{210}Pb (46.5 keV), although they are lower at higher energies indicating the decrease in the magnitude of the self-absorption effects when the energy increases. ^{210}Po and ^{238}U activities determined by alpha-particle spectrometry are also shown in this table for validating purposes.

In order to accomplish fully the validation, we have performed the linear weighted fitting of the gamma data, corrected by self-absorption effects, against the corresponding alpha data at the three gamma-energies analysed. The following results were obtained

Table 4
Major and trace elements (mass fraction in percentage) in the sediments S1–S9 used in the validation test

Sample	Si	Al	Ti	K	Ca	Fe	P	Cu	Zn	Pb	As	μ (cm ² g ⁻¹)	ρ (g cm ⁻³)
S1	17.6	8.5	0.8	1.8	1.3	5.5	0.7	0.11	0.12	0.06	0.03	0.480	0.84
S2	17.5	8.6	0.7	1.8	1.1	5.8	0.7	0.15	0.14	0.09	0.04	0.490	0.75
S3	14.3	7.7	0.8	1.4	0.6	6.5	1.8	0.28	0.23	0.10	0.09	0.505	0.63
S4	11.6	7.4	0.5	1.2	0.9	5.2	2.4	0.22	0.39	0.12	0.13	0.472	0.69
S5	15.8	8.7	0.6	1.6	0.5	6.1	1.7	0.30	0.29	0.15	0.13	0.508	0.65
S6	15.0	7.7	0.6	1.4	0.6	7.1	2.2	0.29	0.24	0.25	0.14	0.535	0.53
S7	13.6	7.0	0.6	1.2	0.6	10.0	2.9	0.29	0.21	0.34	0.24	0.610	0.54
S8	12.5	5.7	0.6	0.8	0.6	10.5	3.0	0.33	0.27	0.71	0.26	0.655	0.56
S9	16.2	8.2	0.8	1.7	0.5	8.7	1.4	0.26	0.18	0.66	0.19	0.620	0.70

For each sediment sample the mass attenuation coefficient at 46.5 keV (²¹⁰Pb) determined through the Bragg's formula and the apparent density are also shown.

Table 5
Activity concentrations (Bq kg⁻¹) in sediment samples determined by gamma spectrometry using the efficiency values obtained directly with the calibration sample (A_{un}), and with the corrected efficiencies values based on the method proposed in this work (A_{corr})

Sample	²¹⁰ Pb (46.5 keV)			²¹⁰ Po	²³⁴ Th (63 keV)			²³⁴ Th (93 keV)			²³⁸ U
	A_{un}	A_{corr}	Δ (%)		A_{un}	A_{corr}	Δ (%)	A_{un}	A_{corr}	Δ (%)	
S1	550 ± 24	314 ± 14	75	353 ± 23	438 ± 19	317 ± 14	38	365 ± 15	306 ± 13	19	319 ± 22
S2	334 ± 15	184 ± 8	81	217 ± 13	272 ± 13	193 ± 9	41	238 ± 11	197 ± 9	21	191 ± 8
S3	849 ± 34	450 ± 18	89	525 ± 21	1007 ± 42	694 ± 29	45	819 ± 34	661 ± 27	24	781 ± 20
S4	1046 ± 42	559 ± 23	87	586 ± 16	1335 ± 55	929 ± 38	44	1125 ± 46	918 ± 38	23	1023 ± 29
S5	525 ± 21	281 ± 11	87	281 ± 9	586 ± 25	406 ± 17	44	480 ± 20	389 ± 16	23	483 ± 17
S6	N.M.	N.M.		N.M.	970 ± 41	654 ± 28	48	792 ± 33	631 ± 26	26	650 ± 51
S7	1081 ± 41	579 ± 22	87	553 ± 25	936 ± 39	642 ± 27	46	768 ± 32	617 ± 26	24	667 ± 22
S8	1171 ± 43	648 ± 24	81	649 ± 29	913 ± 38	637 ± 26	43	744 ± 31	606 ± 25	23	652 ± 33
S9	687 ± 28	404 ± 17	70	408 ± 22	N.M.	N.M.		N.M.	N.M.		N.M.

The relative differences between uncorrected and corrected activities determined by gamma spectrometry (Δ) as well as the ²¹⁰Po and ²³⁸U activities (Bq kg⁻¹) measured by alpha spectrometry in these sediment samples are also shown. Uncertainties are expressed at one sigma level.

(uncertainties expressed at 1 σ level):

$$\begin{aligned}
 {}^{210}\text{Pb}_\gamma &= (-28 \pm 24) + (1.01 \pm 0.07){}^{210}\text{Po}_\alpha, & R^2 &= 0.964, \\
 {}^{234}\text{Th}_{63} &= (21 \pm 19) + (0.90 \pm 0.04){}^{238}\text{U}_\alpha, & R^2 &= 0.982, \\
 {}^{234}\text{Th}_{93} &= (31 \pm 19) + (0.90 \pm 0.05){}^{238}\text{U}_\alpha, & R^2 &= 0.981.
 \end{aligned}
 \tag{7}$$

By other hand, the t -distribution at 95% confidence level for six degrees of freedom (8 data) is 2.45, being the intercept and the slope at this level of confidence, -28 ± 58 and 1.01 ± 0.17 for ²¹⁰Pb; 21 ± 47 and 0.90 ± 0.11 for ²³⁴Th (63 keV) and 31 ± 48 and 0.90 ± 0.11 for ²³⁴Th (93 keV) respectively.

As can be seen, the values of the intercept and the slope are not significantly different from 0 and 1 respectively at 95% level of confidence indicating that there are not systematic differences between the determinations carried out by the two methods.

Finally, it is interesting to remark that although in environmental samples the peak of 92.4 and 92.8 keV of ²³⁴Th are included in a complex multiplet consisting of K–X-rays and γ photopeaks from members of the three natural decay series (Morel et al., 2002; García-Talavera, 2003), in the case treated in this work (calibration and environmental samples with a clear enhancement of ²³⁸U in respect to ²³²Th) the activities obtained from the photopeak at 93 keV agrees very well with those obtained from the photopeak at 63 keV. From the Uranium series, only an X-ray of ^{234m}Pa (94.66 keV, 0.015%) can have a very small influence in this multiplet in the samples treated in this work. Nevertheless, the consideration of this multiplet in environmental samples with normal U/Th mass ratios is not recommended, because the interferences provoked in the ²³⁴Th multiplet can distort the obtained results if they are not taken into account.

6. Conclusions

In this work a simple method for the direct analysis of low-energy gamma emitters in sediment samples with coaxial Ge detectors has been described. The correction factors that need to be applied to account for the differences in self-absorption of the low-energy gamma rays between the calibration and the sediment samples are determined knowing the composition (the mass attenuation coefficient) and the apparent density of each sample.

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