

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/226501618>

Correction by self-attenuation in gamma-ray spectrometry for environmental samples

Article in *Journal of Radioanalytical and Nuclear Chemistry* · November 2006

DOI: 10.1007/s10967-006-0354-8

CITATIONS

26

READS

289

5 authors, including:



Luis Santiago Quindos Poncela

Universidad de Cantabria

91 PUBLICATIONS 1,123 CITATIONS

SEE PROFILE



Carlos Sainz

Universidad de Cantabria

73 PUBLICATIONS 653 CITATIONS

SEE PROFILE



Ismael Fuente

Universidad de Cantabria

33 PUBLICATIONS 199 CITATIONS

SEE PROFILE



Jorge Nicolas

3 PUBLICATIONS 90 CITATIONS

SEE PROFILE

Some of the authors of this publication are also working on these related projects:



RADON CERO-Optimization of constructive solutions against radon gas immission in buildings and development of solution design methodologies [View project](#)



PERFO-RA. Development and design of microperforation techniques as a solution for the protection of the health of users against radon immission in inhabited spaces. [View project](#)

Correction by self-attenuation in gamma-ray spectrometry for environmental samples

L. S. Quindós,¹ C. Sainz,^{1*} I. Fuente,¹ J. Nicolás,¹ L. Quindós,¹ J. Arceche²

¹ Department of Medical Physics, Faculty of Medicine, University of Cantabria, c/Herrera Oria s/n, 39011, Santander, Spain

² Spanish National Institute of Meteorology, Santander, Spain

(Received January 20, 2006)

Analysis of environmental samples by gamma-ray spectrometry needs to include a correction factor by self-attenuation, preferably when working on gamma-radiation with energies below 200 keV. This effect depends on soil characteristics like chemical composition and density, the latter being the most important one. Nevertheless, this fact is not included in many papers concerning the measurement of gamma-emitters. In this note we analyze the process for a good and practical calibration of a gamma-ray system showing the correction due to self-attenuation for the main radionuclides as a function of the density of calibrated samples.

Introduction

Some radioactive elements present in air, water or soils can be used as a valuable tool to understand underlying properties of the environment in which they are present. Atmospheric transport and deposition of air pollution, for example, can be modeled and assessed using radiotracers such as ²¹⁰Pb and ²²²Rn.¹ Another radioactive isotope like ¹³⁷Cs is frequently used for analysing the large-scale and long-term consequences of natural and man-made changes in surface and ground water. Also the evaluation of this radionuclide concentration in soils is very useful to study sedimentation and transport phenomena in several places.²

There are three main sources of radionuclides in environmental samples: naturally occurring sources, technologically-enhanced naturally occurring ones and man-made sources. In all cases, the main problem in the preparation of a radiological characterization protocol of environmental samples concerns the selection of the measurement method. Besides the abovementioned use of radionuclides, the radiological effects derived from radioactivity in environmental samples must be taken into account. For instance the usual presence of uranium in soil samples can lead to internal irradiation of the lung by the alpha-particles emitted from the short-lived decay products of the radon gas (²²²Rn), as well as external irradiation of the body by gamma-rays emitted by the mixture of radionuclides present in the soil.

For the development of laboratory measurements there is a wide variety of well established techniques. Gamma-spectrometry is probably the most common technique used in this field. This technique provides a non destructive method of multielemental analysis and can be used for many purposes such as to assess the mobility and transfer of natural radionuclides in

ecosystems,³ to know the erosion and sedimentation processes by measuring ¹³⁷Cs and ²¹⁰Pb in soil profiles^{4,5} or to evaluate ²²⁶Ra, ²³²Th and ⁴⁰K concentrations in building materials in order to avoid their use as commercial products.⁶

The accurate determination of the photopeak efficiency curve for a given sample matrix represents the most important challenge in gamma-ray spectrometry. This difficulty is specially relevant in soil samples, on which self-absorption effects can be significant for a wide range of gamma-ray energies. Few data are available on studies concerning the correction by self-absorption. It is a general practice to reduce the extent of self-attenuation effects by keeping the sample size limited and focusing on gamma-radiation with energies above 200 keV. However, the greatest influence of self-absorption on efficiency values is always observed below that energy. For the general purpose of gamma-spectrometry, several mathematical or semi empirical methods have been proposed for correcting the attenuation mentioned above.⁷ Also transmission methods for gamma-efficiency determination in soil samples has been proposed.⁸ However, these procedures present some practical difficulties related to the availability and high cost of calibrated sources. The aim of this work is to show first a standard calibration procedure using this kind of sources and second to propose a practical and inexpensive method to use in laboratories in order to consider the influence of self-attenuation in the evaluation of the concentration of each radionuclide.

Experimental

Material

The application of gamma-ray spectrometry for natural gamma-emitting radionuclide analysis is well described in the literature.⁹ In our laboratory,

* E-mail: sainzc@unican.es

measurements of radionuclide activities in environmental samples are undertaken by using a high resolution, low background, hyper pure coaxial gamma-ray Canberra 7229-3020 detector coupled to a Canberra amplifier and multichannel analyser. The detector has a relative efficiency of 20% and a resolution of 1.86 keV, both referred to 1332 keV photons and is surrounded with a lead shielding in a cubic volume with 600 mm edge and 100 mm thick and internally a 1.5 mm thick copper foil to reduce the background counting rate. In all cases, in order to reach radioactive equilibrium between ^{226}Ra , ^{222}Rn and its progeny, the 250 ml PVC cylindrical containers are kept sealed for 1 month before measurement.

^{226}Ra , ^{210}Pb , ^{137}Cs , ^{232}Th and ^{40}K activity concentrations are usually measured in environmental samples at our laboratory by gamma-ray spectrometry. For counting time around 30,000 seconds, the analytical uncertainty of the measurements is approximately $\pm 10\%$. Considering the appropriate corrections for laboratory background, the activity of ^{226}Ra is commonly determined from the 609.3 keV line of ^{214}Bi . ^{40}K activity is calculated from the 1460.8 keV photopeak; ^{232}Th is estimated from the 911.2 keV photopeak of ^{228}Ac , ^{210}Pb activity is determined from the 46.5 keV photopeak and ^{137}Cs is determined from the 661.6 keV line. Of course, for a good quality assurance of the measurements, intercomparison exercises are periodically developed.¹⁰

Method

Five standard samples with apparent densities ranging from 750 to 1900 $\text{kg}\cdot\text{m}^{-3}$ were used to determine photopeak efficiencies by using ten gamma ray energies coming from the present radionuclides (^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{203}Hg , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y and ^{60}Co). These samples (manufactured by AEA Technology QSA GmbH, Germany and calibrated by the Spanish calibration laboratory, CIEMAT), were elaborated from a certified solution containing the previous radioactive isotopes with a number of gamma-lines enough to cover an energy range from 45 to 1460 keV. Each sample was measured during 30,000 seconds, and the photopeak efficiency was determined as a function of the gamma-emission energy, E_γ . These efficiencies ε were fitted to the energy values:

$$\ln \varepsilon = \sum_{i=0}^4 a_i \cdot \left(\ln \frac{E_\gamma}{E_0}\right)^i \quad (1)$$

where a_i is the fitting coefficient, E_γ is given in MeV and $E_0 = 1$ MeV is included to ensure the homogeneity of the equation.

By focussing attention on the energies emitted from the radionuclides of interest ^{210}Pb , ^{214}Bi , ^{137}Cs , ^{228}Ac

and ^{40}K , efficiency versus density was fitted to an exponential decay function, for each gamma-energy, in order to obtain an empirical relationship between both. This fitting function is:

$$\varepsilon(\rho) = \frac{a}{\rho} \cdot (1 - e^{-b \cdot \rho}) \quad (2)$$

where a , b are the fitting parameters and ρ is the sample's density expressed in $\text{kg}\cdot\text{m}^{-3}$.

Results and discussion

Figure 1 shows the typical efficiency curves as a function of the energy for the different densities referred above. As can be seen, the differences between efficiency values for a given energy are greater in the low energy range, on which self attenuation effects are stronger.

The mathematical expression [Eq. (1)] was used, and the fitting coefficients obtained are included in Table 1. By using the data of this table, the relationship between efficiency and density was evaluated for each analyzed radionuclide. The results are shown in Fig. 2 indicating the densities of the calibrated samples used.

Table 2 compiles the parameters corresponding to the efficiency versus density curve fit found. As seen, the self-attenuation correction is bigger for radionuclides with low energy emission, ^{210}Pb , than for those with higher energy emission as ^{40}K .

Taking into account the data shown before, a new and practical method for the evaluation of the self-attenuation correction factor for the ^{210}Pb and ^{137}Cs radionuclides was studied. For this, a low cost calibrated sample IAEA-327 (manufactured by the International Atomic Energy Agency, Vienna, Austria) was used. Table 3 shows the relationship between the results achieved for some radionuclides to the standard source, 1000 $\text{kg}\cdot\text{m}^{-3}$ density, taking into account the corrective factors derived from Eq. (2) and those coming from the reference values reported by the IAEA certificate.¹¹ From the original source, seven different subsamples were compressed with different pressures and placed in standard 250 ml PVC cylindrical containers in order to obtain a variety of densities in the range from 930 to 1540 $\text{kg}\cdot\text{m}^{-3}$.

Table 4 shows, for each subsample, the activity concentration for the two radionuclides of interest, ^{210}Pb and ^{137}Cs , obtained from the net count rate under each photopeak, assuming a constant value for the efficiency not dependent of the density and equal to that evaluated for the standard source of 1000 $\text{kg}\cdot\text{m}^{-3}$. The difference between the measured activity value and the real one was expressed throughout the relative deviations showed in Fig. 3. The strongest influence of density was observed in the photopeak of 46 keV from ^{210}Pb on

which differences as high as 30% were calculated with respect to the reference one. These calculations indicate that all the environmental samples with densities lower

than calibrated will show higher concentrations than real while for those with high densities the calculated concentrations will be lower than the actual values.

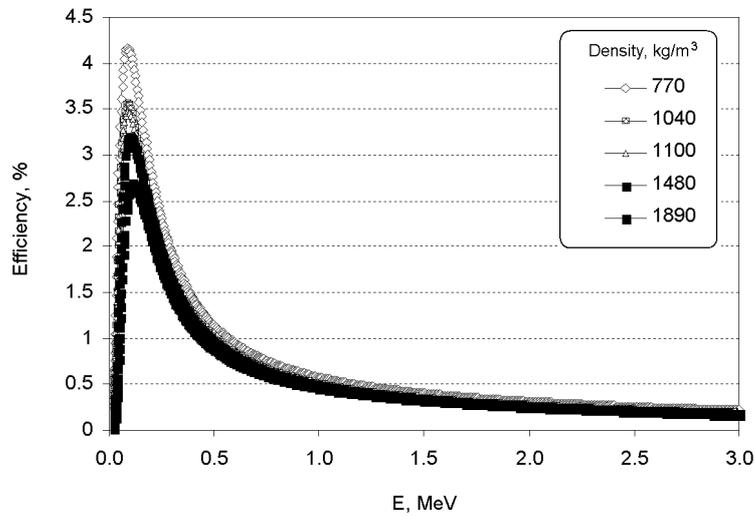


Fig. 1. Efficiency as a function of the photopeak's energy

Table 1. Parameters obtained for each density value corresponding to the efficiency vs. energy polynomial curve fit

Density, kg·m ⁻³	a ₀	a ₁	a ₂	a ₃	a ₄
770	-5,1586999	-0,9345001	0,0807259	-0,0176939	-0,0293190
1040	-5,2806859	-0,9227309	0,0812430	-0,0121974	-0,0272423
1100	-5,2860304	-0,9212684	0,0816052	-0,0115700	-0,0275976
1480	-5,3101161	-0,8939964	0,0884792	-0,0230412	-0,0334821
1890	-5,3774950	-0,8633085	0,0728391	-0,0591201	-0,0477706

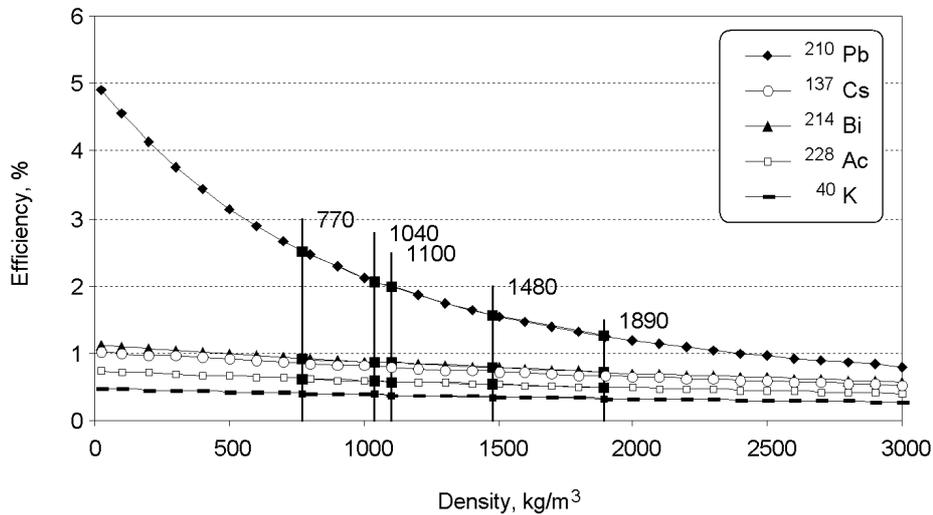


Fig. 2. Exponential fit of the relationship between efficiency and sample density for the selected energies

Table 2. Parameters obtained for each energy value, corresponding to the efficiency vs. density exponential curve fit

Radionuclide	Energy, keV	a , $\text{kg}\cdot\text{m}^{-3}$	Standard uncertainty, $u(a)$, $\text{kg}\cdot\text{m}^{-3}$	b , $\times 10^5 \text{ m}^3\cdot\text{kg}^{-1}$	Standard uncertainty, $u(b)$, $\times 10^5 \text{ m}^3\cdot\text{kg}^{-1}$
^{210}Pb	46.5	2420	88	212.0	20.0
^{214}Bi	609.3	2203	47	51.1	1.5
^{137}Cs	661.6	2063	49	50.1	1.7
^{228}Ac	911.2	1637	61	45.4	2.3
^{40}K	1460.8	1214	53	38.8	2.2

Table 3. IAEA reference values and activity concentration of the radionuclides of interest obtained in the UC laboratory using the efficiency obtained from the standard calibration procedure

	^{210}Pb , $\text{Bq}\cdot\text{kg}^{-1}$	^{137}Cs , $\text{Bq}\cdot\text{kg}^{-1}$	^{214}Bi , $\text{Bq}\cdot\text{kg}^{-1}$	^{40}K , $\text{Bq}\cdot\text{kg}^{-1}$	^{228}Ac , $\text{Bq}\cdot\text{kg}^{-1}$
IAEA data	58.8	24.9	34.1	621	38.7
UC laboratory	60.1	25.1	33.8	634	38.2

Table 4. Activity concentration of ^{210}Pb and ^{137}Cs measured in the seven subsamples assuming a constant value of the efficiency independent from density

Density, $\text{kg}\cdot\text{m}^{-3}$	^{210}Pb , $\text{Bq}\cdot\text{kg}^{-1}$	^{137}Cs , $\text{Bq}\cdot\text{kg}^{-1}$
930	61.7	25.6
1000	58.8	24.9
1100	54.1	25.6
1200	51.7	26.4
1300	48.2	27.1
1400	45.9	27.3
1540	42.3	27.8

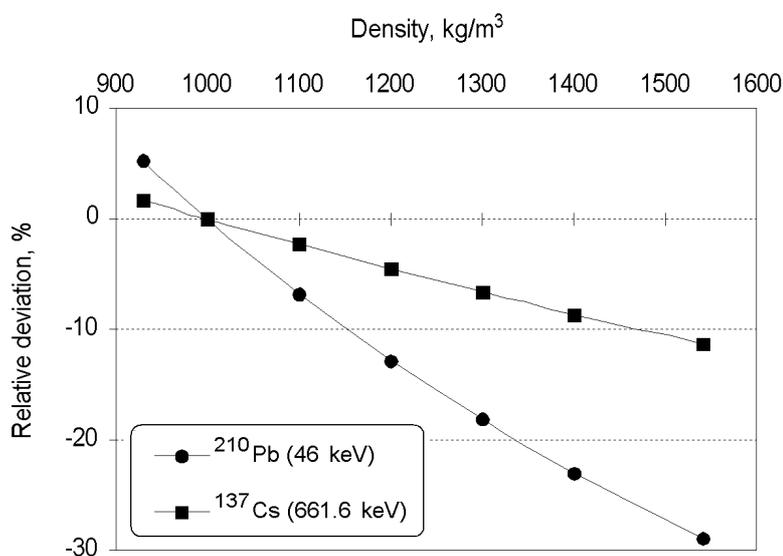


Fig. 3. Relative deviations between corrected activity concentrations with and without self-attenuation correction

For ^{137}Cs , as well as many other radionuclides with higher gamma-emission energies, the experimental data achieved in this practical case shows differences lower than 10% between the reference calibrated values of the sample and those achieved taking into account the correction by self attenuation taken from Fig. 3, which means that in this case the correction is not absolutely necessary.

Data referred in this note show how important is, in order to carry out accurate gamma-spectroscopy measurements, to develop density-dependent efficiency curves for the energies of the different radionuclides measured in the samples to be analyzed. If the self-absorption correction are not made for an specific measurement condition concerning sample size and

composition, either underestimations or overestimations may occur in the obtained results. Under the conditions used in our laboratory, these differences can be as high as 30%, especially when ^{210}Pb or other low energy emitting radionuclide are measured.

If the efficiency calibration is made using the sub-samples prepared from a standard one, all the above calculations can be made for the radionuclides of interest, representing a good and accessible method for reducing the extent of the variability introduced for self attenuation effects.

References

1. P. L. FERNANDEZ, I. GUTIERREZ, L. QUINDOS, J. SOTO, *Nature*, 321 (1986) 586.
2. A. NAVAS, J. SOTO, J. MACHIN, *Eur. J. Soil. Sci.*, 53 (2002) 629.
3. J. B. COWART, W. C. BURNETT, *J. Environ. Qual.*, 23 (1994) 651.
4. R. BLAGOEVA, L. ZIKOVSKY, *J. Environ. Radioact.*, 27 (1995) 269.
5. A. GELEN, O. DIAZ, M. J. SIMON, E. HERRERA, J. SOTO, J. GOMEZ, C. RODENAS, J. BELTRÁN, M. RAMÍREZ, *J. Radioanal. Nucl. Chem.*, 256 (2003) 561.
6. M. TZORTZIS, H. TSERTOS, S. CHRISTOFIDES, G. CHRISTODOULIDES, *J. Environ. Radioact.*, 70 (2003) 223.
7. R. M. W. OVERWATER, P. BODE, J. J. M. DE GOEIJ, *Nucl. Instr. Meth. Phys. Res.*, A324 (1993) 209.
8. J. P. BOLIVAR, R. GARCIA TENORIO, M. GARCIA LEON, *Nucl. Geophys.*, 8 (1994) 485.
9. L. QUINDOS, P. FERNANDEZ, J. SOTO, C. RODENAS, J. GOMEZ, *Health Phys.*, 66 (1994) 194.
10. Consejo de Seguridad Nuclear, Spanish Nuclear Safety Council, Intercomparación analítica entre laboratorios de radiactividad ambiental, 2004.
11. International Atomic Energy Agency, Report on the Intercomparison Run for the Determination of Radionuclides in Soils, IAEA-326 and IAEA-327, IAEA, April, 2001.