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DETERMINATION OF ^{7}BE IN SOIL SAMPLE BY GAMMA SPECTROMETRY FOR EROSION RESEARCHS

Alexander D. Esquivel[1], Geraldo F. Kastner[2], Ângela M. Amaral[2], Roberto Pellacani G. Monteiro[2] and Rubens M. Moreira[2]

[1] Centro de Investigaciones Hidráulicas e Hidrotécnicas (CIHH / UTP – PA)
Universidad Tecnológica de Panamá, Extensión Tocumen, Pabellón C
Vía Domingo Díaz al lado de Pazco, S.A.
alexander.esquivel@utp.ac.pa

[2] Centro de Desenvolvimento da Tecnologia Nuclear (CDTN / CNEN – MG)
Av. Presidente Antônio Carlos 6.627
31270-901 - Belo Horizonte, MG
gfk@cdtn.br
ama@cdtn.br
rpgm@cdtn.br
rubens@cdtn.br

ABSTRACT

Cosmogenic ^{7}Be is a natural radiotracer produced in the stratosphere and troposphere and reached to the Earth surface via wet and dry fallout and hence its measurement for research of erosion in soils is very significant. The ^{7}Be radioanalyse based on gamma spectrometry technique has been a routine methodology for decades and although is the reference procedure is not free of analytical interference. ^{7}Be is a β-γ emitting radionuclide (E_γ = 477.59 keV, $T_{½}$ = 53.12d) and depending on the chemical profile of the soil its determination is susceptible to ^{228}Ac (E_γ = 478.40 keV, $T_{½}$ = 6.15h) interference. The aim of this work was to establish an analytical protocol for the ^{7}Be determination in soil samples from Juatuba-Mg region in different sampling periods of dry and rainy seasons for erosion studies and to establish some methodologies for evaluating and correcting the interference level of ^{228}Ac in the ^{7}Be activity measurements by gamma spectrometry.

1. INTRODUCTION

'Fallout' radionuclides that adsorb strongly to solids have been used for decades to quantify soil dynamics. As a naturally occurring fallout radionuclide useful as soil tracer in soil cosmogenic ^{7}Be, (E_γ= 477.59keV, $T_{½}$=53.12d) [1] is deposited on the earth's surface. Gamma spectrometry allows for the convenient measurement of ^{7}Be in sediment samples without chemical separation steps [2-5]. Larsen and Cutshall [6] were the first to suggest the direct quantification of ^{7}Be, without radiochemical separation, in sediments by high-resolution gamma spectrometry as a way to trace rapid sedimentation in tidal rivers and near-shore environments.

Unfortunately, interfering photopeak of ^{228}Ac (E_γ= 478.40keV, $T_{½}$=6.15h) [1] derived from ^{232}Th-serie nuclide can happen depending on the type of soil. Despite having very low nuclear yield (0.209%) [1], these interference is detectable. ^{7}Be suffers convolution with the 478.40 keV photoemission of ^{228}Ac and this interference is not resolvable with conventional HPGe detectors. Furthermore, ^{228}Ac is a short-lived granddaughter of ^{232}Th. As a consequence of its very short half-life, ^{228}Ac activity is supported in natural materials by secular equilibrium with

its immediate parent ^{228}Ra, half-life 5.75 y [1], which in turn is supported by ^{232}Th, half-life 1.40e10 y [1].

In order to establish an analytical protocol for the ^{7}Be determination in soil samples from Juatuba-Mg region in different sampling periods of dry and rainy seasons for erosion studies three methodologies for evaluating and correcting the interference level of ^{228}Ac in the ^{7}Be activity measurements by gamma spectrometry based on data treatment of spectral and decay analysis were stablished namely, counting area ratios, long time counting and nuclide half-life measurements.

2. EXPERIMENTAL

2.1. Apparatus

The gamma-spectrometry measurements were carried out by using the HPGe detector (5019) with 50% relative efficiency, and DSA-2000 coupled to microprocessor with Genie 2 K software, from Canberra, USA.

2.2. Samples

Approximately 1 kg sample of soil is placed and distributed uniformly on a plastic tray and air dried for 48 hours and then is milled and crushed. After this step the sample is sieved and selected the particle size fraction less than 2 mm which is weighed and packaged in 700mL Marinelli beakers.

2.3 Analytical calibration

Due to the standard soil for calibration purpose wasn't available in our laboratory we use an alternative procedure based on the Monte Carlo software for ^{7}Be determination. This procedure has been used in studies involving analysis by gamma spectrometry [2, 7, 8].

2.4. Half-life methodology

In this methodology the same sample soil was counted in different time periods in order to evaluate de half-life associated to ^{7}Be (477.59keV) and possible interference contribution of ^{228}Ac photopeak energy (478.40kev). The purpose is to verify how much the determined half-life is close to the half-life of ^{7}Be.

2.5. Area ratio methodology

As the studied soil has a spectrum indicating the presence of thorium radionuclides family and hence the presence of the ^{228}Ac photopeaks at different energies, a comparison was made of some representative photopeaks of ^{228}Ac to photopeak countings at energy of 478.40 keV.
A standard soil with ^{232}Th was used for the reference area ratio determinations, according to,

$$R_r = \frac{C_{E\gamma}}{C_{E_{478.40}}} \quad (1)$$

In the equation 1, R_r is the reference area ratio, $C_{E\gamma}$ is the counting area for some representative photopeak of ^{228}Ac and $C_{E_{478.40}}$ the counting area for the photopeak at 478.40 keV. The expectation is that any change in the R_r value obtained in a soil sample analysis indicates the presence of ^{7}Be, considering that the ^{228}Ac is in secular equilibrium with their parents.

2.6. Long time methodology

In this methodology the same sample soil was counted in different long time periods, approximately one year, in order to promote the complete decay of ^{7}Be ($T_{½}$=53.12d) and ^{228}Ac counting if present from secular equilibrium.

3. RESULTS AND DISCUSSION

The half-life methodology was applied to soil sample with representative counting in the range 477.0 – 479.0keV in order to verify the photopeak at 477.59keV behavior. Fig. 1and Fig. 2 show the spectra obtained in time period, equivalent to the half-life of ^{7}Be.

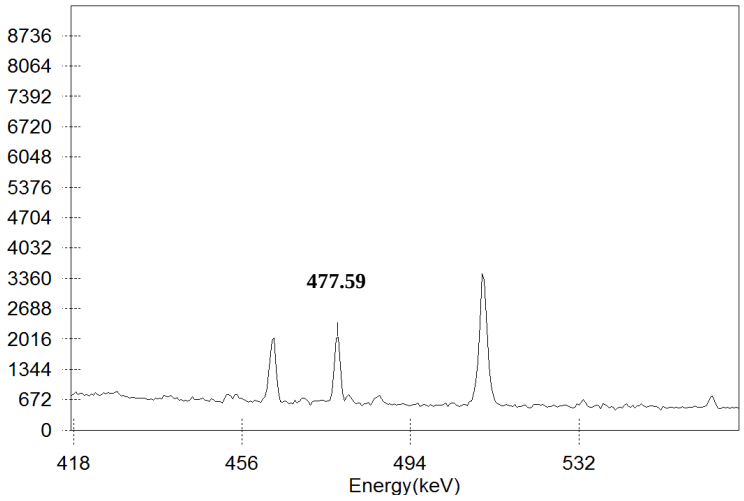

Figure 1: The gamma spectrum of soil sample

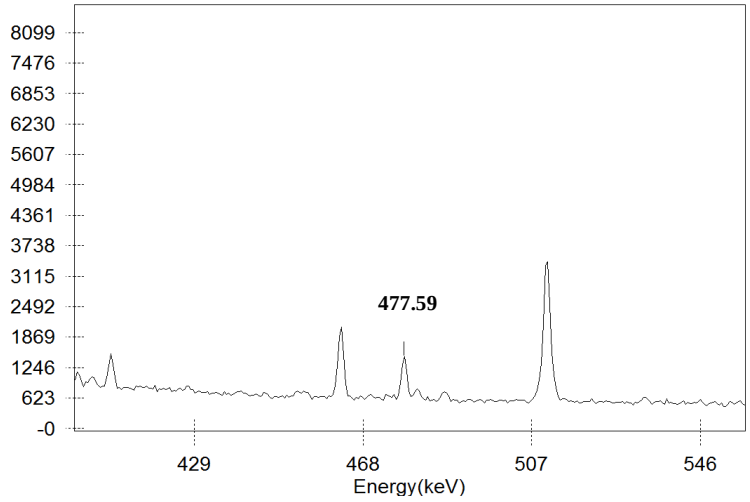

Figure 2: The gamma spectrum of soil sample after one half-life decay

After data analysis of gamma spectra, Fig. 1 and 2, the result of half-life for ^{7}Be, 51.3d was well consistent with the literature value, 53.12d [1]. The data analysis was carried out with equations (2) and (3) and the data in Table 1.

$$A(t) = \frac{N}{\varepsilon I_{\gamma}} \tag{2}$$

$$A_f = A_o e^{-\alpha t} \tag{3}$$

Table 1: Calculation of half-life of Beryllium-7

Counting date	N (cps)	A_o	A_f
13/04/2015	0.0289	8.3424	---
03/06/2015	0.0145	---	4.1872

ε = *0.03317* (counting efficiency to the energy of the gamma ray)
I_{γ} = *0.1044* (absolute probability of transition to gamma ray)

In the area ratio methodology a standard sample, ThO_2 – NBL 106A standard, was used for the reference area ratio, R_r, and the results are show in the Table 2. In the tables 3 and 4 are registered the values of R_s for two sample soils sampled in different seasons, rainy (soil 1) and dry (soil 2).

Is worth mentioning that the methodology area ratio is a new procedure developed among the personnel of the Laboratory of Nuclear Spectrometry (LNS) of the Analytical Techniques Service (SERTA) and Environmental Service (SEMAM) of CDTN. This methodology seeks to provide security to researcher that the results obtained are products of radionuclide under study

and the possible influence that may occur by other isotopes is negligible. Currently work is being done on improving the same and in a scientific manner support the results of its use.

Table 2: The R_r values for ^{228}Ac representative photopeaks of Th standard sample

Photopeak energy (keV)	Counting area	R_r
1,630.63	19000	5.121
1,588.20	38300	10.323
968.97	259000	69.811
964.77	89000	23.989
911.20	442000	119.137
835.71	24100	6.496
794.95	67100	18.086
772.29	22400	6.038
755.32	17000	4.582
674.75	2310	0.623
478.40[a]	3710	---
463.00	107000	28.841
409.46	51700	13.935
338.32	446000	120.216
328.00	98400	26.523
270.25	129000	34.771
209.25	194000	52.291
129.07	136000	36.658
99.51	62400	16.819

a. Interference photopeak energy

Table 3: The soil 1 sample R_s values for ^{228}Ac representative photopeaks

Photopeak energy (keV)	Counting area	R_s	Correlation factor[b] (%)
1,630.63	1990	1.064	20.779
1,588.20	3570	1.909	18.493
968.97	24700	13.209	18.920
964.77	9040	4.834	20.152
911.20	43500	23.262	19.525
835.71	2590	1.385	21.321
794.95	6880	3.679	20.342
772.29	2590	1.385	22.940
755.32	1830	0.979	21.357
674.75	0	0.000	0.000
478.40[a]	1870	-	---

463.00	11300	6.043	20.952
409.46	5240	2.802	20.108
338.32	35900	19.198	15.970
328.00	12100	6.471	24.396
270.25	15900	8.503	24.453
209.25	14200	7.594	14.522
129.07	10400	5.561	15.171
99.51	4220	2.257	13.417

a. Interference photopeak energy
b. $R_s / R_r \times 100$

Table 4: The soil 2 sample R_r values for ^{228}Ac representative photopeaks

Photopeak energy (keV)	Counting area	R_s	Correlation factor[b] (%)
1,630.63	1060	1.568	30.618
1,588.20	2180	3.225	31.238
968.97	15400	22.781	32.632
964.77	5130	7.589	31.634
911.20	25900	38.314	32.159
835.71	1690	2.500	38.485
794.95	4150	6.139	33.943
772.29	1530	2.263	37.486
755.32	1030	1.524	33.252
674.75	0	0.000	0.000
478.40[a]	676	-	---
463.00	7320	10.828	37.545
409.46	3160	4.675	33.545
338.32	22800	33.728	28.056
328.00	7090	10.488	39.544
270.25	16600	24.556	70.623
209.25	8770	12.973	24.810
129.07	7080	10.473	28.571
99.51	2460	3.639	21.636

a. Interference photopeak energy
b. $R_s / R_r \times 100$

For application of long time methodology a sample soil was analyzed two times, immediately after sampling and about seven ^{7}Be half-lives in order to 477.59keV photopeak disappearance and 478.40keV photopeak investigation. No photopeak was observed in the 477.0 – 479.0keV and which indicates that ^{228}Ac if present is with a very low activity. The Fig. 3 and Fig. 4 show the gamma spectra of soil sample.

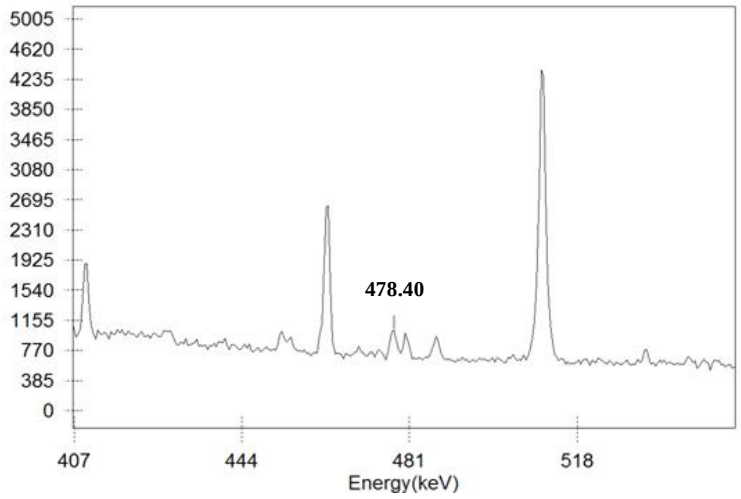

Figure 3: The gamma spectrum of soil sample

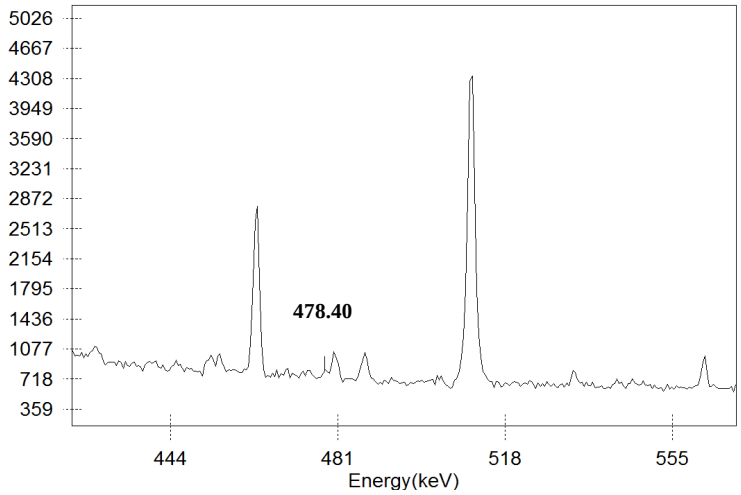

Figure 4: The gamma spectrum of soil sample after one year decay

3. CONCLUSIONS

For the area ratio methodology, the conclusion we can get is that the lowest values for R_s in the samples refers to the presence of ^{7}Be, the cosmogenic contribution. The correlation factor, $R_s / R_r \times 100$, suggests a mean reduction in the R_r values, of 18,49% and 32,54% for samples soil 1 and soil 2, respectively. When the R_s value is close to R_r, the ^{7}Be contribution to the counting area will be smaller, and the correlation factor tends to higher values. The interference of the ^{228}Ac will be more pronounced.

According to the results obtained in the long time and half-life methodologies, it is concluded that the interference of ^{228}Ac, if present, is not significant for soil samples studied.

Although ^{228}Ac interference in the determination of ^{7}Be, it can be used as a tracer. When the activity in 477.0-479.0keV region is very low and the presence of ^{232}Th family is evident, sophisticated deconvolution methods should be adopted [9] because the interference of ^{228}Ac is likely to be quite competitive.

When we associate the results of these three methodologies we can evaluate the interference level of ^{228} Ac on the ^{7}Be and thus to ensure the use of the tracer ^{7}Be for erosion studies in soils where ^{232}Th family may be present.

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REFERENCES

1. "The Berkeley Laboratory Isotopes: Project's: Exploring the Tables of Isotopes," http://ie.lbl.gov/education/isotopes.htm (2000).

2. Pinto, M.V.; Pires, L.F.; Bacchi, O.O.S.; Arthur, R.C.J.; Bruno, I. P.; Reichardt, K., "Spatial variability of ^{7}Be fallout for erosion evaluation," *Radiation Physics and Chemistry*, **Vol. 83**, pp. 1-7 (2013).

3. Kaste, J.M.; Norton, S.A.; Hess, C.T., "Environmental chemistry of berylium-7. In: E. S. Grew (Ed). Berylium: mineralogy, petrology, and geochemistry. Chantilly, VA: Mineralogical Socirty of America," *Reviews in Mineralogy & Geochemistry*, **Vol. 50**, pp. 271-289 (2002).

4. Blake, W.H.; Walling, D.E.; Hee, Q., "Fallout beryllium-7 as a tracer in soil erosion investigations," *Applied Radiation and Isotopes*, **Vol. 51**, pp. 599-605 (1999).

5. Cannizaro F., G.G.; Ranelli, M.; Spitale, M.C.; Tomarchio, E., "Concentration measurements of 7Be at ground level air at Palermo, Italy—comparison with solar activity over a period of 21 years," *Journal of Environmental Radioactivity*, **Vol. 72**, pp. 259-271 (2004).

6. Larsen, I.L.; Cutshall, N.H., "Direct Determination of ^{7}Be in sediments,"*Earth and Planetary Science Letters*, **Vol. 54**, pp. 379–384 (1981).

7. Vidmar, T.; et all, "An intercomparison of Monte Carlo codes used in gamma-ray spectrometry," *Applied Radiation and Isotopes*, **Vol. 66**, pp. 764-768 (2008).

8. Díaz, N.C.; Vargas, M.J., "DETEFF: An improved Monte Carlo computer program for evaluating the efficiency in coaxial gamma-ray detectors," *Nuclear Instruments and Methods in Physics Research A*, **Vol. 586**, pp. 204-210 (2008).

9. Landis, J.D.; Renshaw, C.E.; Kaste, J.M., "Measurement of 7Be in soils and sediments by gamma spectroscopy," *Chemical Geology*, **Vol. 291**, pp. 175-185 (2012).