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Proposal to correct for the effect of background and density in the determination of gamma emitters using sodium iodide detectors

Propuesta para corregir el efecto del fondo y la densidad en la determinación de emisores gamma empleando detectores de yoduro de sodio

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Abstract

This study develops a modification to the spectra decomposition method for sodium iodide scintillation equipment, including correcting the natural background counts for the activity concentration calculation. A comparison of the results obtained between two sodium iodide detectors of 2" × 2" and 3" × 3" dimensions versus a hyper-pure germanium detector of 50% relative efficiency is performed for soil, sediment, and water samples. It is found that background correction improves activity concentration results for higher density samples. The concentration values determined by the different spectrometric systems for the samples analyzed are comparable, which supports the results obtained with the proposed calculation method. The uncertainty associated with the measurement and limits of detection for the NaI(Tl) detectors is higher than those obtained with the GeHp detector due to the operation of the two technologies and the presence of interferences in the regions of interest. The methodological model proposed makes it possible to quantify, with reliable results, low concentration levels of NORM materials and even ^{131}I using sodium iodide detectors, among other analytical applications of general interest.

Keywords: modification method, NORM materials, sodium iodide detector, activity concentration.

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Resumen

Se desarrolla una modificación al método de descomposición de espectros para equipos de centelleo de yoduro de sodio, con la inclusión de una corrección a las cuentas de fondo natural para el cálculo de la concentración de actividad. Se realiza una comparación de los resultados obtenidos entre dos detectores de yoduro de sodio de 2" × 2" y 3" × 3" versus un detector de germanio hiperpuro de 50 % de eficiencia relativa, para muestras de suelo, sedimento y agua. Se encuentra que la corrección del fondo genera una mejora en los resultados de concentración de actividad para las muestras de mayor densidad. Los valores de concentración determinados por los diferentes sistemas espectrométricos para las muestras analizadas son comparables, lo cual soporta los resultados obtenidos con el método de cálculo propuesto. La incertidumbre asociada a la medición y los límites de detección para los detectores de NaI(Tl) es mayor a las obtenidas con el detector de GeHp, debido a la naturaleza del funcionamiento de las dos tecnologías y a la presencia de interferencias en las regiones de interés. El modelo metodológico planteado posibilita cuantificar, con resultados confiables, niveles bajos de concentración de materiales NORM e inclusive la determinación de ^{131}I usando detectores de yoduro de sodio, entre otras aplicaciones analíticas de interés general.

Palabras clave: método modificado, materiales NORM, detector de yoduro de sodio, concentración de actividad.

1. Introduction

Naturally occurring radioactive materials (NORM) are part of the earth's composition and are responsible for 85% of the radiation dose received by humans. Radiation comes mainly from radionuclides of the natural series of uranium (^{238}U) and thorium (^{232}Th), as well as potassium (^{40}K) [1]. The average values of the specific activity of ^{226}Ra, ^{232}Th, and ^{40}K in the earth's crust are 35, 30, and 400 Bq/kg, respectively [2], [3]. Therefore, determining a baseline NORM is of utmost importance for radiological protection of the population and radon mapping. Additionally, the determination of a baseline supports the productive use of some areas of interest, all of which need a large volume of samples requiring the use of a robust, reliable, economical, and fast spectrometric technique.

Solid-state semiconductor (GeHp) and scintillation (NaI(Tl)) detectors are commonly used in the determination of gamma emitters from the natural series associated with NORM materials. Analysis of spectra mainly natural radiation from ^{40}K, ^{235}U, radionuclides from the ^{238}U chain - ^{226}Ra, ^{214}Pb, ^{214}Bi; and those from the ^{232}Th chain - ^{228}Ac, ^{212}Pb, and ^{208}Tl are recorded [4]. The activity concentration (Bq/Kg) and/or radium equivalent activity - Raeq (Bq/Kg) is usually determined, ensuring secular equilibrium between radium and its daughters (after about 30 days or 10 half-life periods of ^{222}Rn) [1], [5], [6].

NaI(Tl) detectors are advantageous in that they have a simple operation, operate at room temperature, and are inexpensive. They have a high detection efficiency suitable for low radiation level materials [7], enabling analysis with shorter detection times and the analysis of a larger number of samples. In terms of disadvantages, Nal(Tl) detectors have a low resolution per energy, so their ability to distinguish between gamma rays of different energies is relatively poor. However, scintillators can be used successfully in the quantitative determination of mixtures containing only natural radionuclides (NORM) [3]. GeHp detectors, on the other hand, have better energy resolution, thus allowing the identification and quantification of complicated mixtures of gamma-emitting radionuclides [8]. Further, Germanium detectors have the disadvantage of working at low temperatures and require liquid nitrogen for proper operation, making them significantly more expensive than NaI(Tl) detectors [9].

There are several methods to compensate for the low spectral resolution of NaI(Tl) detectors [3], among which are: (1) Spectral decomposition using the *Generalized Least*

Squares (GLS) method in the energy range 300-2800 keV, which represents the spectrum as the sum of the background, ^{137}Cs, ^{40}K, ^{226}Ra and/or ^{232}Th; (2) Matrix method for the four energy regions: 600-720; 1350- 1560; 1640-1880; and 2500-2750 keV (method analogous to the spectral deconvolution described by Rybach (1988), Chiozzi et al. (2000), and Meng 2000)) [10], [11]; (3) Matrix method for 12 selected regions in the energy range; (4) Determination of the activity of ^{40}K, ^{226}Ra, ^{232}Th by calculating the 1460, 1764, and 2614 keV peaks; and (5) Determination of the activity of ^{137}Cs, ^{226}Ra, ^{232}Th by calculating the areas of the 662, 609 and 583 keV interference peaks. Additionally, some of the methods of spectra processing have been implemented as Script programs in such software as "Progress" [3].

In the decomposition method, the sample spectrum can be represented as a superposition of the contained radionuclide spectra and the background. The systematic decomposition of spectra into separate components determines the sample's activity [4], [12]. The spectra are processed by analyzing energy interval counts for a known matrix and the mixture of radionuclides such as ^{226}Ra, ^{232}Th, ^{40}K (920 Bq), and/or ^{137}Cs [1]. During processing, corrections to self-absorption are made assuming a linear dependence of the sensitivity coefficients on sample density. However, the sample density is usually not the same as the standard source density, for which correction coefficients are calculated by linear interpolation [13]. Uncertainties are calculated according to the international standard such as ISO 5725 [14].

NaI(Tl) detectors have been widely used in different applications, including: (i) Studies of the number of radionuclides found in milk powder and the total effective dose due to annual intake [15]; (ii) Determination of ^{131}I from thyroid cancer treatments in urban waste water treatment plants and rivers [16]; (iii) Methodologies for rapid screening of building material samples and determination of Raeq [1], [3]; (iv) Studies of geophysical processes in water [5] and investigations on samples of lichens, grasses, soil, and granite [12]; (v) Development of detailed maps of uranium distribution and its uncertainties using the Kriging method [17]; (vi) The influence of geological formations and their structure on the increase of radon concentrations in soil [6]; (vii) Development of a baseline for mapping radon-prone areas [18]; (viii) Assessment of indoor and outdoor environmental radioactivity [19], [20]; (ix) In situ underwater measurements of ^{40}K [21], [22] and tracer applications with ^{40}K [23]; (x) Using Monte Carlo (MC) simulations, the water content in agriculture [24] and the calibration of detectors using different analysis geometries [25]; (xi) Use of GEANT4 code to simulate radionuclide spectra submerged in seawater [26]; and (xii) High-resolution digital terrain models [27].

In this work, a correction to the mathematical calculation model used in the methodology of spectra decomposition to determine the activity concentration is proposed with a low energy resolution sodium iodide equipment. The modification comprises the inclusion of a correction in the natural background counts considering the sample density under analysis. In addition, the correction for self-absorption is accounted assuming a linear dependence from the density of the sample. Finally, a comparison is made between the results obtained with two NaI(Tl) detectors and a GeHp detector for soil, sediment, and water samples from a region of interest, while considering improvements in the spectrum analysis model and an optimization process of the sodium iodide equipment. The contribution of this study is to corroborate the usefulness of the proposed decomposition method and the scintillation technique determining low activity NORM samples. It also benefits the development of further studies, including the determination of a NORM baseline, determination of ^{131}I in environmental samples, and related radiological protection programs, among other applications mentioned above.

2. Methods

The aim is to adjust the decomposition method, where the sample spectrum can be represented as a superposition of the contained radionuclide spectra and the background, including a correction factor for the background measuring that contemplates an adjustment for the sample density. Physically, the photons detected in gamma spectrometry equipment can come from the real sample or reference material and the natural radiation (background). There is a phenomenon of self-attenuation of the sample's radiation and an attenuation of the natural background, both of which are affected by the density of the analyzed material. The self-attenuation is calculated assuming a linear dependence between the density of the analyzed sample and calibration reference material. The natural background counts are corrected using two samples (1 and 2) without radioactivity, B_1 and B_2, with different densities ($\rho_1 < \rho_2$) made of water (1.0 g/cm^3) and viscous polymer (1.45 g/cm^3), respectively (see Figure 1). An adjustment equation is obtained with the B_1 and B_2 materials measuring that allows correcting the value of the background count rate for the analyzed sample density. The only background affected by the sample density is the cosmic radiation which is detected also with the use of a lead shielding. In Figure 1, a greater attenuation of the natural background radiation is observed with increasing density of the material. Consequently, the area under the curve at the characteristic energy from the natural background counts (cts_f) obtained in the detectors will be greater for the lower density material, i.e., that $cts_{f1} > cts_{f2}$. In the calibration process, calculating the efficiency (e) is directly proportional to the number of counts of the reference material and the background. For this case, it would be obtained that $e_1 > e_2$. Additionally, the detection limit is higher as the value of the background counts increases, which affects the analysis of low concentration samples.

The calculation of the activity concentration is especially affected by the attenuation due to the density of the material analyzed. It is important to carry out a correction in the background counts due to the density of the sample, using a material without radioactivity, as well as the self-attenuation correction, given the difference between sample and reference materials density in the calibration process. For this reason, this study carries out a comparison of the concentration results obtained with two NaI(Tl) detectors (with a corrected calculation model) and one GeHp detector for soil (S), sediment (SED), and water (A) samples from an area of geological interest in Boyacá, Colombia.

Soil samples were collected using a Shelby auger and by removing soil profiles on escarpments. In addition, sediment and water samples were taken on the riverbeds of the area rivers. The sampling protocols were proposed using the Spanish Nuclear Safety Commission guidelines [28]-[31]. Many samples were taken at different points in the area of interest, choosing for this work one sample of soil, water, and sediment, respectively.

Aliquots of the three matrices were taken at each sampling point. These samples were packaged in the analysis geometry made of at cylindrical polyethylene recipes, preserved, and individually identified. They were transported in an insulated box at a temperature of 1 to 5 °C from the time they were taken until they were received at the laboratory. The samples were prepared "in situ," taking them to a 100 mL recipes mark and sealed with liquid silicone. To preserve the samples in a liquid matrix, a vacuum filtration was performed to remove particles present in the water. In addition, an acidification process at pH 2 with hydrochloric acid was conducted to avoid the proliferation of microorganisms. Each container was weighed before collection to determine the net weight of the sample. Once the samples were taken to the laboratory, they were stored for a minimum of 25 calendar days to ensure a secular equilibrium between ^{226}Ra and its daughters before analysis. The live measurement times in the sodium iodide equipment were 55 000 and 172 800 s (15.3 and 48 hours) with the goal of optimizing the measuring time and analyzing the limits of detection. In the case of germanium detectors, a sample was analyzed every two days with a live time of 172 800 s (48 hours).

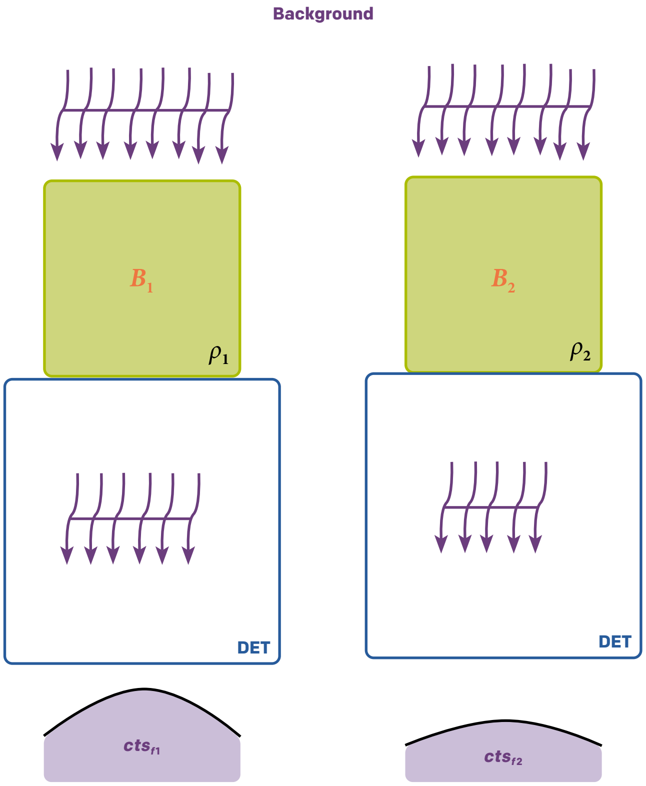

Figure 1. Radiation attenuation of the natural background

The detectors used for the determination of the activity concentration of the samples consist of: A 2"×2" in thallium-doped sodium iodide (NaI(Tl)) CANBERRA spectro-

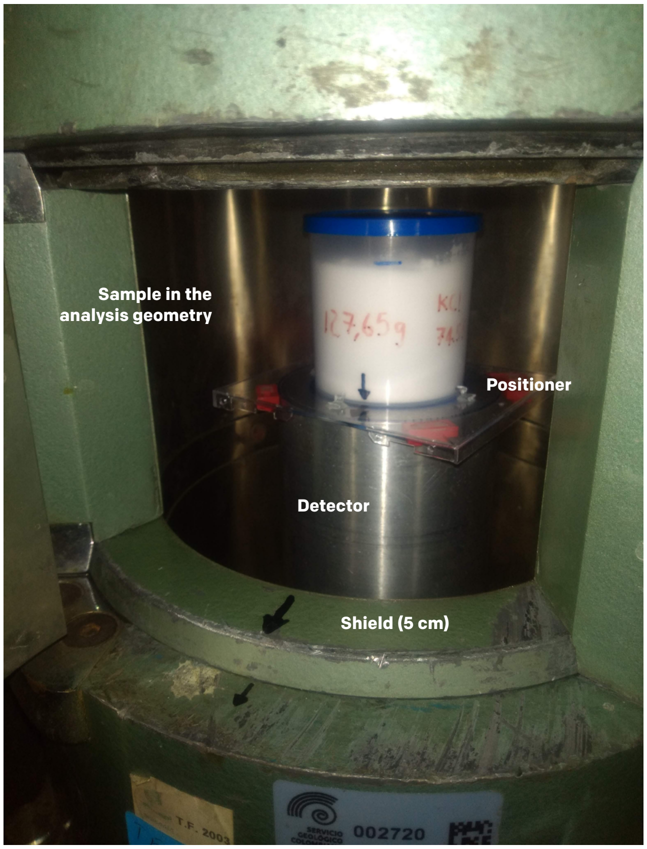

Figure 2. Experimental setup

meter (DETI), resolution 8.5% specific to the 662 keV peak of ^{137}Cs and a 5 cm shield of high purity lead [32]. A 3"×3" in CANBERRA NaI(Tl) detector (DETH), resolution 7.5% specific to the 662 keV peak of ^{137}Cs and a 5 cm shielding of high purity lead. A third CANBERRA N-type hyper-pure germanium (GeHp) detector (DET02), with 50% (r.e), a resolution (FWHM) at an energy of 122 keV ≤ 1.00 and an energy of 1.33 MeV ≤ 1.9 keV with a 10 cm shielding of high purity lead [33]. Efficiency calibration of the DET02, DETI, and DETH spectrometric systems, was performed with IAEA certified reference materials RGU-1 and RGTh-1 [34], and potassium chloride (KCl) at 99.5% purity, all prepared in the same analysis geometry and volume used with the real samples and B_1 - B_2 materials. The samples were placed over the detectors center, always in the same way with a positioner, reaching the best counting statistic and considering a detector acceptance factor equal to one under these conditions (see Figure 2).

The activity concentration (A) from the sample measurement in Becquerel/Kg of DET02 was calculated by equation 1. The characteristic photopeaks were established in each of the spectrometric systems to determine the net detected counts (cts_n) (without background counts) after the measuring time had elapsed. The efficiency (e) was established in a previously calibration using reference materials. The gamma yield γ, the mass of the sample m in kg, the live counting time t_c in seconds, and the decay correction factor f_d were considered.

$$A = \frac{cts_n}{e\gamma m t_c f_d} \tag{1}$$

For the semiconductor detectors, the energies of 351.9 KeV for ^{214}Pb, and 911.2 keV for ^{228}Ac corresponding to the natural series of ^{238}U and ^{232}Th, respectively, and 1460.8 keV for ^{40}K were established.

Efficiency calibration was performed using the GENIE2000 analysis software and its simulation tool, *Geometry Composer* [35]. Since the densities of the samples analyzed are all different, the *Geometry Composer* tool, using a mathematical model based on Monte Carlo, was used to calculate the specific efficiency for the detector and each of the samples analyzed. In addition, GENIE2000 calculated the uncertainty and minimum detectable activity (MDA) values based on standardized mathematical models.

The DETI and DETH detector efficiency calculation model was manual. Maestro acquisition software [36] was used to establish the areas or regions of interest (ROI) in the gamma-ray spectrum (see Figure 3) and their respective counts and associated uncertainties. For the scintillation detectors, the energies of 1764 KeV for ^{214}Bi, 2614.5 keV for ^{208}Tl corresponding to the natural series of ^{238}U and ^{232}Th, respectively, and 364.5 keV for ^{131}I and 1460.8 keV for ^{40}K were established. The amplitude of the regions of interest corresponds to 15% of the energy value on both sides of the maximum peak of the characteristic radionuclide [10] and the total spectrum amplitude ranged between 24 and 3092 keV. The contribution of K in the ROIs of U and Th was not significant, and the RGU-1 standard is almost pure U; thus, Th and K's contribution can be neglected. The K standard contains mainly Cl, I, Br, Na, and thus Th and U's activity can be neglected. In the RGTh-1 standard for Th, there was a negligible amount of K present, while U cannot be ignored.

On the other hand, a series of interferences was observed to the left of the peaks due to the Compton continuum coming from the higher energy events and lower energy peaks

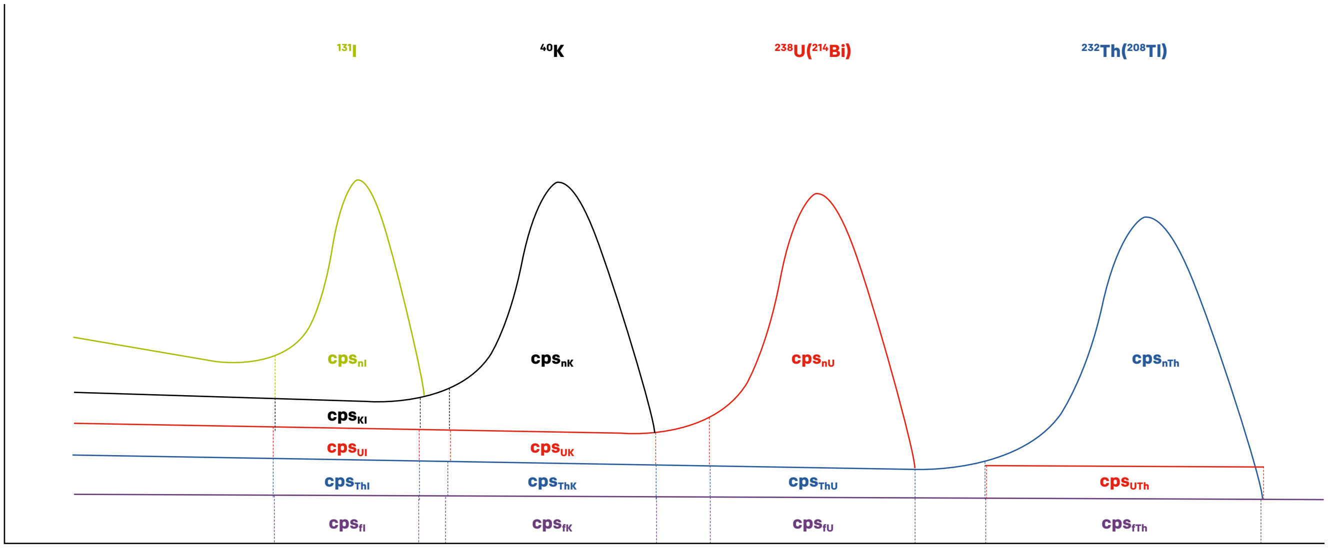

Figure 3. Idealized model of the regions of interest (ROI) and interferences of the decomposition method

of K, U, and Th (see Figure 3). Figure 4 presents a comparative spectrum in counts vs energy obtained with DETH for the background using the B_2 material of density 1.45 g/cm^3 (blue color) and the reference material RGU-1 (green color), where one can see the interfering peaks coming from U in the ROIs of Th, K, and ^{131}I. Additionally, two interfering peaks was observed on the right side of the spectrum on both sides of the thorium region, coming from ^{214}Bi [37]. In the GeHp equipment (DET02), the spectrum obtained for the reference material RGU-1 in counts vs energy shows the peaks coming from U between 1750 and 2640 keV. However, they did not interfere in the 2614 peak of ^{208}Tl due to the high resolution of the detector (see Figure 4). A similar analysis for the Th and K reference materials showed that the lower energy peaks of U and Th generated interference in the K and iodine regions of interest; Th did not generate considerable interference in the U region, and the iodine region was the most affected by the interferences coming from U and Th.

The spectra analyses for DETI and DETH were performed according to the following procedure with an own mathematical development. The sample spectrum can be represented as a superposition of the spectra of radionuclides contained in it and the background (see Figure 3). The activity concentration $[R]$ in (Bq/Kg) for each of the four regions of the sample spectrum is represented as:

$$[R] = \frac{cps_{nR}}{e_R^{\rho_m}} \tag{2}$$

Where R is the analysis regions for the ^{238}U, ^{232}Th, ^{40}K, and ^{131}I in that order, and cps are the counts per second. The term $e_R^{\rho_m}$ in cps (Bq/Kg)$^{-1}$ is defined as the efficiency for the characteristic radionuclide in the R region considering the correction to background and self-absorption by the sample density (ρ_m), which is expressed as:

$$e_R^{\rho_m} = e_R \frac{\rho_m}{\rho_{sR}} \tag{3}$$

Where ρ_m is the density of the sample analyzed in (g/cm^3) and ρ_{sR} is the density of the standard for the characteristic radionuclide in the R region. The quantity e_R in cps (Bq/Kg)$^{-1}$ is defined as the efficiency for the characteristic radionuclide in the R region and is calculated during the calibration of the spectrometer using the standard sources. The correction to the efficiency term is because the density of the sample is usually not the same as the standard, so one assumes a linear dependence of the efficiency term on the density of the samples.

The term cps_{nR} corresponds to the net count rate (n) in R region in cps and is expressed as:

$$cps_{nR} = cps_{tR} - \Sigma_{i \neq R} cps_{iR} - cps_{fR} \tag{4}$$

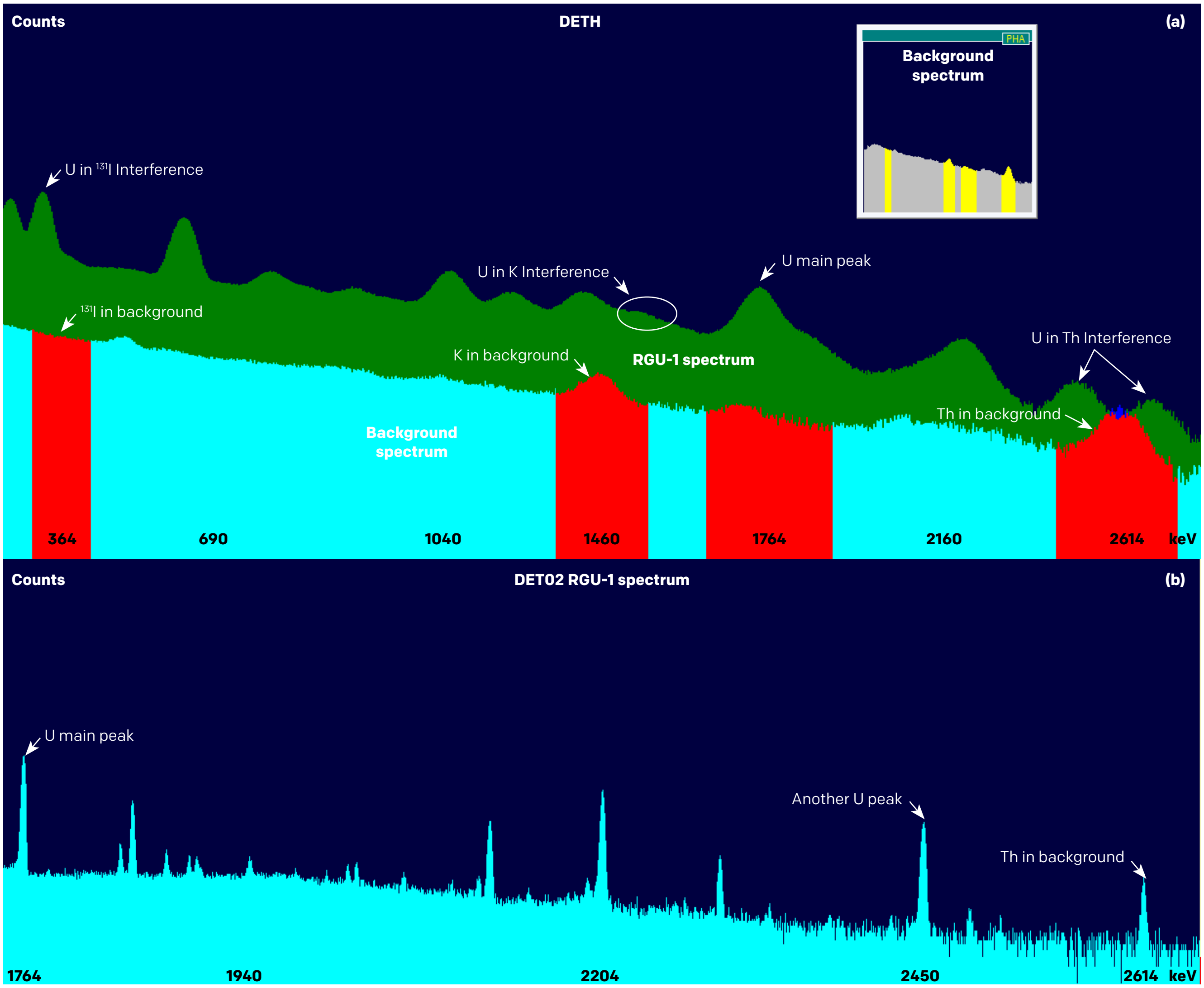

Figure 4. a) Comparative spectrum in counts vs energy (keV) between the background using a material without radioactivity, B_2, (ρ_2 = 1.45 g/cm^3) (in blue color and shown in the small window) and the RGU-1 material (in green color) obtained with DETH; b) RGU-1 spectrum in counts vs energy obtained with DET02 between 1750 and 2640 keV

where i is the radionuclide ^{238}U, ^{232}Th, ^{40}K, and ^{131}I in that order, and cps_{tR} the total count rate (t) in the R region. The term cps_{iR} is the count rate caused by radionuclide i in the R region in cps, which is used for the calculation of the contribution or interference factor of radioisotope i in the R region, C_{iR}, given by the expression:

$$C_{iR} = \frac{cps_{iR}}{cps_{ni}} \tag{5}$$

where cps_{ni} is the net count rate (n) for the radionuclide i in cps.

The term cps_{fR} corresponds to the background count rate (f) in the region R, which is corrected for sample density and is given by the expression:

$$cps_{fR} = cps_{ft}^{corr} \times \theta_{R}^{corr} \tag{6}$$

where cps_{ft}^{corr} is the corrected for density total background count rate (corr), which includes the counts in the total spectrum amplitude. The calculation of this factor considers the ten recent measurements of total background count rate obtained with the use of the two materials without radioactivity (B_1 and B_2) of density 1.0 (ρ_1) and 1.45 (ρ_2) g/cm^3

respectively, for a time of 55 000 s (15.3 h) each, where these values are used to adjust equations 7a and 7b through a linear interpolation. The expression 7a is used to correct the background count rate for sample with density values greater than 1.25 g/cm^3 and expression 7b for lower values. The background counts obtained with the B$_1$ material of ρ_1 is used when the correction for the sample density is not considering.

$$cps_{ft}^{corr} = cps_{fct}^{\rho_2} \times \left[1 + \frac{(\rho_2 - \rho_m)}{(\rho_2 - \rho_1)}(k-1)\right] \tag{7a}$$

$$cps_{ft}^{corr} = \frac{cps_{fct}^{\rho_1}}{\left[k - \frac{(\rho_2 - \rho_m)}{(\rho_2 - \rho_1)}(k-1)\right]} \tag{7b}$$

The constant k is the best linear adjustment to obtained the equations 7a and 7b. The term $cps_{fct}^{\rho_2}$ is the total background count rate in the entire spectrum amplitude obtained prior to sample analysis, using the B$_2$ material of ρ_2 and $cps_{fct}^{\rho_1}$ is the total background count rate using the B$_1$ material of ρ_1.

The term θ_R^{corr} corresponds to the corrected background fraction in the region R, which is expressed as:

$$\theta_R^{corr} = \theta_R^{\rho_1} + (\theta_R^{\rho_2} - \theta_R^{\rho_1}) \times \frac{(\rho - \rho_1)}{(\rho_2 - \rho_1)} \tag{8}$$

where $\theta_R^{\rho_1}$ is the average of the fraction between the background count rate in the region R and the total background count rate obtained with the use of the B$_1$ material of ρ_1 ($cps_{fR}^{\rho_1}$ / $cps_{fct}^{\rho_1}$), the term $\theta_R^{\rho_2}$ is the same fraction between the background count rate, but obtained with the use of the B$_2$ material of ρ_2 ($cps_{fR}^{\rho_2}$ / $cps_{fct}^{\rho_2}$).

Included in the calculation method is a contribution factor due to U interferences in the region of Th, C_{UTh^*}, which is calculated by the ratio of the net counts of thorium and uranium in the thorium ROI (cps_{UTh} / cps_{nTH}) obtained with the reference materials RGTh and RGU respectively. Making use of this factor adjusts the calculation of the thorium activity concentration using the following expression:

$$[Th] = [Th] - C_{UTh^*} \times [U] \tag{9}$$

The value of the detection limit for the proposed decomposition method, according to the theoretical approximations of Currie's work [38], is determined by the following expression:

$$LOD_R = \frac{4{,}66 + 2{,}71 \times \sqrt{(\Sigma_{i \neq R} cps_{iR} + cps_{fR}) \times t}}{e_R^{\rho_m}} \tag{10}$$

where LOD_R is the detection limit for the characteristic radionuclide in the R region given in (Bq/Kg) and t is the measuring time in seconds.

Measurement uncertainties were calculated following the ISO 5725 series [14] and provided with a confidence level of 0.95, consisting of random (statistical) and systematic errors. Systematic errors were mainly due to uncertainties associated with the certification of calibration sources and interferences of radioisotope i in the R region, reflected in the calculation of spectrometer efficiency coefficients, and uncertainties related to the measurement of sample mass. For the determination of the expanded uncertainty for the radionuclide R ($U_{[R]}$), the following expression was used:

$$U_{[R]} = \mathrm{k} * [R] * \sqrt{\frac{U^2_{cps_{nR}}}{(cps_{nR})^2} + \frac{U^2_{e_R^{\rho_m}}}{(e_R^{\rho_m})^2}} \tag{11}$$

where $U^2_{cps_{nR}}$ in cps is the expanded uncertainty of the net count rate in the region R and $U^2_{e_R^{\rho m}}$ in cps (Bq/Kg)$^{-1}$ is the expanded uncertainty of the efficiency for the radionuclide R considering the correction to self-absorption by the sample density (ρ_m).

3. Results and discussion

This study makes a comparison between the activity concentration results obtained for the GeHp detector (DET02) and the sodium iodide detectors (DETI and DETH) with and without using the natural background counts correction proposed in this work. The background counts obtained with the material without radioactivity of ρ_1 was used when the correction for the sample density is not considering. Soil, water, and sediment samples were analyzed during an acquisition time of 172 800 s (48 h). Table 1 presents the concentration values, which shows a percentage variation from 2.5 to 80.5 when comparing the results of K, U, and Th with and without the background correction for water and sediment samples, finding a greater difference in the sample with higher density (SED1).

A coefficient of variation in percentage (% CV) was performed for the K, U, and Th detectable data of water and sediment samples, which are measured with the DET02, DETI and DETH with and without the background correction in the calculation model of sodium iodide detectors. It was observed that concentration values obtained with the three spectrometers for K, U a Th are comparable, where the coefficient of variation is lower to 21.7% when the background correction is included (see Table 1); the results statistics can be improved with a larger number of samples. The DETH, of greater size and efficiency with respect to the DETI, presented uncertainty values closer to those obtained with the DET02. The uncertainty values for K and U were above 10% and for Th less than 7% for both sodium iodide detectors. DET02 recorded uncertainty values below 7% in all cases. The uncertainty values did not decrease with the background correction, given the small contribution of this counting rate in the expanded uncertainty calculation.

The systematic errors of the sodium iodide detectors were mainly due to the uncertainties associated with the certification of the calibration sources and the interferences of the radioisotope *i* in the R region, which is reflected in the calculation of the spectrometer efficiency coefficients. For the reference materials used, RGTh, RGU, and KCl, the uncertainty value for Th corresponded to 2.0%, for U to 0.5%, and for K to 0.6%. On the other hand, the contribution or interference factor, C_{iR}, increased to the left of the spectrum in the lower energy ROIs, causing the uncertainty value to be higher in the K region, before decreasing towards the Th region. The GeHp equipment presented lower measurement uncertainty due to the better calculation statistics associated with calculation software and its high resolution.

The water samples analyzed have non-detectable values of the radionuclides of interest due to the low concentration of U and Th. In addition, the proposed decomposition method presents a higher number of interferences in the left regions of the spectrum with energies below 1460 keV, which may affect the determination of ^{131}I and K. However, as the previous results show, the environmental water samples did not present detectable contents of U, Th, or K, which allows an adequate determination of ^{131}I in water, inasmuch as the latter, being of an anthropic nature, does not usually occur simultaneously with NORM.

Table 2 shows the results of the activity concentration and the detection limit (LOD) of the samples, analyzed with DETI with a measuring time equal to 55 000 and 172 800 s. The equation 10, for LOD, depends on the measuring time and sample interference counts in the interesting regions for K, U and Th. The interference counts avoid a greater decrease

Table 1. Activity concentration of real samples using a correction in the background counts

Analysis	Sample	DET02	DETI	DETH	DET02	DETI	DETH	DET02	DETI	DETH
			K (Bq/kg)			U (Bq/kg)			Th (Bq/kg)	
With correction	S1 ($\rho = 1.6$)	309.0 ± 13.7	261.0 ± 97.3	213.2 ± 30.7	57.7 ± 4.8	37.5 ± 23.7	45.4 ± 6.0	105.0 ± 6.3	84.9 ± 5.4	105.0 ± 6.1
	SED1 ($\rho = 2.1$)	162.0 ± 7.3	148.7 ± 70.1	143.0 ± 21.6	34.5 ± 2.9	27.4 ± 17.1	26.8 ± 4.2	36.8 ± 2.3	38.5 ± 2.8	37.3 ± 2.5
	A1 ($\rho = 1.0$)	< 9.9	< 31.3	< 26.0	3.6 ± 0.8	< 8.6	< 6.8	< 3.2	< 7.9	< 6.2
	% CV S1		18.4			21.7			11.8	
	% CV SED1		6.4			14.5			2.3	
Without correction	S1	309.0 ± 13.7	254.8 ± 98.0	177.5 ± 35.7	57.7 ± 4.8	40.0 ± 23.8	25.2 ± 6.4	105.0 ± 6.3	81.5 ± 5.3	94.5 ± 5.6
	SED1	162.0 ± 7.3	140.2 ± 70.4	113.9 ± 25.7	34.5 ± 2.9	31.3 ± 17.1	< 7.0	36.8 ± 2.3	33.6 ± 2.6	31.3 ± 2.2
	A1	< 9.9	< 31.3	< 26.0	3.6 ± 0.8	< 8.6	< 6.8	< 3.2	< 7.9	6.2
	% CV S1		26.8			39.7			12.6	
	% CV SED1		17.4			-			8.2	
% Variation With vs without correction	S1	-	2.5	20.1	-	6.4	80.5	-	4.2	11.1
	SED1	-	6.0	25.5	-	12.5	282.4	-	14.4	19.2

Table 2. Activity concentration and LOD with measuring time equal to 55 000 and 172 800 s using the DETI

Sample	Measuring time (s)	Concentration of activity			Detection limits		
		K (Bq/kg)	U(Bq/kg)	Th (Bq/kg)	LOD K	LOD U	LOD Th
S1	55 000	250.4 ± 93.9	34.6 ± 21.0	92.5 ± 6.9	65.7	18.1	14.0
	172 800	261.0 ± 97.3	37.5 ± 23.7	84.9 ± 5.4	36.9	10.0	7.8
SED1	55 000	125.2 ± 58.8	21.0 ± 14.4	44.2 ± 4.1	66.4	18.3	14.1
	172 800	148.7 ± 70.1	27.4 ± 17.1	38.5 ± 2.8	35.4	9.6	7.7
A1	55 000	62.6 ± 125.5	< 17.3	16.6 ± 6.8	59.2	16.9	14.3
	172 800	< 31.3	< 8.6	< 7.9	31.3	8.6	7.9

in the value of the LOD when the background correction is used. The table shows that the limits of detection decrease with increasing measuring time and increasing analysis time generates higher counting statistics and measurement accuracy. The uncertainty of the concentration increases when the value is close to the method detection limit, and the concentration is lower than the global average [2], [3].

Table 3 shows the values the range of limits of detection obtained with DETI for a given measuring time. The value of the world average natural concentration presented for K, U, and Th, establishes a methodological reference framework for achieving LOD less than or equal to 50% of the value of each radionuclide for each sample.

Table 3. Range of LOD using DETI

Range of detection limits			
Measuring time (s)	K (Bq/kg)	U(Bq/kg)	Th (Bq/kg)
172 800	31.3-36.9	8.6-10.0	6.9-7.9
55 000	62.6	17.3-18.5	14.2-15.0
Natural reference concentration	400	35	30

The above table shows that for a measuring time of 172 800 s (48 hours), this method obtains a detection limit four times lower than the world average concentration value, and for the time of 55 000 s (15.3 hours), the LOD is less than half of the world average value. NaI(Tl) detectors are characterized by their high detection efficiency suitable for low radiation level materials, using a measuring time of 55 000 s that allows to obtain an appropriate detection limit and the analysis of one sample per day.

4. Conclusions

This study proposes the inclusion of a mathematical correction associated with the background in the decomposition method used to calculate the activity concentration with NaI(Tl) detectors of low energy resolution. The improvement included a correction to the natural background counts considering the sample density and a simplified self-attenuation calculation. A comparison of the concentration values obtained with two sodium iodide detectors and a GeHp detector was carried out for soil, sediment, and water samples from an area of interest. In addition, the effect of varying measuring times on activity concentration, detection limit, and measurement uncertainty was analyzed.

Relevant percentage variations were obtained for the K, U, and Th activity concentration results, including the background counts correction, where the higher density samples present the greatest influence. Comparable activity concentration values between detectors were found for the samples analyzed, which corroborated the results obtained with the proposed calculation method and the inclusion of the background correction for sodium iodide equipment. The measurement uncertainty and limits of detection for the NaI(Tl) detectors were higher than those obtained with the GeHp equipment due to the low resolution, the use of a decomposition method, and the presence of interferences in the regions of interest. Non-detectable values of K, U, Th, and their interferences were recorded in the water samples analyzed, allowing for the determination of ^{131}I in water.

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