Coincidence summing correction factors for ^{238}U and ^{232}Th decay series using the Monte Carlo method.

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Abstract: Environmental samples analyzed in gamma spectrometry laboratories usually contain natural radionuclides such as ^{238}U and ^{232}Th. Using gamma spectrometry techniques is possible to estimate the activity of these radionuclides by measuring the gamma emissions of radionuclides belonging to their decay chain. Nonetheless, some of these radionuclides emit photons in cascade presenting Coincidence Summing (CS), which if not corrected, may affect the final activity quantification. The aim of this work is to apply the Monte Carlo method to calculate the True Summing Correction Factors (TSCFs) for ^{238}U and ^{232}Th decay series for different sample configurations (geometry and matrix) using the GEANT4 toolkit. In order to validate the results provided by GEANT4 using the RDM, the software TRUECOINC has been applied to calculate also the TSCFs. In addition, the influence of the geometry/matrix on the TSCFs is analyzed.

1. Introduction

A common application of gamma ray spectrometry is the quantification of activity in environmental samples containing natural radionuclides such as ^{238}U and ^{232}Th. Often, these radionuclides are in secular equilibrium with their progeny and, therefore, it is possible to quantify their activity by measuring gamma-emitting radionuclides using a High Purity Germanium (HPGe) detector. However, some of them emit photons in cascade within the resolving time of the detector and therefore, the spectrometer may detect one interaction with an energy equal to the sum of all of them. This phenomenon, named Coincidence Summing (CS), affects the experimental activity quantification of the radionuclide. In order to obtain the true activity, a correction factor (True Summing Correction Factor, TSCF) must be applied [Morel et al., 1983].

On the other hand, environmental samples often contain low-level radioactive sources, requiring HPGe detectors with close sample/detector geometries maximizing the measuring solid angle to increase the detection efficiency. However, this configuration also increases the coincidence summing effect thus, the consideration of TSCFs are mandatory if high efficiencies are needed [García-Talavera et al., 2001, Debertin and Schotzig, 1979].

Different methods to overcome this effect have been proposed in recent years, both through empirical [Quintana and Fernández, 1995, Montgomery et al., 1995] and computational techniques. For point sources, numerical expressions requiring only FEP (Full Energy Peak) and total efficiencies can be used to calculate TSCF [Rizzo et al., 2010]. Extended sources, however, become more difficult as the correction factor depends on the photon emission point, needing a more complex technic, either through Monte Carlo [Décombaz et al., 1992, García-Torano et al., 2005, Dryák and Kovár, 2009] or analytical methods [Schima and Hoppes, 1983].

The aim of this work is to apply the Monte Carlo method to calculate TSCFs for the ^{238}U and ^{232}Th natural decay series and for different sample configurations (geometry and matrix) using the GEANT4 toolkit [Agostinelli et al., 2003] along with the Radioactive Decay Module (RDM), following the methodology proposed by Hurtado in 2004 [Hurtado et al., 2004].

2. Materials and Methods

2.1. Methodology

In recent years, many different codes and computational methods have been developed to obtain TSCFs for extended sources.

KORSUM [Debertin and Schotzig, 1979], CORCO [Morel et al., 1983] and CSCOR [Sinkko and Aaltonen, 1985], where some of the first specific codes for TSCFs calculation based on Andrevv et al. (1972) [Andrevv et al., 1972] and Mc Callum et al. (1975) [McCallum and Coote, 1975] works.

ETNA (Efficiency Transfer for Nuclide Activity measurements) applies the efficiency transfer analytical method based in the recursive formula proposed by Andrevv using the NUCLÉIDE decay data [Piton et al., 2000, Lepy et al., 2004].

GESPECOR (Germanium Spectroscopy Correction factor) uses a coupled method of Monte Carlo and analytical techniques for the efficiency calibration and the TSCFs calculation [Sima and Arnold, 2000].

TRUECOINC [Sudár, 2002] applies a combinatorial method for the TSCF computation, requiring as input data the detector efficiencies and the Evaluated Nuclear Structure Datafile (ENSDF).

CYLTRAN, MCNP and GEANT3 are Monte Carlo tools used for peak and total efficiency calibration. Different methods have been developed to TSCFs calculation, combining them with other codes for the decay simulation or just comparing the simulated efficiencies with the experimental ones and considering the counting difference as the CS effect [Helmer and Gehrke, 1997, Dziri et al., 2012, Morel et al., 1983].

In this work, the GEANT4 toolkit is used for both, FEP efficiencies simulation and TSCFs calculation. In order to obtain accurate results, a characterization of the efficiency response for each sample is required. It is well known that the efficiency curve obtained by simulation shows discrepancies with the experimental one when using the manufacturer geometric data [García-Talavera et al., 2000, Jurado-Vargas and Guerra, 2006]. In this frame, a complete characterization of the detector as well as the FEPE calibrations are required to validate TSCFs quantification [Ordóñez et al., 2018].

The detector system simulated consists of a type-N GMX40 HPGe detector from ORTEC (Fig. 1) along with a multi-channel analyzer with 8192 channels, used in the environmental radioactivity laboratory (LRA) from the Universitat Politècnica de València (UPV).

The entire process, including the detector characterization, the FEPE calibration and the TSCFs calculation is determined for five samples with different geometries, matrix composition and densities.

- 25 ml water Petri box (PS25W).
- 100 ml water, sea sand and Zr sand Petri boxes (PS100W, PS100SS and PS100ZrS, respectively).
- 500 ml water Marinelli beaker (MS500W).

The sample geometries have been chosen to study the difference between extended (PS100 and MS500) and closer to point source (PS25) geometries. Moreover, the MS500 allows comparing top with surrounding geometries. On the other hand, the effect of the density on the TSCFs is analyzed comparing three sample compositions or matrices. Water matrix with low density (1.0 g/cm^3), an environmental sample of sea sand with medium density (1.65 g/cm^3) and a non-environmental sample of zirconium sand with high density (3.5 g/cm^3).

The composition of every sand has been analyzed by electronic microscopy. The sea sand sample is composed by silicon dioxide calcium and aluminum (86.57%, 11.48% and 1.95%, respectively). On the other hand, the Zr sand is composed by silicon dioxide and zirconium (57.33% and 42.67%, respectively).

Once the detector model is validated, to calculate correction factors, it is necessary to perform two different simulations for each radionuclide of interest. Monoenergetic sources are simulated, obtaining the efficiency for a given energy without considering the CS effect. On the other hand, a second simulation is performed using the RDM, thus simulating the complete decay of the radionuclide and considering the CS effect. Finally, the TSCFs have been calculated as the ratio between the photo-peak area obtained in the first simulation by the photo-peak area of the second one, considering the gamma yield of each photon-energy. Fig. 2 summarizes the methodology followed for the TSCFs calculation.

In order to validate the results provided by GEANT4 using the RDM, the software TRUECOINC [Sudár, 2002] has been applied to calculate also the TSCFs. TRUECOINC calculates the TSCFs through an analytical expression based on the combinatorial method carried out by [Daróczy et al., 1982]. This method requires additionally the full energy peak efficiency curve as well as the total-to-peak efficiency curve to characterize the response of the detector. These curves have been obtained using GEANT4, simulating only monoenergetic sources.

To study the influence of the matrix and the solid angle on the TSCFs, a statistical test has been applied. Accordingly, PS100W, PS100SS and PS100ZrS have been compared between them and the same procedure is carried out for PS100W, PS25W and MS500W, following this expression:

$$u = \frac{|V_1 - V_2|}{\sqrt{u_1^2 + u_2^2}} \sim N(0,1) \quad (1)$$

where u represents the statistic used to compare the TSCFs (V_1, V_2) taking into account their uncertainties (u_1, u_2) [Brookes et al., 1979].

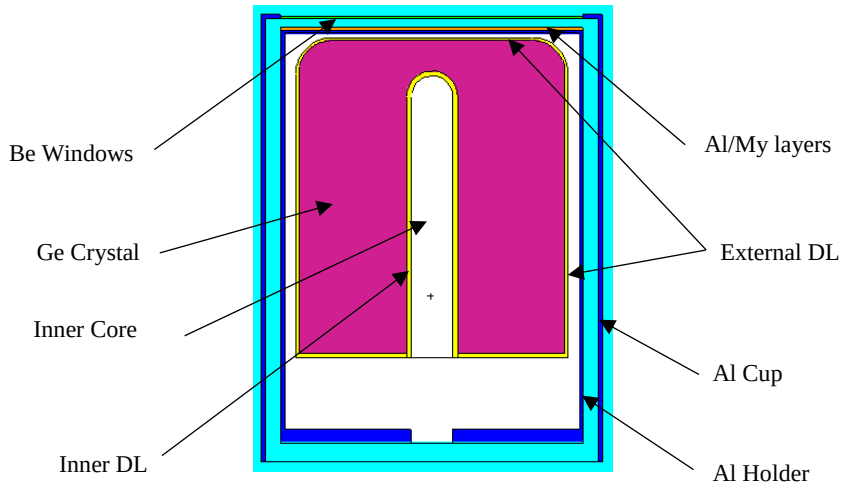

Fig. 1: GMX40 HPGe detector model. The scheme is not to scale.

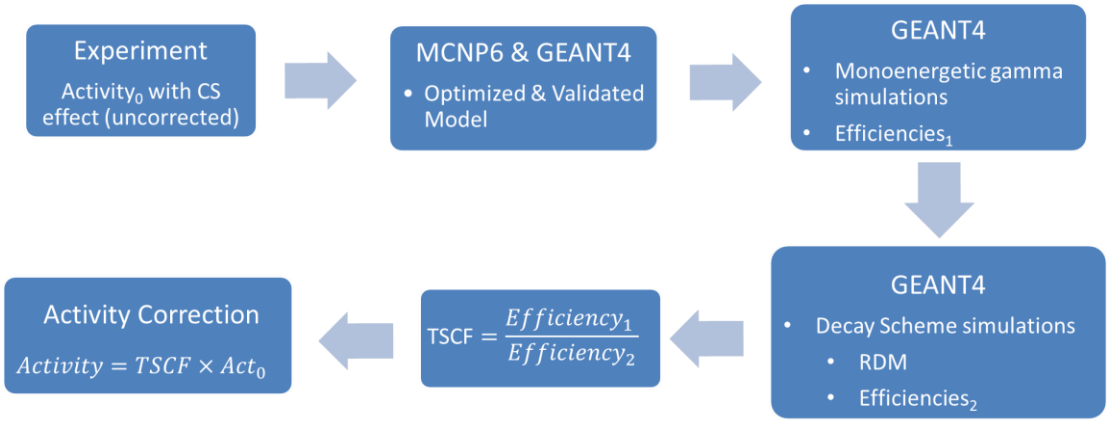

Fig. 2: TSCF calculation

2.2. *GEANT4 Simulations*

The number of histories in each simulation has been established in 20 million to achieve good statistics. The physics processes activated are the following:

- Compton and Rayleigh scattering.
- Pair production.
- Auger electron production.
- Photoelectric effect for photon.
- Ionization processes and Bremsstrahlung for secondary particles.

The Radioactive Decay Module (G4RadioactiveDecay class) is used to simulate the decay of the radionuclides and the coincidence summing effect, using branching ratios from the Evaluated Nuclear Datafile (ENSDF)

2.3. *Decay Series*

In order to quantify the activity of ^{238}U and ^{232}Th by γ-ray spectrometry techniques, a standard practice is to analyze their progeny. However, the problem arises when selecting the appropriate radionuclides, since not all of them are measurable, either because their gamma emissions are too weak or with a very low energy or even because their gamma ray overlaps with the photo-peaks of other radionuclides.

Regarding the ^{238}U decay series, there are gamma-emitting radionuclides such as ^{234}Th and ^{234}Pa^{m} but they present high uncertainties when measuring with γ-ray spectrometry. Analyzing the ^{238}U progeny, ^{214}Bi is found suitable to be measured because of its intense gamma emissions (609.3 and 1120.3 keV). However, both photons have CS effect, being necessary to apply TSCFs. Another acceptable radionuclide to be measured is ^{214}Pb, which does not present such effect. In a closed system ^{214}Pb and ^{214}Bi reach secular equilibrium with ^{226}Ra within 1 month, thus, ^{226}Ra activity can be obtained from them. Assuming that ^{226}Ra is also in secular equilibrium with ^{238}U, at least in solid matrices, the activity of the precursor can be estimated.

Concerning the ^{232}Th decay series, ^{212}Bi and ^{208}Tl are suitable to be measured using their main emissions at 727.3 keV and 583.2 keV, respectively. Likewise ^{214}Bi, they have summing coincidence and TSCFs must be calculated.

Assuming secular equilibrium, ^{228}Th activity can be obtained from the latter radionuclides and thus, the precursor activity can be estimated.

3. Results

Table 1-3 list the efficiencies and the TSCFs obtained for each radioisotope and energy under study. The RDM efficiency represents the efficiency simulated with the G4RadioactiveDecay class (GEANT4), considering the true coincidence summing effect. The gamma efficiency, on the other hand, represents the efficiency for a given energy, without considering such effect.

The TSCFs calculated by TRUECOINC are in good agreements with those obtained with GEANT4, showing lower values in all cases except for ^{212}Bi with the PS100ZrS. The mean differences when comparing TRUECOINC with GEANT4 are -1.91%, -0.14% and -2.10% for ^{214}Bi, ^{212}Bi and ^{208}Tl, respectively. This ensures the goodness of the RDM performance for the radionuclides under study.

Results show that the PS25 geometry has the highest TSCFs with values around 1.19 and 1.21 for ^{214}Bi (Table 1), 1.05 for ^{212}Bi (Table 2) and 1.23 for ^{208}Tl (Table 3). On the other hand, the MS500 geometry presents the lowest TSCFs with values around 1.12 and 1.14 for ^{214}Bi, 1.16 for ^{208}Tl and 1.04 for ^{212}Bi. The PS100 shows intermediate values but closer to the MS500 geometry.

The TSCFs for ^{212}Bi at 727.3 keV obtained for the PS100 and MS100 samples are almost 1.0 (Table 2), so for these geometries and matrices, the coincidence summing effect could be neglected, but not for the PS25, with a TSCF of 1.05.

Comparing the results of the PS100 for water, sea sand and Zr sand (Table 1-3), it is found that TSCFs are very similar for both, uranium and thorium series. The p-values listed in Table 4 are much higher than 0.05 for the PS100W-PS100SS and for the PS100SS-PS100ZrS, which suggest that these matrix compositions/densities have no effect on the TSCFs [Giubrone et al., 2016]. On the other hand, when comparing water with Zr sand, which have very different densities (Table 4), it is found that 1120.3 and 583.2 keV show p-values lower than 0.05 and higher for 609.3 and 727.3 keV. An in-depth study on the influence of high density matrices is required.

Regarding the geometry of the sample, the TSCFs show significant differences when comparing PS100W with PS25W and PS25W with MS500W (Table 1-3), with p-values much lower than 0.05 (Table 5). Accordingly, the solid angle between the sample and the detector is of relevance. On the other hand, PS100W and MS500W have similar TSCFs, with p-values higher than 0.05 for ^{212}Bi and ^{208}Tl and lower than 0.05 for ^{214}Bi, so the solid angle effect is greater when comparing extended to more likely point source geometries. The PS100W-MS500W p-values indicate that the surrounding geometry barely affects the TSCFs calculation.

Table 1: TSCFs for ^{214}Bi (609.3 and 1120.3 keV); ^{238}U decay chain.

Sample	Energy (keV)	RDM Efficiency	Gamma Efficiency	TSCF GEANT4 (uncertainty)	TSCF TRUECOINC	Difference
PS25W	609.3	0.0354	0.0420	1.187 (±0.003)	1.166	-1.8%
	1120.3	0.0211	0.0255	1.211 (±0.005)	1.190	-1.7%
PS100W	609.3	0.0240	0.0270	1.126 (±0.003)	1.106	-1.8%
	1120.3	0.0146	0.0169	1.158 (±0.006)	1.119	-3.4%
PS100SS	609.3	0.0229	0.0259	1.134 (±0.008)	1.114	-1.8%
	1120.3	0.0144	0.0164	1.138 (±0.02)	1.125	-1.7%
PS100ZrS	609.3	0.0197	0.0222	1.130 (±0.003)	1.112	-1.6%
	1120.3	0.0127	0.0145	1.140 (±0.006)	1.121	-1.7%
MS500W	609.3	0.0225	0.0252	1.119 (±0.003)	1.105	-1.3%
	1120.3	0.0139	0.0159	1.144 (±0.006)	1.117	-2.3%

Table 2: TSCFs for ^{212}Bi (727.3 keV); ^{232}Th decay chain.

Sample	RDM Efficiency	Gamma Efficiency	TSCF GEANT4 (uncertainty)	TSCF TRUECOINC	Difference
PS25W	0.0344	0.0362	1.053 (±0.005)	1.053	-0.0%
PS100W	0.0227	0.0235	1.037 (±0.006)	1.034	-0.3%
PS100SS	0.0217	0.0226	1.039 (±0.006)	1.036	-0.3%
PS100ZrS	0.0190	0.0196	1.030 (±0.007)	1.035	0.5%

MS500W	0.0212	0.0220	1.040 (±0.006)	1.034	-0.6%

Table 3: TSCFs for ^{208}Tl (583.2 keV); ^{232}Th decay chain.

Sample	RDM Efficiency	Gamma Efficiency	TSCF GEANT4 (uncertainty)	TSCF TRUECOINC	Difference
PS25W	0.0356	0.0437	1.226 (±0.004)	1.222	-0.3%
PS100W	0.0241	0.0280	1.160 (±0.004)	1.131	-2.5%
PS100SS	0.0231	0.0269	1.165 (±0.004)	1.140	-2.1%
PS100ZrS	0.0196	0.0230	1.170 (±0.004)	1.133	-3.2%
MS500W	0.0226	0.0261	1.156 (±0.004)	1.129	-2.4%

Table 4: u statistic (p-value); Matrix Comparison; Calculated for GEANT4 values

Radionuclide	Energy (keV)	PS100W-PS100SS	PS100W-PS100ZrS	PS100SS-PS100ZrS
^{214}Bi	609.3	0.94 (0.175)	0.96 (0.168)	0.45 (0.327)
	1120.3	1.12 (0.132)	2.11 (0.017)	0.14 (0.446)
^{208}Tl	583.2	0.92 (0.179)	1.87 (0.030)	0.96 (0.168)
^{212}Bi	727.3	0.29 (0.387)	0.73 (0.231)	1.00 (0.158)

Table 5: u statistic (p-value); Geometry Comparison; Calculated for GEANT4 values

Radionuclide	Energy (keV)	PS100W-PS25W	PS100W-MS500W	PS25W-MS500W
^{214}Bi	609.3	16.12 (0.000)	1.70 (0.044)	17.69 (0.000)
	1120.3	6.74 (0.000)	1.71 (0.044)	8.52 (0.000)
^{208}Tl	583.2	13.19 (0.000)	0.78 (0.218)	13.92 (0.000)
^{212}Bi	727.3	2.06 (0.020)	0.41 (0.340)	1.57 (0.058)

4. Conclusions

GEANT4 toolkit, along with the Radioactive Decay Module, has been used to calculate the TSCFs of ^{214}Bi and ^{212}Bi / ^{208}Tl from the ^{238}U and ^{232}Th decay series, respectively.

The LRA works with different matrices (water, sea sand and zirconium sand) and different geometries (small petri of 25 ml, large petri of 100 ml and Marinelli beaker of 500 ml) for the estimation of the activity of ^{238}U and ^{232}Th natural series. Regarding the radionuclides studied in this paper, all of them emitting high energy photons, it has been determined that the effect of the density in the TSCFs is not significant as compared to the effect of the geometry and hence, the solid angle. The p-values obtained when comparing the PS100W with the PS100ZrS show that high density matrices still need further study.

The conclusions obtained for these radionuclides can be extrapolated to other matrices of different density (biota, soils, sediments, etc.). However, these conclusions cannot be generalized for radionuclides with low energy photon emitters and both the composition and density must be precisely characterized in such cases.

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