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Validation of a new true coincidence summing correction algorithm for Genie to Monte-Carlo simulations and measurements

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Abstract
A new algorithm for calculation of true coincidence summing correction factors has been developed for the Genie software suite, a nuclear material characterization software packaged offered by Mirion Technologies. The calculated correction factors have been compared to measured correction factors for eight single-radionuclide standard sources as well as calculated correction factors from Monte-Carlo simulations using the G4ARES framework based on Geant4 and MCNP-CP. The calculated correction factors agree to within a few percent with each other and with the measured correction factors within 5% for most emissions, while a few emissions have up to 10% difference. Radionuclides decaying with electron capture and/or having high internal conversion coefficients were the most challenging radionuclides.

Keywords True coincidence summing · Gamma spectrometry · Monte-Carlo radiation transport

Introduction

True coincidence summing (TCS) occurs when a detector detects more than one gamma- or X-ray from a single decay of a radionuclide. This changes the probability of registering a count in the full-energy peak from a gamma emission. If not corrected for, it can lead to incorrect activity determination. The effect is largest for high-efficiency geometries that offer the best detection sensitivity and shortest feasible measurement times. It is also highly dependent on the decay of the radionuclide being measured.

There are two fundamentally different approaches of calculating the TCS correction factors. The first consists of finding the full energy peak and total efficiencies and a representation of the decay schema to calculate the probability of detecting multiple photons from a single decay [1, 2]. The second one is to use a Monte-Carlo radiation transport code [3–6] which tracks gamma- and X-rays through a geometry and tallies the counts in a peak from an emission. The simulation is run twice: once with all emissions emitted in a single event and once with emissions in different events. The correction factor is the ratio of the two simulations [7]. The first approach relies on an accurate method to calculate the efficiencies while the second approach relies on an accurate model of the detector and sample. Both methods rely on the radionuclide decay data. Efforts have been made to compare different methods to each other [8–11] with similar results for a few radionuclides.

Automated utilities that provide reliable and accurate TCS corrections are a necessity to offer timely and accurate results from radiometric sample analysis. Mirion has developed a new algorithm to calculate TCS correction factors for consideration as a future enhancement in the Genie software. The new algorithm is expected to improve upon TCS corrections for radionuclides with complex decay, and detectors with very high efficiency geometries, and to include more radionuclides in the TCS corrections library than previously.

Comparison of calculated to measured correction factors is the ultimate test of the quality of the TCS correction factor calculations. Measuring correction factors are challenging and require a high-quality efficiency calibration, a validated detector model, and measurement of radionuclides with known activities. TCS correction factors have been measured for eight radionuclides at Pacific Northwest National Laboratory (PNNL). The new proposed algorithm in Genie has

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been compared to these measured values as well as calculated values from G4ARES [4], MCNP-CP [5, 12] and the current algorithm in Genie v3.4 [1, 13].

Theory

The new proposed TCS algorithm in Genie contains three parts: radionuclide decay data processing, full energy peak and total efficiency calculation, and TCS correction factor calculation. The algorithm has two radionuclide decay libraries: in one of them the decay data is taken from Evaluated Nuclear Structure Data Files (ENSDF) [14] while in the other the decay data is taken from Decay Data Evaluation Project (DDEP) [15] for radionuclides included in DDEP and from ENSDF for radionuclides where DDEP data is not available. For both libraries the atomic data is taken from Firestone [16]. In all calculations for this article, ENSDF was used to be consistent with the other calculation methods. The parsing algorithm steps through the decay of each radionuclide and determines all the possible decay chains using a recursive acyclic forward graph algorithm. The probability that the chain occurs and the transitions it contains are stored in an intermediate database. If the chain goes through an excited state that has a half-life of more than 1 μ s, the chain is broken into two. Photons emitted before the decay of the excited states are assumed not to be in coincidence with the photons emitted after the decay of the excited state. The database also contains data for the transitions, including internal k and l shell conversion coefficients for gamma transitions and k and l shell electron capture probabilities for electron capture transitions. The internal conversion coefficients for the l subshells are calculated using the BrIcc software [17] and the l subshell electron capture probabilities are calculated as in Bambynek [18]. Parsing of the radionuclide decays is done once and stored for use in the third part of the algorithm. The second part of the algorithm is calculating full-energy peak and total efficiencies for a set of virtual point sources distributed in a volume source. The calculation is performed using the ISOCS/LabSOCS [19] software.

The TCS correction factor, CF, is defined as the probability of registering a count in the FEP with TCS divided by the probability of registering a count in the full-energy peak (FEP) if TCS did not occur. Using this definition, the uncorrected activity for a radionuclide needs to be divided by the correction factor. The probability of registering a count in the FEP without TCS from an emission i is

$$P_{noTCS_i} = \varepsilon_i I_i = \varepsilon_i c_i \sum_{j=1}^{n} \delta_{ij} d_j \tag{1}$$

where ε is the FEP efficiency, c is the probability of a gamma being emitted when the transition occurs, δ is 1 if the transition is part of the chain and 0 otherwise, I_i is the intensity of the emission, the sum is over all decay chains for the radionuclide, and d is the probability of the chain occurring. The probability of detecting a count in the FEP with TCS is the sum of two terms: 1) detecting the full energy of the gamma ray of the transition of interest and no energy from any other transition in the decay chain. 2) the probability of detecting two or more gamma or X-rays that sums into an energy that is close enough to the energy of the gamma ray of interest that can't be separated, additionally no energy from any other transition is detected. The first term is called summing out because it removes counts from the full-energy peak and the second term is called summing in because it adds counts to the full-energy peak. The probability of detecting a count from summing out for a chain is

$$P_{SumOut} = c_i \varepsilon_i d_j \prod_{\substack{k=1,\\ k \neq i}}^{n} \left(1 - P_{E_k}\right) \tag{2}$$

where P_{E_k} is the probability that any energy is detected from transition k. The probability of detecting a count from summing in of multiple gamma rays is

$$P_{SumInGamma} = \prod_{j=1}^{k} c_j \varepsilon_j \left(\sum_{l=1}^{n} \delta_{kl} d_l \prod_{\substack{o=1,\\ o \neq j,k}}^{m} \left(1 - P_{E_o}\right) \right) \tag{3}$$

where the first product is over the gamma rays that sum to the energy of the gamma ray of interest and P_{E_o} is the probability of detecting any energy from transition o. The most common case for this is from cross-over transitions where the gamma ray of interest skips levels and the summing gamma rays are from the skipped levels. However, there are cases where the summing gamma rays are from other parts of the radionuclide decay. It is also possible that one or more of the photons summing into the energy of the transition of interest is from an X-ray. This case is slightly different because an X-ray can be emitted from any transition. The probability then becomes

$$P_{SumInXray} = \prod_{j=1}^{n} c_j \varepsilon_j \left(\sum_{\substack{k=1,\\ k \notin n}}^{m} \left(g_k \varepsilon_k \prod_{\substack{l=1,\\ l \neq k}}^{o} \left(1 - P_{E_l}\right)\right) \right) \tag{4}$$

where g_k is the probability that X-ray is emitted from the transition k. For k X-rays the probability of registering a count in a k X-ray full energy peak with TCS are

$$P_{SumOutXray} = d_j f_i \varepsilon_i \sum_{l=1}^{n} \left(h_l \prod_{\substack{k=1 \\ k \neq j}}^{n} \left(1 - P_{E_k}\right) \right) \tag{5}$$

where f_i is the probability of emitting the k X-ray from transition i and h_l is the probability of creating a k-shell vacancy from the transition l. The algorithm sums over all possible chains in the decay. Next, we need to define the probability of detecting any energy from a transition. For α and β^- decays the detection probability is assumed to be 0. For internal transitions the probability is

$$P_{IT} = \frac{1}{1+\alpha_t}\left(\eta_t + \alpha_k \sum_{i=1}^{n} P_i \eta_i + \alpha_{L_1} \sum_{j=1}^{m} P_j \eta_j + \alpha_{L_2} \sum_{k=1}^{o} P_k \eta_k + \alpha_{L_3} \sum_{l=1}^{p} P_l \eta_l\right) \tag{6}$$

where η is the total efficiency for the gamma or X-ray emission, α is the internal conversion coefficient either total or for the k or l subshells. For electron capture and β^+ transitions the probability of detecting any energy is

$$P_{\beta^+} = 2\beta_{\beta^+}\eta_{511} + \beta_{EC} \left(\beta_k \sum_{i=1}^{n} P_i \eta_i + \beta_{L_1} \sum_{j=1}^{m} P_j \eta_j + \beta_{L_2} \sum_{k=1}^{o} P_k \eta_k + \beta_{L_3} \sum_{l=1}^{p} P_l \eta_l\right) \tag{7}$$

where β_{β^+} and β_{EC} are the β^+ and electron capture branching ratios, the β_k and β_{l_x} are the k and l subshell electron capture probabilities, and the P s are the emission probabilities of the X-rays. The factor of 2 for the β^+ contribution stems from the two photons emitted from the positron annihilation. Finally, the correction factor is calculated for ~ 1000 points randomly sampled from the volume source; the TCS correction factor for the sample is the ratio of the sum of the probabilities with TCS to the sum of the probability without TCS:

$$CF = \frac{\sum_{i=1}^{n} P_{TCS_i}}{\sum_{i=1}^{n} P_{noTCS_i}} \tag{8}$$

Experimental

Measurements of radionuclide standards that exhibits TCS were performed at PNNL using the Gamma Alpha Beta Radio-Isotope EvaLuator (GABRIEL) [20] instrument shown in Fig. 1. The GABRIEL instrument consists of two BEGe5030 detectors combined with a 20 mL plastic liquid scintillation vial containing the source, together with two PMTs sandwiched between the detectors. Data is acquired in list-mode using a digitizer employing pulse height analysis firmware for the gamma detectors and a digitizer employing charge integration and pulse shape discrimination firmware for the acquisition of the PMT signals. This configuration

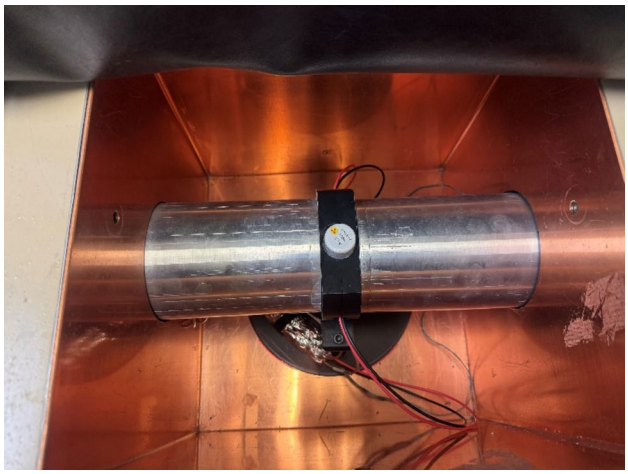

Fig. 1 Detector setup used for measuring TCS correction factors

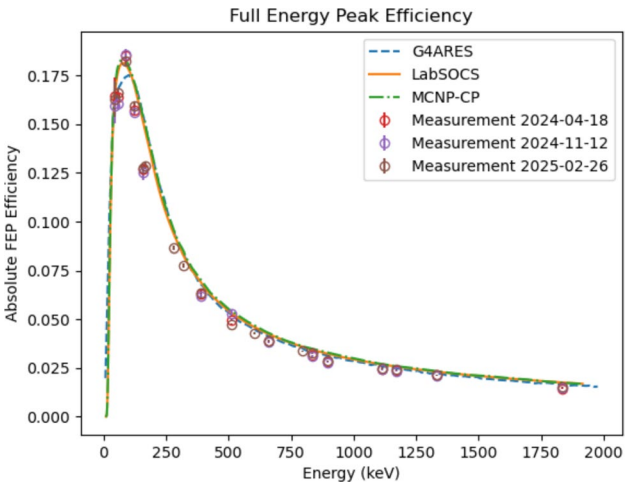

Fig. 2 FEP efficiencies measured and calculated using G4ARES, ISOCS/LabSOCS, and MCNP-CP

is more complicated to model compared to normal sample counting with a single detector. Measurements were performed of a mixed gamma calibration standard for efficiency calibration and for single radionuclide standards of Sb-125, Ba-133, Cs-134, Eu-152, Eu-154, Gd-153, Ho-166 m, and Th-229 for establishing measured TCS correction factors.

Efficiency calibration measurements were performed using a NIST-traceable mixed gamma source in April and November of 2024 and February of 2025. Gamma emissions spanning the energy range 46.5 to 1836 keV were used for the efficiency calibration. Figure 2 shows the measured FEP efficiencies together with the efficiencies calculated using ISOCS/LabSOCS, G4ARES and MCNP-CP. The mixed gamma sources contained Co-57, Te-123 m, Cs-139, Y-88, and Co-60 which require TCS corrections for the measured FEP efficiencies. The corrections were performed using the

correction algorithm in Genie 2000 v3.4.1 [1]. Te-123 m is not present in the TCS library in Genie 2000 v3.4.1 and is therefore not corrected for TCS in this efficiency calibration. TCS correction factors for emissions close to the 159 keV emission can therefore be overestimated. The single radionuclide standards were analyzed using this measured efficiency calibration and the activities were reported without a TCS correction. The correction factor was then determined by taking the ratio of the calculated activity and the certified activity of the standard. The 1-sigma uncertainty quoted on all the calibration standard certificates was 1.2%, and the peak area uncertainty was about 1% for most emissions, and the sample positioning repeatability was about 2% which gives a combined 1-sigma uncertainty of about 2.5% in the best measurements.

Monte-Carlo simulations

Monte-Carlo radiation transport simulation methods differ from the method developed and presented here. Transport-based methods rely on tracking individual particles through a geometry and tallying the energy deposited in the active volume of the detector crystal. The TCS correction factors can be calculated by comparing two separate tallies, either conducted in the same simulation or in two different simulations. One where all particles from a single decay are tracked in the same event and one simulation where they are tracked in separate events. Monte-Carlo calculations of TCS can also include angular correlations between emitted gamma rays from the decay. The full measurement setup was modeled in Geant4 v10.7.4 with RadioactiveDecay5.6 and PhotonEvaporation5.7 nuclide data libraries [3]. This model was developed using the G4ARES (Geant4 Advanced Radio-Emission Corrections) framework [4]. Typical statistical uncertainties of the correction factors calculated by G4ARES and MCNP-CP are between 0.1 to 1%. The full model can be seen in Fig. 3. is the G4ARES model included much more detail compared to a simplified model used for the ISOCS/LabSOCS and MCNP-CP calculations shown in Fig. 4.

Results and discussion

Figures 5, 6, and 7 show the measured and calculated correction factors for the eight single-radionuclide standards. All correction factors and their uncertainties are tabulated in the supplementary information for this article. For emissions with an energy outside the energy range of the efficiency calibration or where the statistical uncertainty in the measurement was too large, the ratio was compared to G4ARES instead of the measured correction factor.

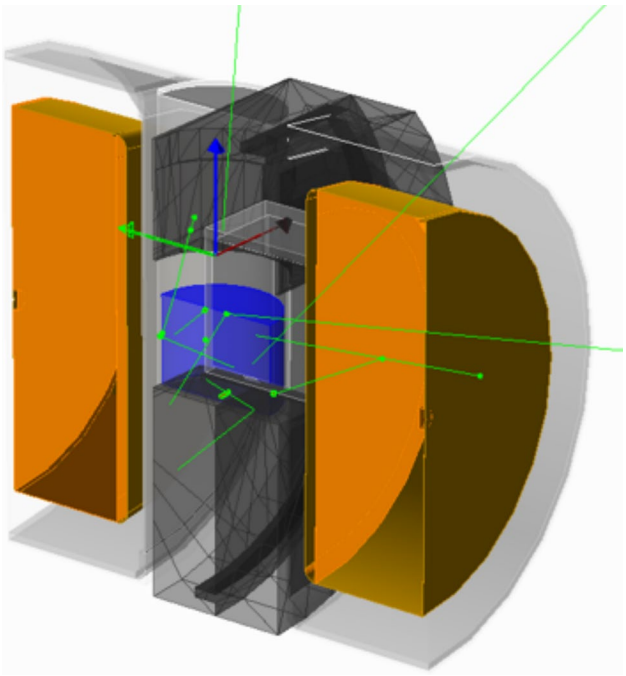

Fig. 3 Cross section of the Geant4 model of the measurement setup. The green lines show the tracks of individual gamma rays through the geometry

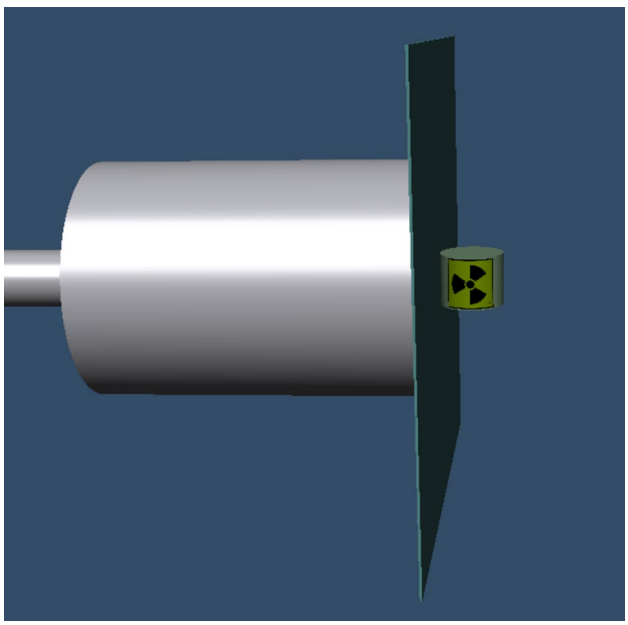

Fig. 4 Simplified model of the measurement setup used in ISOCS/LabSOCS and MCNP-CP

The library in Genie v3.4 does not contain all emissions. A correction factor cannot be calculated for the missing emissions and the ratio for Genie v3.4 is removed from the figures.

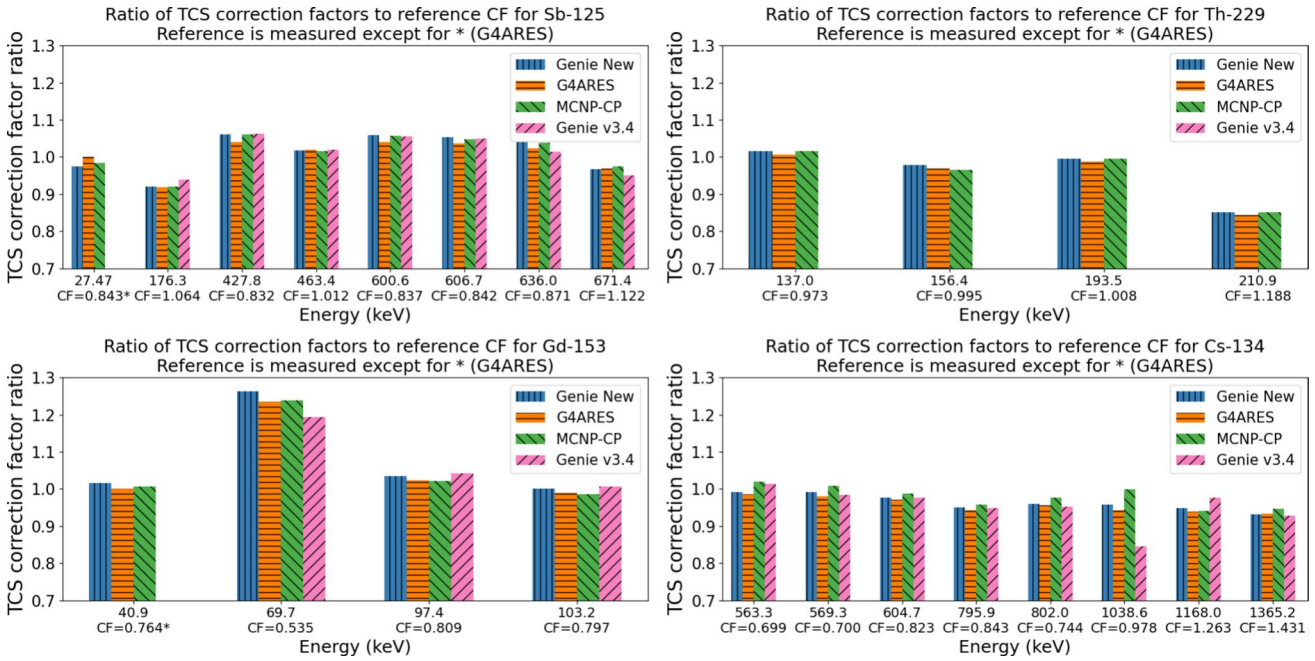

Fig. 5 The ratio of the modeled to measured correction factors for Sb-125, Th-229, Gd-153, and Cs-134. The x-axis indicates the energy of the emission and the value of the reference TCS correction factor. For emissions marked with an asterisk the measured correction factor is not available, and the ratio is to the correction factor calculated by G4ARES instead. When an emission is missing from the library for the Genie v3.4 algorithm the bar is missing from the figure

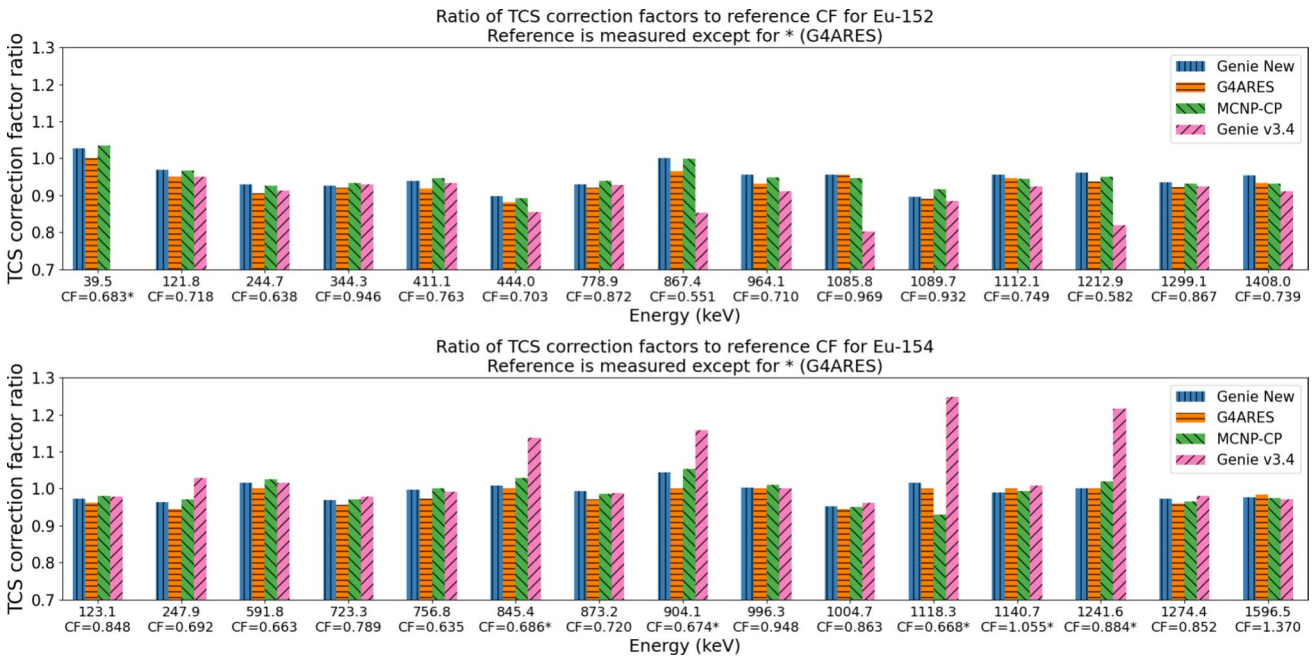

Fig. 6 The ratio of the modeled to measured correction factors for Eu-152 and Eu-154. The x-axis indicates the energy of the emission and the value of the reference TCS correction factor. For emissions marked with an asterisk the measured correction factor is not available, and the ratio is to the correction factor calculated by G4ARES instead. When an emission is missing from the library for the Genie v3.4 algorithm the bar is missing from the figure

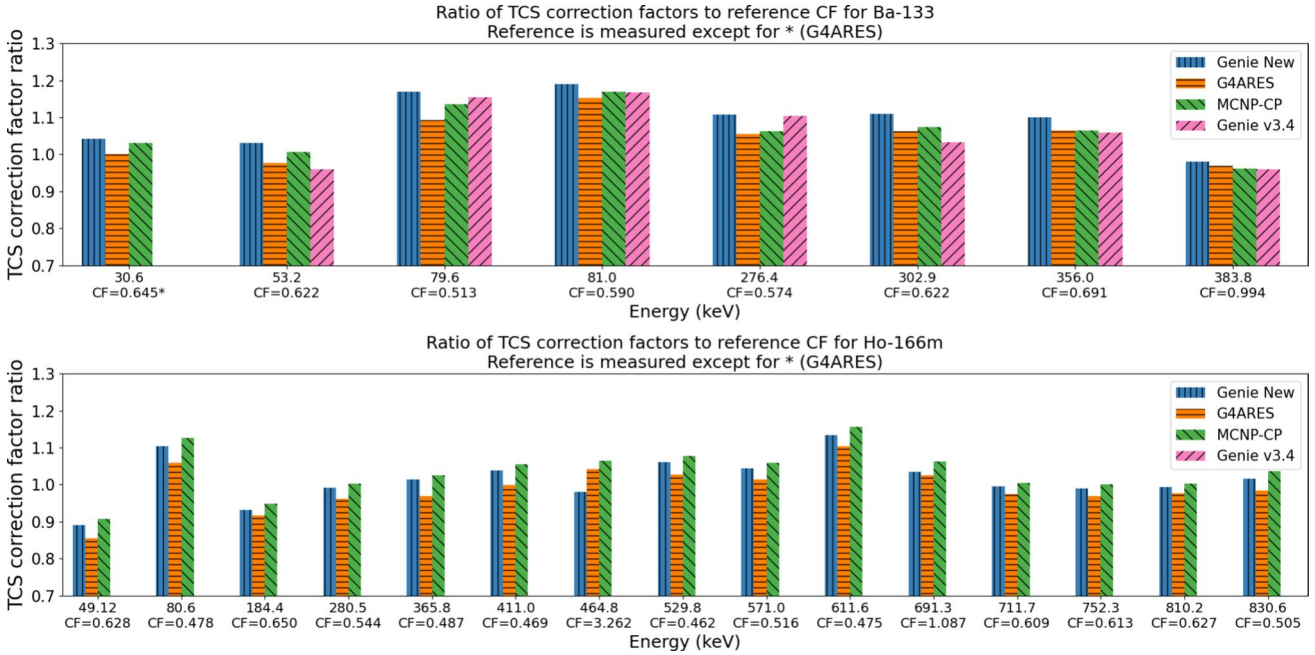

Fig. 7 The ratio of the modeled to measured correction factors for Ba-133 and Ho-166 m. The x-axis indicates the energy of the emission and the value of the reference TCS correction factor. For emissions marked with an asterisk the measured correction factor is not available, and the ratio is to the correction factor calculated by G4ARES instead. When an emission is missing from the library for the Genie v3.4 algorithm the bar is missing from the figure

The first conclusion is that the calculated correction factors for both k X-rays and gamma-rays from the four different algorithms are within 3% of each other. For Ba-133 and Ho-166 m a few emissions can differ with up-to 5–7% between the new Genie algorithm and G4ARES. The likely explanation is that G4ARES includes the second detector and angular correlations. The exceptions are a few low intensity emissions for the Genie v3.4 algorithm where the difference can be over 10%. This demonstrates one of the limitations of this algorithm. The measured and calculated TCS corrections agree within the uncertainties for most emissions for Sb-125, Cs-134, Eu-154, and Th-229. These radionuclides decay with β^- or α (Th-229) and have small internal conversion coefficients. Gd-153 decays via electron capture and the agreement are within 2% for the two most intense emissions at 97 and 103 keV. The modeled correction factor is overestimated for the 69 keV emission for all models.

Sb-125

The transitions from the two lowest excited states in Sb-125 have small contribution to TCS because of the long half-life of the second excited state in Sb-125. A measured correction factor above 1 for the 176 keV emission for Sb-125 is unexpected and not reflected in any of the modeled correction factors because there are no gamma rays emitted that could sum into this energy. The 176 keV emission is close to the Te-123 m emission at 158 keV that was not corrected for TCS in the efficiency calibration and this is part of the explanation of the larger than expected measured correction factor.

Ba-133

The agreement between the measured and calculated TCS corrections is slightly worse for Ba-133 but on the order of the uncertainties for most emissions. Ba-133 decays via electron capture and the X-ray and gamma ray emissions have high total efficiencies. This makes the TCS correction large, which is reflected in a correction factor that is significantly lower than 1. The contribution from scatter in the second detector that is only modeled in the G4ARES modeled is largest for the energy range emitted from Ba-133 and most likely explains the majority of could explain the differences between the modeled correction factors. One of the most intense gamma-lines emitted by Ba-133, 81 keV, is not always discernably separable from the smaller 79.6 keV emission. In this study, high resolution BEGe detectors were used which allowed for both emissions to be visible but the proximity and multiplet structure of these two gamma-lines increased the overall uncertainty in the correction factor.

Eu-152

Eu-152 decays with both electron capture and β^-, has a complex decay schema, and has high internal conversion factors. This makes it a challenging radionuclide to calculate the correction factor for. The correction factor ranges from nearly one and all the way down to 0.55. The modeled correction factors agree to within a few percent of each other while the measured and calculated correction factors differ by as much as 10%. It's currently unclear why the agreement varies between the emissions.

Ho-166 m

Ho-166 m decays by β^-, has a complex decay scheme, and high internal conversion coefficients; it is also challenging to calculate the correction factor for. Most emissions have large corrections, and the correction factor can be as low as 0.5 and as high as 3.2. Ho-166 m also contains an example of two gamma emissions that sum into a non-crossover gamma emission. The sum of the energies from the 184.411 keV and 280.464 keV is only 43 eV from the 464.832 keV emission. The 464.832 keV emission originates from the 1376 keV excited state while the 184.411 and 280.464 keV emissions originates from the 265 and 545.5 keV excited states, respectively. All three calculation methods can reproduce the measured correction factor within the uncertainties for this emission. Ho-166 m is the only radionuclide in the set for which the emitted X-rays are within the energy range of the efficiency calibration and the calculated factor for all models for this X-ray is about than 10% lower than the measured correction factor. Overall, the calculated correction factors are within 5% except for the 80.6 keV, 184.4 keV and 611.6 keV emissions, which are within 10% of the measured correction factors. There is a note in the ENSDF record that the intensity of the 80.6 keV emission appears to be too low, which could be a possible reason for the difference between the calculated and measured correction factors.

Overall, the G4ARES calculation generally gives higher correction factors compared to the new Genie algorithm and MCNP-CP. Angular correlation of the emitted gamma rays could be the reason for this. Angular correlation is included in G4ARES, but it is not included in the Genie algorithm.

Conclusions

True coincidence summing correction factors have been measured for eight radionuclides and compared to the new TCS correction algorithm for consideration in future releases of Genie software, as well as Monte-Carlo simulations using the G4ARES framework based on Geant4, MCNP-CP and the algorithm in Genie v3.4. All models perform well with most correction factors within the uncertainty of the measurement. Best performance for all models was achieved for radionuclides decaying via β^- and low internal conversion coefficients. The new proposed Genie algorithm performs as well as or better than the previous Genie algorithm. The most notable differences are its ability to calculate correction factors for more radionuclides and X-rays, and the better performance for low intensity emissions. G4ARES has the most realistic detector model, including both detectors and the PMTs; it also includes angular correlations between emitted gamma-rays. This could explain the differences observed between the calculation methods, in particular for Ba-133 and Ho-166 m. Improvements in these methods offer opportunities for further refinement to nuclear data by studying detector response predictions like TCS correction tools to measured results. Refinements like this are necessary to further reduce the uncertainty limitations, precision, and accuracy of non-destructive gamma-ray analysis.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10967-025-10534-z.

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Declarations

Conflict of interest Henrik Persson and Kara E. Phillips are employees of Mirion Technologies.

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