Monte Carlo modelling of live-timed anticoincidence (LTAC) counting for Cu-64

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Abstract

The radionuclide copper-64 is a promising candidate for nuclear medicine, but its complex decay creates challenges in the primary standardization of its activity. Monte Carlo simulations of live-timed anticoincidence (LTAC) counting of $^{64}$Cu were used to calculate corrections to extrapolation intercepts, resulting in improved activity determinations. A small correction (-0.33 %) to the linear extrapolation of LTAC data acquired with a $\gamma$-gate over the 1346 keV gamma peak was determined. We discuss the physical origin of the correction. We also use experimental data to demonstrate a Monte Carlo scaling that allows for inclusion of data acquired with a $\gamma$-gate set over the annihilation photon peak(s).

Keywords: Beta-gamma coincidence method; anticoincidence; liquid scintillation; Monte Carlo simulations
Highlights

- Monte Carlo simulations of $^{64}$Cu LTAC found a 0.33 % correction to the linear extrapolation.
- The intercept of a linear extrapolation is insensitive to some model deficiencies.
- A Monte Carlo scaling approach allows the use of data acquired with a γ-gate set over the annihilation photon peak(s).
1. Introduction

Copper-64 decays by positron emission, beta decay, and electron capture with a half-life of 12.7004(20) h (Bé et al., 2011). Interest in $^{64}$Cu stems mostly from its promise as a “theranostic” radionuclide (Smith, 2004). Copper nimbly coordinates to a vast assortment of complexing agents, allowing for very specific chemical targeting of biologically active sites. Its $\beta^-$ branch ($\approx 17.5 \%$) allows for quantitative imaging by positron emission tomography (PET), while its $\beta^-$ and $\beta^+$ branches ($\approx 56 \%$) provide significant local therapeutic dose.

The National Institute of Standards and Technology (NIST) recently performed a primary standardization of $^{64}$Cu activity (Bergeron et al., 2017). The standard was realized by live-timed anticoincidence (LTAC) counting, gating on the 1346 keV gamma peak as described by Kawada (1986) and others (Wanke et al., 2010; Sahagia et al., 2012; Bé et al., 2012; Havelka and Sochorová, 2014). The very small emission probability of the 1346 keV $\gamma$-ray (0.4748 %; Bé et al., 2011) requires longer-than-typical count times and makes the measurement somewhat difficult. Furthermore, the experiment relies on an inefficiency determined with the minority electron capture branch of a complex decay scheme. If the difference in K/L/M electron capture ratios is not considered, then the intercept can be expected to be slightly biased (Funck and Nylandstedt Larsen, 1983; Fitzgerald et al., 2015).

Recently, the value of Monte Carlo simulations to determine corrections to coincidence extrapolations has become well-established (Dias et al., 2006; Dias et al., 2013; Bobin et al., 2016; Fitzgerald 2016). Herein, we describe Monte Carlo simulations of $^{64}$Cu activity determinations by LTAC at NIST. The simulations predict that a small correction to the
extrapolated intercept is required. In addition to calculating extrapolation corrections, we applied a scaling method similar to that described by Dias et al. (2013). We show that the Monte Carlo simulations may be used to achieve a more accurate and potentially more precise activity for $^{64}$Cu by LTAC.

2. Methods

The $4\pi\beta-\gamma$ anticoincidence apparatus used in this work (Lucas, 1998; Fitzgerald and Schultz, 2008) contains a $4\pi$ liquid scintillation (LS) detector consisting of a hemispherical LS vial optically coupled to a single photomultiplier tube. The LS source sits inside a NaI(Tl) well detector, which provides the $\gamma$-ray channel. Events from both detectors are stored in list mode as time-amplitude pairs, which are then analyzed offline using the live-timed anticoincidence method (Bryant, 1962; Baerg et al., 1976; Fitzgerald et al., 2015) with adjustable LS thresholds, $\gamma$-ray gates, and extending dead-times.

The $^{64}$Cu data were acquired as part of a larger set of measurements, spanning multiple $^{64}$Cu solutions and multiple methods to be reported separately (Bergeron et al., 2017). For the present paper a small, representative subset of those data was considered.

The Geant4-based (Agostinelli et al., 2003) Monte Carlo simulation model (Fitzgerald, 2016) of the apparatus and analysis method was modified for the present work. The scintillation photon yield was adjusted to 5000 MeV$^{-1}$ with the Birks parameter, $kB$, set to 0.008 cm MeV$^{-1}$ (previously found to be appropriate for the Ultima Gold scintillation cocktail (PerkinElmer,
Waltham, MA)\(^1\) used here) in order to match the shape of the simulated LS spectrum to the experiment. The NaI simulation was modified to account for the non-linear energy response of the detector (Knoll 2000, p. 333), which is important for reproducing the relative positions of the \(^{64}\text{Cu}\) \(\gamma\)-ray peak and the positron annihilation sum peak. To accomplish this, the energy of the \(^{64}\text{Cu}\) \(\gamma\)-ray in the simulation was lowered to 1312 keV, thereby matching the peak positions in the simulated and experimental spectra (Figure 1). Additionally, pulse pileup was included in the simulation assuming a pileup resolving time of 1 \(\mu\)s. The experimental and simulated spectra are shown for the LS and NaI(Tl) detectors in Figure 1a and 1b, respectively, along with the various components of the simulated spectra.\(^2\)

Three NaI(Tl) gates, G1, G2, and G3, were set in both the experiment and simulation around the 511 keV, 1022 keV, and 1346 keV peaks, respectively. The ratio of anti-coincident to total counts in each gate gave the inefficiency parameters \(Y_1\), \(Y_2\), and \(Y_3\), used for activity determinations. Previous standardizations of \(^{64}\text{Cu}\) have followed Kawada (1986), plotting \(N_{\text{LS}}\) vs. \(Y_3\) and taking the activity \((N_0)\) to be the intercept \((N_{\text{LS}}(0))\) using a linear extrapolation (Wanke et al., 2010; Sahagia et al., 2012; Bé et al., 2012; Havelka and Sochorová, 2014),

\[
N_{\text{LS}} = N_0(1 - Y_3). \tag{1}
\]

\(^1\) Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

\(^2\) The very low emission probability for the 1346 keV \(\gamma\)-ray incentivized simulating each branch separately and then combining the results in a spreadsheet. This approach simplified the analysis of branch-specific contributions. Furthermore, simulations including all branches required \(> 50 \cdot 10^6\) histories to remove statistical artifacts in the \(N_{\text{LS}}\) vs. \(Y_3\) plots.
Using this extrapolation without further correction ignores the difference in K/L/M electron capture ratios between the two electron capture branches, which is expected to cause a bias in the intercept (Funck and Nylandstedt Larsen, 1983). This approach also ignores any effect of the positron or beta branch inefficiencies being non-linear functions of $Y_3$. Further, the pure extrapolation ignores photon contributions to $Y_3$ from annihilation in flight of the ($E_{\beta^+, \text{max}} = 653$ keV) positron, for which Kossert, et al. (2014) estimated a probability of 0.6 % in a similar LS cocktail.

In this work, the determination of source activity follows Fitzgerald (2016) with some improvements. Two basic methods were used. In the first method, which we call “correction”, the data were extrapolated using one or more of the 3 NaI(Tl) gates. In the case of a multi-gate fit, we define an effective inefficiency parameter,

$$Y_{\text{eff}} = \sum_{i=1}^{3} a_i Y_i$$

where the sum of the weighting parameters $a_i$ is normalized to 1. An extrapolation was then performed, replacing $Y_3$ in Equation 1 with $Y_{\text{eff}}$. Then, a correction factor to the activity, $F$, was determined by repeating the experiment in the Geant4 simulation in which the “activity” in the simulation was fixed to 1 and the intercept of the extrapolation of the simulated data$^3$ is $\tilde{N}_0$.

$$F = \frac{1}{\tilde{N}_0}$$

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$^3$ Symbols topped with a *tilde* represent simulated values.
The corrected activity is then determined by

\[ N'_0 = F N_0 \]  

(4)

The application of the correction method to $^{64}$Cu data is described in Section 3.1.

The second method, which we call “scaling”, was to scale the simulated $\tilde{N}_{LS}$ by a factor $S$ to match the experimental data. The activity is then given by $S$. To accomplish this, a spline fit of the simulated $\tilde{N}_{LS}$ vs. $\tilde{Y}$ data was used to interpolate between points to estimate $\tilde{N}_{LS}(Y)$. A least-squares fit was then performed of $S\tilde{N}_{LS}$ vs. $N_{LS}$ with $S$ as the only free parameter. In addition to this single parameter scaling (1PS) approach, a variant scaling method was implemented in which the $a_i$ weighting parameters were allowed to vary along with $S$. In practice, the least squares minimizations tended to result in $a_1 = 0$ so that only the $a_2: a_3$ ratio and $S$ actually needed to be scaled; the result was a two-parameter scaling (2PS) approach.

3. Results and Discussion

3.1. Correction methods

Linear and quadratic extrapolations were carried out with a variety of $a_i$ values and $E_{LS}$ domains.

3.1.1. $Y_1$ and $Y_3$ extrapolations

Figure 2 shows simulated data for G1 and G3 over the full range of $E_{LS}$, illustrating the ranges over which $N_{LS}$ varies linearly with $Y_i$, showing where discontinuities emerge as counts from $\beta^{+/-}$. 
or electron capture events dominate the slope, and giving a feel for the elements being combined
when $Y_{\text{eff}}$ are considered (*vide infra*).

Table 1 shows the results of linear and quadratic extrapolations with data from two versions of
the Monte Carlo simulation for the $Y_1$ and $Y_3$ extrapolations. The 2016 version lacks the
modifications accounting for the non-linear energy response of the detector included in the 2017
version (see Section 2). As Figure 3a demonstrates, the 2016 simulation does not reproduce the
slope of the $N_{\text{LS}}$ vs. $Y_3$ plot. It is a testament to the robustness of the classical extrapolation
 technique that the $N_0$ values from the two models are so similar.

The intercepts from high-$E_{\text{LS}}$ $Y_1$ extrapolations have sometimes been used to estimate $(P_{\beta^+} +
P_{\beta^-})$ or to calculate an activity (Christmas et al., 1983; Sahagia et al., 2012). Our simulations
(using DDEP values for $P_{\beta^+}$ and $P_{\beta^-}$) are consistent with the findings of Sahagia et al. (2012),
Wanke et al. (2010), and others in finding that the $Y_1$ extrapolations do not provide a reliable
measure of $(P_{\beta^+} + P_{\beta^-}) = 0.1752 + 0.3848 = 0.5600$ (data from DDEP, Bé et al., 2011). While
the annihilation peak data are not useful for extrapolation, we show in Section 3.2 that they can
give good results with scaling approaches.

A linear fit to the low-$E_{\text{LS}}$ data vs. $Y_3$, resulted in an intercept uncertainty of 0.08 %. However, a
small oscillatory trend was found in the residuals. Performing the same extrapolation with the
2017 simulation gives the same trend in the residuals and produces an intercept of 1.0033 (see
Table 1), leading to $F = 0.9967$. 
It was expected that $F < 1$ due to the difference in K/L/M electron capture ratios between the capture branch to the ground state ($g$), and the branch to the 1346 keV state ($e$). The latter branch is the one whose inefficiency is being measured by $Y_3$, yet the former branch contains 99\% of the electron capture decays. The correction factor for this effect, assuming only K-events are detected during the extrapolation and considering only the electron capture branches (excluding positron and beta), is (Funck and Nylandstedt Larsen, 1983; Chauvenet et al., 1987; Fitzgerald et al., 2015)

$$F = \left[ 1 + g \frac{P^g_K - P^e_K}{P^e_K} \right]^{-1} \quad (5)$$

This amounts to $F = 0.9955(22)$, where the uncertainty is assuming uncorrelated uncertainties in $P^e_K$ and $P^g_K$ taken from DDEP (Bé et al., 2011). If the extrapolation were mostly over L-capture events, then $F \rightarrow 1$. In practice, L-events make a small contribution to the extrapolation (see Figure 1) and the presence of the positron and beta branches also push $F$ toward 1.

To further understand the cause of the simulated $F$, we ran the simulation for subsets of the decay scheme. The results are shown in Table 2 and indicate that branch $g$ is the largest contributor to the correction. Simulating branch $e$ by itself and using Equation 3, produced $F = 0.9944 (5)$, which is in good agreement with the result from Equation 5. Here, only the uncertainty from the extrapolation is listed, though the total uncertainty would include that from the nuclear data (0.0022).

3.1.2. $Y_{\text{eff}}$ extrapolations
The extrapolation data are shown in Figure 3a and residuals from linear fits are shown in Figure 3b. When all three $a_i$ values were allowed to vary in a multi-parameter extrapolation, it was found that having both $a_1$ and $a_2$ was redundant. In most cases, the resulting least-squares fit produced $a_1 \ll a_2$, and eliminating $a_1$ from the fit had no consequence. Traditionally, it is ill-advised to extrapolate over a non-smooth region of $N_{LS}$ vs. $Y$ (Baerg, 1965), however this was also attempted as a stringent test of our analysis methods.

The residuals of a linear fit were minimized with $a_1 = 0.47$ and $a_2 = 0.53$ (Figure 3). With these coefficients, linear and quadratic fits to the simulation data indicated respective values for $F$ of 0.9789 and 0.9660 (Table 3).

3.2. Scaling methods

The scaling methods were applied to the same data as the extrapolation correction methods. The single-parameter scaling (1PS) method was applied with $Y_1$ and $Y_3$ using the full range of experimental data and using the data subsets over which the inefficiency plots are nearly linear (see Figure 2). The linear ranges for $Y_1$ and $Y_3$ were the same as for the correction methods (see Table 3). Table 4 shows the 1PS results for both the 2016 Monte Carlo and the improved version. The 1PS using the $Y_1$ data performs reasonably well over the linear subset with both models. The 2016 model does not perform as well with the $Y_3$ data, as expected based on the slope mismatch evident in Figure 3a. Neither model achieves acceptable $S_{1PS}/N_0$ (the ratio of the activity determined by 1PS and by uncorrected extrapolation of the experimental data) when using the full range of experimental data, which is not surprising given that $\chi^2$ ranged from 0.01
(for $Y_1$ with the 2017 model) to 0.21 (for $Y_3$ with the 2016 model). With the near-linear subsets, $\chi^2$ ranged from $4 \cdot 10^{-7}$ (for $Y_1$ with the 2017 model) to $7 \cdot 10^{-4}$ (for $Y_3$ with the 2016 model).

While the single-gate 1PS method was only effective over a limited range of $Y_i$, we found that the full range of data could be used in a multi-gate scheme. The 1PS method was implemented with several $a_i$ weighting coefficients and in this way, the dependence of the recovered activity on the $Y_{\text{eff}}$ weighting was established. All of the $Y_{\text{eff}}$ minimizations gave activities closer to the pure linear extrapolation ($S_{1\text{PS}}/N_0$ closer to 1) and lower $\chi^2$ than any pure $Y_i$ ($i = 1, 2, 3$). The lowest values of $\chi^2$ corresponded to $S_{1\text{PS}}/N_0$ very slightly less than 1 (see Supplemental Figure S1).

We also allowed the $a_i$ weighting coefficients to vary along with $S$ (see 2PS entries in Table 5). For the 2017 model the least squares minimization resulted in $a_1 = 0$ and $a_2/a_3 = 2.75$ and gave $S_{1\text{PS}}/N_0 = 0.99963(6)$, where the expressed uncertainty is the sum of the squares of the fit residuals ($N = 22$ efficiency points). For the 2016 model, the minimization resulted in much heavier weighting of $Y_2$ and $S_{1\text{PS}}/N_0$ farther from 1. Dramatic improvement can be seen in Table 5 when moving from the 2016 model to the 2017 model. For the single-parameter scaling method, about 80% of that improvement was due to the inclusion of the non-linear energy response of the NaI(Tl), with the rest due to the inclusion of pileup.

Figure 5 shows the fits and residuals associated with the 1PS-S calculations with the 2016 and 2017 models and the 2PS calculations with the 2016 model. The 2016 2PS and 2017 1PS-$N_0$ residuals are small, but definitely show some trending.
In another exercise, $S_{1PS}/N_0$ was set to 1 and the $a_i$ weighting coefficients allowed to vary (see 1PS-$a_i$ entries in Table 5). For the 2017 model, $a_2/a_3$ was very similar to the one recovered in the 1PS-$N_0$ case, with marginally larger $\chi^2$. Fit uncertainties were consistently smaller with $a_i$ that gave $S_{1PS}/N_0$ close to 1 (Figure S1).

One of the largest uncertainty components in the NIST LTAC standardization (Bergeron, 2017) was “model dependence”, estimated by considering several factors, the largest of which was the difference between a linear and a quadratic extrapolation. As Table 3 shows, applying the Monte Carlo correction factors brings the linear and quadratic extrapolations into much better agreement.

We considered that the Monte Carlo 1PS approach might allow for lower total combined uncertainty. Since the $G_1$ and $G_2$ include many more counts than $G_3$, we might also expect improved uncertainty on the determination of $Y$ due to improved counting statistics. A direct comparison of the linear extrapolation and 1PS methods was made for a subset of 20 activity determinations. In the 1PS method, $a_2/a_3$ was set to 2.75, the value that gave the best $\chi^2$ for the averaged data (vide supra). For this subset, $S_{1PS}/N_0 = 0.9959$, with the relative standard deviations on 20 linear extrapolations ($N_0; s = 0.90\%$) significantly greater than on 20 1PS minimizations ($S_{1PS}; s = 0.54\%$). The fit uncertainties associated with both the linear extrapolation and the 1PS are negligible ($\approx 3\cdot10^{-4}\%$ and $\approx 6\cdot10^{-3}\%$, respectively) compared to other sources of uncertainty. It appears possible that the 1PS approach, using data from multiple
γ-gates, may yield a more precise $^{64}$Cu activity than the linear extrapolation using only the 1346 keV γ-gate.

The treatment of a subset of experimental data also allows us to comment on the agreement between the 1PS and extrapolation correction Monte Carlo approaches by providing us with a measure of the statistical uncertainty. Calculating the ratio of the activity determined by scaling to the activity determined by pure linear extrapolation of the $Y_3$ data ($S_{1PS}/N_0$) for each of the 20 data subsets gave an average value of 0.9959(15), where the uncertainty is the standard deviation of the mean for the 20 determinations of the ratio. Correcting each $N_0$ using the Monte Carlo-determined $F$ to give $\tilde{N}_0^T$, we could compare the two Monte Carlo approaches, finding $S_{1PS}/\tilde{N}_0^T = 0.9986(15)$. Within statistical uncertainties, the two approaches are consistent.

Table 3 allows a broader comparison of the correction and scaling approaches to Monte Carlo corrections. In general, the two approaches give results that are in agreement to within the fit uncertainties. The $Y_{\text{eff}}$-based correction methods give very large fit uncertainties as they attempt to fit a linear or quadratic function over a discontinuity. The other Monte Carlo approaches give activities in very tight accord. Even including the $Y_{\text{eff}}$-based correction results, the standard deviation on the seven methods is 0.25%. The corresponding uncorrected extrapolations carry a standard deviation of 2.01%. Of course, it is the uncorrected $Y_3$ extrapolation that is generally considered to be the most appropriate, making the 2.01% standard deviation something of a “straw-man”, set up to make the Monte Carlo corrections look good—which they do. The normalization of the values in Table 3 by $N_0$ provides a more relevant comparison of the potential impact of the Monte Carlo corrections. The average Monte Carlo-corrected activity is
0.9981·\(N_0\). The uncertainty on the correction would be similar in magnitude to the correction itself.

4. Conclusions

We have improved our Monte Carlo model of the NIST LTAC system to better account for the non-linear energy response of the NaI(Tl) detector and applied the new model to measurements of \(^{64}\text{Cu}\). We explored both extrapolation correction and scaling approaches. We calculated that a correction factor, \(F = 0.9967\), should be applied to the intercept acquired by gating over the 1346 keV \(\gamma\)-ray. This is understood in terms of the contributions from individual branches, and arises principally from the Funck correction (Funck and Nylandstedt Larsen, 1983). The small correction implied by our calculations implies a small but systematic bias in previous standardizations; its magnitude is smaller than typical stated uncertainties.

One remarkable conclusion that can be drawn from our calculations concerns the robustness of the traditional LTAC extrapolation approach against model deficiencies, as illustrated by the consistency of intercepts realized with two versions of our Monte Carlo model giving different \(Y_3\) v. \(N_{LS}\) slopes. In this instance, the different slopes came from the treatment of the energy response of the NaI(Tl) detector. Similar effects would be expected from other slight differences in conditions, such as gate settings or peak shifts during an experiment due to pileup or gain shifting. Our simulations show that the extrapolation should be stable against such effects. Further, applying the correction factor not only improves the accuracy of the result, but reduces the difference between linear and quadratic extrapolation intercepts.
We also considered scaling approaches, fitting the simulated data to the experiment by varying a parameter, $S$, equal to the activity. The scaling approach allows for the use of data acquired with a gate over the annihilation photon peak(s), improving counting statistics. The improved precision on the inefficiency determination results in reduced uncertainty due to counting statistics and as such may result in a slight improvement in the uncertainty on $^{64}$Cu activity. In the recent NIST primary standardization of $^{64}$Cu (Bergeron et al., 2017), the largest uncertainty component was the source-to-source variability (followed by model dependence). We do not expect the Monte Carlo scaling approach to significantly improve this component, and so its overall impact on the total combined standard uncertainty will be relatively small. We have shown that the veracity of the model itself can be assessed by comparing the shapes of the experimental and simulated extrapolation curves, revealing subtle effects such as non-linear energy response and pulse-pileup in the NaI(Tl) detector.

Acknowledgements

We are grateful to the NIST team responsible for the primary standardization of Cu-64, including Jeffrey Cessna, Leticia Pibida, and Brian Zimmerman.

References


Tables

Table 1 Results of linear and quadratic extrapolations carried out with data from the 2016 and 2017 versions of the LTAC Monte Carlo simulation. The fits were performed with $\tilde{N}_{lS}$ values estimated by spline for each $Y_i$. The extrapolations were performed over the regions $Y_1 \approx 0.34$ to $0.99$ and $Y_3 \approx 0.32$ to $0.79$. Experimental results, normalized by the intercept of a linear extrapolation of low $E_{lS}$ $Y_3$ data, are given for comparison.

<table>
<thead>
<tr>
<th>$\tilde{N}_0$ (2016)</th>
<th>$Y_1$</th>
<th>$Y_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>linear</td>
<td>0.4834</td>
<td>1.0020</td>
</tr>
<tr>
<td>quadratic</td>
<td>0.5174</td>
<td>0.9971</td>
</tr>
<tr>
<td>$\tilde{N}_0$ (2017)</td>
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<td>$Y_3$</td>
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<td>quadratic</td>
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<td>$Y_1$</td>
<td>$Y_3$</td>
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<tr>
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<td>1</td>
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<tr>
<td>quadratic</td>
<td>0.5199</td>
<td>0.9919</td>
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Table 2  Correction factors for various parts of the $^{64}$Cu decay scheme. In each case, the electron capture (EC) transition to the excited state ($e$) was included.

<table>
<thead>
<tr>
<th>Branch</th>
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<td>EC $e$</td>
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<td>EC $e$ + EC$g$</td>
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<td>EC $e$ + $\beta^+$</td>
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<td>ALL</td>
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Table 3  Results for 7 activity determinations using the same experimental data, covering two ranges of $E_{LS}$ (given in keV), and both the correction (Corr.) and scaling methodologies. All results are scaled by the uncorrected intercept ($N_0$) of the linear extrapolation of the $Y_3$ data. For the Corr. methods, $A = N_0$ and $A' = \bar{N}_0$. For the scaling methods, $A = S$. The stated uncertainties, $u_c$, are calculated from the fit residuals.

<table>
<thead>
<tr>
<th>Method</th>
<th>$a_2$</th>
<th>$a_3$</th>
<th>$E_{LS}$ Start</th>
<th>$E_{LS}$ End</th>
<th>Description</th>
<th>$(A/N_0)$ / %</th>
<th>$F$</th>
<th>$(A'/N_0)$ / %</th>
<th>$u_c$ / %</th>
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<tbody>
<tr>
<td>1</td>
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<td>9</td>
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<td>99.67</td>
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<td>AVE</td>
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<td></td>
<td></td>
<td>2.01</td>
<td>SD</td>
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Table 4  Single parameter scaling (1PS) results for the 2016 and 2017 versions of the LTAC Monte Carlo simulation. All results are normalized by the intercept of a linear extrapolation of low $E_{ls} Y_3$ data, are given for comparison.

<table>
<thead>
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<th>$S_{1PS}/N_0$</th>
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<td></td>
<td>$Y_1$</td>
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<tr>
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<tr>
<td>subset</td>
<td>1.0061</td>
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Table 5  Results of single parameter and two-parameter scaling (1PS and 2PS) applied with the 2016 and 2017 versions of the LTAC Monte Carlo simulation. In the 1PS-S approach, $a_2/a_3$ was fixed at 2.75, the value indicated in the 2PS minimizations. In the 1PS-$a_i$ approach, $S$ was set equal to $N_0$, the uncorrected intercept of the $Y_3$ extrapolation. In the 2PS approach, both $S$ and $a_i$ were allowed to vary.

<table>
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<th>$a_2 / a_3$</th>
<th>$\chi^2$</th>
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Figure captions

Figure 1  (a) Experimental (open circles) and simulated LS spectra normalized to give $P = 1$ at $E_{LS} = 0$. The K EC (blue long dashes), L EC (blue dots), $\beta^+$ (red dashed), and $\beta^-$ (green dot/dash) contributions to the total (bold gray line) are shown. The continuous shape of the EC spectra is a result of the low resolution of LS. (b) Simulated NaI(Tl) spectrum normalized to an arbitrarily set number of histories, $N$, with the same color scheme as in (a). The bright green boxes show the positions of the three $\gamma$-gates.

Figure 2  Simulated data for $Y_1$ and $Y_3$.

Figure 3  (a) Experimental (open black symbols) and simulated (closed red symbols) data used in extrapolations with $Y_{eff} = Y_3$ (circles) and $Y_{eff} = 0.47Y_2 + 0.53Y_3$ (diamonds). The blue stars are $Y_3$ data from the 2016 Monte Carlo model, showing the sensitivity of the slope to the model veracity. (b) Fit residuals from linear extrapolations to the data in panel a. A large trend is visible in the residuals for the $Y_{eff} = 0.47Y_2 + 0.53Y_3$ data, both in the experiment and simulation.

Figure 4  Activity results using extrapolation of the experimental data (open squares), simulation-correction of that data (solid red squares) and scaling of the simulation to the data (red circles). Clearly the Monte Carlo-based analyses are more consistent than the pure experimental extrapolations. Uncertainty bars are from least-squares
fit results, not shown for corrected data. Methods are described in Sections 3.1 and 3.2 and summarized in Table 3.

**Figure 5** Experimental $N_{LS}/N_0$ (open black circles) and simulated $\tilde{N}_{LS}/S$ (closed red diamonds) data for $Y_{\text{eff}}$ with (a) $a_2/a_3 = 2.75$ using the 2016 Monte Carlo simulation, (b) $a_2/a_3 = 19.1$ using the 2016 Monte Carlo simulation, and (c) $a_2/a_3 = 2.75$ using the 2017 Monte Carlo simulation. The weighting ratio used in panels (a) and (c) was found to be optimal from a 2PS minimization with the 2017 model, while the ratio in panel (b) was found to be optimal from a 2PS minimization with the 2016 model. In each panel, the residuals ($r = \frac{\tilde{N}_{LS}}{S} - \frac{N_{LS}}{S}$) are shown (gray triangles).

**Figure S1** Activities determined by scaling ($S_{xPS}$, where $x = 1$ or 2), normalized by the uncorrected linear extrapolation of $Y_3 (N_0)$, with different $Y_i$ weights. Open diamonds represent 1PS varying $S$ only with $Y_1 = 0$ (orange), $Y_1 = 1$ (green), and $Y_2 = 0$ (blue). The black star represents the results of 2PS, while the blue closed triangle shows fixed $S$ with varying $Y_i$ weights. The corresponding $\chi^2$ of the scaling fits are shown to the right. The best fits cluster around $S_{xPS}/N_0 \approx 1$. 

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