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A Proposed Methodology for Gamma Radiation Activity Correction Using Monte Carlo Simulation

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Abstract

This study presents a methodology for correcting gamma radiation activities in the MAPRP (US DOE's Mixed Analyte Performance Evaluation Program) proficiency test, focusing on water, hay, and soil samples. Utilizing the MCNP code, we simulated the response of a high-purity Germanium detector (HPGe) detector, emphasizing the importance of accurate detector geometry modeling. Our simulations addressed discrepancies between measured and calculated efficiencies, which exceeded 50% for low gamma energies and decreased with higher energies. By adjusting parameters such as crystal dimensions and dead layer thickness, we achieved a significant reduction in efficiency discrepancies to less than 8%. The recalculated activities, based on optimized detector specifications, showed good agreement with reference values, falling within the 20% acceptability threshold set by MAPEP. The findings underscore the efficacy of Monte Carlo simulations in improving the accuracy of environmental radioactivity measurements and highlight the potential of this approach in addressing gamma spectrometry issues. Our methodology offers a robust solution for correcting unacceptable and warning results, ensuring reliable and precise activity estimations.

Keywords: HPGe detectors, Gamma spectrometry, Monte Carlo simulation, Detector efficiency

1. INTRODUCTION

The National Center for Energy and Nuclear Sciences and Technique CNESTEN, a prominent institution in Morocco, plays a critical role in planning and implementing comprehensive environmental monitoring programs. These programs cover a wide range of radioanalytical techniques, including alpha and gamma spectrometry and gross alpha-beta counting. HPGe-based detectors are frequently used for environmental radioactivity measurements due to their notable sensitivity and energy resolution. However, the experimental determination of the detector's response requires standardized sources containing a mix of radionuclides in the same counting geometry as the measured sample [1]. Following ISO 17025 standards, CNESTEN routinely participates in proficiency tests organized by international organizations such as IAEA and MAPEP [2]. These tests yield varied results—acceptable, acceptable with a warning, or unacceptable—contributing to the ongoing pursuit of quality in environmental radioactivity assessments [3].

Monte Carlo-based codes have emerged as valuable tools for determining the full energy peak efficiency of HPGe detectors due to their ability to simulate the detection process by tracking particles through a sensitive volume. However, discrepancies between Monte Carlo code efficiency results and experimental values have been reported in the literature [4–7]. These differences are often attributed to incorrect information about crystal sizes provided by manufacturers.

Adjustments to detector characteristics are necessary to reconcile Monte Carlo code efficiencies with measured values. This usually entails modifying crystal parameters (length and diameter), dead layer thickness, end-cap thickness, and end-cap-to-crystal distance. Determining the dead layer thickness is especially important and challenging in geometry modeling. Several techniques have been employed to determine the dead layer thickness of HPGe detectors. Modarresi et al. and Boson et al. used the transmission method with a collimated gamma source to study dead layer thickness across several detector types [2]. Loan et al. utilized the two-line method, calculating the ratios of two efficiency area counts for gamma rays and X-rays emitted by the same radioisotope [8]. This method allows for easy analysis of the exterior dead layer using X-rays, while gamma rays are more suited for examining the inner dead layer. Furthermore, Ródenas et al., Huy, and Chham et al. evaluated dead layer thickness by comparing experimental and calculated efficiencies [4,9,10].

This study presents an approach for correcting gamma radiation activity, with a particular emphasis on improving the critical parameter of full energy peak efficiency in HPGe detectors. The MCNP code is used to simulate the detector's response, underscoring the necessity of regular calibration for accurate radionuclide activity estimation [11,12]. The obtained results were compared with experimental efficiencies derived from MAPEP acceptable activities. These activities were measured for three samples: water, hay, and soil, contained in different Marinelli geometries, as they are the most commonly used for environmental samples. The primary goal is to enhance the accuracy of the detector's initial parameters, which are provided by the manufacturer and can change over time due to aging. This is achieved by adjusting parameters related to dead layer thickness and crystal lengths, which are affected by lithium-ion diffusion. Detailed MCNP simulations with these modified settings aim to rectify discrepancies observed in unacceptable and warning activities identified in the MAPEP's Proficiency Test Reports.

2. METHODOLOGY AND INPUT PARAMETERS

2.1.Experimental setup

The experimental setup involved utilizing a p-type HPGe detector with an intrinsic efficiency of 30%, as part of the MAPEP proficiency test. The detector configuration comprised an aluminum holder, 0.76 mm thick, securing the crystal. A 5 mm gap separated the front of the crystal from the window, with the aluminum end cup window measuring approximately 1.5 mm thick. The dimensions of the crystal were as follows: height 64 mm, diameter 57 mm, with a core hole diameter and depth of 10.1 mm and 52 mm, respectively (see Figure 1). External and internal surfaces of the semiconductor underwent treatment with lithium and boron ion diffusion for the creation of p and n contacts, resulting in outer and inner electrodes measuring approximately 0.5 mm and 0.3 µm, respectively. These electrode layers were considered inert. As outlined by Dryak, Kovar, and Gudelis, radiation absorbed within the dead layers failed to contribute to full-energy peaks due to either charge trapping or delayed release [7]. In this study, the experiment aimed to assess the gamma radiation levels in samples of hay (mass = 94.5 g), soil (mass = 627 g), and water (volume = 1 L) contained in different Marinelli geometries, as they are the most commonly used for environmental samples as presented in Figure 2.

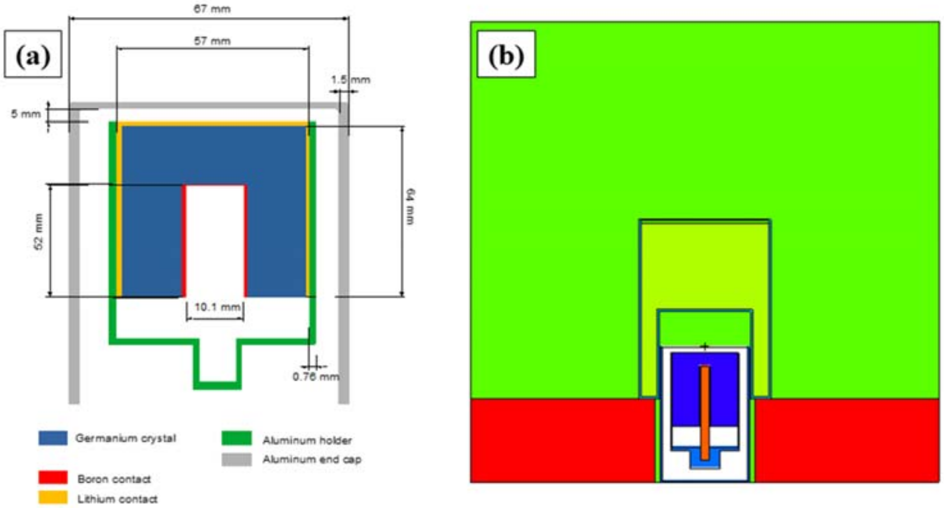
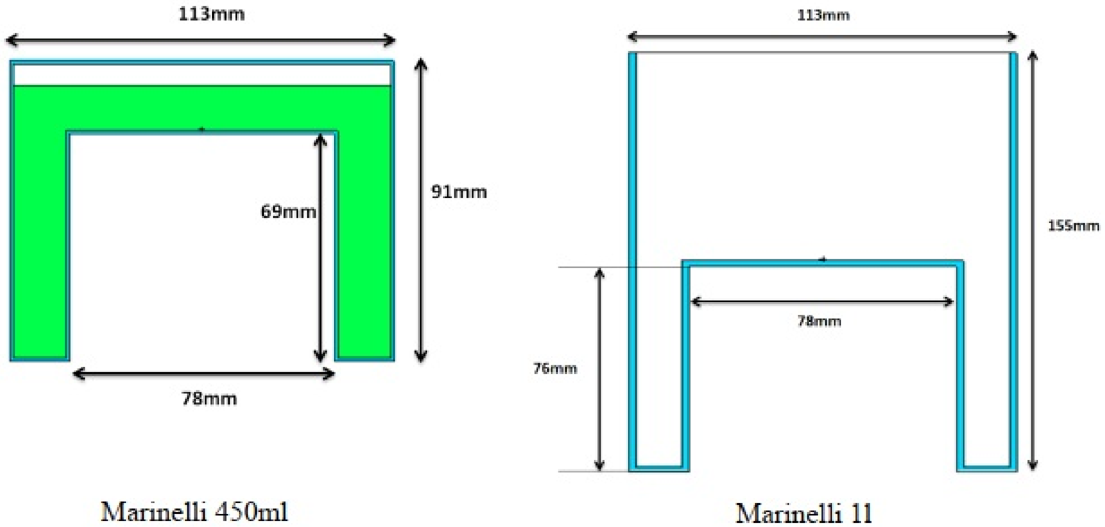


Figure 1. (a) Illustration of the counting geometry using initial parameters given by the manufacturer (b) Cross-sectional views of the MCNP model.



Marinelli 450ml

Marinelli 1l

Figure 2. Dimensions of Various Marinelli Bottles.

2.1. Simulation tool

Monte Carlo-based codes are widely utilized to simulate phenomena governed by probability density functions, such as gamma spectrometry which commonly employs HPGe detectors. Various Monte Carlo codes are employed to compute the efficiency of the full energy peak deposited in the germanium detector [1,6]. These codes track emitted rays at different energies, simulating their interactions with encountered materials from creation to disappearance while considering their respective interaction probabilities. In this study, the Monte Carlo N-Particle (MCNP) radiation transport code was employed to simulate the detector response. MCNP is known it's for extensive cross-section data libraries for neutron, photon, and electron interaction calculations.

Throughout this ongoing study, the detector responses in various evaluated scenarios were simulated using the Pulse Height Tally, denoted as F8. This tally represents the ratio of absorbed particles to emitted ones, considering 1E7 photon histories. The simulation was conducted using the detector's dimensions provided by the manufacturer and appropriate variance reduction techniques. The relative errors associated with each tally ranged from 0.1% to 1%. Subsequently, the calculated efficiencies were compared with experimental values.

3. RESULTS AND DISCUSSIONS

3.1. Efficiency study and optimization of detector specifications

The obtained values of detector efficiency (ε), based on manufacturer specifications, for low gamma energies (E < 120 keV) were found to be over 50% higher compared to MAPEP reference values. As energy increased, these discrepancies decreased, falling to within 15-30%.

As mentioned in the introduction, numerous studies have reported significant discrepancies between experimental and calculated results when using initial parameters provided by the manufacturer. To reconcile Monte Carlo simulation results with experimental values, adjustments to the detector's characteristics are necessary. Typically, these adjustments involve modifying crystal dimensions (length and diameter), the thickness of dead layers, the thickness of the end cap, and the distance from the end cap to the crystal.

Van der Graaf, Dendooven, and Brandenburg (2014) found that differences between measured and calculated efficiencies for high energies reached 10-20%, and were even more pronounced (60-80%) for low energies [13]. These differences were attributed to absorbing materials affecting low energies and increases in dead layer thickness (by factors of 1.8 and 3 for the top and lateral sides, respectively) compared to initial specifications, primarily due to the ongoing diffusion of lithium ions within the crystal. Modest but necessary adjustments in the end cap to crystal top surface distance (from 3 to 5 mm) and the crystal diameter (from 60.4 to 61.8 mm) were also required for greater accuracy.

Huy (2010) reported an 18.5% reduction in the efficiency of an HPGe detector over 13 years of operation. These decreases corresponded to an increase in dead layer thickness (from 0.35 mm to 1.46 mm), causing additional shielding and a reduction in the active volume of germanium. Each effect impacts a specific energy range: at low energies, the primary cause of decreased efficiency is the shielding effect, while at high energies, both the reduction in active volume and the shielding effect contribute to decreased efficiency [10].

To examine the impact of each parameter on efficiency, we conducted a study using different gamma rays (59.5, 122.06, 661.6, and 1332.5 keV) that represent key points on the energy-dependent efficiency curve. First, we examined the variation in efficiency while keeping one dead layer fixed and varying the other. Second, we used the initial specifications and varied either the length or diameter of the crystal. The results are presented in Figure 3.

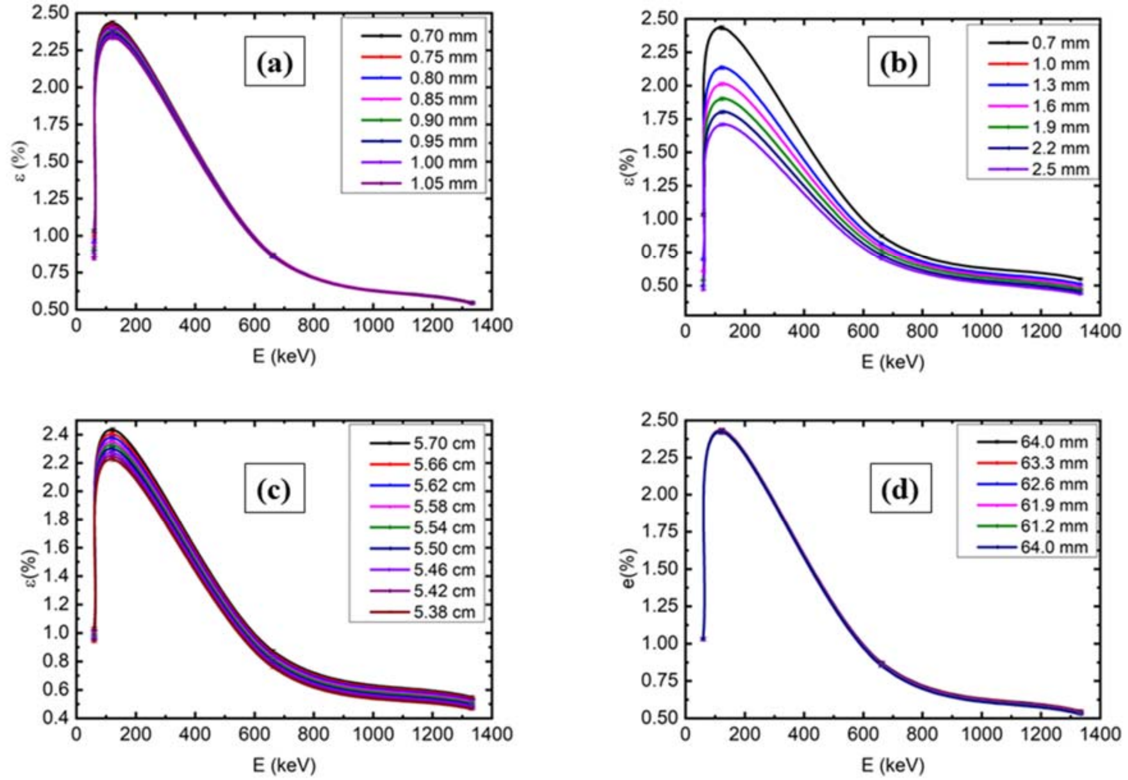


Figure 3. (a) Variation of efficiency with energy for different front dead layer thicknesses (lateral dead layer = 0.7 mm), (b) Variation of efficiency with energy for different lateral dead layer thicknesses (top dead layer = 0.7 mm), (c) Variation of efficiency with energy for different values of crystal diameter, and (d) Variation of efficiency with energy for different values of crystal length.

Starting with Figure 3(d), we observe that there is no significant variation in efficiency when considering different crystal lengths. This is primarily due to the minimal contribution of backscattered rays to the full energy peak. In contrast, the variation in crystal diameter, as illustrated in Figure 3(c), reveals a substantial efficiency difference across all energy levels, which can be attributed to the volume effect. Regarding dead layer variations, it is evident that even a small variation in the front dead layer (Figure 3(a)) predominantly affects energies below 400 keV. The increased dead layer thickness results in greater absorption due to the shielding effect. For the lateral dead layer (Figure 3(b)), there is a noticeable impact on efficiency across all energy levels, with higher efficiency observed for thinner dead layers. This effect is more pronounced at lower energies, again due to the shielding effect. To underscore these findings, we plotted the variation of efficiency with different dead layer thicknesses and crystal dimensions results presented are in Figure 4.

As can be seen, efficiency is less sensitive to variations in crystal length around the certified value. This is mainly because, near the initial length specified by the manufacturer, the interaction of photons occurs entirely within the crystal across the energy spectrum for the sample geometries considered. The efficiency for energies below 100 keV is more sensitive to variations in the top dead layer due to the shielding effect, which causes increased absorption within the inactive volume. Above 100 keV, efficiency is less sensitive to top dead layer variations compared to lateral dead layer and crystal diameter variations. Furthermore, crystal diameter impacts efficiency more significantly than the lateral dead layer across the energy spectrum. This can be attributed to the solid-angle effect or the variation in active volume outweighing the shielding effect induced by the dead layer. For lateral dead layer variation, we observed an exponential decrease in efficiency for both 59.5 keV and 122.06 keV. Additionally, efficiency decreases slightly linearly for higher energies (661.6 keV), while it remains constant for the 1332.5 keV peak. All these findings are illustrated in Figure 5, which shows the absolute values of efficiency variation slopes, obtained for different crystal characteristics, depending on energy. In the optimization procedure, we selected the most impactful parameters: crystal diameter, front dead layer, and lateral dead layer. The values of these parameters that minimized discrepancies between calculated and measured efficiencies were set as follows:

- The top dead layer thickness was increased from 0.5 mm to 0.9 mm.
- The lateral dead layer thickness was increased from 0.5 mm to 2.2 mm.
- The active volume was reduced from 160 cm³ to 155 cm³.

This optimal configuration resulted in a good agreement between Monte Carlo and experimental results, with differences reduced to around 8%.

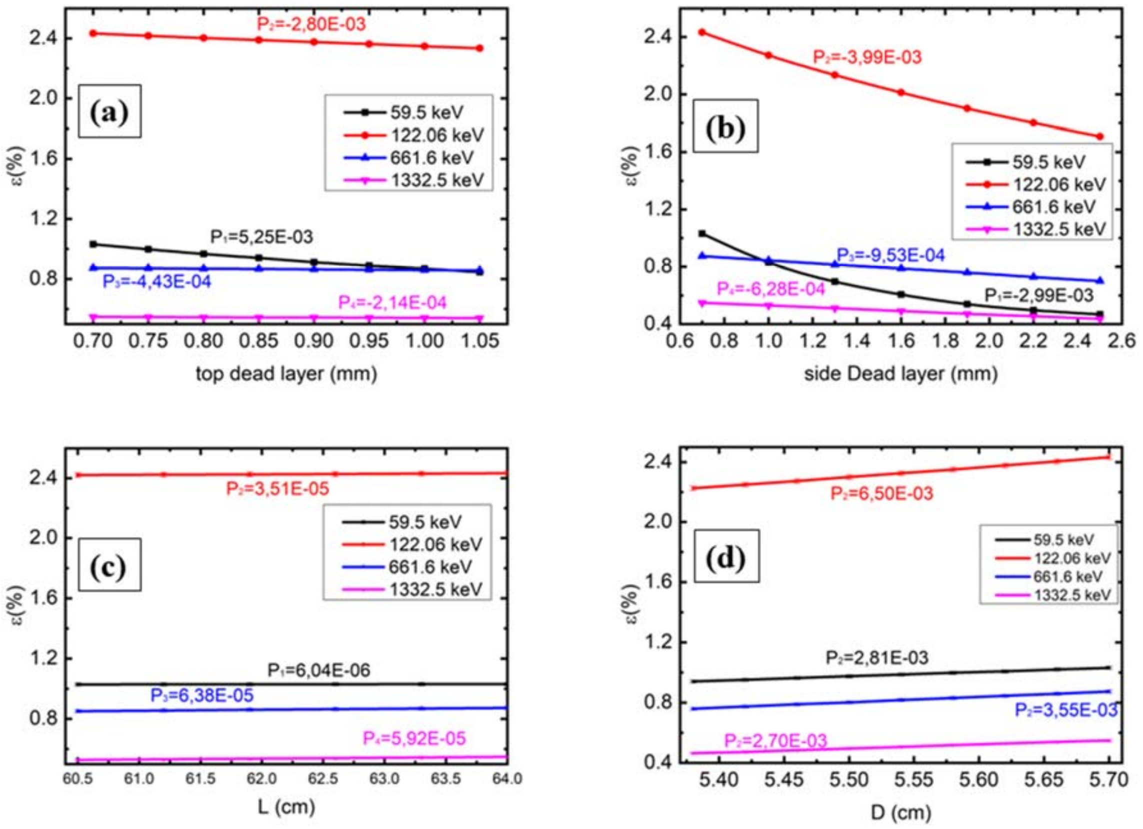


Figure 4. (a) Variation of efficiency with different front dead layer thicknesses for various energies, (b) Variation of efficiency with different lateral dead layer thicknesses for various energies, (c) Variation of efficiency with different crystal lengths for various energies, and (d) Variation of efficiency with different crystal diameters for various energies.

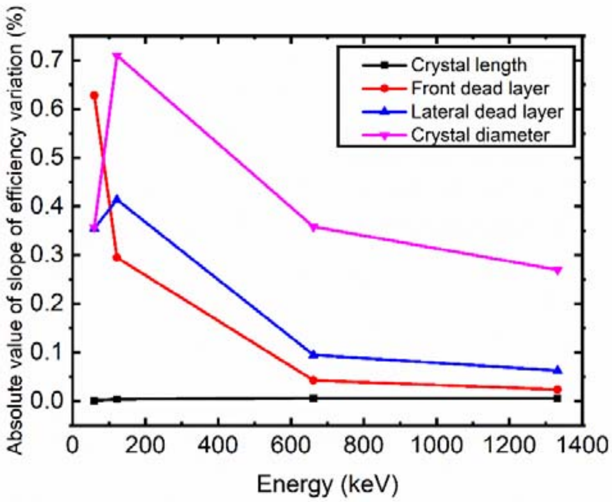


Figure 5. Absolute values of the slopes of efficiency variation obtained for different crystal characteristics at various energies.

3.2. Activity correction

The simulation was conducted considering each sample's specific properties: density, mass composition, and volume. We then recalculated the associated activity using Equation (1) below, incorporating experimentally acquired parameters while substituting measured efficiency with the computed one. The obtained activities and related biases are presented in Table 1.

$$A = \frac{N_{\text{counts}}}{\varepsilon_{\text{cal}}.\,I.\,T_c} \qquad (1)$$

Where N is the number of counts, I is the ray intensity, T_c is the counting time, and ε_{cal} is the calculated efficiency.

In comparison to the certified activities, the experimental results in this study show a bias exceeding 100% for multiple gamma rays, likely due to errors in the parameters used for activity computation. The results obtained using MCNP simulation (Table 1) are in good agreement with the acceptable threshold set by the MAPEP organization (20%). Various correction factors need to be considered to reduce differences between measured and reference activities. For example, periodic detector calibration (once a week or every two weeks) is essential. Additionally, the auto-absorption factor should be evaluated experimentally and applied as a corrective measure to the measured quantities.

Tabel 1. Measured and corrected activities.

Campaign	Matrix	Element	E(keV)	Reference A(Bq)	Measured A(Bq-Bq/kg)	Corrected A (Bq-Bq/kg)	Bias (%) Measured-Reference	Bias (%) Corrected-Reference
MAPEP30	Soil	Am-241	59.5	68	50.61	70.32	-25.57	3.41
	Hay	Cs-134	604.8	6.04	50.76	4.89	740.4	-19.10
		Cs-137	661.6	4.74	47.5	4.95	902.11	4.35
		Co-57	122.06	10.1	102.41	9.18	913.96	-9.06
		Co-60	1332.5	6.93	66.04	6.40	852.96	-7.69
		Mn-54	834.8	8.62	82.55	7.34	857.66	-14.83
		Zn-65	1115.5	7.86	78.37	6.49	897.07	-17.49
MAPEP31	Soil	Am-241	59.5	85.5	39.05	85.01	-54.33	-0.58
		Cs-134	604.8	622	265.59	548.42	-57.3	-11.83
	Hay	Cs-134	604.8	7.38	5.47	5.94	-25.88	-19.53

4. Conclusion

This paper presents a procedure to correct the activities obtained in the MAPRP proficiency test for different samples, namely water, hay, and soil. The unacceptable and warning results were corrected using the MCNP code. The simulation involved accurately modeling the counting geometry of an HPGe detector by studying the relationship between its response and its characteristics. The optimal geometry reduced discrepancies between simulated and measured efficiencies from more than 50% to less than 8%. This modified counting geometry allowed the activities to converge with reference results within 20%, which is the acceptability threshold set by the MAPEP organization. As a result, we consider the procedure followed in this study to be a powerful and effective tool for correcting unacceptable and warning activities. Finally, Monte Carlo simulation could be considered a reliable method for addressing any gamma spectrometry issue.

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