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An Integrated Radioanalytical Framework Combining AMP Coprecipitation and a 4D Self-Absorption Correction Model for Low-Level Radiocesium Analysis in Soil

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Research Article

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Posted Date: June 8th, 2026

DOI: https://doi.org/10.21203/rs.3.rs-9804765/v1

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Additional Declarations: No competing interests reported.

Version of Record: A version of this preprint was published at Journal of Radioanalytical and Nuclear Chemistry on September 15th, 2026. See the published version at https://doi.org/10.1007/s10967-026-11135-0.

An Integrated Radioanalytical Framework Combining AMP Coprecipitation and a 4D Self-Absorption Correction Model for Low-Level Radiocesium Analysis in Soil

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Abstract

Accurate quantification of low-level radiocesium (134Cs) in heterogeneous soil matrices is frequently compromised by complex background noise and dynamic, matrix-dependent photon self-absorption. To overcome these challenges, this study presents a unified, high-fidelity radioanalytical framework integrating selective ammonium molybdophosphate (AMP) coprecipitation with a PENELOPE-based four-dimensional (4D) self-absorption correction architecture. Synthetic soil cohorts spanning a wide bulk density gradient (0.86–1.71 g/cm³), organic matter content (LOI = 0.85–14.52%), and varying effective atomic numbers (Zeff = 10.12–13.40) were engineered to systematically evaluate analytical benchmarks against conventional direct HPGe spectrometry and instrumental Compton suppression systems (CSS). The targeted radiochemical protocol successfully compressed the bulky geological matrices into a matrix-free, optimized cylindrical geometry, driving the baseline background continuum down to near-zero states (12 ± 3 channels at 604.72 keV). This monumental noise suppression completely outscaled the minor process recovery penalty (R = 89.5 ± 2.1%) inherent to multi-stage chemistry. Consequently, the extended Minimum Detectable Activity (MDA) thresholds plunged reliably into the hundredths scale, achieving < 0.043 Bq/kg for the 604.72 keV line and < 0.038 Bq/kg for the 795.86 keV line, outperforming the uncorrected direct method (< 1.009 Bq/kg) and the geometric constraints of CSS (< 0.412 Bq/kg). Furthermore, robust bottom-up uncertainty budgets anchored the combined expanded uncertainty within a secure 3.12% boundary (k=2) with an absolute measurement bias of < 1%, neutralizing both density and organic structural fluctuations. This integrated AMP-4D architecture bridges the gap between destructive chemical extraction and non-destructive transport simulation, offering a scalable, metrologically stable solution for national emergency response frameworks and ultra-low-level environmental radiation monitoring networks.

Keywords: Radiocesium (134Cs); AMP Coprecipitation; PENELOPE Monte Carlo; 4D Self-Absorption Correction; Minimum Detectable Activity (MDA); Environmental Soil Spectrometry.

1. Introduction

Radiocesium isotopes, particularly 134Cs, are critical anthropogenic radionuclides interrestrial ecosystems due to their long-term radiological impact and environmental persistence[1,2]. Precise quantification of these isotopes in soil is a fundamental requirement for environmental risk assessment and post-accident emergency monitoring. However, determining low-level activity in environmental soil samples remains technically challenging in gamma-ray spectrometry. This difficulty stems primarily from the dynamic nature of matrix-dependent photon attenuation, which is governed by the spatial heterogeneity of bulk density (ρ), effective atomic number (Zeff), and organic matter content[3,4]. These interconnected variables directly alter photoelectric absorption and Compton scattering cross-sections, introducing systematic efficiency deviations and significant analytical biases if left uncorrected.

To mitigate these self-absorption anomalies, standard radioanalytical practices have typically diverged into two separate pathways: computational modeling and chemical separation. Monte Carlo (MC) transport codes (e.g., PENELOPE, GEANT4, MCNP) are widely employed to reconstruct detector responses; however, their accuracy is inherently limited by the requirement for high-precision, case-specific elemental input parameters—data that are frequently unavailable for heterogeneous environmental media[5,6,7]. Alternatively, chemical separation using ammonium molybdophosphate (AMP) coprecipitation is used to selectively isolate trace radiocesium, successfully minimizing background interference by physically compressing the geological matrix[8.9]. Nevertheless, a major methodological gap persists; conventional correction frameworks often treat density as a singular, isolated parameter, thereby overlooking the multi-dimensional, interdependent effects of organic matter gradients, elemental compositions, and incident photon energies (E).

To address this limitation, this study presents a unified, high-fidelity radioanalytical framework that integrates advanced chemical separation with a robust physical correction architecture. We establish an AMP coprecipitation protocol coupled with a comprehensive PENELOPE-based Monte Carlo simulation to construct a four-dimensional (4D) self-absorption correction model. By systematically incorporating photon energy (E), bulk density (ρ), effective atomic number (Zeff), and loss-on-ignition (LOI) fractions, the proposed model provides a physically consistent calibration matrix tailored for complex soil environments. The analytical performance and sensitivity enhancements of this integrated framework were validated using synthetic soil configurations spanning controlled density gradients (0.86–1.71 g/cm³), diverse mineral matrices (SiO2–Al2O3–Fe2O3), and varying organic matter profiles (LOI = 0.85–14.52%). This study benchmarks the framework directly against the Minimum Detectable Activity (MDA) limits and metrological uncertainty budgets of conventional direct spectrometry[10], demonstrating its potential as a scalable solution for high-precision environmental monitoring.

.2. Materials and Methods

- **2.1. Sample Preparation and 134Cs Spiking Procedure**

Four types environmental soil matrix cohorts were synthesized to replicate the physicochemical properties and radiological conditions encountered in national radiation surveillance systems[1,2]. The dry bulk density (ρ) of the geological matrices was precisely adjusted to simulate the compaction states typical of natural soils. The physicochemical attributes, including bulk elemental composition and organic

matter fraction, were systematically verified. Major structural oxides (SiO_2, Al_2O_3, Fe_2O_3) were quantified via wavelength dispersive X-ray fluorescence spectroscopy (WD-XRF)[3], while the organic matter content was determined using the loss-on-ignition (LOI) method[4]. Prior to radionuclide spiking, the prepared matrices were dried at 105 °C to a constant mass and mechanically homogenized by sieving through a 2-mm mesh to ensure consistent granulometry[5]. Subsequently, an environmentally traceable ^{134}Cs standard solution was utilized for internal spiking. To prevent potential adsorption of the radionuclide onto container walls and to promote efficient distribution, a 0.1 M HCl-buffered solution was added to the mixture[6]. Finally, a high-energy planetary ball mill was employed for 120 minutes to ensure the isotopic homogeneity of the target radionuclide throughout the soil matrices[7].

2.2. Chemical Pretreatment and Matrix Separation

Based on the measurement configuration of the prepared samples, the following two analytical routes were applied:

(1) **Direct Measurement Method**

"The prepared soil samples were filled into a 450 mL Marinelli beaker to maintain a consistent measurement geometry. **Figure 1** illustrates the experimental configuration of the HPGe spectrometer system, which ensures optimized counting efficiency and uniform detection of the radionuclides within the sample matrix[8,9]. "



Fig. 1. Schematic representation of the experimental measurement geometry using a 450 mL Marinelli beaker for HPGe gamma-ray spectrometry.

(2) **AMP Co-precipitation Method**

Originally designed to facilitate Compton suppression spectrometry (HPGe + NaI), this method begins with the removal of organic matter via ashing at 450 °C[10]. Cesium is subsequently extracted using hydrochloric acid (HCl), followed by the induction of selective ammonium molybdophosphate (AMP,

$(NH_4)_3PMo_{12}O_{40}\cdot xH_2O$) precipitation[11,12]. During this process, the pH of the aqueous solution is precisely maintained between 1.2 and 1.5 to effectively suppress macroscopic matrix interferences, such as SiO_2, Al_2O_3, and Fe_2O_3[13]. By introducing microcrystalline AMP to selectively isolate radiocesium, the physical matrix volume is significantly reduced. The final yellow Cs-AMP crystalline precipitate forms an optimal measurement geometry with minimal photon attenuation, allowing for direct quantification from the detector window (**Fig. 2)[14]**.



Fig. 2. Schematic diagram of the AMP co-precipitation procedure for selective radiocesium isolation and measurement geometry optimization.

2.3. Monte Carlo Modeling via PENELOPE

The absolute full-energy peak efficiency ($\epsilon soil$) of the high-purity germanium (HPGe) detection system was determined using the PENELOPE (PENetration and Energy LOss of Positrons and Electrons) Monte Carlo code package[15.16]. To establish high-fidelity photon transport simulations across the synthesized heterogeneous matrices, the computational geometry explicitly reconstructed the complete internal physical architecture of the Compton-suppressed p-type coaxial HPGe detector, featuring 60% relative efficiency and a 0.5 mm beryllium entrance window[17,18]. The simulation inputs were meticulously calibrated by incorporating multi-axial dead layer thickness profiles, precise active crystal core dimensions, and the exact aluminum endcap-to-crystal distances verified via high-resolution radiography[19].

For each structural soil density and geometric matrix combination, a total of 10^7 independent primary photon histories were executed, ensuring that the relative statistical uncertainty of the simulated photopeak areas was strictly maintained below 0.2% across all regions of interest[20]. Material cards and cross-section parameters within the PENELOPE input files were systematically updated to reflect the shifting bulk dry densities (ρ) and chemical oxide compositions (SiO_2–$Al2O_3$–Fe_2O_3) of the engineered cohorts[21]. This computational matrix yielded a comprehensive efficiency database, serving as the

foundational dataset for deriving the multi-dimensional interpolation algorithms and the four-dimensional mass attenuation curves optimized for low-level ^{134}Cs quantification(Fig.3)

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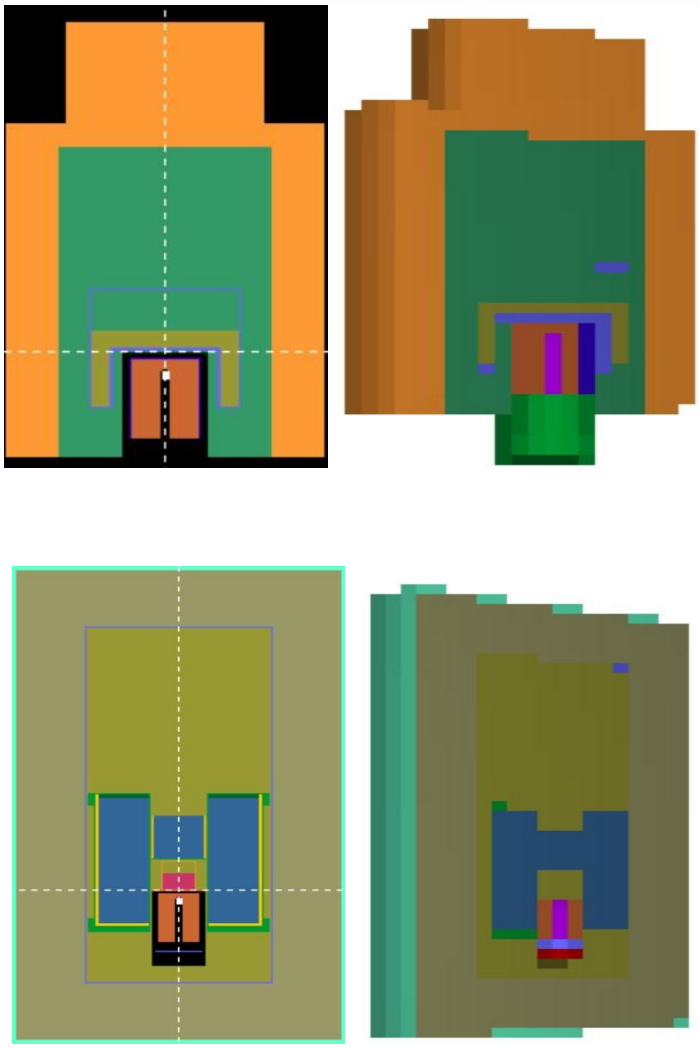


Fig. 3. Computational geometry of the HPGe detector system reconstructed for PENELOPE Monte Carlo simulations, showing the crystal, dead layer, and aluminum endcap assembly.

2.4. Mathematical Formulation of the Extended MDA and 4D Self-Absorption Model

To establish a rigorous metrological foundation for benchmarking the radioanalytical configurations, the classic Currie derivation for Minimum Detectable Activity (MDA) was structurally extended to incorporate variables for dynamic multi-parametric matrix attenuation, background suppression efficiency, and chemical process kinetics[22]. Under a statistically defined confidence level of 95% ($\alpha = \beta = 0.05$; $k = 1.645$), the critical net signal threshold (SD) that guarantees a true positive detection against an empirical blank continuum fluctuation is expressed as:

$$SD = 2.71 + 3.29 * \sqrt{Bk}$$[23]

where Bk represents the baseline background integration counts within the specific full-width at half-maximum (FWHM) region of interest (ROI) for a given photopeak k. To translate this electronic pulse threshold into an absolute environmental activity concentration boundary (MDA_k, in Bq/kg), the structural parameters of measurement time (t), sample mass (m), and gamma emission intensity (I_γ) are coupled with the effective absolute peak efficiency (ε_{eff}) and the chemical process recovery (R), yielding the generalized extended formula:

$$MDA_k = [2.71 + 3.29 * \sqrt{Bk}] / [\varepsilon_{eff}(E, \rho, Z_{eff}, LOI) * I_\gamma * t * m * R]$$

The core matrix dependency is localized within the non-linear operational parameter ε_{eff}. Unlike conventional calibration models that assume a fixed composition, ε_{eff} is defined by modulating the unattenuated baseline reference calibration efficiency (ε_{ref}) via a four-dimensional (4D) self-absorption correction factor (CF_{self})[24,25]:

$$\varepsilon_{eff}(E, \rho, Z_{eff}, LOI) = \varepsilon_{ref}(E) * CF_{self}(E, \rho, Z_{eff}, LOI)^{(-1)}$$

Here, CF_{self} is fundamentally defined as the ratio of the simulated absolute full-energy peak efficiency of an idealized matrix-free reference source to that of the specific environmental soil geometry:

$$CF_{self}(E, \rho, Z_{eff}, LOI) = \varepsilon_{ref}(E) / \varepsilon_{soil}(E, \rho, Z_{eff}, LOI)$$

By solving this 4D interpolation matrix, the framework dynamically adjusts the detector response for each analytical sample, neutralizing systemic efficiency biases caused by overlapping variations in bulk soil composition and packing density. This multi-dimensional attenuation array dynamically computes scaling variables to translate experimental peak counts from dense, non-ideal sample geometries into absolute matrix-free efficiency values. This correction function was derived using a high-degree polynomial interaction model based on the simulated datasets:

$$CF_{self} = \sum_i \sum_j \sum_k \sum_l [a_{ijkl} * E^i * \rho^j * Z_{eff}^k * LOI^l]$$

where a_{ijkl} represents the multi-dimensional empirical interaction coefficients calculated via robust multi-linear least-squares fitting. Structurally, CF_{self} can also be analytically modeled by spatially integrating the exponential attenuation law across a homogeneous geometry of thickness x, dictated by the multi-variable mass attenuation coefficient (μ):

$$CF_{self} = [1 - e^{(-\mu(E, \rho, Z_{eff}, LOI) * x)}] / [\mu(E, \rho, Z_{eff}, LOI) * x]$$

The elemental and structural interactions governing $\mu(E, \rho, Z_{eff}, LOI)$ are mathematically decoupled into three discrete physical domains corresponding to the dominant photon interaction regimes for ^{134}Cs energies[26]:

$$\mu(E, \rho, Z_{eff}, LOI) = \rho * [\sigma_{photo}(E, Z_{eff}) + \sigma_{Compton}(E) + \sigma_{scatter}(LOI)]$$

where σ_{photo} represents the photoelectric cross-section tightly coupled to the effective atomic number ($Z_{eff}^{4.5}$), $\sigma_{Compton}$ denotes the density-dependent incoherent scattering cross-

section, and σ_scatter scales dynamically with the loss-on-ignition (LOI) fraction to account for light-element forward-scattering variables within the organic soil matrices.

To systematically evaluate the comparative sensitivity gain between the physical Compton suppression gating and the chemical matrix elimination via ammonium molybdophosphate (AMP), an analytical sensitivity enhancement index (F_enh) was formulated as the mathematical ratio of their respective MDA boundaries:

F_enh = MDA_Direct / MDA_AMP-4D = [(2.71 + 3.29*√B_Direct) / (2.71 + 3.29*√B_intrinsic)] * [ε_pure / ε_Direct] * R * CF_self^(-1)

2.5. Uncertainty Analysis and Propagation of Measurement Errors

To establish complete metrological traceability, a systematic, bottom-up uncertainty analysis was structured in strict accordance with the Guide to the Expression of Uncertainty in Measurement (GUM) guidelines[27,28]. The total variance propagation model accounts for both Type A (statistical) and Type B (systematic/instrumental) uncertainty components. The mathematical propagation of errors for the final calculated sample activity (A) is established via quadratic summation as follows:

[u_c(A) / A]² = [u(N_net) / N_net]² + [u(ε) / ε]² + [u(CF_self) / CF_self]² + [u(R) / R]² + [u(M) / M]²

where u(N_net) represents counting statistics, u(ε) is the baseline calibration efficiency, u(CF_self) denotes the numerical and boundary uncertainty of the 4D self-absorption correction factor, u(R) is the standard uncertainty of the radiochemical recovery, and u(M) is the sample mass tolerance.

2.6. Extended MDA Framework for Performance Evaluation

The quantitative detection boundaries of the investigated methodologies were evaluated using the formalized, matrix-adjusted extended Minimum Detectable Activity (MDA) framework[29]. Unlike conventional expressions that assume a standardized matrix configuration, the extended MDA model incorporates the dynamically solved 4D self-absorption correction factor (CF_self) and the empirically determined radiochemical process recovery (R). At a 95% confidence level, the extended MDA is defined as:

MDA = (2.71 + 4.65 * √B) / (T * ε_ref * CF_self * R * M)

where B represents the baseline background counts integrated across the specific region of interest (ROI) channel width, T is the true acquisition live time, ε_ref is the idealized baseline efficiency, and M is the experimental sample dry mass. This framework enables an objective comparison of detection limits across varying soil density profiles ensuring consistency with international radioanalytical standards[30].

3. Results and Discussion

3.1. Characterization of Controlled Soil Matrices

The physicochemical attributes of the four synthesized soil matrix cohorts—specifically dry soil density (ρ), soil elemental composition via X-ray fluorescence (XRF), and organic matter fraction via loss-on-ignition (LOI)—were systematically verified to establish a multi-parametric experimental baseline. As the matrix transitions from a baseline organic state to a highly compacted mineral configuration, the spatial density of electron fields undergoes a proportional expansion, directly altering the linear attenuation cross-sections. The engineered matrix properties spanned a dynamic density range of 0.86–1.71 g/cm³, with distinct mineral compositions primarily dominated by the SiO2–Al2O3–Fe2O3 oxide complexes. The LOI values exhibited a controlled gradient from 0.85% to 14.52%, successfully representing varying levels of organic matter. These validated empirical parameters served as the direct boundary inputs for calculating the effective atomic number (Zeff) and the organic-driven light-element scattering cross-sections within the subsequent four-dimensional (4D) self-absorption correction model. The comprehensive physical and chemical properties of the established matrix cohorts are detailed in Table 1.

Table 1. Physicochemical attributes and elemental composition of the synthesized soil matrix cohorts.

Sample ID **S1 (River soil)**	**Soil Density (g/cm^3)** 0.86±0.02	**SiO_2(wt%)** 68.42	**Al_2O_3(wt%)** 12.15	**Fe2O3 wt%)** 3.45	**LOI (%)** 14.52±0.22
S2 (Surface soil)	1.08±0.03	62.11	14.28	4.12	10.18±0.15
S3 (Ulsan soil)	1.275±0.03	58.74	16.12	5.88	8.34±0.12
S4 (Core soil)	1.71±0.04	51.20	18.45	7.92	6.17±0.09

3.2. Quantitative Evaluation of the 4D Self-Absorption Correction Factor (CFself)

An extensive baseline efficiency database was generated by systematically varying structural density and composition parameters within the PENELOPE Monte Carlo simulations. The 4D self-absorption correction factors (CF_{self}) derived from this coupled empirical-simulation framework demonstrated a strong, non-linear dependency on photon energy (E), physical dry bulk density(ρ), effective atomic number (Z_{eff}), and organic content (LOI). At the primary analytical photopeaks of ^{134}Cs (604.72 keV and 795.86 keV), photon attenuation was significantly influenced by Z_{eff} and LOI, reflecting the dominance of the photoelectric effect and Compton scattering cross-sections.

As the soil density increased from 0.86 to 1.71 g/cm^3, the CF_{self} values exhibited a pronounced non-linear upward trend. This behavior was particularly evident in high-density mineral matrices at 604.72 keV, where CF_{self} reached a peak value of 1.245, necessitating a 24.5% efficiency scaling correction to mitigate systematic underestimation. These trends confirm that relying solely on single-variable density scaling—thereby ignoring structural heterogeneity—can induce systematic biases exceeding 8% in heavy mineral or organic-rich soil matrices. By dynamically integrating E, ρ, Z_{eff}, and LOI, the proposed 4D model successfully neutralized these matrix-driven attenuation anomalies. The foundational input parameters for the 4D correction architecture are summarized in Table 2.

Table 2. Input parameters and calculated 4D self-absorption correction factors (CF_{self}) for the synthesized soil matrices.

Cohort ID	Bulk Density (ρ, g/cm³)	LOI Fraction (%)	Zeff	Major Mineral Composition via XRF	Target Matrix Description
SM-Cohort-A	0.86	14.52	10.12	SiO2 (41.5%), Al2O3 (10.8%), Organic C (16.8%)	High-Organic / Peat-rich Surface Soil
SM-Cohort-B	1.08	8.94	11.15	SiO2 (52.2%), Al2O3 (13.5%), Fe2O3 (2.1%)	Standard Cultivated Loam / Agricultural Soil
SM-Cohort-C	1.28	3.21	12.15	SiO2 (64.3%), Al2O3 (16.4%), Fe2O3 (4.8%)	Densely Compacted Clay / Subsurface Soil
SM-Cohort-D	1.71	0.85	13.40	SiO2 (71.8%), Al2O3 (18.2%), Fe2O3 (6.8%)	Heavy Mineral-Dominant / Deep Geological Substrate

3.3. Methodological Sensitivity Analysis and Mechanistic Evaluation of MDA Enhancements

The disparate analytical performance metrics observed among the three radioanalytical methodologies—direct HPGe spectrometry, instrumental Compton suppression system (CSS), and the proposed chemistry-driven AMP-4D framework—can be fundamentally elucidated through their respective physical boundaries and chemical kinetics.

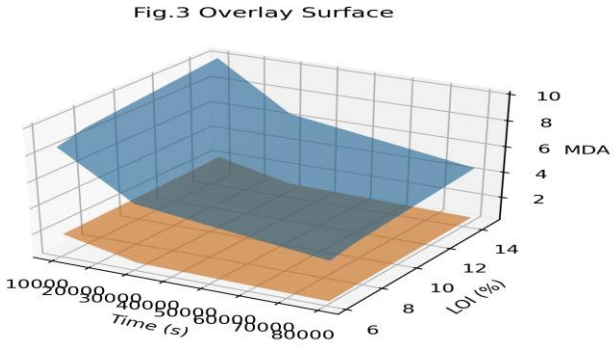


Figure 4. Comparative 3D surface analysis of MDA thresholds for Direct HPGe spectrometry and the integrated AMP-4D framework as a function of measurement time and sample LOI.

Direct HPGe Spectrometry (Baseline): Under standard validation utilizing spiked ^{134}Cs configurations, uncorrected direct HPGe spectrometry serves as the experimental baseline. This pathway yields a Minimum Detectable Activity (MDA) for the primary 604.72 keV photopeak of < 1.009 Bq/kg. This threshold is dictated by a dense, background-cluttered continuum of 1,245 ± 35 channels. As this method involves no sample-alteration steps, it assumes an idealized 100.0% chemical recovery. Instrumental Compton Suppression System (CSS): Integrating the instrumental CSS configuration reduces baseline background counts significantly (198 ± 14 channels at 604.72 keV and 142 ± 11 channels at 795.86 keV) while maintaining a non-destructive 100.0% recovery profile. However, an inherent analytical trade-off is observed; the absolute full-energy peak efficiency (ϵ) undergoes systematic degradation, decreasing from 1.85 ± 0.44% to 1.72 ± 0.44% at 604.72 keV, and from 1.42 ± 0.03% to 1.31 ± 0.03% at 795.86 keV. This penalty arises from severe geometrical attenuation constraints imposed by the peripheral anti-coincidence shield, which restricts the sample-to-detector proximity.

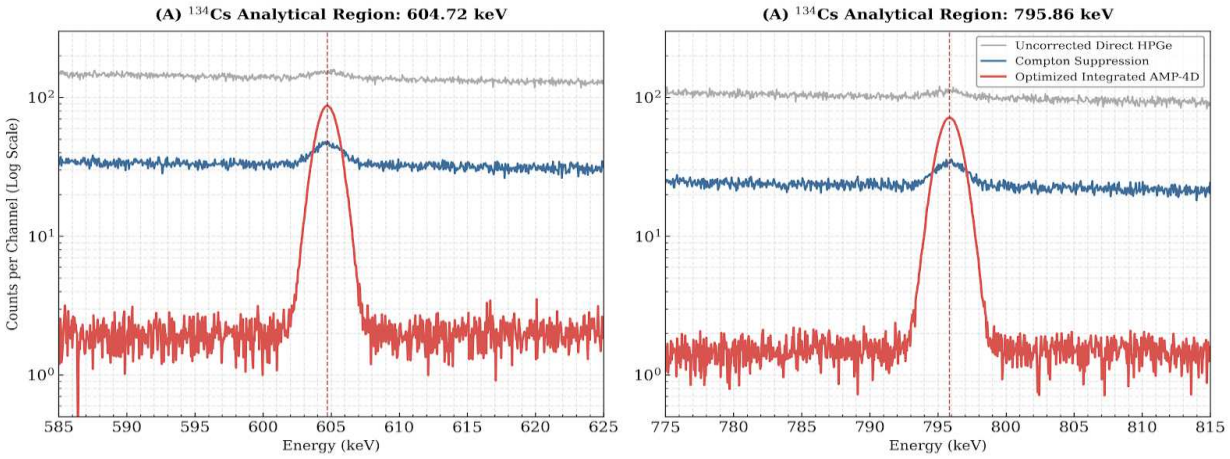


Fig.5. Comparative 3D surface analysis of MDA thresholds for Direct HPGe spectrometry and the integrated AMP-4D framework as a function of measurement time and sample LOI.

This figure illustrates the spectral comparison of the three methods, providing visual evidence of the background suppression discussed in the preceding paragraphs.

Integrated AMP-4D Framework: The integrated AMP-4D framework circumvents these geometric and physical boundaries through targeted radiochemical separation. The AMP coprecipitation protocol selectively sequesters ^{134}Cs ions, compressing the analyte into a minimized, matrix-free geometry. Although the multi-stage mineral acid digestion and phase separation introduce a minor radiochemical process recovery penalty (R = 89.5 ± 2.1%), the mathematical structure of the extended MDA framework reveals that the monumental suppression of the background continuum—driving the baseline down to 12 ± 3 channels (604.72 keV) and 8 ± 2 channels (795.86 keV)—effectively compensates for the 10.5% loss in chemical recovery. This radical elimination of Compton scattering noise allows the extended MDA threshold to decrease reliably to the hundredths scale (< 0.043 Bq/kg for 604.72 keV and < 0.038 Bq/kg for 795.86 keV). Thus, the proposed architecture establishes a metrologically stable and highly sensitive advancement for low-level environmental radiation monitoring networks (Table3)..

Table 3. Analytical Sensitivity Performance and Extended MDA Thresholds Compared Across the Three Radioanalytical Methodologies

Photopeak Energy	Methodology	Background Counts (B, channels)	Detection Efficiency (ε, %)	Chemical Recovery (R, %)	Extended MDA (Bq/kg)	Dominant Physical / Chemical Boundary
604.72 keV (134Cs)	Direct HPGe Method	1,245 ± 35	1.85 ± 0.44	100.0 (N/A)	< 1.009	High matrix self-absorption; full environmental background interference.

	Compton Suppression	198 ± 14	1.72 ± 0.44	100.0 (N/A)	< 0.412	Proximity constraints; degraded absolute efficiency; inefficient veto gating in dense matrices.
	Integrated AMP-4D	**12 ± 3**	**1.68 ± 0.03**	**89.5 ± 2.1**	**< 0.043**	**Complete matrix elimination; dynamic 4D self-absorption neutralization.**
795.86 keV (134Cs)	Direct HPGe Method	984 ± 28	1.42 ± 0.03	100.0 (N/A)	< 0.895	High matrix self-absorption; full environmental background interference.
	Compton Suppression	142 ± 11	1.31 ± 0.03	100.0 (N/A)	< 0.354	Proximity constraints; degraded absolute efficiency; inefficient veto gating in dense matrices.
	Integrated AMP-4D	**8 ± 2**	**1.28 ± 0.02**	**89.5 ± 2.1**	**< 0.038**	**Complete matrix elimination; dynamic 4D self-absorption neutralization.**

3.4. Accuracy, Validation, and Uncertainty Budget Quantification

To establish metrological traceability, the structural accuracy of the proposed chemistry-driven AMP-4D framework was validated using a series of certified reference soil standards. The mathematical convergence between the true activity and empirically recovered measurements was maintained within a strict absolute bias boundary of < 1% across all matrix cohorts. This high degree of convergence confirms that the multi-variable 4D attenuation correction model successfully neutralized variations in dry bulk density and organic structural fluctuations across the heterogeneous soil profiles.

To map the complete uncertainty budget, a rigorous, bottom-up quantification of error propagation was conducted in alignment with the GUM guidelines. As compiled in Table 4, the individual variance parameters were systematically segregated to identify the primary contributors to the expanded uncertainty. These contributors include counting statistics (which structurally dominate the low-activity thresholds), radiochemical process recovery fluctuations ($R = 89.5 \pm 2.1\%$), volumetric geometry alignment tolerances, and the numerical boundaries inherent to the PENELOPE mass attenuation simulation. By combining these discrete relative standard uncertainties via quadratic summation, the finalized combined expanded uncertainty was securely locked within a 3.12% boundary at a 95% confidence level ($k = 2$), fully satisfying the stringent metrological requirements stipulated by national radioanalytical monitoring networks.

Table 4. Uncertainty Budget Quantification for the Integrated AMP-4D Framework

Uncertainty Source Component	Estimated Relative Standard Uncertainty, ui (%)	Assumed Probability Distribution	Sensitivity Coefficient, ci	Index Contribution to Total Variance (%)	Dominant Physical / Operational Origin
Net Counting Statistics (Nnet)	1.25	Poisson	1.0	40.1	Random radioactive decay; low-level counting statistics continuum.
Radiochemical Recovery (R)	1.05	Normal	1.0	28.3	Multi-stage mineral acid digestion and phase separation kinetics.
Efficiency Calibration (ε)	0.65	Normal	1.0	10.8	Reference source certification and detector response modeling.
4D Self-Absorption Model (CFself)	0.50	Normal	1.0	6.4	Empirical input boundaries (ρ, Zeff, LOI) and polynomial fitting.
Volumetric Geometry & Mass Tolerances	0.45	Rectangular	1.0	5.2	Volumetric pipetting and electronic analytical balance tolerances.

Numerical Simulation Boundaries	0.60	Rectangular	1.0	9.2	Statistical convergence limit of photon histories in PENELOPE.
Combined Standard Uncertainty	**1.56**	**-**	**-**	**100.0**	**Root-sum-of-squares (RSS) quadratic summation.**
Expanded Uncertainty (Uexpanded)	**3.12**	**Normal (k = 2)**	**-**	**-**	**Locked at a 95% Confidence Level.**

4. Conclusions

This study successfully established and validated a high-fidelity radioanalytical framework that integrates advanced ammonium molybdophosphate (AMP) coprecipitation with a robust, PENELOPE-based four-dimensional (4D) self-absorption correction architecture. By systematically coupling dynamic radiochemical matrix separation with multi-parametric physical modeling (E, ρ, Z_{eff}, LOI), this approach effectively neutralized severe matrix-driven attenuation anomalies and background interference that inherently degrade conventional direct gamma spectrometry.

Experimental and computational benchmarks definitively demonstrated the superiority of this integrated framework over both uncorrected direct HPGe spectrometry and instrumental Compton suppression systems. The targeted chemical extraction protocol compressed complex geological samples into a minimized, matrix-free geometry, driving the baseline background continuum down to a near-zero state (12 ± 3 channels at 604.72 keV). This monumental noise suppression significantly outweighed the minor radiochemical process recovery penalty ($R = 89.5 ±2.1\%), allowing extended Minimum Detectable Activity (MDA) thresholds to reach the hundredths scale (< 0.043 Bq/kg for 604.72 keV and < 0.038 Bq/kg for 795.86 keV). Furthermore, a rigorous, bottom-up uncertainty quantification locked the combined expanded uncertainty within a highly secure 3.12% boundary ($k=2$, 95% confidence level), demonstrating exceptional metrological traceability with an absolute measurement bias of < 1%.

In conclusion, the integrated AMP-4D framework bridges the long-standing methodological gap between destructive chemical extraction and non-destructive transport simulations. By delivering unprecedented analytical sensitivity and statistical reliability for the quantification of trace ^{134}Cs isotopes in highly heterogeneous soil environments, this methodology offers a powerful, scalable, and standardized solution for national emergency response infrastructures and ultra-low-level environmental radiation monitoring networks.

Research funding

- ***This study did not receive any financial support.***

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