

Methodology for the simultaneous determination of the parameters associated with the k_0 method of neutron activation analysis and experimental determination of the k_0 and Q_0 parameters for the $^{158}\text{Gd}(n,\gamma)^{159}\text{Gd}$ reaction

Rafael Vanhoz Ribeiro^{*,}, Maurício Moralles, Mauro da Silva Dias

Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP, Av. Prof. Lineu Prestes 2242, São Paulo, 05508-000, Brazil

ARTICLE INFO

Keywords:

k_0 -NAA
Covariance analysis
Levenberg–marquardt optimization
Neutron activation analysis
Uncertainty quantification

ABSTRACT

A nonlinear adjustment framework for the calculation of k_0 -NAA parameters is presented, integrating covariance analysis with the Levenberg–Marquardt optimization algorithm. This approach enables the simultaneous adjustment of multiple parameters, explicitly incorporating their uncertainties and correlations within a unified fitting procedure. Experimental validation was performed using data from irradiations conducted in 2010, 2015, and 2023, resulting in new values of k_0 and Q_0 for several radionuclides. In particular, the adjustment framework enabled the determination of k_0 values for radionuclides and energies previously absent from the reference database. The results were compared with reference data, demonstrating improved reliability and precision in neutron activation analysis. Furthermore, the framework presented here enables the inclusion of different combinations of targets, energies, and irradiation dates in a single adjustment, providing greater flexibility for future applications.

1. Introduction

The k_0 -standardization method of neutron activation analysis (k_0 -NAA) is a widely adopted approach for the quantitative determination of elemental concentrations without the need for individual standards (International Atomic Energy Agency (IAEA), 2024; De Corte, 1987). Given its extensive application across diverse fields, ranging from pet food categorization (de Lima et al., 2022) to the analysis of trace elements in the fingernails of breast cancer patients (Huynh et al., 2020), maximizing its precision is essential to ensure reliable analytical results.

The method is based on nuclear constants and comparator irradiation, typically using gold as a reference, allowing the calculation of concentrations from measured gamma-ray activities through well-established nuclear and spectral parameters. By incorporating corrections related to neutron spectrum characteristics, detector efficiency, and irradiation conditions, k_0 -NAA provides a robust and flexible strategy for multi-element analysis with improved accuracy and reproducibility across different experimental facilities. In this context, uncertainty evaluation plays a central role in ensuring the reliability of the calculated results.

Uncertainty evaluation in k_0 -NAA requires special attention due to the large number of parameters involved and the potential correlations between them. Previous studies have investigated methodologies for uncertainty estimation. Smodiš and Bučar (2006) demonstrated a methodology for evaluating uncertainties in concentration results based on the Guide to the Expression of Uncertainty in Measurement (GUM). Other studies have focused on improving analytical performance through uncertainty optimization (Kubešová and Kučera, 2015; Benedik and Repinc, 2011; Robouch et al., 2000). However, even among contemporary studies, very few works have managed to resolve this issue by comprehensively addressing all correlations and parameters inherent to the method.

The uncertainty calculations become more complex due to the presence of a larger number of parameters, in addition to the potential correlations that may exist among them. A comparison performed by Kubešová and Kučera (2011) between the two most widely used software packages for k_0 -NAA showed a noticeable difference in their uncertainty estimates, arising from differences in the calculation methods, which introduces a certain level of complexity in determining whether the uncertainties are being obtained in a compatible and reliable manner.

* Corresponding author.

E-mail addresses: rafaelvanhoz@alumni.usp.br (R. Vanhoz Ribeiro), moralles@ipen.br (M. Moralles), msdias@ipen.br (M. da Silva Dias).

<https://doi.org/10.1016/j.apradiso.2026.112568>

Received 7 December 2025; Received in revised form 9 March 2026; Accepted 14 March 2026

Available online 19 March 2026

0969-8043/© 2026 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

Dias et al. (2010) introduced a rigorous statistical model for addressing k_0 -NAA uncertainties through a covariance analysis approach. This strategy provided a more rigorous evaluation of k_0 and its uncertainties, effectively mitigating the limitations of earlier methods that overlooked or oversimplified parameter correlations. Currently, there is increasing interest in the application of the k_0 -NAA method for the calculation of elemental concentrations. As a result, various analytical approaches and specialized software packages have been recently developed to support and optimize its application (Di Luzio and D'Agostino, 2023, 2022; Dung et al., 2022; Barradas et al., 2022; Chahidi et al., 2023; Dias et al., 2023). The International Atomic Energy Agency (IAEA) has conducted an international benchmarking exercise among neutron activation analysis software packages, aiming to elucidate similarities and differences in the uncertainty estimation procedures adopted by various software packages (International Atomic Energy Agency, 2023). Variations in computational models can yield inconsistent analytical outcomes, a major concern when analyzing highly sensitive materials, such as clinical or forensic samples (Datta et al., 2020), that demand strict reliability and precision. Furthermore, because k_0 factors for a specific element's gamma peaks are mutually dependent, and measurements from different spectra of the same sample are likewise correlated, the process of uncertainty propagation is inherently more complicated than computing the mass fraction itself. This complexity persists whether the goal is to determine classical accuracy and precision, evaluate GUM-compliant uncertainty, or calculate an expanded uncertainty (International Atomic Energy Agency, 2023).

Classical implementations typically estimate α , k_0 , and Q_0 in separate steps, propagating uncertainties only partially or assuming weak interdependence between parameters. In addition, earlier experimental determinations of k_0 and Q_0 parameters frequently rely on local fitting strategies or simplified statistical models that do not explicitly incorporate the full covariance structure arising from detector efficiency calibration, neutron spectrum characterization, and nuclear data uncertainties. As a consequence, correlations between experimental observables may be underestimated, potentially leading to biased parameter estimates or underestimated uncertainties.

In recent years, the application of the Levenberg–Marquardt and other computational optimization methods has been widely reported in the nuclear physics literature, demonstrating their effectiveness for parameter estimation, nonlinear fitting of experimental data, and optimization of nuclear models (Özdoğan et al., 2024, 2023; Okutan et al., 2026, 2021). These optimization techniques can also contribute to improving uncertainty calculations in the k_0 -NAA method by enabling the simultaneous adjustment of correlated parameters and supporting covariance-based uncertainty propagation, leading to more consistent results.

The Levenberg–Marquardt algorithm is a well-established technique for solving nonlinear least-squares problems and is particularly suitable for complex physical models in which experimental uncertainties and nonlinear relationships must be treated consistently. In the context of k_0 -NAA parameter estimation, it enables the simultaneous adjustment of multiple correlated parameters while preserving numerical stability and ensuring efficient convergence, thereby supporting rigorous uncertainty propagation within covariance-based frameworks.

Although other optimization techniques may be advantageous in problems characterized by highly multimodal objective functions or poorly defined initial conditions, the present adjustment problem is structurally constrained by physically well-defined parameters and analytically derived gradients. Under these conditions, the Levenberg–Marquardt method provides a computationally efficient and deterministic solution, eliminating the need for stochastic exploration of the parameter space while remaining fully consistent with established practices in nuclear data analysis.

1.1. Covariance-based adjustment framework

This study aims to improve the estimation of parameters involved in the k_0 -NAA method. The adjustment framework adopted in this study, hereafter referred to as *AKQFit*, is based on Levenberg–Marquardt nonlinear optimization for the simultaneous determination of α , k_0 , and Q_0 .

A large portion of previous applications, as discussed, still handle parameter optimization in a sequential manner, with limited scope or assuming only partial correlations. The *AKQFit* adjustment strategy developed in this work is intended to address these limitations by integrating covariance analysis with the Levenberg–Marquardt algorithm within a unified global fitting model. Unlike traditional approaches, *AKQFit* enables the simultaneous determination of α , k_0 , and Q_0 , explicitly incorporating all uncertainties and their mutual correlations in a single step, resulting in a more realistic parameter estimation aligned with contemporary nuclear data evaluation practices.

The *AKQFit* approach incorporates parameter uncertainties and their mutual correlations within a covariance-based formalism, extending the covariance analysis previously developed by Dias et al. (2010). In this framework, uncertainties are treated as intrinsic components of the adjustment procedure, allowing consistent propagation of covariance effects throughout the parameter estimation process.

The main contribution of this work is the application of a covariance-based adjustment framework to the treatment of partial uncertainties associated with the k_0 method, with explicit consideration of equations that are not usually handled simultaneously in integrated adjustment procedures. This framework enables a more consistent treatment of uncertainties and parameter correlations than conventional methods. The adopted adjustment framework was evaluated through analysis of both historical irradiation data and new experimental irradiations specifically performed at the IPEN-CNEN/SP IEA-R1 research reactor. The results contribute to improving the reliability of parameter calculations within the k_0 -NAA community.

Although modern analytical methodologies increasingly rely on advanced computational tools, the literature still contains relatively few integrated adjustment frameworks for the simultaneous determination of the α , k_0 , and Q_0 parameters within the k_0 -NAA formalism. In this context, the present study describes a covariance-based global adjustment framework that integrates covariance analysis and nonlinear least-squares optimization into a single procedure for the simultaneous estimation of key k_0 -NAA parameters. The approach employs a Levenberg–Marquardt regression scheme to improve parameter estimation while explicitly accounting for parameter correlations.

The contributions of this work provide the scientific community with evaluated values of k_0 and Q_0 that were previously unavailable or considered questionable for practical use (KELLETT, 2009). This advances the reliability and precision of neutron activation analysis.

1.2. Practical implementation of the adjustment procedure

For practical application, the adjustment framework described here can be summarized as a sequential workflow that links experimental observables to the nonlinear adjustment procedure. First, the experimental quantities required for the k_0 -NAA formalism are evaluated, including specific activities, efficiency corrections, neutron spectrum parameters, and associated uncertainties. These quantities are then used to construct the observation vector $\tilde{Y}$ and the planning matrix $\mathbf{X}$ according to the mathematical relations described in the next sections.

The covariance matrix V_y is assembled by considering the main uncertainty contributions, such as counting statistics, detector efficiency calibration, neutron flux parameters, and nuclear data. Correlations between parameters are incorporated whenever applicable, ensuring consistency with the global adjustment strategy.

Once the matrices are defined, the nonlinear least-squares problem is solved using a Levenberg–Marquardt optimization scheme, allowing

the simultaneous estimation of α , k_0 , and Q_0 . The adjustment procedure iteratively updates the parameter vector until convergence criteria based on residual minimization are satisfied.

This workflow provides a practical pathway for implementing the adjustment framework in different experimental environments while preserving the covariance structure required for consistent uncertainty propagation.

2. Methodology

The proposed method involves analyzing and determining correlations between parameters through their functional relationships. Constructing the covariance matrix requires substantial computational effort. The calculations were performed using the R programming language because of its computational efficiency and practical analytical resources.

The implementation of the method consisted of identifying the set of equations linking the three parameters: α , Q_0 , and k_0 . The parameter α describes the neutron spectrum shape factor, characterizing deviations of the epithermal neutron flux from the ideal $1/E$ dependence. The quantity k_0 represents the nuclear constant used in the k_0 -NAA standardization method, relating the gamma-ray emission probability and nuclear parameters of a given radionuclide to those of the gold comparator. The parameter Q_0 corresponds to the ratio between the resonance integral and the thermal neutron cross section, describing the relative contribution of epithermal neutrons in the activation process. In order to enable a unified adjustment procedure, the equations were algebraically reorganized by grouping terms dependent solely on experimental observables on the left-hand side, while terms containing the adjustment parameters were placed on the right-hand side. This formulation allowed the construction of the matrices required for the least-squares solution within a matrix formalism. Based on the equations for α , k_0 , and Q_0 (De Corte, 1987), a vector $\vec{Y}$ was defined to incorporate the three relations simultaneously.

$$\vec{Y} = \begin{bmatrix} \frac{(A_{sp,i})Cd}{\epsilon_{p,i} F_{Cd,i} G_{ep,i}} \\ \frac{(F_{Cd,Au} R_{Cd,Au} - 1) G_{th,i} G_{ep,Au}}{(F_{Cd,i} R_{Cd,i} - 1) G_{th,Au} G_{ep,i}} \\ \frac{(A_{sp,i})Cd}{F_{Cd,i}} \cdot \frac{G_{th,Au} \epsilon_{p,Au}}{A_{sp,Au} - \frac{(A_{sp,Au})Cd}{F_{Cd,Au}}} \\ \frac{(A_{sp,Au})Cd}{F_{Cd,Au}} \cdot \frac{G_{th,i} \epsilon_{p,i}}{A_{sp,i} - \frac{(A_{sp,i})Cd}{F_{Cd,i}}} \end{bmatrix} = \begin{bmatrix} e^{\alpha} \cdot E_{r,i}^{2\alpha} \cdot k_{0Au,i,j} \left[\left(\frac{Q_{0,i} - 0.429}{E_{r,i}^{\alpha}} \right) + \frac{0.429}{(2\alpha+1) \cdot 0.55^{\alpha}} \right] \\ \left(\frac{Q_{0,i} - 0.429}{E_{r,i}^{\alpha}} + \frac{0.429}{(2\alpha+1) \cdot 0.55^{\alpha}} \right) \\ \left(\frac{Q_{0,Au} - 0.429}{E_{r,Au}^{\alpha}} + \frac{0.429}{(2\alpha+1) \cdot 0.55^{\alpha}} \right) \\ k_{0Au,i,j} \end{bmatrix} \quad (1)$$

All symbols appearing in Eq. (1) are defined as follows. The terms $A_{sp,i}$ and $(A_{sp,i})Cd$ represent the specific activity of the sample for the i th radionuclide measured without and with cadmium cover, respectively, while $\epsilon_{p,i}$ corresponds to the absolute peak detection efficiency of the HPGe detector. The factor $F_{Cd,i}$ denotes the cadmium correction factor and $R_{Cd,i}$ represents the cadmium ratio. The parameters $G_{ep,i}$ and $G_{th,i}$ correspond to the correction factors associated with epithermal and thermal neutrons, respectively. The energy $E_{r,i}$ denotes the effective resonance energy for each gamma transition. The index i refers to the analyzed sample, whereas Au indicates the gold comparator used in the k_0 -NAA formalism. The quantity a represents the intercept of the regression line of $Y_i = a + \alpha X_i$ for the classical α calculation, where $X_i = \ln E_{r,i}$ and:

$$Y_i = \ln \left(\frac{(E_{r,i})^{-\alpha} (A_{sp,i})Cd}{k_{0Au,i,j} \cdot \epsilon_{p,i} \cdot F_{Cd,i} \cdot Q_{0,i}(\alpha) \cdot G_{ep,i}} \right) \quad (2)$$

Although a does not represent an independent nuclear parameter, it is incorporated into the nonlinear adjustment formulation to preserve mathematical consistency and proper covariance propagation. The term $Q_{0,i}(\alpha)$ represents the resonance integral to thermal cross-section ratio corrected for the epithermal flux deviation parameter α . The numerical constants (0.429 and 0.55) arise from the conventional Westcott formalism adopted in k_0 -NAA and are associated with the epithermal flux correction factors as described in De Corte (1987). All symbols follow the standard notation commonly adopted in k_0 -NAA.

The upper- $\vec{Y}$ (Y^{upper}) refers to the α equation, the middle- $\vec{Y}$ (Y^{middle}) refers to the Q_0 equation, and the lower- $\vec{Y}$ (Y^{lower}) refers to the k_0 equation. The indices i , j , and Au refer to the sample, the gamma transition energy of the sample, and gold (comparator), respectively.

The vector $\vec{A}$, which contains the adjustment parameters, is:

$$\vec{A} = \begin{bmatrix} a \\ \alpha \\ Q_{0,i} \\ k_{0,i,j} \end{bmatrix} \quad (3)$$

The planning matrix X is formed by the partial derivatives of the functions with respect to the parameters:

$$X = \begin{bmatrix} \frac{\partial Y^{upper}}{\partial a} & \frac{\partial Y^{upper}}{\partial \alpha} & \frac{\partial Y^{upper}}{\partial Q_{0,i}} & \frac{\partial Y^{upper}}{\partial k_{0,i,j}} \\ 0 & \frac{\partial Y^{middle}}{\partial a} & \frac{\partial Y^{middle}}{\partial Q_{0,i}} & 0 \\ 0 & 0 & 0 & \frac{\partial Y^{lower}}{\partial k_{0,i,j}} \end{bmatrix} \quad (4)$$

The indices i and j depend on the amount of data and gamma transitions of the isotope, making it necessary to expand matrix X in indices i and j accordingly. The matrix V_y is composed of the variances and covariances of each parameter of $\vec{A}$. For its construction, all non-zero covariance terms of each equation from the first part of $\vec{Y}$ were considered. Therefore, the matrix takes the form:

$$V_y = \begin{bmatrix} \text{cov}(Y^{upper}, Y^{upper}) & \text{cov}(Y^{upper}, Y^{middle}) & \text{cov}(Y^{upper}, Y^{lower}) \\ \text{cov}(Y^{middle}, Y^{upper}) & \text{cov}(Y^{middle}, Y^{middle}) & \text{cov}(Y^{middle}, Y^{lower}) \\ \text{cov}(Y^{lower}, Y^{upper}) & \text{cov}(Y^{lower}, Y^{middle}) & \text{cov}(Y^{lower}, Y^{lower}) \end{bmatrix} \quad (5)$$

The complexity of the V_y matrix depends on the number of samples included in the analysis. As the number of analyzed elements increases, additional parameters must be considered, leading to a corresponding increase in the size of the V_y matrix (see Fig. 1).

This structure allows the use of comparators from different irradiations (different Au), thereby enabling the inclusion of all combinations of targets, energies, and dates in a single adjustment, with all their possible correlations. This adds even more value to the initial proposal, providing great flexibility and comprehensiveness to the adjustment.

2.1. The nonlinear adjustment of parameters

The Levenberg–Marquardt method is a well-established optimization algorithm for parameter estimation in nonlinear mathematical models (Levenberg, 1944; Marquardt, 1963), particularly suited for complex curve fitting and optimization problems. To apply the method, the values of $\vec{A}$ were initially defined using the conventional method based on COVAR (Dias et al., 2010), and, with the experimental data, the values of $\vec{Y}_{exp}$ (left side of $\vec{Y}$ of Eq. (1)) and the adjusted values of $\vec{Y}_{adju}$ (right-hand side of $\vec{Y}$ of Eq. (1)) were obtained. The difference between $\vec{Y}_{exp}$ and $\vec{Y}_{adju}$ was:

$$\vec{D}_{actual} = \vec{Y}_{exp} - \vec{Y}_{adju} \quad (6)$$

The Levenberg–Marquardt method was then applied to minimize the difference between the experimental and adjusted values, iteratively adjusting the parameters in $\vec{A}$ to find the best fit. The method

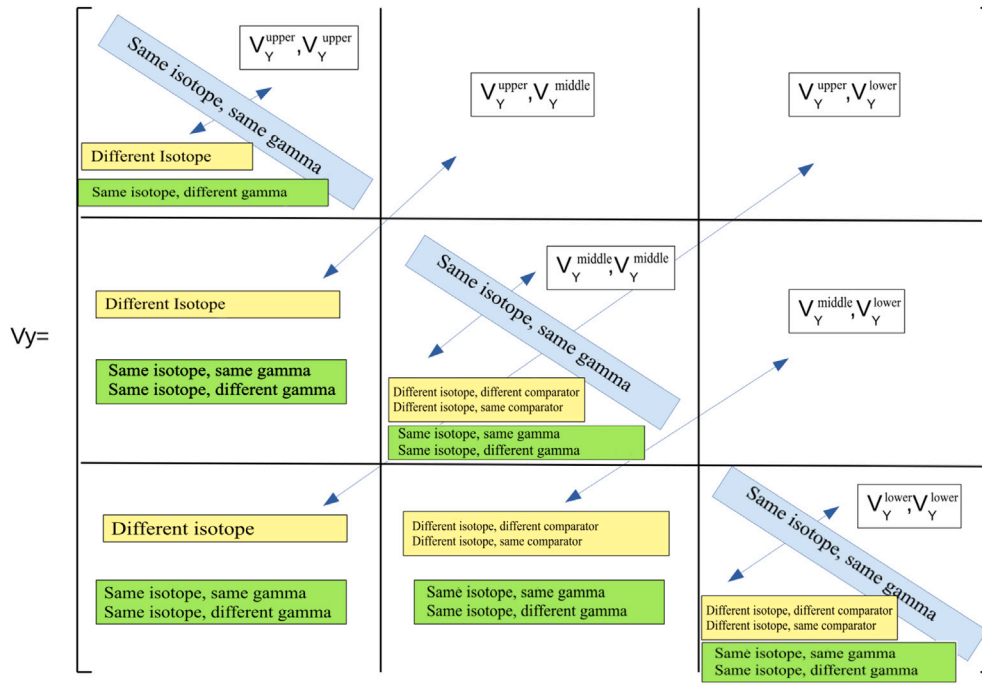


Fig. 1. Planning of the derivatives of the V_y matrix, divided into 9 quadrants, each with its correction condition.

combines the gradient descent and Gauss–Newton methods, allowing efficient convergence to the optimal parameter values. The adjustment of the parameters was performed using the following equation:

With matrix X from Eq. (4) and the matrix V_y from Eq. (5), the matrix $R(\lambda)$ was constructed. The matrix $R(\lambda)$ consists of the matrix $X^T \cdot V_y^{-1} \cdot X$, with its diagonal multiplied by $1 + \lambda$, where λ is the damping parameter of the Levenberg–Marquardt algorithm. The parameter λ , which is an adjustment coefficient, varies during the iterations of the method, increasing or decreasing accordingly. Thus, the expression for $\bar{A}_{new}$ was obtained as follows:

$$\bar{A}_{new} = \bar{A}' + \Delta \bar{A} \quad (7)$$

where

$$\Delta \bar{A} = R(\lambda)^{-1} X^T V_y^{-1} \bar{Y}^T \quad (8)$$

and $\bar{Y}_{adj(new)}$ is calculated by:

$$\bar{Y}_{adj(new)} = X \cdot \Delta \bar{A} \quad (9)$$

2.2. Determination of the gamma-ray detection efficiency curve

The HPGe gamma-ray efficiency curve has been determined using standard sources ^{133}Ba , ^{60}Co , ^{137}Cs , and ^{152}Eu supplied by the IAEA, in the energy range from 121 to 1408 keV. In order to minimize the influence of cascade summation corrections, the radioactive source was positioned approximately 17.9 cm from the front face of the HPGe detector.

2.3. Sample preparation

Zirconium samples (Aldrich Chemical Company, purity 99.98%, thickness 0.25 mm), alloys of Au (0.10% in Al, IRMM-530RC) and Gd (SPEX Certificate, 1000(1) mg/L concentration) were used in the irradiations. Furthermore, irradiation campaigns performed in 2010 (Dias et al., 2010) and 2015 (Barros et al., 2019), originally carried out within doctoral research projects at IPEN, were combined with the 2023 experimental data to create a larger dataset with more points for

the adjustment. This procedure was possible because all irradiations were performed at exactly the same position in the reactor, allowing a common value of α to be assumed for all campaigns. The results indicated that this assumption was consistent with the experimental observations.

In the present work, a single α parameter was assumed for all irradiation campaigns, since the samples were irradiated at the same reactor position under comparable operating conditions. In k_0 -NAA, the parameter α characterizes the epithermal neutron spectrum shape and is primarily determined by the irradiation channel rather than by the individual sample. Therefore, the use of a common α value is justified when experiments are performed in a stable and well-characterized irradiation site. Small variations in reactor conditions are expected to introduce only minor deviations, which are effectively absorbed within the covariance-based adjustment framework. In this context, the application of the AKQFit approach is appropriate, as it was originally developed for situations involving consistent irradiation geometries and shared neutron spectrum characteristics.

3. Results

3.1. HPGe efficiency curve

Fig. 2 shows the experimental behavior of the peak efficiency of the HPGe spectrometer as a function of gamma-ray energy. Markers represent the experimental data points, while the dotted curve corresponds to a fourth-degree polynomial fit applied on the logarithmic scale (log–log). The parameters and corresponding covariance matrix of the polynomial fit are shown in Table 1.

The peak efficiency as a function of the gamma-ray energy can be calculated using the polynomial equation. The dimensionless fitting parameters (a_i) and their corresponding covariance matrix are presented in Table 1.

3.2. AKQFit

The results obtained by the proposed AKQFit method are presented in Tables 2 and 3. The Zeta-Score ζ_i was introduced to evaluate

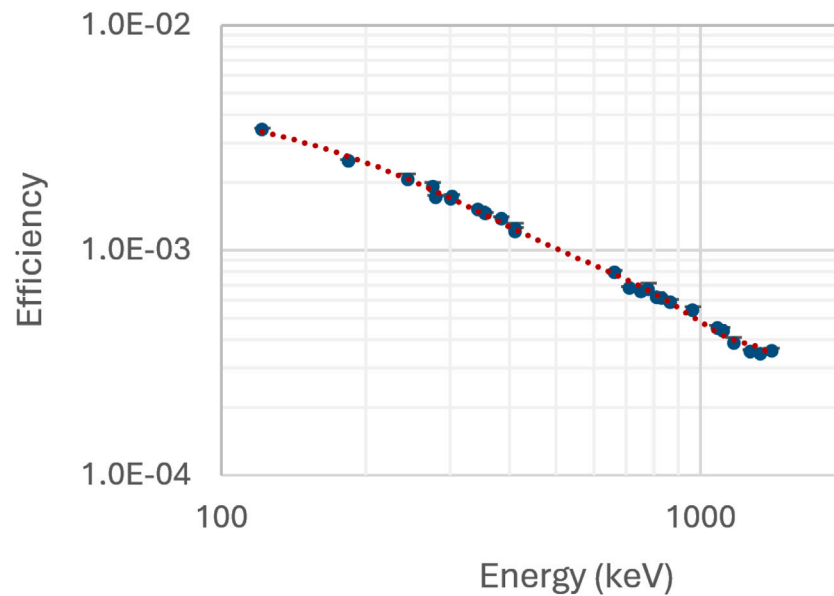


Fig. 2. Experimental HPGe peak efficiency, as a function of the gamma-ray energy. The energy interval is 121–1408 keV, in log–log scale.

Table 1

Efficiency parameters and corresponding covariance matrix of the polynomial fitting with 24 points. The reduced chi-squared was $\chi^2/L = 1.290$, where L is the number of degrees of freedom of the fit.

Fitting parameters	Value	Covariance matrix					
a_0	-3.356×10^{-1}	3.039×10^{-1}					
a_1	3.383×10^{-1}	-2.597×10^{-2}	1.722×10^{-2}				
a_2	6.145×10^{-2}	6.355×10^{-3}	-4.227×10^{-3}	1.041×10^{-3}			
a_3	6.904×10^{-3}	-6.840×10^{-4}	4.564×10^{-4}	-1.127×10^{-4}	1.222×10^{-5}		
a_4	2.868×10^{-4}	2.700×10^{-5}	-1.800×10^{-5}	4.531×10^{-6}	-4.924×10^{-7}	1.987×10^{-8}	

the dispersion of the value obtained from AKQFit compared to the k_0 -Database (Ghent University, 2020), and is expressed as:

$$\zeta_i = \frac{X_i - X_j}{\sqrt{u^2(X_i) + u^2(X_j)}} \quad (10)$$

where

X_i is the obtained value

X_j is the assigned value

$u(X_i)$ is the uncertainty of its result X_i

$u(X_j)$ is the uncertainty of its result X_j

A threshold of $|\zeta| < 3.0$ was adopted, corresponding to a confidence level of 99%, minimizing the probability of type I errors in the comparison.

The experimental determination of Q_0 values for radionuclides ^{140}La and ^{56}Sc , for which uncertainty data were previously unavailable in the k_0 -Database, were $1.21(13)$ and $4.99(6) \times 10^{-1}$, respectively. The Zeta-Score could not be calculated due to the absence of uncertainty data in the k_0 -Database. For ^{159}Gd , the obtained Q_0 value was $2.89(11) \times 10^1$, demonstrating excellent agreement with the reference value, as indicated by a Zeta-Score close to zero.

Table 3 presents the experimentally determined k_0 values from irradiations conducted in 2010, 2015, and 2023, comparing the values derived from AKQFit with those of the established k_0 -Database. The calculated Zeta-Scores indicate the level of agreement between these two sets of values. Generally, the majority of radionuclides exhibited Zeta-Scores within ± 2 , indicating satisfactory agreement and supporting the reliability of the AKQFit framework. However, some of the radionuclides of the 2015 irradiation exhibited a high absolute Zeta-Score, suggesting discrepancies that could warrant further methodological review or a reevaluation of the values associated with the database, as verified by Barros et al. (2019). For ^{159}Gd , the k_0 value obtained

Table 2

Q_0 values for radionuclides from 2010 (Dias et al., 2010), 2015 (Barros et al., 2019), and 2023 (this work) irradiations. The numbers inside parentheses correspond to uncertainties in the last digits. Agreement was considered for Zeta-Scores below 3.0 (99% confidence interval).

Year	Radionuclide	AKQFit	k_0 -Database (Ghent University, 2020)	Zeta-Score
2010	^{95}Zr	5.01(13)	5.31(17)	0.42
	^{65}Zn	1.94(7)	1.90(9)	0.35
	$^{69\text{m}}\text{Zn}$	2.71(8)	3.19(4)	-1.35
	^{140}La	1.21(13)	1.24()	
	^{60}Co	2.16(5)	1.99(6)	-0.24
	^{56}Sc	$4.99(6) \times 10^{-1}$	$4.30() \times 10^{-1}$	
2015	^{75}Se	$1.22(4) \times 10^1$	$1.10(3) \times 10^1$	2.78
	^{192}Ir	3.21(11)	3.70(7)	-3.50
	^{114}In	$2.44(6) \times 10^1$	$2.44(5) \times 10^1$	0.04
	^{187}W	$1.29(6) \times 10^1$	$1.37(3) \times 10^1$	-1.20
2023	^{95}Zr	5.26(16)	5.31(17)	-0.23
	^{159}Gd	$2.89(11) \times 10^1$	$2.99(9) \times 10^1$	-0.69

with AKQFit at 363.5 keV was $8.58(15) \times 10^{-4}$, close to the reference value from the k_0 -Database. The Zeta-Score of 0.47 obtained indicates a high concordance between the experimental result and the database reference.

Additionally, it was noted that the reference database lacked the k_0 value for ^{187}W at an energy of 625 keV, which highlights the significance of the new data obtained in this study. The agreement between the values obtained by AKQFit and the reference values was evaluated using the Zeta-Score parameter, where absolute values below 2.0 indicate satisfactory agreement. However, elevated Zeta-Score values

Table 3

k_0 values for radionuclides from 2010 (Dias et al., 2010), 2015 (Barros et al., 2019), and 2023 (this work) irradiations. The numbers inside parentheses correspond to uncertainties in the last digits. Agreement was considered for Zeta-Scores below 3.0 (99% confidence interval).

Year	Radionuclide	Energy (keV)	AKQFit	k_0 -Database (Ghent University, 2020)	Zeta-Score
2010	^{95}Zr	724.2	$8.86(11) \times 10^{-5}$	$8.90(12) \times 10^{-5}$	-0.24
		756.7	$1.09(26) \times 10^{-4}$	$1.10(14) \times 10^{-4}$	-0.20
	^{65}Zn	1115.5	$5.76(4) \times 10^{-3}$	$5.72(2) \times 10^{-3}$	0.88
		438.6	$3.88(9) \times 10^{-4}$	$3.98(24) \times 10^{-4}$	-1.00
	^{140}La	328.7	$2.86(3) \times 10^{-2}$	$2.87(3) \times 10^{-2}$	-0.21
		487.0	$6.33(4) \times 10^{-2}$	$6.37(6) \times 10^{-2}$	-0.47
		815.8	$3.29(2) \times 10^{-2}$	$3.32(2) \times 10^{-2}$	-0.84
		1596.2	$1.33(2) \times 10^{-1}$	$1.34(1) \times 10^{-1}$	-0.15
	^{60}Co	1173.2	1.30(23)	1.28(9)	0.10
		1332.5	1.33(10)	1.32(7)	0.17
	^{46}Sc	889.0	1.20(11)	1.22(8)	-1.25
		1120.0	1.21(15)	1.22(9)	-0.49
2015	^{75}Se	121.12	$2.14(3) \times 10^{-3}$	$2.18(3) \times 10^{-3}$	-0.93
		136.0	$7.55(15) \times 10^{-3}$	$7.33(9) \times 10^{-3}$	1.28
		264.66	$7.61(12) \times 10^{-3}$	$7.53(9) \times 10^{-3}$	0.54
		279.54	$3.29(5) \times 10^{-3}$	$3.18(4) \times 10^{-3}$	0.68
		400.7	$1.52(3) \times 10^{-3}$	$1.44(2) \times 10^{-3}$	2.61
		295.96	1.17(2)	1.14(2)	
		316.51	3.38(4)	3.26(4)	2.33
		468.07	1.96(3)	1.87(2)	3.55
	^{114}In	190.3	$1.02(23) \times 10^{-3}$	$1.06(8) \times 10^{-3}$	-2.03
		558.4	$2.72(31) \times 10^{-4}$	$2.86(20) \times 10^{-4}$	-2.83
		725.2	$2.72(29) \times 10^{-4}$	$2.90(17) \times 10^{-4}$	-4.78
		479.53	$3.21(6) \times 10^{-3}$	$2.97(3) \times 10^{-3}$	3.47
	^{187}W	551.53	$7.32(12) \times 10^{-3}$	$6.91(4) \times 10^{-3}$	3.24
		618.36	$9.16(7) \times 10^{-3}$	$8.65(4) \times 10^{-3}$	2.83
		625.51	$1.58(3) \times 10^{-3}$		
		685.77	$3.99(8) \times 10^{-2}$	$3.71(2) \times 10^{-2}$	3.50
		772.89	$5.99(12) \times 10^{-3}$	$5.61(4) \times 10^{-3}$	3.08
2023	^{95}Zr	724.2	$8.94(14) \times 10^{-5}$	$8.90(12) \times 10^{-5}$	0.22
		756.7	$1.11(2) \times 10^{-4}$	$1.10(14) \times 10^{-4}$	0.39
	^{159}Gd	363.5	$8.58(15) \times 10^{-4}$	$8.49(13) \times 10^{-4}$	0.47

were observed for some radionuclides, such as Se, In, and Ir, justifying further experimental measurements for future database updates.

4. Conclusion

This study addresses previous methodological limitations through AKQFit, a unified nonlinear covariance-based adjustment framework for neutron activation analysis. Unlike sequential or partially correlated treatments, AKQFit explicitly embeds the covariance matrix into a single nonlinear least-squares optimization procedure, allowing for the simultaneous determination of the α , Q_0 , and k_0 parameters. By accommodating experimental datasets from multiple irradiation campaigns under a common formalism, this strategy extends previous approaches and could improve internal consistency and enhance uncertainty propagation. The analysis demonstrated that AKQFit can yield realistic values by incorporating all correlations and covariances in all three principal parameters of k_0 -NAA, demonstrating the effectiveness of the proposed framework when compared to traditional methods. This work also directly contributes to the refinement of the k_0 -Database by introducing new and updated reference values for ^{159}Gd , ^{187}W , ^{140}La and ^{56}Sc .

Nevertheless, a current limitation of the algorithm must be acknowledged. The present model assumes a constant α parameter across all combined irradiations. Consequently, if the irradiation position within the reactor changes or if the true α value exhibits significant variability between experiments, the global adjustment may introduce inaccuracies by converging on an averaged α . The Levenberg–Marquardt algorithm presents some inherent limitations that should be considered in nonlinear adjustment problems. The method remains fundamentally a local optimization technique, meaning that convergence to the global

minimum is not guaranteed if the objective function contains multiple minima or if the initial parameter estimates are poorly chosen. In addition, the performance of the Levenberg–Marquardt algorithm depends on the proper scaling of parameters and residuals, since large differences in magnitude may lead to numerical instability or slow convergence. Consequently, careful initialization, parameter normalization, and validation of convergence criteria are essential to ensure consistent results when applying the method to complex nuclear data adjustment problems.

For future studies, we suggest extending the AKQFit framework to address this limitation, as well as to include the calculation of elemental concentrations and the addition of a new parameter: the thermal-to-epithermal neutron flux ratio. Furthermore, future work will also focus on a deeper analysis of parameter correlations and the evaluation of convergence robustness under varying experimental conditions. Finally, re-implementing the framework in Python may facilitate the use of more advanced computational data-analysis techniques and broaden its applications within the NAA community.

CRediT authorship contribution statement

Rafael Vanhoz Ribeiro: Writing – original draft. **Maurício Morales:** Supervision. **Mauro da Silva Dias:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

$$\frac{\partial Y^{middle}}{\partial \alpha} = \frac{\left(\frac{\ln(E_{r,i})(Q_{0,i}+0.429)}{E_{r,i}^\alpha} + \frac{0.2565}{(2\alpha+1)0.55^\alpha} - \frac{0.858}{(2\alpha+1)^2 0.55^\alpha} \right)}{\left(\frac{(Q_{0,Au}-0.429)}{E_{r,Au}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right)} - \quad (A.5)$$

$$\frac{\left(\frac{\ln(E_{r,Au})(Q_{0,Au}+0.429)}{E_{r,Au}^\alpha} + \frac{0.2565}{(2\alpha+1)0.55^\alpha} - \frac{0.858}{(2\alpha+1)^2 0.55^\alpha} \right) \left(\frac{(Q_{0,i}-0.429)}{E_{r,i}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right)}{\left(\frac{(Q_{0,Au}-0.429)}{E_{r,Au}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right)^2} \quad (A.6)$$

Box 1.

Appendix A. Partial derivatives of the Y^{upper} , Y^{middle} , and Y^{lower} equationsA.1. Partial derivatives of Y^{upper}

Partial derivative of Y^{upper} with respect to α

$$\frac{\partial Y^{upper}}{\partial \alpha} = e^a \cdot E_{r,i}^{2\alpha} \cdot k_{0Au,i,j} \left[\frac{(Q_{0,i} - 0.429)}{E_{r,i}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right] \quad (A.1)$$

Partial derivative of Y^{upper} with respect to α

$$\begin{aligned} \frac{\partial Y^{upper}}{\partial \alpha} = & 2e^a \cdot \ln(E_{r,i}) E_{r,i}^{2\alpha} \cdot k_{0Au,i,j} \left[\frac{(Q_{0,i} - 0.429)}{E_{r,i}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right] \\ & + e^a E_{r,i}^{2\alpha} \cdot k_{0Au,i,j} \left[\frac{\ln(E_{r,i})(Q_{0,i} + 0.429)}{E_{r,i}^\alpha} + \frac{0.2565}{(2\alpha+1)0.55^\alpha} \right. \\ & \left. - \frac{0.858}{(2\alpha+1)^2 0.55^\alpha} \right] \end{aligned} \quad (A.2)$$

Partial derivative of Y^{upper} with respect to Q_0

$$\frac{\partial Y^{upper}}{\partial Q_{0,i}} = e^a \cdot E_{r,i}^{2\alpha} \cdot k_{0Au,i,j} \quad (A.3)$$

Partial derivative of Y^{upper} with respect to k_0

$$\frac{\partial Y^{upper}}{\partial k_{0Au,i,j}} = e^a \cdot E_{r,i}^{2\alpha} \left[\frac{(Q_{0,i} - 0.429)}{E_{r,i}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right] \quad (A.4)$$

A.2. Partial derivatives of Y^{middle}

Partial derivative of Y^{middle} with respect to α (see Eqs. (A.5) and (A.6) in Box 1).

Partial derivative of Y^{middle} with respect to Q_0

$$\frac{\partial Y^{middle}}{\partial Q_{0,i}} = \frac{1}{E_{r,i}^\alpha \left[\frac{(Q_{0,Au}-0.429)}{E_{r,Au}^\alpha} + \frac{0.429}{(2\alpha+1)0.55^\alpha} \right]} \quad (A.7)$$

A.3. Partial derivatives of Y^{lower}

Partial derivative of Y^{lower} with respect to k_0

$$\frac{\partial Y^{lower}}{\partial k_{0,i,j}} = 1 \quad (A.8)$$

Appendix B. Construction of the covariance matrix V_y

The covariance matrix V_y was constructed by explicitly incorporating the eighteen analytical expressions presented above, which include all diagonal and off-diagonal terms associated with the upper, intermediate, and lower components under the different isotope and gamma-transition configurations. The eighteen covariance expressions correspond to the complete expansion of Eq. (5) under all isotope/transition configurations.

1. (Y^{upper} , Y^{upper}) – diagonal term

The quantities that contain uncertainties are $(A_{sp,i})_{Cd}$, $\epsilon_{p,i}$, $F_{Cd,i}$, and $G_{ep,i}$. Therefore:

$$\begin{aligned} V_y(Y_i^{upper}, Y_i^{upper}) = & \left(\frac{\partial Y_i^{upper}}{\partial (A_{sp,i})_{Cd}} \right)^2 \sigma_{(A_{sp,i})_{Cd}}^2 + \left(\frac{\partial Y_i^{upper}}{\partial \epsilon_{p,i}} \right)^2 \sigma_{\epsilon_{p,i}}^2 \\ & + \left(\frac{\partial Y_i^{upper}}{\partial F_{Cd,i}} \right)^2 \sigma_{F_{Cd,i}}^2 + \left(\frac{\partial Y_i^{upper}}{\partial G_{ep,i}} \right)^2 \sigma_{G_{ep,i}}^2 \end{aligned} \quad (B.1)$$

2. (Y^{upper} , Y^{upper}) – off-diagonal term, different isotopes

Considering two different isotopes, the only correlated term corresponds to the detector efficiency of each gamma transition. Therefore,

$$V_y(Y_i^{upper}, Y_j^{upper}) = \left(\frac{\partial Y_i^{upper}}{\partial \epsilon_{p,i}} \right) \left(\frac{\partial Y_j^{upper}}{\partial \epsilon_{p,j}} \right) \text{COV}(\epsilon_{p,i}, \epsilon_{p,j}) \quad (B.2)$$

3. (Y^{upper} , Y^{upper}) – off-diagonal term, same isotope, different gamma transition

The expression for V_y in this case is given by

$$\begin{aligned} V_y(Y_i^{upper}, Y_j^{upper}) = & \left(\frac{\partial Y_i^{upper}}{\partial \epsilon_{p,i}} \right) \left(\frac{\partial Y_j^{upper}}{\partial \epsilon_{p,j}} \right) \text{COV}(\epsilon_{p,i}, \epsilon_{p,j}) \\ & + \left(\frac{\partial Y_i^{upper}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{upper}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\ & + \left(\frac{\partial Y_i^{upper}}{\partial G_{ep,i}} \right) \left(\frac{\partial Y_j^{upper}}{\partial G_{ep,j}} \right) \sigma_{G_{ep,i}}^2 \end{aligned} \quad (B.3)$$

4. (Y^{middle} , Y^{middle}) – diagonal term

$$\begin{aligned} V_y(Y_i^{middle}, Y_i^{middle}) = & \left(\frac{\partial Y_i^{middle}}{\partial R_{Cd,i}} \right)^2 \sigma_{R_{Cd,i}}^2 + \left(\frac{\partial Y_i^{middle}}{\partial R_{Cd,Au}} \right)^2 \sigma_{R_{Cd,Au}}^2 \\ & + \left(\frac{\partial Y_i^{middle}}{\partial F_{Cd,i}} \right)^2 \sigma_{F_{Cd,i}}^2 + \left(\frac{\partial Y_i^{middle}}{\partial G_{ep,i}} \right)^2 \sigma_{G_{ep,i}}^2 \\ & + \left(\frac{\partial Y_i^{middle}}{\partial G_{th,i}} \right)^2 \sigma_{G_{th,i}}^2 + \left(\frac{\partial Y_i^{middle}}{\partial F_{Cd,Au}} \right)^2 \sigma_{F_{Cd,Au}}^2 \\ & + \left(\frac{\partial Y_i^{middle}}{\partial G_{ep,Au}} \right)^2 \sigma_{G_{ep,Au}}^2 + \left(\frac{\partial Y_i^{middle}}{\partial G_{th,Au}} \right)^2 \sigma_{G_{th,Au}}^2 \end{aligned} \quad (B.4)$$

5. $(Y^{\text{middle}}, Y^{\text{middle}})$ – off-diagonal term, different isotopes

$$\begin{aligned}
V_y(Y_i^{\text{middle}}, Y_j^{\text{middle}}) = & \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{ep,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{ep,Au}} \right) \sigma_{G_{ep,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2
\end{aligned} \quad (B.5)$$

6. $(Y^{\text{middle}}, Y^{\text{middle}})$ – off-diagonal term, same isotope, different gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{middle}}, Y_j^{\text{middle}}) = & \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{ep,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{ep,Au}} \right) \sigma_{G_{ep,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{ep,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{ep,j}} \right) \sigma_{G_{ep,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{th,j}} \right) \sigma_{G_{th,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial R_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial R_{Cd,Au}} \right) \sigma_{R_{Cd,Au}}^2
\end{aligned} \quad (B.6)$$

7. $(Y^{\text{lower}}, Y^{\text{lower}})$ – diagonal term

$$\begin{aligned}
V_y(Y_i^{\text{lower}}, Y_i^{\text{lower}}) = & \left(\frac{\partial Y_i^{\text{lower}}}{\partial A_{sp,i}} \right)^2 \sigma_{A_{sp,i}}^2 + \left(\frac{\partial Y_i^{\text{lower}}}{\partial A_{sp,Au}} \right)^2 \sigma_{A_{sp,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial (A_{sp,i})_{Cd}} \right)^2 \sigma_{(A_{sp,i})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right)^2 \sigma_{(A_{sp,Au})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial F_{Cd,i}} \right)^2 \sigma_{F_{Cd,i}}^2 + \left(\frac{\partial Y_i^{\text{lower}}}{\partial F_{Cd,Au}} \right)^2 \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial G_{th,i}} \right)^2 \sigma_{G_{th,i}}^2 + \left(\frac{\partial Y_i^{\text{lower}}}{\partial G_{th,Au}} \right)^2 \sigma_{G_{th,Au}}^2
\end{aligned} \quad (B.7)$$

8. $(Y^{\text{lower}}, Y^{\text{lower}})$ – off-diagonal term, different isotopes

$$\begin{aligned}
V_y(Y_i^{\text{lower}}, Y_j^{\text{lower}}) = & \left(\frac{\partial Y_i^{\text{lower}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \sigma_{(A_{sp,Au})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial A_{sp,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial A_{sp,Au}} \right) \sigma_{A_{sp,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial \epsilon_{p,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial \epsilon_{p,Au}} \right) \sigma_{\epsilon_{p,Au}}^2
\end{aligned} \quad (B.8)$$

9. $(Y^{\text{lower}}, Y^{\text{lower}})$ – off-diagonal term, same isotope, different gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{lower}}, Y_j^{\text{lower}}) = & \left(\frac{\partial Y_i^{\text{lower}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \sigma_{(A_{sp,Au})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial A_{sp,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial A_{sp,Au}} \right) \sigma_{A_{sp,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial \epsilon_{p,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial \epsilon_{p,Au}} \right) \sigma_{\epsilon_{p,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial G_{th,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,j}} \right) \sigma_{G_{th,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{lower}}}{\partial \epsilon_{p,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial \epsilon_{p,j}} \right) \text{COV}(\epsilon_{p,i}, \epsilon_{p,j})
\end{aligned} \quad (B.9)$$

10. $(Y^{\text{upper}}, Y^{\text{middle}})$ – upper and intermediate parts, different isotopes

$$V_y(Y_i^{\text{upper}}, Y_j^{\text{middle}}) = 0 \quad (B.10)$$

11. $(Y^{\text{upper}}, Y^{\text{middle}})$ – upper and intermediate parts, same isotope, same gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{upper}}, Y_j^{\text{middle}}) = & \left(\frac{\partial Y_i^{\text{upper}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial G_{ep,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{ep,j}} \right) \sigma_{G_{ep,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial (A_{sp,i})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial (A_{sp,j})_{Cd}} \right) \sigma_{(A_{sp,i})_{Cd}}^2
\end{aligned} \quad (B.11)$$

12. $(Y^{\text{upper}}, Y^{\text{middle}})$ – upper and intermediate parts, same isotope, different gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{upper}}, Y_j^{\text{middle}}) = & \left(\frac{\partial Y_i^{\text{upper}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial G_{ep,i}} \right) \left(\frac{\partial Y_j^{\text{middle}}}{\partial G_{ep,j}} \right) \sigma_{G_{ep,i}}^2
\end{aligned} \quad (B.12)$$

13. $(Y^{\text{upper}}, Y^{\text{lower}})$ – upper and lower parts, different isotopes

$$V_y(Y_i^{\text{upper}}, Y_j^{\text{lower}}) = 0 \quad (B.13)$$

14. $(Y^{\text{upper}}, Y^{\text{lower}})$ – upper and lower parts, same isotope, same gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{upper}}, Y_j^{\text{lower}}) = & \left(\frac{\partial Y_i^{\text{upper}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial G_{th,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,j}} \right) \sigma_{G_{th,i}}^2
\end{aligned}$$

$$\begin{aligned}
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial (A_{sp,i})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,j})_{Cd}} \right) \sigma_{(A_{sp,i})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial \epsilon_{p,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial \epsilon_{p,j}} \right) \sigma_{\epsilon_{p,j}}^2
\end{aligned} \quad (\text{B.14})$$

15. ($Y^{\text{upper}}, Y^{\text{lower}}$) – upper and lower parts, same isotope, different gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{upper}}, Y_j^{\text{lower}}) & = \left(\frac{\partial Y_i^{\text{upper}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{upper}}}{\partial \epsilon_{p,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial \epsilon_{p,j}} \right) \sigma_{\epsilon_{p,j}}^2
\end{aligned} \quad (\text{B.15})$$

16. ($Y^{\text{middle}}, Y^{\text{lower}}$) – intermediate and lower parts, different isotopes

$$\begin{aligned}
V_y(Y_i^{\text{middle}}, Y_j^{\text{lower}}) & = \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2
\end{aligned} \quad (\text{B.16})$$

17. ($Y^{\text{middle}}, Y^{\text{lower}}$) – intermediate and lower parts, same isotope, same gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{middle}}, Y_j^{\text{lower}}) & = \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial (A_{sp,Au})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \sigma_{(A_{sp,Au})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial A_{sp,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial A_{sp,Au}} \right) \sigma_{A_{sp,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,j}} \right) \sigma_{G_{th,i}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial (A_{sp,i})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,j})_{Cd}} \right) \sigma_{(A_{sp,i})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial A_{sp,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial A_{sp,j}} \right) \sigma_{A_{sp,i}}^2
\end{aligned} \quad (\text{B.17})$$

18. ($Y^{\text{middle}}, Y^{\text{lower}}$) – intermediate and lower parts, same isotope, different gamma transition

$$\begin{aligned}
V_y(Y_i^{\text{middle}}, Y_j^{\text{lower}}) & = \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,Au}} \right) \sigma_{F_{Cd,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,Au}} \right) \sigma_{G_{th,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial (A_{sp,Au})_{Cd}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial (A_{sp,Au})_{Cd}} \right) \sigma_{(A_{sp,Au})_{Cd}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial A_{sp,Au}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial A_{sp,Au}} \right) \sigma_{A_{sp,Au}}^2 \\
& + \left(\frac{\partial Y_i^{\text{middle}}}{\partial F_{Cd,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial F_{Cd,j}} \right) \sigma_{F_{Cd,i}}^2
\end{aligned}$$

$$+ \left(\frac{\partial Y_i^{\text{middle}}}{\partial G_{th,i}} \right) \left(\frac{\partial Y_j^{\text{lower}}}{\partial G_{th,j}} \right) \sigma_{G_{th,i}}^2 \quad (\text{B.18})$$

Data availability

Data will be made available on request.

References

- Barradas, N.P., Farjallah, N., Vieira, A., et al., 2022. Artificial neural networks for NAA: proof of concept on data analyzed with k_0 -based software. J. Radioanal. Nucl. Chem. 332, 3421–3429.
- Barros, L.F., Ribeiro, R.V., Dias, M.S., Morales, M., Semmler, R., Yamazaki, I.M., Koskinas, M.F., 2019. Determination of k_0 and Q_0 for ^{74}Se , ^{113}In , ^{186}W and ^{191}Ir targets applying covariance analysis. Appl. Radiat. Isot. 154, 108846.
- Benedik, L., Repinc, U., 2011. Evaluation of measurement uncertainty components associated with results of radiochemical neutron activation analysis for determination of uranium traces. Accred. Qual. Assur. 16, 637–642.
- Chahidi, H., Bounouira, H., Amsil, H., et al., 2023. Combination of k_0 -IAEA and KayWin for the characterization of neutron flux parameters at Triga Mark II research reactor, Morocco. J. Radioanal. Nucl. Chem. 332, 3499–3505.
- Datta, A., Garg, A.N., Sharma, V., Acharya, R., 2020. Quantification of minor and trace elements in raw and branded turmeric samples using instrumental neutron activation analysis utilizing apsara-u reactor for possible applications to forensic science. Radioanal. Nucl. Chem. 325, 967–975.
- De Corte, F., 1987. The K_0 Standardization Method—a Move To the Optimization of Neutron Activation Analysis (Ph.D. thesis). Rijksuniversiteit, Gent.
- de Lima, R.C., De Nadai Fernandes, E.A., Mazola, Y.T., et al., 2022. Pet food categorization by neutron activation analysis and data science. J. Radioanal. Nucl. Chem. 331, 5433–5440.
- Di Luzio, M., D'Agostino, G., 2022. Developments of the k_0 -NAA measurement model implemented in k_0 -INRIM software. J. Radioanal. Nucl. Chem. 331, 4251–4258.
- Di Luzio, M., D'Agostino, G., 2023. The k_0 -INRIM software version 2.0: presentation and an analysis vademecum. J. Radioanal. Nucl. Chem. 332, 411–420.
- Dias, M.S., Cardoso, V., Koskinas, M.F., Yamazaki, I.M., 2010. Determination of the neutron spectrum shape parameter α in k_0 -NAA methodology using covariance analysis. Appl. Radiat. Isot. 68, 592–595.
- Dias, M.S., Semmler, R., Koskinas, M.F., et al., 2023. k_0 -IPEN: A new software for INAA. Radioanal. Nucl. Chem. 332, 3401–3409.
- Dung, H.M., Thien, T.Q., Van Doanh, H., et al., 2022. Development of a PC program for the k_0 -based epithermal neutron activation analysis. J. Radioanal. Nucl. Chem. 332, 3469–3474.
2020. K_0 -Database 2020. Ghent University and Hungarian Academy of Sciences, Available at: https://www.kayzero.com/k0naa/k0naaorg/Nuclear_Data_SC/Entries/2020/8/24/Update_of_k0-database_1-128.html.
- Huynh, P.T., Tran, T.P.N., Dinh, B.T., et al., 2020. Analysis of trace elements in the fingernails of breast cancer patients using instrumental neutron activation analysis. J. Radioanal. Nucl. Chem. 324, 663–671.
2023. Intercomparison of K_0 -NAA Software Packages, IAEA-TECDOC-2026. INTERNATIONAL ATOMIC ENERGY AGENCY, IAEA, Vienna.
2024. Neutron Activation Analysis. International Atomic Energy Agency (IAEA), Available at: <https://www.iaea.org/topics/neutron-activation-analysis> (Accessed 16 2026).
- KELLETT, M.A., 2009. Reference database for neutron activation analysis. In: IAEA Nuclear Data Section Co-Ordinated Research Project. Available at: <https://www.nds.iaea.org/naa/index.html>.
- Kubešová, M., Kučera, J., 2011. Comparison of kayzero for windows and k_0 -IAEA software packages for k_0 standardization in neutron activation analysis. Nucl. Instrum. Methods Phys. Res. A 654, 206–212.
- Kubešová, M., Kučera, J., 2015. How to calculate uncertainties of neutron flux parameters and uncertainties of analysis results in k_0 -NAA? J. Radioanal. Nucl. Chem. 293, 87–94.
- Levenberg, K., 1944. A method for the solution of certain nonlinear problems in least squares. Quart. Appl. Math. 2, 164–168.
- Marquardt, D.W., 1963. An algorithm for least-squares estimation of nonlinear parameters. SIAM J. Appl. Math. 11, 431–441.
- Okutan, Murat, Üncü, Yiğit Ali, Sevim, Gençay, Şekerci, Mert, Kiliçarslan, Tolga, Demir, Bayram, Özdoğan, Hasan, 2021. Estimation of (n, p) reaction cross sections at 14.5 +0.5 MeV neutron energy by using artificial neural network. Appl. Radiat. Isot. 170, 109584.
- Okutan, Murat, Üncü, Yiğit Ali, Sevim, Gençay, Şekerci, Mert, Kiliçarslan, Tolga, Demir, Bayram, Özdoğan, Hasan, 2026. Artificial neural network-based prediction of production cross sections of the medical radioisotopes ^{67}Ga and ^{89}Zr . Appl. Radiat. Isot. 230, 112461.

- Özdoğan, Hasan, Üncü, Yiğit Ali, Şekerci, Mert, Kaplan, Abdullah, 2023. Estimations for (n, a) reaction cross sections at around 14.5MeV using Levenberg–Marquardt algorithm-based artificial neural network. *Appl. Radiat. Isot.* 192, 110609.
- Özdoğan, Hasan, Üncü, Yiğit Ali, Şekerci, Mert, Kaplan, Abdullah, 2024. Neural network predictions of (a, n) reaction cross sections at 18.5 ± 3 MeV using the Levenberg–Marquardt algorithm. *Appl. Radiat. Isot.* 204, 111115.
- Robouch, P., Arana, G., Eguskiza, M., Pommé, S., Etxebarria, N., 2000. Uncertainty budget for k_0 -NAA. *J. Radioanal. Nucl. Chem.* 245, 195–197.
- Smodiš, B., Bučar, T., 2006. Overall measurement uncertainty of k_0 -based neutron activation analysis. *J. Radioanal. Nucl. Chem.* 269, 311–316.