

# QUANTIFICATION OF UNCERTAINTY APPLIED TO RADIOLOGICAL CHARACTERIZATION OF RADIOACTIVE WASTE

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*Abstract.* Characterization of radioactive waste involves establishing the list of radionuclides together with their activities. The estimated activity levels are compared with the limits given by the national authority in the field. Therefore, quantifying the uncertainty that affects the concentration of radionuclides is essential for estimating the acceptability of waste in the final repository, as well as for controlling the stages of segregation and radiological characterization. The approach proposed in this study aims to estimate the uncertainties involved in gamma-ray spectrometry, which is utilized for characterizing radioactive waste.

*Key words:* radiological characterization, radioactive waste, radionuclide, uncertainty, decommissioning, key radionuclide, hard to detect radionuclide

## 1. INTRODUCTION

A comprehensive radiological characterization process that spans the entire life cycle of a nuclear facility offers valuable insights into all activities and their timelines, ranging from the design, construction, operation, transition, decommissioning, and waste management phases, all the way to final disposal [1]. By carrying out radiological characterization at every stage of the facility's life cycle, pertinent questions can be answered, and informed decisions can be made. Each stage of the life cycle of a nuclear facility requires a distinct approach for radiological characterization, ensuring that relevant information is available to make the best choices for subsequent phases of the life cycle. Multiple objectives must be considered when performing radiological characterizations at every stage of the nuclear facility's life cycle, and the level of uncertainty in the information obtained must be acceptable. The uncertainty level is influenced by the scale of the characterization activities and the precision and accuracy of the results. The risks associated with the characterization objectives must be taken into account as well. For instance, high uncertainties and risk levels may be tolerable when rough estimates of radioactive waste volumes are required before commencing the decommissioning process. However, much lower levels of uncertainty are acceptable

when worker protection is at stake, as an incorrect decision can have severe consequences. Therefore, it is crucial to ensure that the required information for each characterization objective is considered, and the level of uncertainty in this information is appropriate. Practically, this may entail employing identical characterization techniques but varying detection limits and measurement uncertainties, depending on the characterization objectives.

For example, in the case of the VVR-S research nuclear reactor at Magurele, a radiological characterization was carried out prior to the decommissioning process, which required all design information regarding the composition of construction materials and operational data necessary for estimating contamination/activation levels. These pieces of information were used to optimize the additional characterization data required and when it needed to be prepared. All this information formed the basis for the safe and secure decommissioning of the facility and management of resulting radioactive waste.

The focus of this paper is to propose a method for estimating measurement uncertainties in gamma-ray spectrometry, which is commonly used in the radiological characterization of radioactive waste. The methodology for quantifying measurement uncertainty strategies is applied in the radioactive waste management of the Radioactive Waste Management Department (DMDR) at IFIN-HH.

The results of this study have a significant impact in the field of expertise, as they cover many important aspects in assessing radionuclide uncertainties encountered in radioactive waste through the experience and lessons learned in radiological characterizations carried out in the decommissioning project of the VVR-S nuclear research reactor successfully completed at the end of 2020.

## 2. UNCERTAINTY ESTIMATION FOR RADIOLOGICAL CHARACTERIZATION OF RADIOACTIVE WASTE

The characterization of radioactive waste poses a significant challenge due to its complex nature. At Radioactive Waste Management Department (DMDR) from IFIN-HH, the Radionuclide, Physico-Chemical, Mechanical and Structural Characterization Laboratory (DMDR-Lab) employs gamma spectrometry to estimate the specific activity of easy to measure radionuclides [2]. Radionuclides that emit X-rays,  $\beta$  particles, and low-energy gamma rays, and are hard to detect, are analysed using radiochemical methods or through calculations and Monte Carlo simulations [3, 4]. The specific activities are then compared with the limits prescribed in national regulations. To ensure compliance with these limits, the producer of radioactive waste must assess the distribution of the relevant quantities, taking into account the associated uncertainties [5, 6].

For illustrative purposes, Table 1 presents some of the radionuclides that are easy to measure and those that are hard to detect, resulting from the operation of the VVR-S Magurele nuclear research reactor.

Table 1

Radionuclides resulted from operation of VVR-S nuclear research reactor

Radionuclid	Emitted radiation	Life time (years)	Observation
Activation products			
<sup>3</sup> H	Beta (100%)	12.3	Hard to detect
<sup>14</sup> C	Beta (100%)	573E+01	–
<sup>41</sup> Ca	EC	1.03E+05	Hard to detect
<sup>55</sup> Fe	EC, X	2.7	Hard to detect
<sup>63</sup> Ni	Beta	10E+02	Hard to detect
<sup>60</sup> Co	Beta, gamma	5.3	Key radionuclide for activation products
<sup>134</sup> Cs	Beta, gamma	2	–
<sup>152</sup> Eu	EC, X, beta, gamma	13.5	–
<sup>154</sup> Eu	Beta, gamma, X	8.6	–
<sup>155</sup> Eu	Beta, gamma, X	4.76	–
Fission products			
<sup>90</sup> Sr+ <sup>90</sup> Y	Beta	28.7	–
<sup>137</sup> Cs	Beta, gama	30	Key radionuclide for fission products
Actinides			
<sup>238</sup> Pu	Alpha	87.7	–
<sup>239</sup> Pu	Alpha	2.41E+04	–
<sup>241</sup> Pu	Beta	14.3	–
<sup>241</sup> Am	Alpha, gamma	432	Key radionuclide for activide products

### 3. ASSESSMENT OF UNCERTAINTIES FOR EASY TO MEASURE RADIONUCLIDES IN RADIOACTIVE WASTE PACKAGES

The main source of uncertainty related to easy to measure radionuclide activity (such as <sup>60</sup>Co, <sup>137</sup>Cs, <sup>241</sup>Am from Table 1) comes from various factors, including the weight and density of the radioactive waste packages, the distribution of activity within the packages, the geometry of the radioactive materials contained within the packages, and the relative position of the detector and the package. To minimize uncertainty, sorting of the radioactive materials introduced into a package should be done based on the dose rate level, which helps limit the effects of hotspots [7]. Additionally, compacting large-volume objects and filling the packages with smaller-sized radioactive materials are effective techniques for achieving uniformity of materials inside the radioactive waste packages.

Regarding the activity estimated through gamma-ray spectrometry of identified radionuclides in radioactive waste packages, it is calculated for each identified peak. There are several methods to determine whether a nuclide is present in the spectrum or not, and if it is not present, then the minimum detectable activity must be reported.

The radionuclide is reported as being present in the spectrum if the following considerations are true: the first peak with the highest intensity of the radionuclide present in the library is present in the spectrum and has an uncertainty below the maximum allowed limit; all peaks marked as key lines are present in the acquired spectrum; the limit fraction test is passed [8]. If the radionuclide is not present, then the value of the minimum detectable activity is reported.

The activity of a radioactive waste package ( $\Lambda_{em}$ ) containing easy to measure radionuclides can be expressed as a function of several parameters such as: the net area ( $R_n$ ) of the peak in the acquired spectrum obtained by subtracting the background ( $B$ ) from the total area ( $R_t$ ), the net weight of materials inside the package ( $m$ ), the emission probability for the specific energy of the identified radionuclide in the spectrum ( $Y$ ), the acquisition time of the spectrum ( $t$ ), the detection efficiency ( $\varepsilon$ ), and the correction factor ( $f_c$ ) [9, 10]. Therefore, it is possible to obtain:

$$\Lambda_{em} = f(R_n, m, Y, t, \varepsilon, f_c) = \frac{R_n}{Yt\varepsilon} \quad (1)$$

Focusing on aspects relevant to characterizing low or medium-level radioactive waste and assuming that the activity is estimated solely from a peak without any additional interferences, a straightforward equation for determining the relative standard uncertainty of the activity is presented as follows:

$$\frac{u(\Lambda_{em})}{\Lambda_{em}} = \sqrt{\left(\frac{u(R_n)}{R_n}\right)^2 + \left(\frac{u(Y)}{Y}\right)^2 + \left(\frac{u(t)}{t}\right)^2 + \left(\frac{u(\varepsilon)}{\varepsilon}\right)^2} \quad (2)$$

The sum of pulses in a given channel range determines the total area of a peak as follows:

$$R_t = \sum_{i=l}^h C_i \quad (3)$$

where:  $R_t$  represents the total area,  $C_i$  is the count of pulses in channel  $i$ ,  $l$  is the leftmost channel of the peak, and  $h$  is the rightmost channel of the peak.

This approach to peak area calculation offers sufficient accuracy, even for small peaks, since it is less susceptible to variations in the peak shape between the measured spectrum and the shape of a peak measured using a calibration source. The net area is calculated by subtracting the background area from the total area between the selected channels, where the background area is calculated using the following formula:

$$B = \frac{B_l - B_r}{2} W \quad (4)$$

where  $B_l$  is the background on the left side of the peak,  $B_r$  is the background on the right side of the peak, and  $W$  is the width of the peak.

To calculate the statistical counting uncertainty, one needs to take the square root of both the uncertainty in the total area and the uncertainty in the background. Similarly, the uncertainty in the total area can be found by taking the square root of the total area. However, estimating the uncertainty in the background area is not as simple. This uncertainty can be calculated by multiplying the uncertainty in the channels used at the beginning and end of the area of interest by the ratio of the number of channels in the peak and the number of channels used to calculate the background.

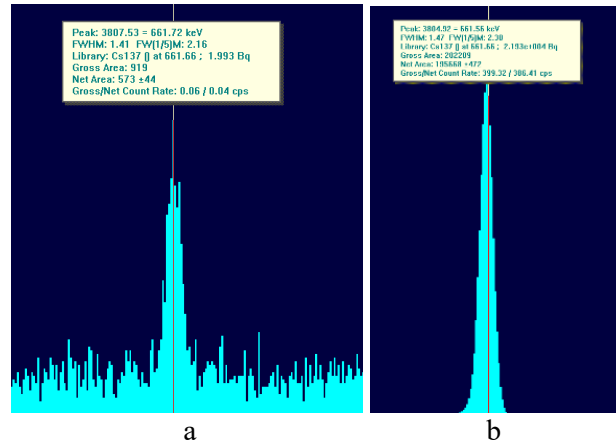


Fig. 1 –  $^{137}\text{Cs}$  peak with: a) high uncertainty of the net area; b) low uncertainty of the net area.

For broader or smaller peaks (with a lower counting rate), there is a higher uncertainty in the obtained value for the background area. Figure 1a shows a peak of  $^{137}\text{Cs}$  (661.66 keV) with a smaller net area, while Fig. 1b shows the same peak of  $^{137}\text{Cs}$  with a larger net area. The differences in net area, background area, and associated uncertainties can be easily observed. Therefore, the net area uncertainty is calculated as follows:

$$u(R_n) = \sqrt{(u(R_t))^2 + (u(B))^2} \quad (5)$$

where:  $u(B) = \sqrt{\frac{BW}{B_l + B_r}}$  and  $u(R_t) = \sqrt{R_t}$ .

When data is analysed, the option exists to present either the counting uncertainty or the total uncertainty in the report, and they can be expressed for 1, 2, or 3 sigma. Counting uncertainty pertains to the uncertainty in the peak area resulting from statistical uncertainties. If it is necessary to express the counting uncertainty for the net area of the peak, it can be converted into a percentage. The same

percentage can then be utilized to describe the counting uncertainty for the value of activity obtained.

As a result, to ascertain the total uncertainty (1 sigma), it is necessary to take the square root of the sum of the squared individual uncertainties of the components that contribute to the value of activity for the radioactive waste package, as follows:

$$u_t = \sqrt{u_{cnt}^2 + u_{nd}^2 + u_{rs}^2 + u_{abs}^2 + u_{rad}^2 + u_{\varepsilon}^2 + u_{geo}^2 + u_{ud}^2} \quad (6)$$

where:  $u_{cnt}$  is the counting uncertainty,  $u_{nd}$  is the additional uncertainty due to the normal distribution evaluated as  $u_{nd}^2 = \left(\frac{\text{input}_{nd}}{100}\right)^2$  which  $\text{input}_{nd}$  the value entered by the user;  $u_{rs}$  is the uncertainty due to random summing evaluated as  $u_{rs}^2 = \frac{|f_{rs}-1|}{100}$  which  $f_{rs}$  the correction factor for random summation,  $u_{abs}$  is the uncertainty due to absorption corrections evaluated as  $u_{abs}^2 = \frac{|f_{abs}-1|}{100}$  which  $f_{abs}$  the correction factor for absorption,  $u_{rad}$  is the uncertainty due to the radionuclide evaluated as  $u_{rad}^2 = \left(\frac{\text{input}_{rad}}{200}\right)^2$  which  $\text{input}_{rad}$  the uncertainty of the scheme factor for the main energy of each radionuclide in the library used in the analysis the value entered by the user (at 2 sigma),  $u_{\varepsilon}$  is the uncertainty due to the efficiency,  $u_{geo}$  is the uncertainty due to geometry corrections evaluated as  $u_{geo}^2 = 0.015^2$  (in practice, it is 1.5% for 1 sigma),  $u_{ud}$  is the uncertainty due to uniform distribution evaluated as  $u_{ud}^2 = (\text{input}_{ud})^2$  with  $\text{input}_{ud}$  the value entered by the user. All uncertainties are evaluated for 1 sigma, except for  $u_{ud}$ . If  $u_{ud}$  is not used in the evaluation of the total uncertainty, then it is considered as 0.

The uncertainty of detection efficiency is evaluated differently depending on the type of fitting used to assess the calibration curve in effectiveness. The components used to evaluate effectiveness include errors caused by the uncertainty of the calibration source and the uncertainty due to the fitting performed.

Polynomial fitting is a mathematical technique used to fit a polynomial equation to a set of data points. This technique involves finding the coefficients of a polynomial equation that best describes the relationship between the input and output variables of the data. The equation for a polynomial fit of degree  $n$  can be written as:  $y = c_0 + c_1x + c_2x^2 + c_3x^3 + \dots + c_nx^n$ , where  $y$  is the dependent variable,  $x$  is the independent variable, and  $c_0, c_1, c_2, \dots, c_n$  are the coefficients of the polynomial. The degree of the polynomial,  $n$ , determines the number of coefficients needed to describe the relationship between  $x$  and  $y$ . The coefficients are usually calculated using a method such as least squares regression, which minimizes the sum of the squared errors between the actual data points and the predicted values from the polynomial equation.

In general, the efficiency curve for measuring samples through gamma spectrometry is obtained by polynomial fitting expressed as follows:

$$\ln \varepsilon = \sum a_k x^n, \quad k = 1, \dots, m \quad (7)$$

where  $\varepsilon$  is the efficiency at the energy  $E$  of interest,  $\ln$  is the natural logarithm,  $a_k$  are the  $k$  fitting parameters. The sum is of all the fitting parameters,  $k = 1, \dots, m$ , not the calibration points. For polynomial fitting  $x = E$ ,  $n = 2 - k$ ,  $m = 6$ .

If a parameter is defined as  $y = \ln \varepsilon$ , the following expression is obtained:

$$y = \sum a_k x^n, \quad k = 1, \dots, m \quad (8)$$

As a result, the efficiency evaluated based on the efficiency fitting curve can be expressed using the following formula:

$$y_i(x_i) = \sum a_k x_i^n, \quad k = 1, \dots, m \quad (9)$$

The uncertainty of the fitting polynomial is given by:

$$u_y^2 = \sum \sum \delta_{ij} \left( \frac{\partial y}{\partial a_i} \right) \left( \frac{\partial y}{\partial a_j} \right), \quad i, j = 1, \dots, m \quad (10)$$

where  $y$  is the polynomial expressed in Eq. 8 and  $a_i$  is a fitting parameter. The covariance error  $\delta_{ij}$  is used instead of  $u_{a_i a_j}^2$ , and furthermore, the covariance error is symmetric,  $\delta_{12} = \delta_{21}$ .

Once  $x^{i1} = \frac{\partial y}{\partial a_i}$ , it follows from Eq. 8:

$$u_y^2 = \sum \sum \delta_{ij} x^{(i_1 + j_1)}, \quad i, j = 1, \dots, m \quad (11)$$

where, for the case of polynomial fitting  $i_1 = 2 - i$ ,  $j_1 = 2 - j$ .

In the end, the calibration uncertainty is:

$$u_{calib} = \varepsilon u_y \quad (12)$$

where  $\varepsilon$  is the efficiency according to the fitting curve at the energy  $E$  evaluated from Eq. 7, and  $u_y$  is evaluated from Eq. 11. If  $u_{calib}$  is 0, then both the above-curve error,  $u_{sup}$ , and the below-curve error,  $u_{bel}$ , are used.

In the case of polynomial fittings, the calibration uncertainty is evaluated as follows:

$$u_{calib} = u_{sup}. \quad (13)$$

### 3.1. REPORTED UNCERTAINTY FOR MEASURING A PACKAGE WITH RADIOACTIVE WASTE

For measurements of radioactive waste, it is not possible to provide a mathematical solution for the attenuation location in the matrix or measurement geometry, as there are many crucial parameters for each waste package that are not measured. However, practical experience gained in radiological characterization through gamma-ray spectrometry obtained during the decommissioning of the VVR-S nuclear research reactor from Magurele, can be used to make estimations based on accumulated knowledge to provide realistic uncertainties. Practical experience involves measuring a large number of radioactive waste packages to empirically verify the location of activity within the package, so that the effect of geometry can be evaluated. The model assumes that the matrix density is homogeneously distributed within the package and that the measured activity is located within it.

$$u_t = \sqrt{u_{peak}^2 + u_{matrix}^2 + u_{geom}^2} \quad (14)$$

where:  $u_t$  is the total uncertainty [%],  $u_{peak}$  is the uncertainty due to counting rate,  $u_{matrix}$  is the uncertainty due to matrix,  $u_{geom}$  is the uncertainty due to geometry.

The uncertainty due to the matrix is evaluated by assuming that the attenuation and path length of gamma radiation can vary by up to 40%. Therefore, the following is defined:

$$u_{matrix} = \frac{\left(\frac{M}{1-e^{-M}}\right) - \left(\frac{N}{1-e^{-N}}\right)}{\frac{N}{1-e^{-N}}} 100 \quad (15)$$

where:  $M = \mu\rho dc$ ,  $\mu$  is the mass attenuation coefficient [ $\text{cm}^2/\text{g}$ ],  $\rho$  is the matrix density [ $\text{g}/\text{cm}^3$ ],  $d$  is the container diameter [cm],  $c$  is a constant (0.834 for a cylinder) [11, 12, 28, 29], and  $N = 1.4M$ .

It is important to mention that when materials in the package have a high density, the correction factor  $F_c$  is approximated as follows:

$$F_c \approx \mu\rho d. \quad (16)$$

Measurements of radioactive waste packages that contain uranium, for example, can exhibit such a high self-attenuation of gamma rays that it is very difficult to quantify.

The uncertainty due to geometry varies with the shape of the package. For cylindrical or parallelepiped packages, it is possible to assign an uncertainty assuming that the activity is distributed in the outer part of the package



(approximately  $\frac{1}{4}$  of the thickness towards the outside), based on experience. The uncertainty can be evaluated by taking into account the homogeneity of the materials as follows:

$$u_{geom} = \frac{(F_{co} - F_{cf})}{F_{co}} 100 \quad (17)$$

where  $F_{co}$  is the correction factor for homogeneous materials,  $F_{cf}$  is the correction factor when the activity is located in the front or farthest point of the container. When packages are measured in rotation, the correction factor reflects whether the activity is in the outer half of the container. Therefore, the uncertainty due to geometry will be smaller.

### 3.2. TECHNIQUES FOR MINIMIZING UNCERTAINTY

One can employ various techniques to minimize measurement uncertainties in radioactive waste packages. These methods focus on reducing correction factors. Proper placement of the detector is essential to achieve accurate measurements of the radioactive waste packages. Increasing the distance between the detector and the waste package decreases the error caused by geometrical corrections. Statistical counting uncertainty is the primary factor that contributes to the total measurement uncertainty. If the activity of the waste package is too low, the acquisition time needs to be increased to meet the required sensitivity level.

The attenuation correction factor is one of the most critical correction factors that need to be minimized. To minimize the correction due to attenuation in the matrix, the measurements of parallel packages should be performed on the surface with the smallest thickness. Conversely, if measurements are taken on the surface with the highest thickness, the matrix thickness will increase the attenuation in the matrix and increase the associated uncertainty.

The detector-waste package position should be chosen to avoid corrections due to the collimator. The techniques mentioned above can be used to minimize measurement uncertainties associated with radioactive waste packages.

## 4. ASSESSMENT OF UNCERTAINTIES FOR HARD TO DETECT RADIONUCLIDES

Radionuclides that emit alpha and beta radiation, such as  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{63}\text{Ni}$ , and  $^{240}\text{Pu}$ , or those that emit low-energy X-rays, like  $^{55}\text{Fe}$ , are classified as hard-to-measure radionuclides. Measuring their activity requires destructive and complex methods like radiochemical analysis, making them difficult to measure. To evaluate

the activity of such radionuclides, the scaling factor method can be used. This method is based on the relationship between the activity of easy-to-measure radionuclides and that of hard-to-measure ones [13]. In the case of radioactive waste packages, the activity of hard-to-measure radionuclides is estimated by gamma spectrometry using the scaling factors obtained from the activity of easy-to-measure radionuclides [14, 15, 16, 17, 18, 19].

The International Standard ISO 21238:2007 [3] provides a general methodology for the empirical determination of scaling factors to evaluate the activity of hard-to-measure radionuclides in low or intermediate level radioactive waste packages. These international standards provide guidance on the use of scaling factors in characterizing waste produced in nuclear facilities.

The procedure for radiochemical analysis of hard-to-measure radionuclides involves sampling, sample treatment (pre-concentration), radiochemical separation of the radionuclide of interest, preparation of the source, and measurement of the radionuclide. The main methods of radiochemical analysis include alpha spectrometry, which is used for the identification and quantification of alpha-emitting radionuclides; liquid scintillation counting, which is particularly used for the measurement of beta-emitting radionuclides or radionuclides that decay through electron capture (*e.g.*, radionuclides found in the nuclear industry such as  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{55}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ , etc.).

Regarding the experimental aspect, the method of scaling factors involves verifying the reliability and reproducibility of the correlation between the activity of hard-to-measure radionuclides and that of key radionuclides in representative samples. This correlation is then used to estimate the activity of the hard-to-measure radionuclides by measuring the activity of the key radionuclides in each type of material or package of radioactive materials [3, 13].

An experimental example in this regard is the estimation of the radioactive inventory of the VVR-S research nuclear reactor block, which formed the basis for its decommissioning and contained both easily measurable and hard-to-measure radionuclides. For hard-to-measure radionuclides, the specific activity was evaluated using the scaling factor method, using  $^{60}\text{Co}$  as the key radionuclide. The estimation of scaling factors was based on theoretical calculations, gamma spectrometric results obtained from collected samples, and data from specialized literature [20–23].

Therefore, the activity values of difficult-to-measure radionuclides can be estimated by measuring the activity of key radionuclides in each type of material (*e.g.*,  $^{60}\text{Co}$  for activation products). If activation occurs under similar conditions, the activity distribution of the sampled materials should be log-normal. This is observed for both emitters and non-emitters. The distribution of scaling factors, calculated as the ratio of the activity of difficult-to-measure radionuclides to the activity of key radionuclides, is also log-normal [24].

For log-normally distributed scale factors, a good estimator of the central tendency is the geometric mean ( $\bar{G}_{FS}$ ) [3], defined as follows:

$$\bar{G}_{FS} = \exp\left(\frac{\sum_{i=1}^n \ln(FS_i)}{n}\right) = \sqrt[n]{\prod_{i=1}^n FS_i} \quad (18)$$

where  $FS_i$  is the scaling factor for sample  $i$  and represents the ratio of the activity of the difficult-to-measure radionuclide for sample  $i$  to the activity of the key radionuclide [25].

The uncertainty of the geometric mean  $u(\bar{G}_{FS})$  can be evaluated as the standard error of the geometric mean of the scaling factor [13, 26, 27] as follows:

$$u(\bar{G}_{FS}) = \bar{G}_{FS} \frac{ds(\ln(FS_i))}{\sqrt{n-1}} \quad (19)$$

where  $\bar{G}_{FS}$  is obtained from Eq. 18,  $ds(\ln(FS_i))$  is the estimated standard deviation of the natural logarithm of the scale factors, and  $n - 1$  is the degrees of freedom of the sample.

The uncertainty of the activity of the hard-to-measure radionuclide ( $u(\Lambda_{hd})$ ) in a package  $i$  or in a quantity/category of material can be evaluated according to the error propagation formula in Eq. 2. In case multiple measurements are performed on a package or a batch of materials, then the standard deviation of the mean activity of the hard-to-measure radionuclide ( $\Lambda_{hd}$ ) obtained from  $n$  measurements is given by:

$$\frac{1}{u^2(\Lambda_{hd})} = \sum_{i=1}^n \frac{1}{u_i^2(\Lambda_{hd})} \quad (20)$$

where  $u_i(\Lambda_{hd})$  is the standard deviation for  $i$  measurement evaluated from Eq. 2.

The standard uncertainty for the geometric mean of the scale factors is evaluated with Eq. 19. Regarding the linear model, only values above the detection limit should be used to make these estimates.

The combined relative standard uncertainty  $u_{comb}$  of the activity of the difficult-to-measure radionuclides in the batch is given by:

$$\frac{u_c(\Lambda_{hd,i})}{\Lambda_{hd,i}} = \sqrt{\frac{u^2(\bar{G}_{FS})}{\bar{G}_{FS}^2} + \frac{u^2(\Lambda_{em,i})}{\Lambda_{em,i}^2}} \quad (21)$$

where  $\Lambda_{em,i}$  is the activity evaluated for the key radionuclide for the  $i$  package or batch of materials.

## 5. DISCUSSION AND CONCLUSIONS

A crucial aspect of radioactive waste storage is the ability to determine the specific concentrations of radionuclides present in the waste. However, many important long-lived radionuclides found in radioactive waste are difficult to measure using non-invasive techniques due to being alpha or beta emitters, or having low energy levels. To identify these radionuclides, chemical methods are used to separate different radionuclides in the sampled materials for measurement. However, such techniques are complex and impractical for most radioactive wastes. Therefore, alternative methods such as gamma spectrometry are needed to evaluate the radiological characteristics of gamma emitters, which can provide information about radionuclides that are otherwise difficult to measure.

The scaling factor method is an alternative method widely used to evaluate the activity of difficult-to-measure radionuclides. The method is based on the correlation between easily measured gamma-emitting radionuclides called key radionuclides and the difficult-to-measure radionuclides. The activity of difficult-to-measure radionuclides in radioactive waste packages/lots is estimated through gamma spectrometric measurements and the use of scaling factors. The development and use of scaling factors depend on a number of steps, and each step introduces uncertainties in the final result.

The activity of hard-to-measure radionuclides evaluated using scaling factors has an uncertainty that depends on the physical parameters of the radioactive waste package on one hand, and on the gamma spectrometric measurements used to evaluate the activity of key radionuclides on the other hand. The uncertainties associated with the radioactive waste package are related to the conditioning process and the package contents, and these uncertainties involve all parameters required to evaluate the activity of radionuclides easily measured through gamma spectrometry. On the other hand, uncertainties associated with the measurement itself can be evaluated based on suitable references or standards.

This study has explored various aspects concerning the analysis and assessment of uncertainties in radiological characterization of radioactive waste. Specifically, it has addressed the evaluation of uncertainties for easily measurable radionuclides present in radioactive waste packages, as well as those associated with the measurement of these packages. The work has also delved into techniques aimed at minimizing such uncertainties, while also providing insights into uncertainties linked to difficult-to-measure radionuclides.

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## REFERENCES

1. Nuclear Energy Agency, Organisation for Economic Co-Operation and Development, NEA No. 7373, *Radiological Characterisation from a Waste and Materials End-State Perspective: Practices and Experience*, 2017.
2. IAEA, *Strategy and methodology for radioactive waste characterization*, Technical Report IAEA-TECDOC-1537, Vienna, 2007.
3. ISO 21238, *Nuclear energy – Nuclear fuel technology – Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants*, Geneva, 2007.
4. ISO 16966, *Nuclear energy – Nuclear fuel technology – Theoretical activation calculation method to evaluate the radioactivity of activated waste generated at nuclear reactors*, Geneva, 2013.
5. IAEA, *Classification of radioactive waste: a safety guide*, Technical Report Safety, series no. 111-G-1.1, Vienna, 1994.
6. Lowenthal, M.D., *Waste-acceptance criteria and risk-based thinking for radioactive waste classification*, *Waste Manag.* **18** (4), 249–256, 1998.
7. Rzyski, B.M., Suarez, A.A., *Evaluation of homogeneity of radioactive waste forms: statistical criteria*, *Nucl. Chem. Waste Manag.* **8** (3), 211–215, 1988.
8. IAEA, *Determination and Interpretation of Characteristic Limits for Radioactivity Measurements, Decision Threshold, Detection Limit and Limits of the Confidence Interval*, Analytical Quality in Nuclear Applications Series No. 48, IAEA/AQ/48, 2017.
9. Gilmore, G., *Practical gamma-ray spectrometry*, John Wiley and Sons, Inc., The Atrium, Southern Gate, Chichester, West Sussex, PO19 8SQ, England, 2008.
10. Knoll, G.F., *Radiation Detection and Measurement*, John Wiley and Sons, Inc, New York, 2010.
11. ISOTOPIC Supervisor software user manual, *Advanced Measurement Technology*, Inc., Ortec, 2008.
12. ISOTOPIC Supervisor software user manual, *Advanced Measurement Technology*, Inc., Ortec, 2014.
13. IAEA, Nuclear Energy Series No. NW-T-1.18, *Determination and use of scaling factors for waste characterization in nuclear power plants*, Technical Report, 2009.
14. IAEA, *Characterization of Radioactive Waste Forms and Packages*, Technical Reports Series No. 383, IAEA, Vienna, 1997.
15. IAEA, *Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes*, Technical Reports, Series No. 389, IAEA, Vienna, 1998.
16. IAEA, *Review of the Factors Affecting the Selection and Implementation of Waste Management Technologies*, IAEA-TECDOC-1096, IAEA, Vienna, 1999.
17. IAEA, *Development of Specifications for Radioactive Waste Packages*, IAEA-TECDOC-1515, Vienna, 2006.
18. IAEA, *Innovative Waste Treatment and Conditioning Technologies at Nuclear Power Plants*, IAEA-TECDOC-1504, Vienna, 2006.
19. IAEA, *Management of Problematic Waste and Material Generated During the Decommissioning of Nuclear Facilities*, Technical Reports, Series No. 441, Vienna, 2006.
20. Ionescu, E., Gurau, D., Stanga, D., Dului, O.G., *Decommissioning of the VVR-S research reactor – Radiological characterization of the reactor block*, *Romanian Reports in Physics* **64** (2), 387–398 (2012).
21. Ancius, D., Ridicas, D., Remeikas, V., Plukis, A., Plukiene, R., Cometto, M., *Evaluation of the activity of irradiated graphite in the Ignalina Nuclear Power Plant RBMK-1500 Reactor*, *Nucleonika* **50** (3), 113–120 (2005).

22. Cometto, M., Ridikas, D., Aubert, M.C., Damoy, F., Ancius, D., *Activation Analysis of Concrete and Graphite in the Experimental Reactor RUS*, Radiation Protection Dosimetry **115** (1–4), 104–109 (2005).
23. Kinno, M., Kimura, K., Ishikawa, T., Miura, T., Ishihama, S., Hayasaka, N., Nakamura, T., *Correlation between tritium and  $^{152}\text{Eu}$  induced in various types of concrete by thermal neutron irradiation*, Journal of Nuclear Science and Technology **39** (3), 215–225 (2002).
24. Cochran, W.G., *Sampling Techniques*, John Wiley and Sons Inc, New York, 1963.
25. Zaffora, B., Magistris, M., Saporta, G., La Torre, F., *Statistical sampling applied to the radiological characterization of historical waste*, EPJ Nucl. Sci. Technol. **34** (2), 2016.
26. Norris, N., *The standard errors of the geometric and harmonic means and their application to index numbers*, Ann. Math. Stat. **11** (4), 445–448, 1940.
27. Harding, B., Tremblay, C., Cousineau, D., *Standard errors: a review and evaluation of standard error estimators using Monte Carlo simulations*, Quant. Methods Psychol. **10** (2), 107–123 (2014).
28. Manual, Ortec, Advanced Measurement Technology, 2006.
29. Maestro32 Software User Manual, Software Version 6.0, Advanced Measurement Technology, Inc., 2006.