Research on Calculation Methods for Important Parameters in Radioactivity Measurement

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Abstract

Radioactive measurements are widely employed across various fields of nuclear technology applications. Measurement uncertainty, confidence intervals, detection limits, and other related parameters constitute important aspects of radioactive measurements, with their calculated results directly influencing relevant decision-making. This study investigates key parameters in α-particle activity concentration measurements using both the partial derivative method and the Monte Carlo method: firstly, the sources of uncertainty in measurement results are analyzed, followed by the calculation of α-particle activity concentration uncertainty, confidence intervals, decision thresholds, and detection limits using both partial derivative and MC (Monte Carlo) methods. The results demonstrate that when the uncertainty of input quantities exceeds 10%, the relative deviation between the confidence intervals and uncertainty results obtained by the two computational methods surpasses 15%; when the relative uncertainty of input quantities is relatively small, the detection limit is approximately twice the decision threshold. While the partial derivative method is widely applied, it fails to account for the probability distribution forms of input quantities and is unsuitable for models involving complex and special input quantities; consequently, the Monte Carlo method can be employed to obtain more reliable computational results, and these two methods can be applied in a complementary manner.

Full Text

Preamble

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Research on Calculation Methods of Important Parameters in Radioactivity Measurement

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Abstract

[Background] Radioactivity measurement is widely used in various fields of nuclear technology application. Measurement uncertainty, confidence interval, and detection limit are important parameters in radioactivity measurement, and their calculation results directly affect relevant decision-making. [Purpose] This study investigates important parameters in alpha particle activity concentration measurement using both partial derivative and Monte Carlo methods. [Methods] Based on alpha activity concentration measurement, the sources of uncertainty in measurement results were first analyzed. The measurement uncertainty, confidence limits, decision threshold, and detection limit of alpha particle activity concentration were then derived and calculated using partial derivative and MC methods under different input modes. [Results] The results show that when input uncertainty exceeds 10%, the relative deviation between confidence intervals and uncertainty results obtained by the two methods is greater than 15%. When the relative uncertainty of input quantities is small, the detection limit is approximately twice the decision threshold. [Conclusions] The partial derivative method yields more reliable calculation results, and these two methods can be applied complementarily.

Keywords: Activity concentration, Uncertainty, Decision threshold, Detection limit, Monte Carlo

Introduction

Radioactivity measurement is critically important in nuclear reactor operation, environmental radiation monitoring, nuclear medicine, and other fields. Its purpose is to determine whether operational conditions are abnormal or to support certain decisions. By comparing measurement results with specific limits, we can determine whether nuclear facilities are operating normally or whether radiation doses exceed standards, enabling appropriate measures to be taken. Essential parameters associated with measurement results include measurement uncertainty and confidence intervals [1-2]. Without considering measurement uncertainty, results cannot be evaluated for reliability and thus cannot be properly applied. Underestimating or overestimating measurement uncertainty may lead to inappropriate actions: in radioactive measurement, underestimating uncertainty may result in inadequate protective measures, increasing the risk of excessive radiation exposure to personnel or equipment, while overestimating uncertainty may lead to excessive protective actions, causing waste of social resources. The detection limit is an important parameter for equipment, and considerable research has been conducted in this area [3-5]. Different radioactive monitoring devices operating under natural background or interference from other nuclides require slightly different factors for calculating detection limits. Many studies simply use formulas to derive detection limit measurement methods without considering the uncertainty of various equipment parameters and the applicable scope of derived formulas, which may lead to incorrect results.

The GUM (Guide to the Expression of Uncertainty in Measurement) [6-7] mentions two methods for calculating measurement uncertainty: one is the partial derivative calculation method, and the other is the MC method. The partial derivative method involves calculating first-order derivatives of each input component related to the output quantity. This method is widely used in practice, but if the distribution of individual input components is not Gaussian, this method cannot predict the distribution form of output results, and the reliability of results requires further investigation. MC can specify arbitrary probability density distributions for each input component and directly provide the probability density distribution of output results. In radioactive alpha activity concentration monitoring, this study considers different probability density distribution forms of input components and uses both methods to calculate and compare uncertainty and confidence interval results to select a more appropriate calculation method.

1 Calculation Model

This study primarily measures the activity concentration of alpha particles in gas. Gas flows through a filter paper at a certain flow rate, depositing alpha radioactive particles onto the filter paper. A PIPS (Passivated Implanted Planar Silicon) detector assembly is used to detect alpha particle radioactive counts on the filter paper. The activity concentration AC of alpha particles in the gas is calculated as follows:

$$AC = \frac{N_g/t_g - N_b/t_b}{V \cdot \varepsilon \cdot f} = \frac{R_g - R_b}{V \cdot \varepsilon \cdot f}$$

where AC represents activity concentration ($\mathrm{Bq \cdot L^{-1}}$); N_g represents total counts within measurement time t_g; N_b represents background counts within measurement time t_b; R_g and R_b represent total count rate and background count rate, respectively ($\mathrm{s^{-1}}$); V represents sampling volume (L); ε represents detection efficiency; and f represents self-absorption coefficient.

In this model, time and count uncertainties are not considered. V and ε follow Gaussian distributions, while f has both Gaussian and rectangular distribution forms. The specific input quantities in the calculation model are shown in Table 1 .

2 Partial Derivative Method

According to the standard uncertainty calculation formula, the standard uncertainty expression for activity concentration is derived as equation (3):

$$u_c(AC) = \sqrt{\left(\frac{\partial AC}{\partial V}\right)^2 u^2(V) + \left(\frac{\partial AC}{\partial \varepsilon}\right)^2 u^2(\varepsilon) + \left(\frac{\partial AC}{\partial f}\right)^2 u^2(f)}$$

If we define $R = R_g - R_b$, then equation (3) simplifies to:

$$u_c(AC) = AC \cdot \sqrt{\left(\frac{u(V)}{V}\right)^2 + \left(\frac{u(\varepsilon)}{\varepsilon}\right)^2 + \left(\frac{u(f)}{f}\right)^2}$$

According to ISO 11929-2019 [8-10] and combining equations (1)-(4), the expressions for the decision threshold and detection limit of alpha particle activity concentration are derived for cases where both Type I and Type II error probabilities are 5%, as shown in equations (5) and (6):

$$y^* = k_{1-\alpha} \cdot u_c(0)$$

$$y^\# = (k_{1-\alpha} + k_{1-\beta}) \cdot u_c(y^\#)$$

where the confidence level is 95% and $k_{1-\alpha} = k_{1-\beta} = 1.65$.

Based on Table 1 and equations (1)-(4), the intermediate calculation values are obtained as shown in Table 2 .

From Table 1 and equation (4), the variation of activity concentration relative uncertainty and uncertainty with self-absorption coefficient uncertainty are shown in Figures 1 Figure 1: see original paper and 1(b), respectively. According to equations (5), (6) and Tables 1-2, the values of decision threshold and detection limit under different activity concentration relative uncertainties are obtained, as shown in Figure 2 [Figure 2: see original paper].

As seen in Figure 1 and equation (4), activity concentration uncertainty increases with increasing self-absorption coefficient uncertainty. Figure 2 and equations (5)-(6) show that the decision threshold is independent of the relative uncertainty of activity concentration, while the detection limit varies as a hyperbolic curve with relative uncertainty. When the relative uncertainty of activity concentration is large (\$ 0.61), *thedetectionlimitbecomesnegative*; *whentherelativeuncertaintyissmall*(\$0.1), the detection limit is approximately twice the decision threshold. The calculation of decision threshold does not consider the probability density distribution forms of V, ε, and f.

Using the partial derivative method, the lower and upper limit values of the 95% confidence interval under different self-absorption coefficient relative uncertainties are shown in Figure 3 [Figure 3: see original paper]. In Figure 3, LP_1 and UP_1 represent the lower and upper limits of the 95% confidence interval calculated by the partial derivative method. The results show that the partial derivative method does not consider special distribution forms of input quantities. The lower confidence limit decreases with increasing relative uncertainty of f, while the upper limit increases with increasing relative uncertainty. When the relative uncertainty of f exceeds 0.7, the lower confidence limit becomes less than zero.

3 Monte Carlo Simulation Method

The partial derivative method requires specialized mathematical skills, and calculations can be cumbersome for complex mathematical models. Compared to the partial derivative method, the MC method is a more intuitive numerical approach that simulates calculation results based on the probability density distribution functions of input quantities. For input quantities with arbitrary probability density distributions such as Gaussian, rectangular, triangular, etc., MC can provide the probability density distribution of output results and measurement uncertainty through extensive random sampling [11-13].

Based on equation (1) and the calculation results in Table 2, equation (1) can be transformed into equation (7):

$$y = \frac{R}{V \cdot \varepsilon \cdot f}$$

where y represents the activity concentration output value. According to the different probability density distribution forms of input quantities in equation (7), the required output values can be obtained through extensive sampling simulation.

When using 10^4 samples in MC simulation, the resulting uncertainty is approximately 1%. To achieve higher calculation accuracy, 10^5 samples were used in all simulation processes. With 10^5 samples repeated 20 times to calculate activity concentration y, the obtained relative uncertainty is 0.3%, significantly better than the 1% uncertainty obtained with 10^4 samples.

The sampling volume V and detection efficiency ε follow Gaussian distributions, as shown in Figures 4 Figure 4: see original paper and 4(b). The self-absorption coefficient f follows both Gaussian (Figure 4(c)) and rectangular distributions (Figure 4(d)), with relative uncertainties of 1%, 10%, 20%, 30%, 40%, and 50% under both distribution forms. The resulting activity concentration y distribution forms are shown in Figures 5 Figure 5: see original paper and 5(b).

Figure 5 demonstrates that the probability density distribution of output results obtained by the MC method depends not only on input uncertainty but also on the probability density distribution form of inputs. The differences in output results from different input distributions increase with increasing input uncertainty.

When sampling volume V and detection efficiency ε have fixed relative uncertainties, and the self-absorption coefficient f follows Gaussian and rectangular distributions, the variation curves of activity concentration standard uncertainty with f relative uncertainty are shown in Figures 6 Figure 6: see original paper and 6(b).

Figure 6 shows that when the self-absorption coefficient follows a Gaussian distribution, its relative uncertainty ($ $0.3) causes a sharp increase in standard uncertainty (Figure 6a). When the self-absorption coefficient follows a rectangular distribution, the increase in relative uncertainty causes a relatively slow increase in standard uncertainty (Figure 6b).

Using the MC method, the lower and upper limits of the 95% confidence interval under different relative uncertainties when the self-absorption coefficient follows Gaussian and rectangular distributions are shown in Figure 7 [Figure 7: see original paper]. In Figure 7, LP_2 and UP_2 represent the lower and upper limits of the 95% confidence interval for output result Y calculated by the MC method when f follows a Gaussian distribution; LP_3 and UP_3 represent the corresponding limits when f follows a rectangular distribution.

The results indicate that under both distribution modes of f, the upper confidence limit of activity concentration increases sharply with increasing uncertainty in input values, with different magnitudes of change.

4 Results and Analysis

When $t_g = t_b$, equation (5) simplifies to equation (8):

$$y^* = k_{1-\alpha} \cdot \frac{R_b}{V \cdot \varepsilon \cdot f} \cdot \sqrt{\frac{1}{t_b}}$$

From equation (6), it can be seen that when the relative uncertainties of input quantities such as sampling volume V, detection efficiency ε, and self-absorption coefficient f are small, equation (6) can be simplified to equation (9):

$$y^{\#} = 2 \cdot y^*$$

From equations (8) and (9), under fixed sampling volume, detection efficiency, self-absorption coefficient, and background measurement time, both decision threshold and detection limit are related to background.

The relative deviations between lower and upper confidence limits calculated by the partial derivative method and MC method under different f relative uncertainties are shown in Figure 8 [Figure 8: see original paper]. In Figure 8, ΔLP and ΔUP represent the relative deviations of lower and upper confidence limits calculated by the two methods, respectively.

Figure 8 and comparisons with Figures 1b, 6a, and 6b show that for this activity concentration calculation model, the relative deviations of upper confidence limits and uncertainties calculated by the partial derivative method and MC method increase with increasing input uncertainty. If negative values are not considered, the relative deviation of lower confidence limits first increases and then decreases with increasing input uncertainty. When the relative uncertainty of f exceeds 10%, the relative deviations of upper confidence limits and uncertainties calculated by the two methods exceed 15%.

If the calculation model contains trigonometric or exponential functions, the results from the partial derivative method differ significantly from MC simulation results because the partial derivative method does not consider the influence of input probability density functions on the output probability density function, while the MC method with extensive sampling can compensate for this limitation.

Important parameters in radioactivity measurement can be obtained using both partial derivative and MC methods. The partial derivative method is widely used for calculating measurement uncertainty. For simple calculation models like alpha activity concentration measurement, when input uncertainties are small, the standard uncertainty and confidence intervals calculated by the partial derivative method are relatively accurate. However, this method does not consider the effect of input density distribution functions on output density distribution functions and has limitations for complex calculation models. For complex models and large input uncertainties, the partial derivative method is less accurate than the more intuitive numerical MC method. When calculating important parameters in radioactivity measurement, it is necessary to consider the uncertainties and probability density distribution forms of each parameter in the input to select an appropriate calculation method for reliable results.

Author Contribution Statement

ZHOU Xuemei was responsible for conceptualization, drafting, and revision of the final manuscript; LUO Kun and LAI Wei designed the algorithms; LIAO Feng and DU Bingjie processed the data; LIU Wei provided critical review of intellectual content and research funding.

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Note: Figure translations are in progress. See original paper for figures.