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Evaluation of a gamma technique for the assay of radioactive waste drums using two measurements from opposing directions

Tran Quoc Dung *, Nguyen Duc Thanh, Luu Anh Tuyen, Lo Thai Son, Phan Trong Phuc

Centre for Nuclear Techniques, 217 Nguyen Trai St., D.1, Hochiminh City, Viet Nam

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ABSTRACT

A technique using two measurements from opposing directions for the assay of the activity of radioactive waste drums, mainly consisting of organic materials, has been studied. A model for the calculation of systematic errors is given by simulating the measurement system. The calculated values are in good agreement with the experimental values. This confirms the validity of the model and proofs the good performance of this technique in practice. These results may provide guidelines for setting up a measuring system for the assay the radwaste drums.

1. Introduction

Segmented gamma scanner (SGS) is a traditional tool for the isotopic composition measurement and for the determination of the activity level in gamma-contaminated waste drums (Bjork, 1987; Sprinkle and Hsue, 1987). The systematic error of this technique is still large because of several factors: non-uniform distribution of radioactive source within the drums frequently causes the largest error (Estep, 1990; Dung, 1997a, b; Levai et al., 2000); non-uniform distribution of non-radioactive materials (matrix) (Prettyman et al., 1996; Dung, 1998); particle size of the nuclear material, the lump effect, specially for uranium and plutonium assay (Prettyman et al., 1996; Anh and Dung, 2001); the drum-to-detector distance and the noisy data (Anh et al., 2005; Dung, 2006), etc.

Another measuring technique has been suggested and studied (Cesana et al., 1993) for the assay of the drums containing low-density waste, mainly consisting of organic materials such as contaminated paper, rags, protective clothing, shoes, etc. The measuring arrangement consists of two identical detectors set at equal distances from two bases of the drum. This technique was developed mainly because of three reasons: first, the measurement is usually limited to rather hard gamma rays emitted by Cs-137, Cs-134, Co-60, etc., the mass-absorption coefficients are nearly independent of the atomic number of matrix, and the linear attenuation coefficients are very low (typically 0.01–0.03 cm^{-1}) because of the low waste density (0.2–0.4 g/cm^3). Therefore, the gamma attenuation in whole drum can be considered by means of an average linear attenuation coefficient, independent of the position of the source in the drum; second, the number of drums to be examined is supposed to be very large, so that a detailed scanning by SGS is practically impossible; third, the measuring arrangement is very simple, so it can be used for any situation.

However, in this measurement the drum must lie down. Thus, it requests extra time and is not convenient for use in practice. Moreover, the systematic error is still very large if the activity is distributed in an extended region. To overcome these disadvantages a modification to the measuring arrangement with the geometric coefficient for cylindrical sample was proposed (Dung, 1997a, b, 2005).

In order to develop this technique, the systematic errors of the method have been evaluated by both experiments and calculations. The influence of several factors of importance, such as the spatial distribution of sources, attenuation coefficients, and homogeneity of matrix on the measuring results, was investigated. The results of this study, presented in this paper, may provide the guidelines for setting up a measuring system for assaying the radwaste drums.

2. Measuring principle

The measuring principle was proposed by Cesana et al. (1993) and modified by Dung (1997a, b). Two identical detectors are set perpendicularly to the drum axis at equal distances (see Fig. 1). If this distance is large enough so that the detector

* Corresponding author. Tel.: +84 8 8368865; fax: +84 8 8367361.
E-mail address: trandungquoc@gmail.com (T.Q. Dung).

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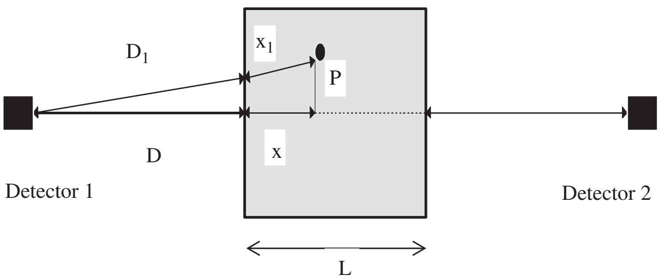

Fig. 1. Waste drum counting geometry. Two identical detectors are set perpendicularly to the drum axis at its middle point at equal distances.

size and the height of the drum can be neglected. The efficiency of Detector 1 for a point source located at point P can be expressed as

$$\varepsilon_1 = \frac{\alpha}{(D_1 + x_1)^2} e^{-\mu x_1}$$

where coefficient α is a function of the gamma-ray energy and the intrinsic efficiency of detector, D is the effective distance, and μ is the linear attenuation coefficient in the waste mixture.

Suppose that D is large enough than L, the width of the drum. So, $x_1 \sim x$, $D_1 \sim D$ and

$$\varepsilon_1 = \frac{\alpha}{(D_1 + x_1)^2} e^{-\mu x_1} \approx \frac{\alpha}{(D + x)^2} e^{-\mu x} = \frac{\alpha}{D^2} \frac{e^{-\mu x}}{(1 + (x/D))^2}$$

If $x/D \ll 1$, by using the series expansion of exponential function $e^x \approx 1+x$, so $(1+(x/D))^{-2} \approx e^{-2x/D}$ and

$$\varepsilon_1 \approx \frac{\alpha}{D^2} \frac{e^{-\mu x}}{(1 + (x/D))^2} \approx \frac{\alpha}{D^2} e^{-\mu x} e^{-2x/D} \approx \frac{\alpha}{D^2} e^{-(2/D+\mu)x} \tag{1}$$

If all the activity I concentrates in a very thin layer at a depth x, the count rate of Detector 1 can be expressed as

$$C_1 = I\varepsilon_1 = \frac{I\alpha}{D^2} e^{-\mu_1 x}$$

with $\mu_1 = 2/D + \mu$. Similarly, the count rate of Detector 2 is

$$C_2 = I\varepsilon_2 = \frac{I\alpha}{D^2} e^{-\mu_1 (L-x)}$$

and

$$(C_1 C_2)^{1/2} = I(\varepsilon_1 \varepsilon_2)^{1/2} = I \frac{\alpha}{D^2} e^{-\mu_1 L/2}$$

From the above equations, the geometric mean of the efficiencies is defined as

$$G = \frac{(C_1 C_2)}{I} = \frac{\alpha}{D^2} \exp(-\mu_1 L/2) \tag{2}$$

i.e. the geometric mean of the efficiencies does not depend on the source position. Thus

$$I = \frac{(C_1 C_2)^{1/2}}{G} \tag{3}$$

Before the estimation of the total activity the values of G in Eq. (3) must be determined. The values of α/D^2 versus gamma-ray energy can be determined by using appropriate standard sources. The attenuation coefficient can be evaluated by computation based on the fact that the mass attenuation coefficients of most organic materials are nearly equal to that of water μ_H. Thus the linear attenuation coefficients can be determined by the expression $\mu = \mu_H \rho$, where ρ is the density of the waste assayed. The value of G can also be experimentally determined by using a calibrated source placed next to the edge of the drum.

In the general case of an activity randomly disposed, the drum can be subdivided into many sheets. Supposing that I_i is the activity of the ith sheet at x_i, then the value of the coefficient G_T can be given as

$$G_T = \frac{\alpha \exp(-\mu_1 L/2)}{D^2} \left\{ 1 + 2 \sum_i \sum_{j>i} \frac{I_i I_j}{I^2} \times [\cosh[\mu_1 (x_i - x_j)] - 1] \right\}^{1/2} \tag{4}$$

where $I = \sum_i I_i$. From Eqs. (2) and (4) it can be concluded that the value of G approaches the value of G_T when the coefficient μ_1 is low or/and the total activity is concentrated in a small region, i.e. $x_i \sim x_j$.

Since the detectors view the curved surface of the drum, L is assumed to be a "mean thickness" of the drum and equal to $0.823d$, where d is the diameter of the drum and 0.823 is the geometrical coefficient (Augustson and Reilly, 1974; Sprinkle and Hsue, 1987). In other words, the drum is assumed to be a rectangular prism whose thickness is equal to $0.823d$. The activity I in the drum can be determined as expression (3) with

$$G = \frac{\alpha}{S^2} \exp -(\mu_1 \times 0.823 \times R) \tag{5}$$

where $S = K - 0.823R$; K is the distance from the detector to the centre of drum and R is the radius of drum, and

$$\mu_1 = 2/S + \mu \tag{6}$$

The proposed coefficient G in formula (5) is based on the approximations (Cesana et al., 1993; Dung, 1997a, b). The geometric mean of the efficiencies for the sources at different positions of vertical axis is considered to be the same. The systematic errors are caused mainly by the horizontal inhomogeneity of the source in the drum because an important assumption of this technique is that the activity is distributed in a small region of the drum volume. Therefore, these systematic errors have to be estimated.

3. Study and results

In principle, the measurement using two identical detectors at the same distances from the detectors to get count rate C_1 (of Detector 1) and C_2 of (of Detector 2) as given above is the s ame as two successive measurements with one detector: the first measurement gives C_1, and after turning the drum by 180° the second measurement gives C_2. Hence, when having only one detector the drum can be measured 2 times in two directions opposite to each other at the same distances from the detector.

The measuring arrangement is shown in Fig. 2. The detector sets perpendicularly to the drum axis at its middle point at equal distances from the curved surface of the drum.

Based on the measuring principles the calculations of activity incorporated in the drum were carried out and checked by experiment. In the experiment, a standard drum was used. Point sources Co-60 of 0.36 MBq and Cs-137 of 0.64 MBq were located at four specified radial positions in the different measurements as shown in Fig. 2. Matrices with various shapes and densities were made from clothing materials. The gamma-ray measurements

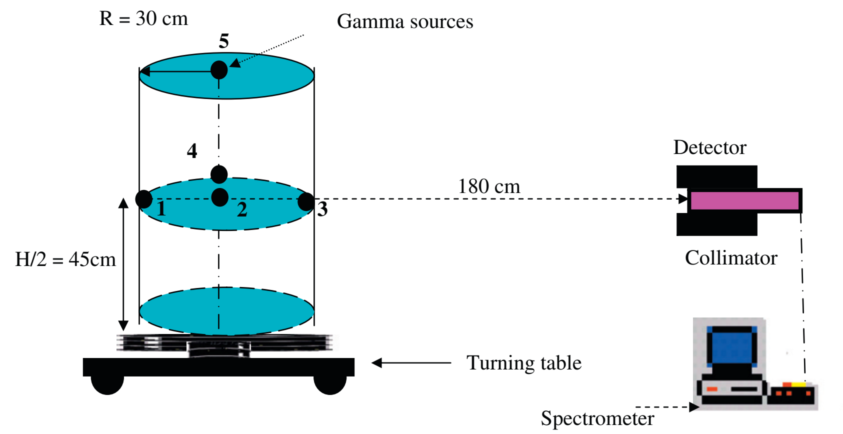

Fig. 2. Waste drum counting geometry. The drum is measured 2 times. In the second measurement the drum is rotated 180°.

were performed at energies of 611, 1173, and 1332 keV emitted by the Cs-137 and Co-60 sources. These data have resulted in a range of the average linear attenuation coefficients from 0.010 to 0.034 cm^{-1}. A high-purity germanium (HPGe) detector GEM50P4 and a standard digital spectrometer, model DSPEC.JR-312-Ortec, were used to record the gamma spectra. The GAMMAVISION 32 version 6.1 and COLEGRAM version 2.01 software packages were used for analysing the spectra.

In the measurements the statistical errors in the countings present usually the less uncertainty because this error can be under the direct control of the user. Frequent normalization and background checks can reduce the errors to low and acceptable levels. The most important sources of uncertainty in non-destructive techniques are the systematic errors related to the properties of the sample itself. The quality of the measurement depends on the assumption that the conditions under which the calibration has been done are identically reproduced during the measurement. Here the systematic errors of the proposed technique related to the properties of the sample itself, such as the spatial distribution of sources, attenuation coefficients, the homogeneity of matrix, etc., have to be investigated. The aims of our study were to verify the modelled systematic error by the experiments and to estimate the maximum systematic errors of this technique that provide the limitations to a precise determination of the nature and the extent of the radioactive contamination in the assay of radwaste drums.

The effects of the following parameters on the relative errors are estimated:

- The non-uniformity of the source.
- The density of matrices.
- The non-uniformity of matrices.

The results are given in the tables below. The experimental and calculated values of activity of the gamma sources in the drum are presented. The statistical uncertainties in the measurements are also given. The two main cases considered were as follows:

Point sources in homogeneous matrix: Here the error caused by a heterogeneous distribution of the radioactive sources can be estimated. Table 1a presents the result for the cases of point sources in homogeneous matrix. In Table 1b, the measured and calculated values with true value of the activity are compared. The errors that are caused mainly by the horizontal inhomogeneity of the source in the drum, such as the case of two sources with the same activity in positions 1 and 2, 1 and 3, can be evaluated.

Point sources in heterogeneous matrix: In principle, this is the worst case for the proposed system. For the experiments of the non-uniform distribution of matrices, the segment is divided into two parts with different attenuations. The value of the average attenuation coefficients μ was used to calculate the factor μ_1 in expression (6) and G by expression (5). The average attenuation coefficients were measured by the transmission technique with calibrated sources. The cases considered are illustrated in Fig. 3. The results are given in Table 2.

In the tables the pair of experimental and calculated values is given one by one. When the experimental value is not in agreement with the calculated value, they are displayed in bold italic.

In addition, numerous counting experiments with point sources in the different matrices were carried out. In the experiments many small pieces of matrix with different attenuation coefficients in the range from 0.011 to 0.034 cm^{-1} and sources were randomly arranged. The results give the values of systematic errors, not exceeding 30%.

4. Discussion

The experimental results show that the calculation model is satisfactory in view of the approaches for application. The results in the tables show that the experimental and calculated values are in agreement with 82% of the cases considered.

When the distance between the centre of the drum and the detectors is equal to about 4 times the half of the height of the drum, the maximum variation is less than 10% as shown in the case of the source at the bottom or top of the drum (position 5). As a result, the sources at different positions of vertical axis can be considered to be identical.

The results demonstrate that generally the proposed expression (5) satisfies the characteristic of the method: the more the total activity is distributed in a small range the higher the measurement accuracy is, found to be about 3–12% for almost all cases. The large errors (maximum value ~41%) occur when two sources with the same activity are in opposite positions as 1 and 3 positions (see Table 1b).

However, there are some exceptions. The reason is that the drum is assumed to be well approximated by a rectangular prism having thickness equal to 0.823 times of the drum diameter. This assumption leads to a difference between the path lengths of gamma ray in the drum (i.e. the real case) and in the prism (i.e. the idealised case), and it results in the difference between

Table 1
The measured and calculated values for homogenous matrix

(a) Point sources

Case of study		Source in position 1		Source in position 2		Source in position 4	
Gamma energy (keV)	Linear attenuation coefficient (cm^{-1})	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)
Cs-137, activity	0.016	0.623 ± 0.019	0.634 ± 0.006	0.592 ± 0.018	0.608 ± 0.009	0.793 ± 0.021	0.813 ± 0.012
0.64 MBq,	0.024	0.589 ± 0.017	0.608 ± 0.005	0.666 ± 0.020	0.582 ± 0.011	0.877 ± 0.025	0.864 ± 0.014
$E\gamma = 661$ keV	0.031	0.589 ± 0.018	0.582 ± 0.090	0.646 ± 0.019	0.557 ± 0.012	0.915 ± 0.027	0.947 ± 0.019
Co-60, activity	0.013	0.356 ± 0.012	0.360 ± 0.004	***0.323 ± 0.014***	***0.346 ± 0.006***	0.428 ± 0.018	0.421 ± 0.007
0.36 MBq,	0.020	0.348 ± 0.011	0.347 ± 0.003	0.331 ± 0.015	0.334 ± 0.005	0.468 ± 0.019	0.463 ± 0.008
$E\gamma = 1173$ keV	0.023	0.346 ± 0.013	0.342 ± 0.005	0.328 ± 0.017	0.328 ± 0.006	0.482 ± 0.021	0.479 ± 0.009
Co-60, activity	0.011	0.356 ± 0.011	0.364 ± 0.004	***0.324 ± 0.016***	***0.349 ± 0.005***	0.418 ± 0.017	0.410 ± 0.007
0.36 MBq,	0.019	0.349 ± 0.012	0.349 ± 0.004	0.328 ± 0.016	0.335 ± 0.006	0.461 ± 0.020	0.457 ± 0.008
$E\gamma = 1332$ keV	0.021	0.346 ± 0.014	0.346 ± 0.005	0.335 ± 0.018	0.331 ± 0.007	0.472 ± 0.021	0.468 ± 0.009

(b) Point sources in different positions

Case of study		Source in position 5		Two sources in position 1 and 2		Two sources in position 1 and 3	
Gamma source	Linear attenuation coefficient (cm^{-1})	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)
Cs-137, activity	0.016	0.518 ± 0.023	0.544 ± 0.011	0.710 ± 0.022	0.685 ± 0.005	0.736 ± 0.023	0.774 ± 0.012
0.64 MBq,	0.024	0.557 ± 0.025	0.518 ± 0.015	***0.742 ± 0.023***	***0.691 ± 0.006***	0.813 ± 0.025	0.832 ± 0.014
$E\gamma = 661$ keV	0.031	0.486 ± 0.024	0.493 ± 0.018	***0.819 ± 0.026***	***0.704 ± 0.013***	0.883 ± 0.027	0.902 ± 0.017
Co-60, activity	0.013	***0.295 ± 0.012***	***0.313 ± 0.004***	0.353 ± 0.011	0.382 ± 0.002	0.414 ± 0.017	0.425 ± 0.007
0.36 MBq,	0.020	0.290 ± 0.014	0.296 ± 0.006	0.374 ± 0.013	0.384 ± 0.004	0.445 ± 0.020	0.449 ± 0.008
$E\gamma = 1173$ keV	0.023	0.281 ± 0.017	0.292 ± 0.007	0.378 ± 0.014	0.385 ± 0.005	0.472 ± 0.021	0.464 ± 0.010
Co-60, activity	0.011	***0.288 ± 0.015***	***0.317 ± 0.003***	0.374 ± 0.012	0.382 ± 0.003	0.410 ± 0.018	0.421 ± 0.007
0.36 MBq,	0.019	0.324 ± 0.019	0.299 ± 0.007	0.367 ± 0.014	0.385 ± 0.004	0.421 ± 0.020	0.446 ± 0.009
$E\gamma = 1332$ keV	0.021	0.317 ± 0.021	0.295 ± 0.009	0.374 ± 0.013	0.385 ± 0.004	0.468 ± 0.021	0.454 ± 0.009

count rates of the real case and of the idealised case. Two typical situations are given here.

- A point source is placed at the centre of drum: since the path length (equal to $R = 29$ cm) of gamma ray in the drum is larger than that (equal to $0.823R = 23.87$ cm) in the prism, the count rates are reduced. The results bias to smaller value. The third column of Table 1a (a source in position 2) shows that the maximum error is 13% instead of +2.2% as expected.
- A point source is placed next to the edge of the drum (a source in position 4): contrary to the above case, here the path length (equal to 10.1 cm) of gamma ray in the drum is 2 times smaller than that (equal to 24.2 cm) in the prism, the count rates increase. So, the results bias to larger value. The last column of Table 1a shows that the maximum error is +48% instead of +1.5% as expected.

The data in Table 2 show that the effect of the non-uniform matrix to the systematic errors is small. Therefore, non-uniformity in chemical composition and density of matrix can be disregarded, and the gamma-ray attenuation for a given energy can be well approximated by means of an appropriate average linear attenuation coefficient.

In comparison with the SGS, the well-known technique that has been widely used to assay radioactive waste drums, the technique proposed in this paper has some particular advantages. The latter is more sensitive than the former (about 4–6 times), resulting in reduced statistical errors. This advantage arises from the fact that, while the proposed technique requires that the drum be counted once, the SGS technique requires a number of countings at different heights for each individual segment (in SGS a drum is divided usually into five to eight segments).

The measuring time of this technique is 5–8 times shorter than that of SGS technique. In this method, the measurement experimental apparatus is very simple and the calibration procedure and data handling are more straightforward than for the SGS technique.

As far as systematic errors are concerned, for the low-density radwaste, the maximum error (~40%) of this method is slightly larger than that (~30%) of the SGS technique. It is necessary to emphasize here that in many cases, when the attenuation coefficients are large, the errors of SGS can be from −88% to 450%. Therefore, the maximum error (~40%) of this technique can be acceptable.

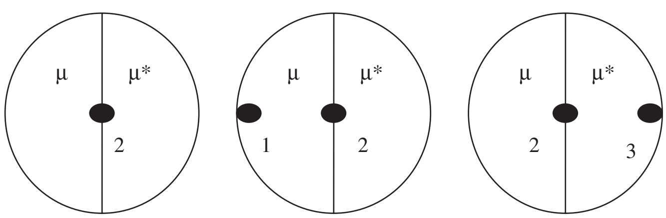

Fig. 3. Cases of the errors caused by non-uniform matrix having attenuation coefficients μ and μ^*: one source in position 2 and two sources in positions 1, 2 and 2, 3.

5. Conclusion

The above studies show the advantages of the proposed technique. It also confirms that this method can be developed for the assay of low-density radioactive waste, mainly consisting of organic materials. Combination of this and the SGS technique should give satisfying results in any situation. Based on the above investigation, the systematic errors can be estimated to fulfill the requirements of waste disposal regulators.

Table 2
The measured and calculated values of the point sources in heterogeneous matrix

Gamma source	Case of study	Source in position 2		Two sources in positions 1 and 2		Two sources in positions 2 and 3	
	Linear attenuation coefficient (cm^{-1}) μ–μ^*	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)	Experimental value of activity (MBq)	Calculated value of activity (MBq)
Cs-137, activity 0.64 MBq	0.017–0.026	0.602 ± 0.020	0.589 ± 0.007	***0.710 ± 0.021***	***0.672 ± 0.008***	0.734 ± 0.021	0.717 ± 0.009
	0.017–0.034	0.587 ± 0.018	0.582 ± 0.006	0.686 ± 0.019	0.659 ± 0.009	0.786 ± 0.023	0.755 ± 0.010
	0.026–0.034	0.589 ± 0.019	0.563 ± 0.007	0.702 ± 0.022	0.678 ± 0.011	***0.783 ± 0.024***	***0.730 ± 0.011***
	0.03–0.034	***0.582 ± 0.017***	***0.557 ± 0.007***	0.725 ± 0.021	0.710 ± 0.009	0.765 ± 0.021	0.736 ± 0.010
Co-60, activity 0.36 MBq	0.013–0.02	0.324 ± 0.014	0.338 ± 0.002	0.374 ± 0.012	0.374 ± 0.004	0.389 ± 0.012	0.392 ± 0.005
	0.013–0.025	0.320 ± 0.017	0.335 ± 0.003	0.367 ± 0.011	0.371 ± 0.003	0.403 ± 0.013	0.400 ± 0.004
	0.02–0.025	***0.306 ± 0.018***	***0.328 ± 0.003***	0.371 ± 0.012	0.378 ± 0.004	0.382 ± 0.011	0.392 ± 0.003
	0.022–0.025	0.306 ± 0.019	0.328 ± 0.004	0.378 ± 0.013	0.382 ± 0.004	0.378 ± 0.012	0.392 ± 0.005
	0.013–0.021	0.317 ± 0.019	0.338 ± 0.003	0.364 ± 0.012	0.374 ± 0.003	0.382 ± 0.011	0.392 ± 0.005
	0.013–0.024	0.313 ± 0.016	0.331 ± 0.003	0.367 ± 0.013	0.367 ± 0.004	0.396 ± 0.014	0.396 ± 0.004
	0.021–0.024	0.335 ± 0.018	0.328 ± 0.004	0.364 ± 0.011	0.382 ± 0.004	***0.374 ± 0.012***	***0.392 ± 0.004***
	0.022–0.024	0.335 ± 0.017	0.328 ± 0.004	0.371 ± 0.013	0.385 ± 0.003	0.371 ± 0.011	0.389 ± 0.004

Note: In general, the absolute error of the quantity A is displayed as ΔA. The uncertainties on the data in these tables have been calculated based on expressions (3), (5), and (6). Therefore, the transmission of the error of the activity (I) can be calculated by following formula:

$$\left(\frac{\Delta I}{I}\right)^2 = \frac{1}{4}\left[\left(\frac{\Delta C_1}{C_1}\right)^2 + \left(\frac{\Delta C_2}{C_2}\right)^2\right] + \left(\frac{\Delta \alpha}{\alpha}\right)^2 + (0.832R)^2 \times \left\{4\frac{(\Delta K)^2}{K^4} + \frac{1}{d^2}\left[\left(\frac{\Delta \alpha}{\alpha}\right)^2 + \left(\frac{\Delta C_1}{C_1}\right)^2 + 4\left(\frac{\Delta K}{K}\right)^2\right] - \right\}$$

Here the value of d is 30 or 60 cm. It depends on the position of the sources.

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