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Original Article

Source and LVis based coincidence summing correction in HPGe gamma-ray spectrometry

Jieun Lee, HyoJin Kim, Yong Uk Kye, Dong Yeon Lee, Jeung Kee Kim, Wol Soon Jo, Yeong-Rok Kang*

Research Center, Dongnam Institute of Radiological and Medical Sciences, Busan, 46033, South Korea

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ABSTRACT

The activity of gamma-ray emitting nuclides is calculated assuming that each gamma-ray is detected individually; thus, the magnitude of the coincidence summing signal must be considered during activity calculations. Here, the correction factor for the coincidence summing effect was calculated, and the detection efficiencies of two HPGe detectors were compared. The CANBERRA Inc. GC4018 high-purity Ge detector provided an estimate for the peak-to-total ratio using a point source to determine the coincidence summing correction factor. The ORTEC Inc. GEM60 high-purity Ge detector uses EFFTRAN in LVis to obtain the parameters of the detector and source model and the gamma-gamma and gamma-X match estimates, in order to determine the coincidence summing correction factor. Nuclide analyses, radioactivity comparisons, and analyses of reference material samples were performed utilizing certified reference materials to accurately determine the detection efficiencies. For both Co-60 and Y-88, the detection efficiency for a point source increased by an average of at least 12–13%, whereas the detection efficiency determined using LVis increased by an average of at least 13–15%. The calculated radioactivity values of the certified reference material and reference material samples were accurate to within 3% and 6% of the measured values, respectively.

1. Introduction

Given the long-term implications of the Chernobyl and Fukushima incidents, accurate and precise evaluations of the artificial radioactivity in environments, in particular, gamma nuclide analyses, have become significantly important. Gamma-ray spectrometry is an analytical method used to identify and quantify gamma-ray emitting isotopes in a variety of materials. In general, calibration of the HPGe detector efficiency is achieved using multiple gamma-ray standards that cover the entire energy range [1]. When the detector is close to the radioactive source, several radionuclides are detected owing to the coincidence summing effect, which is a result of the nuclides exhibiting relatively short half-lives and decaying within the time resolution of the detector. Therefore, the efficiency curves obtained from multiple radionuclides are different from those obtained from a radionuclide that emitting energy at a constant magnitude. If two or more gamma-rays are emitted from an excited nucleus in a cascade, the detector will consider these as a single event if the events are recorded within the resolution time of the detector. This is termed as the true coincidence summing effect, and it results in inaccurate count rates and erroneous results during data analyses. It should be noted that the coincidence correction effect is rarely accurate and routinely overestimates or underestimates the true activity of any seafarer or sample. Coincidence correction methods include peak-to-total (P/T) ratio calculations for a single gamma-ray emitting seafarer, Monte Carlo simulations, and true coincidence correction. In particular, the development of these correction methods has become an area of interest. Kolotov [2], Blaauw [3], and Rizzo and Tomarchio [4] reported a numerical calculation method for simultaneous synthesis in a theoretical system using a point source, whereas Hurtado [5], Vidmar [6], He [7], Badawi [8] and Kastlander [9] presented a novel approach to Monte Carlo simulation analyses [2–9].

In this study, the coincidence summing correction factor was calculated using a number of methods, and it was applied to the efficiency correction of a detector for authentication. The radioactivity of samples was validated with respect to the radioactivity of a

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* Corresponding author. Tel:. +82 51 720 5828; fax: +82 51 720 5826.
E-mail address: yeongrok@dirams.re.kr (Y.-R. Kang).

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standard reference substance.

2. Materials and methods

In this study, two detectors were used: a CANBERRA GC4018 high-purity Ge detector and an ORTEC GEM60 high-purity Ge detector. A certificated reference material was employed for the energy and efficiency calibrations of these two HPGe detector. For the GC 4018 HPGe detector, the coincidence summing correction factor was calculated using a point source. By contrast, the GEM 60 HPGe detector uses EFFTRAN in LVis to obtain the parameters of the detector and source model and the gamma-gamma and gamma-X estimates, in order to determine the coincidence summing correction factor. The coincidence summing correction factors can be obtained using any suitable and appropriately modified mathematical algorithm applicable to the sample. The full-energy peak efficiency values obtained before and after applying the coincidence summing corrections were compared. Lastly, deterministic efficiency calibration was applied during the nuclide analyses of the certificated reference material and reference material samples to validate its accuracy and thus confirm the radioactivity values.

2.1. Certificated reference material (CRM) and reference material (RM)

The CRM used for the energy and efficiency calibrations of the HPGe detectors was purchased from Korea Institute of Standards Science (KRISS). The materials used in the detector and sample models are presented in Fig. 1. The samples were placed directly on the detector housing. The calibration material contained multiple gamma-ray standard sources and was prepared in a cylindrical U8 beaker (90 mL). Calibration values were obtained using standard reference solutions containing Am-241, Cd-109, Co-57, Ce-139, Cr-51, Sn-113, Sr-85, Cs-137, Co-60, and Y-88. The detected gamma-ray energy ranged from approximately 59.54 keV–1836.05 keV. Table 1 lists the certified values of the reference nuclides and the photon emission intensity and activity. The sample was an RM sample obtained from KRISS. The water-based sample was obtained through the acid-treatment of a gamma-ray emitting, mixed nuclide substance. The sample was added to the cylindrical U8 beaker, which was then sealed. The sample was placed directly on the detector housing. The overall measurement time was 300,000 s for both the GC4018 and the GEM60 HPGe detectors. It was found that the sample contained traces of Cs-134, Cs-137, Cr-51, and Co-60. Table 2 lists the experimental conditions for the sample.

Table 1
Certified reference material values and uncertainties.

Nuclide	Energy (keV)	Photon emission intensity (%)	Activity (Bq)[a]
Am-241	59.54	35.92	612 ± 25
Cd-109	88.03	3.66	3357 ± 140
Co-57	122.06	85.49	141.4 ± 5.7
	136.47	10.71	
Ce-139	165.86	79.90	198.6 ± 8.0
Cr-51	320.08	9.89	29138 ± 1200
Sn-113	391.70	64.97	503 ± 21
Sr-85	514.00	98.5	726 ± 29
Cs-137	616.66	84.99	308 ± 13
Co-60	1173.23	99.85	359 ± 15
	1332.49	99.9826	
Y-88	898.04	93.7	973 ± 39
	1836.05	99.346	

[a] Confidence level: approx. 95%, $k = 2$.

2.2. High-purity Ge detector

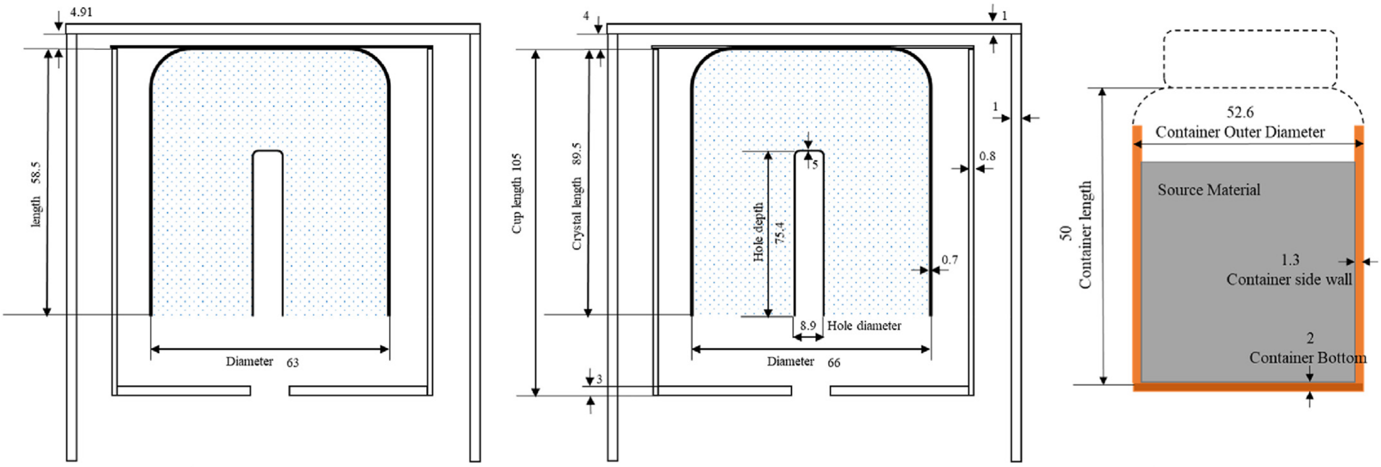
The specific activity of each sample was measured using two p-type HPGe detectors. The first of these was the GC4018 detector, which featured a diameter of 63 mm and a relative efficiency of 40%. The second was the GEM60 detector, which exhibited a diameter of 66 mm and a relative efficiency of 60%. The specifications of both these detectors are listed in Table 3.

2.3. Coincidence summing effect and correction methodology

Energy and efficiency calibrations of the GC4018 and GEM60 detectors were performed using certified reference materials. The certificated reference source was placed in a container, to which Co-60 and Y-88 were added. The gamma-rays emitted from the source

Table 2
Experimental conditions.

Detector	GC4018/GEM60
Time	300000 s
Container Material	Polyethylene
Reference Data	2020-06-01



(a) GC 4018 HPGe detector (b) GEM 60 HPGe detector (c) CRM & RM samples

Fig. 1. Schematic presentation of the setup for the case of CRM & RM sample. The (a), (b) and (c) images show the physical characteristics of the GC4018 HPGe detector, the GEM 60 detector and the CRM and RM sample, respectively. Note that the figures are not to scale. All dimensions are given in millimeters (mm).

Table 3
Detector specifications.

Detector	GC4018	GEM60
Corporation	CANBERRA	ORTEC
Operating Voltage	+3500 V	+1700 V
Relative Efficiency	40%	60%
Detector Diameter	63 mm	66 mm
Resolution	1.8 keV at 1.33 MeV	2.14 keV at 1.33 MeV
Coincidence correction method	Point Source	LVis software

exhibited a low peak efficiency at their point of detection, owing to the coincidence summing effect. Therefore, it was essential to correct the coincidence summing effect caused by the limited resolution time of the detector during gamma-ray detection. The detector calculates the coincidence summing correction coefficient using several methods. In the case of the GC4018 detector, Genie-2000 was initially used to measure the radioactivity of the point source; the software was then used to analyze the background correction, alongside the major gamma-ray energies detected. Lastly, these values were used to determine the P/T ratio. The algorithm used in Genie-2000 is based on the study of Kolotov [2], among others, which focused on the calculation of correction factors for volume sources. The total correction factor was determined by integrating the data gathered from each energy position, as recorded from the point source, alongside the total correction factor recorded from the previous seafarer; the following formula was adopted:

$$COI_{g,i} = \left(1 - L_g\right) \times \left(1 - S_a\right)$$

where L_g is the probability of summing out, and S_a is the probability of summing, as reported in Ref. [10]. The point sources used to determine the coincidence summing correction factors were Cd-109, Co-57, Sn-113, Cs-137, Mn-54, and Zn-65. Each of these nuclides emitting energy at a single intensity; these were placed 10 cm from the detector. The GEM60 detector can be operated such that Gamma Vision and LVis run simultaneously, and the data are evaluated under the Gamma Vision operating conditions, as defined by the initial parameter set. In EFFTRAN, the efficiency transfer method is used to calculate the efficiencies; therefore, calibration with a standard source is required. The two virtual total efficiencies are computed using the code, and the point source full energy peak efficiency is measured. The result of the method is the required full energy peak efficiency of the volume source [6]. The measured efficiency can then be transferred to a sample that differs from the standard one in terms of size, composition, and density. The coincidence library contains decay data on 300 radionuclides. True coincidence summing correction factors are provided considering the gamma-rays and gamma-gamma coincidences. Information pertaining to the geometry of both the detector and the sample are added to the system; thus, the initial parameters are set. Subsequently, energy calibration, coincidence summing effect, and geometry correction calculations are performed using the software to obtain the geometry correction and correction library correction coefficients. The resulting correction is then applied to the efficiency of the detector [11].

3. Experimental results and discussions

The coincidence summing correction factor was calculated using the point source alongside LVis and then applied for the efficiency calibration of the detector, in order to facilitate an accurate comparison of the radioactivity exhibited by the certified reference materials. For both methods, the detection efficiency was corrected using Co-60 and Y-88. To this end, the radioactivity of both the sample substance and the nuclide were recorded, and the values were compared.

3.1. *Efficiency and activity of certificated reference material*

The full energy peak efficiency was calculated for a certified reference material. The calculated coincidence summing correction factor was applied to the efficiency correction, which was calculated using the following formula:

$$\varepsilon = \frac{n(E)}{A \times P_\gamma(E)}$$

where n(E) represents the net count, A is the total source activity for all radionuclides emitting gamma photons with varying energies (E), and $P_\gamma(E)$ is the photon emission intensity. The uncertainty factor was considered during the determination of the following parameters: net count, activity, photon emission intensity, correction for decay during detection, and coincidence summing correction factor. Table 4 and Fig. 2 show the comparative detector efficiencies. As can be observed in Fig. 2, the peak efficiency reaches a maximum at approximately 122 keV and then decreases monotonically as the energy increases. The detection efficiencies of the Am-241, Cd-109, Co-57, Ce-139, Cr-51, Sn-113, Sr-85, and Cs-137 nuclides within the certified reference materials were compared. For single energy-emitting nuclides, the coincidence summing effect is negligible. As can be seen from Fig. 2, after applying the coincidence summing effect correction, the corrected detection efficiencies of the Co-60 and Y-88 nuclides increased. The fitted parameters A0, A1, A2, A3, A4, and A5 were then obtained via polynomial fitting, and the results are presented in Table 5. Finally, the efficiency equation was established based on the known fitted parameters; this enabled efficiency evaluations at particular energies for Co-60 and Y-88. In the case of the GC 4018 HPGe detector, the detection efficiency of Co-60 and Y-88 increased by 13% after applying the coincidence summing effect correction. For the GEM 60 HPGe detector, the detection efficiency of Co-60 and Y-88 increased by 13–15% after applying the coincidence summing effect correction. Furthermore, the detection efficiency improved by 13% or more once the coincidence summing effect exhibited by both the point source and LVis was considered. Table 6 presents the radioactivity exhibited by the certified reference materials after applying the non-coincidence and coincidence summing correction. When using the detection efficiency corrected for the coincidence summing effect, it coincided with the radioactivity value of the certificate. It was confirmed that the process of calculating the radioactivity of the certified reference material considering the coincidence summing effect was accurate to within 3% of the measured value. Table 7 and Fig. 3 present the correction factors calculated based on the point source and LVis for both Co-60 and Y-88, which are the most commonly used radionuclides for efficiency curve calibration. At the gamma energies of Co-60 and Y-88, the coincidence corrected factor obtained using LVis was higher than that determined based on the point source.

Table 4
Detection efficiency before and after applying coincidence summing correction.

Energy (keV)	GC 4018		GEM 60	
	Non-corrected Efficiency (x10^{-3})	Corrected Efficiency (x10^{-3})	Non-corrected Efficiency (x10^{-3})	Corrected Efficiency (x10^{-3})
59.54	26.21 ± 1.11	26.21 ± 1.11	25.65 ± 0.64	25.75 ± 0.64
88.03	53.75 ± 2.70	53.75 ± 2.70	53.01 ± 1.32	52.53 ± 1.31
122.06	62.30 ± 2.51	62.30 ± 2.51	58.88 ± 1.47	59.86 ± 1.49
136.47	58.65 ± 1.56	58.65 ± 1.57	58.49 ± 1.46	59.01 ± 1.47
165.86	56.51 ± 1.81	56.87 ± 1.84	55.63 ± 1.39	60.03 ± 1.50
320.08	35.53 ± 0.93	35.53 ± 0.92	37.73 ± 0.94	37.35 ± 0.93
391.70	30.55 ± 0.73	30.55 ± 0.72	32.55 ± 0.81	32.05 ± 0.80
514.00	26.13 ± 0.58	26.13 ± 0.59	26.65 ± 0.66	27.53 ± 0.69
661.66	21.35 ± 0.45	21.35 ± 0.47	22.19 ± 0.55	23.56 ± 0.59
898.04	15.14 ± 0.36	17.02 ± 0.39	17.84 ± 0.45	19.96 ± 0.50
1173.23	12.40 ± 0.32	14.08 ± 0.36	14.71 ± 0.37	16.65 ± 0.42
1332.49	11.24 ± 0.28	12.78 ± 0.33	13.37 ± 0.33	15.32 ± 0.38
1836.05	8.73 ± 0.34	9.89 ± 0.39	10.32 ± 0.26	12.37 ± 0.31

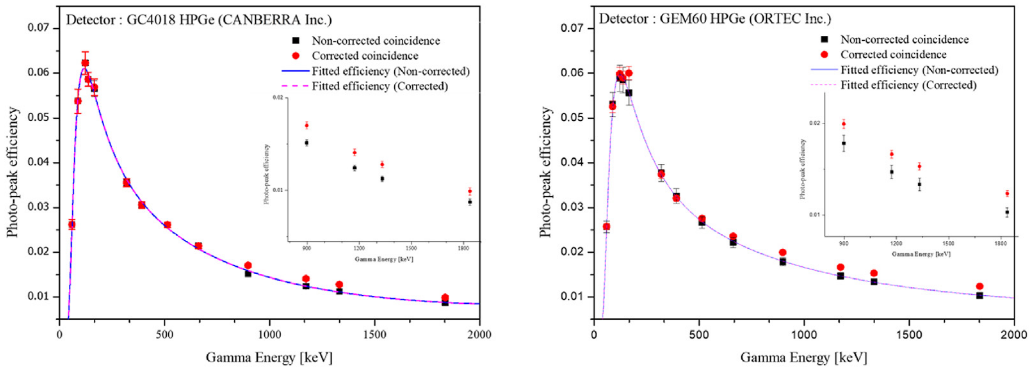


Fig. 2. Detection efficiency before and after applying coincidence summing correction. The straight dashed lines represent the fitting efficiencies for 13 energies of gamma emitters, squares denote non-corrected coincidence photo-peak efficiencies, and the circles denote corrected coincidence photo-peak efficiencies corresponding to these energies.

Table 5
Fitting parameters of Genie-2000 and LVis.

Parameter	Genie-2000		LVis	
	Non-corrected	Corrected	Non-corrected	Corrected
A_0	−173.5948	−134.7169	−125.0725	−94.2299
A_1	327.5706	244.2335	226.6677	160.3946
A_2	−247.4782	−176.8424	−164.4718	−108.2885
A_3	93.1028	63.5223	59.3130	35.8313
A_4	−17.4687	−11.3510	−10.6598	−5.8224
A_5	1.3067	0.8071	0.7635	0.3707

Table 7
Coincidence correction factors calculated for Co-60 and Y-88.

Nuclide	Energy (keV)	Genie-2000	LVis
Y-88	898.04	0.939	1.154
Co-60	1173.23	0.924	1.165
Co-60	1332.49	0.921	1.170
Y-88	1836.05	0.92	1.171

Table 6
Comparison between certificated and measured radioactivity values.

Nuclide	Certification Activity (Bq)	GC4018 Detector		GEM60 Detector	
		Non-corrected Activity (Bq)	Corrected Activity (Bq)	Non-corrected Activity (Bq)	Corrected Activity (Bq)
Am-241	612 ± 25	612 ± 27	612 ± 27	650 ± 33	631 ± 35
Cd-109	3357 ± 140	3355 ± 192	3355 ± 193	3340 ± 174	3328 ± 186
Co-57	141 ± 6	140 ± 4	140 ± 4	143 ± 6	140 ± 6
Ce-139	199 ± 8	204 ± 7	204 ± 7	196 ± 8	198 ± 9
Cr-51	29138 ± 1200	28484 ± 741	28980 ± 753	28526 ± 1170	28140 ± 1323
Sn-113	503 ± 21	492 ± 12	495 ± 12	493 ± 19	502 ± 22
Sr-85	726 ± 29	734 ± 18	749 ± 19	749 ± 27	750 ± 32
Cs-137	308 ± 13	324 ± 7	310 ± 7	326 ± 12	314 ± 14
Co-60	359 ± 15	324 ± 10	357 ± 6	350 ± 13	354 ± 15
Y-88	973 ± 39	951 ± 25	974 ± 20	992 ± 48	969 ± 52

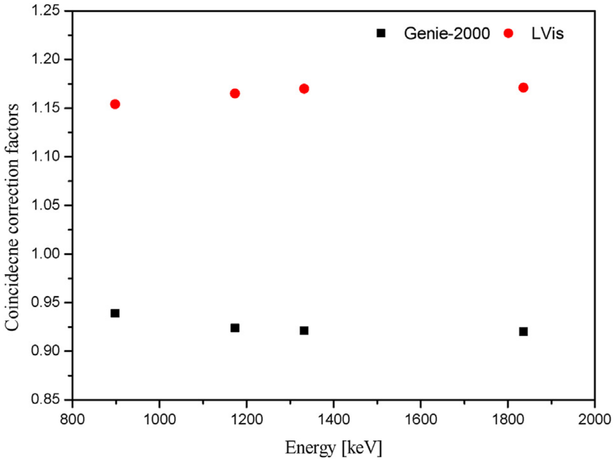


Fig. 3. Coincidence correction factors of the point source and Lvis.

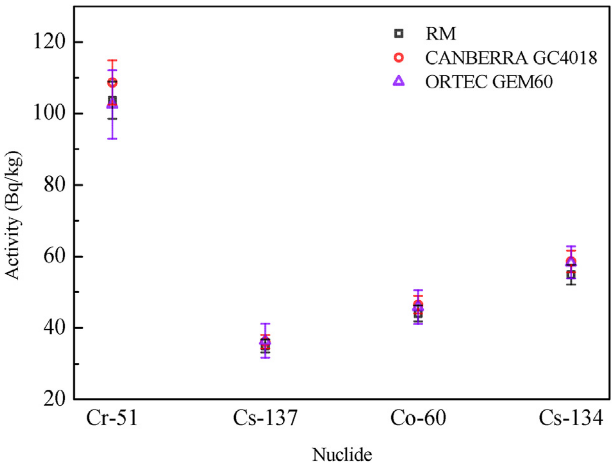


Fig. 4. Comparison of the radioactivity values obtained from the RM samples.

3.2. Radioactivity of RM samples

Fig. 4 shows the measurement and analysis results for the RM sample material when using both detectors; the detectors were calibrated for energy and detection efficiencies. The radioactivity of the four nuclides in each of the samples was recorded using both the detectors, the results of which are presented in Table 8 and Fig. 4. The uncertainty ($k = 2$) in the recorded radioactivity values was 1.90–6.17% and 4.58–9.58% for the GC4018 and GEM60 detectors, respectively.

As a KOLAS-certified testing/calibration institution, the Z -scores of the RM samples were evaluated using the statistical evaluation method (ISO13528: 2015). The Z -score is a standardized measure of performance; it is calculated using the participant result, assigned value, and the combined standard uncertainties for the result and the assigned values. Z -scores can be interpreted in the same manner as Z -scores and by using the same critical values of 2,0 and 3,0, depending on the proficiency testing scheme design. In the case of the GC4018 detector, the Z -scores ranged from 0.97 to 1.49, and the evaluation result was deemed satisfactory. In the case of the GEM60 detector, the Z -scores ranged from 0.91 to 1.47, and the evaluation result was deemed satisfactory. The parameters considered when calculating the radioactivity of the sample substances included the peak count rate, full energy peak efficiency, photon emission intensity, measurement time, sample weight, coincidence summing coefficient, and decay correction coefficient. The uncertainty assessments are presented in Table 9. The uncertainty arising due to the detection efficiency of the detector has the largest contribution, amounting to 2.5%. By contrast, the uncertainty due to the coincidence summing correction was significantly smaller, at 0.1%.

Table 9
Uncertainty factor.

Uncertainty Contribution	Probability (%)
Peak area evaluation	1.0
Efficiency	2.5
Photon emission intensity	0.1
Time	0.1
Weight	0.01
Coincidence summing	0.1
Decay of radionuclide	0.01

4. Conclusions

Energy and efficiency calibrations were achieved using high-purity, p-type Ge detectors produced by CANBERRA and ORTEC. The results were obtained through the analyses of certified reference materials. In the case of the GC4018 detector, a point source was measured to determine the coincidence summing factor via P/T calibration; by contrast, the GEM60 detector uses EFFTRAN in LVis to obtain the parameters of the detector and source model match estimates to determine the coincidence summing correction factor. By determining the coincidence summing effect using the point source and LVis and applying it to the efficiency correction for the detector, the detection efficiency was increased by at least 13% for both the Co-60 and Y-88 nuclides. The calculated radioactivity of the certified reference material was within 3% of the measured radioactivity value. Following the RM sample analysis, the radioactivity values obtained for the RM sample materials were within 6% of the measured radioactivity values obtained from the GC4018 and GEM60 detectors. It was found that only LVis could correct the coincidence summing effect easily and efficiently, without requiring additional measurements of the point source. Even when sources with different sizes were used, the detection efficiency corrected for the coincidence summing effect can be obtained and applied, such that it is feasible and expected to be utilized for various environmental samples and experimental conditions.

Table 8
Sample nuclide radioactivity and analysis.

Nuclide	RM		GC4018		GEM60	
	Activity (Bq/kg)	Uncertainty ($k = 2$)	Activity (Bq/kg)	Uncertainty ($k = 2$)	Activity (Bq/kg)	Uncertainty ($k = 2$)
Cr-51	103.7	5.2	108.71	6.17	102.5	9.58
Cs-137	35.0	1.8	36.17	1.90	36.44	4.72
Co-60	44.1	2.2	46.47	2.47	45.81	4.72
Cs-134	54.9	2.8	58.61	3.02	58.32	4.46

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.

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