



Validation of an efficiency calibration procedure for a coaxial n-type and a well-type HPGe detector used for the measurement of environmental radioactivity



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ABSTRACT

To obtain reliable measurements of the environmental radionuclide activity using HPGe (High Purity Germanium) detectors, the knowledge of the absolute peak efficiency is required. This work presents a practical procedure for efficiency calibration of a coaxial n-type and a well-type HPGe detector using experimental and Monte Carlo simulations methods. The method was performed in an energy range from 40 to 1460 keV and it can be used for both, solid and liquid environmental samples. The calibration was initially verified measuring several reference materials provided by the IAEA (International Atomic Energy Agency). Finally, through the participation in two Proficiency Tests organized by IAEA for the members of the ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity) the validity of the developed procedure was confirmed. The validation also showed that measurement of ²²⁶Ra should be conducted using coaxial n-type HPGe detector in order to minimize the true coincidence summing effect.

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1. Introduction

Many approaches have been proposed in the literature for efficiency calibration of HPGe detectors when environmental samples are measured [1–3]. Set of standard point sources having single energy emissions, standard solution of mixed radionuclides and Certified Reference Materials commercially available, are commonly employed. In most of these cases, the activity measured by this method has to be corrected for summation effects induced by photons emitted in coincidence and also for self-absorption when the measured sample has a different matrix (density or composition) than the source used in the calibration process. A direct calibration can be performed using standard radioactive sources of the same geometrical dimensions, density, and chemical composition, compared with the samples of interest. However, standards are not often available for all environmental matrices or for all radionuclides of interest.

In addition, theoretical and computational methods have also been employed for efficiency calibration, coincidence-summing and self-absorption corrections. In several studies have been used general

and specific software based on Monte Carlo (MC) codes like Geant, MCNPX, EFFTRAN or DETEF [4–7]. A good agreement between experimental and calculated data can be reached using these codes; in addition the working time can be reduced considerably. In these cases, main limitations reside in the precise knowledge about the characteristics of the experimental geometry and sample compositions [8]. Generally, the efficiency values obtained experimentally and by MC simulation based on nominal values of the parameters supplied by the manufacturer show significant differences due to the inaccuracy in some critical parameter like the thickness of the dead layer or the active volume. The optimization of these parameters can result in a substantial decrease of the deviations between the experimental and calculated values [9,10]. However, even when precise geometrical data are available, it is necessary to refine the model by feeding it back with experimental results when accuracy is desired. This is because some parameters involved in the detector response cannot easily be assessed. They include the distribution of the electrical field in the crystal, its mounting and dimensions and properties of the dead layers [11,12].

One effective procedure to overcome these difficulties is to use an efficiency transfer function from reference geometry to other source configurations, using MC calculations and experimental measurements. This procedure consists of calculating the full energy peak efficiency (FEPE) by an energy dependent transfer

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factor, which is derived by comparing the direct calculated FEPEs with the source experimental values at a reference position. In the literature, different authors have reported differences below 5% after using the transfer function [13,14]; even when there was no adjustment of parameters of the detectors.

Independent of the chosen calibration method (experimental or computational), there is a group of limitations which will be required to overcome. However, the combination of these techniques can be a potential tool as a practical and cheap method for routine measurement purpose in many laboratories. The goal of this study is to develop a simple procedure for efficiency calibration of two different HPGe detectors, complementing experimental and MC simulation methods. The main advantages of this approach is that it can be applied to coaxial detectors as well as well detectors and it can be used for different environmental matrices. The method was verified measuring several IAEA reference materials and finally validated through the participation in two ALMERA Proficiency Tests organized by IAEA for the ALMERA network members.

2. Materials and methods

2.1. Detectors

Two high purity germanium (HPGe) detectors were used for experimental measurements. Detector 1 (D1) was a coaxial n-type detector (model NGC 3019, from DSG Detector Systems GmbH) with epoxy-carbon window and 31.5% of relative efficiency. It was coupled to an electronic chain, including a multichannel analyzer type TMCA. Detector 2 (D2) was a well-type HPGe detector (model EGPC100 P-15, from Canberra) with an absolute efficiency of 12.1% at 661 keV. The data acquisition system of this detector consists of a PSC822 pre-amplifier, Canberra amplifier model 7245 and electronic card MCA 5000 which includes a 7602 ADC with 8192 channels and InterFast multichannel analyzer. Both detectors are surrounded by a cylindrical low-background chamber made with the following elements from outside to the inner region: 240 mm of steel, 37 mm of lead, 1 mm of aluminum and 1 mm of copper. In both detectors the gamma spectra were recorded and analyzed using WinnerTM 6.0 software. The detectors resolution and energy calibration is periodically verified for stability using a set of point sources (²⁴¹Am, ¹³⁷Cs, ⁶⁰Co and ²²⁶Ra).

2.2. Monte Carlo simulation

The first step of the calibration procedure was to obtain the efficiency calibration curves for both detector (D1 and D2) by Monte Carlo simulation methods. It was employed the code MCNPX 2.6. The efficiency response for both detectors have been previously reported in the studies [6,15]. Nominal values of the parameters supplied by the manufacturer are described in those studies.

For detector D1 the Monte Carlo efficiency transfer method was applied using directly the manufacturer supplied data in all MC calculations. The efficiencies were calculated using the expression $\epsilon_x = \epsilon_{ref} (\epsilon_x^{MC} / \epsilon_{ref}^{MC})$ where ϵ_x is the efficiency for a particular geometry and energy, ϵ_{ref} is the experimental efficiency for a reference case, and ϵ_{ref}^{MC} and ϵ_x^{MC} are calculated efficiencies (via Monte Carlo) for the reference case and the geometry of interest, respectively. The characteristics of the reference source and source-detector configuration are described in [6]. We used the detector model described in [6] to compute the efficiencies of the samples that were measured here (ϵ_x^{MC}) and we used the same values of ϵ_{ref} and ϵ_{ref}^{MC} measured and calculated in this work, respectively.

For detector D2 the Monte Carlo efficiency calculations were made directly. In this case, a tuning of some critical parameters of the detector was made. This tuning showed that the thickness of

external dead layer (EDL) and distance between the Ge crystal and the Al end cap (DGA) are critical parameters and they were optimized. For the rest of parameters we used the nominal values.

We used the pulse-height tally (F8) per photons emitted from the source to compute the absolute efficiency and we generally obtained relative errors lower than 1% with a number of histories about 10^5 – 10^6 and 12–14 min of computational times for each energy. This computational time per energy allows us to build an efficiency calibration curve in approximately 4 h. Therefore, the proposed calibration method is good for practical application in everyday measurements. All MC calculations covered the energy range 40–1460 keV.

2.3. True coincidence summing corrections

The second step consisted of the determination of the coincidence summing correction factors (TCSs). For this purpose a simple experimental technique was applied. The corrections were estimated by measuring of a sample, containing the radionuclides of interest with summing effect and one single-emitter radionuclide as reference, in two detector geometries (near and far) [16,17]. The coincidence summing correction factor is defined by the relations $TCS = \left(\frac{R_r}{R_s}\right)_h / \left(\frac{R_r}{R_s}\right)_g$ where R_r is the count rates of the single-emitter radionuclide of reference and R_s is the count rates of the radionuclide to correct. The ratios $\left(\frac{R_r}{R_s}\right)_h$ and $\left(\frac{R_r}{R_s}\right)_g$ are calculated at height h from the detector where coincidence summing is negligible and over the detector (indicated by g), respectively. This method is simple to use and it is independent of the sample activity which contribute to minimize several uncertainties.

Water solution of unknown activity with the following radionuclides of interest: ¹³⁴Cs, ¹⁵²Eu, ⁶⁰Co and ¹³⁷Cs, was used. For the reference we used ¹³⁷Cs in the middle energy of the spectra (661.7 keV). For detector D1 the sample was measured at 10 cm from the end cap and over the detector while for detector D2 the sample was measurement out of the well (at 6 cm from the top of the well) and into the well of the detector. In order to calculate the corrections factors, the TCS expression was applied to the following energy lines: ¹³⁴Cs (604.7 and 795.7 keV), ¹⁵²Eu (344.3 keV) and ⁶⁰Co (1173.2 and 1332.5 keV). It should be noted that no correction was applied to ²²⁶Ra and ²³²Th because the available water source did not contain these radionuclides. All measurements were carried out for about 6–10 h ensuring a meaningful statistics in each source to detector configuration and in all cases the dead time corrections were intrinsically taken into account by the software WinnerTM.

2.4. Experimental verification

The experimental verification included the internal validation of MC efficiency curves and the participation in two ALMERA Proficiency Tests: IAEA-TEL-2013-04 and IAEA-TEL-2014-04 [18].

The validations of the efficiency calibration curves obtained by Monte Carlo simulation was performed with high purity KCl and several Certified Reference Materials (CRMs) provided by IAEA [19]: Uranium Ore IAEA-RGU-1, soil IAEA-375, IAEA-326 and IAEA-444, marine sediments IAEA-300 and IAEA-306, and grass IAEA-373. For detector D1 the samples were encapsulated in a cylindrical container of polystyrene of 1.8 mm thickness with an internal diameter of 72 mm and filling height of 20 mm. The measurement geometry of detector D2 was a cylindrical polystyrene vial of 1 mm thickness with an internal diameter of 10 mm and filling height of 45 mm. Both capsules do not allow radon diffusion. In the sample IAEA-RGU-1, the radionuclides ²²⁶Ra and ²¹⁰Pb are in secular equilibrium. The specific activity of ⁴⁰K was determined considering the elemental weights for KCl and ⁴⁰K natural abundance.

The 2013 ALMERA proficiency test (PT) consisted of three water samples and one flour sample. The participating laboratories were

requested to analyze man-made and natural gamma emitting radionuclides in water samples, and ^{134}Cs and ^{137}Cs in the flour sample. The subsequently PT (ALMERA 2014) consisted of three water samples, one seaweed sample, one sediment sample and one water sample from oil field. In this case, the participating laboratories were requested to analyze anthropogenic and natural gamma-emitter radionuclides in the water samples, gamma-emitter radionuclides in the seaweed and sediment samples, and ^{226}Ra in the water sample from oil field. One water sample containing known radioisotopes and activity concentrations was provided in each PT for quality control purpose.

The measurements were carried out in the low-background systems described above; placing the samples over the front end cap or into the well of the detectors D1 and D2, respectively. For ^{226}Ra activity measurement the samples were sealed and kept for a minimum of three weeks, to ensure that secular equilibrium between ^{226}Ra and radon daughters had been achieved. The activity of the samples was determined via its daughters ^{214}Pb (351.9 and 295.2 keV) and ^{214}Bi (609.3 keV). The ^{232}Th activity was determined from the activity of ^{212}Pb (238.6 keV), ^{208}Tl (583.2 keV) and ^{228}Ac (911.2 keV). Two gamma-energies were also analyzed for ^{134}Cs (604.7 and 795.7 keV) and ^{60}Co (1173.2 and 1332.5 keV). In all cases the activity concentration was reported as the arithmetic mean. ^{238}U activity was determined from the activity of ^{234}Th (63.3 keV) and only one gamma-energy was used for ^{152}Eu (344.3 keV). Finally, to determine ^{40}K , ^{137}Cs , ^{241}Am and ^{210}Pb activities, the well-known 1460.7, 661.7, 59.54 and 46.54 keV gamma-energies were used, respectively. Activities of ^{60}Co , ^{134}Cs and ^{152}Eu were corrected applying the calculated TCSs coefficients. Also, decay-corrections were applied to reference materials and samples provided by ALMERA PTs with the reference time given in their Certificates or with the one requested by the PT instructions.

The determined activity concentrations and its combined standard uncertainty were expressed in Bq/kg on a dry-mass for flour, seaweed and sediment samples and as Bq/kg for water samples. The measured results uncertainties were reported as a combined standard uncertainty at 1 sigma level. Uncertainties were calculated using the law of uncertainty propagation taking into account the following components: sample and reference source counting statistics, background correction, photopeak efficiency fitting, Monte Carlo statistical uncertainties and coincidence summing correction. In our laboratory this procedure was accredited by the Normalization National Office for ISO-NC-17025, and it is recognized by the IAEA through the ARCAL XXVI IAEA Regional Project since 2005.

The use of Monte Carlo codes makes necessary to have a detailed description of the geometric intrinsic characteristics of the detector and geometry characteristics of the sample container, which were described above. But also, the sample chemical composition was needed. The major elemental composition of CRMs used in this study is known and it is available in their respective reports [19]. For sediment we used the chemical composition of the IAEA-SL-3 reference material (lake sediment from Austria) [19]. For flour we used the following composition: C (40%), O (40%) and H (20%) and for seaweed C (42%), O (50%), H (5%) and N (3%). Finally, for KCl and H_2O their elemental weights were considered. Once each sample of interest was directly implemented for MC calculations the self-absorption corrections were not necessary.

3. Results and discussions

3.1. Efficiency calibration and internal verification

The efficiency response of detector D1 and D2 was reported in previous researches. Monte Carlo efficiency transfer method was applied for D1 using directly the nominal parameters of the

detector [6]. The method was conducted in the 40–1408 keV energy range and was obtained a good agreement between measurement and calculated values in three reference materials: DL-1a, IAEA-375 and IAEA-RG-1. In the present study, the efficiency transfer calculations were extended to other IAEA reference materials, environmental samples and liquid samples.

On the other hand, only the response at 46.54 keV was studied for detector D2 [15], while in this work the whole energy range was studied. For this detector, a tuning of some critical parameters was made in order to optimize the efficiency response at middle and high energies. We compared the calculated and measured efficiency values for different filling heights of the sample into the measurement geometry at two energies: 661.7 keV (measuring ^{137}Cs in the IAEA-375 reference material) and 1460 keV (measuring ^{40}K in KCl). The tuning was previously applied by Morera-Gómez et al. in [15] for the low energy of 46.54 keV from ^{210}Pb in DL-1a. Fig. 1 show that a good agreement between the calculated and measured efficiency values was achieved using 0.75 mm for external dead layer (EDL), 12 mm for distance between the Ge crystal and the Al end cap (DGA), and the nominal values of the rest of parameters. With the optimizations, the average of relative deviations between computer and experimental data decreased from 2.9% to 0.99% at the energy of 661.7 keV and from 6.4% to 2.0% at 1460 keV. These adjusted parameters were used in this work for all MC calculations in detector D2.

The efficiencies calculated in the entire energy range (40–1460 keV) were loaded to the software for gamma-ray spectrometry

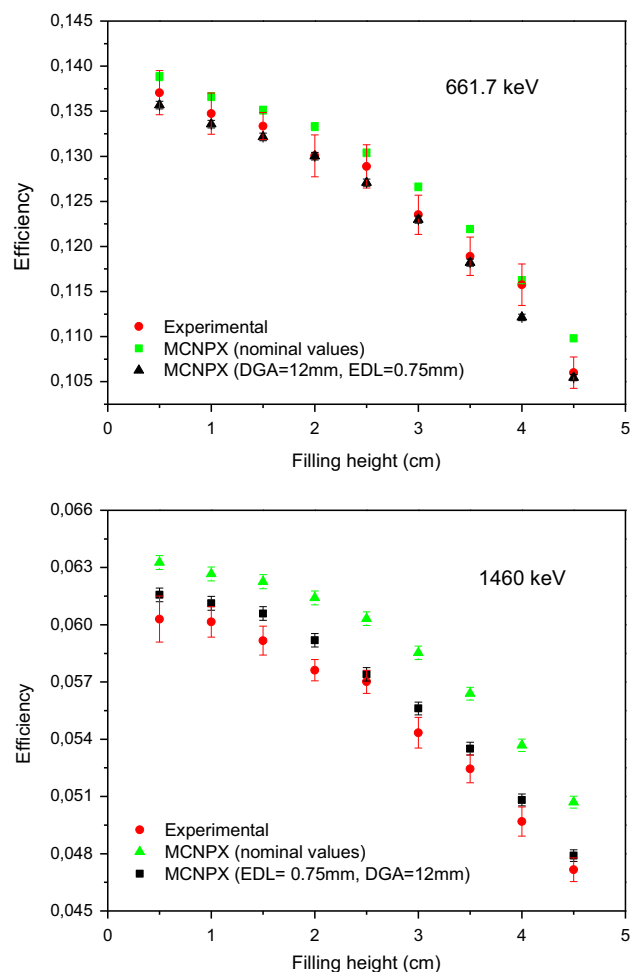


Fig. 1. Calculated and measured efficiencies for different filling heights at 661.7 and 1460 keV in detector D2. The calculations via MCNPX were made for different values of external dead layer (EDL, nominal value=0.5 mm) and the distance between the Ge crystal and the Al end cap (DGA, nominal value=11 mm).

Table 1

Comparison between reported and measured activity concentration (Bq/kg) in several IAEA Reference Materials and high purity KCl. All uncertainties are reported with 95% confidence level.

Isotopes	Reported activity (R)	Measured activity (A)		(A/R-1)100%	
		Detector D1	Detector D2	D1	D2
IAEA-375					
⁴⁰ K	424 ± 8	430 ± 28	417 ± 14	1.4	−1.7
¹³⁷ Cs	5280 ± 80	5224 ± 160	5233 ± 120	1.1	−0.9
²²⁶ Ra	20 ± 2	21.1 ± 2.1	23.3 ± 2.3	14.5	16.5
²³² Th	20.5 ± 1.4	20.7 ± 1.7	19.3 ± 3.6	1.0	−5.9
IAEA-RGU-1					
²¹⁰ Pb	4940 ± 30	4781 ± 200	4980 ± 250	−3.2	0.8
²²⁶ Ra	4940 ± 30	4727 ± 150	5025 ± 160	−4.3	1.7
²³⁸ U	4940 ± 30	4933 ± 170	4895 ± 150	−0.1	−0.9
IAEA-300					
⁴⁰ K	1059 ± 19	1068 ± 29	1085 ± 39	0.8	2.5
¹³⁷ Cs	1067 ± 17	1069 ± 21	1061 ± 26	0.2	−0.6
²¹⁰ Pb	360 ± 20	339 ± 16	359 ± 19	−5.8	−0.3
²³⁸ U	64.7 ± 4.0	− ^a	64.9 ± 6.8	−	0.3
IAEA-306					
²²⁶ Ra	66 ± 10.5	65.7 ± 7.2	69.0 ± 4.8	−0.5	4.5
⁴⁰ K	785 ± 35	773 ± 38	795 ± 46	−1.5	1.3
²³² Th	49 ± 4.5	47.2 ± 5.2	47.6 ± 7.2	−3.7	−2.9
²³⁸ U	77 ± 15	76 ± 14	74 ± 14	−1.8	−3.5
²¹⁰ Pb	435 ± 50	461 ± 34	469 ± 36	6.0	7.8
IAEA-326					
⁴⁰ K	580 ± 56	562 ± 34	557 ± 20	−3.1	−4.0
¹³⁷ Cs	137.5 ± 2.9	136.7 ± 6.4	135.2 ± 3.3	−0.6	−1.7
²²⁶ Ra	32.6 ± 2.4	28.9 ± 2.5	28.7 ± 2.1	−11.3	−11.9
²³² Th	39.4 ± 3.9	37.0 ± 2.8	38.1 ± 2.3	−6.0	−3.4
²¹⁰ Pb	45.7 ± 4.9	47.3 ± 6.0	44.5 ± 5.9	3.5	−2.6
²³⁸ U	29.4 ± 3.4	31.6 ± 4.3	27.7 ± 7.8	7.5	−5.8
IAEA-373					
¹³⁷ Cs	12,350 ± 220	12,190 ± 383	− ^b	−1.3	−
IAEA-444					
¹³⁷ Cs	68.5 ± 1.4	− ^b	69.1 ± 4.4	−	0.9
KCl					
⁴⁰ K	16,358 ± 245 ^c	− ^b	16,129 ± 504	−	−1.4
Mean				3.6	3.5
Standart deviation				3.7	3.9

^a Not detected.

^b Not measured.

^c Calculated activity concentrations considering the elemental weights for KCl, ^{40}K natural abundance and the sample mass.

Winner™ 6.0, the data was fitted to a 4-order polynomial function and they were saved in independent files for each detector and each matrix of interest. Later, these files were used for the corresponding measurements. The differences between the MC calculated efficiency and the fitted efficiency for selected gamma-energies used in the calibration procedure were less than 2% for the entire energy range.

The results for the internal validation of the efficiency calibration curves are given in Table 1. The reported and measured activity concentration show a good agreement for all radionuclides, which are distributed throughout the energy range we studied here. The mean of relative deviation in % between the certified and measured activity concentration are 3.6% and 3.5% for detector D1 and D2 respectively. In all cases the relative deviations are less than 8% except for ^{226}Ra in IAEA-375 and IAEA-326, which may be affected by the summing effect in the sample-detector configuration. It should be noted that no corrections are applied to the gamma-energy lines used for the ^{226}Ra measurement. Also, it is important to note that deviations are more significant for detector D2. In this detector the true coincidence summing effect depends heavily on the measurement geometry and increases when the detection solid angle increases. Nevertheless, in both cases the measured values are found within the combined standard uncertainty of the certified activity concentration. In others CRMs like IAEA-306 and IAEA-RGU-1 there

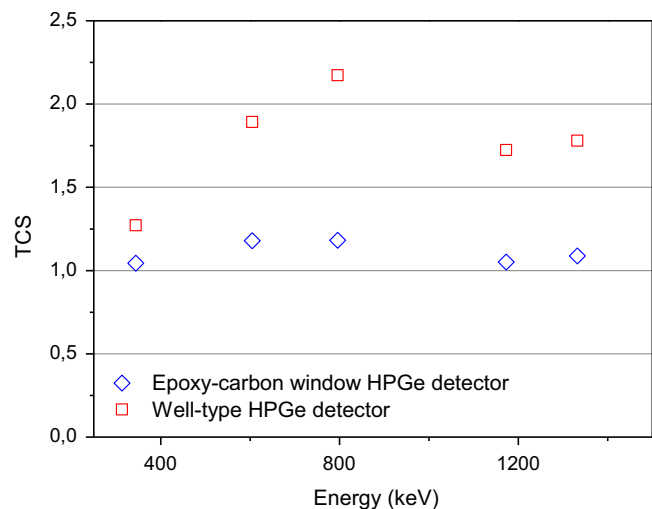


Fig. 2. Calculated coincidence summing correction factors (TCS).

are no apparent coincident summing effects for ^{226}Ra . Similar results are found for ^{232}Th in IAEA-306, IAEA-326 and IAEA-375.

Lower discrepancies can be observed for the single gamma emitters ^{40}K , ^{137}Cs and ^{210}Pb ; and also for ^{238}U , for which several deviations less than 1% are reported. These results are totally satisfactory for environmental sample measurement. Therefore, the calibrations efficiency curves obtained by MC simulations were internally validated for the entire spectra region and for different sample matrices.

3.2. Coincidence summing correction factors

Fig. 2 shows the coincidence summing correction factors TCS calculated for ^{60}Co , ^{134}Cs and ^{152}Eu . It is observed that the TCSs are higher when measurements are performed in the well-type HPGe detector. As it noted above, this is because the sample-detector solid angle is much higher (near 4π geometry of measurement) and increases the probability of coincidence summing occurrence. For detector D1 (with epoxy-carbon window) the solid angle is much lower and correction factors are close to unity for the gamma-energy 344.3 keV (^{152}Eu), 1173.2 and 1332.5 keV (^{60}Co).

Additionally, the correction factors obtained for the 1173.2 and 1332.5 keV (^{60}Co) are very similar to each other for both detectors (3% of relative deviation). This shows that the correction factor for the 1173.2 keV gamma-energy could be used to correct the 1332.5 keV gamma-energy efficiency. This fact has been reported in the literature when experimental and semi-empirical methods were used [20].

3.3. Validation through participation in ALMERA proficiency tests

IAEA proficiency tests and interlaboratory comparison exercises are organized on a regular basis specifically for the members of the ALMERA network. The participation in these exercises is a requirement of the accreditation body for in-house methods [1]. The participation in ALMERA Proficiency Tests IAEA-TEL-2013-04 and IAEA-TEL-2014-04 represent the first times that the MC simulation methods were employed by our laboratory.

The performance of the participant laboratories are evaluated for accuracy and precision according to the IAEA evaluation criterias. The final score according to these criterias will be “Accepted” when both accuracy and precision achieved “Accepted” status, “Non Accepted” when the accuracy is “Non Accepted” and “Warning” when accuracy is “Accepted” but the precision is “Non Accepted”.

Table 2 shows the analytical performance evaluations of the PTs for each radionuclide in the different samples. The reported results for IAEA-TEL-2013-04 were measured in detector D1 while the

Table 2

Analytical performance evaluations of the ALMERA Proficiency Tests IAEA-TEL-2013-04 and IAEA-TEL-2014-04.

Sample code and description	Analyte	Target value (Bq/kg)	Target unc. (Bq/kg)	Reported value (Bq/kg)	Reported Unc. (Bq/kg)	Rel. Bias (%)	MARB (%)	Accuracy	P (%)	Precision	Final score
IAEA-TEL-2013-04											
1-Spiked water	¹³⁴ Cs	24.4	0.21	26.63	0.97	9.14	15	A	3.43	A	A
	¹³⁷ Cs	50.3	0.29	53.2	1.8	5.77	15	A	3.74	A	A
2-Spiked water	²⁴¹ Am	40.2	0.17	39.9	0.91	−2.26	20	A	2.35	A	A
	⁶⁰ Co	50.9	0.42	52.8	2.1	3.73	15	A	4.06	A	A
	¹⁵² Eu	49.9	0.41	51.0	1.7	2.20	15	A	3.43	A	A
4-Spiked flour	¹³⁴ Cs	70.58	0.6	75.1	2.7	6.40	15	A	3.69	A	A
	¹³⁷ Cs	153.95	0.92	163.1	5.4	5.94	15	A	3.36	A	A
IAEA-TEL-2014-04											
1-Spiked water	¹³⁴ Cs	21.4	0.2	22.40	0.67	4.67	20	A	3.13	A	A
	¹³⁷ Cs	12.06	0.1	11.95	0.5	−0.91	20	A	4.27	A	A
2-Spiked water	²⁴¹ Am	16.25	0.2	16.19	0.37	−0.97	20	A	2.60	A	A
	¹⁵² Eu	50.05	0.41	41.4	1.2	−17.28	20	A	3.01	N	W
4-Seaweed	¹³⁴ Cs	8.27	0.2	7.98	0.30	−3.51	25	A	4.47	A	A
	¹³⁷ Cs	22.96	0.45	21.93	0.63	−4.49	20	A	3.48	A	A
5-Sediment	¹³⁷ Cs	12	0.4	11.9	0.25	−0.83	20	A	3.94	A	A
	²²⁶ Ra	37.5	3.2	22.66	0.96	−39.57	25	N	9.53	N	N

results reported in IAEA-TEL-2014-04 were measured in detector D2. In the first PT the final score of “Accepted” was achieved for all radionuclides in all samples. In the second, the final score of “Accepted” was achieved for all radionuclides except for ¹⁵²Eu in spiked water 2 and ²²⁶Ra in water from oil field, with a final score of “Warning” and “Non Accepted”, respectively. Both results were underestimated compared with the target values given by the IAEA in the Laboratory Report, although for ¹⁵²Eu the accuracy was evaluated of “Accepted”. In these cases, the radionuclides are apparently affected by coincidence summing effect, mainly the ²²⁶Ra for which no correction was performed.

In general, most of results showed a satisfactory accuracy and precision evaluation. For all the results evaluated of “Accepted” the reported relative bias were less than 10% and in the entire energy range several relative bias less than 2.5% were reported for both detectors. These external evaluations reaffirm the validation of the efficiency calibration of the two HPGe spectrometric systems used for the gamma measurements in different environmental samples and also for both natural and artificial radionuclides. However, ²²⁶Ra measurement must be improved.

Measurements validation in different HPGe detectors is very important for any laboratory dealing with environmental samples. The routine use of coaxial and well-type HPGe detectors can complement very well the task of measuring samples with different characteristics and containing different radionuclides. Coaxial detectors are very useful to reduce the counting time by increasing the detector efficiency with high sample volumes and short sample to detector distances, while well detectors are essential to reach low minimum detectable activities or statistical uncertainties for small sample quantities. But, as we have seen, it is crucial take into account, mainly for these latest, the true coincidence summing effect to obtain accurate activity results. In order to improve the ²²⁶Ra determination, we recommended measuring the samples containing this radionuclide in the coaxial n-type HPGe detector when no corrections are performed. For the same purpose, experimental and mathematical methods reported

in the literature to correct the coincidence summing effect can also be implemented [21,22].

4. Conclusions

A simple efficiency calibration procedure was implemented for a coaxial n-type and a well-type HPGe detector. For the calibrations were employed experimental and Monte Carlo simulation methods in an energy range from 40 to 1460 keV. This procedure was internally validated through gamma measurements of several IAEA reference materials: IAEA-RGU-1, IAEA-375, IAEA-326, IAEA-444, IAEA-300, IAEA-306 and IAEA-373, and KCl. The reported and measured activity concentration showed a good agreement for all radionuclides, and the mean of relative deviation between the certified and measured activities were 3.6% and 3.5% for coaxial n-type and well-type HPGe detector, respectively. Finally, the method was validated through the participation in two ALMERA Proficiency Tests. The internal and external validations confirm the reliability of the efficiency calibration of the two HPGe detectors for different environmental samples and also for natural and artificial radionuclides. We recommended using coaxial n-type HPGe detector for ²²⁶Ra determination in order to minimize the true coincidence summing effect if no correction is performed. The results reported in this study have contributed to keep the accredited category for the gamma spectrometry test in our laboratory.

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