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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  $^{226}\text{Ra}$  should be conducted using coaxial n-type HPGe detector in order to minimize the true coincidence summing effect.

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

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14 **Abstract**

15 To obtain reliable measurements of the environmental radionuclide activity using HPGe  
16 (High Purity Germanium) detectors, the knowledge of the absolute peak efficiency is  
17 required. This work presents a practical procedure for efficiency calibration of a coaxial n-  
18 type and a well-type HPGe detector using experimental and Monte Carlo simulations  
19 methods. The method was performed in an energy range from 40 to 1460 keV and it can  
20 be used for both, solid and liquid environmental samples. The calibration was initially  
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25 confirmed. The validation also showed that measurement of <sup>226</sup>Ra should be conducted  
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27 effect.

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29 correction; Proficiency test

## 30 **1. Introduction**

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

42 In addition, theoretical and computational methods have also been employed for efficiency  
43 calibration, coincidence-summing and self-absorption corrections. In several studies have  
44 been used general and specific software based on Monte Carlo (MC) codes like Geant,  
45 MCNPX, EFFTRAN or DETEF [4–7]. A good agreement between experimental and  
46 calculated data can be reached using these codes; in addition the working time can be  
47 reduced considerably. In these cases, main limitations reside in the precise knowledge  
48 about the characteristics of the experimental geometry and sample compositions [8].  
49 Generally, the efficiency values obtained experimentally and by MC simulation based on  
50 nominal values of the parameters supplied by the manufacturer show significant  
51 differences due to the inaccuracy in some critical parameter like the thickness of the dead  
52 layer or the active volume. The optimization of these parameters can result in a substantial  
53 decrease of the deviations between the experimental and calculated values [9,10].  
54 However, even when precise geometrical data are available, it is necessary to refine the  
55 model by feeding it back with experimental results when accuracy is desired. This is  
56 because some parameters involved in the detector response cannot easily be assessed.  
57 They include the distribution of the electrical field in the crystal, its mounting and  
58 dimensions and properties of the dead layers [11,12].

59 One effective procedure to overcome these difficulties is to use an efficiency transfer  
60 function from reference geometry to other source configurations, using MC calculations  
61 and experimental measurements. This procedure consists of calculating the full energy  
62 peak efficiency (FEPE) by an energy dependent transfer factor, which is derived by  
63 comparing the direct calculated FEPEs with the source experimental values at a reference  
64 position. In the literature, different authors have reported differences below 5% after using  
65 the transfer function [13,14]; even when there was no adjustment of parameters of the  
66 detectors.

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

## 77 **2. Materials and Methods**

### 78 *2.1. Detectors*

79 Two high purity germanium (HPGe) detectors were used for experimental measurements.  
80 Detector 1 (D1) was a coaxial n-type detector (model NGC 3019, from DSG Detector  
81 Systems GmbH) with epoxy-carbon window and 31.5% of relative efficiency. It was  
82 coupled to an electronic chain, including a multichannel analyzer type TMCA. Detector 2  
83 (D2) was a well-type HPGe detector (model EGPC100 P-15, from Canberra) with an  
84 absolute efficiency of 12.1% at 661 keV. The data acquisition system of this detector  
85 consists of a PSC822 preamplifier, Canberra amplifier model 7245 and electronic card  
86 MCA 5000 which includes a 7602 ADC with 8192 channels and InterFast multichannel  
87 analyzer. Both detectors are surrounded by a cylindrical low-background chamber made  
88 with the following elements from outside to the inner region: 240 mm of steel, 37 mm of  
89 lead, 1 mm of aluminum and 1 mm of copper. In both detectors the gamma spectra were  
90 recorded and analyzed using Winner<sup>TM</sup> 6.0 software. The detectors resolution and energy

91 calibration is periodically verified for stability using a set of point sources ( $^{241}\text{Am}$ ,  $^{137}\text{Cs}$ ,  
92  $^{60}\text{Co}$  and  $^{226}\text{Ra}$ ).

### 93 *2.2. Monte Carlo simulation*

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

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

108 For detector D2 the Monte Carlo efficiency calculations were made directly. In this case, a  
109 tuning of some critical parameters of the detector was made. This tuning showed that the  
110 thickness of external dead layer (EDL) and distance between the Ge crystal and the Al end  
111 cap (DGA) are critical parameters and they were optimized. For the rest of parameters we  
112 used the nominal values.

113 We used the pulse-height tally (F8) per photons emitted from the source to compute the  
114 absolute efficiency and we generally obtained relative errors lower than 1% with a number  
115 of histories about  $10^5$ – $10^6$  and 12-14 minutes of computational times for each energy. This  
116 computational time per energy allows us to build an efficiency calibration curve in  
117 approximately 4 hours. Therefore, the proposed calibration method is good for practical  
118 application in everyday measurements. All MC calculations covered the energy range 40 –  
119 1460 keV.

### 120 *2.3. True coincidence summing corrections*

121 The second step consisted of the determination of the coincidence summing correction  
122 factors (TCSs). For this purpose a simple experimental technique was applied. The  
123 corrections were estimated by measuring of a sample, containing the radionuclides of  
124 interest with summing effect and one single-emitter radionuclide as reference, in two  
125 detector geometries (near and far) [16,17]. The coincidence summing correction factor is  
126 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  
127 radionuclide of reference and  $R_s$  is the count rates of the radionuclide to correct. The ratios  
128  $\left(\frac{R_r}{R_s}\right)_h$  and  $\left(\frac{R_r}{R_s}\right)_g$  are calculated at height  $h$  from the detector where coincidence summing is  
129 negligible and over the detector (indicated by  $g$ ), respectively. This method is simple to  
130 use and it is independent of the sample activity which contribute to minimize several  
131 uncertainties.

132 Water solution of unknown activity with the following radionuclides of interest:  $^{134}\text{Cs}$ ,  $^{152}\text{Eu}$ ,  
133  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ , was used. For the reference we used  $^{137}\text{Cs}$  in the middle energy of the  
134 spectra (661.7 keV). For detector D1 the sample was measured at 10 cm from the end cap  
135 and over the detector while for detector D2 the sample was measurement out of the well  
136 (at 6 cm from the top of the well) and into the well of the detector. In order to calculate the  
137 corrections factors, the TCS expression was applied to the following energy lines:  $^{134}\text{Cs}$   
138 (604.7 and 795.7 keV),  $^{152}\text{Eu}$  (344.3 keV) and  $^{60}\text{Co}$  (1173.2 and 1332.5 keV). It should be  
139 noted that no correction was applied to  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  because the available water  
140 source did not contain these radionuclides. All measurements were carried out for about 6  
141 to 10 hours ensuring a meaningful statistics in each source to detector configuration and in  
142 all cases the dead time corrections were intrinsically taken into account by the software  
143 Winner<sup>TM</sup>.

#### 144 2.4. Experimental verification

145 The experimental verification included the internal validation of MC efficiency curves and  
146 the participation in two ALMERA Proficiency Tests: IAEA-TEL-2013-04 and IAEA-TEL-  
147 2014-04 [18].

148 The validations of the efficiency calibration curves obtained by Monte Carlo simulation was  
149 performed with high purity KCl and several Certified Reference Materials (CRMs) provided  
150 by IAEA [19]: Uranium Ore IAEA-RGU-1, soil IAEA-375, IAEA-326 and IAEA-444, marine

151 sediments IAEA-300 and IAEA-306, and grass IAEA-373. For detector D1 the samples  
152 were encapsulated in a cylindrical container of polystyrene of 1.8 mm thickness with an  
153 internal diameter of 72 mm and filling height of 20 mm. The measurement geometry of  
154 detector D2 was a cylindrical polystyrene vial of 1 mm thickness with an internal diameter  
155 of 10 mm and filling height of 45 mm. Both capsules do not allow radon diffusion. In the  
156 sample IAEA-RGU-1, the radionuclides  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are in secular equilibrium. The  
157 specific activity of  $^{40}\text{K}$  was determined considering the elemental weights for KCl and  $^{40}\text{K}$   
158 natural abundance.

159 The 2013 ALMERA proficiency test (PT) consisted of three water samples and one flour  
160 sample. The participating laboratories were requested to analyze man-made and natural  
161 gamma emitting radionuclides in water samples, and  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in the flour sample.  
162 The subsequently PT (ALMERA 2014) consisted of three water samples, one seaweed  
163 sample, one sediment sample and one water sample from oil field. In this case, the  
164 participating laboratories were requested to analyze anthropogenic and natural gamma-  
165 emitter radionuclides in the water samples, gamma-emitter radionuclides in the seaweed  
166 and sediment samples, and  $^{226}\text{Ra}$  in the water sample from oil field. One water sample  
167 containing known radioisotopes and activity concentrations was provided in each PT for  
168 quality control purpose.

169 The measurements were carried out in the low-background systems described above;  
170 placing the samples over the front end cap or into the well of the detectors D1 and D2,  
171 respectively. For  $^{226}\text{Ra}$  activity measurement the samples were sealed and kept for a  
172 minimum of three weeks, to ensure that secular equilibrium between  $^{226}\text{Ra}$  and radon  
173 daughters had been achieved. The activity of the samples was determined via its  
174 daughters  $^{214}\text{Pb}$  (351.9 and 295.2 keV) and  $^{214}\text{Bi}$  (609.3 keV). The  $^{232}\text{Th}$  activity was  
175 determined from the activity of  $^{212}\text{Pb}$  (238.6 keV),  $^{208}\text{Tl}$  (583.2 keV) and  $^{228}\text{Ac}$  (911.2 keV).  
176 Two gamma-energies were also analyzed for  $^{134}\text{Cs}$  (604.7 and 795.7 keV) and  $^{60}\text{Co}$   
177 (1173.2 and 1332.5 keV). In all cases the activity concentration was reported as the  
178 arithmetic mean.  $^{238}\text{U}$  activity was determined from the activity of  $^{234}\text{Th}$  (63.3 keV) and only  
179 one gamma-energy was used for  $^{152}\text{Eu}$  (344.3 keV). Finally, to determine  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$   
180 and  $^{210}\text{Pb}$  activities, the well-known 1460.7, 661.7, 59.54 and 46.54 keV gamma-energies  
181 were used, respectively. Activities of  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$  and  $^{152}\text{Eu}$  were corrected applying the  
182 calculated TCSs coefficients. Also, decay-corrections were applied to reference materials

183 and samples provided by ALMERA PTs with the reference time given in their Certificates  
184 or with the one requested by the PT instructions.

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

194 The use of Monte Carlo codes makes necessary to have a detailed description of the  
195 geometric intrinsic characteristics of the detector and geometry characteristics of the  
196 sample container, which were described above. But also, the sample chemical  
197 composition was needed. The major elemental composition of CRMs used in this study is  
198 known and it is available in their respective reports [19]. For sediment we used the  
199 chemical composition of the IAEA-SL-3 reference material (lake sediment from Austria)  
200 [19]. For flour we used the following composition: C (40%), O (40%) and H (20%) and for  
201 seaweed C (42%), O (50%), H (5%) and N (3%). Finally, for KCl and H<sub>2</sub>O their elemental  
202 weights were considered. Once each sample of interest was directly implemented for MC  
203 calculations the self-absorption corrections were not necessary.

### 204 **3. Results and discussions**

#### 205 *3.1. Efficiency calibration and internal verification*

206 The efficiency response of detector D1 and D2 was reported in previous researches.  
207 Monte Carlo efficiency transfer method was applied for D1 using directly the nominal  
208 parameters of the detector [6]. The method was conducted in the 40 to 1408 keV energy  
209 range and was obtained a good agreement between measurement and calculated values  
210 in three reference materials: DL-1a, IAEA-375 and IAEA-RG-1. In the present study, the  
211 efficiency transfer calculations were extended to other IAEA reference materials,  
212 environmental samples and liquid samples.



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

227 The efficiencies calculated in the entire energy range (40 to 1460 keV) were loaded to the  
228 software for gamma-ray spectrometry Winner<sup>TM</sup> 6.0, the data was fitted to a 4-order  
229 polynomial function and they were saved in independent files for each detector and each  
230 matrix of interest. Later, these files were used for the corresponding measurements. The  
231 differences between the MC calculated efficiency and the fitted efficiency for selected  
232 gamma-energies used in the calibration procedure were less than 2% for the entire energy  
233 range.

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

246 activity concentration. In others CRMs like IAEA-306 and IAEA-RGU-1 there are no  
247 apparent coincident summing effects for  $^{226}\text{Ra}$ . Similar results are found for  $^{232}\text{Th}$  in IAEA-  
248 306, IAEA-326 and IAEA-375.

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

### 254 3.2. Coincidence summing correction factors

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

262 Additionally, the correction factors obtained for the 1173.2 and 1332.5 keV ( $^{60}\text{Co}$ ) are very  
263 similar to each other for both detectors (3% of relative deviation). This shows that the  
264 correction factor for the 1173.2 keV gamma-energy could be used to correct the  
265 1332.5 keV gamma-energy efficiency. This fact has been reported in the literature when  
266 experimental and semi-empirical methods were used [20].

### 267 3.3. Validation through participation in ALMERA proficiency tests

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

273 The performance of the participant laboratories are evaluated for accuracy and precision  
274 according to the IAEA evaluation criterias. The final score according to these criterias will  
275 be "Accepted" when both accuracy and precision achieved "Accepted" status, "Non

276 Accepted” when the accuracy is “Non Accepted” and “Warning” when accuracy is  
277 “Accepted” but the precision is “Non Accepted”.

278 Table 2 shows the analytical performance evaluations of the PTs for each radionuclide in  
279 the different samples. The reported results for IAEA-TEL-2013-04 were measured in  
280 detector D1 while the results reported in IAEA-TEL-2014-04 were measured in detector  
281 D2. In the first PT the final score of “Accepted” was achieved for all radionuclides in all  
282 samples. In the second, the final score of “Accepted” was achieved for all radionuclides  
283 except for  $^{152}\text{Eu}$  in spiked water 2 and  $^{226}\text{Ra}$  in water from oil field, with a final score of  
284 “Warning” and “Non Accepted”, respectively. Both results were underestimated compared  
285 with the target values given by the IAEA in the Laboratory Report, although for  $^{152}\text{Eu}$  the  
286 accuracy was evaluated of “Accepted”. In these cases, the radionuclides are apparently  
287 affected by coincidence summing effect, mainly the  $^{226}\text{Ra}$  for which no correction was  
288 performed.

289 In general, most of results showed a satisfactory accuracy and precision evaluation. For all  
290 the results evaluated of “Accepted” the reported relative bias were less than 10% and in  
291 the entire energy range several relative bias less than 2.5% were reported for both  
292 detectors. These external evaluations reaffirm the validation of the efficiency calibration of  
293 the tow HPGe spectrometric systems used for the gamma measurements in different  
294 environmental samples and also for both natural and artificial radionuclides. However,  
295  $^{226}\text{Ra}$  measurement must be improved.

296 Measurements validation in different HPGe detectors is very important for any laboratory  
297 dealing with environmental samples. The routine use of coaxial and well-type HPGe  
298 detectors can complement very well the task of measuring samples with different  
299 characteristics and containing different radionuclides. Coaxial detectors are very useful to  
300 reduce the counting time by increasing the detector efficiency with high sample volumes  
301 and short sample to detector distances, while well detectors are essential to reach low  
302 minimum detectable activities or statistical uncertainties for small sample quantities. But,  
303 as we have seen, it is crucial take into account, mainly for these latest, the true  
304 coincidence summing effect to obtain accurate activity results. In order to improve the  
305  $^{226}\text{Ra}$  determination, we recommended measuring the samples containing this  
306 radionuclide in the coaxial n-type HPGe detector when no corrections are performed. For

307 the same purpose, experimental and mathematical methods reported in the literature to  
308 correct the coincidence summing effect can also be implemented [21,22].

#### 309 **4. Conclusions**

310 A simple efficiency calibration procedure was implemented for a coaxial n-type and a well-  
311 type HPGe detector. For the calibrations were employed experimental and Monte Carlo  
312 simulation methods in an energy range from 40 to 1460 keV. This procedure was internally  
313 validated through gamma measurements of several IAEA reference materials: IAEA-RGU-  
314 1, IAEA-375, IAEA-326, IAEA-444, IAEA-300, IAEA-306 and IAEA-373, and KCl. The  
315 reported and measured activity concentration showed a good agreement for all  
316 radionuclides, and the mean of relative deviation between the certified and measured  
317 activities were 3.6 and 3.5 % for coaxial n-type and well-type HPGe detector, respectively.  
318 Finally, the method was validated through the participation in two ALMERA Proficiency  
319 Tests. The internal and external validations confirm the reliability of the efficiency  
320 calibration of the two HPGe detectors for different environmental samples and also for  
321 natural and artificial radionuclides. We recommended using coaxial n-type HPGe detector  
322 for  $^{226}\text{Ra}$  determination in order to minimize the true coincidence summing effect if no  
323 correction is performed. The results reported in this study have contributed to keep the  
324 accredited category for the gamma spectrometry test in our laboratory.

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Figure 1

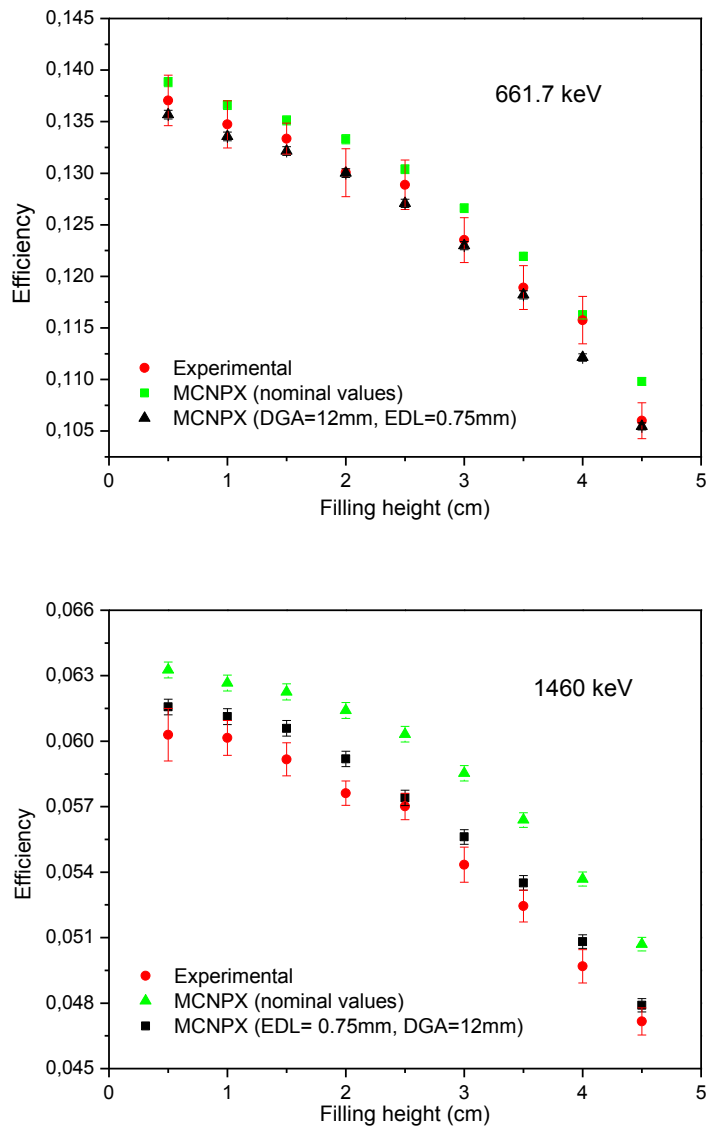


Fig. 1. Calculated and measured efficiencies for different filling heights at 661.7 and 1460 keV in detector D2. The calculations via MCNPX were made for different values of external dead layer (EDL, nominal value= 0.5mm) and the distance between the Ge crystal and the Al end cap (DGA, nominal value=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

Figure 2

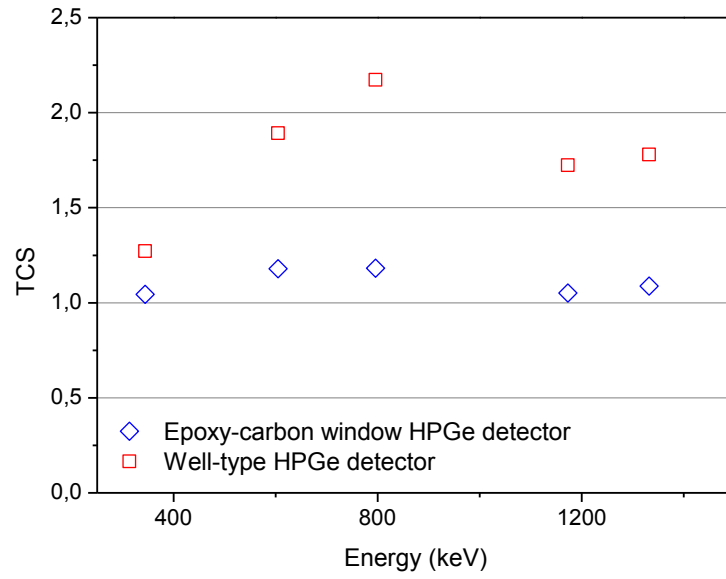


Fig. 2. Calculated coincidence summing correction factors (TCS).



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
<b>IAEA-375</b>					
<sup>40</sup> K	424 ± 8	430 ± 28	417 ± 14	1.4	-1.7
<sup>137</sup> Cs	5280 ± 80	5224 ± 160	5233 ± 120	1.1	-0.9
<sup>226</sup> Ra	20 ± 2	21.1 ± 2.1	23.3 ± 2.3	14.5	16.5
<sup>232</sup> Th	20.5 ± 1.4	20.7 ± 1.7	19.3 ± 3.6	1.0	-5.9
<b>IAEA-RGU-1</b>					
<sup>210</sup> Pb	4940 ± 30	4781 ± 200	4980 ± 250	-3.2	0.8
<sup>226</sup> Ra	4940 ± 30	4727 ± 150	5025 ± 160	-4.3	1.7
<sup>238</sup> U	4940 ± 30	4933 ± 170	4895 ± 150	-0.1	-0.9
<b>IAEA-300</b>					
<sup>40</sup> K	1059 ± 19	1068 ± 29	1085 ± 39	0.8	2.5
<sup>137</sup> Cs	1067 ± 17	1069 ± 21	1061 ± 26	0.2	-0.6
<sup>210</sup> Pb	360 ± 20	339 ± 16	359 ± 19	-5.8	-0.3
<sup>238</sup> U	64.7 ± 4.0	- <sup>a</sup>	64.9 ± 6.8	-	0.3
<b>IAEA-306</b>					
<sup>226</sup> Ra	66 ± 10.5	65.7 ± 7.2	69.0 ± 4.8	-0.5	4.5
<sup>40</sup> K	785 ± 35	773 ± 38	795 ± 46	-1.5	1.3
<sup>232</sup> Th	49 ± 4.5	47.2 ± 5.2	47.6 ± 7.2	-3.7	-2.9
<sup>238</sup> U	77 ± 15	76 ± 14	74 ± 14	-1.8	-3.5
<sup>210</sup> Pb	435 ± 50	461 ± 34	469 ± 36	6.0	7.8
<b>IAEA-326</b>					
<sup>40</sup> K	580 ± 56	562 ± 34	557 ± 20	-3.1	-4.0
<sup>137</sup> Cs	137.5 ± 2.9	136.7 ± 6.4	135.2 ± 3.3	-0.6	-1.7
<sup>226</sup> Ra	32.6 ± 2.4	28.9 ± 2.5	28.7 ± 2.1	-11.3	-11.9
<sup>232</sup> Th	39.4 ± 3.9	37.0 ± 2.8	38.1 ± 2.3	-6.0	-3.4
<sup>210</sup> Pb	45.7 ± 4.9	47.3 ± 6.0	44.5 ± 5.9	3.5	-2.6
<sup>238</sup> U	29.4 ± 3.4	31.6 ± 4.3	27.7 ± 7.8	7.5	-5.8
<b>IAEA-373</b>					
<sup>137</sup> Cs	12350 ± 220	12190 ± 383	- <sup>b</sup>	-1.3	-
<b>IAEA-444</b>					
<sup>137</sup> Cs	68.5 ± 1.4	- <sup>b</sup>	69.1 ± 4.4	-	0.9
<b>KCl</b>					
<sup>40</sup> K	16358 ± 245 <sup>c</sup>	- <sup>b</sup>	16129 ± 504	-	-1.4
<b>Mean</b>				3.6	3.5
<b>Standart deviation</b>				3.7	3.9

<sup>a</sup>Not detected

<sup>b</sup>Not measured

<sup>c</sup>Calculated activity concentrations considering the elemental weights for KCl, <sup>40</sup>K natural abundance and the sample mass

Table 2

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	<sup>134</sup> Cs	24.4	0.21	26.63	0.97	9.14	15	A	3.43	A	A
	<sup>137</sup> Cs	50.3	0.29	53.2	1.8	5.77	15	A	3.74	A	A
2-Spiked water	<sup>241</sup> Am	40.2	0.17	39.9	0.91	-2.26	20	A	2.35	A	A
	<sup>60</sup> Co	50.9	0.42	52.8	2.1	3.73	15	A	4.06	A	A
	<sup>152</sup> Eu	49.9	0.41	51.0	1.7	2.20	15	A	3.43	A	A
4-Spiked flour	<sup>134</sup> Cs	70.58	0.6	75.1	2.7	6.40	15	A	3.69	A	A
	<sup>137</sup> Cs	153.95	0.92	163.1	5.4	5.94	15	A	3.36	A	A
IAEA-TEL-2014-04											
1-Spiked water	<sup>134</sup> Cs	21.4	0.2	22.40	0.67	4.67	20	A	3.13	A	A
	<sup>137</sup> Cs	12.06	0.1	11.95	0.5	-0.91	20	A	4.27	A	A
2-Spiked water	<sup>241</sup> Am	16.25	0.2	16.19	0.37	-0.97	20	A	2.60	A	A
	<sup>152</sup> Eu	50.05	0.41	41.4	1.2	-17.28	20	A	3.01	N	W
4-Seaweed	<sup>134</sup> Cs	8.27	0.2	7.98	0.30	-3.51	25	A	4.47	A	A
	<sup>137</sup> Cs	22.96	0.45	21.93	0.63	-4.49	20	A	3.48	A	A
5-Sediment	<sup>137</sup> Cs	12	0.4	11.9	0.25	-0.83	20	A	3.94	A	A
6-Water from oil field	<sup>226</sup> Ra	37.5	3.2	22.66	0.96	-39.57	25	N	9.53	N	N

## Highlights

- An efficiency calibration for a coaxial and a well-type HPGe detector was performed
- The calibration was made using experimental and Monte Carlo simulations methods
- The procedure was verified measuring several reference materials provided by IAEA
- Calibrations were validated through the participation in 2 ALMERA Proficiency Tests