

## Determination of the Volume Efficiency of Hyper Pure Germanium (HPGe) Detector Using Ra- 226 Point Source

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### ABSTRACT

In the present work, a previous mathematical method to determine the volume efficiency of Hyper Pure Germanium (HPGe) detector for cylindrical shape sources at different distances is introduced and the volume efficiency is calculated using the KCl method covering the energy range from 186 to 2614 keV. Correction factors  $F_1$  are suggested for KCl solution and  $F_2$  due to the presence of medium between the sample and the detector. To validate these results eighteen soil samples were collected from Ras Shakier, Egypt. The samples were filled in 100 ml polyethylene containers, weighed, sealed and stored for about one month in order to allow radioactive equilibrium to be reached (secular equilibrium). This step ensured that radon gas and its daughters remain in the samples. The samples were measured at different distances. The activities were determined using volume efficiency from the mathematical method and the KCl method using two Ra-226 point sources. Two samples were also measured in Environmental Pollution Lab at the Egyptian Atomic Energy Authority using the standard calibration source of U-238 RGU-1 and Th-232 RGTh-1. The specific activity results were compared with the mathematical method calculation to confirm the mathematical method.

**Key words:** Volume efficiency, Ra-226 source, Hyper Pure Germanium (HPGe), Soil samples

### Introduction

Gamma –ray spectrometry is one of the most widely used techniques to determine the concentrations of natural and artificial radionuclides in environmental samples. Its major advantages are being non-destructive, used for multi-element analysis, and simplified regarding sample preparation, i.e. mostly no need for any radiochemical separation processes (Jurado *et al.*, 2002; Khater and Ebaid 2008). In the case of low-level radio environmental measurements at close geometry, much care has to be dedicated to the efficiency calibration of the spectrometer used because the full-energy peak efficiency is a complicated function of many parameters, i.e. gamma-ray energy, the detector dimensions, the source dimensions, self absorption, detector and source solid angle, as well as sample density.

The efficiency calibration of HPGe detectors for volume source geometry has been discussed by many authors (Mahmoud, (2001 and 2007); Julio *et al.*, (2006); Gharbi, (2011)).

Mahmoud (2001 and 2007) used direct mathematical expressions to calculate the photo peak efficiency, the coincidence correction and the source self-absorption for the cases of closed end HPGe detector and Marinelli beaker sources. In 2007 he calculated the full-energy peak efficiency obtained by an analytical formula and then compared it with the experimental results (after performing the coincidence corrections), for extended sources. Julio *et al.*, (2006) derived a theoretical method of obtaining the volumetric efficiency of a cylindrical homogeneous source by multiplying the point–source efficiency by a geometric and an attenuation factor. This method has been validated experimentally. Jackman and Biegalski (2009) presented a review article of different methods and software for predicting Germanium detector absolute full-energy peak efficiencies, using the relative method, the Mont Carlo method and the semi-empirical method.

Gharbi, (2011) explores with the inhomogeneity effects of the source on the full energy peak efficiency determination with HPGe gamma spectrometry. Nie *et al.*, (2012) proposed an empirical formula for the HPGe efficiency calibration of bulky sources in cylindrical geometry at different source-to-detector distances.

As mentioned before an accurate determination of the absolute efficiency for  $\gamma$ - spectrometers is of great importance especially in low-level measurements of environmental samples. Therefore the aim of the present work is studying the efficiency for a HPGe detector for volumetric samples using a mathematical method which depends on the experimental absolute efficiency using a point source Ra-226 at different source to detector distances.

To validate these results the activity concentrations of 18 soil samples will be estimated by the efficiencies using the mathematical method and also those from the efficiency curves obtained using KCl to normalize a relative and an absolute efficiency to absolute volume efficiency curves. In addition, the results of two samples studied using standard materials.

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## Theoretical treatment and experimental technique

### Theoretical Treatment

The HPGe detector can be considered as a point detector since the energy of the photons is absorbed after they have penetrated a certain distance into the crystal. Based on this idea, some investigators have proposed the concept of a point – detector position (Mahmoud (2001); Longoria and Benitez, 1997)

In the present work, the volume efficiency for the HPGe detector was determined using the mathematical method. That method starts from the point detector efficiency equation (Klaus and Richard, 1988):

$$\epsilon_p(\mathbf{r}, \mathbf{d}) = \frac{\epsilon_p(0, \mathbf{d}) * \mathbf{d}^2}{(\mathbf{r}^2 + \mathbf{d}^2)} \quad (1)$$

$$\mathbf{d} = \mathbf{d}_s + \mathbf{d}_c$$

Where  $d_s$  the distance from sample to the surface of the detector,

$d_c$  represent detector surface to detector end cap and  $\epsilon_p(0, d)$  is the absolute efficiency for point source at vertical line from effective point.

In case of an area source with radius  $R_s$  and with uniformly distributed activity; the absolute efficiency  $\epsilon_a(d)$  is obtained from the integration (Klaus and Richard, 1988):

$$\epsilon_a(\mathbf{d}) = \frac{2}{R_s^2} \int_0^{R_s} \epsilon_p(\mathbf{r}, \mathbf{d}) * \mathbf{r} * \mathbf{d}\mathbf{r} \quad (2)$$

Where the effect of self attenuation was neglected, the absolute efficiency volume is calculated from the following equation:

$$\epsilon_v(\mathbf{d}) = \frac{1}{h} \int_d^{d+h} \epsilon_a(\mathbf{x}) \mathbf{d}\mathbf{x} = \frac{2}{R_s^2} * \frac{1}{h} \int_d^{d+h} \int_0^{R_s} \epsilon_p(\mathbf{r}, \mathbf{x}) \mathbf{r} \mathbf{d}\mathbf{r} \mathbf{d}\mathbf{x} \quad (3)$$

Where:  $h$  is the sample height. The sample height was calculated from the equation:

$$V = \pi r^2 h$$

Where:  $V$  is the sample volume. Maple 12 program is used to solve equ. 3 at  $d_s = 3, 5, 7$  and  $9$  cm. The best advantages of putting the source at a large distance from the detector are smaller coincidence – summing corrections and less sensitivity of the detection efficiency to small changes in source shape and positioning.

### Experimental Technique

In this work, an ORTEC Hyper Pure Germanium gamma ray spectrometer was used. The HPGe crystal has a diameter of 49.3 mm and a length of 47.1 mm, the end cap to crystal distance is 3 mm, and the absorbing beryllium layer is 0.5 mm. The detector is coupled to an ORTEC model 572A spectroscopy amplifier and an IBM compatible PC with a MCA card (Maestro-32) mounted in it. The resolution is 1.9 keV at the 1332.5 keV gamma transition of Co-60. To reduce the background a lead cylinder together with a concentric copper cylinder, with a movable cover, shielded the detector.

### Experimental work

The mathematical method starts from the experimental absolute efficiency for a point source Ra-226 with activity 0.1  $\mu\text{Ci}$ . The point source was located on the detector axis on contact with detector surface. The experimental absolute efficiency was determined for an energy range from 186.1 to 2450 keV. Also, the experimental absolute efficiency was determined at different distances (3, 5, 7 and 9 cm) between the detector and the Ra-226 source (air medium) to study the effect of the distance on mathematical method results.

The efficiency calibration of the HPGe detector, using the KCl solution has been done through two stages. In the first stage the relative photo peak efficiency curve of the detector was obtained using Ra-226 point source (unknown activity). In the second stage, the relative efficiency curve was normalized to an absolute volume efficiency curve using KCl solution (EL-Tahawy *et al.*, 1992).

To validate these results eighteen soil samples were collected from Ras Shakier, Egypt. The samples were filled in 100 ml polyethylene containers, weighed, sealed and stored for about one month in order to allow radioactive equilibrium to be reached (secular equilibrium). This step ensured that radon gas and its daughters remain in the samples. The samples were measured at different distances. The activities were determined using volume efficiency from the mathematical method and the KCl method using two Ra-226 point sources.

The activities of two soil samples (7, 17) were measured in Environmental Pollution Lab in Egyptian Atomic Energy Authority (EAEA) for soil samples. Standard calibration sources (powder) of U-238 (RGU-1) and Th-232 (RGTh-1) were poured in a similar plastic container up to the same volume as the samples and the results were compared with the activities using the mathematical method.

### Results of volume efficiency from mathematical method

The absolute efficiency for the Ra-226 point source (0.1  $\mu\text{Ci}$ ) is determined for gamma energies 186.1, 241.9, 295.1, 352, 609.3, 768.4, 934, 1120.3, 1238.1, 1729.6, 1764.5, 2204.1 and 2447.7 keV, at different distances as shown in Fig. (1).

Fig (2) shows the volume efficiency which determined from eq (3) at different distances for each gamma energy.

Equation (4) represents the the fitting equation by using the ORIGIN 6.1 software program for different gamma energies

$$Y = \varepsilon_p(0, d) = y_0 + A_1 e^{-x/t_1} \quad (4)$$

Where  $y_0$ ,  $A_1$  and  $t_1$  are the fitting parameter .

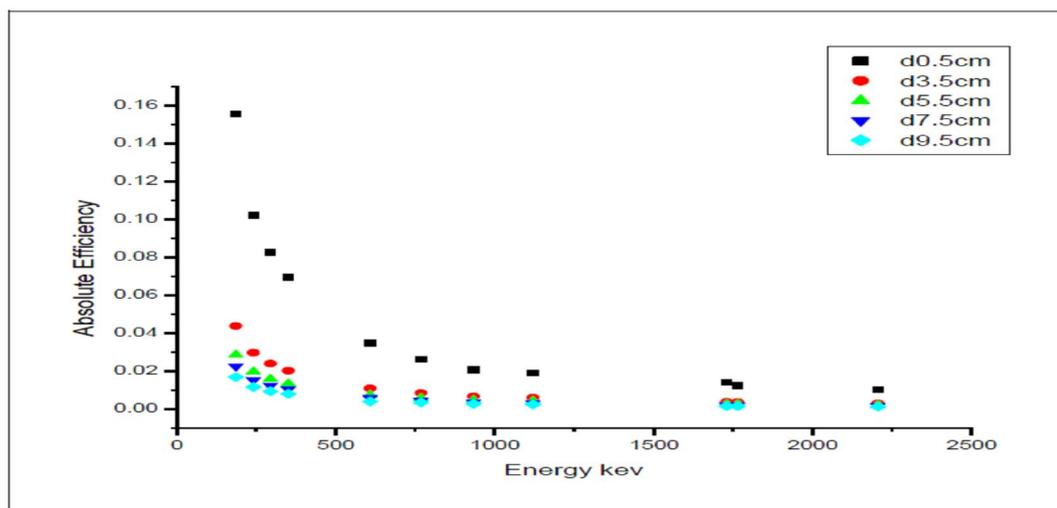


Fig.1. Absolute efficiency at all source- detector distances for the Ra-226 source (0.1 $\mu\text{Ci}$ )

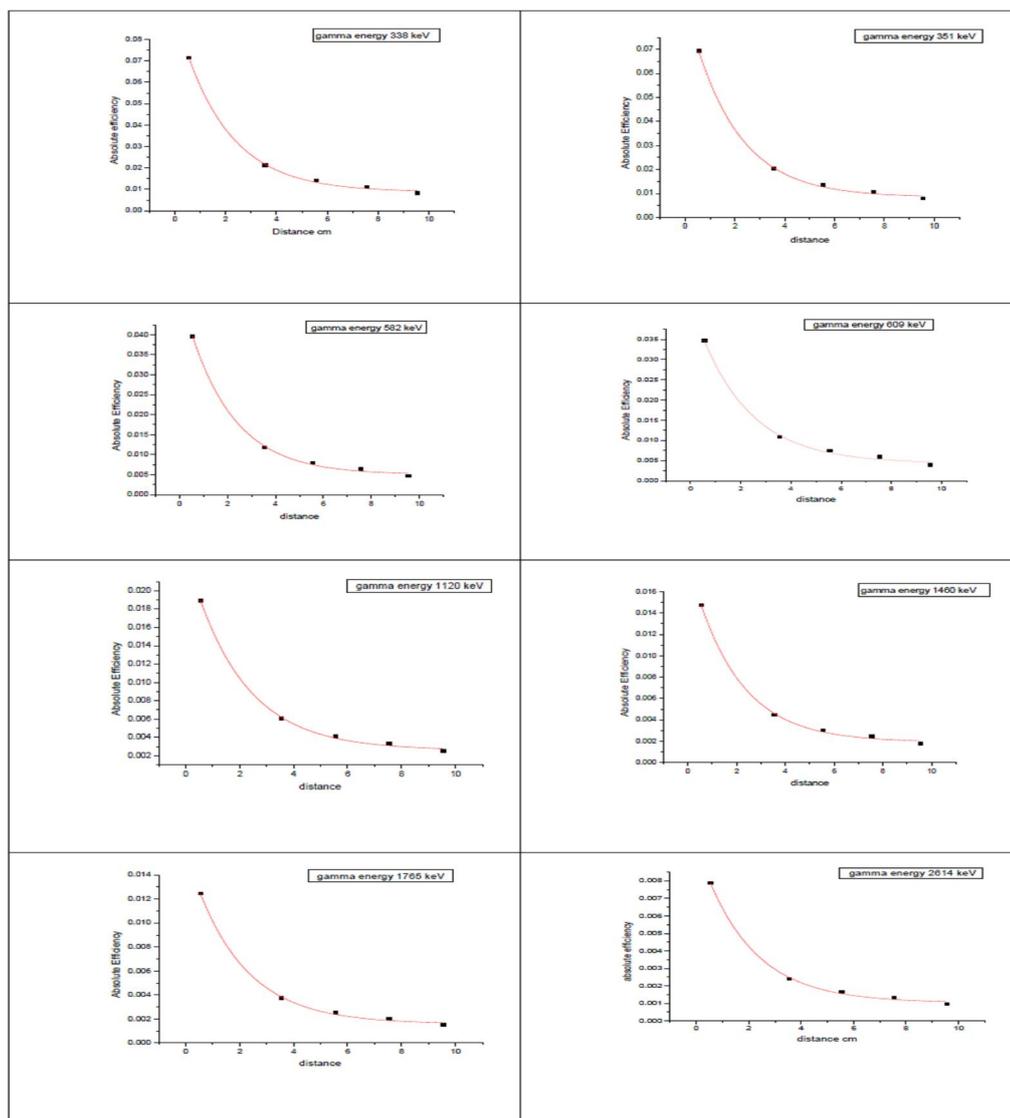


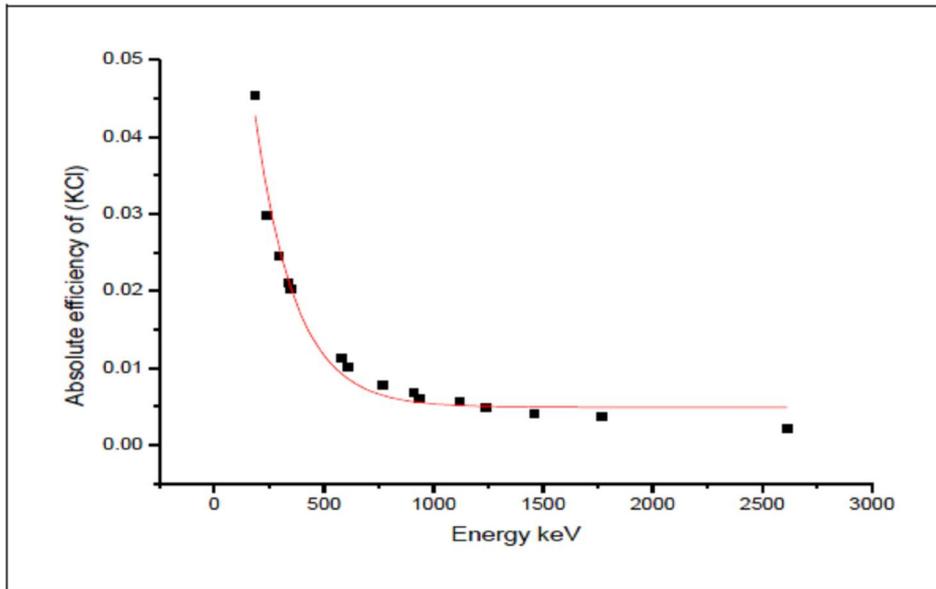
Fig. (2): The relation between the absolute efficiency and distance for different gamma energies.

#### Validation to Mathematical Method

The relative efficiency curve of the detector was normalized to an absolute efficiency curve using the KCl method. This was used to normalize a relative efficiency curve obtained from a Ra-226 point source (unknown activity) to absolute volume efficiency curve as shown in Fig.(3). There was a difference between the volume efficiency using the KCl method and the mathematical equation (which contain the height of the used samples). We suggested that the difference may due to the density difference and therefore a correction was made. The correction factor  $F_1$  is expressed as:

$$F_1 = \frac{\rho_{KCl}}{\rho_{sample}} \quad (5)$$

Where:  $\rho_{KCl}$  and  $\rho_{sample}$  are the densities of KCl and the sample, respectively.



**Fig. (3).The absolute efficiency normalized by KCl (100ml) & energy.**

For validation of the Mathematical Method, the activity concentrations of 18 soil samples were determined using the efficiencies of the two methods and the results were compared. Table (2) shows the results of specific activities for different soil samples (after correction) on contact with the surface of the detector. From these results, it is noticed that the deviation between the two methods ranges between 1 –15 % but reached 25 % in 3 soil samples.

The histograms in Fig. (4) shows activity concentration for all samples using the two methods on contact with the surface of the detector. Sample six has maximum activity for U-238 and Th-232. The differences between the two methods are referred to the presence of other errors like random errors, self-absorption and systematic errors.

Table (3) shows the results of activity concentrations for the samples (1, 6, and 17) at different distances. It was found that there is no agreement between the results at different distances. These differences may be due to the soil density differences, also due to the presence of medium (air volume) between the sample and the detector where the volume of the air increases as the distance increases, therefore we proposed a correction factor  $F_2$  as:

$$F_2 = \frac{\rho_{total}}{\rho_{sample}} = \frac{\rho_{air} + \rho_{sample}}{\rho_{sample}} \quad (6)$$

Where  $\rho_{sample}$  is the sample density and  $\rho_{air}$  is air density.

Table (4) shows the activities after using the proposed correction factor  $F_2$ . It was found that the deviation ranged between 3 – 15 %.

Also to confirm the mathematical method by using the standard calibration source at

The activity concentrations for samples (EAEA) using standard calibration sources of U-238 RGU-1 and Th-232 RGTh-1 to verify the mathematical method. The results are shown in Table (5).

Also to confirm the mathematical method by using the standard calibration source of U-238 RGU-1 and Th-232 RGTh-1. Two samples 7, 17 were also measured in Environmental Pollution Lab at the Egyptian Atomic Energy Authority. The specific activity were compared with the mathematical methods calculation as shown in Table (5).

**Table 1.** The fitting parameters for all gamma energies.

Energy keV	Fitting parameters		
	$Y_0$	$A_1$	$t_1$
186	0.01803	0.18543	1.83458
338	0.00878	0.08361	1.89814
351	0.00847	0.08152	1.87693
582	0.005	0.04619	1.89811
609	0.00432	0.03982	2.03374
911	0.00312	0.02847	1.91459
1120	0.00261	0.02157	1.97706
1460	0.00191	0.01709	1.91505
1765	0.0016	0.01448	1.90392
2614	0.00104	0.00908	1.92863

**Table 2.** Comparison of specific activities for soil samples on contact with the surface of the detector after correction with  $F_1$ .

Sample No	Mean Activity Concentrations in (Bqkg <sup>-1</sup> ) by Mathematical Method			Mean Activity Concentrations in (Bqkg <sup>-1</sup> ) by KCl Method* $F_1$		
	238U	232Th	40K	238U	232Th	40K
1	147.69±3.79	46.53±2.75	787.54± 14.70	149.91±5.78	48.08±3.03	841.72±16.18
2	29.50±1.95	29.88±2.39	737.11±12.43	35.71±2.65	34.76±3.05	890.90±15.02
3	22.08±1.65	21.35±1.96	540.27±9.94	27.44±2.38	25.92±2.61	687.05±12.64
4	35.42±2.37	36.72±2.65	790.70±15.13	44.97±3.46	48.70±3.72	1016.14±19.44
5	30.46±1.94	32.55±2.03	800.06±12.59	40.81±2.89	42.39±2.72	1022.54±16.10
6	474.42±10.93	95.27±5.79	755.90±17.93	433.15± 10.32	85.72±4.72	634.46±13.99
7	50.13±2.46	37.60±2.15	859.55±14.34	65.31±3.27	44.98±2.78	1059.21±17.67
8	28.95±2.12	29.53±2.17	906.20±13.22	32.8±2.66	36.08±2.87	1113.67±16.24
9	55.97±3.98	55.95±4.03	1318.81±22.89	49.02±3.22	42.05±2.88	936.56±16.25
10	16.00±1.59	17.58±1.85	1094.58±15.60	19.67±2.02	20.16±2.28	1220.16±17.39
11	61.95±4.60	54.01±5.20	1443.23±30.32	54.27±3.65	43.82±3.35	1029.07±21.54
12	41.23±2.58	44.17±2.46	1075.97±17.66	45.07±2.88	44.00±2.89	1060.38±17.41
13	25.61±1.87	22.91±1.89	601.89±12.04	30.81±2.72	28.02±2.56	777.28±15.55
14	21.34±1.80	32.31±2.44	1006.26±16.54	23.80±2.03	33.34±2.68	1005.92±16.53
15	20.57±1.84	24.22±2.63	1047.07±15.50	22.11±2.34	28.10±3.15	1167.86±17.29
16	22.64±2.08	17.35±1.75	1087.31±15.44	27.89±2.66	20.68±2.07	1207.08±17.15
17	28.41±2.02	29.95±2.34	906.03±14.79	32.85±2.51	31.81±2.44	944.93±15.91
18	37.12±3.18	28.56±2.78	658.72±14.48	34.30±2.35	24.11±2.34	525.39±11.55

**Table 3.** Comparison of specific activities for soil samples at different distances by Mathematical Method and KCl method

Sample No.	Average value	Activity Concentrations in (Bqkg <sup>-1</sup> ) by Mathematical Method					Activity Concentrations in (Bqkg <sup>-1</sup> ) by KCl Method*factor" $F_1$ "
		0.5 cm	3.5 cm	5.5 cm	7.5 cm	9.5 cm	
1	<sup>238</sup> U	147.69±3.79	63.14±3.08	47.69±2.75	36.26±2.04	25.16±1.97	149.91±5.78
	<sup>232</sup> Th	46.53±2.75	17.25±1.38	8.56±1.01	7.13±0.95	7.55±0.69	48.08±3.03
	<sup>40</sup> K	787.54± 14.70	336.46±11.69	222.41±10.08	187.56±6.53	174.72±7.54	841.72±16.18
6	<sup>238</sup> U	474.42±8.34	227.18±7.59	175.23±6.47	137.83±7.34	105.73±6.52	433.15±7.61
	<sup>232</sup> Th	95.27±4.08	40.67±3.86	31.75±3.10	21.86±2.70	16.47±2.53	85.72±3.55
	<sup>40</sup> K	755.90±16.67	371.57±11.91	269.22±9.94	168.47±6.86	121.43±8.09	634.46±13.99
17	<sup>238</sup> U	28.41±2.13	12.50±1.13	14.21±1.61	6.91±0.86	4.72±0.56	32.85±2.51
	<sup>232</sup> Th	29.95±2.27	11.38±1.36	9.09±1.29	7.62±1.46	5.13±0.97	31.81±2.44
	<sup>40</sup> K	906.03±15.25	389.74±10.28	295.78±9.86	219.82±6.70	194.50±7.49	944.93±15.91

**Table 4.** Specific activities for soil samples at different distances after corrections ( $F_2$ ).

Sample No.	Average value	Activity Concentrations in (Bqkg <sup>-1</sup> ) by Mathematical Method* correction factor ( $F_2$ )					Activity Concentrations in (Bqkg <sup>-1</sup> ) by KCl Method*Factor" $F_1$ "
		0.5cm	3.5 cm	5.5 cm	7.5 cm	9.5 cm	
1	<sup>238</sup> U	147.69±3.79	150.33±7.25	153.83±8.96	151.08±8.51	132.42±9.98	149.91±5.78
	<sup>232</sup> Th	46.53±2.75	41.07±3.27	27.6±3.30	29.70±3.95	39.73±3.50	48.08±3.03
	<sup>40</sup> K	787.54± 14.70	801.09± 27.57	717.45± 32.89	781.5± 27.20	919.57± 38.22	841.72±16.18
6	<sup>238</sup> U	474.42±8.34	540.9±17.89	565.25±21.09	574.29±30.55	556.47±33.02	433.15±7.61
	<sup>232</sup> Th	95.27±4.08	96.83±9.10	102.41±10.13	91.08±11.24	86.68±12.84	85.72±3.55
	<sup>40</sup> K	755.90±16.67	884.69±28.06	868.45±32.41	776.95±28.57	639.10±45.18	634.46±13.99
17	<sup>238</sup> U	28.41±2.13	29.76±2.66	45.83±4.85	28.79±3.59	24.84±2.69	32.85±2.51
	<sup>232</sup> Th	29.95±2.27	27.09±3.21	29.32±4.20	31.75±6.08	27±4.95	31.81±2.44
	<sup>40</sup> K	906.03±15.25	927.95±22.22	954.12±21.08	915.91±21.47	1023.68±20.55	944.93±15.91

**Table 5.** Comparison of specific activity for soil samples 7, 17 which measured at Environmental Pollution Lab. and the mathematical method.

Sample No	Activity Concentrations in (Bqkg <sup>-1</sup> ) by mathematical method			Activity Concentrations in (Bqkg <sup>-1</sup> ) in EAEA		
	238U	232Th	40K	238U	232Th	40K
7	50.13	37.60	859.55	45.35	34.86	1033
17	28.41	29.95	906.03	25.72	32.26	1025

### Conclusion

In this work, a previous mathematical method was used to determine the volume efficiency for HPGe detector. A proposed factor  $F_1$  is introduced to correct the difference in the densities between the samples and KCl solution. From the obtained results, it can be concluded that the mathematical method with the proposed factor  $F_2$  described in this work can be used to calculate the volume efficiency at different distances. Therefore the activity concentrations can be calculated at different distances.

### Acknowledgements

The authors would like to express their deepest gratitude to Professor Dr. Sanaa Atia Mohamed Prof. Assistant in Radioactive Pollution Environment Unit, Egyptian Atomic Energy Authority (EAEA) for allowing the measurement of samples 7, 17. The authors also thank Prof. Dr. Samia Abdel Malak professor of nuclear physics at physics department Faculty of Women for Arts, Science and Education. Ain Shams University for the helpful discussion during this work.

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