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Effect of the Radon Background Fluctuation on the Low Activity Measurement of the ^{214}Pb, ^{214}Bi and ^{226}Ra

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Radon background fluctuation,
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Abstract: In this study, radon background fluctuation effect on the measurement of ^{214}Pb, ^{214}Bi and ^{226}Ra low level activity concentrations were investigated in ventilated laboratory. For this purpose, detectors which had 150% and 20% efficiencies were used. In spite of the air ventilation, radon background fluctuation effect on the low level activity measurement was observed. 20% efficiency detector measured activity discrepancies were 5.7% for ^{214}Pb, 8.7% for ^{214}Bi and 5.8% for ^{226}Ra. 150% efficiency detector measured activity discrepancies were 13.8% for ^{214}Pb, 11.5% for ^{214}Bi and 12.7% for ^{226}Ra.

Radon Doğal Fon Dalgalanmasının ^{214}Pb, ^{214}Bi ve ^{226}Ra Düşük Radyoaktivite Ölçümü Üzerindeki Etkisi

Anahtar Kelimeler
Radon doğal fon dalgalanması,
Düşük aktivite,
^{214}Pb,
^{214}Bi,
^{226}Ra

Özet: Bu çalışmada, radon doğal fon dalgalanmasının havalandırma sistemli laboratuvarda ^{214}Pb, ^{214}Bi and ^{226}Ra düşük seviyeli aktivite konsantrasyonlarının ölçümü üzerindeki etkisi araştırıldı. Bu amaçla, %150 ve %20 verimli dedektörler kullanıldı. Havalandırmaya rağmen, düşük seviyeli aktivite ölçümleri üzerinde radon doğal fon dalgalanma etkisi gözlendi. %20 verimli dedektör için ölçülen aktivite farklılığı ^{214}Pb için %5.7, ^{214}Bi için %8.7 ve ^{226}Ra için %5.8 ölçülmüştür. %150 verimli dedektör için ölçülen aktivite farklılığı ^{214}Pb için %13.8, ^{214}Bi için %11.5 ve ^{226}Ra için %12.7 ölçülmüştür.

1. Introduction

The background spectrum is using for Quality Assurance/Quality Control purposes and for background correction in the spectrum evaluation algorithm. Therefore background contribution to the gamma spectrometric measurement is important. For this reasons background spectrums are obtaining regularly. Background spectrum of an HPGe detector arises from the cosmic radiation, building materials surrounding it and radioactive impurities in the detector and shielding materials. The naturally occurring radionuclides are ^{238}U and ^{232}Th decay series and ^{40}K. Main contributions to the natural background spectrum are these primordial radionuclides [1-4]. Generally the activity levels of primordial radionuclides are low but there are encountered high activity concentrations.

Another contribution to the background is radon gas and its daughter nuclides. Radon is a noble gas and concentration in the laboratory changes. Radon permeates in the detector environment and affects the peak count rates in the spectrum through its daughter nuclides. Additionally radon background fluctuation affects the Compton continuum. Radon fluctuation in the laboratory can affect considerably the activity determination and reduces its reliability. The most serious effect of the radon fluctuation in laboratory is on the measured activity concentration of the ^{214}Pb, ^{214}Bi daughters of the ^{226}Ra.

Generally environmental sample contains low content of artificial and natural radionuclides. The determination of low level activities of natural radionuclides in these samples has problem of natural background subtraction. Low detection limits provide more accurate and precise measurements of radionuclides in environmental samples. So to improve low detection limit gamma-ray spectrometry laboratories pay special attention to minimize the radiation background of detectors [5-7].

Measurement sensitivity in energy regions not containing the peaks of interested radionuclides will be affected by radon fluctuations in the background.

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Also the minimum detectable activity (MDA) of the photopeaks of the radionuclides in the spectrum will be affected by radon fluctuation in the background. Commercially available gamma spectrometry analysis software packages include peaked background corrections. But this software packages do not consider fluctuation of the radon background level in the laboratory due to the seasonal and climate changes.

In the present work radon background fluctuation effect on the activity measurements in the ventilated gamma spectrometry laboratory is investigated. Background measurements are taken in different days. Each background spectrums is used to evaluate ^{214}Pb, ^{214}Bi and ^{226}Ra activity concentration in IAEA-SOIL-375 soil sample. Also MDA of the each radionuclide is calculated.

2. Materials and Methods

Saraykoy Nuclear Research and Training Center (SNTRC) Gamma Spectrometry Laboratory (GML) operates 10 coaxial HPGe detectors with relative efficiencies 10–150% and DSA-1000 - DSPEC multichannel analyzer. GML accredited by Turkish Accreditation Agency (TURKAK), a member of International Laboratory Accreditation Cooperation, in 2009. GML has taken part in comparison tests and has been inspected by TURKAK experts annually since then. GML has two entrances and two ventilation system mounted on the windows. Both ventilation systems are on 24 hours and 365 days.

Two software programs GAMMAVISION-32 and GENIE-2000 have been used for data analyses. Detectors which were chosen for statistical comparison of background fluctuation are presented in Table 1.

Table 1. Specifications of detectors

Detector	#1	#2
Manufacturer	ORTEC	CANBERRA
Model No.	GEM150P4	GR2020
Type	p-type	n-type
Resolution (1.33MeV)	2.31	1.80
%Rel. Eff. (1.33MeV)	150%	20%
Crystal diameter [mm]	94.8 mm	50 mm
Crystal length [mm]	87.2 mm	51 mm

Before the measurements, energy calibration and peak shape calibration was done by using peaks of ^{241}Am, ^{137}Cs and ^{60}Co radionuclides which were obtained from a certified radioactive standard point sources. IAEA-RGU was used as standard reference material and IAEA-SOIL-375 was used as sample for activity measurement. Activity concentrations are directly proportional to the net count rate of the samples. Therefore, to determine activity concentrations of samples, the net count rate of reference material and the net count rate of samples were compared with each other and calculated at 2s statistical uncertainty. According to the IAEA-SOIL-375 certificate ^{226}Ra activity was 20 Bq/kg. Considering that ^{226}Ra in equilibrium with the daughter ^{214}Pb and ^{214}Bi, activity of the ^{214}Pb and ^{214}Bi was 20.0 Bq/kg. This value appropriate for to determine the effect of the radon fluctuation on the low level activity.

The activity concentrations of the samples were determined from radionuclide's own energies or gamma ray photopic of their decay products. The activity concentration of ^{214}Pb was calculated from 295.2, 351.9 keV gamma-ray energies. The activity concentration of ^{214}Bi was calculated from 609.3 keV. The activity concentration of ^{226}Ra was calculated from the 295.2, 351.9 keV gamma-ray energies of ^{214}Pb and 609.3 keV of ^{214}Bi.

The activity concentrations of the samples are calculated by Equation 1 :

$$A = \frac{N}{\varepsilon_\gamma \ P_\gamma \ t \ M} \quad (1)$$

where N corresponds the net peak area of gamma-ray energy, ε_γ denotes the absolute efficiency, P_γ is the gamma-ray yield per decay, t and M denotes the counting time and sample mass, respectively. Minimum Detectable Activity (MDA) calculations were performed by Equation 2 [8]:

$$MDA = (1.64)\sigma_n/\varepsilon.P.t.w \quad (2)$$

where σ_n stands for standard deviation of the background in the region of interest and equal square root of the number of count for the background spectrum; ε is absolute efficiency; P is emission probability of gamma decay; t and w denote measurement time and weight of the dried sample respectively.

3. Results and Discussion

Background measurements are planned daytime, nighttime, all day time and longtime for the Det#01 and longtime for the Det#02. Each background spectrums is used to evaluate ^{214}Pb, ^{214}Bi and ^{226}Ra activity concentrations in IAEA-SOIL-375 soil sample. Daytime, nighttime, all day time and longtime measurements were carried out for Det#01. Counting times were 28000, 57000, 83000 and 260000 sec respectively. Measured activity, MDA and deviation from reference values are shown in Table 2, 3, 4, 5, 6.

As can be seen at tables, biggest variation of the activity measurement was daytime measurements for Det#01.Table 2 shows the results of the measurements. Minimum and maximum value of the activity measurements for ^{214}Pb, ^{214}Bi and ^{226}Ra are 21.4-23.2 Bq/kg, 20.3-22.6 Bq/kg and 20.9-22.9 Bq/kg respectively. Maximum deviations from

reference value are 13.8% for ^{214}Pb, 11.5 % for ^{214}Bi and 12.7 % for ^{226}Ra.

Measurement average value of the ^{226}Ra was 21.9 Bq/kg. Upper level of ^{226}Ra activity value in the IAEA-Soil-375 was 22.0 Bq/kg in 95% confidence interval. In table 2, there are 6 values for ^{214}Pb, 3 values for ^{214}Bi and 6 values for ^{226}Ra outside the acceptable range. Deviation from reference value for average measurement value of the ^{226}Ra was 8.6%.

Minimum and maximum value of the activity measurement for nighttime measurements at Det#01 for ^{214}Pb, ^{214}Bi and ^{226}Ra are 20.5-22.6 Bq/kg, 19.7-21.6 Bq/kg and 20.1-22.0 Bq/kg respectively. Table 3 shows the results of the measurements.

Maximum deviations from reference value are %11.5 for ^{214}Pb, 7.4% for ^{214}Bi and 9.5% for ^{226}Ra. Measurement average value of the ^{226}Ra was 21.0 Bq/kg. Deviation from reference value for measurement average value of the ^{226}Ra was 5%.

For the allday time measurements minimum and maximum values of the activity measurement for ^{214}Pb, ^{214}Bi and ^{226}Ra are 20.5-22.8 Bq/kg, 19.5-21.9 Bq/kg and 20.0-22.3 Bq/kg respectively. Table 4 shows the results of the measurements. Deviations from reference value are 12.3% for ^{214}Pb, 8.7% for^{214}Bi and 10.3% for ^{226}Ra. Measurement average value of the ^{226}Ra was 21.2 Bq/kg. Deviation from reference value for measurement average value of the ^{226}Ra was 6%.

Smallest variation of the activity measurements was longtime measurements for Det#01. Table 5 shows the results of the measurements. For the longtime measurement minimum and maximum value of the activity measurement for ^{214}Pb ^{214}Bi and ^{226}Ra are 19.4-21.7 Bq/kg, 18.8-21.1 Bq/kg and 19.1-21.4 Bq/kg. Maximum deviations from reference value are 7.8 % for ^{214}Pb, -6.4% for ^{214}Bi and 6.5% for ^{226}Ra. Average measurement value of the ^{226}Ra is 20.5 Bq/kg. When we compared these values with the reference value no value was outside the acceptable range. Average measurement value was the closest one to thereference value. Deviation from reference value for average measurement value of the ^{226}Ra was 2.5%.

Due to the low detector efficiency only long time measurements were carried out for Det#02. Counting times were 260000 sec. Table 6 shows the results of the measurements. Minimum and maximum value of the activity measurement for ^{214}Pb, ^{214}Bi and ^{226}Ra are 19.5-21.2 Bq/kg, 18.4-20.5 Bq/kg and 18.9-20.7 Bq/kg respectively. Maximum deviation from reference value 5.7% for ^{214}Pb, -8.7% for ^{214}Bi and 5.8% for ^{226}Ra. When we compared these values with the reference values, these values were in the acceptable range. Average measurement value of the ^{226}Ra was 19.9 Bq/kg. Deviation from reference value was 0.5%.

Table 2. Measured activities, deviation from the reference values and MDA values for Det #01(Daytime).

	Number of the measurement									
	1	2	3	4	5	6	7	8	9	10
^{214}Pb										
Activity (Bq/kg)	22.7	22.9	21.4	22.4	21.7	22.8	21.7	23.0	21.7	23.2
Deviation (%)	11.9	12.7	6.5	10.7	7.8	12.3	7.8	13.0	7.8	13.8
MDA	0.6	0.6	0.7	0.6	0.7	0.6	0.8	0.5	0.7	0.5
^{214}Bi										
Activity (Bq/kg)	21.6	22.5	20.7	22.0	21.1	21.3	20.3	21.8	20.7	22.6
Deviation (%)	7.4	11.1	3.4	9.1	5.2	6.1	1.5	8.3	3.4	11.5
MDA	1.1	1.0	1.2	1.0	1.0	1.0	1.1	0.9	1.0	0.9
^{226}Ra										
Activity (Bq/kg)	22.1	22.7	21.1	22.2	21.4	22.0	20.9	22.4	21.2	22.9
Deviation (%)	9.5	11.9	5.2	9.9	6.5	9.1	4.3	10.7	5.7	12.7
MDA	0.8	0.8	0.9	0.8	0.8	0.8	0.9	0.7	0.8	0.7

Table 3. Measured activities, deviation from the reference values and MDA values for Det #01(Nighttime).

	Number of the measurement									
	1	2	3	4	5	6	7	8	9	10
^{214}Pb										
Activity (Bq/kg)	20.5	21.6	21.6	22.6	21.2	22.2	20.7	20.7	21.5	21.2
Deviation (%)	2.4	7.4	7.4	11.5	5.7	9.9	3.4	3.4	7.0	5.7
MDA	0.5	0.5	0.4	0.5	0.5	0.4	0.5	0.6	0.5	0.5
^{214}Bi										
Activity (Bq/kg)	19.7	20.8	20.4	21.4	20.5	21.6	20.0	19.8	21.2	20.4
Deviation (%)	-1.5	3.8	2.0	6.5	2.4	7.4	0.0	-1.0	5.7	2.0
MDA	1.0	0.7	0.8	0.9	0.8	0.8	0.8	0.8	0.8	0.8
^{226}Ra										
Activity (Bq/kg)	20.1	21.2	21.0	22.0	20.8	21.9	20.3	20.2	21.3	20.8
Deviation(%)	0.5	6	5	10	4	9.5	1.5	1	6.5	4
MDA	0.7	0.6	0.6	0.7	0.6	0.6	0.7	0.7	0.7	0.7

Table 4. Measured activities, deviation from the reference values and MDA values for Det #01(Allday time).

	Number of the measurement									
	1	2	3	4	5	6	7	8	9	10
^{214}Pb										
Activity (Bq/kg)	22.8	20.5	21.4	22.4	21.2	22.6	21.9	21.8	21.9	20.5
Deviation (%)	12.3	2.4	6.5	10.7	5.7	11.5	8.7	8.3	8.7	2.4
MDA	0.2	0.4	0.3	0.3	0.4	0.4	0.3	0.4	0.4	0.4
^{214}Bi										
Activity (Bq/kg)	21.9	19.6	20.4	20.9	20.7	21.8	20.8	20.5	21.2	19.5
Deviation (%)	8.7	-2.0	2.0	4.3	3.4	8.3	3.8	2.4	5.7	-2.6
MDA	0.5	0.6	0.7	0.8	1.0	0.5	0.6	0.6	0.5	0.7
^{226}Ra										
Activity (Bq/kg)	22.3	20.1	20.9	21.6	20.9	22.2	21.3	21.1	21.5	20.0
Deviation (%)	10.3	0.5	4.3	7.4	4.3	9.9	6.1	5.2	7.0	0.0
MDA	0.4	0.5	0.5	0.6	0.7	0.4	0.5	0.5	0.4	0.5

Table 5. Measured activities, deviation from the reference values and MDA values for Det #01(Longtime).

	Number of the measurement									
	1	2	3	4	5	6	7	8	9	10
^{214}Pb										
Activity (Bq/kg)	21.3	21.0	21.7	21.1	19.4	19.7	21.3	19.9	21.4	21.7
Deviation (%)	6.1	4.8	7.8	5.2	-3.1	-1.5	6.1	-0.5	6.5	7.8
MDA	0.3	0.2	0.1	0.2	0.4	0.3	0.2	0.3	0.2	0.2
^{214}Bi										
Activity (Bq/kg)	20.2	20.7	20.8	20.7	18.8	19.5	20.4	19.4	21.1	21.1
Deviation (%)	1.0	3.4	3.8	3.4	-6.4	-2.6	2.0	-3.1	5.2	5.2
MDA	0.7	0.4	0.2	0.3	0.5	0.4	0.3	0.4	0.4	0.3
^{226}Ra										
Activity (Bq/kg)	20.7	20.8	21.3	20.9	19.1	19.6	20.8	19.6	21.2	21.4
Deviation (%)	3.4	3.8	6.1	4.3	-4.7	-2.0	3.8	-2.0	5.7	6.5
MDA	0.4	0.3	0.2	0.3	0.4	0.4	0.3	0.3	0.3	0.3

Table 6. Measured activities, deviation from the reference values and MDA values for Det #02

	Number of the measurement									
	1	2	3	4	5	6	7	8	9	10
^{214}Pb										
Activity (Bq/kg)	19.9	20.5	21.2	19.6	19.6	20.0	20.3	20.9	19.5	21.1
Deviation (%)	-0.5	2.4	5.7	-2.0	-2.0	0.5	1.5	4.3	-2.6	5.2
MDA	0.9	0.7	0.8	0.9	0.8	0.8	0.7	0.6	0.6	0.7
^{214}Bi										
Activity (Bq/kg)	19.2	19.3	20.1	18.4	19.4	19.8	19.8	20.5	19.3	20.1
Deviation (%)	-4.2	-3.6	0.5	-8.7	-3.1	-1.0	-1.0	2.4	-3.6	0.5
MDA	0.6	0.7	0.7	0.7	0.6	0.7	0.6	0.6	0.6	0.7
^{226}Ra										
Activity (Bq/kg)	19.6	19.9	20.6	18.9	19.5	19.9	20.0	20.7	19.4	20.6
Deviation (%)	2.0	0.5	2.9	5.8	2.6	0.5	0.0	3.4	3.1	2.9
MDA	1.0	0.8	1.0	1.0	0.8	0.9	0.9	0.8	0.8	0.9

4. Conclusions

Variations of the count rates caused by the changes of the radon levels in laboratory have significant effects on the measured activity concentrations of ^{214}Pb, ^{214}Bi and ^{226}Ra. 150% and 20% detectors were used to evaluate this effect. GML is ventilated laboratory but effect of the radon background fluctuations on the low activity measurement of the ^{214}Pb, ^{214}Bi and ^{226}Ra is observed.

Measured radioactivity concentrations discrepancies for the 150% efficiency detector are bigger than 20% efficiency detector. For 20% efficiency detector measured activity concentration discrepancies were 5.7% for ^{214}Pb, 8.7% for ^{214}Bi and 5.8% for ^{226}Ra. For 150% efficiency detector maximum measured activity concentration discrepancies were 13.8% for ^{214}Pb, 11.5% for ^{214}Bi and 12.7% for ^{226}Ra. Minimum radon background fluctuation for Det#01 and Det#02 is longtime counting. Therefore longtime background counting is appropriate for activity concentrations determination or mean value of the long term monitored background fluctuation can be taken.

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