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Gamma-ray Exposure Rate Monitoring by Energy Spectra of NaI(Tl) Scintillation detectors

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ABSTRACT

Background: Nuclear facilities in South Korea have generally adopted pressurized ion chambers to measure ambient gamma ray exposure rates for monitoring the impact of radiation on the surrounding environment. The rates assessed with pressurized ion chambers do not distinguish between natural and man-made radiation, so a further step is needed to identify the cause of abnormal variation. In contrast, using NaI(Tl) scintillation detectors to detect gamma energy rates can allow an immediate assessment of the cause of variation through an analysis of the energy spectra. Against this backdrop, this study was conducted to propose a more effective way to monitor ambient gamma exposure rates.

Materials and Methods: The following methods were used to analyze gamma energy spectra measured from January to November 2016 with NaI detectors installed at the Korea Atomic Energy Research Institute (KAERI) dormitory and Hanbat University. 1) Correlations of the variation of rates measured at the two locations were determined. 2) The dates, intervals, duration, and weather conditions were identified when rates increased by 5 $nSv{\cdot}h^{-1}$ or more. 3) Differences in the NaI spectra on normal days and days where rates spiked by 5 $nSv{\cdot}h^{-1}$ or more were studied. 4) An algorithm was derived for automatically calculating the net variation of the rates.

Results and Discussion: The rates measured at KAERI and Hanbat University, located 12 kilometers apart, did not show a strong correlation (coefficient of determination = 0.577). Time gaps between spikes in the rates and rainfall were factors that affected the correlation. The weather conditions on days where rates went up by 5 nSv·h-1 or more featured rainfall, snowfall, or overcast, as well as an increase in peaks of the gamma rays emitted from the radon decay products of ^{214}Pb and ^{214}Bi in the spectrum. This study assumed that ^{214}Pb and ^{214}Bi exist at a radioactive equilibrium, since both have relatively short half-lives of under 30 minutes. Provided that this assumption is true and that the gamma peaks of the 352 keV and 1,764 keV gamma rays emitted from the radionuclides have proportional count rates, no man-made radiation should be present between the two energy levels. This study proved that this assumption was true by demonstrating a linear correlation between the count rates of these two gamma peaks. In conclusion, if the count rates of these two peaks detected in the gamma energy spectrum at a certain time maintain the ratio measured at a normal time, such variation can be confirmed to be caused by natural radiation.

Conclusion: This study confirmed that both ^{214}Pb and ^{214}Bi have relatively short half-lives of under 30 minutes, thereby existing in a radioactive equilibrium in the atmosphere. If the gamma peaks of the 352 keV and 1,764 keV gamma rays emitted from these radionuclides have proportional count rates, no man-made radiation should exist between the two energy levels.

Keywords: Gamma ray exposure rates, Radiation monitoring, NaI(Tl) scintillation detector, Gamma energy spectrum

Introduction

Monitoring gamma exposure rates with NaI(Tl) scintillation detectors is beneficial in many ways. Commercial 76 mm Φ×76 mm NaI(Tl) scintillation detectors have a higher efficiency than pressurized ion chambers (PICs), which are commonly used for gamma ray detection in South Korea. They are also much more effective in ascertaining causes of variation in gamma exposure rates, as they can measure gamma ray energy spectra. Japan and the Baltic-Nordic countries have chosen to use NaI(Tl) scintillation detectors to monitor gamma exposure rates for this reason [1, 2]. China is also planning to use dosimeters equipped with NaI detectors for its radiation monitoring system that is currently being deployed.

NaI(Tl) scintillation detectors have been adopted in some facilities, including nuclear power plants, in South Korea for measuring ambient gamma exposure rates, but are mostly used for simply monitoring variation in count rates in the gamma energy spectrum. The city of Daejeon is the only municipal government in South Korea attempting to measure gamma energy spectra to identify causes of variation in gamma exposure rates [3]. There are two major obstacles to analyzing the spectra measured by NaI detectors for monitoring gamma exposure rates. First, a single measurement spectrum may include 500 to 1,000 channels of data, meaning that the total number of spectra created annually from 15-minute interval measurements can reach as many as 35,040 per each detector. Analyzing so many spectra is only possible if automated. The second obstacle has to do with the fact that NaI detectors are temperature-sensitive. The values measured by NaI(Tl) scintillation detectors have been reported to decrease when temperature rises, at the rate of –0.0068/°C.[1)] In other words, if the peak value at 0°C is at channel 500, it will move to channel 432 at 20°C. Such fluctuation in the peak channels requires energy calibration for each measured spectrum. The 60 keV gamma ray emitted from ^{241}Am is used for calibration. The radionuclides that are important for monitoring ambient radiation near nuclear power plants are ^{134,137}Cs and ^{131}I. These nuclides, however, release gamma rays whose energy levels are 300 keV or more, and the 60 keV gamma ray emitted from ^{241}Am does not have a great impact on the nuclides' gamma spectrum. Calibration is also possible using the gamma peaks of 352 keV, 1,460 keV, and 2,614 keV emitted from ^{214}Pb, ^{40}K, and ^{208}Tl, respectively, all of which are naturally occurring radionuclides that exist within the earth's crust or soil.

NaI spectra contain a considerable amount of information on ambient gamma ray energy, such as gamma exposure rates, radiation concentration levels in the soil or crust, and the increase in gamma exposure rates during rainfall. Gamma exposure rates and radiation concentration levels have been investigated in many studies in South Korea. This study was conducted to understand the causes of variation in gamma exposure rates using gamma energy spectra. To this end, we studied gamma ray energy spectra measured by the 76 mm Φ×76 mm NaI detectors installed at the dormitory of the Korea Atomic Energy Research Institute (KAERI) and Hanbat National University campus, both located in the city of Daejeon, over a period of 11 months in 2016. These spectra contain the general traits of gamma exposure rate variation, based on which we suggested a method to monitor gamma ray exposure rates more effectively.

Materials and Methods

1. Installation of NaI detectors

The dosimeters installed at the KAERI and Hanbat University campuses were products developed by SI Detection, a Korean business.

They were equipped with 76 mm Φ×76 mm NaI detectors that measure gamma energy spectra up to 3 MeV with 1,024 channels. The devices were installed approximately a meter above the ground, as seen in Figure 1, with sunshades cover-

Fig. 1. Ambient gamma energy spectrum measurement device.

1) Csaba M. Rozsa, Temperature compensated scintillation detector and method, US 6407390 B1(2000).

ing the top to prevent abrupt changes in temperature of the detector. Gamma energy spectra were measured every 15 minutes and the results, which included breakdowns per channel and spectrum analysis reports, were sent to a server in Daejeon. The spectrum analysis reports contained energy calibration data and gamma ray exposure rates calculated from the spectra.

2. Energy calibration

The NaI ambient gamma energy spectra measured outdoors demonstrated peaks from the 352 keV gamma ray emitted from the natural radiation nuclide ^{214}Pb, the 1,460 keV gamma ray emitted from the nuclide ^{40}K, and the 2,614 keV gamma ray emitted from the nuclide ^{208}Tl as shown in Figure 2. We found that channel shift of each peak maintained the same proportion, which provides grounds for using a certain gamma ray peak for energy calibration. Such an energy calibration method, in which measured spectra are used, are valuable when man-made radiation peaks are included in a spectrum. Most of the gamma energy emitted from nuclear facilities is under 2,000 keV, meaning that spectra under 2,000 keV can be disturbed by man-made radiation. In other words, the peaks of the 2,614 keV gamma ray often detected in NaI ambient gamma energy spectra are not disturbed by man-made radiation, and therefore can be used for energy calibration.

Let us assume that the energy calibration function of a spectrum measured at the temperature of T(°C), where man-made radiation can be ignored, is as follows:

$$C(T) = a_1 + a_2E + a_3E^2 \quad (1)$$

Here, E stands for gamma energy, C(T) for the peak channel of gamma energy E, and a_1, a_2, a_3 are constants. If the peak of gamma energy E measured at a different temperature of T'(°C) has the value of C'(T'), the energy calibration function will be as follows:

$$C'(T') = k[a_1 + a_2E + a_3E^2] \quad (2)$$

From this, we can deduce that $k = \frac{C'(T')}{C(T)}$. The proportional constant k for the energy calibration of the spectrum measured at temperature T' can be derived using the 2,614 keV gamma ray peak, which can again be used to deduce other gamma energy peaks.

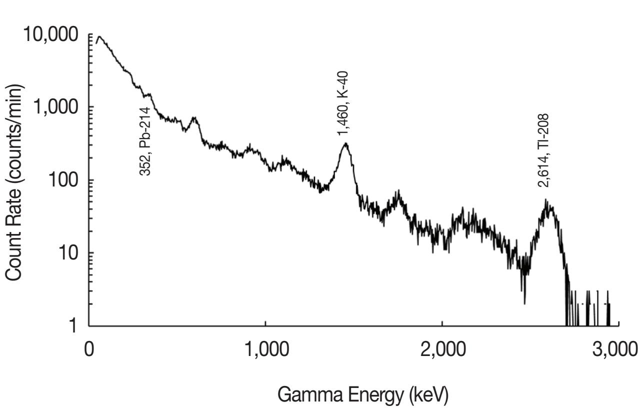

Fig. 2. Peaks used for energy calibration.

3. Monitoring the variation of ambient gamma exposure rates

Ordinary ambient gamma exposure rates measured over 15 minutes or longer using PIC dosimeters or NaI detectors varied within deviations of 2 nSv·h^{-1} among nearby devices, with the rates spiking during rainfall and returning to normal a few hours later. In this case, the variation of gamma exposure rate can be calculated by comparing the currently measured gamma exposure rate with the immediately previous measured value. If this variation is greater than the normal variation magnitude, the variation in the next measurement is calculated based on the gamma exposure rate measured before the fluctuation. This calculation method of gamma exposure variation is useful for automatically analyzing time series gamma exposure rate. Figure 3 shows the flow chart for monitoring the variation of ambient gamma exposure rates that was used in this study.

This simple flow chart is particularly valuable because it

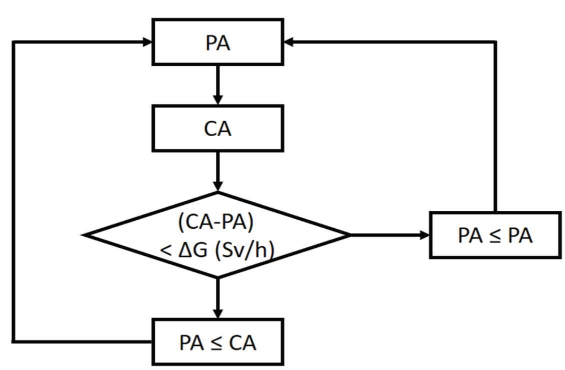

Fig. 3. Flow chart for monitoring the variation of ambient gamma exposure rates. PA, Previous ambient gamma exposure rate; CA, Current ambient gamma exposure rate; ΔG, In continuous ambient gamma exposure rate measurements, the difference between the current measured value and the previous measured value.

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only represents the fluctuation gap, even when measured in different regions.

4. Evaluating the natural variation of ambient gamma radiation exposure rates

Variation in ambient gamma exposure rates in normal environments is mostly caused by changes in the concentration of ^{214}Pb and ^{214}Bi, radon decay products that exist in the atmosphere. The two radionuclides have relatively short half-lives of 26.8 and 19.9 minutes, respectively. This is a reason why they are likely to exist at radioactive equilibrium s within the atmosphere. A variety of different gamma rays are emitted from these nuclides; ^{214}Pb emits gamma rays under 352 keV of energy, while ^{214}Bi emits gamma rays of 609 keV or higher. If the nuclides are at radioactive equilibrium, the ratio of the numbers of the gamma rays emitted from each will remain the same. The ratio between the peak count rate of the 352 keV gamma ray and that of the 1,764 keV gamma ray will stay consistent within the energy spectrum unless man-made gamma rays are present with energy levels between the two.

This study selected count rates between 330 keV and 374 keV for the 352 keV gamma energy peak, and from 1,650 keV to 1,878 keV for the 1,764 keV peak as shown in the Figure 4, the combined area of which accounted for a 99% or higher confidence interval (3 σ) of the peak area.

Results and Discussion

Table 1 presents the characteristics of ambient gamma exposure rates measured by the dosimeters installed at the KAERI dormitory and Hanbat University between January 2 and November 30, 2016, as shown in Figure 5. The dosimeters estimated gamma exposure rates by measuring gamma energy spectra from 76 mm Φ × 76 mm NaI detectors; thus, the rates do not account for the contribution of cosmic radiation (32 nSv·h^{-1}) [4], and only represent the contribution of radiation emitted from radionuclides within the earth's crust and atmosphere. As seen in Table 1, the values measured at the two locations had similar averages, at 113.1 nSv·h^{-1} and 104.4 nSv·h^{-1}, respectively. The standard deviation for the variation of gamma exposure rates at Hanbat University was around 2 nSv·h^{-1} higher than that of the other location. This can be explained by the different weather conditions between the two stations.

The highest recorded rates at each location were 213.9 nSv·h^{-1} for the KAERI dormitory at 17:30, August 9, and 233.7 nSv·h^{-1} for Hanbat University at 06:45, June 24.

Figure 6 demonstrates the relationship between the rates measured at the two locations. The x-axis represents the KAERI rates and the y-axis represents the rates measured at Hanbat University. It is clear in the graph that the numbers from the two locations are linearly proportional, with a coefficient of determination of 0.5732. However, the proportional

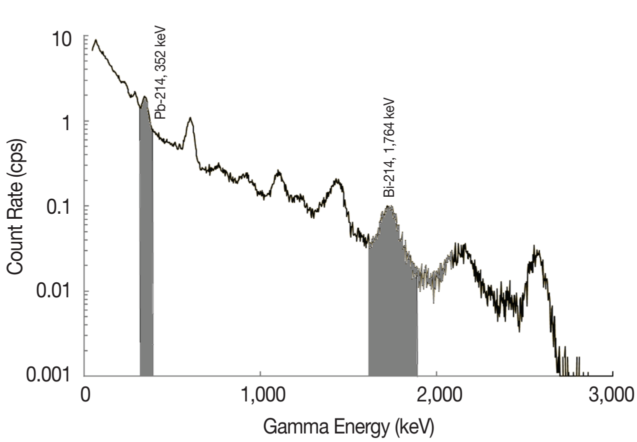

Fig. 4. The gamma ray peaks emitted from ^{214}Pb and ^{214}Bi that were selected to evaluate the variation of natural radiation.

Table 1. Characteristics of the Exposure Rates Measured by the Gamma Dosimeters Installed at the Dormitory of the KAERI and Hanbat National University

Location	KAERI dormitory	Hanbat University
Number of collected data points	31,967 (97.40%)	31,967 (97.40%)
Average exposure rate (nSv·h^{-1})	113.1	104.4
Standard deviation (nSv·h^{-1})	4.9	6.9
Maximum exposure rate (nSv·h^{-1})	213.9	233.7
Maximum increase in the exposure rate (nSv·h^{-1})	100.8	129.3

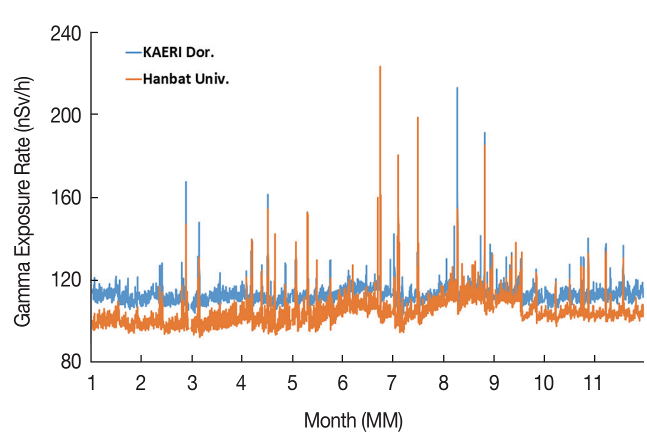

Fig. 5. The ambient gamma exposure rates measured at the Korea Atomic Energy Research Institute dormitory and Hanbat National University.

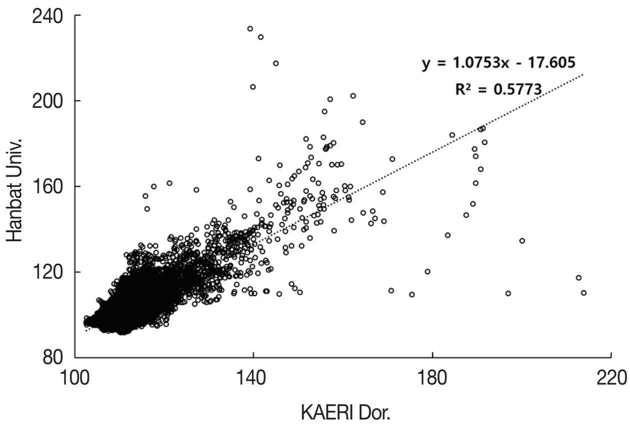

Fig. 6. Relationship between the gamma exposure rates measured at the Korea Atomic Energy Research Institute dormitory and Hanbat National University.

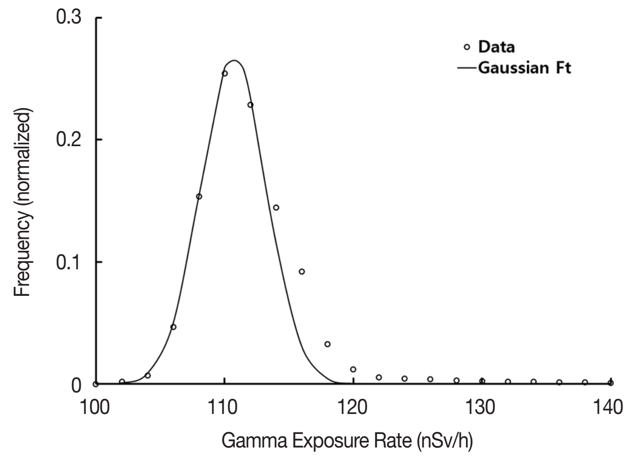

Fig. 8. Distribution of ambient gamma exposure rates measured at the Korea Atomic Energy Research Institute dormitory.

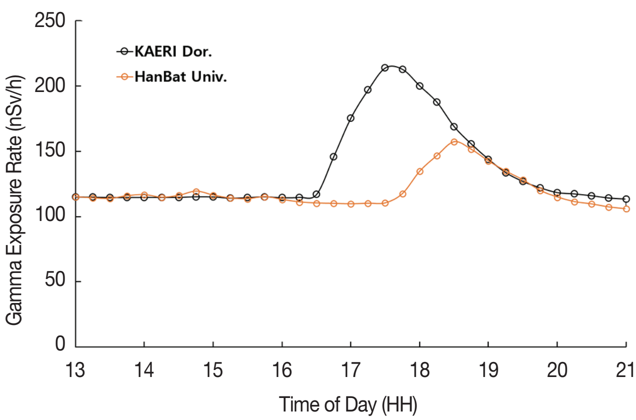

Fig. 7. Ambient gamma exposure rates measured on August 9, 2016.

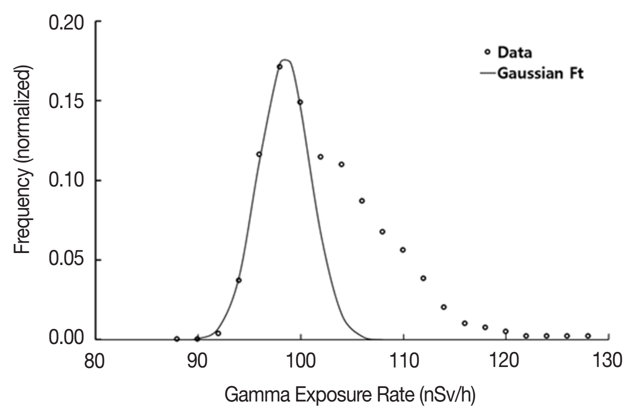

Fig. 9. Distribution of ambient gamma exposure rates measured at Hanbat University.

relationship seemed to weaken as the rates increased. An analysis of data from the period of increased rates suggests that this had to do with rainfall and other weather conditions. In other words, the different timing of rainfall in the two locations led to discord in the peak points or an increase in one location and not in the other. Figure 7 shows an example where the rates in the two locations peaked inconsistently. The graph demonstrates a case in which the rates at Hanbat University spiked some time after a rise at the KAERI location.

Figures 8 and 9 present the distribution of the measured gamma exposure rates. The graphs indicate that the measured rates followed a normal distribution curve at lower values, but gradually moved away from it as they increased. It was found that 85.7% of the KAERI rates followed a normal distribution curve, compared to only 56.2% of the values measured at Hanbat University. This considerable gap implies that there were other factors that determined this variation, in addition to statistical deviations during the measuring process. One such factor is known to be weather conditions. Radionuclides in the atmosphere consist of radon and its decay products emitted from the earth's crust, whose proliferation is reduced during temperature inversion events, leading to increased concentrations of radon and its decay products around the surface. Radon decay products that exist in the upper atmosphere also descend to the surface during rainfall, boosting atmospheric radiation levels emitted from radon decay products near the surface.

Table 2 lists the amount and duration of increases on days that saw a 5 $nSv{\cdot}h^{-1}$ or more increase in the rates, using the algorithm developed in Figure 3 to understand the patterns of variation. The average variation of the rates was set at 2 $nSv{\cdot}h^{-1}$. The number of days where the rates increased by 5 $nSv{\cdot}h^{-1}$ or more was 37 and 39 for KAERI and Hanbat University, respectively, and 29 were the same days. The average

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Table 2. Date and Weather on Days When the Exposure Rate Increased by More Than 5 nSv·h-1

Date		KAERI		Hanbat		Weather
Month	Day	Duration (h)	Increase (nSv·h-1)	Duration (h)	Increase (nSv·h-1)	
January	23	0.3	5.8	-	-	Snow
February	12	4.3	13.0	-	-	Rainfall
	13	2.8	17.2	4.5	15.3	Rainfall
	14	-	-	0.8	7.8	Rainfall
	26	3.8	18.1	0.5	6.3	Snow
	28	3.3	48.0	3.3	39.5	Snow
	29	-	-	2.0	15.6	Snow
March	5	4.5	36.4	0.8	7.8	Rainfall
April	6-7	15.0	22.6	62.0	38.5	Rainfall
	16-17	10.0	42.6	7.5	37.0	Rainfall
	18	1.8	9.9	1.8	11.7	Rainfall
May	3	7.3	23.5	9.0	30.9	Rainfall
	10	0.6	9.2	15.3	42.8	Rainfall
	24	4.8	10.3	-	-	Rainfall
June	7	2.8	10.5	2.3	18.5	Rainfall
	15	-	-	0.8	8.5	Rainfall
	20	-	-	1.0	9.3	Rainfall
	22	2.3	14.6	3.8	52.0	Rainfall
	24	10.3	35.0	6.5	98.3	Rainfall
	30	1.0	13.5	-	-	Cloudy
July	1	3.3	26.1	4.3	17.6	Rainfall
	1-2	-	-	1.3	8.4	Rainfall
	3-4	10.3	33.5	14.3	61.4	Rainfall
	6	2.3	9.3	2.3	8.1	Rainfall
	15-16	10.8	43.8	15.3	94.9	Rainfall
August	7	2.3	33.3	-	-	Cloudy
	9	3.3	97.0	2.3	47.0	Cloudy
	18	-	-	1.5	21.6	Cloudy
	23	3.3	16.9	1.0	8.7	Cloudy
	26	4.8	73.4	4.8	75.9	Rainfall
	29	2.3	22.1	2.0	12.5	Rainfall
September	2	1.5	8.9			Rainfall
	5	2	7.9	1.5	7.2	Rainfall
	8	2	13.4			Rainfall
	10	2	9.3	2.5	12	Rainfall
	12	3	20	4	16.6	Rainfall
	14			2.5	24.1	Rainfall
	17			3.5	11.4	Rainfall
	17	1	7.3	3	12.6	Rainfall
	18			2	9.1	Rainfall
	27			8.5	13.5	Rainfall
October	23	11	17.8	7	19.7	Rainfall
	25	2.5	7.7	2.5	9.6	Rainfall
	28	30	27.6	2.5	12	Rainfall
November	7	3	12.4	14.5	26.1	Rainfall
	11	2	9.3	2	8.5	Rainfall
	11. 18	5	14.2			Rainfall

KAERI, Korea Atomic Energy Research Institute.

duration of the increase 4.9 hours and 5.8 hours, and the longest duration was 30 and 62 hours, respectively. The common weather conditions on days of rate spikes included snow, rain, or overcast conditions, as shown in Table 2.

The ambient gamma exposure rates were calculated from the gamma energy spectrum assessed with 76 mm $\Phi \times 76$

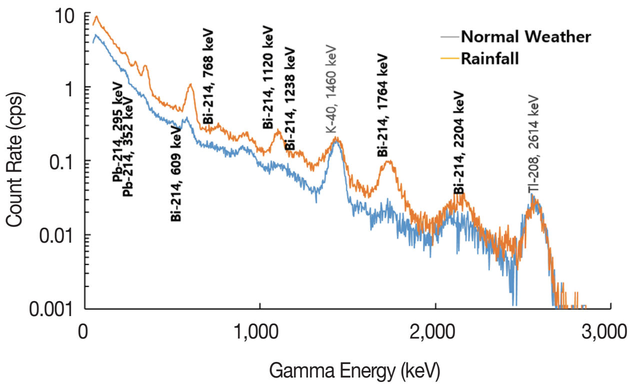

Fig. 10. Peaks of gamma rays emitted from ^{214}Pb and ^{214}Bi in the gamma-ray energy spectrum of the NaI detector.

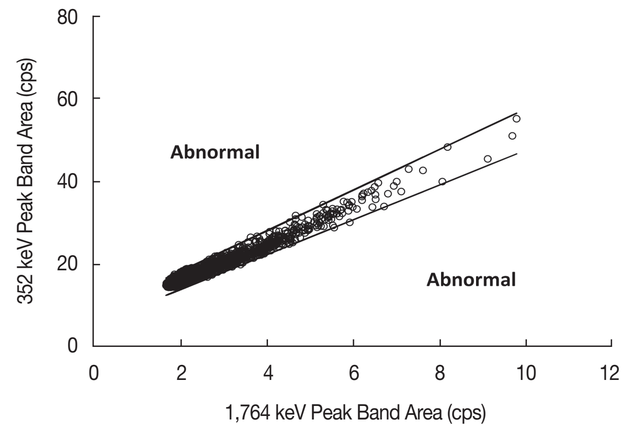

Fig. 11. The ratio of the 352 keV peak counting rate to the 1,764 keV peak counting rate.

mm NaI detectors. Figure 10 is the record of the spectrum measured over 15 minutes starting at 24:00 on January 2, 2016 at the KAERI dormitory. The number of gamma rays detected with the NaI detectors per second was approximately 622. This spectrum shows the peaks of gamma rays emitted from the natural radioactive nuclide series of ^{40}K, ^{238}U, and ^{232}Th. These include the 242 and 352 keV gamma rays emitted from the ^{238}U series nuclide ^{214}Pb, the 1,764 keV gamma ray emitted from 214Bi, the 2,614 keV gamma ray emitted from the ^{232}Th series nuclide ^{208}Tl, and the 1,460 keV gamma ray emitted from ^{40}K.

A report published by the National Council on Radiation Protection and Measurements proposed a method of estimating the level of contribution of each radionuclide series to the ambient gamma exposure rate, using peaks of the 1,460 keV, 1,764 keV, 2,614 keV gamma rays emitted from the ^{40}K, ^{238}U, ^{232}Th nuclides or their decay products [5]. The gamma rates emitted from the ^{40}K, ^{238}U, and ^{232}Th nuclide series calculated using this method were 43 nSv·h^{-1}, 22 nSv·h^{-1}, and 48 nSv·h^{-1}, totaling 113 nSv·h^{-1}. The total rate across Daejeon would be 145 nSv·h^{-1}, after factoring in the contribution of cosmic radiation of 32 nSv·h^{-1} (6). This is 1 nSv·h^{-1} less than the rate of 146 nSv·h^{-1} that was measured at the PIC installed at the KAERI dormitory. In conclusion, the level of contribution of non-natural radiation to the gamma exposure rates was negligible.

Figure 10 combines the spectra measured on sunny and rainy days. This illustrates that the peaks of gamma rays emitted from ^{214}Pb and ^{214}Bi were distinctly high on rainy days. On both sunny and rainy days, the peaks spiked simultaneously when concentration levels of radon decay products increased at the ground surface. Therefore, the relationship between the areas of the peaks serves as the basis for determining whether an increase in radiation levels was caused by natural radiation.

The graph shown in Figure 11 highlights the ratio of the peak counting rates of gamma rays emitted from the two nuclides, ^{214}Bi and ^{214}Pb, measured between January 2, 2016 and August 31, 2016. Clearly, the 352 keV peak counting rate is proportional to that of the 1,764 keV peak, meaning that only natural radiation existed between the two energy levels and that no man-made radiation was detected at KAERI.

Conclusion

Below are the conclusions derived from analyzing gamma ray exposure rate data obtained from January to November 2016 from NaI detector-equipped dosimeters installed at the KAERI dormitory and Hanbat University.

- The rates measured at the two locations, 12 kilometers apart, did not always maintain a proportional relationship (coefficient of determination = 0.577).
- The rates increased by 5 nSv·h^{-1} or more on rainy, cloudy, or snowy days; such increases generally lasted for less than 10 hours, with the longest increase being 4 consecutive days.
- Analyses of gamma energy spectra detected on rainy, cloudy, or snowy days confirmed that gamma rays emitted from the radon decay products of ^{214}Pb and ^{214}Bi were the agents contributing to the rate increases.
- The algorithm we developed for calculating the net variation of gamma rates using the normal variation of rates would be beneficial for monitoring gamma exposure

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rates in various regions.

- The 1,764 keV peak count rates maintained a proportional ratio to the 352 keV count rates, which can be used to monitor gamma exposure rates.

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