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Technical Data

Long-Term Continuous In-Situ Measurement of Gamma Rays Using Ge Semiconductor Detector

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We developed a long-term continuous measurement method for *in-situ* gamma-ray measurement using a Ge semiconductor detector (*in-situ* Ge measurement), and performed continuous measurement for 36 months by the method. A measurement system was successfully stably operated for a long period, and it was confirmed that artificial radioactive nuclides were able to be detected at high sensitivity. A counting rate of each of ^{214}Pb and ^{214}Bi was increased about twice in correspondence to slight increase in environmental radiation dose rate of about 10% as a measurement value of a monitoring post using a NaI (Tl) detector. The problem of discriminating between natural radiation and radiation from man-made source was solved by this system.

KEY WORDS: Ge semiconductor detector, *in-situ*, artificial radioactive nuclide, nuclear power plant, nuclear medicine, radon progeny.

I INTRODUCTION

According to the Agreement about Securing Safety,[1] Shimane prefecture performs monitoring such as measurement of environmental radiation dose rate at 11 monitoring posts around the nuclear power plant. However, artificial radioactive nuclides emitted from the nuclear power plant may not be grasped by the measurement of environmental radiation dose rate because the amount of the nuclides is slight, and therefore the nuclides are largely evaluated by a result of gamma-ray spectrometry using an environmental sample in a laboratory. However, the method hardly provides representing concentration on site promptly in response to emission from the nuclear power plant.

In contrast, the *in-situ* Ge measurement may decrease sampling error caused by non-uniformity of soil compared with measurement in laboratory, and enables determination of a representative value of radioactivity in a radius range of about 10 m in a measurement time about one tenth of measurement time of laboratory measurement, and furthermore enables calculation of radioactivity concentration for each nuclide in soil or on a soil surface, and enables evaluation of dose rate for each series or for each nuclide. In 1972, Beck et al., members of U. S. Health and Safety Laboratory, established the measurement method[2] (HASL method) and reported with HASL-258. In Japan, SAKAI,[3] TERADA[4] and the like, actively studied the measurement in Japan Atomic Energy Research Institute and had various findings, and consequently they established the basis of *in-situ* Ge measurement in Japan. Local governments having a nuclear power plant appreciated this high-resolution measurement method, and then began to use the method for the monitoring. However, the method was used only for batch measurement because of expensive equipment, low weather resistance, necessary replenishment of liquid nitrogen, and, as the most important reason, occurrence of peak drift due to variation in air temperature. Therefore, only several examples of the *in-situ* Ge continuous measurement have been given in the past,[5～7] including one in investigation of influence of accident of the Chernobyl nuclear power plant by Shimane prefecture.[5] However, it has been confirmed from the examples that useful information is obtained by the *in-situ* Ge measurement. In addition, the number of nuclides, as objectives, of the *in-situ* measurement have been extremely increased compared with HASL-258 since a report[8] on an *in-situ* measurement method was published from International Commission on Radiation Units and Measurement (ICRU), and currently the measurement may be performed in various environmental fields.

In this work, we performed long-term continuous measurement aiming to realtime discrimination of gamma-rays by means of *in-situ* Ge measurement while taking into account a practical application of the measurement to monitoring of a nuclear power plant in the future.

We report sufficient weather resistance and a protection

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method of instruments, compensation of long-term drift by improving software, and automatic analysis of data, database construction, and realtime graphic representation of data.

II MEASUREMENT INSTRUMENTS AND MEASUREMENT METHOD

- Measurement period: September 2006~August 2009
- Ge semiconductor detector: portable-P-type Ge, relative efficiency of 113%, energy resolution of 1.9 keV.
- MCA: Inspector 2000 (Canberra).
- Analysis program: Genie2000 and *in-situ* measurement program (Canberra), database (MDB).
- Analysis method: HASL method (including ICRU53) and ISOCS.
  In the Ge semiconductor detector used for the present measurement, a response function was determined by efficiency simulation calculation of MCNP (Monte Carlo Calculation Code) in a space around the Ge detector.
  This is called ISOCS (In-Situ Object Calibration Software) in Canberra,
- NaI (Tl) detector: sphere 3 inches in diameter, resolution of 7%, low potassium glass.
- Analysis method: G (E) function method[9] (G (E) method) and response matrix method[10] (RM method).

September 2006, a portable Ge semiconductor detector was fixed by a tripod with a distance of 100 cm from the ground to a crystal center in grass of the Nishihamasada measurement station in the Nuclear Power Environment Center (**Fig. 1**).

A vinyl sheet to prevent precipitation protected each section of the detector, and silica gel was placed in the periphery of connectors to prevent moisture. The detector was placed within a four-leg tarp to prevent direct sunlight, heavy wind, rain and snow. Dewar container of liquid nitrogen held 7 liters, and liquid nitrogen was replenished at a frequency of twice a week because consumption of liquid nitrogen was about 1.5 liters a day.

A data processor was installed within a small tent before March 2007, and cables were led through an industrial hose 100 mm in diameter for reducing change in temperature and protecting the cables so that the data processor was connected to the detector. Data were able to be analyzed even within the center by wireless LAN. Later, the data were able to be analyzed even within a building of the Nishihamasada measurement station by using long cables.

Fig. 1 *In-situ* Ge measurement scene in the Nishihamasada monitoring post, September 2006.

In the Nishihamasada measurement station, environmental radiation dose rate and weather were continuously measured at intervals of two minutes by using the spherical NaI (Tl) detector of 3 inches in diameter and an ionization chamber, and concentration of radon, concentration of radon progeny, and total alpha/beta concentration were continuously measured every hour.

Any analysis was performed by manual operation except for continuous spectrum sampling for the first six months. Moreover, drift of about 1.5 keV at the maximum occurred in ^{40}K, and therefore mistakes frequently occurred in identification. Therefore, energy was manually recalibrated for each file for reanalysis. Since the Auto Gain Adjust function as an automatic gain adjustment method of Genie 2000, and the Stabilizer function thereof were not able to be incorporated in the continuous measurement, a tracking program for a gamma-ray peak (1,460.8 keV) of ^{40}K was used to cope with the drift, so that drift of the gamma-ray peak was successfully kept within 0.2 keV, and identification accuracy was remarkably improved.

In and after March 2007, a program for sequentially processing a measurement analysis sequence and database construction was completed, so that power saving was able to be achieved, and therefore a measurement interval was changed from 60 min to 10 min.

Measurement values of the Ge semiconductor detector were basically analyzed by the HASL method. Uranium-series (U) nuclides and thorium-series (Th) nuclides were evaluated in such a manner that assuming that radioactive equilibrium was established, dose rate of each series was obtained from individual spectra of nuclides in the series, and dose rate was evaluated for each series by obtaining gamma-ray emission rate weighted average.

A ground surface was assumed to be a flat infinite-plane, and natural radioactive nuclides (U series, Th series and ^{40}K) were assumed to be constant in concentration in a depth direction because they were derived from components of soil respectively.

Fallout nuclides generally show vertical distribution approximated by an exponential function satisfying $\beta = 4.8$ g/cm^2 ($\alpha = 0.33$ cm^{-1}: $\rho = 1.6$ g/cm^3). However, in the case of such distribution, when new fallout is evaluated, radioactive concentration is overestimated about three times.[11] Therefore, artificial radioactive nuclides and ^{7}Be were assumed to be distributed over the ground surface. Density of soil was assumed to be 1.6 g/cm^3.

III MEASUREMENT RESULTS AND CONSIDERATION

1. Calibration of instruments

In July 2006, energy calibration, zero degree efficiency calibration, and angle calibration were performed using nine

nuclides of ^{241}Am, ^{133}Ba, ^{137}Cs, ^{57}Co, ^{60}Co, ^{203}Hg, ^{54}Mn, ^{22}Na and ^{88}Y of about 400 kBq each.

In ISOCS efficiency calibration, efficiency was produced assuming that the earth was a disk 50 m in radius and 30 cm in depth.

A used Ge semiconductor crystal was 77 mm in length and 81.5 mm in diameter, namely, had a small direction dependence, L/D = 0.94. However, the Ge semiconductor was corrected in angle by a HASL angle calibration file during analysis.

Figure 2 shows angle dependence of the Ge semiconductor detector. While direction dependence was substantially not shown at high energy, efficiency was reduced in oblique incidence at low energy of 60 keV.

This seems to be due to shielding by an aluminum crystal holder of 1.5 mm thick circumferentially holding the Ge semiconductor crystal. At energy of 122 keV, efficiency was highest in oblique incidence of about 50 degrees, namely, higher in oblique incidence. This seems to be because apparent section area of the Ge semiconductor crystal is effective for efficiency compared with shielding of a mount cap in the energy band.

Rough minimum limit values of measurement of the detector were calculated by Currie MDA method (calculated with a reliability of 95%), and shown in **Table 1**. From the table, it is known that when fallout of ^{137}Cs occurs, the ^{137}Cs may be detected even if increase in dose rate may not be confirmed in a monitoring post. The same is true in the case of another artificial radioactive nuclide.

2. Artificial radiation source irradiation experiment

In February 2007, an irradiation experiment was conducted with calibration radiation sources (8 nuclides, 50 to 441 kBq) in order to verify artificial-radioactive-nuclide detection ability of the *in-situ* Ge measurement system.

Eight irradiation distances of 0.5 to 15 m were set, and measurement time was set to 600 sec. Furthermore, analysis of data of the 3-inch spherical NaI (Tl) detector was performed by the response matrix method (RM method).

A clear difference in resolution was seen between them. In the Ge system, all irradiated nuclides were determined. On the other hand, in the NaI (Tl) system, peaks of several nuclides were often superimposed into one broad peak, and thereby determination of the nuclides was substantially impossible.

Figure 3 shows calculation results of dose rate by the *in-situ* Ge, the G (E) method and the RM method respectively.

Total dose rate including natural radiation was the same among the three methods, showing that the dose rate was

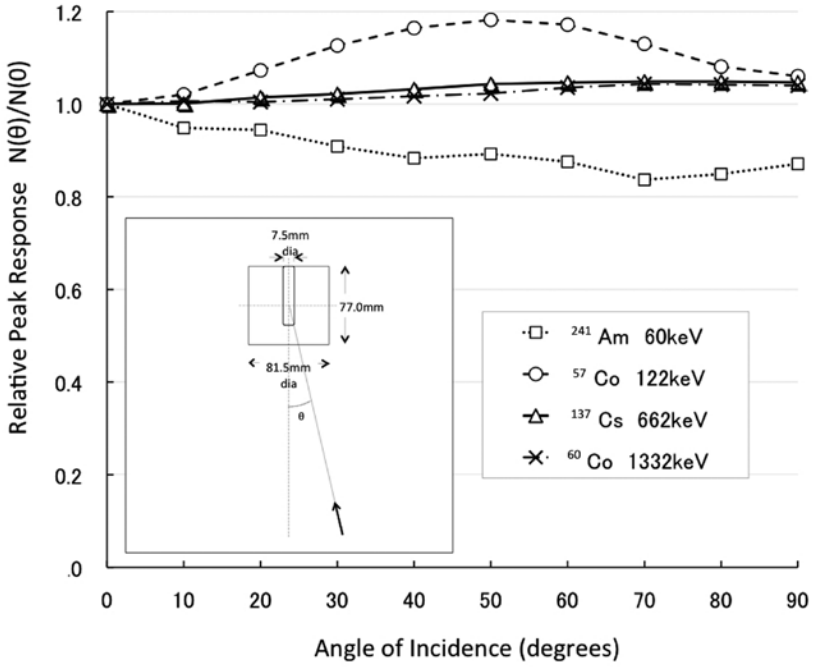

Fig. 2 Angular dependence of peak detection efficiency. The direction of the right under is zero degree.

Table 1 Detection limit of this *in-situ* Ge measurement system.

	^{137}Cs		^{7}Be		^{40}K	
Measurement time (min)	Dose rate (nGy/h)	Surface density (Bq/m^2)	Dose rate (nGy/h)	Surface density (Bq/m^2)	Dose rate (nGy/h)	Soil concentration (Bq/kg)
10	0.04	50	0.08	500	0.33	8
60	0.02	20	0.03	200	0.17	4

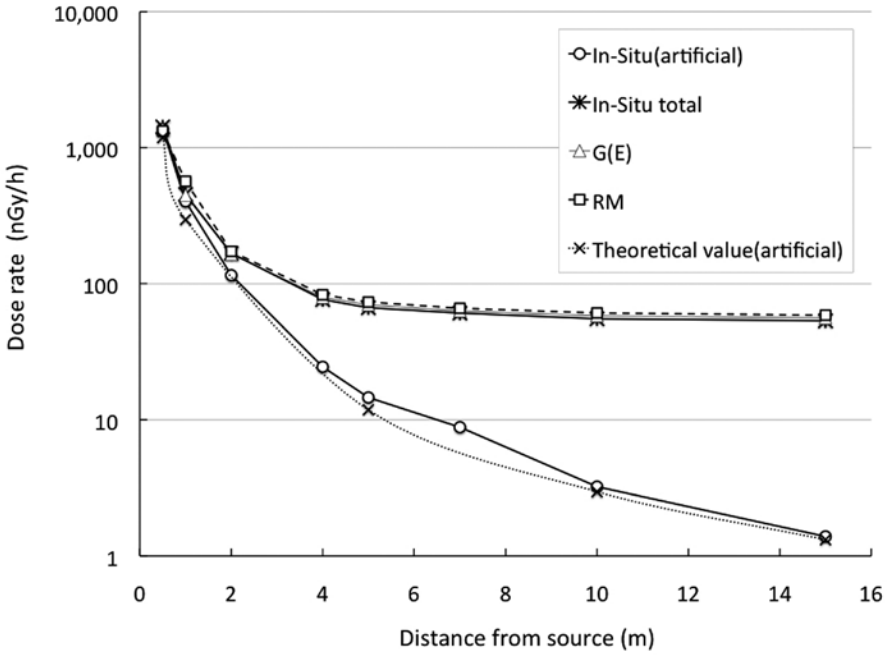

Fig. 3 Difference by dose rate analysis technique. Upper lines are results analyzed by the G(E) function method, the RM method and the *in-situ* Ge method. The Lower lines are theory dose rate from the point sources and dose rate from the artificial radioactive nuclide by the *in-situ* Ge method.

correctly calculated by the G (E) method or the RM method despite the fact that irradiation was performed in a different way from the ground-surface-distributed radiation source (isotropic planar radiation source) assumed in the *in-situ* Ge calculation.

Even when only the artificial radioactive nuclides were focused, theoretical dose rate given by each radiation source was in good agreement with total dose rate of each artificial radioactive nuclide obtained by the *in-situ* Ge measurement.

3. Radiation discrimination from nuclear medicine test subject

In November 2007, a staff member was administrated with ^{201}Tl for a nuclear medicine test and the member underwent radioactive measurement using the *in-situ* Ge system. Urgent request of such measurement was made without notice, and the measurement was performed with no experiment design.

The member was administrated with ^{201}Tl of 74 MBq two days before the measurement.

The subject dosed with radioisotope was separated by 8 m from a monitoring post, and ratio for detection pulse to pass DBM circuit (pass rate) through the monitoring post showed incidence of gamma-rays of approximately 170 keV.

If low-energy gamma-rays did not reach the post, and gamma-rays of 167 keV, maximum energy of ^{201}Tl, largely reached the post, such pass rate is considered to be appropriate. However, nuclide determination may not be performed only from pass rate.

At that time, increase in dose rate of 7 nGy/h was shown in the monitoring post. However, the amount of radioactivity given to the monitoring post, which was calculated based on residual radioactivity of the subject, 44 MBq, was corresponding to increase in dose rate of 8 nGy/h at the monitoring post.

Figure 4 shows measurement spectra of the *in-situ* Ge measurement in that case. Only the maximum energy of 167 keV of ^{201}Tl is increased, and neighboring lines of 70 and 80 keV are accurately separated and qualified.

In the *in-situ* Ge measurement, since ^{201}Tl was neither found in the HASL nuclear data file, nor in the ICRU53 nuclear data file, it was uniquely added for analysis.

In particular, the ICRU53 nuclear data file does not include ^{67}Ga, ^{111}In and ^{123}I often used for nuclear medicine tests, and therefore when a nuclide of the subject is qualified by *in-situ* Ge measurement, nuclear data of them need to be added.

The positron emission tomography (PET), which is expected to be widely used in the future, emits gamma-rays of 511 keV (annihilation gamma), and such gamma-rays are somewhat difficult to be discriminated. However, the gamma-rays are likely to be determined based on variation in high-energy gamma-rays of more than 1 MeV.

Furthermore, it is likely that gamma-rays of ^{85}Kr (514 keV) emitted from a reprocessing plant are able to be separated from the annihilation gamma-rays and qualified.

4. Influence of tarp on measurement values

Measurement was performed while the four-leg tarp was provided on the *in-situ* Ge system. A nuclide such as ^{214}Pb or ^{214}Bi, causing increase in dose rate during precipitation, was given from a drop from a leg of the tarp or the tarp rather than

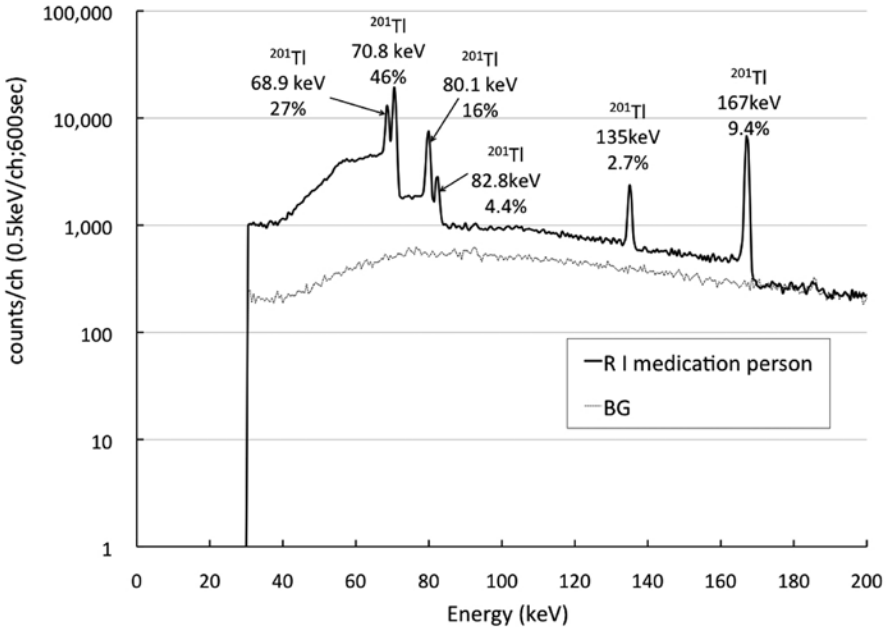

Fig. 4 Ge spectrum of the ^{201}Tl administrated person.
He stood upright at the position where the distance from the detector was 2.5 m.

from a ground surface, and geometry of radiation sources was thus different from that in typical *in-situ* measurement. Therefore, influence of the tarp was investigated.

On December 22, 2008, the tarp was broken by strong wind and removed. To investigate influence of the tarp on that occasion, waterproof was enhanced, and measurement was continued without a tarp.

Measurement values of a monitoring post, which was located 5 m away from the system in a lateral direction, was used, and the amount of increase in dose rate due to precipitation was compared to counting rate of ^{214}Bi being a causative nuclide. Data were sampled in a period from December 16 to 22 in a case with tarp, and in a period from December 25 to 31 in a case without tarp.

While the monitoring post was apt to vary in baseline of dose rate due to variation in water content of soil because the monitoring post was placed on the ground, the baseline was constant in the month.

Figure 5 shows a result of the measurement. ^{214}Bi was investigated using two gamma-rays having low energy of 609 keV and high energy of 1,765 keV.

From the result, counting rate is not significantly changed by presence of a tarp in each of ^{214}Bi of 609 keV and ^{214}Bi of 1,765 keV, and therefore it can be regarded that even if a tarp is provided, approximately the same measurement as the typical *in-situ* measurement may be performed.

5. Evaluation of radioactive nuclide in atmosphere by ISOCS

ISOCS was used for measurement of radon progeny in atmosphere.

In analysis of *in-situ* Ge measurement by the HASL method, objects are limited to be in and on the ground, and other objects may not be analyzed. However, in ISOCS, since efficiency of an object having an optional shape or optional composition may be obtained. We assumed that there was the detector in the center position of the circle of 100 m in radius, and efficiency of the hemisphere (an inverted bowl shape) 100 m in radius covering there was produced.

As a background spectrum of terrestrial gamma radiations, a spectrum, which appeared when radon and its daughter nuclides in atmosphere were decreased to substantially zero in the latter half of a long period without precipitation, was used, and the spectrum was automatically subtracted from a spectrum for each measurement to form a gamma-ray spectrum due to atmosphere.

Concentration of each of ^{214}Pb (RaB) and ^{214}Bi (RaC) was obtained from the formed gamma-ray spectra and the efficiency of the hemisphere 100 m in radius. **Fig. 6** shows part of the obtained concentration in a time series manner.

As shown in **Fig. 6**, when mixing of air was active due to solar radiation in the daytime, and thus concentration of each radon progeny were likely to be substantially uniform in an atmospheric boundary layer, measurement values of the radon progeny by a dust monitor substantially corresponded to analytical values by the *in-situ* Ge measurement, so that effectiveness of ISOCS was confirmed.

In contrast, when a ground inversion layer was formed from the beginning of night to dawn, the measurement values sometimes corresponded to or greatly differed from the analytical values.

This is because ISOCS analysis greatly depends on concentration distribution of a nuclide in a shape used for efficiency calculation in radon progeny analysis. For example,

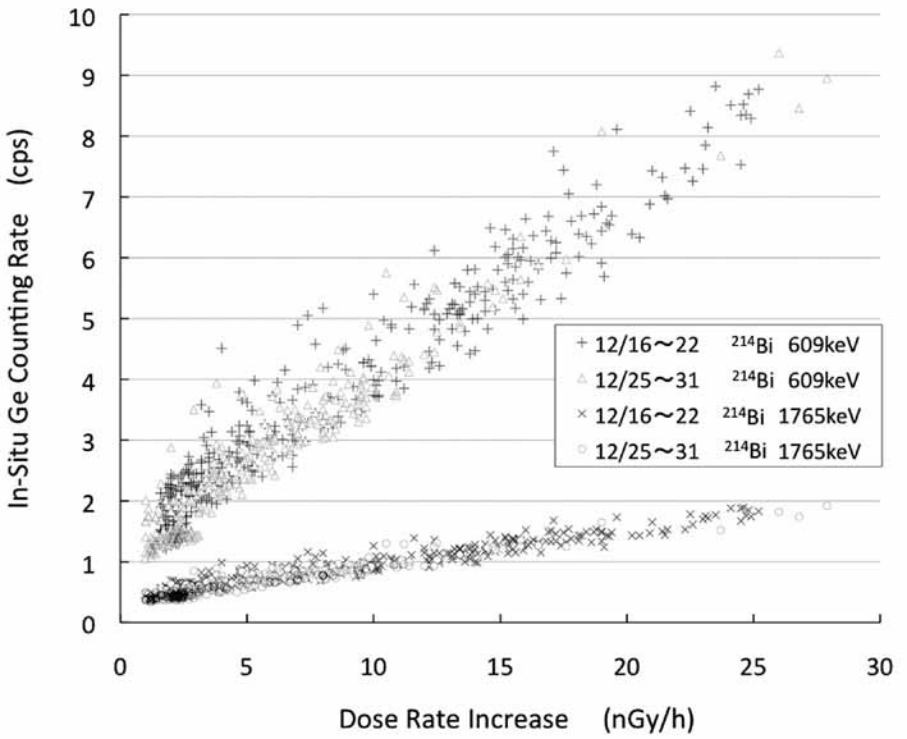

Fig. 5 Influence on measurements with tarp. Relation between increase dose rate by the NaI (Tl) detector and counting rate of radon progeny by the *in-situ* Ge.

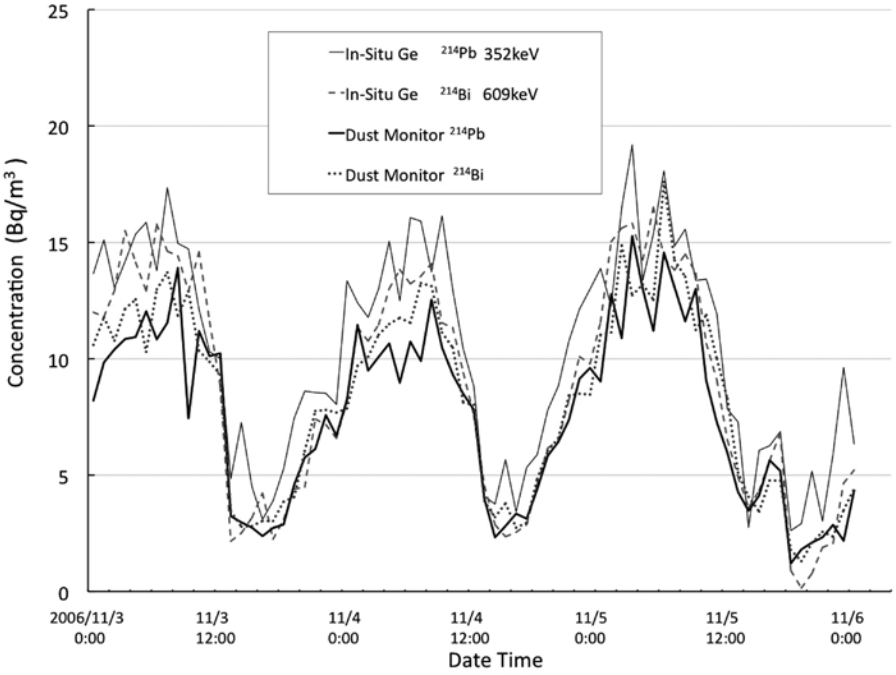

Fig. 6 Comparison between radon progeny concentration analyzed by the ISOCS and dust concentration measured by direct suction of air.

in the case of dawn of November 4, large difference in concentration was considered to exist between the ground surface and 100 m in the sky.

However, this suggests that ISOCS efficiency having a concentration gradient is automatically produced, and gamma-rays having high and low energy of one nuclide are analyzed,

thereby height of the inversion layer may be obtained.

6. Increase in dose rate and equilibrium between radon daughter nuclides (^{214}Bi/^{214}Pb) during precipitation

In the rainy season in 2008, rain intermittently fell from June 21 to 23.

Figure 7 shows environmental radiation dose rate (Nishihamasada monitoring post), precipitation intensity, ^{214}Pb concentration, ^{214}Bi concentration, and concentration ratio between them in the period.

In the Nishihamasada monitoring post, dose rate increased with deposition of the radon progeny (^{214}Pb and ^{214}Bi) on the ground, and as dose rate increased 10%, counting rate (equivalent to concentration) of each of ^{214}Pb and ^{214}Bi increased about twice, and as dose rate increased 20%, the counting rate increased about three times.

While dose rate was monitored in this way, the causative nuclides of increase in dose rate were extremely sensitively monitored.

As shown in the figure, the ratio of ^{214}Bi/^{214}Pb was approximately one during increase of dose rate immediately after start of precipitation (even in another season), and even when rain intermittently fell again during decay of ^{214}Bi after the end of the precipitation, the ratio showed a substantially radioactive equilibrium state during precipitation.

The ratio of ^{214}Bi/^{214}Pb increased with start of precipitation, and reached a maximum value, 1.4 to 1.6, at the end of precipitation.

However, such a numeral value included influence of terrestrial gamma radiations, and when the ratio of ^{214}Bi/^{214}Pb was estimated from true increasing amount subtracted with background of terrestrial gamma radiations, the ratio varied from 1 to 2.5.

This is because the radon progeny deposited in substantially radioactive equilibrium are in a relationship of ^{214}Pb (half-life; 26.8 m) ^{214}Bi (half-life; 19.9 m), and therefore the radioactive equilibrium is broken so that concentration of ^{214}Bi is constantly high before return to background.

When the radon progeny deposit with precipitation and the dose rate rise, since ^{214}Bi contributes to increase in dose rate about seven times more than ^{214}Pb,[12, 13] dose rate increased by precipitation may be roughly grasped by evaluating only ^{214}Bi. However, such deviation from radioactive equilibrium appears as error in the strict sense.

When it is assumed that radon progeny including ^{218}Po (RaA) are taken into cloud droplets mainly by rainout, a certain time for growth of an ice crystal to a raindrop, and a certain precipitation falling time (a water drop 1 mm in diameter has a terminal velocity of 4 m/s, and therefore falls onto the ground in eight minutes from 2,000 m above the ground) are necessary before depositing on the ground, and therefore the ratio of ^{214}Bi/^{214}Pb varies during precipitation due to various conditions.

Although further verification is necessary, it can be estimated that the observation result indicates large contribution of washout.

IV CONCLUSION

As natural radioactive nuclides, U-series nuclides (^{214}Pb, ^{214}Bi and ^{210}Pb,), Th-series nuclides (^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl), ^{40}K and ^{7}Be were detected. But ^{210}Pb was not frequently detected because its energy is low.

During precipitation, only ^{214}Pb and ^{214}Bi were increased in counting rate, showing accurate discrimination of nuclides contributing to increase in dose rate.

During no precipitation, dose rate for each series obtained by the HASL method was approximately in agreement with dose rate obtained by each of the G (E) method using NaI and the RM method.

In the measurement of dose rate by the *in-situ* Ge system, when radon progeny fell onto the ground surface during precipitation, large error occurred because the nuclides were analyzed assuming that they were uniformly distributed in a depth direction of soil. In the case of artificial radioactive nuclides, we irradiated to detector from one point of horizontal direction. Even if the nuclides were analyzed assuming that they were distributed over the ground surface, the analytical dose rate was corresponding to the theoretical dose rate.

Similarly, in the case of radiation sources in atmosphere, concentration of each radiation source was able to be accurately calculated by analysis using ISOCS.

Accuracy of nuclide identification by the present measurement method was confirmed by measurement of the subject dosed with radioisotope for a nuclear medicine test.

An object of the monitoring around nuclear facility imposed on a local government is to confirm a fact that exposed dose caused by the nuclear facility is sufficiently lower than 1 mSv/y When a standard of the exposed dose is temporarily assumed to be only for exposed dose contributed by environmental radiation, and set to 0.05 mSv/y being the dose objectives of a nuclear power plant, dose rate of about 6 nGy/h needs to be discriminated.

However, increase in dose rate of several tens nanogray per hour is not unusual during precipitation, so that even if dose rate increases by about several nanogray per hour due to plume derived from nuclear facility, such increase in dose rate is hardly detected.

Local governments that measure the environmental radiation around nuclear facility have tried to discriminate gamma-rays caused by the facility by analyzing pass rate through a DBM circuit of a pulse detected by a NaI (Tl) detector, counting rate of a single channel analyzer (SCA), and NaI spectra.

However, such discrimination has not completely achieved. This is because error tends to occur in NaI spectrum analysis because a radiation equilibrium relationship between radon progeny (RaB-RaC), which cause increase in dose rate, greatly varies due to precipitation, and artificial radioactive nuclides are hardly determined because of low resolution of the NaI (Tl) detector. Therefore, it seems that whether increase in dose rate of about several nanogray per hour is due to a natural or artificial radioactive nuclide may be distinguished only by a method using the Ge detector having high resolution at present.

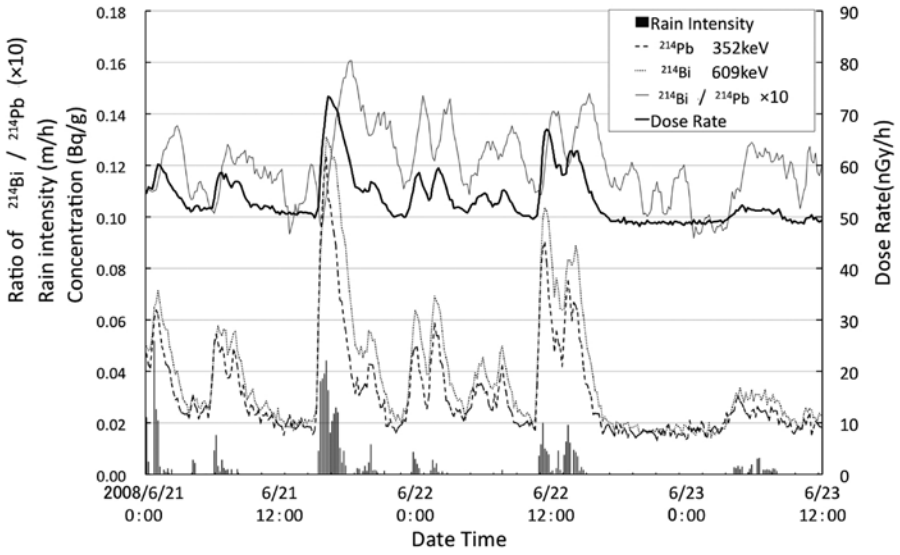

Fig. 7 The trend of radon progeny concentration by the *in-situ* Ge measurement. $^{214}Bi/^{214}Pb$ ratio, precipitation intensity and dose rate were compared.

Even if dose rate given by artificial radioactive nuclides is in order of 10^{-2} nGy/h at 1 m above the ground, the dose rate may be detected by using the present *in-situ* Ge system.

As hereinbefore, gamma-ray spectrometry in outdoor environment using the Ge semiconductor detector is the most useful means for grasping slight accumulation of artificial radioactive nuclides emitted from nuclear facility, for evaluating exposed dose, and for promptly determining nuclides emitted in an accident.

In the present work, *in-situ* Ge measurement was able to be continuously and stably performed for such a long term, which shows that the Ge semiconductor detector may be used for continuous monitoring of environmental radiation around nuclear facility. From a viewpoint of grasping exposed dose caused by the facility, this is revolutionary in the sense that only artificial radioactive nuclides are expected to be monitored as objects for monitoring environmental radiation in a normal condition as in sample analysis. We consider that this shows a direction of a peripheral monitoring system of nuclear facility in the future.

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