Study on Method for Simultaneous Determination of Ambient Dose Equivalent Rates and Activity Concentration in Air for Environmental Radiation Monitoring

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For proper environmental radiation monitoring, a method to simultaneously determine ambient dose equivalent rate and radioactivity concentration in the air by using a newly developed scintillation spectrometer, namely a CeBr3 spectrometer was investigated. The performance of the proposed method, including energy dependence, angular dependence and the linearity of the spectrometer (i.e. the dose rate dependence of its response), was verified by a series of measurements, conducted according to the procedure of inter-comparison of detectors used for early warning network performed by the European Radiation Dosimetry Group (EURADOS). Measurement results show that the proposed method is suitable for environmental radiation monitoring purposes. After thorough tests, the investigation on obtaining activity concentration in air from the pulse height spectrum of γ-ray was demonstrated in the laboratory by using a point-like sealed 133Ba source to simulate an artificial increase of ambient dose equivalent rate due to a radioactive cloud containing 131I and 133Xe. The photon fluence rate was obtained from the pulse height spectrum by using the unfolding method, and the activity concentration in air for radionuclides of interest could be estimated from the obtained photon fluence rate by applying the conversion coefficient evaluated via a Monte Carlo calculation.

Key Words: CeBr3, ambient dose equivalent-H*(10), emergency radiation monitoring, radiation protection, Monte Carlo calculation

1. Introduction

The authors propose a method for the simultaneous determination of ambient dose equivalent rate and radioactivity concentration in air by using a newly developed scintillation spectrometer. To collect radiological information more effectively, environmental γ-ray spectrometry is recognized as a powerful method. For instance, the NaI(Tl) scintillation spectrometers have been used for monitoring the surroundings of nuclear facilities in Japan.1, 2) In European countries, the so-called spectro-dosimetry system for the radiological early warning networks has been developed and ready for installation.3) Installing the NaI(Tl) scintillation spectrometers around nuclear facilities depends mainly on the emergency radiological monitoring strategy in Japan, which focuses on information about abnormal
increase of dose rates. Determination of radioactive concentration in air of radioiodine from the pulse height spectra obtained the NaI(Tl) scintillation spectrometers has been published in several years after the nuclear accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP).\(^1\), \(^2\), \(^4\)

Just after the nuclear accident at the FDNPP, a real-time dose rate mapping system called KURAMA was developed.\(^5\) The KURAMA system helped decision makers establish the rehabilitation strategy from the contamination due to the radioactive materials released from the FDNPP. However, the KURAMA and its successor model KURAMA-II didn’t release the information about radioactivity concentration in real-time. Some of the radiation surveillance systems, such as D3S ID gamma neutron detector\(^6\) enables to show dose rate and identify the radioisotopes involved in the increase of dose equivalent rate. This also can’t determine the accurate radioactive concentration in air for radioisotopes identified by the system.

The authors focused on how the ambient dose equivalent rate and radioactivity concentration in air could be estimated from the same pulse height spectrum obtained using a \(\gamma\)-ray spectrometer, to obtain radiological information almost in real time. A newly developed CeBr\(_3\) scintillation spectrometer was selected for use because of the better energy resolution of \(\gamma\) rays than that of a conventional NaI(Tl) scintillator and relatively low self-contamination. Kessler et al. proposed to introduce several types of scintillation spectrometers including the CeBr\(_3\) for the new generation spectro-dosimeter for the early warning networks in European countries.\(^3\) They developed the conversion coefficients to directly convert the pulse height spectra to the ambient dose equivalent rates and verified the energy and linearity dependence in the reference fields.\(^3\) In this study, the \(G(E)\) function\(^7\) and the unfolding methods were selected for the dosimetry. To determine the activity concentration in air, photon fluence rate was obtained from the pulse height spectrum by using the unfolding method, and activity concentration in air of radionuclides of interest were estimated from the obtained photon fluence rate by applying the conversion coefficient evaluated by means of Monte Carlo (MC) calculations.\(^8\) The newly developed method was verified according to the procedure of inter-comparison of detectors used for early warning network performed by the European Radiation Dosimetry Group (EURADOS).\(^9\) After the thorough performance test, the simultaneous determination of \(H^\#(10)\) and radioactivity concentration in air was demonstrated using a point-like sealed \(^{133}\)Ba source in the laboratory, instead of actual use of a radioactive plume containing \(^{131}\)I and/or \(^{133}\)Xe.

2. Materials and methods

To demonstrate whether or not the proposed method for simultaneous determination of ambient dose equivalent rates and activity concentrations in air of radionuclides is working properly during emergencies, the followings have to be accounted; 1) proper determination of photon fluence rate (by a scintillation spectrometer) from a radioactive plume which will be treated by a volume source containing a variety of radionuclides and 2) proper estimation of radioactivity concentration in air for radionuclides from obtained photon fluence rate spectrum by the proposed method.

The method to determine ambient dose equivalent rate, \(H^\#(10)\) and photon fluence rate should meet the general requirements of dosimeter for environmental monitoring specified in the IEC 60846-1.\(^10\) The items to be tested are the energy dependence, linearity of ambient dose equivalent rate and the angular dependence of the scintillation spectrometer. Proper measurement of pulse height spectrum even under conditions where an attenuation of direct component of \(\gamma\) rays inside a plume due to air is predominant.
should be demonstrated as well. The determination of ambient dose equivalent rate in the reference field for environmental radiation measurement was conducted after the performance test of the CeBr₃ scintillation spectrometer. During the measurement in the reference field, we could detect terrestrial γ rays coming from a semi-infinite source with a variety of radionuclides such as ⁴⁰K, U-series and Th-series. Gamma rays reaching the measurement point consisted of direct γ rays attenuated by soil and air and indirect components due to the Compton scattering by air and soil. In this study, radiation monitoring regarding the radioactive plume is assumed to be carried out inside the plume. For estimating radioactivity concentration in air, a photon fluence rate from the volume source with dimensions of several hundred meters in radius containing mixed radionuclides should be properly determined. It would be an incontrovertible evidence that the proposed method could work properly to monitor γ rays from the radioactive plume if reasonable ambient dose equivalent rate in the reference environmental field would be obtained.

As for the proper estimation of radioactivity concentration in air for radionuclides from measured pulse height spectra by a scintillation spectrometer, we proposed the methodology to apply an appropriate conversion coefficient to a measured photon fluence rate spectrum by a scintillation spectrometer. In this study, appropriate conversion coefficients from photon fluence rate at the height of 1 m above ground to activity concentration in air were calculated by a MC calculation code with the same assumptions as in Hirayama et al. (2014) and Namito et al. (2012). In this study, simple assumption about the plume’s dimension was introduced, because rapid and timely release of the information about radionuclides involved and their activity concentration in air is indispensable in case of a real nuclear accident.

2.1 Instrument

In this study, we employed a cylindrical CeBr₃ scintillation spectrometer (OKEN 44/DM: CeBr3) with a diameter of 2.54 cm and a height of 2.54 cm coupled with a PC-driven multi-channel analyzer (MCA) (ITECH Instruments VENUS). Gamma-ray spectrometry with a suitable energy resolution is required to determine the activity concentration in air of artificial radionuclides generated in a nuclear accident at the FDNPP, such as ¹²²Te, ¹³¹I, ¹³²I, ¹³⁴Cs, ¹³⁶Cs and ¹³⁷Cs. Compared to a conventional NaI(Tl) scintillation detector (around 7% relative energy resolution for 662-keV photon), the CeBr₃ scintillation spectrometer has better energy resolution (4–5% for 662-keV photons) and it will be able to distinguish between the events from each nuclide better, such as 605-keV photons from ¹³⁴Cs and 662-keV photons from ¹³⁷Cs. Moreover, the measurement system used comprises of simple and light units of a spectrometer with MCA and turns out to be an easy-to-handle system. No external power supply and no coolant such as LN2 are required.

2.2 The G(E) function method

In this study, the dose rate was evaluated in terms of ambient dose equivalent rate, \( H^\#(10) \). Ambient dose equivalent rates were evaluated from the pulse height spectra obtained using the CeBr₃ scintillation spectrometer. The G(E) function of the scintillation spectrometer facilitates the direct estimation of dose rate from the measured pulse height spectra. In this study, simple assumption about the plume’s dimension was introduced, because rapid and timely release of the information about radionuclides involved and their activity concentration in air is indispensable in case of a real nuclear accident.

\[
H^\#(10) = \frac{\int_{E_{\text{min}}}^{E_{\text{max}}} N(E) \times G(E) dE}{E_{\text{max}} - E_{\text{min}}}
\]

where \( E_{\text{min}} \) and \( E_{\text{max}} \) are the lower and upper energy limits of the incident γ rays, respectively. N(E) is a pulse height spectrum of the incident γ rays with an energy E, as measured using the spectrometer. The
operator, that is the function \( G(E) \), is defined as a function of \( E \) to satisfy equation (1). The function \( G(E) \) for a scintillation spectrometer was expressed as the polynomial expression (2) in this study:

\[
G(E) = \sum_{k=1}^{k_{\text{max}}} H(k) \times g(E)^{k-m-1} \tag{2}
\]

where \( k \) is the term of equation (2), \( k_{\text{max}} \) is the number of the term, \( H(k) \) is the fitting parameter for equation (2), \( g(E) (= \log_{10} E \text{ in this case}) \) is a function that is selected appropriately, and \( m \) is a constant that is selected as required. The function \( G(E) \) for \( H \ast(10) \) was determined for the CeBr\(_3\) scintillation spectrometer by using the calculated responses of the spectrometer to mono-energetic \( \gamma \) rays ranging from 40 keV to 13 MeV. The set of response functions was calculated using the electron-photon MC simulation code EGS4.\(^{16}\) For better accuracy, detailed geometrical structure of the CeBr\(_3\) scintillation spectrometer was introduced into calculations. The photon-fluence-to-ambient-dose-equivalent conversion coefficient was taken from the International Commission on Radiological Protection (ICRP) publication 74.\(^{17}\) The calculated \( G(E) \) function of the CeBr\(_3\) scintillation spectrometer is shown in Fig. 1. The ambient dose equivalent rates obtained using the \( G(E) \) function were experimentally verified in a reference radiation field including energy and angular dependence, as described below.

2-3 The unfolding method

In addition to the \( G(E) \) function method, the photon fluence rate was evaluated from the pulse height spectrum by using the unfolding method. The response matrix was prepared before the measurement to apply the unfolding method to the obtained pulse height spectra. The response matrix of the scintillation spectrometer was calculated using the MC calculation code, MCNP-4C.\(^{8}\) The CeBr\(_3\) spectrometer geometry was modeled in detail. Mono-energetic photons entered from the direction perpendicular to the cylinder axis of the spectrometer. The photon fluence spectrum from the CeBr\(_3\) spectrometer was obtained by de-convoluting the measured pulse height spectra using the MAXED\(^{18}\) unfolding code. The ambient dose equivalent rates were then estimated by multiplying the photon fluence rate with the fluence-to-ambient-dose-equivalent conversion coefficients provided by ICRP 74.\(^{16}\) The average deviation due to the unfolding process and the uncertainties associated with the response matrix (statistical errors in the MC calculation and uncertainty of the standard source) were accounted for in the total uncertainty. The unfolding method was verified thoroughly in the reference radiation fields operated by the German National Metrological Laboratory, Physikalisch-Technische Bundesanstalt (PTB).

2-4 Determination of activity concentration in air

We propose the method for determining activity concentration in air by multiplying the photon fluence rate at a point of interest with an appropriate conversion coefficient. In this study, photon fluence at a certain height was evaluated from the pulse height spectrum by using the unfolding method, as mentioned above. The area that was identified and

![Fig. 1 Pulse height to ambient dose equivalent rate conversion function, \( G(E) \), for the cylindrical CeBr\(_3\) scintillation spectrometer with dimension of 2.54 cm \( \times \) 2.54 cm.](image)
quantified as the peak in the photon fluence rate spectrum was calculated and provided as the “photon fluence rate” emitted from the radionuclide of interest.

In radiological emergencies, radioactive plumes are known to move and disperse according to atmospheric dynamics. Generally, the radioactive plume to be monitored comprises of various kinds of radionuclides with non-homogeneous distribution inside the plume. During emergencies, decision making whether the member of the public is evacuated should be rapidly implemented. From the viewpoint, a fast and rough estimation of the radioactivity concentration in air is needed.

The conversion coefficient from photon fluence rate to activity concentration in air was calculated using the PHITS code.\(^\text{19}\) It was assumed that the activity concentration in air for the radionuclides originated from a radioactive plume of homogeneous density. In other words, the radioactive plume was treated as a cylinder with a radius of 2 km and a height of 2 km with uniform distribution of radionuclides and with its base at ground level. To reduce computational time more efficiently, the transformed geometry that converts the surface source with a point detector into point source with a surface detector set at the height of interest was applied,\(^\text{11, 20}\) as shown in Fig. 2. A linear source measuring 2 km in length from the surface of the ground to the top of the air volume was selected and a surface detector with a radius of 2 km was selected. The height of the surface detector for detecting \(\gamma\) rays from the source was set at the height of 1.0 m. The calculation was verified by comparing the results with the values estimated by Hirayama et al.\(^\text{12}\)

The activity concentration in air, \(C_{\text{air}}\) was then estimated by applying the evaluated photon fluence rate and the calculated conversion coefficients according to the following equation (3):

\[
\frac{\phi}{F_{\text{air}}} = C_{\text{air}}
\]

where, \(\phi\) is the photon fluence rate evaluated using the unfolding method from the pulse height spectrum of the CeBr\(_3\) spectrometer (photons cm\(^{-2}\)s\(^{-1}\)); \(F_{\text{air}}\) is the conversion coefficient from unit radioactivity of a radionuclide (such as \(^{133}\text{Xe}\) and \(^{131}\text{I}\) contained inside the radioactive plume (photons cm\(^{-2}\)s\(^{-1}\))/(Bq cm\(^{-3}\))) to photon fluence rate at 1.0 m above ground.; The proposed method was verified experimentally using a point-like sealed \(^{133}\text{Ba}\) source with a nominal activity of 0.95 MBq (as of September 2018), instead of using an actual radiation plume. An artificial increase of the radiation level due to a radioactive plume passing by was simulated using a point-like sealed source in the laboratory to comply with the Japanese regulation of prevention from hazards against radiation and radioactive materials. Barium-133 mainly emits \(\gamma\) rays with energies of

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**Fig. 2** Schematic drawing of the calculation geometry employed in this study. The original and transformed calculation geometry is shown (Color online).
81 keV and 356 keV, and these energies are close to those of the \(\gamma\) rays emitted by \(^{133}\text{Xe}\) (81 keV) and \(^{131}\text{I}\) (364.5 keV), respectively. This enables to perform the test for the determination of photon fluence rates from each radionuclide independently at a single measurement. Iodine-131 has several \(\gamma\)-ray emissions which may interrupt the determination of \(^{133}\text{Xe}\) concentration. The influence of the scattered photons by the Compton scattering due to air should be paid attention as well. However, the performance test for deconvolution of photon fluence rate from the pulse height spectrum can be easily made by making use of a single-line due to gamma rays from a point-like sealed \(^{133}\text{Ba}\) source. Therefore, a point-like sealed \(^{133}\text{Ba}\) source is appropriate for testing the performance of the proposed method.

2.5 Performance test of the proposed method

The proposed method was experimentally verified for both dosimetry and determination of radioactivity concentration in air. The performance test for dosimetry was focused on how precisely the ambient dose equivalent rate can be determined considering the wide range of \(\gamma\)-ray energies and dose rate levels of several tens of nSv h\(^{-1}\). The performance tests made in the study include 1) the energy dependence, 2) linearity dependence, 3) inherent background, 4) angular dependence, 5) evaluation of environmental ambient dose equivalent rate, 6) response to the artificial dose rate increase using the plume simulator, and 7) radioactive plume simulation by \(^{133}\text{Ba}\) point-like source.

We made the thorough performance test in these reference fields above mentioned for the development. The IEC60846-1 specifies the criterion of the variation of the response of the CeBr\(_3\) scintillation detector for energy and angular response and the criterion is from 0.71 to 1.67.\(^{10}\) As for the linearity of the response, the criterion is recommended as ranging between \(-13\%\) and \(+18\%\) from 5\(\mu\)Sv h\(^{-1}\) to 1 Svh\(^{-1}\). The obtained responses by the following performance test should satisfy the requirement from the IEC60846-1. For the routine calibration of dosemeters for environmental radiation monitoring, testing in ordinary \(\gamma\)-ray calibration fields which cover down to 2–3 \(\mu\)Sv h\(^{-1}\) is enough for the purpose.

Five different types of \(\gamma\)-ray reference fields, namely, \(^{241}\text{Am},\) \(^{57}\text{Co},\) \(^{137}\text{Cs},\) \(^{226}\text{Ra},\) and \(^{60}\text{Co}\), were used for the energy dependence test. The \(^{137}\text{Cs}\) \(\gamma\)-ray reference field was selected for checking the linearity of the evaluated ambient dose equivalent rate. The Cs \(\gamma\)-ray reference field covers the range of 10–120 nSv h\(^{-1}\) in terms of \(H^*(10)\), which perfectly matches the range of ambient dose equivalent rate normally observed in the environment. A series of the energy and linearity tests were performed in an underground laboratory (UDO II) operated by PTB.\(^{21}\) Inside the underground laboratory, the ambient dose equivalent rate is estimated to be around 1.5 nSv h\(^{-1}\), and the radon concentration in air was of the order of 3 Bq m\(^{-3}\). An overnight measurement (15 h) was made to evaluate the inherent background of the CeBr\(_3\) scintillation spectrometer in the underground laboratory as performed by Kessler et al.\(^{3}\) The angular dependence and the maximum ambient dose equivalent rate measurable of the CeBr\(_3\) scintillation spectrometer were investigated at the facility of radiation standards in Japan Atomic Energy Agency (JAEA).

In addition to the laboratory measurement, the ambient dose equivalent rate due to \(\gamma\) rays from the natural radionuclides was evaluated using the proposed method in the reference site operated by PTB. The ambient dose equivalent rate in the reference field was evaluated precisely by PTB and compared to that by the proposed method for verification. To evaluate \(H^*(10)\) from the terrestrial \(\gamma\) rays in the reference site precisely, the influences of the inherent background of the CeBr\(_3\) scintillation spectrometer and the secondary cosmic radiation should be
properly eliminated from the measured pulse height spectrum. The influence of the secondary cosmic-ray component was determined on a platform floating on a lake. Thus, no terrestrial $\gamma$ rays were present. In addition, the measurements of ambient dose equivalent rates from point-like sealed radioactive sources were conducted at a free-field site in PTB by simulating a “radiological incident” using the so-called “plume simulator.”

3. Results and discussion

3.1 Performance test of the proposed method in terms of dosimetry

3.1.1 Energy and linearity dependence of the CeBr$_3$ scintillation spectrometer

The tests of energy dependence and linearity of dosimetry, and the inherent background of the CeBr$_3$ spectrometer were conducted in UDO II. Table 1 presents a comparison of the ambient dose equivalent rate evaluated using the CeBr$_3$ scintillation spectrometer. The CeBr$_3$ scintillation spectrometer was checked thoroughly in the energy range between 60 and 1250 keV. In Table 1, the results obtained using the G(E) function and the unfolding methods are summarized. The Results show that ambient dose equivalent rates are well obtained using both methods (G(E) function and unfolding methods).

Irradiating the spectrometer in the $^{226}$Ra $\gamma$-ray reference field well simulated the measurement of terrestrial $\gamma$ rays from natural radionuclides in the environment. Even in this case, the ambient dose equivalent rates evaluated by both methods and the reference value were in good agreement (within 6%).

| Source, E (keV) | $^{241}$Am, 59.6 | $^{57}$Co, 122 | $^{137}$Cs, 662 | $^{226}$Ra, 744 | $^{60}$Co, 1250 |
|----------------|-----------------|--------------|----------------|----------------|----------------|
| Reference $H'(10), H$ (nSv h$^{-1}$) | 135 ± 3 | 15.8 ± 0.2 | 105 ± 1 | 283 ± 3 | 75.1 ± 0.6 |
| evaluated $H'(10)$ using the $G(E)$ function, $M_{GE}$ (nSv h$^{-1}$) | 140 ± 2 | 17.4 ± 1.4 | 105 ± 1 | 285 ± 4 | 79.8 ± 0.4 |
| Response, $R_M$ | 1.039 | 1.10 | 0.993 | 1.008 | 1.06 |

(b) the unfolding method

| Source, E (keV) | $^{241}$Am, 59.6 | $^{57}$Co, 122 | $^{137}$Cs, 662 | $^{226}$Ra, 744 | $^{60}$Co, 1250 |
|----------------|-----------------|--------------|----------------|----------------|----------------|
| Reference $H'(10), H$ (nSv h$^{-1}$) | 135 ± 3 | 15.8 ± 0.2 | 105 ± 1 | 283 ± 3 | 75.1 ± 0.6 |
| evaluated $H'(10)$ by the unfolding method, $M_U$ (nSv h$^{-1}$) | 118 ± 1 | 16.1 ± 0.2 | 102 ± 1 | 267 ± 3 | 79.8 ± 0.7 |
| Response, $R_M$ | 0.879 | 1.02 | 0.970 | 0.943 | 1.06 |
rates evaluated by both methods were within 13% of the reference $H^*(10)$. This fully satisfies the requirement of the dosimeter for radiation protection purposes, ranging from 0.71 to 1.67 as specified by the IEC60846-1\(^{10}\) and also the Japanese Industrial Standards JISZ4333.\(^{22}\)

Linearity in terms of the ambient dose equivalent rate in the Cs $\gamma$-ray reference field was verified in UDO II. The linearity of the ambient dose equivalent rate evaluated using the CeBr$_3$ scintillation spectrometer in conjunction with the G(E) function and the unfolding methods in the $^{137}$Cs $\gamma$-ray reference field is summarized in Table 2. As given in Table 2, the evaluated ambient dose equivalent rates agreed well with the reference ambient dose equivalent rates from 10 nSv h$^{-1}$ to 100 nSv h$^{-1}$. This also meets the requirement from the IEC60846-1.\(^{10}\) These findings show that ambient dose equivalent rate normally observed in the environment could be determined properly using both methods.

### 3.1.2 Influence of the inherent background of the CeBr$_3$ scintillation spectrometer

The inherent background of the CeBr$_3$ scintillation spectrometer was determined using the unfolding method and be 7.9 nGy h$^{-1}$ in terms of the air kerma rate and 9.2 nSv h$^{-1}$ in terms of the ambient dose equivalent rate, respectively. Both values were estimated using the same pulse height spectrum obtained in the overnight measurement in the underground laboratory. It is known that the scintillation crystal of CeBr$_3$ contains a trace amount of radioactive $^{227}$Ac, which can be ascribed to the crystalline fabrication process.\(^{14, 23}\) The total count rate per volume due to the self-contamination was estimated to be 0.12 counts/s/cm$^3$, which was more than two times larger than shown in the table 2 of the reference 14. The total count rate per volume depends on each crystal, ranging from 0.022 $\pm$ 0.001 counts/s/cm$^3$ to 0.051 $\pm$ 0.004 counts/s/cm$^3$ for 2.54-cm in dia.$\times$2.54-cm in length CeBr$_3$ scintillation crystal. The

### Table 2 The linearity of $H^*(10)$ evaluated by pulse height spectrum from CeBr$_3$ scintillation spectrometer. The Cs $\gamma$-ray reference field was chosen. The G(E) function and the unfolding methods were employed for evaluating ambient dose equivalent rate, $H^*(10)$. The coverage factor is $k = 1$

#### (a) the G(E) function method

| Source, source ID    | $^{137}$Cs, 137Cs, UG50 | $^{137}$Cs, 137Cs, UG52 | $^{137}$Cs, 137Cs, WV98 |
|----------------------|-------------------------|-------------------------|-------------------------|
| Evaluation of $H^*(10)$ using the G(E) function, $M_{G(E)}$ | 10.7 $\pm$ 0.1 | 49.8 $\pm$ 0.4 | 105 $\pm$ 1 |
| Response, $R M_{G(E)}/H$ | 1.00 | 1.03 | 0.993 |

#### (b) the unfolding method

| Source, source ID    | $^{137}$Cs, 137Cs, UG50 | $^{137}$Cs, 137Cs, UG52 | $^{137}$Cs, 137Cs, WV98 |
|----------------------|-------------------------|-------------------------|-------------------------|
| Evaluation of $H^*(10)$ using the unfolding method, $M_U$ | 10.7 $\pm$ 0.1 | 49.8 $\pm$ 0.4 | 105 $\pm$ 1 |
| Response, $R M_U/H$ | 0.950 | 0.991 | 0.970 |
measured results of the same type of scintillator crystal by Kessler et al.\textsuperscript{3)} was determined to be 9.8 nSv h\textsuperscript{−1} in terms of the ambient dose equivalent rate. Comparing the pulse height spectrum due to the inherent background with that obtained by the Guss et al.,\textsuperscript{23)} peaks around 2 MeV (0.002 cps) were almost twice as shown in Fig. 7 (0.001 cps) in the reference 23. As a result of the comparison of measured results with those in references, the inherent background of the CeBr\textsubscript{3} scintillation spectrometer obtained in this study was found to be consistent.

3.1.3 Angular dependence of the CeBr\textsubscript{3} scintillation spectrometer

To fulfill the general requirements for dosimeters for environmental monitoring, the angular dependence of the CeBr\textsubscript{3} scintillation spectrometer was experimentally investigated as well. The symmetry axis of the cylindrical shaped CeBr\textsubscript{3} crystal was set as 0 degree. Sealed $^{133}$Ba, $^{137}$Cs, and $^{226}$Ra radioactive sources were used for the test and measurements were performed in steps of 15 degrees in the plane of the symmetry axis. The results are shown in Fig. 3. Only counting errors of each values were accounted for. The relative responses for $\gamma$ rays (normalized to 90 degrees) above 356 keV are within 10% range. For the lowest $\gamma$ ray energy of 81 keV a higher angular dependence is observed (between 0.95 and 1.18), which is expected due to the cylindrical shape of the crystal and the short path length of the $\gamma$ rays. Comparing the measure results with those obtained by Kessler et al.,\textsuperscript{7)} angular dependences for both CeBr\textsubscript{3} spectrometers have similar tendencies for each energy region. Although the same type of the scintillation spectrometer was selected, the normalized responses for $^{241}$Am source evaluated by Kessler et al.\textsuperscript{3)} were larger than those measured in the study. This might be explained by the presence of the outer Aluminum housing for weather protection. The CeBr\textsubscript{3} spectrometer used by Kessler et al. has a housing, since the CeBr\textsubscript{3} spectrometer in this study are tested in uncovered condition. As for results obtained by $\gamma$ rays with an energy of 662 keV, measured results obtained by both institutes were Only counting errors were accounted for.

The result still fully satisfies the requirements for dosimeter for radiation protection purposes, as specified in IEC 60846-1 (from 0.71 to 1.67).\textsuperscript{10)} During the measurements of terrestrial $\gamma$ rays and in the free-field site described later, the CeBr\textsubscript{3} scintillation spectrometer was set up so that the cylinder axis of the spectrometer was be aligned perpendicular to the ground surface. The measured results of the angular dependence imply that no significant difference for evaluated photon fluence rate and $H^*(10)$ could be observed even in the situation where $\gamma$ rays come and enter from omni-direction on the ground.

3.1.4 Ambient equivalent dose rate for environmental background

After the thorough tests of the energy dependence, the angular dependence and the linearity of the proposed method, natural background radiation was measured at the reference site in PTB. Before the environmental measurement at the reference site,
the effect of the secondary cosmic-ray component was estimated to be 2.5 nSv h\(^{-1}\) and that of the self-contamination component to be 9.2 nSv h\(^{-1}\). Both components were excluded after obtaining the photon fluence rate spectra. The ambient dose equivalent rate due to \(\gamma\) rays from natural radionuclides was then evaluated to be 49.4\(\pm\)0.8 nSv h\(^{-1}\) by the unfolding method. These results are consistent with the reference value (51.8\(\pm\)4.0 nSv h\(^{-1}\)) of the ambient dose equivalent rate due to terrestrial radiation measured using a GE Reuter Stokes reference ionization chamber. This strongly supports that our proposed method could obtain consistent photon fluence rate spectra from direct and indirect components of \(\gamma\) rays emitted by natural radionuclides in semi-infinite volume sources such as the ground.

3.1.5 Response to the artificial dose rate increase using the plume simulator

In addition to the “static” measurements mentioned above, irradiation using the so-called plume simulator, which facilitates the irradiation of \(\gamma\) rays with a remote control, was performed at the free-field reference site, a large lawn at the PTB premises. The cylindrical axis of the CeBr\(_3\) scintillation spectrometer was set perpendicular to the ground surface and the spectrometer was exposed to uncollimated photon radiation of sealed \(^{137}\)Cs, \(^{60}\)Co, and \(^{226}\)Ra sources to test its ability to detect small

| radionuclides | PTB reference by RS-001 \(H_e'(10)\) (nSv h\(^{-1}\)) (A) | CeBr\(_3\) scintillation spectrometer, by the unfolding method \(H_e'(10)\) (nSv h\(^{-1}\)) (B) | En value |
|--------------|---------------------------------|---------------------------------|--------|
| \(^{226}\)Ra | 400 \(\pm\) 12                   | 386 \(\pm\) 29                  | 0.46   |
| \(^{137}\)Cs | 260 \(\pm\) 10                   | 264 \(\pm\) 5                   | 0.39   |
| \(^{137}\)Cs | 68.2 \(\pm\) 2.6                 | 67.0 \(\pm\) 1.1                | 0.40   |
| \(^{137}\)Cs | 144 \(\pm\) 5                    | 146 \(\pm\) 3                   | 0.37   |
| \(^{60}\)Co  | 252 \(\pm\) 12                   | 251 \(\pm\) 8                   | 0.04   |
artificial increases above the natural background. The spectrometer and the PTB reference instrument were placed in a circle of radius 5.0 m around the sealed $\gamma$-ray source, as shown in Fig. 4. The reference of each instrument was positioned 1.0 m above the ground (reference height).

The obtained ambient dose equivalent rates are summarized in Table 3. The plume simulator reproduces an artificial increase of $H^*(10)$ due to radionuclides which may be contained within a radioactive plume passing by, and the test items were, 1) detection of artificial increases above the natural background, 2) identification of radionuclides involved, and 3) changes in precise ambient dose equivalent rate due to abovementioned increases. The unfolding method was employed. The unfolding method facilitates the identification of the radionuclides involved and the quantification of the resulting ambient dose equivalent rate properly. For comparison, the $E_n$ values were calculated according to the following equation (4):

$$E_n = \frac{|\dot{H}^*(10)_{\text{PTB}} - \dot{H}^*(10)_{\text{JAEA}}|}{\sqrt{U^2_{\text{PTB}} + U^2_{\text{JAEA}}}}$$

(4)

where, $\dot{H}^*(10)_{\text{PTB}}$ is the reference value for the ambient dose equivalent rate by PTB, $\dot{H}^*(10)_{\text{JAEA}}$ is the measured ambient dose equivalent rate by the proposed method, and $U_{\text{PTB}}$ and $U_{\text{JAEA}}$ are the associated expanded uncertainties by each institute. In the study, the criterion for the $E_n$ value was taken as 1.0. Evaluated $E_n$ values range from 0.04 to 0.46. This shows the ambient dose equivalent rates obtained by the proposed method are identical to the reference values provided by the PTB.

3.2 Radioactivity concentration in air

3.2.1 Conversion coefficients for determining radioactivity concentration in air

An attempt to estimate activity concentration in air from measured pulse height spectra was implemented concurrently with the performance test at the free-field site in PTB. Instead of using an actual radioactive plume, a point-like sealed radioactive source of $^{133}$Ba was selected, to reproduce an artificial increase of the radiation level above the natural background (BG) due to a radioactive plume in the laboratory. From the de-convoluted photon fluence spectrum, the photon fluence rate of each $\gamma$ ray of interest was calculated, and the radioactivity concentration in air was determined by multiplying the

| Plume height in meter (m) | Total volume of the radioactive plume (cm$^3$) | Photon fluence rate at 1.3 m height per unit radioactivity concentration of $^{131}$I (cm$^2$ s$^{-1}$ (Bq cm$^{-3}$)) |
|---------------------------|---------------------------------------------|--------------------------------------------------------------------------------------------------|
| 50                        | $6.283 \times 10^{14}$                       | $2689 \pm 12$                                                                                    |
| 100                       | $1.257 \times 10^{15}$                       | $3288 \pm 14$                                                                                    |
| 200                       | $2.513 \times 10^{15}$                       | $3595 \pm 15$                                                                                    |
| 500                       | $6.283 \times 10^{15}$                       | $3707 \pm 49$                                                                                    |
| 1000                      | $1.257 \times 10^{16}$                       | $3675 \pm 29$                                                                                    |
| 2000                      | $2.513 \times 10^{16}$                       | $3616 \pm 97$                                                                                    |
photon fluence rate with the conversion coefficient of the activity concentration in air according to equation (3).

The calculation of the conversion coefficient was verified by comparing the results with the estimates obtained by Hirayama et al.\textsuperscript{11)} Table 4 shows variations in calculated photon fluence rate at the reference height of 1.3 m per unit radioactivity concentration of $^{131}\text{I}$ (photons cm\textsuperscript{-2} s\textsuperscript{-1}/(Bq cm\textsuperscript{-3})) with respect to the plume height. The values were calculated for $\gamma$ rays with an energy of 364.5 keV from $^{131}\text{I}$. Statistical errors were considered in quantifying the uncertainty of each value. Calculated photon spectra at 1.3 m above ground from the radioactive plume uniformly containing $^{131}\text{I}$ were also shown in Fig. 5. The obtained value was almost identical to those in Fig. 4 of Hirayama et al.\textsuperscript{11)} As a result, the calculation method was found to be consistent for the purpose. Results from the MC calculations listed in Table 4 also imply that the first 200 m of the plume thickness made a significant contribution to the photon fluence rate at 1.3 m height above ground. Results also suggest that the radioactive materials existing in the region of more than 500 m would less affect the calculated photon fluence rate at 1.3 m height above ground. As shown in Table 4, the estimated plume height affects the results of activity concentration in air of radionuclides of interest.

In Table 5, the variation of the photon fluence rate at 1.0 m height per unit radioactivity concentration in air with respect to plume height for 81-keV $\gamma$ ray from $^{133}\text{Xe}$ was summarized. The photon fluence rate at 1.0 m height per unit photon emission was calculated within 4\% regardless of the plume height. Considering the purpose of the proposed method is focused on timeliness and rapid release of radiological information for members of the public, the assumption of the plume height as 2 km (\(=2000\) m) will meet the

![Fig. 5 Calculated photon fluence rate spectrum at 1.3 m height above ground of $^{131}\text{I}$ uniformly distributed in the radioactive plume. The radius of the radioactive plume was 2000 m (\(=2\) km) and the height was set to 2000 m (\(=2\) km) (Color online).](image)

| Plume height in meter (m) | Photon fluence rate at 1.0 m height per unit radioactivity concentration (photons cm\textsuperscript{-2} s\textsuperscript{-1}/(Bq cm\textsuperscript{-3})) |
|---------------------------|--------------------------------------------------------------------------------|
| 100                       | 1088 ± 6                                                                         |
| 200                       | 1144 ± 8                                                                         |
| 500                       | 1132 ± 10                                                                        |
| 1000                      | 1122 ± 24                                                                        |
| 2000                      | 1126 ± 27                                                                        |
requirement for emergency radiation monitoring strategy. It should be also pointed out that the plume’s dimensions such as height, radius and position above ground should be investigated to evaluate precise activity concentrations, retrospectively.

3.2.2 Benchmark of determination of determining radioactivity concentration in air

The test measurement was conducted in the laboratory as shown in Fig. 6. The point-like sealed $^{133}$Ba source with a nominal activity of 0.95 MBq (as of September 2018) was selected for reproducing an increase of ambient dose equivalent rate due to passing a radioactive plume containing $^{131}$I and $^{133}$Xe. By changing the distance between source and the spectrometer, the intensity of $\gamma$ rays to be identified was changed. The photon fluence rate was derived from the net pulse height spectrum by using the unfolding method.

Fig. 7 shows the pulse height spectrum due to $\gamma$-rays from a $^{133}$Ba source obtained by the 2.54 cm $\times$ 2.54 cm cylindrical CeBr$_3$ scintillation spectrometer. The pulse height spectrum was obtained by the point-like $^{133}$Ba source with nominal activity of 0.95 MBq (Color online).

Fig. 8 Example of photon fluence rate spectrum evaluated from the pulse height spectrum from the $^{133}$Ba by the unfolding method. The pulse height spectrum shown in Fig. 8 was used (Color online).
using the CeBr₃ scintillation spectrometer. The background component was subtracted from measured pulse height spectrum before the application of the unfolding method. Peaks due to γ rays around 80 keV could be clearly identified in the pulse height spectrum. The unfolded photon fluence rate spectrum was then obtained, as shown in Fig. 8. The $H^*(10)$ by the G(E) function method was estimated to be $84.7 \pm 1.4$ nSv h⁻¹. Comparing with the theoretically estimated $H^*(10)$ (86.0 nSv h⁻¹), the proposed method correctly determines the $H^*(10)$ due to a small increase in artificial radionuclides.

The results of photon fluence rates for γ rays with energies of 81 keV and 356 keV are listed in Table 6. The theoretical photon fluence rates for γ rays of each different energy were estimated considering the inverse square law of the distance and the attenuation due to dry air. As listed in the table, the photon fluence rates evaluated using the unfolding method were found to be consistent with the theoretically estimated values. This also supports that the proposed method could derive consistent photon fluence rate of γ rays emitted from each radionuclide.

From the MC calculation, the photon fluence rate per unit radioactivity concentration of $^{131}$I and $^{133}$Xe uniformly distributed in the plume were estimated to be $3606 \pm 46$ and $1149 \pm 27$ cm⁻² s⁻¹/(Bq cm⁻³) at a height of 1.0 m, respectively. Each conversion coefficient was estimated to γ rays at energies of 354 keV for $^{131}$I and 80 keV for $^{133}$Xe, respectively. In this study, $^{133}$Ba point-like source resembles an artificial increase of radiation level due to $^{131}$I and $^{133}$Xe. If evaluated photon fluence rates were to come from a radioactive plume containing $^{131}$I and $^{133}$Xe, activity concentrations in air of each radionuclide would be estimated according to equation (3).

$$ \frac{4.45 \pm 0.05}{3606 \pm 46} \left( \frac{\text{cm}^{-2} \text{s}^{-1}}{\text{Bq cm}^{-3}} \right) = 0.00123 \pm 0.00002 \frac{\text{Bq}}{\text{cm}^3} \quad \text{for } ^{131}\text{I} $$

$$ \frac{2.67 \pm 0.20}{1149 \pm 27} \left( \frac{\text{cm}^{-2} \text{s}^{-1}}{\text{Bq cm}^{-3}} \right) = 0.00232 \pm 0.00018 \frac{\text{Bq}}{\text{cm}^3} \quad \text{for } ^{133}\text{Xe} $$

Then, activity concentrations in air of each radionuclide were estimated to be $0.00123 \pm 0.00002$ Bq cm⁻³ for $^{131}$I and $0.00232 \pm 0.00018$ Bq cm⁻³ for $^{133}$Xe, respectively.

Table 7 summarizes the photon fluence rates obtained from the pulse height spectra due to γ rays from $^{133}$Ba. The background component was subtracted from the measured pulse height spectra before the deconvolution of the pulse height spectra. The photon fluence rate evaluated using the unfolding method was 10% lower than the theoretically estimated value. This might be explained by the fact that fewer γ rays entered the spectrometer and led to relatively larger uncertainty of unfolded photon fluence rate. A CeBr₃ spectrometer with a larger crystal can facilitate the determination of the photon fluence.
rate with reduced uncertainty.

3.3 Applicability of proposed method to radiological emergencies

In the present study, the authors proposed a method for simultaneous determination of dose rates and radioactivity concentrations in air of radionuclides from the same pulse height spectra by using spectrometers. The study particularly focused on the determination of the activity concentrations in air.

For dosimetry, the detection of artificial increases of ambient dose equivalent rate above the natural background was demonstrated with the proposed method to identify the radionuclides involved and to determine the ambient dose equivalent rate due to the increase. Minimum detectable dose equivalent rate was experimentally confirmed to be approximately 10 nSv h\(^{-1}\) from the results of the linearity of the spectrometer for \(\gamma\)-rays from \(^{137}\)Cs. The accumulation time was set as 600 s for the measurement. If measurement duration were set to be 10 min., the evaluated minimum detectable ambient dose equivalent rate is approximately one-fifth of the typical terrestrial BG (~50 nSv h\(^{-1}\)). For 600 s measurement intervals, the minimum detectable counting rate was estimated to be 0.4 cps for the peak area of \(^{137}\)Cs and would be equivalent to an increase of 6 nSv h\(^{-1}\). This is sensitive enough to detect artificial increases in terms of dose equivalent rate due to dispersion of artificial radionuclides. The maximum measurable \(H^*\) (10) for the CeBr\(_3\) scintillation spectrometer was also experimentally verified in the \(^{137}\)Cs \(\gamma\) ray calibration field and evaluated to be 101.3 \(\mu\)Sv h\(^{-1}\). This is quite satisfactory, comparing to the reference value (100 \(\mu\)Sv h\(^{-1}\)).

The authors proposed to introduce the method for simultaneous determination of the activity concentration in air from photon fluence rate for \(\gamma\) rays involved in abnormal increases of radiation level. Based on the measurements using the point-like sealed radioactive source of \(^{133}\)Ba in the laboratory, activity concentrations in air of each radionuclide would be equivalent to 0.00123 ± 0.00002 Bq cm\(^{-3}\) for \(^{131}\)I and 0.00232 ± 0.00018 Bq cm\(^{-3}\) for \(^{133}\)Xe, respectively. The radioactive concentrations in air of \(^{131}\)I just after the nuclear accident in the FDNPP determined by the filter sample collected by the air sampler were evaluated to be between 2.0 × 10\(^{-6}\) Bq cm\(^{-3}\) and 0.00137 Bq cm\(^{-3}\) in Tokai, Ibaraki Japan.\(^{24}\) The highest values of radioactivity concentration in air of \(^{133}\)Xe were also measured using the LaBr\(_3\) detector and to be in the range of 0.12 Bq cm\(^{-3}\) and 0.032 Bq cm\(^{-3}\) at the expressway in Fukushima prefecture.\(^{12}\) The observed activity concentrations strongly support the assertion that the proposed method can properly determine activity concentration in air of \(^{131}\)I and \(^{133}\)Xe.

Considering the measurement time of the proposed method, the minimum detectable activity concentration in air is much higher than other methods.

| Distance from the \(^{133}\)Ba source (m) | Theoretically estimated \(H^*(10)\) (nSv h\(^{-1}\)) | Evaluated \(H^*(10)\) by the G(E) function method (nSv h\(^{-1}\)) | Evaluated photon fluence rate by the unfolding method (81 keV) (cm\(^{-2}\) s\(^{-1}\)) | Evaluated photon fluence rate by the unfolding method (356 keV) (cm\(^{-2}\) s\(^{-1}\)) |
|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|
| 1.0                             | 85.0                             | 84.7 ± 1.4                       | 2.67 ± 0.20                      | 4.45 ± 0.05                      |
| 1.5                             | 37.8                             | 34.9 ± 0.9                       | 0.917 ± 0.014                    | 1.24 ± 0.05                      |
such as air volume sample method taken in the reference 23. From the viewpoint of the prompt release of the radiological information, the proposed method enables to release the information about radioactivity concentration in air in about 10 min.

For the purpose of the rapid and timely response to the radiological accident, the determination of $^{131}$I and $^{133}$Xe was focused on in the study. A several different species of radionuclides such as $^{134}$Cs and $^{137}$Cs observed in the pulse height spectrum would also be identified and determined by means of the proposed method. Because introducing the unfolding method can minimize the influence of the overlapping of the Compton continuum region to the peak area of interest in the pulse height spectrum.

4. Summary

Simultaneous determination of the ambient dose equivalent rate and activity concentration in air was performed using a newly developed CeBr$_3$ scintillation spectrometer to obtain radiological information more effectively. The G(E) function and the unfolding methods were employed for dosimetry. To determine the activity concentration in air, the photon fluence rate was obtained from the pulse height spectrum by using the unfolding method, and the activity concentrations in air of radioisotopes of interest, mainly of $^{131}$I and $^{133}$Xe, were estimated from the obtained photon fluence rate by applying the conversion coefficient evaluated by MC calculation. The developed method was verified according to the EURADOS procedure for inter-comparison of detectors used for early warning networks. After a thorough performance test, a demonstration of the simultaneous determination of $H^*(10)$, and radioactivity concentration in air was performed using a point-like sealed $^{133}$Ba source that reproduces an artificial increase of radiation level above the natural BG due to the passing of a radioactive plume containing $^{131}$I and $^{133}$Xe.

The results obtained in the thorough performance tests satisfied the requirements of the dosimeter used for radiation protection. The proposed method can detect artificial increases in $H^*(10)$ above the natural background comparable to the value of $H^*(10)$, that is $60\text{ nSv h}^{-1}$ within 4%, and identify the radionuclides involved. The minimum detectable dose equivalent rate was experimentally confirmed to be approximately $10\text{ nSv h}^{-1}$ for $\gamma$ rays from $^{137}$Cs, when measurement duration were to set to be 10 min. The maximum measurable $H^*(10)$ for the CeBr$_3$ scintillation spectrometer was also experimentally verified in the $^{137}$Cs $\gamma$ ray calibration field and evaluated to be $101.3\text{ \mu Sv h}^{-1}$.

To estimate the radioactivity concentration in air of $^{131}$I and $^{133}$Xe from measured pulse height spectrum by the CeBr$_3$ scintillation spectrometer, the conversion coefficient from photon fluence rate to the activity concentration in air was calculated and compared with that obtained by Hirayama et al. for verification. The calculation method was found to be consistent based on a comparison of the calculated results for $^{131}$I. The photon fluence rates obtained from the measured pulse height spectra due to $\gamma$ rays from $^{133}$Ba, which mimics artificial increases of radiation levels due to $^{131}$I and $^{133}$Xe, were evaluated properly by the unfolding method. The photon fluence rates of $\gamma$ rays with energies of 356 keV and 81 keV were then determined to be $1.24\pm0.06\text{ cm}^{-2}\text{s}^{-1}$ and $0.917\pm0.014\text{ cm}^{-2}\text{s}^{-1}$, respectively. If radioactive plumes containing $^{131}$I and $^{133}$Xe were to be involved, these would be equivalent to $(3.44\pm0.17)\times10^{-4}\text{ Bq cm}^{-3}$ for $^{131}$I and $(7.98\pm0.12)\times10^{-4}\text{ Bq cm}^{-3}$ for $^{133}$Xe. The results strongly support the assertion that the proposed method could properly estimate the activity concentration in air of $^{131}$I and $^{133}$Xe during emergency.

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要　旨
環境放射線モニタリングにおける線量当量率及び空気中放射性物質濃度の同時決定法に関する検討

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原子力事故時等緊急時を想定した環境放射線モニタリングを確実に行うため、近年開発されたCeBr$_3$シンチレーション検出器を用い、波高分布の一回の測定結果から、核種同定に加えて、空気中放射能濃度及び線量当量率の同時評価法を開発した。開発された手法は、エネルギー応答、線量率直線性及び角度依存性について、欧州放射線防護グループ（EURADOS）において実施される環境放射線モニタリング用線量計の特性試験で実施される試験法に従って行った。試験の結果、著者らが開発した手法が、国際規格（IEC）及びJIS規格で示された基準（エネルギー及び角度応答特性で−29%〜+67%）に対し、基準を十分に満たす性能が示された。さらに、$^{133}$Ba点源を用い、特に$^{131}$I及び$^{133}$Xeを含んだ放射性プルーム通過による線量率増加及びそれらの空気中放射能濃度についても適切に評価できるか、実験室内での測定で検証した。その結果、著者らの開発した手法で得られた光子フルエンス率に、モンテカルロ計算コード PHITS で得られた換算係数を適応することで、妥当な空気中放射能濃度を推定できることが示された。