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# Attempt to estimate the background pulse height spectrum of the CeBr<sub>3</sub> scintillation spectrometer due to terrestrial gamma ray components: Application in environmental radiation monitoring

Munehiko Kowatari <sup>a,d,\*</sup>, Yoshihiko Tanimura <sup>a</sup>, Patrick Kessler <sup>b</sup>, Annette Röttger <sup>c</sup>

- a Department of Radiation Protection, Nuclear Science Research Institute, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki, 319-1195, Japan
- b Department of Radiation Oncology, University of Wurzburg, Josef-Schneider-Str. 11, 97080, Wurzburg, Germany
- <sup>c</sup> Physikalisch-Technische Bundesanstalt, Bundesallee 100, Braunschweig, 38116, Germany
- d Center for Advanced Radiation Emergency Medicine, National Institutes for Quantum and Radiological Science and Technology, 4-9-1 Anagawa, Inage-ku, Chiba, 263-8555, Japan

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### ABSTRACT

This article describes an attempt to estimate the background (BG) pulse height spectrum (PHS) of the CeBr<sub>3</sub> scintillation spectrometer due to terrestrial gamma ray components. In radiological emergencies, there are some cases which the prior measurement of the background components can't be made. By subtracting the BG components from the measured gross PHS, timely and accurate information about the artificial radionuclides involved in the emergencies would be effectively drawn to protect the member of the public and first responders. A Monte Carlo calculation was conducted to simulate the PHS by gamma rays from natural radionuclides uniformly distributed in the ground. The calculated PHS were verified to be consistent with measured PHS. Derived dose equivalent rate from the calculated PHS were compared with reference value and found to be in good agreement with each other. The study revealed that applying the calculated PHS to the in-situ gamma ray spectrometry components hardly affected the determination of surface deposition density on soil for radioactive resium

# 1. Introduction

The authors demonstrate the simulation of the pulse height spectrum (PHS) obtained by the  $CeBr_3$  scintillation spectrometer used for emergency radiation monitoring. During radiological emergencies, timely and reliable radiological information such as surface contamination density on soil and dose equivalent rate,  $H^*(10)$ , in affected areas should be measured. Prior measurements of the background components (BG) before an emergency is important to properly estimate how much the artificial radionuclides are involved in an artificial increase of dose equivalent rate. However, the BG measurement isn't in general implemented before a radiological emergency as long as the  $H^*(10)$  wasn't periodically monitored at measurement points. It would be informative if the BG components of the PHS obtained from the scintillation spectrometer used for emergency radiation monitoring could be estimated without the prior measurement.

In the study, the authors attempted to reconstruct the PHS of the CeBr<sub>3</sub> scintillation spectrometer due to natural radionuclides by means of a Monte Carlo (MC) calculation. From the calculated PHS, we estimated the  $H^{*}(10)$  and compare it with the reference  $H^{*}(10)$  obtained by the well-calibrated ionization chamber at the reference site for verification of the proposed method. After the verification of the method, we applied the proposed method to the determination of the surface contamination density on soil. An in-situ gamma ray spectrometry at the field of Japan Atomic Energy Agency (JAEA) was performed, for estimating surface contamination densities on soil of <sup>134</sup>Cs and <sup>137</sup>Cs. By subtracting the estimated BG from the "gross" PHS obtained by in-situ gamma ray spectrometry, the "net" PHS due to radioactive Cs were calculated. The derived surface contamination densities on soil of <sup>134</sup>Cs and <sup>137</sup>Cs from both "gross" and "net" PHS were then compared if the subtraction of the calculated BG component hamper the proper determination of the surface contamination density on soil.

E-mail address: kowatari.munehiko@qst.go.jp (M. Kowatari).

<sup>\*</sup> Corresponding author. Center for Advanced Radiation Emergency Medicine, National Institutes for Quantum and Radiological Science and Technology, 4-9-1 Anagawa, Inage-ku, Chiba, 263-8555, Japan.

A CeBr $_3$  scintillation crystal employed in the study has an excellent energy resolution. A LaBr $_3$ (Ce) scintillation crystal shows higher detection efficiency and energy resolution compared to a conventional NaI(Tl) scintillation crystal (Quarati, F.G.A., et al., 2013). However, the presence of the radioactive contaminant,  $^{138}$ La in the LaBr $_3$ (Ce) crystal leads the higher PHS due to the inherent background component. This keeps the LaBr $_3$ (Ce) detector away from the introduction of the environmental radiation measurement. Instead of the LaBr $_3$ (Ce) detector, CeBr $_3$  scintillation spectrometer were selected for the determination of activity concentration in sea water (Tsabaris et al., 2019). This is because they have relatively lower self-contamination (Lutter et al., 2013, Quarati, F.G.A., et al., 2013) and similar energy resolution, compared with a LaBr $_3$ (Ce) scintillation detector.

### 2. Materials and methods

#### 2.1. Instrument and in-situ gamma ray spectrometry

We employed a cylindrical CeBr $_3$  scintillation spectrometer (OKEN 44/DM:CeBr $_3$ ) 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) shown in Fig. 1. Compared to a conventional NaI(Tl) scintillation detector (around 7% relative energy resolution for 662-keV gamma rays (W.E. van Eijk., 2001)), the CeBr $_3$  scintillation spectrometer has better energy resolution (4–5% for a 662-keV gamma ray (Quarati, F.G.A., et al., 2013)) and can distinguish between the events from each radioactive Cs, such as 605-keV photons from  $^{134}$ Cs to 662-keV photons from  $^{137}$ Cs (Kowatari et al., 2019). Moreover, the measurement system used comprises of simple and light units of a scintillation spectrometer with MCA and turns out to be an easy-to-handle system and suitable for radiological emergency. In addition, no external power supply and no coolant such as LN $_2$  are required.

# 2.2. Dosimetry and in-situ gamma ray spectrometry

The authors have investigated the applicability of newly developed scintillation detectors to in-situ gamma ray spectrometry (Kowatari et al., 2015, 2019). A newly developed scintillation spectrometer, CeBr<sub>3</sub> is revealed to be a promising candidate for the purpose, because of its simplified measurement system and its energy resolution enough to distinguish peaks due to gamma rays from  $^{134}\mathrm{Cs}$  and  $^{137}\mathrm{Cs}$  (its progeny,  $^{137\mathrm{m}}\mathrm{Ba}$ ). In the same study, we developed the method to estimate Air kerma rate and  $H^*(10)$  from the measured PHS by means of the so-called G(E) function method and the unfolding method (Kowatari et al., 2019).



Fig. 1. The cylindrical CeBr $_3$  scintillation spectrometer with dimension of 2.54 cm  $\times$  2.54 cm with the PC-driven multi-channel Analyzer.

Before measuring the  $H^*(10)$ , both methods were thoroughly verified in accredited underground gamma ray reference fields operated in German national metrological laboratory, Physikalisch-Technische Bundesanstalt (PTB) (Dobrowski and Neumaier, 2010; Kowatari et al., 2010). The results obtained though the test was found to be consistent with the reference values and reported to another article (Kowatari et al., 2020). The G(E) function for the CeBr<sub>3</sub> scintillation detector enable to directly estimate air kerma rate or  $H^*(10)$  from the measured PHS, which was evaluated from calculated response functions using EGS4 code (Nelson et al., 1985). In order to apply the unfolding method to environmental radiation monitoring, the response matrix and the appropriate unfolding code were also prepared prior to environmental measurement. Response matrix for the CeBr3 scintillation detectors was calculated using the Monte Carlo calculation code, MCNP-4C (Briesmeister, 2000). Photon fluence spectrum from the spectrometer was evaluated by unfolding the measured spectra using MAEXD (Reginatto et al., 2002) code and Air kerma rate and  $H^*(10)$  were then estimated by multiplying the photon fluence rate with fluence-to-air-kerma conversion coefficients provided by ICRP 74 (ICRP, 1996).

The in-situ gamma ray spectrometry using the CeBr $_3$  scintillation spectrometer was performed in the same manner as the ICRU 53 (ICRU, 1994). The CeBr $_3$  scintillation spectrometer coupled with a PC-driven MCA was fixed at a height of 1.0 m from ground surface in an open and plain field in Tokai Research Branch of JAEA. Measurement time was set to 7200 s. The conventional peak area method was chosen to estimate surface contamination densities on soil for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from the obtained PHS. The surface deposition density on soil,  $A_a$  (Bq cm $^{-2}$ ) was determined by dividing a net peak counting rate,  $C_f$  (cps) by a peak counting rate per activity,  $N_f/A_a$  (cps (Bq cm $^{-2}$ ) $^{-1}$ ), according to the following Eq. (1)-A and (1)-B.

$$A_a = C_f / \left( N_f / A_a \right), \tag{1A}$$

and

$$\left(N_{f}/A_{a}\right) = \left(\frac{N_{f}}{F_{\phi}} \times \phi_{Aa}\right) \tag{1B}$$

The peak efficiency of the  $CeBr_3$  scintillation spectrometer was obtained by measuring PHS by gamma rays from point sources ( $^{152}Eu$ ,  $^{133}Ba$  and  $^{137}Cs$ ) and shown in Fig. 2. The uncertainties of each peak efficiency were evaluated by considering counting errors of each gamma ray and uncertainties of each source activity (c.a., 2%, k=1). Measurements were performed at the height of 1.0 m from the point source

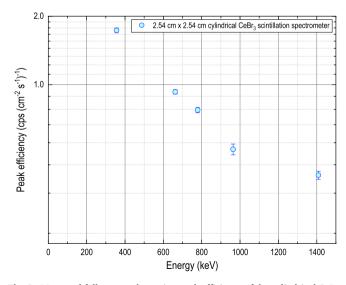


Fig. 2. Measured full energy absorption peak efficiency of the cylindrical  ${\sf CeBr}_3$  scintillation spectrometer.

put on the floor. Peak counting rates per activity,  $N_f/A_a$  for each point sources were derived from multiplying  $N_f/F_{\varphi}$  with photon fluence rate per unit surface deposition density,  $\varphi_{Aa}$  (photons cm<sup>-2</sup> s<sup>-1</sup> (Bq cm<sup>-2</sup>)<sup>-1</sup>). Values  $\varphi_{Aa}$  are independent of the detector used and taken from Table A.1 in ICRU 53.

$$A_a = F_{\phi/\phi_{Aa}},\tag{2}$$

To evaluate the surface contamination density on soil for  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ , the migration from the ground surface and the depth profile of radioactive Cs in soil should be accounted for. The parameter called the relaxation mass per unit area,  $\beta$  (g cm $^{-2}$ ), is introduced, to express the depth profile of radioactive Cs in soil. In the study,  $\beta$  was set to 5, because almost nine years have passed since initial deposition of radionuclides took place at the measurement date. The uncertainties of the obtained surface contamination densities on soil for  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  from the gross and net PHS were calculated by considering the counting errors of each peak and uncertainties accompanying each peak efficiency.

# 2.3. Calculation and verification of the PHS of the $CeBr_3$ scintillation spectrometer

The PHS due to gamma rays from each natural radionuclide, such as  $^{40}$ K, U-series and Th-series was estimated by simulating the gamma ray fluence rate at the height of 1.0 m from the ground using the Monte Carlo (MC) calculation code called PHITS (Sato et al., 2013). To verify the simulated PHS due to natural radionuclides, we compared the simulated PHS with the measured PHS at the environmental reference site operated by Physikalisch-Technische Bundesanstalt (PTB).

To estimate the PHS of the  $CeBr_3$  due to the terrestrial gamma rays from natural radionuclides, gamma ray fluence rate spectra at the height of 1.0 m for natural radionuclides in soil per unit radioactivity concentration in soil were estimated. The gamma ray fluence rate per unit activity concentration in soil was calculated, by assuming the ground was treated as a cylinder with a diameter of 4 km and a depth of 20 m with uniform distribution of radionuclides. 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 (Lemercier et al., 2008; Namito et al., 2012). A linear source measuring 20 m in length from the bottom to the surface of the ground was selected and a surface detector with a diameter of 4 km was set at the height of 1.0 m.

The responses of the CeBr $_3$  scintillation spectrometer for each radionuclide were then estimated. All calculated responses were summed up and fitted to the measured PHS by means of the least square method. The PHS due to gamma rays at 1.0 m height of the ground from each natural radionuclide were estimated also by the MC calculation. The CeBr $_3$  scintillation spectrometer was modelled in detail and the response of the spectrometer was obtained in the unit of counting rate per activity concentration in soil (cps (Bq g $^{-1}$ )) $^{-1}$ ) for each natural radionuclide. Potassium-40 ( $^{40}$ K), U-series ( $^{226}$ Ra,  $^{214}$ Bi,  $^{214}$ Pb, and  $^{210}$ Pb) and Th-series ( $^{228}$ Ac,  $^{212}$ Bi,  $^{212}$ Pb, and  $^{208}$ Tl) are taken into consideration. Both U- and Th-series were assumed to keep radioactive equilibrium in the MC calculation and activity concentrations in soil for each radionuclide follows their branch ratios. The calculated PHS for each radionuclide were summed up and compared with the measured PHS. The simulated PHS were fitted by the least square method to reproduce the measured PHS. The inherent and cosmic-ray components were also accounted for the synthesized PHS.

In addition to natural BG components, the inherent and cosmic-ray components are included in the measured PHS. The inherent background of the CeBr<sub>3</sub> scintillation spectrometer was determined by an overnight measurement (15 h) in the underground laboratory operated by PTB (Dombrowski and Neumaier, 2010). Inside the underground laboratory, the ambient dose equivalent rate is estimated to be around

 $1.5~{\rm nSv~h^{-1}}$ , and the radon concentration in air was of the order of 3 Bq m $^{-3}$ . The influence of the secondary cosmic-ray component was determined on a platform floating on a lake. No significant terrestrial gamma rays were presented there (Dombrowski et al., 2009). The measured PHS on a floating platform was also accounted for the simulated PHS.

### 3. Results and discussions

# 3.1. Synthesis of the PHS of natural BG component

The PHS due to terrestrial gamma rays was estimated by synthesizing the PHS due to gamma rays emitted from each natural radionuclide using the MC. To verify the simulated PHS by the proposed method, the PHS was compared with measured natural BG obtained at the reference site in PTB. Before the environmental measurement at the reference site. the influence of the secondary cosmic-ray component to the PHS was assessed. The corresponding ambient dose equivalent rate,  $H^*(10)$  was estimated to be 2.5 nSv h<sup>-1</sup>. From the measured results in the underground laboratory, the  $H^*(10)$  due to the inherent component was also estimated to be 9.18 nSv h<sup>-1</sup>. The scintillation crystal of CeBr<sub>3</sub> is known to contain a trace amount of radioactive <sup>227</sup>Ac, which can be ascribed to the crystalline fabrication process (Quarati et al., 2013). The total count rate per volume was estimated to be 0.12 counts/s/cm<sup>3</sup>, which was more than two times larger than that obtained by Quarati, F.G.A., et al., 2013. The total count rates due to the self-contamination ranged from 0.022  $\pm$  $0.001 \text{ counts/s/cm}^3 \text{ to } 0.051 \pm 0.004 \text{ counts/s/cm}^3$ . The total count rate varies each crystal by each crystal. The measured results of the same type of scintillator crystal was determined to be 9.8 nSv h<sup>-1</sup> in terms of the ambient dose equivalent rate (Kessler et al., 2017). Comparing the pulse height spectrum due to the inherent background with that obtained by the Guss et al. (Guss, et al., 2009), peaks around 2 MeV (0.002 cps) were almost twice as shown in Figure 7 (0.001 cps). Considering the difference of the contamination level of each crystal reported by Quarati et al., the evaluated inherent component of the CeBr<sub>3</sub> scintillation spectrometer was found to be consistent.

Fig. 3 shows the measured PHS of natural BG component at the reference site in PTB. There is no peak due to artificial radionuclides identified in the measured PHS. In Fig. 3, the synthesized PHS by the MC calculations is superimposed. The measured and synthesized PHS were found to be in good agreement with each other. The PHS due to self-contamination component would hamper the PHS simulation. It should be pointed out that a contamination-free scintillation spectrometer such as NaI(Tl) and CsI(Tl) would be chosen if the proposed method was more properly applied.

Table 1 lists estimated activity concentrations in soil for natural

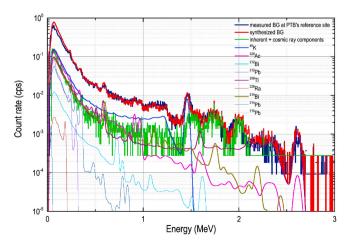


Fig. 3. The measured and simulated pulse height spectra of the cylindrical  $CeBr_3$  scintillation spectrometer. Measurement of the PHS were performed in the environmental reference site in PTB.

radionuclides for  $^{40}$ K, U-series and Th-series. These were obtained from the least square method applied of the calculated PHS to the measured PHS. By multiplying nominal activity concentration in soil shown in Table 1 with air kerma rate per unit activity per unit mass in the table 5.2 in ICRU 53, air kerma rate due to natural BG was estimated to be 34 nGy  $\,h^{-1}$ . The air kerma rate obtained from the measured PHS was evaluated to be 38.4  $\pm$  2.2 nGy  $\,h^{-1}$ , by applying the G(E) function method (Moriuchi and Miyanaga, 1966). Both were in good agreement, considering the uncertainties accompanying the measured PHS and estimated nominal activity concentrations in soil.

Table 2 summarizes the comparison of the  $H^*(10)$  due to gamma rays from natural radionuclides obtained at the reference site in PTB. By excluding the inherent and secondary cosmic-ray components, the  $H^*(10)$  due to gamma rays from natural radionuclides was evaluated to be  $49.4 \pm 0.8$  nSv h $^{-1}$  from the measured PHS. The result is consistent with the reference value ( $51.8 \pm 4.0$  nSv h $^{-1}$ ) measured using the GE Reuter Stokes ionization chamber. As shown in Table 2, the  $H^*(10)$  due to BG components was also estimated from the simulated PHS by applying the G(E) function method. The  $H^*(10)$  was derived to be  $51.0 \pm 0.2$  nSv h $^{-1}$  from the synthesized PHS. Only statistical errors accompanying the MC calculation were accounting for the uncertainty of the  $H^*(10)$  from the simulated PHS. The results imply that the proposed method can quantitively reproduce the PHS from terrestrial gamma rays as a BG component.

# 3.2. Application of the synthesized BG component to the determination of surface contamination density on soil of radioactive Cs

The authors attempt to apply the proposed method to the in-situ gamma ray spectrometry. Fig. 4 shows the measured and synthesized PHS by the proposed method. The synthesized PHS due to natural BG component without <sup>134</sup>Cs and <sup>137</sup>Cs were then simulated by the verified method. Comparing Fig. 3 obtained in the environmental reference site in PTB, peaks due to radioactive Cs were clearly shown in the measured PHS obtained by the in-situ gamma ray spectrometry performed in the field in JAEA. Fig. 5(A) and (B) and (C) also shows the comparison of peak areas of the measured, synthesized and subtracted PHS for each radioactive Cs and <sup>40</sup>K. Only the measured and the synthesized PHS were shown in the peak areas of 40K. From both measured and subtracted PHS, surface contamination densities on soil for  $^{134}\mathrm{Cs}$  and  $^{137}\mathrm{Cs}$ were then determined by the peak area method. Counting errors for each peak of radioactive Cs and the uncertainty of peak efficiency curve were accounted for the uncertainty of surface contamination density on soil. Results were listed in Table 3. The obtained surface contamination densities on soil of radioactive Cs were almost identical to the results obtained by in-situ gamma ray spectrometry using the HP-Ge detector.

The subtraction of the BG PHS from the "gross" measured PHS hardly affect the determination of surface contamination density on soil of  $^{137}$ Cs. However, the results of surface contamination densities on soil for  $^{134}$ Cs were found to be different. This can be explained by the decline of the baseline of the BG of the PHS. The baseline obtained by measurement fluctuates according to the statistical errors from the nature of

**Table 1** The estimated activity concentrations in soil for natural radionuclides for  $^{40}$ K, Useries and Th-series.

Radionuclide	Estimated activity concentration in soil <sup>a</sup> (Bq kg <sup>-1</sup> )	Air kerma rate per unit activity per unit mass from ICRU 53 $\mu \text{Gy } h^{-1}$ per Bq g $^{-1}$
<sup>40</sup> K	430	0.0417
U-series	5.2	$0.462^{b}$
Th-series	22	$0.604^{b}$

<sup>&</sup>lt;sup>a</sup> Values were estimated from the fitting by the least square method by calculated PHS.

Table 2 Comparison of measured and estimated ambient dose equivalent rates at the reference site in PTB. (k=1).

Reference $\dot{H}^{*}(10)$ (nSv h <sup>-1</sup> )	Measured $\overset{.}{H}^{*}(10)$ by the unfolding method (nSv $h^{-1}$ )	Estimated $\dot{H}^*(10)$ from the synthesized PHS (nSv $h^{-1}$ )
$51.8 \pm 4.0$	$49.4 \pm 0.8$	$51.0\pm0.2$

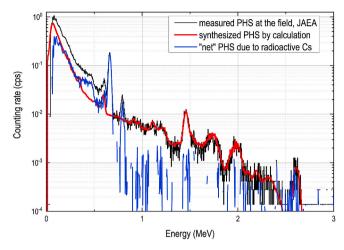


Fig. 4. Measured pulse height spectrum at the field in JAEA by the in-situ gamma ray spectrometry.

radiation measurement. On the other hand, the synthesized PHS of the natural BG component can be ideally estimated regardless of measurement time. In some case, the baseline with energy around 800 keV would be excessively subtracted, which might lead to wrongly determine the surface contamination density on soil for  $^{134}\mathrm{Cs}.$ 

The minimum detectable activities (MDAs) for each radioactive Cs were calculated by the following equations in the ISO 11929 (ISO, 2019);

$$MDA\left(Bq\ cm^{-2}\right) = L_D/\left(N_f/A_a\right),\tag{3a}$$

and

$$L_D(cps) = 2k \times \sqrt{n_0 \left(\frac{1}{t_g} + \frac{1}{t_0}\right)} + \frac{k^2}{t_g},$$
 (3b)

where, L<sub>D</sub> (cps): detectable limit taken from the ISO 11929 (ISO, 2019).

 $n_0$  (cps): the net counting rate of the BG region

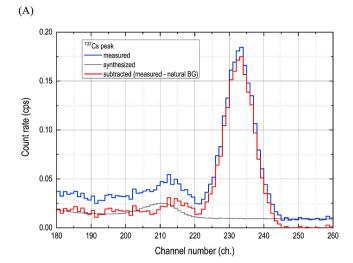
k: coverage factor (this study k = 1.645).

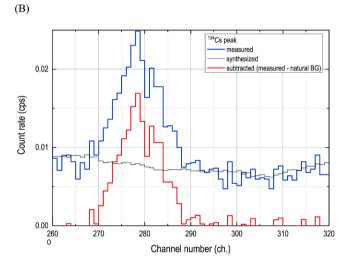
 $t_{\sigma}$  (sec): the measurement time of the BG (this study 7200 s).

 $t_0$  (sec): the measurement time (this study 7200 s).

The MDAs for each radioactive Cs were summarized in Table 4. The MDAs of surface contamination density on soil for <sup>134</sup>Cs and <sup>137</sup>Cs were derived from the synthesized PHS and listed in the table. For each radioactive Cs, the MDAs obtained by measured and synthesized PHS were in good agreement with each other, considering their uncertainties. This implies that the MDAs estimated from the measured PHS might be less affected by radioactive Cs themselves in this measurement. The comparison of MDA of radioactivity concentration in soil for <sup>40</sup>K was listed in the same table. It should be noted that the inherent component affects the determination of activity and ant the MDA for <sup>40</sup>K. The influence of the inherent component to the determination of  $^{40}$ K peak area was reported (Tsabaris et al., 2019). Counting rate due to the intrinsic self-contamination of the CeBr3 crystal (5.08-cm dia. and 5.08-cm length) was estimated by subtracting the counting rates due to  $^{40}$ K and  $^{214}$ Bi from the total counting rate of energy window for  $^{40}$ K peak area. For the estimation of the MDA for terrestrial background

<sup>&</sup>lt;sup>b</sup> Radioactive equilibrium is assumed, and the total amount of its decay products is accounted.





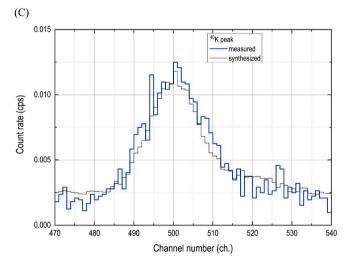


Fig. 5. (A), (B) and (C) Comparison of the peak areas of the measured, synthesized and subtracted PHS in the energy regions of radioactive Cs and  $^{40}$ K.

component, the influence of the inherent component of the  $CeBr_3$  detector should be accounted. Introducing the other type of scintillation spectrometers with low self-contamination such as  $SrI_2$  and GGAG or the CdZnTe semiconductor spectrometer would also be preferable for the purpose of low counting-rate measurement of environmental radiation monitoring.

Radionuclide	Surface contamination density estimated from the measured "gross" PHS (Bq cm <sup>-2</sup> )	Surface contamination density estimated from the subtracted "net" PHS (Bq $\mathrm{cm}^{-2}$ )
<sup>137</sup> Cs <sup>134</sup> Cs	$3.94 \pm 0.06$ $0.46 \pm 0.03$	$\begin{array}{c} 3.85 \pm 0.05 \\ 0.38 \pm 0.02 \end{array}$

**Table 4** Comparison of minimum detectable activities (MDA) of surface contamination density on soil for each radioactive Cs and radioactivity concentration in soil of  $^{40}$ K. The MDAs were calculated according to the ISO 11989-1. (k=1).

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Radionuclide	MDA estimated from the measured "gross" PHS (Bq cm <sup>-2</sup> )	MDA estimated from the synthesized PHS (Bq cm <sup>-2</sup> )
<sup>137</sup> Cs <sup>134</sup> Cs	$\begin{array}{c} 0.069 \pm 0.011 \\ 0.064 \pm 0.011 \end{array}$	$\begin{array}{c} 0.068 \pm 0.011 \\ 0.065 \pm 0.011 \end{array}$
Radionuclide <sup>40</sup> K	MDA estimated from the measured "gross" PHS (Bq g $^{-1}$ ) $0.047\pm0.010$	MDA estimated from the synthesized PHS (Bq g $^{-1}$ ) 0.049 $\pm$ 0.010

The MDAs obtained by measured and synthesized PHS were in good agreement with each other as well. This showed that the proposed method could properly reproduce the PHS which may be obtained from the CeBr<sub>3</sub> scintillation spectrometer. These also support that the proposed method might help derive a proper MDA for each artificial radionuclide without any knowledge of natural BG components.

## 4. Summary and limitation of the study

The study mainly focuses on the estimation of contamination level by unknown BG during a radiological emergency. No information about the natural BG components were available if prior measurement of the BG was conducted. The authors tried to estimate the pulse height spectrum (PHS) due to natural BG components of the CeBr<sub>3</sub> scintillation spectrometer, by performing the MC calculations. For verification, the synthesized PHS due to the natural BG component was then compared with measured PHS obtained at the reference site for environmental radiation monitoring operated by the PTB. The synthesized PHS were in quite good agreement with measured PHS. The inherent component due to self-contamination of the CeBr<sub>3</sub> spectrometer is found to be appropriately accounted for estimation.

After verification, we applied the synthesized PHS derived from the proposed method to determine the surface contamination density on soil for radioactive Cs for in-situ gamma ray spectrometry. In the measured PHS obtained at the field in JAEA, peaks for radioactive Cs were clearly identified. By subtracting the synthesized PHS due to terrestrial gamma rays from the measured gross PHS, the net PHS due to gamma rays from radioactive Cs was derived. Form the peak area method, the determined surface contamination densities on soil for <sup>137</sup>Cs from each PHS were found to be in good agreement with each other. This also supports that the proposed method is applicable to estimate the PHS without any prior knowledge of the natural BG component.

The MDAs of surface contamination density on soil for  $^{134}$ Cs and  $^{137}$ Cs were also derived from the synthesized PHS obtained from the MC calculation. The MDAs for each radioactive Cs estimated from measured and simulated PHS were in good agreement with each other. In this measurement, the MDAs estimated from the measured PHS might be less affected by radioactive Cs themselves. The results also show that the simulated PHS might give information about the BG without any influence due to artificial radionuclides.

In an emergency, background measurement is difficult owing to interference by the artificial radionuclides to be monitored. By applying the proposed method, the influence of the BG component to the measured PHS would be minimized in radiological emergencies. In the

study, we successfully subtracted the synthesized PHS from the gross PHS measured in the in-situ gamma ray spectrometry and the components due to radioactive Cs were clearly identified. Future challenges of the proposed method include 1) investigation about applicability of the proposed method to other artificial radionuclides such as <sup>131</sup>I on the ground, 2) estimation of the BG components from the concrete or asphalt surface and 3) application to the areal or on-vehicle emergency radiation monitoring using a spectrometer.

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# Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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