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Apparatus development for measurement of ^{134}Cs and ^{137}Cs radioactivity of soil contaminated by the Fukushima Daiichi Nuclear Power Plant accident

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Abstract

We developed an apparatus containing a NaI(Tl) scintillator to measure the ^{134}Cs and ^{137}Cs radioactivity of soil contaminated by the Fukushima Daiichi Nuclear Power Plant accident. The unfolding method with the least-squares technique was used to determine the radioactivity. Detector responses for each radionuclide in soil were calculated with EGS5 code for the unfolding method. The radionuclides that were measured were ^{40}K , ^{134}Cs , ^{137}Cs , ^{208}Tl , ^{214}Bi , and ^{228}Ac . The measured spectrum agreed well with the spectrum calculated from the response matrix and measured radioactivities. The unfolding method allows us to use the NaI(Tl) scintillator despite the overlap of peaks.

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1. Introduction

Large amounts of radionuclides were released into the atmosphere by the Fukushima Daiichi Nuclear Power Plant accident that occurred on March 11, 2011. Released radionuclides were deposited over a wide area from the Tohoku region to the northern Kanto region (Endo et al., 2012; Saito et al., 2015; Steinhauser et al., 2014; Kinoshita et al., 2011; Yasunari et al., 2011). Deposited radionuclides have caused high dose rates.

To decrease the dose rate in populated areas, surface soil has been removed as one of the decontamination methods. The volume of removed materials, including soil, was estimated to be 150–280 Mm³ by the Japanese ministry of the Environment (Ministry of the Environment, 2011). The soil will be divided into groups based on the radioactivity concentration and stored separately. A detector system is required for measuring the radioactivity concentration of this huge amount of soil. Measured radionuclides are ^{134}Cs and ^{137}Cs because the radionuclides have not yet decayed out whereas short-lived radionuclides such as ^{129}Te , ^{129m}Te , ^{131}I , and ^{132}I , have already disappeared.

Gamma-ray spectroscopy can be used to measure the radioactivity of ^{134}Cs and ^{137}Cs , which are γ -ray emitting radionuclides. One of the simple methods is the derivation from counts of each full energy peak by γ -rays in a measured spectrum. However, a detector with a sufficiently high energy resolution is essential to prevent the peaks from overlapping.

The unfolding method (Knoll, 2010) could be also used to measure the radioactivity. Although the unfolding method makes the measurements more complex, it does not matter if the peaks overlap. Thus, a detector with poor energy resolution could be used, which would reduce manufacturing and maintenance costs.

We developed an apparatus to measure the radioactivity concentration of the radionuclides ^{134}Cs and ^{137}Cs in soil with a uniform radioactivity distribution. In this paper, we mainly report a method for determining the radioactivity with this apparatus. The unfolding method with the least-squares technique was used to determine the radioactivities despite the overlap of peaks arising from the poor energy resolution of the scintillator. The measured radioactivities were compared with those measured with a high-purity germanium (HPGe) detector to verify the accuracy of the method.

2. Measurement principle

Soil with a uniform radioactivity distribution can be measured with the apparatus. To obtain uniform soil samples, an industrial mixer is placed at the front of the apparatus, and mixed soil is fed into the apparatus. The weight and volume of the soil were measured before the radioactivity was measured.

A simple sectional view of the apparatus is shown in Fig. 1. A $\phi 76.2 \times 76.2 \text{ mm}^3$ NaI(Tl) scintillator connected to a photomultiplier is used as the γ -ray detector. The soil volume in the apparatus is more than 0.240 m^3 . Because the detector is surrounded by the soil, the solid angle of the detector is large. Thus, a large amount of γ -rays from the soil can be detected in a

short time. Furthermore, the soil behaves as a shielding material to γ -rays from outside of the apparatus. The transmission rate is calculated to be 0.16 for 0.662 MeV γ -rays to the soil with a density of 1.0 g/cm³ and a thickness of 24.2 cm, which is the minimum effective thickness of the soil in the apparatus. The transmission rate of the apparatus is more decreased by the construction materials of the apparatus.

The radioactivities of radionuclides in the soil are determined by using the unfolding method with the least-squares technique (Knoll, 2010; Perey, 1978; Matzke, 2002). The radioactivity of the i th radionuclide included in the soil, A_i , and the count rate of the j th channel of measured spectrum, N_j , are related by

$$N_j = \sum_i R_{ij} A_i, \quad (1)$$

where R is the response matrix and R with a variable j is the detector response for the i th radionuclide. The radionuclides measured are ⁴⁰K, ¹³⁴Cs, ¹³⁷Cs, ²⁰⁸Tl, ²¹⁴Bi, and ²²⁸Ac. To obtain the radioactivities, the simple least-squares technique was used. Chi-squared, χ^2 , is written as

$$\chi^2 = \sum_j \left(\frac{N_j - \sum_i R_{ij} A_i}{\sigma_{N_j}} \right)^2, \quad (2)$$

where σ_{N_j} is the statistical uncertainty of the count rate of the j th channel in the measured spectrum. The fitting region of the measured spectrum, indicated by subscript j , was limited from 100 channels (0.5 MeV) to 550 channels (2.9 MeV). The radioactivities gave initial values before they were measured. In order to minimize the chi-squared, Minuit (James and Roos, 1975) implemented in ROOT (Antcheva et al., 2009) was used.

3. Detector responses for each radionuclide

EGS5 code (Hirayama et al., 2005) was used to calculate the detector response for each radionuclide in soil. In the particle transport and reaction for the calculations, the options for the Rayleigh scattering, photoelectron angle distribution, K and L-edge fluorescence, and K and L-Augur electrons were enabled. The cutoff energy for photons, electrons, and positrons was set to 1 keV.

The detector responses for each radionuclide in the soil were calculated. In addition to ^{134}Cs and ^{137}Cs , the four natural soil radionuclides ^{40}K , ^{208}Tl , ^{214}Bi , and ^{228}Ac were also considered in the detector response because of their large contribution to the fitted region of the spectrum. Because ^{134}Cs , ^{208}Tl , ^{214}Bi , and ^{228}Ac emit cascade γ -rays simultaneously, this emission was reproduced by tracing the decay scheme to include the coincidence summing effect (Kajimoto et al., 2015). The current decay schemes for ^{134}Cs (Sonzogni, 2004), ^{208}Tl (Martin, 2007), ^{214}Bi (Wu, 2009), and ^{228}Ac (Abusaleem, 2014) were implemented in EGS5 code.

The constructed geometry is shown in Fig. 1. The γ -rays were generated in the soil. The source positions were decided at random, and the source probability per soil volume was set as constant.

The detector responses were determined from the distribution of deposition energy to the scintillator. The deposition energy for each source was calculated. To consider the energy resolution of the detector, the deposition energy was recalculated using a random in accordance with a Gaussian function. Only the statistical uncertainty of the number of photoelectrons was considered as the contribution to the energy resolution. The standard

deviation, σ , of the Gaussian function was determined as

$$\sigma = \frac{k}{\sqrt{E}}, \quad (3)$$

where E is the deposition energy (MeV) in the scintillator, and k is a constant. The value of k was determined from the measurement of the full width at half maximum with γ -rays sources, and was 2.7×10^{-2} . As an example, the energy resolution at 0.662 MeV was 5%. The deposition energy was converted to the channel number in the measurement. For the conversion, a calibration curve was obtained based on photopeaks in a measured spectrum. The photopeaks were 0.662 (^{137}Cs), 0.796 and 0.802 (^{134}Cs), 1.461 (^{40}K), and 2.615 (^{208}Tl) MeV.

Figure 2 shows the calculated detector responses for each radionuclide. The shapes of each detector response were different. The shape difference facilitates the determination of radioactivities.

4. Measurement by a HPGe detector

The radioactivities of soil measured with the apparatus were also measured with an n-type coaxial HPGe detector (EG&G Ortec, GMX30200P) to verify the accuracy of the proposed apparatus. The HPGe detector was surrounded by lead blocks and iron shielding to shield the detectors from background radiation.

The soil (50–70 g) was encased in a U9 type container. The radioactivities were measured by placing the U9 container on the endcap of the HPGe detector.

The statistical and systematic uncertainties were considered as the error of the measured values. The statistical error for the counts of peaks of interest

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116 was lower than 0.7%. The error was dominated by systematic uncertainties
117 because radioactivities in a standard volume source used to measure full-
118 energy peak efficiency have an uncertainty of about 5% at a 95% confidence
119 level. The uncertainty was about 3% at one sigma.

120 **5. Results and discussion**

121 Figure 3 shows a measured spectrum, which exhibits statistical uncer-
122 tainty. The measurement time was 300 s. The 0.605 (^{134}Cs) and 0.662 (^{137}Cs)
123 MeV peaks overlap because of the poor energy resolution of the NaI(Tl) scin-
124 tillator.

125 For comparison, the spectrum calculated from the response matrix and
126 measured radioactivities based on Eq. (1) is also shown in Fig. 3. The
127 calculated spectrum agreed well with the measured data between 100 and 550
128 channels because the detector response for natural radionuclides was added to
129 the response matrix. These results suggest that the apparatus can measure
130 the radioactivities of natural radionuclides. There is a small discrepancy
131 from 40 to 100 channels that may arise from other natural radionuclides in
132 the soil.

133 Table 1 summarizes the radioactivity concentrations of ^{134}Cs and ^{137}Cs
134 measured by using the apparatus and the HPGe detector for eight soils with
135 different radioactivities. The measurement time of the apparatus was 300 s.
136 Each radioactivity has an uncertainty occurring in minimizing the chi-squared.
137 The uncertainties were less than 0.8%.

138 The ratios of the radioactivity concentration obtained by the apparatus to
139 that obtained by the HPGe detector are also shown in Table 1. The difference

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10 between the concentrations obtained by each detector was a maximum of 9%.
11 The difference was larger than the uncertainty of the HPGe detector, which
12 was about 3%, and it was probably caused by the variation in soil density
13 in the measurement. Because the measurements with the apparatus and the
14 HPGe detector were taken more than 1 month apart, the water content of
15 the soil may have changed.
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22 6. Summary

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25 We developed an apparatus to measure the ^{134}Cs and ^{137}Cs radioactivity
26 of soil contaminated by the Fukushima Daiichi Nuclear Power Plant accident.
27 The apparatus consisted of a NaI(Tl) scintillator surrounded by a large soil
28 sample. There were advantages of the short measurement time and the soil
29 to be a shielding material due to the structure.
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33 The unfolding method with the least-squares technique was used to deter-
34 mine the radioactivities. The detector responses for each radionuclide were
35 calculated with EGS5 code for the unfolding method. In addition to ^{134}Cs
36 and ^{137}Cs , the detector responses for ^{40}K , ^{208}Tl , ^{214}Bi , and ^{228}Ac were pre-
37 pared as natural radionuclides in soil because γ -rays from these radionuclides
38 make a large contribution to the measured spectrum.
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46 The measured spectrum showed good agreement with that calculated
47 from the response matrix and radioactivities determined with the unfolding
48 method. Therefore, the apparatus can be used to measure radioactivities for
49 ^{134}Cs and ^{137}Cs , and also natural radionuclides.
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53 To verify the accuracy of our method, we compared the radioactivities
54 measured by using the apparatus and the HPGe detector. The radioactivities
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for eight soil samples measured with both detectors agreed to within 9%.

We confirmed that the unfolding method with the least-squares technique was effective for determining the radioactivities of soil contaminated with ^{134}Cs and ^{137}Cs . The unfolding method allows us to use a scintillator with poor energy resolution. Other radionuclides could be measured by adding the corresponding detector responses.

Acknowledgements

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Figure 1: Sectional view of the apparatus. In the measurement, the soil is filled to above the minimum filling height. The soil volume is 0.240 m^3 at the minimum filling height.

Figure 2: Detector responses for ^{40}K , ^{134}Cs , ^{137}Cs , ^{208}Tl , ^{214}Bi , and ^{228}Ac .

Figure 3: Measured and calculated spectra from the apparatus. The calculated spectrum was determined from the response matrix and the measured radioactivities. The radioactivity concentrations of ^{134}Cs and ^{137}Cs were 304 and 858 Bq/kg, respectively.

Table 1: Radioactivity concentration (Bq/kg) of ^{134}Cs and ^{137}Cs measured by using the apparatus and the HPGe detector, and the ratio of radioactivity concentrations obtained by the apparatus to those obtained by the HPGe detector. Each value measured by using the apparatus has an uncertainty less than 0.8%, which occurs in minimizing the chi-squared. The HPGe detector values have an uncertainty of about 3%.

Soil	^{134}Cs (Appar.)	^{137}Cs (Appar.)	^{134}Cs (HPGe)	^{137}Cs (HPGe)	^{134}Cs Ratio	^{137}Cs Ratio
#1	2.81×10^2	7.85×10^2	2.84×10^2	8.60×10^2	0.99	0.91
#2	3.04×10^2	8.57×10^2	2.90×10^2	8.53×10^2	1.05	1.01
#3	6.75×10^2	1.86×10^3	6.47×10^2	1.89×10^3	1.04	0.98
#4	4.72×10^2	1.29×10^3	4.55×10^2	1.31×10^3	1.04	0.98
#5	5.80×10^2	1.65×10^3	5.48×10^2	1.57×10^3	1.06	1.05
#6	1.31×10^3	3.53×10^3	1.25×10^3	3.61×10^3	1.05	0.98
#7	1.34×10^3	3.65×10^3	1.30×10^3	3.72×10^3	1.03	0.98
#8	2.06×10^3	5.50×10^3	1.88×10^3	5.53×10^3	1.09	1.00