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Articles

Efficiency Calibration of Ge Detector for ^{131}I and ^{134}Cs in Soil Samples and a Simplified Calculation of Cascade Summing Corrections for Volume Source

M. Asai,*,[a] Y. Kaneya,[a,b] T. K. Sato,[a] K. Tsukada,[a] K. Ooe,[a] N. Sato,[a] and A. Toyoshima[a]

[a]*Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan*
[b]*Graduate School of Science and Engineering, Ibaraki University, 2-1-1 Bunkyo, Mito, Ibaraki 310-8512, Japan*

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To measure radioactivities in soil contaminated by the accident of the Fukushima Daiichi Nuclear Power Plant, efficiency calibration of Ge detectors for ^{131}I, ^{134}Cs, and ^{137}Cs in volume sources was investigated. Gamma-ray detection efficiencies for these nuclei were determined precisely using standard soil samples containing ^{134}Cs, ^{137}Cs, ^{175}Hf, and ^{88}Zr. These standard sources were prepared by admixing radioactive solutions with soil, and point sources were made from the same solutions to determine their radioactivity concentrations. The efficiency for the 364 keV γ ray of ^{131}I was extracted from the experimental efficiencies of ^{137}Cs and ^{175}Hf. Cascade summing corrections for γ rays of ^{134}Cs in soil samples were evaluated experimentally. To easily calculate the cascade summing corrections for volume sources, we examined a simplified method using averaged efficiencies, and evaluated its validity through a comparison of the calculated correction factors with the experimental ones.

1. Introduction

The accident of the Fukushima Daiichi Nuclear Power Plant dispersed a huge amount of radionuclides in the natural environment, and part of them were deposited on the ground surface in the extensive region of east Japan. Assessments of the deposition densities of radionuclides and their geographical distributions provide important quantitative bases for estimations of radiation exposures of the residents, transfer of radionuclides into agricultural products, costs of decontaminations, etc. The Japanese government in collaboration with a few hundreds of researchers in universities and associations carried out detailed measurements of radionuclides deposited on soil over the whole area of Fukushima prefecture.[1] Independent of this work, we carried out measurements of radionuclides in soil samples collected in the north and east area of Ibaraki prefecture and in the east area of Tochigi prefecture. The soil samples were collected on May 21–22 and June 22 in 2011, and γ rays originating from the β^- decay of ^{131}I, ^{134}Cs, and ^{137}Cs were observed with Ge detectors, whose details are described in a separate paper.[2]

To measure radioactivities in soil with a Ge detector, we have to know γ-ray detection efficiencies of the Ge detector for soil samples. Typically soil samples have a large volume and density. Thus, to determine the efficiencies of Ge detectors, it is necessary to prepare the standard soil sample which contains a known amount of radionuclides and whose shape, density, and elemental contents are approximately the same as those of soil samples.

At first, we used a standard soil sample distributed by IAEA to measure the efficiencies. This soil sample contains 4.0–60 Bq kg^{-1} of ^{241}Am, ^{109}Cd, ^{134}Cs, ^{137}Cs, ^{54}Mn, and ^{60}Co. However, these radioactivity concentrations were too weak to determine the detector efficiencies precisely. Moreover, this standard source does not contain radionuclides which emit γ rays with energies at the range of 90–600 keV. This makes it difficult to determine the efficiency for the 364 keV γ ray of ^{131}I.

A mixed γ-ray standard source which resembles soil samples in shape, density, and elemental contents is commercially available. However, typically it does not contain ^{131}I and ^{134}Cs. The efficiency at 364 keV for ^{131}I is extracted from the efficiency calibration curve determined by using γ rays of, e.g., ^{203}Hg (279 keV), ^{51}Cr (320 keV), ^{113}Sn (392 keV), ^{85}Sr (514 keV), and ^{137}Cs (662 keV). However, these radionuclides except ^{137}Cs are not always available because of their short half-lives.

For ^{134}Cs, even if the efficiency calibration curve is determined precisely, one has to take account of the influence of the cascade summing effect which reduces or increases observed γ-ray peak counts, especially when the sample is placed at a close source-to-detector distance. For point sources, correction factors for the cascade summing effect are easily calculated using total efficiencies and full-energy peak efficiencies of Ge detectors determined experimentally.[3-6] For volume sources, however, the efficiencies are different at different parts of the source, which leads to different summing corrections at each part. This forces us to confront a difficulty in obtaining the total and full-energy peak efficiencies at any parts of the volume source.

The correction factor for a certain γ ray depends only on the decay scheme of the nuclide and on the geometry of the source and the detector. Thus, if we can prepare the standard source which contains the same radionuclide and has the same geometry (shape, density, and elemental contents) as those of soil samples, we can determine the contents of radionuclides in soil samples only through a comparison of observed γ-ray peak counts between the standard source and a soil sample, because the cascade summing corrections for both the standard source and a soil sample are identical, and thus they are cancelled out.

The aim of the present study is to determine γ-ray detection efficiencies of Ge detectors for ^{131}I, ^{134}Cs, and ^{137}Cs in soil samples, and to evaluate the cascade summing corrections for ^{131}I and ^{134}Cs in soil samples which has a large volume and density. For this purpose, we make standard soil samples containing ^{134}Cs, ^{137}Cs, ^{175}Hf, and ^{88}Zr. Contents of ^{134}Cs and ^{137}Cs radioactivities in soil samples are determined through a comparison of observed γ-ray peak counts between the standard soil samples and other samples. For ^{131}I, we propose a method to derive the efficiency at 364 keV from the experimental efficiencies at 662 keV of ^{137}Cs and at 343 keV of ^{175}Hf. Comparing the efficiencies determined by using the standard soil sample of ^{134}Cs with those of ^{137}Cs and ^{175}Hf, we evaluate the cascade summing

*Corresponding author. E-mail: asai.masato@jaea.go.jp. Fax: +81-29-282-5927.

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corrections for the γ rays of ^{134}Cs in soil samples experimentally. To easily calculate the cascade summing corrections for volume sources, we examine a simplified method using averaged efficiencies, and evaluate its validity through a comparison of the calculated correction factors with the experimental ones.

2. Experiments and Results

2.1. Preparation of standard soil samples. The standard soil samples were prepared as follows:

1) Hydrochloric or nitric acid solution containing ^{134}Cs, ^{137}Cs, ^{175}Hf, or ^{88}Zr is prepared.
2) A small part of the solution is precisely pipetted and admixed with soil. Then, the soil is packed into a standard sample container.
3) Another small part of the solution is pipetted on a Ta disk and evaporated to dryness to make a point source.
4) Radioactivity concentration in the solution is determined by measuring γ rays in the point source using a Ge detector whose efficiency is determined precisely.
5) Radioactivity in the standard soil sample is calculated from the above concentration and the volume of the pippeted solution.

To prepare the hydrochloric acid solution containing ^{134}Cs and ^{137}Cs, we used commercially available ^{134}Cs and ^{137}Cs solution. The ^{88}Zr and ^{175}Hf nuclei were produced in the ^{89}Y(p,2n)^{88}Zr and ^{175}Lu(p,n)^{175}Hf reactions by bombarding a metallic foil of Y (or Lu) with a 32 MeV (or 13 MeV) proton beam using the 20 MV tandem accelerator at Japan Atomic Energy Agency (JAEA). The irradiated target was dissolved with conc. HCl solution, and ^{88}Zr (or ^{175}Hf) were chemically separated from Y (or Lu) with anion-exchange chromatography.[7] The effluent containing ^{88}Zr (or ^{175}Hf) from the column was evaporated to dryness, and then it was dissolved in 4 M HNO_3 solution.

The soil was taken from a field at Tokai-mura in Ibaraki prefecture on April 2, 2011. Since the surface of the field was contaminated, we used the soil taken from the underground at more than 60 mm below the ground surface. The soil was stuffed into a standard sample container (made of plastic, with 48 mm in diameter and 59 mm in height), and ^{134}Cs and ^{137}Cs radioactivities in the soil were measured with a Ge detector prior to the admixture of the radioactive solutions. Then, the soil was moved into a glass dish with a 100 mm diameter, and the radioactive solution was dropped on the surface of the soil to be distributed as broad as possible, but not to touch the glass wall. The volume of the solution admixed with the soil was 2000 μL for ^{134}Cs, 300 μL for ^{137}Cs, 2000 μL for ^{88}Zr, and 500 μL for ^{175}Hf. To evaporate unwanted solution, the surface of the soil was heated under an infrared lamp for approximately 10 min. Then, the soil was moved into a plastic bag, and shaken in it to make a uniform distribution of the radionuclides. After the soil was moved back into the standard container, radioactivities remained in the plastic bag were measured with a Ge detector; they were less than 0.4% of the total radioactivities admixed with the soil. Finally, the container was sealed with a container cap and with another plastic bag.

The point source of ^{175}Hf was prepared by dropping 100 μL of the ^{175}Hf solution on a Ta disk and evaporating it to dryness. For ^{88}Zr, ^{134}Cs, and ^{137}Cs, since the volume of the solution was more than 100 μL, the solution was made absorbed into a small piece of glass filter on a Ta disk, and gently evaporated on a hot plate at 115 °C. This procedure was repeated several times. Total volume of the solution dropped on the Ta disk was 200 μL for ^{88}Zr, 300 μL for ^{137}Cs, and 300 μL and 1500 μL for ^{134}Cs. Each source was sealed with a plastic tape and in a small plastic bag.

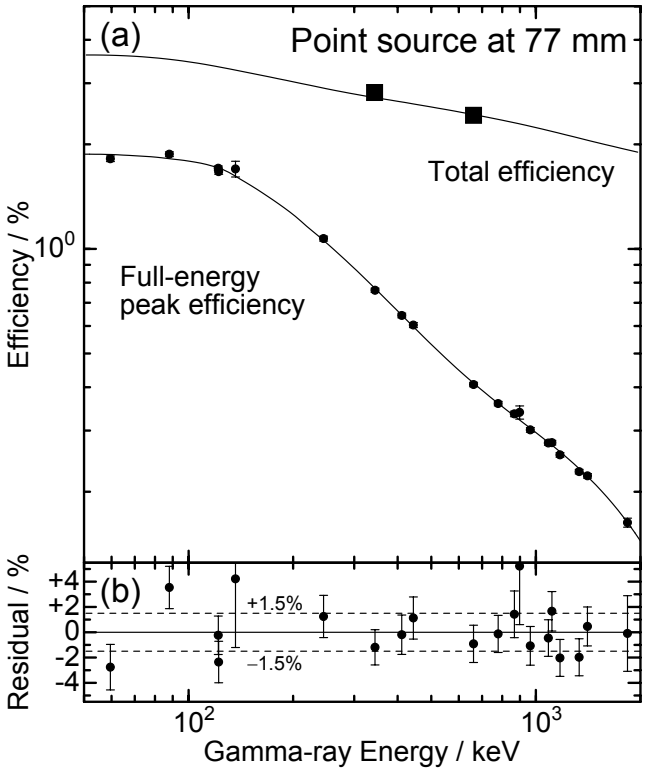

Figure 1. (a) Total and full-energy peak efficiencies for point sources placed at a source-to-detector distance of 77 mm. (b) Residuals of the measured full-energy peak efficiency values to the fitted curve.

2.2. Radioactivity measurement for point source. Radioactivities in the point sources were measured with a 28% n-type coaxial Ge detector (ORTEC GMX). The source was placed at the source-to-detector distance of 77 mm to reduce the influence of the cascade summing effect. Full-energy peak efficiencies of the detector were determined using a ^{152}Eu source and a mixed γ-ray standard source containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{137}Cs, ^{60}Co, and ^{88}Y. The disintegration rate of ^{152}Eu in the source was guaranteed with an uncertainty of ±1.3% (1σ). Those of the mixed γ-ray standard source were ±1.5% (1σ). Gamma-ray peak counts observed in the decay of ^{152}Eu, ^{60}Co, and ^{88}Y were influenced by the cascade summing effect. The correction factors were calculated using total efficiencies and full-energy peak efficiencies determined in this work, and using information on the decay schemes. The deduced full-energy peak efficiencies are plotted in Figure 1 together with a fitted curve. For γ rays with energies of 120–1840 keV, a fitted curve was obtained through a least-squares fitting of the following equation to the experimental efficiencies:

$$\varepsilon_f(E) = a_1 + \sum_{n=1}^{3} a_{2n} \exp(-a_{2n+1}E), \quad (1)$$

where $\varepsilon_f(E)$ is the full-energy peak efficiency at the γ-ray energy E, and a_n are the fitting parameters. The efficiency curve in the <100 keV region was calculated using absorption coefficients of γ rays in Ge and other materials existing between Ge and the source, and was connected with the fitted curve in the >120 keV region. Uncertainties of the fitted curve were evaluated to be ±1.5% for 240–1500 keV γ rays from the deviations between the fitted curve and the experimental values.

Total efficiencies of the Ge detector, which are used to calculate the cascade summing corrections, were determined experimentally for the 343 and 662 keV γ rays using the ^{175}Hf and ^{137}Cs point sources, respectively. For ^{175}Hf, contributions

of weak γ-ray lines other than the 343 keV one in the decay scheme of ^{175}Hf were subtracted from the total counts, which were estimated from the observed γ-ray peak counts and peak-to-total ratios determined in this work. A total efficiency curve was calculated by a method described in Reference 8 using total absorption coefficients of γ rays. This calculation systematically underestimates the total efficiencies by a factor of 1.5–2.0, because the contribution of scattered γ rays off the Pb shield and other materials surrounding the detector are not taken into account in this calculation. Thus, we scaled the calculated efficiency curve to fit the experimental efficiencies as shown in Figure 1.

Using these efficiency curves, disintegration rates A (in unit of Bq) of ^{134}Cs, ^{137}Cs, ^{175}Hf, ^{88}Zr, and ^{88}Y (the daughter nuclide of ^{88}Zr) in the point sources were determined with the following equation:

$$A = N_i/I_i/\varepsilon_f(E_i)/C_i, \quad (2)$$

where N_i is the observed peak count rate of the γ ray i in unit of cps (counts per second), I_i is the emission probability of the γ ray i, E_i is the energy of the γ ray i, and C_i is the correction factor of cascade summing for the γ ray i. For ^{134}Cs, a decay scheme containing 11 γ transitions[9] depicted in Figure 2 was used to calculate the cascade summing corrections, where all the combinations of cascading γ rays including triple and quadruple cascades were taken into account. For ^{175}Hf, a decay scheme containing 8 γ transitions[9] was employed. Summing with characteristic X rays following internal conversion (IC) and electron capture (EC) is fully taken into account, while angular correlations and summing with Bremsstrahlung X rays and β rays are not taken into account because their contributions are small under the present experimental condition. The deduced correction factors for ^{134}Cs are summarized in Table 1. The correction factors for the 393 keV and 662 keV γ rays of ^{88}Zr and ^{137}Cs, respectively, were 1.0 because their initial levels have long lifetimes.

From the deduced disintegration rates of the radionuclides in the point sources, we have derived the radioactivity concentrations in the solution, and determined the disintegration rates of ^{134}Cs, ^{137}Cs, ^{175}Hf, ^{88}Zr, and ^{88}Y in the standard soil samples with uncertainties of ±1.8–4.0%.

2.3. Efficiency determination for soil sample. Figure 3 shows a geometry for the measurement of soil samples in the

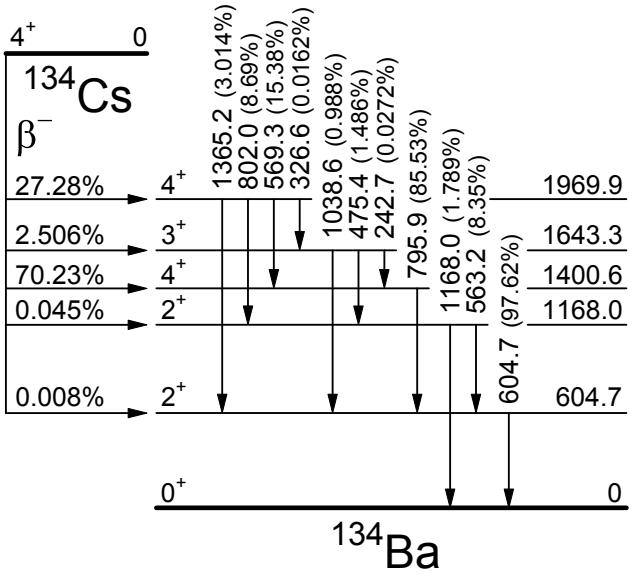

Figure 2. Decay scheme of ^{134}Cs,[9] with which we calculated the cascade summing corrections for the γ rays of ^{134}Cs.

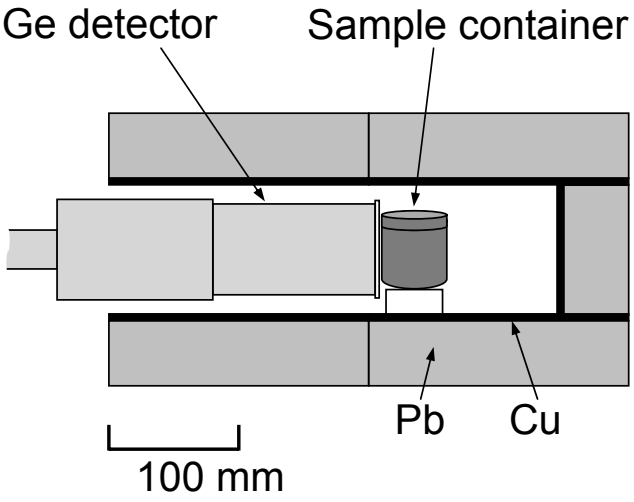

Figure 3. Schematic drawing of our detector setup for measurements of soil samples.

TABLE 1: Correction factors of cascade summing for γ rays in the decay of ^{134}Cs and ^{131}I, calculated for the point source placed at the source-to-detector distance of 77 mm and 4 mm, and for the volume source of soil samples (48 mm in diameter and ~55 mm in height) placed at 4 mm from the detector window. For the volume source, the correction factors are calculated by the simplified method using averaged efficiencies

Nuclide	E_γ / keV	Calculated correction factors of cascade summing: C		
		Point source at 77 mm	Soil sample	Point source at 4 mm
^{134}Cs	475.4	0.955	0.898	0.684
	563.2	0.952	0.891	0.662
	569.3	0.952	0.891	0.664
	604.7	0.971	0.933	0.782
	795.9	0.971	0.932	0.781
	802.0	0.956	0.899	0.686
	1038.6	0.986	0.970	0.912
	1168.0	1.013	1.037	1.149
	1365.2	1.028	1.075	1.284
^{131}I	80.2	0.972	0.933	0.775
	284.3	0.970	0.941	0.810
	364.5	1.0007	1.0017	1.009
	637.0	1.0001	1.0003	1.0016

present work. We used a horizontal-type Ge detector with a relative efficiency of 28% (ORTEC GMX). The sample container was placed so that its radial side should face the front window of the Ge detector. The distance between the window and the sample container was 4 mm.

To verify the homogeneity of radioactivity distributions in the standard soil samples, the direction of the sample container facing the detector was changed several times by axially rotating the container at 45° (or 90°) steps. Figure 4 shows relative γ-ray count rates for ^{134}Cs and ^{137}Cs observed at the 8 different directions. It was found that the observed γ-ray count rates fluctuated within ±2%, indicating that the good homogeneity was achieved for the standard soil samples. To minimize the influence of this fluctuation, we adopted the average count rates to deduce detector efficiencies.

Full-energy peak efficiencies determined by measuring the γ rays of ^{134}Cs, ^{137}Cs, ^{175}Hf, ^{88}Zr, and ^{88}Y in the standard soil samples are shown in Figure 5. Open circles plotted in Figure 5(a) denote the efficiencies without cascade summing corrections, while closed circles in Figure 5(b) denote the efficiencies with cascade summing corrections whose details are described in the next section. A solid curve in Figure 5(a) shows a full-energy peak efficiency curve obtained by fitting eq 1 to the corrected efficiencies, which is also drawn in Figure 5(b). Although the efficiencies plotted in Figure 5(a) are largely scattered because of the cascade summing effect, we have to use these non-corrected efficiencies in the analysis of each γ ray to deduce the ^{134}Cs radioactivity, because the cascade summing corrections are compensated between the measurements of soil samples and the ^{134}Cs standard. Using these standard soil samples and resultant efficiencies, we can deduce the radioactivities of ^{134}Cs and ^{137}Cs in soil samples precisely.

The full-energy peak efficiency for the 364 keV γ ray of ^{131}I was extracted from the experimental efficiencies at 343 and 662 keV measured with the ^{175}Hf and ^{137}Cs standard soil samples, respectively. For this purpose, the cascade summing correction for the 343 keV γ ray have to be done properly. The 343 keV γ ray of ^{175}Hf is summed with the 89 keV γ ray and Lu *K* X rays emitted following EC and IC; it is estimated that 97% of the summing events results from the sum with Lu *K* X rays. Fortunately, the events arising from the sum of the 343 keV γ-ray peak with the Lu *K* X rays (including full-energy peaks and scattered components) are clearly visible in the spectrum as shown in Figure 6. By adding these summing events with the 343 keV peak counts, we can obtain the corrected efficiency for the 343 keV γ ray. The efficiency curve was obtained through a fitting of a linear function in a log-log plot to the experimental efficiencies at 343 and 662 keV, which is drawn by a dashed line in Figure 5(a). As shown in Figure 5(a), this straight line well reproduces the full-energy peak efficiency curve within the energy range of 300–660 keV, leading to the precise determination of the full-energy peak efficiency for the 364 keV γ ray of ^{131}I.

The comparison of this fitted line (dashed one) with the experimental efficiencies allows us to estimate the correction factors of cascade summing for the γ rays of ^{134}Cs in soil samples. The efficiency for the 605 keV γ ray of ^{134}Cs is found to be 6.7% lower than the fitted line, which corresponds to the

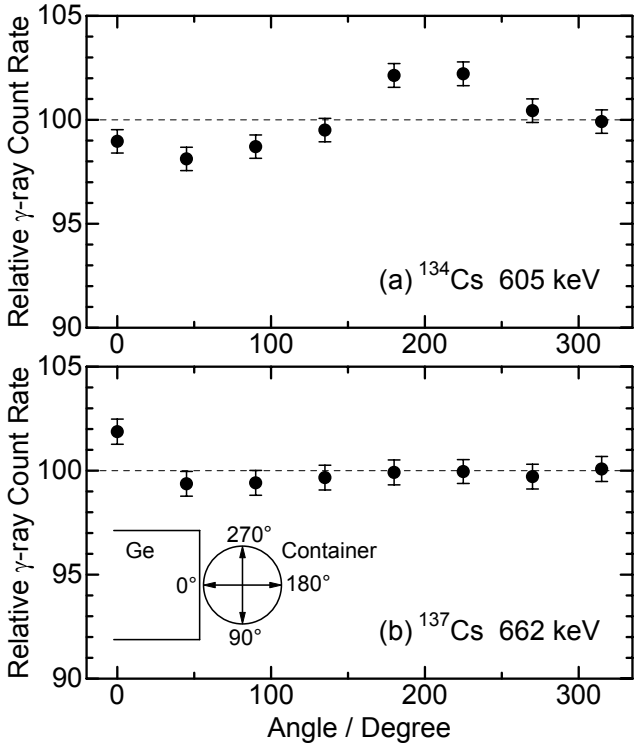

Figure 4. Relative γ-ray count rates observed at 8 different directions of the sample container for (a) ^{134}Cs and (b) ^{137}Cs in the standard soil samples. The geometry of the detector and the sample container is also drawn in the figure.

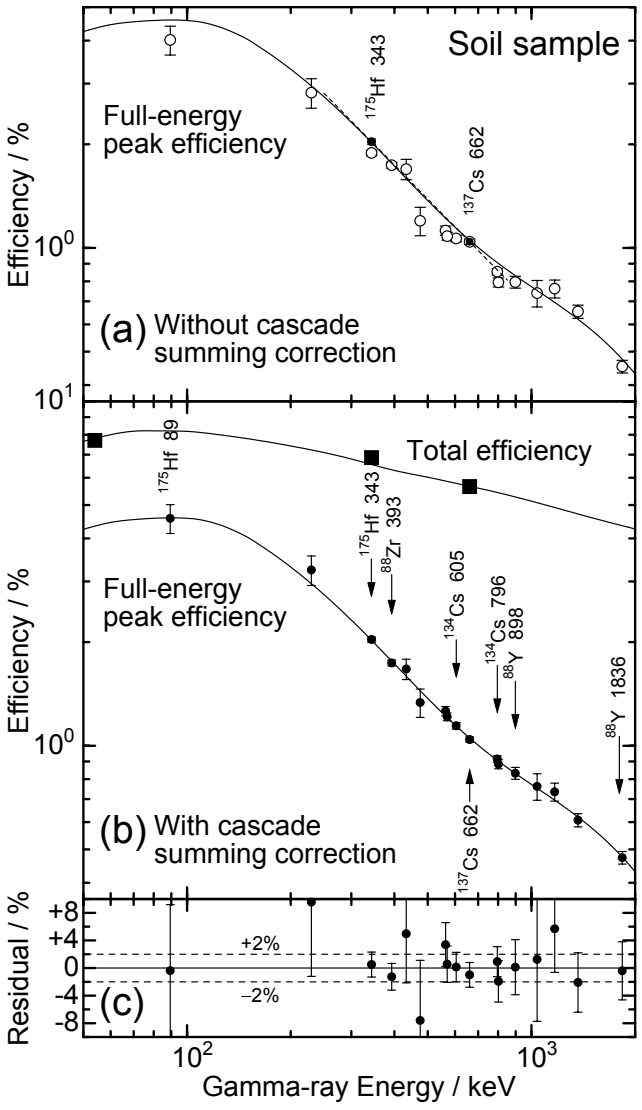

Figure 5. Total and full-energy peak efficiencies for soil samples. Open circles plotted in (a) denote the efficiencies without cascade summing corrections, while closed circles in (b) denote the efficiencies with cascade summing corrections. A full-energy peak efficiency curve in (a) and (b) is obtained by fitting eq 1 to the corrected efficiencies. A dashed line in (a) is obtained by fitting a linear function in a log-log plot to the experimental efficiencies at 343 and 662 keV, where the efficiency at 343 keV is corrected for the cascade summing effect. Residuals of the corrected full-energy peak efficiency values to the fitted curve are given in (c).

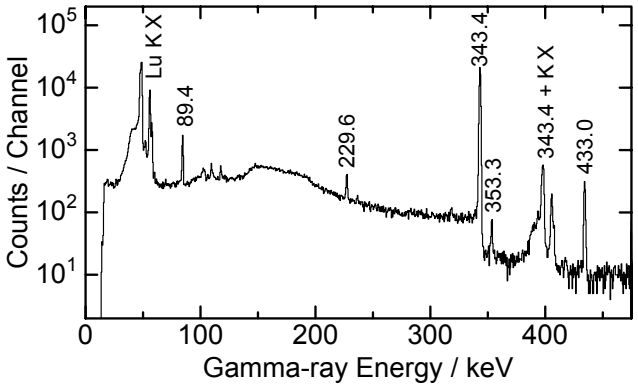

Figure 6. Gamma-ray spectrum observed with the ^{175}Hf standard soil sample.

correction factor of 0.933 for the 605 keV γ ray. Those for the 563 and 569 keV γ rays are deduced to be 0.914 and 0.891, respectively. These correction factors are compared with the calculated ones in the next section.

To determine the full-energy peak efficiency at 364 keV, it should be better to use the 393 keV γ ray of ^{88}Zr because no cascade summing correction is needed for this γ ray. However, because of the weak radioactivity of the ^{88}Zr point source, we could not obtain the accurate values of the ^{88}Zr and ^{88}Y radioactivities in this work. Thus, we did not use the ^{88}Zr and ^{88}Y data to determine the efficiency at 364 keV in this work. Only relative values were used to obtain a fitted curve in the next section.

3. Discussions

3.1. Cascade summing corrections for ^{134}Cs in soil sample. If one can prepare a standard soil sample of ^{134}Cs like this work, no cascade summing correction is needed to determine ^{134}Cs radioactivities in soil samples. On the other hand, if one cannot prepare a standard soil sample of ^{134}Cs, one has to calculate the correction factors of cascade summing for ^{134}Cs. As already mentioned in Section 1, the exact calculation of cascade summing corrections for a volume source is very difficult because it is hard to experimentally obtain total and full-energy peak efficiencies at any parts of the volume source. Thus, it is worthwhile proposing an easier method to estimate the correction factors as an alternative to the exact calculation.

Here, we examine the method to calculate correction factors for a volume source using averaged efficiencies.[10] Consider the decay scheme consisting of two γ transitions (γ_1 and γ_2) in a one-to-one cascade like ^{60}Co. The correction factor for γ_1 for a point source is calculated as

$$C_1 = 1 - \varepsilon_T(E_2), \tag{3}$$

where $\varepsilon_T(E_2)$ is the total efficiency for a point source at the γ-ray energy E_2. For a volume source, the correction factor is calculated with the following equation:[6]

$$C_1 = 1 - \frac{1}{V}\int \varepsilon_f(E_1, r)\varepsilon_T(E_2, r)dV \Big/ \frac{1}{V}\int \varepsilon_f(E_1, r)dV$$

$$= 1 - \frac{1}{V}\int \varepsilon_f(E_1, r)\varepsilon_T(E_2, r)dV \Big/ \bar{\varepsilon}_f(E_1), \tag{4}$$

where $\int \varepsilon(r)dV/V$ means the integration of the efficiency $\varepsilon(r)$ at the position r over the whole volume of the source, dV is the volume element at each position, and V is the total volume of the source. $\bar{\varepsilon}$ denotes the efficiency averaged over the whole volume of the source, which is equal to $\int \varepsilon(r)dV/V$, and is equivalent to the experimental efficiency measured with a standard soil sample. In the present simplified calculation, we replace $\int \varepsilon_f(E_1, r)\varepsilon_T(E_2, r)dV/V$ into $\bar{\varepsilon}_f(E_1)\bar{\varepsilon}_T(E_2)$, that is,

$$\frac{1}{V}\int \varepsilon_f(E_1, r)\varepsilon_T(E_2, r)dV \simeq \bar{\varepsilon}_f(E_1)\bar{\varepsilon}_T(E_2), \tag{5}$$

and convert eq 4 into

$$C_1 \simeq 1 - \bar{\varepsilon}_T(E_2), \tag{6}$$

which is equivalent to eq 3 for a point source.

This approximation (eq 5) systematically underestimates $\int \varepsilon_f(E_1, r)\varepsilon_T(E_2, r)dV/V$ by ~15% in the present experimental condition. However, if the correction factor C_1 is 0.9, the ~15% deviation induces only 1.5% error to the final result, which is accurate enough for most of purposes of environmental radioactivity measurements. The accuracy of this approximation was investigated by several authors[11-13] through Monte Carlo simulations or numerical calculations for various nuclides and geometries. Although the difference between the exact calculation and the simplified one was found to be significant for cascades involving low-energy X/γ rays, the differences for high-energy γ-ray cascades are estimated to be typically of the order of a few per cent for geometries with averaged total efficiencies of <10%.

There are several sophisticated codes available to calculate cascade summing corrections for volume sources, which are based on full Monte Carlo simulations or numerical calculations using calculated efficiencies (References 11, 12, and 14 and references therein). However, the recent intercomparison exercise[14] revealed that the calculated correction factors for a volume source were largely scattered between laboratories by more than ±10%. This large scattering is probably due to incorrect total efficiencies used in the calculations. On the other hand, the present simplified method utilizes experimental averaged efficiencies $\bar{\varepsilon}_f$ and $\bar{\varepsilon}_T$. This is another advantage of this simplified method, because the $\bar{\varepsilon}_f$ and $\bar{\varepsilon}_T$ can be obtained with good accuracies using standard sources, which ensures the reliability and the accuracy of the deduced correction factors.

To confirm the validity and accuracies of this simplified method, we calculate the correction factors for the γ rays of ^{134}Cs, ^{175}Hf, and ^{88}Y. The total efficiencies $\bar{\varepsilon}_T(E)$ are determined using the experimental efficiencies measured with the ^{175}Hf and ^{137}Cs standard soil samples at 343 and 662 keV, and at Lu *K* X-ray energy which is extracted from the events originating from the sum of the 343 keV γ-ray peak with the Lu *K* X rays observed in the ^{175}Hf spectrum. The total efficiency curve fitted to the experimental data is shown in Figure 5(b). The deduced correction factors for ^{134}Cs are summarized in Table 1, and the corrected full-energy peak efficiencies for ^{134}Cs, ^{175}Hf, and ^{88}Y are plotted in Figure 5(b) by closed circles.

The full-energy peak efficiencies which seemed to be largely scattered without corrections turn out to be in line with a smooth fitted curve. This fitted curve also coincides with the dashed line in Figure 5(a) at the energy range of 300–660 keV. The calculated correction factors for the 569 and 604 keV γ rays of ^{134}Cs agree very well with the experimental ones. Although the correction factors calculated for the 796, 802, 1039, 1168, and 1365 keV γ rays of ^{134}Cs are very different each other, the corrected full-energy peak efficiencies all come close to the fitted line within a few % deviations. These results demonstrate that this simplified calculation is accurate enough to estimate correction factors of cascade summing for volume sources if required uncertainty level is not lower than a few per cent.

3.2. Cascade summing corrections for ^{131}I in soil samples. This simplified calculation is also applied to the cascade summing corrections for ^{131}I in soil samples. The decay scheme of ^{131}I contains 19 γ transitions, among which the 364 keV γ ray

has the largest intensity of 81.7%.[9] Using this decay scheme, we calculate the correction factors for the four intense γ rays of 80, 284, 364, and 637 keV of ^{131}I as summarized in Table 1. The correction factor for the 364 keV γ ray is 1.0017, indicating that the cascade summing correction for the 364 keV γ ray is negligible.

3.3. Cascade summing corrections for other geometry. The correction factors summarized in Table 1 are valid only for the specific geometry of the sources and the detector in the present work. However, we can imagine that the correction factors would resemble if total efficiencies are similar between one geometry and another. Thus, it is very useful to summarize the correction factors for various geometries, because using those values other researchers can also obtain a rough estimate of the correction factors for their own geometry. For this purpose, we present another result of the efficiency calibration and cascade summing corrections for the geometry of a short source-to-detector distance.

Here, we measured efficiencies of the Ge detector for a point source placed at a source-to-detector distance of 4 mm. This condition is supposed to be the ones to measure weak radioactivities in environmental samples such as filter paper at the closest geometry. A 4-mm thick plastic plate was inserted between the source and the detector to absorb β particles. Full-energy peak efficiencies were measured by using a mixed γ-ray standard source containing ^{109}Cd, ^{57}Co, ^{137}Cs, and ^{60}Co, and the point sources of ^{134}Cs, ^{137}Cs, and ^{175}Hf prepared in Section 2.1. A total efficiency curve was calculated with the method described in Section 2.2 to reproduce the experimental efficiencies measured with the ^{175}Hf and ^{137}Cs sources. The total efficiency and the full-energy peak efficiencies with and without cascade summing corrections are shown in Figure 7 together with fitted curves. Although the efficiencies without cascade summing corrections are largely scattered, the corrected efficiencies are all in line with a smooth fitted curve, indicating that the cascade summing corrections are appropriate. The correction factors calculated for the γ rays of ^{134}Cs and ^{131}I are summarized in Table 1.

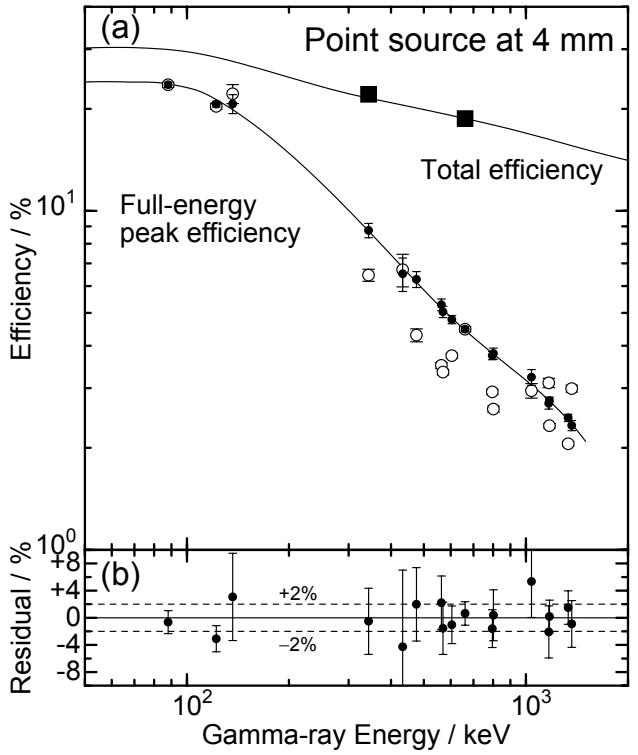

Figure 7. (a) Total and full-energy peak efficiencies for point sources placed at a source-to-detector distance of 4 mm. Open circles denote the efficiencies without cascade summing corrections, and closed circles denote the efficiencies with cascade summing corrections. A full-energy peak efficiency curve is obtained by fitting eq 1 to the corrected efficiencies. (b) Residuals of the corrected full-energy peak efficiency values to the fitted curve.

4. Conclusion

To measure radioactivities in soil contaminated by the accident of the Fukushima Daiichi Nuclear Power Plant, γ-ray detection efficiencies of Ge detectors for ^{131}I, ^{134}Cs, and ^{137}Cs in soil samples were determined precisely using standard soil samples of ^{134}Cs, ^{137}Cs, ^{175}Hf, and ^{88}Zr. The standard soil samples were prepared by ourselves by admixing solutions containing ^{134}Cs, ^{137}Cs, ^{175}Hf, and ^{88}Zr with soil. Contents of ^{134}Cs and ^{137}Cs radioactivities in soil samples were determined through a comparison of observed γ-ray peak counts between the standard soil samples and other samples, which eliminates the influence of the cascade summing effect for ^{134}Cs. For ^{131}I, we proposed a method to derive the efficiency at 364 keV from the experimental efficiencies at 343 keV of ^{175}Hf and at 662 keV of ^{137}Cs. Uncertainties of the deduced efficiency values for ^{131}I, ^{134}Cs, and ^{137}Cs were evaluated to be ±2.7–3.5%.

The cascade summing corrections for the γ rays of ^{134}Cs in soil samples were evaluated experimentally from the deviations among the experimental efficiencies of ^{134}Cs, ^{137}Cs, and ^{175}Hf. To easily calculate the cascade summing corrections for volume sources, we examined a simplified method using averaged efficiencies, and evaluated its validity through a comparison of the calculated correction factors with the experimental ones. The calculated correction factors agreed very well with the experimental ones, demonstrating the validity of this method.

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