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Application of a 60×60 Response Matrix for a NaI(Tl) Scintillator to Fallout from the Fukushima Reactor Accident

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A 60×60 response matrix for a NaI(Tl) scintillator was derived to unfold pulse height distributions of gamma rays due to fallout released in the Fukushima nuclear reactor accident. The unfolded energy spectra yielded information concerning radioactivities and dose rates in the fallout field. The accuracy of the evaluated radioactivities and dose rates was discussed by comparing with other data.

Key words: response matrix method, fallout, Fukushima

1. Introduction

A 22×22 response matrix for a NaI(Tl) scintillator has often been used so far in order to unfold natural environmental gamma ray pulse height distributions[1]. The energy bin widths of this matrix, however, were too wide to distinguish between peaks which are quite near to one another when analyzing pulse height distributions due to artificial radionuclides such as the Fukushima fallout.

The purpose of this paper is to evaluate radioactivities of and dose rates due to individual radionuclides released from the Fukushima reactors by using a response matrix having narrower energy bin widths.

2. Method

Procedures are as follows. First of all, we derive a 60×60 response matrix for a NaI(Tl) scintillator. Next, operating this matrix to a pulse height distribution, we obtain a high resolution energy spectrum, from which we pick out primary gamma ray fluxes emitted from individual nuclides. Moreover, we calculate radioactivities per unit area from the primary fluxes by using constants evaluated assuming that the fallout forms an infinite plane source on the ground. Finally, we calculate the dose rates due to the radioactivities with use made of conversion factors for the plane source.

2.1. Derivation of the response matrix

We modified a Monte Carlo program SPHERIX[2] so as to derive a 60×60 matrix. An isotropic gamma ray incidence was assumed and 100,000 histories were traced for each energy bin.

2.2. Unfolding

While we derived a 60 × 60 response matrix for a 2"ϕ ×2" and that for a 3"ϕ ×3" scintillator, we will deal with only the latter in this paper as an example.

A pulse height distribution measured by Suzuki[3] on the roof of a building of Minato-ku, Tokyo on March 23, 2011 is used as a test sample of fallout. Figure 1 shows the pulse height distribution. Spectroscopic identification of nuclides in the figure was made in reference to articles of websites

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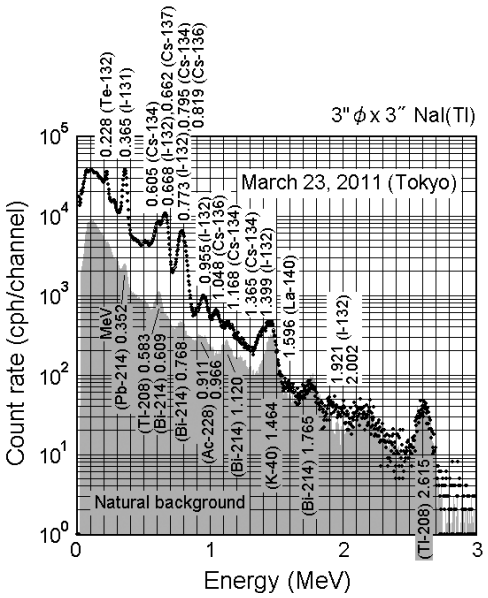

Fig. 1. Pulse height distribution.

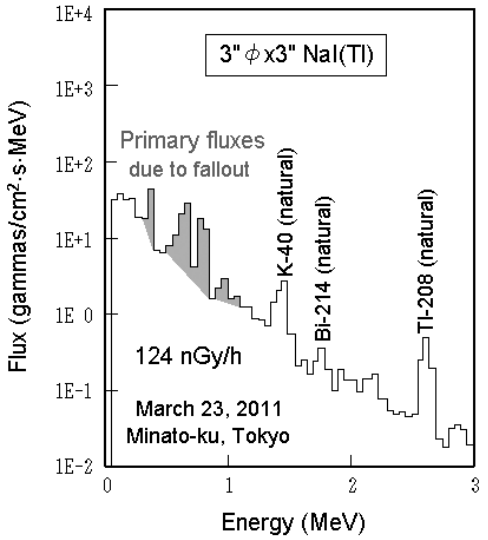

Fig. 2. Unfolded spectrum.

and newspapers on the accident. A natural background spectrum is also shown in the figure for reference.

Figure 2 shows the unfolded spectrum obtained by operating the 60 × 60 matrix to the pulse height distribution given in Figure 1. Primary gamma ray fluxes due to the fallout are evaluated by linearly interpolating between the values at nearest bins outside the region occupied by the fallout peaks as shown in the figure.

2.3. Conversion of primary flux into radioactivity

Table 1 gives basic constants with respect to the radionuclides released from the Fukushima reactors, which were picked up from ICRU Report[4]. The 2nd column of the table represents the gamma ray fluxes at 1 m above ground level per unit activity per unit area for a uniformly

Table 1. Conversion of radioactivity into primary gamma ray flux and dose rate for an infinite plane source.

Basie data (from ICRU53)		
*Flux of primary γ at 1 m above GL per unit activity per unit area		
Nuclide		Plane souroe
Energy (MeV)	* /(s.Bq)	μGy/h per kBq/m^2
Te-132		0.00129
0.2282	1.63	
I-131		0.00174
0.3645	1.57	
0.6370	0.15	
Cs-137		0.00268
0.6616	1.84	
Cs-134		0.00685
0.5693	0.30	
0.6046	1.99	
0.7958	1.78	
I-132		0.00988
0.5227	0.33	
0.6302	0.27	
0.6677	2.03	
0.7726	1.59	
0.9546	0.39	
Cs-136		0.00908
0.8185	2.09	
1.0480	1.72	

distributed plane source with infinite extension. These fluxes have to be assigned into the respective energy bins. How to assign them is described in Ref.1). Table 2 gives the result, where Te-132 is omitted, since it is assumed to be in radioactive equilibrium with I-132.

According to Table 2, the energies of gamma rays emitted from the nuclides other than I-131 are overlapping within the same energy bins. Then, we calculate the activities of individual radionuclides by solving simultaneous linear equations of 5 unknowns. Table 3 gives the coefficients of the simultaneous equations.

Thus, we can calculate the dose rates by multiplying the activities by the values of the 3rd column of Table 1.

3. Result

We developed a calculation program TICCIC which includes all the procedures described in the preceding section. Giving a table of pulse height distribution and inputting the peak channels of I-131 (0.3645 MeV) or of Cs-134 (0.795 MeV) and of Tl-208 (2.615 MeV), we obtain radioactivities and dose rates due to fallout. Table 4 gives an example of output data for the test sample appeared in Figure 1.

4. Discussion

4.1. Total dose rate

Table 2. Assignment of a line spectrum into each energy bin.

No.	E(MeV)	I-131	Cs-137	Cs-134	I-132	Cs-136
3	0.15-0.20					
4	0.20-0.25					
5	0.25-0.30					
6	0.30-0.35	0.33				
7	0.35-0.40	1.24				
8	0.40-0.45					
9	0.45-0.50				0.02	
10	0.50-0.55			0.03	0.32	
11	0.55-0.60			1.08		
12	0.60-0.65	0.11	0.49	1.18	0.54	
13	0.65-0.70	0.04	1.35		1.76	
14	0.70-0.75				0.08	
15	0.75-0.80			1.04	1.51	0.27
16	0.80-0.85			0.74		1.82
17	0.85-0.90					
18	0.90-0.95				0.16	
19	0.95-1.00				0.23	
20	1.00-1.05					0.93
21	1.05-1.10					0.79

Table 3. 5×5 matrix to calculate nuclide concentrations.

	Bin number				
Nuclide	6,7	12,13	15,16	18,19	20,21
I-131	1.57	0.15			
Cs-137		1.84			
Cs-134		1.18	1.78		
I-132		2.30	1.51	0.39	
Cs-136			2.09		1.72

Table 4. Example of output data (Minato-ku, Tokyo: March 23, 2011)

TICCIC
KGER3.CSV
Roof of a bldg

Activity (kBq/m2)	
Te-132	3.75E+00
I-131	1.18E+01
Cs-137	2.92E+00
Cs-134	4.06E+00
I-132	3.75E+00
Cs-136	2.66E–01

Dose rate(nGy/h)	
Te-132	4.83E+00
I-131	2.06E+01
Cs-137	7.82E+00
Cs-134	2.78E+01
I-132	3.70E+01
Cs-136	2.41E+00
Total	1.01E+02

Dose ratio(%)	
Te-132	5
I-131	21
Cs-137	8
Cs-134	28
I-132	37
Cs-136	2

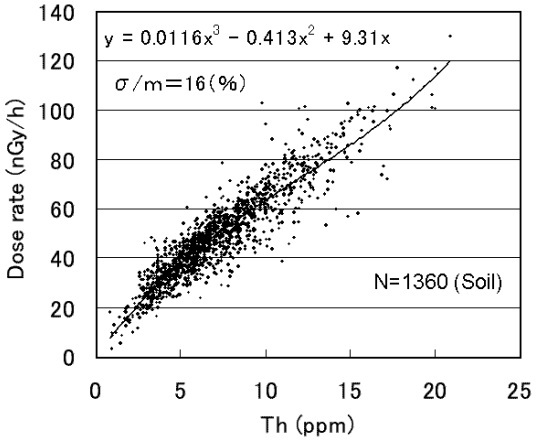

Fig. 3. Relationship between thorium content and dose rate in Japan.

The Radiation Earth Science Laboratory has a calculation program DEF (22×22 matrix), which can distinguish between artificial dose rate and natural component inferred from the photo-peaks due to natural radionucles[1)], i.e.. K-40 (1.464 MeV), Bi-214 (1.765 MeV) and Tl-208 (2.615 MeV). Although the DEF program can not always provide us with clear information about fallout radionuclide because of its wider energy bin widths as stated in the preceding section, it enables us to calculate at least fallout dose rates. Let us compare TICCIC with DEF calculated dose rates to know more about characteristics of TICCIC calculations.

A channel vs. energy calibration for DEF has been done by using the photo-peaks of K-40 and Tl-208 for natural radiation fields. However, we can not apply this method for fallout fields, since the photo-peaks of Cs-134 (1.365 MeV) and I-132 (1.399 MeV) exist extremely near to the K-40 peak (See Fig. 1).

We, therefore, modify the DEF program so as to be able to use a peak of I-131 (0.364 MeV) or of Cs-134 (0.795 MeV). Besides, a further modification is made so that the natural background level (BG) can be inferred from Tl-208 peak alone, since there are no fallout nuclides around there. Figure 3 shows a relationship between thorium (Th) contents estimated from Tl-208 peak[1)] and the corresponding natural gamma ray dose rates. Subtraction of background level from DEF estimation, i.e., DEF-BG, results in the fallout component. As is seen in the figure, the dose rate varies non-linearly with the thorium content because of a lithologic requirement.

Figure 4 shows the plots of dose rate estimated by TICCIC against that by DEF-BG for field data collected so far. As is seen in the figure, the values obtained by TICCIC are generally lower than those by DEF-BG. The reason for this is considered as follows. The TICCIC estimations are made under the assumption of an ideal infinite plane source. In reality, however, the field measurements are always accompanied by a ground roughness effect and/or carried out for somewhat quasi-exponentially distributed sources under the ground.

Figure 5 shows a result of Monte Carlo calculation for uniformly distributed Cs-137 slab sources with infinite extension as an example. In the figure, the term D

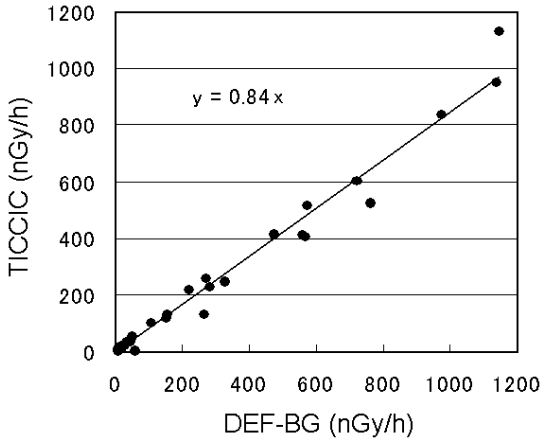

Fig. 4. Comparison between TICCIC and DEF estimates.

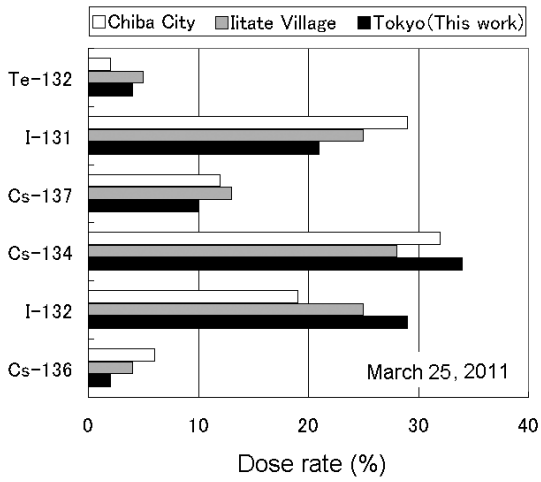

Fig. 6. Dose rate contribution.

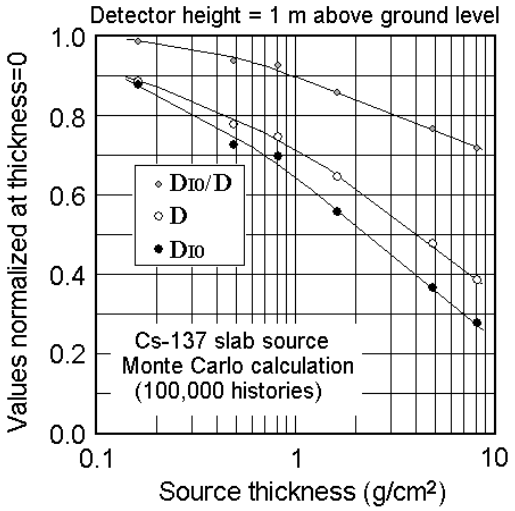

Fig. 5. Slab source calculations. The term D_{10} represents the calculation based on the constant given in ICRU 53 for an infinite plane source.

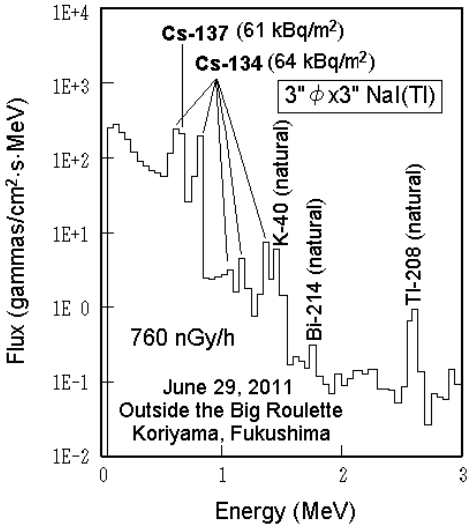

Fig. 7. Unfolded spectrum for Cs-134 and Cs-137 along with natural radionuclides.

represents a total dose rate due to primary and scattered components. The calculations designated by D_{10} were performed by multiplying the Monte Carlo calculated primary fluxes from the slab source by the conversion factors for a plane source given in ICRU Report[4] (See, Table 1). Judging from the regression coefficient, 0.84, given in Figure 4, an equivalent thickness of uniform slab source is inferred to be about 2 g/cm^2 on average in actual fields for Cs-137 according to Figure 5. This may be applicable to Cs-134 as well, since gamma ray energies emitted by Cs-137 and Cs-134 are close to each other.

4.2. Dose rate contribution of each nuclide

Figure 6 shows a comparison of dose rate contributions of Tokyo data with those taken at Chiba City, Chiba[5] and Iitate Village, Fukushima[6] on March 25. Although the soil of Iitate was sampled on March 31 and the measurement at Tokyo was carried out on March 23, those data have been corrected taking into account the decay of the nuclides. The agreement between them looks fairly good as a whole. It is hoped that the accuracy will be evaluated in more detail by obtaining more data in the future.

4.3. Radioactivity ratio of Cs-134 to Cs-137

The activity of Cs-134 has been reported to be nearly equal to that of Cs-137 for the Fukushima fallout with Ge(Li) measurements[5-8] in the field until July, 2011.

Figure 7 shows an example of unfolded data taken at Koriyama, Fukushima. In this case, the activity ratio is almost unity by chance. However, a mean value of the ratios is found from estimations by the present method for 24 sites in east Japan to be 1.26 ± 0.12, which is a little higher than Ge(Li) measurements. Further comparisons would be necessary to check whether or not this is caused by the deference between methods of evaluation.

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