Article

Estimation of Radionuclide Concentration in Plume Using Pulse Height Distribution Measured by $LaBr_3$ Scintillation Detector and Its Response to Radionuclides in Plume Calculated with egs5

Hideo HIRAYAMA [1,*], Hiroshi MATSUMURA [1], Yoshihito NAMITO [1] and Toshiya SANAMI [1]

[1] *High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba-shi, Ibaraki 305-0801, Japan*

A method was presented to estimate radionuclide concentration in plume using the pulse height distribution measured by a $LaBr_3$ scintillation detector and its calculated response to radionuclides in plume with egs5. Radionuclide concentration was estimated from the ratio between the peak count rates corresponding to each radionuclide in the measured pulse height distribution on an expressway on March 15 and in the calculated one from each radionuclide in plume using the egs5 Monte Carlo code. The pulse height distribution reconstructed based on the estimated concentrations agrees well with the measured one at the time that the contribution from radionuclides deposited on a ground surface is negligible.

KEYWORDS: radionuclide concentration, iodine 131, plume, Monte Carlo, egs5, $LaBr_3$ scintillator

I. Introduction

The radionuclide concentrations in plumes dispersed over a wide area due to the accident that occurred at the Fukushima Daiichi Nuclear Power Plant (hereinafter referred to as "plume") are important in evaluating the internal radiation dose from inhalation. Although the concentration of I-131 is particularly important from the perspective of radiation-induced thyroid cancer risk assessment, the direct measurement of radionuclide concentrations in plumes was conducted only in limited places far from the Fukushima Daiichi Nuclear Power Plant. Therefore, the most direct method is estimation from the density of I-131 deposited on the soil surface. However, since the half-life of I-131 is as short as about eight days, the measurement data are extremely limited compared to that of Cs-134 and Cs-137. As a result, estimating from an atmospheric dispersion model and air dose rate, as well as estimating from the analysis of I-139 with a very long half-life, have been attempted. Although it is essential to perform evaluation based on an atmospheric dispersion model to estimate the concentration distribution over a wide area, to increase accuracy, it would be desirable to obtain the data on the concentration of radionuclides,

* Corresponding author, E-mail: hideo.hirayama@kek.jp
DOI : 10.15669/fukushimainsights.Vol.4.233

Originally published in Transactions of the Atomic Energy Society of Japan (ISSN 1347–2879), Vol. 12, No. 4, p.304–310 (2013) in Japanese. (Japanese version accepted: October 1, 2013)

such as I-131, contained in the plumes, but not the air dose rate and the amount of deposition on the soil surface. In this paper, we attempted to estimate the concentrations of radionuclides contained in the plumes that passed overhead using the pulse height distribution measured by Matsumura, et al. at expressway service areas (SA), parking areas (PA), some interchanges (IC), and junctions (JCT) with a $\phi 1.5'' \times 1.5''$ $LaBr_3$ scintillation spectrometer (manufactured by Canberra, IN1KL-1, hereinafter referred to as "$LaBr_3$ detector") on March 15, 2011 [1]. We calculated the detector response of $LaBr_3$ to the radionuclides in plumes by applying a method [2] whereby the photon flux from a plane isotropic source is converted into a point isotropic source, and a plane detector to an egs5 [3] electromagnetic cascade Monte Carlo code to estimate the concentrations of radionuclides in the plumes from the ratio between the peak count rates per Bq/cm^3, corresponding to each radionuclide calculated and the corresponding peak count rate measurements.

II. Calculation of the Response of $LaBr_3$ Detector with egs5

1. Photon Spectra of Radionuclides Contained in a Plume 1.3 m above the Ground Surface

The distribution of radionuclides when a plume arrived was assumed to correspond to the case where there was a plane isotropic source spreading infinitely and horizontally from the ground surface to the plume height (h_0) with a uniform density.

We calculated the photon spectra 1.3 m above the soil surface, assuming that the vertical distribution was uniform from the soil surface to h_0, by applying the method [2] of converting the photon flux from a plane isotropic source into a point isotropic source, and a plane detector to egs5 [3]. To validate this method, **Figure 1** shows the comparison between the result of the analytical method described in EPA-402-R-93-081 [4] and the result of calculation with egs5 for the scattered photon spectrum in an infinite air system in which one 100-keV photon is emitted per cubic meter every second. In the calculation with egs5, an air density of 0.001205 g/cm^3 was used under 1 atm (atmospheric pressure) at 20°C (hereinafter referred to as "NTP state"). As the simulation of an infinite system, h_0 was set to ten times of the mean free path of a 100-keV photon in the air. Both the shape and size of the spectra were found to be identical. Figure 1 also shows the result of calculating the photon spectrum at a height of 1.3 m taking the soil into

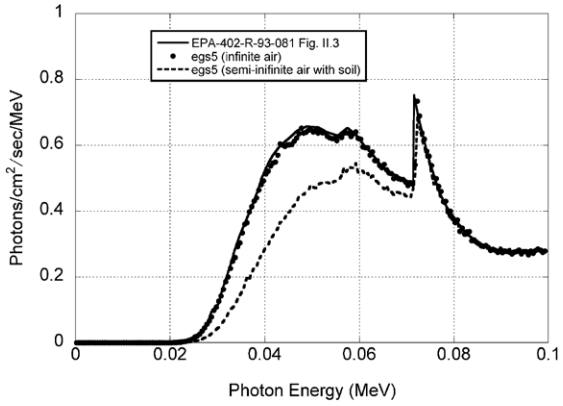


Figure 1 Comparison of energy spectrum of scattered photons for submersion in a 1 Bq/m^3 100 keV contaminated air source

consideration. When the soil was taken into consideration, the calculation result was doubled so that the total of the radiation source would be the same. Although there was little change in the area close to the energy of the radiation source, it was found that there was less backscattering in a remote area when the soil was taken into account. The number of unscattered photons was 5.52×10^{-3} photons/cm^2/sec in the case of EPA-402-R-93-081. The result of egs5 where the soil was not considered was 5.50×10^{-3} photons/cm^2/sec, and the result of egs5 at a height of 1.3 m considering the ground was 6.03×10^{-3} photons/cm^2/sec. We can see that they are identical.

To see the impact of plume height h_0 from the plume containing I-132, which emits high energy gamma rays, **Figure 2** shows the comparison of spectra at a height of 1.3 m from the soil surface between the case where the height is 500 m and the case where the height is 2,219 m, which is ten times of the mean free path for the gamma ray with the highest energy in air, and is regarded as infinite in thickness. It was found that there was little difference between either case. Although the plume height is said to be between 100 m and 1,000 m, based on the result shown in Figure 2, the plumes up to 500 m in height are to be studied.

Figure 3 shows the calculated photon spectra at a height of 1.3 m above the soil surface from plumes 100, 200, and 500 m high containing I-131 at a concentration of 1 Bq/cm^3 in the air in NTP state. The figure shows that the scattering portion increases as the plume height

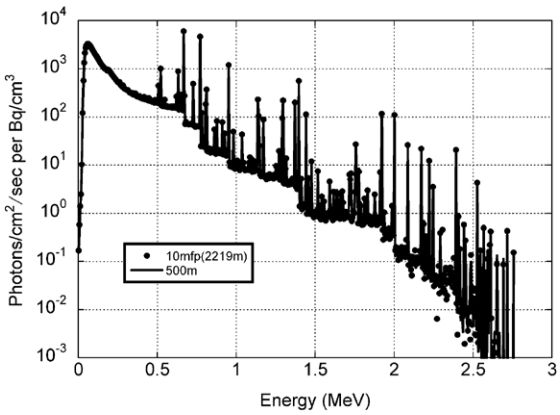


Figure 2 Comparison of photon spectrum at 1.3 m above soil surface from I-132 contained in plumes 500 m and 2,219 m (10 mfp) high

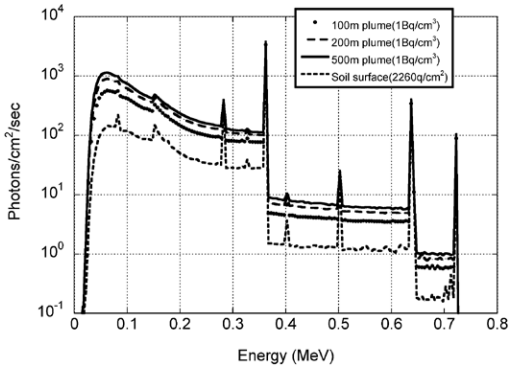


Figure 3 Comparison of photon spectrum at 1.3 m above soil surface from I-131 contained in plume or deposited on soil surface

h_0 increases, but there is little difference between 200 m and 500 m. Figure 3 also shows the calculated photon spectrum at a height of 1.3 m from the soil surface when I-131 is distributed uniformly over a wide area on the soil surface at a concentration of 2,260 Bq/cm^2, where the peak gamma ray count rate of 0.364 MeV becomes almost identical to the result for a plume 500 m high. In the case of ground surface distribution, it was found that the ratio of scattered photons to unscattered photons that were emitted from a radionuclide and then were not subjected to scattering was small compared with that from the plume.

The density of air varies depending on the weather conditions. **Figure 4** shows the air density dependency of the spectra for a plume 200 m high containing I-131. If the density of the air is increased by half without changing the composition, the spectrum changes a little in shape but varies almost inversely with the density, as it is 1/1.44 at the peak of 0.365 MeV. This is because the attenuation in the air increases due to the increase in the density.

2. Response of $LaBr_3$ Detector

We calculated the absorption energy in the detector based on the assumption that photons that have the spectrum calculated in Section II-1 in the air in the NTP state enter a $\phi 1.5'' \times 1.5''$ $LaBr_3$ detector uniformly, and also corrected the resolution using the detector performance achieving a FWHM of 3.5% relative to 0.662 MeV photons. Based on these calculations, we determined the detector response in the case where various nuclides were contained at a concentration of 1 Bq/cm^3. **Figure 5** shows the detector response according to I-131, I-132, Cs-134, Te-132, and Xe-133 when the plume height is 100, 200, and 500 m. In the case of nuclides other than Xe-133, the detector response is also shown for a height of 1.3 m from the soil surface, where respective nuclides are distributed uniformly over a wide area on the soil surface at a density which makes major peak count rates almost identical. The lower the energy of the gamma rays emitted from various types of radionuclides, the smaller the contribution made by distance becomes, showing that the plume height has a smaller impact. The detector response to Te-132 shows peaks around 0.04 and 0.03 MeV only in the case of soil surface distribution. These peaks are due to the gamma rays emitted by the decay of Te-132 and the K X-ray of I. Since the contribution from a region near the detector is relatively large in the case of soil surface distribution, the contribution of the low-energy gamma rays and X-rays is reflected in the pulse height distribution.

Since La, which is a major constituent of the detector, contains 0.09% of La-138 with a

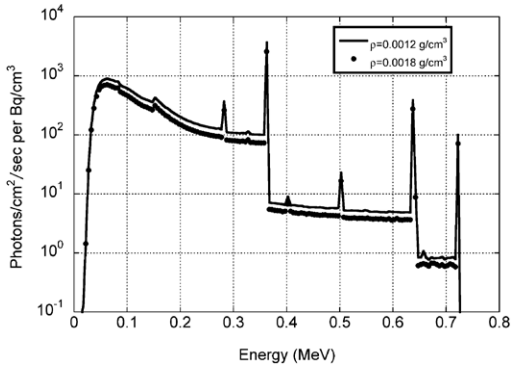
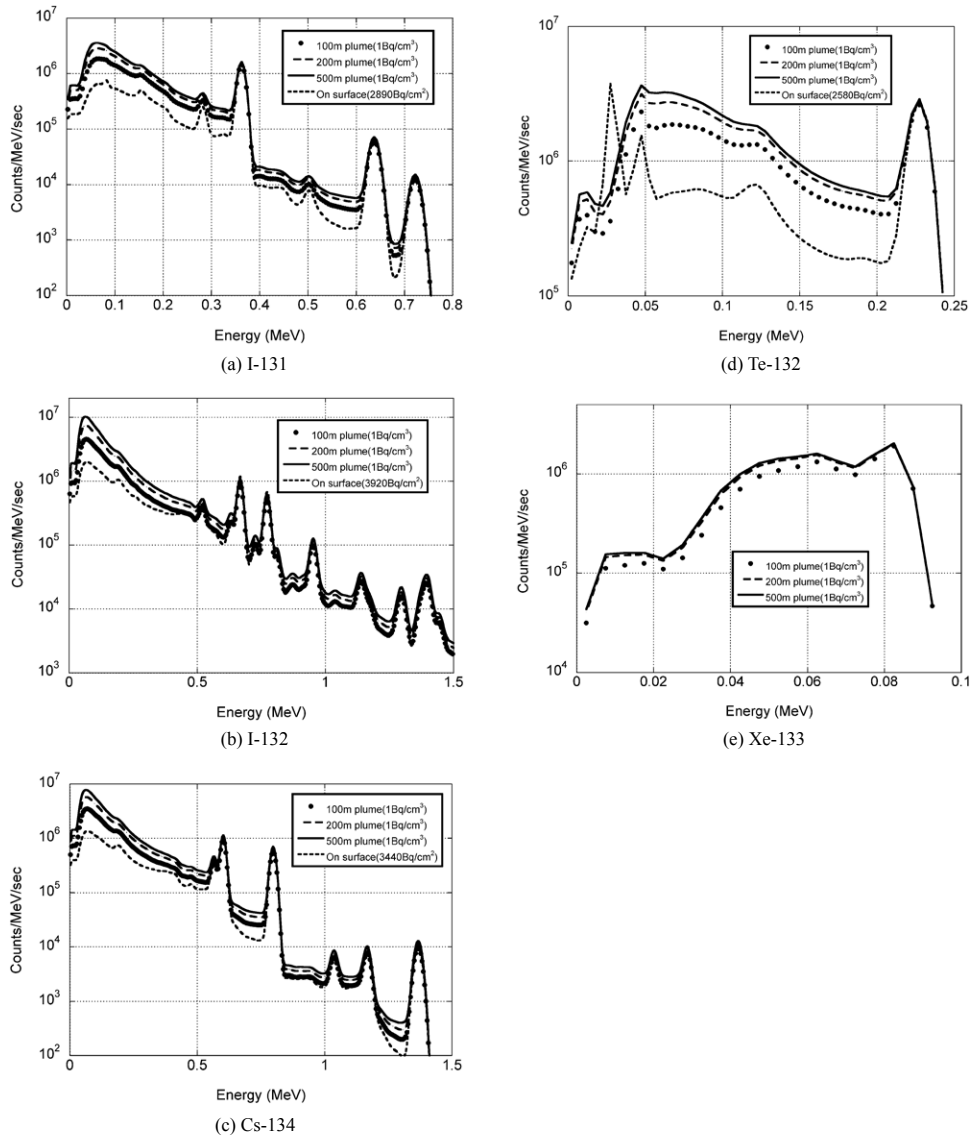


Figure 4 Air density dependence of photon spectrum at 1.3 m above soil surface from 200 m plume containing I-131



(a) I-131 (d) Te-132

(b) I-132 (e) Xe-133

(c) Cs-134

Figure 5 Comparison of $LaBr_3$ response 1.3 m above soil surface from I-131(a), I-132(b), Cs-134(c), Te-132(d) and Xe-133(e) contained in plume or deposited on soil surface

half-life of 1.06×10^{11} years, the $LaBr_3$ detector has a unique background consisting of the gamma and beta rays emitted as a result of the decay of La-138. **Figure 6** shows the background pulse height distribution of the detector, which was calculated with egs5 based on the decay mode of La-138, assuming that La-138 was distributed uniformly in the $LaBr_3$ detector. The detector response calculated had very similar amplitude and shape to the measurement value obtained by placing the $LaBr_3$ detector in a lead shield, and it can be said that the result of calculation with egs5 reproduced the background due to decay of La-138. In the comparison of pulse height distributions performed in Chapter III, the pulse height distributions calculated with egs5 include this background unique for the $LaBr_3$ detector.

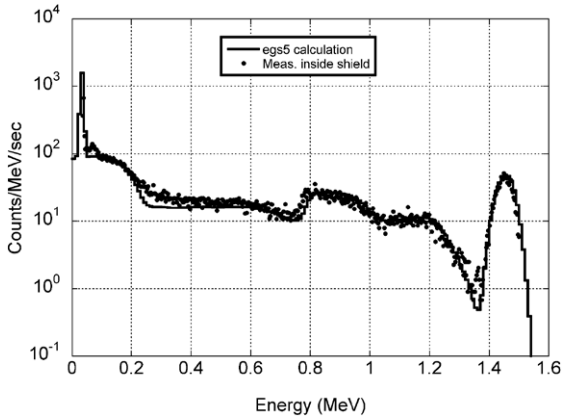


Figure 6 Comparison of $LaBr_3$ detector response due to La-138 between egs5 calculation and measurement inside Pb shield

III. Concentration of Radionuclides in Plume

1. Method of Estimating Radionuclide Concentrations

From the measurements of the pulse height distributions, Te-132, I-132, I-131, Xe-133, Cs-134, and Cs-136 were detected. To estimate the concentrations of respective radionuclides, we used the peaks corresponding to the energies shown in **Table 1**, which were considered to contain fewer gamma rays emitted from other nuclides.

The concentrations of respective nuclides were estimated from the pulse height distribution of the $LaBr_3$ detector according to the following procedures:

(1) To determine the peak count rate from which the tail of the nuclide identified from the measurement value is deducted (A).

(2) To determine the corresponding peak count rate from which the tail is deducted from the detector responses calculated for respective nuclides, as is the case with the measurement value (B). This value is the peak count rate per Bq/cm^3.

(3) The radionuclide concentration (Bq/cm^3) is obtained from B/A.

2. Comparison of the Pulse Height Distributions of $LaBr_3$

Figures 7 and **8** show the comparison between the measured pulse height distributions and the reconstructed distributions using the detector responses in the case where the plume height was 200 m, and where radionuclides deposited uniformly on the ground surface over a wide area at Koriyama-Higashi IC of the Ban-Etsu Expressway (March 15, 15:25–15:26) and at Abukuma-Kogen SA of the Ban-Etsu Expressway (March 15, 15:51–15:52). The reconstruction was based on the assumption that Cs-137, with the same concentration as Cs-134, was contained in the plume. Since Xe-133 does not deposit on soil surfaces, it was considered that Xe-133 existed in the plumes even in the case of calculation of soil surface contamination. In the comparison at Koriyama-Higashi IC, the calculated values had a tendency to be overestimated in the range from 0.081 MeV to 0.228 MeV, but it can be said that the measured pulse height distributions were very similar to the reconstructed distributions. On the other hand, in the comparison at Abukuma-Kogen SA, the situation is different from Koriyama-Higashi IC in that the scattering portion of the pulse height distribution reconstructed at low energy was

Table 1 Gamma-ray energies used for analysis

Radionuclide	Gamma-ray energy
Xe-133	81keV
Te-132	228 keV
1-131	365 keV
Cs-134	605 keV
1-132	955 keV
Cs-136	1,048 keV

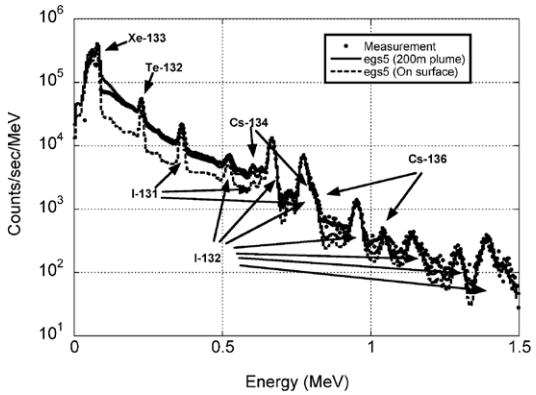


Figure 7 Comparison of $LaBr_3$ pulse-height distribution between measurement at Koriyama-higashi IC at 15:25 March 15, 2011 and egs5 calculation

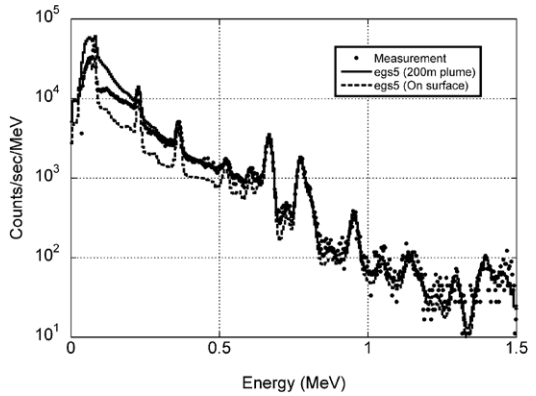


Figure 8 Comparison of $LaBr_3$ pulse-height distribution between measurement at Abukuma-kogen SA at 15:51 March 15, 2011 and egs5 calculation

significantly larger than the measured one.

Based on the results of the analysis of radionuclides mention in the next section 3, it was clear that the concentrations varied markedly with time in both areas, which were located west of the Fukushima Daiichi Nuclear Power Plant. The measurement at Abukuma-Kogen SA was performed after a certain period of time had elapsed following the passage of the plume containing high concentrations of radionuclides. Therefore, it was surmised that this difference could be attributed to the contribution of radionuclides deposited on ground surfaces, etc. when

the passing plume containing high concentrations of radionuclides was relatively large.

3. Estimation of the Concentrations of Radionuclides in Plumes

From the measurement values obtained on March 15, we estimated the concentrations of radionuclides from the pulse height distributions measured at six places, shown in **Table 2**, which were located 38.3–57.4 km west of the Fukushima Daiichi Nuclear Power Plant. **Table 3** shows the concentrations of radionuclides, and measured peak count rates corresponding to respective radionuclides contained in the plumes. Although the radionuclide concentration becomes higher as the effective height of the plume is lower, the concentration is as low as 30% in the case of I-132, which means the largest difference occurs at 100 m and 500 m. The lower

Table 2 Place and time used for the estimation of radionuclide concentration in plume

Place of measurement	Time	Measurement time	Direction and distance from Fukushima No. 1 NPP
Adatara SA	15:03	60 sec.	West 56.0 km
Motomiya IC	15:11	60 sec.	West 57.4 km
Koriyama-higashi IC	15:22	60 sec.	West 52.4 km
Miharu PA	15:33	60 sec.	West 47.5 km
Funehiki-Miharu IC	15:42	60 sec.	West 41.7 km
Abukuma-kogen SA	15:51	60 sec.	West 38.3 km

Table 3 Estimated upper limit of radionuclide concentration inside plume

Nuclide	Concentration of radionuclide Bq/cm^3					
	Adatara SA (15:03)			Motomiya IC (15:11)		
	100 m plume	200 m plume	500 m plume	100 m plume	200 m plume	500 m plume
Te-132	0.00224	0.00209	0.00206	0.00732	0.00683	0.00675
1-131	0.00190	0.00173	0.00169	0.00564	0.00514	0.00503
1-132	0.00290	0.00245	0.00226	0.00966	0.00814	0.00749
Xe-133	0	0	0	0	0	0
Cs-136	0.000102	0.0000867	0.0000810	0.000418	0.000355	0.000331
Cs-134	0.000268	0.000237	0.000227	0.00105	0.000924	0.000888
Nuclide	Concentration of radionuclide Bq/cm^3					
	Koriyama-Higashi IC (15:25)			Miharu PA (15:33)		
	100 m plume	200 m plume	500 m plume	100 m plume	200 m plume	500 m plume
Te-132	0.0103	0.00962	0.00952	0.00425	0.00396	0.00392
1-131	0.00847	0.00772	0.00755	0.00374	0.00340	0.00333
1-132	0.0138	0.0116	0.0107	0.00559	0.00471	0.00434
Xe-133	0.120	0.118	0.118	0.03306	0.03238	0.03238
Cs-136	0.000418	0.000355	0.000331	0.000158	0.000134	0.000125
Cs-134	0.00170	0.00150	0.00145	0.000945	0.000835	0.000802
Nuclide	Concentration of radionuclide Bq/cm^3					
	Funehiki-Miharu IC (15:42)			Abukuma-kogen SA (15 :51)		
	100 m plume	200 m plume	500 m plume	100 m plume	200 m plume	500 m plume
Te-132	0.00353	0.00329	0.00325	0.00285	0.00266	0.00263
1-131	0.00329	0.00300	0.00293	0.00212	0.00193	0.00189
1-132	0.00478	0.00402	0.00370	0.00345	0.00291	0.00268
Xe-133	0.0317	0.0311	0.0311	0.0124	0.0122	0.0122
Cs-136	0.000195	0.000166	0.000155	0.000102	0.0000867	0.0000810
Cs-134	0.000733	0.000647	0.000622	0.000697	0.000616	0.000592

the energy of the gamma rays emitted from radionuclides, the smaller the difference is. **Figure 9** shows the variation in concentration with time, provided the effective plume height is 200 m. Although it seems that Xe-133, which does not deposit on the soil surface, etc. best reflects the plume condition, the energy of the gamma rays emitted from Xe-133 is as low as 81 keV. As a result, the detection limit of the concentration is high due to the scattered photons from other nuclides, and it may possibly be zero at a place where the concentration of Xe-133 is low. From this figure, it is estimated that the plume that traveled westwards arrived at a place located approximately 50 km from Fukushima Daiichi Nuclear Power Plant at around 3:00 p.m., and then reached its highest concentration around 3:25 p.m. Furthermore, the concentration of I-131 dropped to about a quarter of the maximum concentration after 3:50 p.m.

Since it was drizzling during the measurement, it is considered that some radionuclides contained in the plume deposited on the ground surface as the plume passed. Therefore, the concentrations shown in Table 3 are the upper limits of the concentrations of radionuclides contained in the plumes. Since the measurement described in reference 1 used for estimation is not a continuous measurement at a fixed point, it is difficult to estimate the contribution of deposition directly from the measured pulse height distributions. In addition, although it is considered possible to estimate the contribution of the deposition on the soil surface from the difference in the ratio between unscattered and scattered photons (Figure 7), if the measurement is performed over a wide area consisting of a single material, such as soil, it is too difficult to apply this method because the measurement sites located on an expressway are quite different from such conditions. At Adatara SA on the Tohoku Expressway, which is one of the six estimation sites, the measurement was also performed on March 16 (twice), March 17 and April 8. Although the measurements were performed at the same service area, they were not performed continuously at a fixed point. Therefore, we attempted to estimate the influence of deposition from the change in the point of measurement under the assumption that measurements were not conducted at the same exact point. **Table 4** shows the measured peak count rates of the respective nuclides, and **Figure 10** shows the change over time in the peak count rates of Te-132, I-131, Xe-133, and Cs-134. Since the peak count rates of these nuclides increased on the evening of March 16, it would appear that another plume arrived during this period. It was also found that gaseous I-131 as well as particulate Te-132 and Cs-134 showed different change trends over time. In the case of I-131 and Cs-134, the values for which the half-life was corrected from the values measured on April 8 based on the assumption of 100% deposition on the soil surface were larger than the values measured on March 17. It is possible

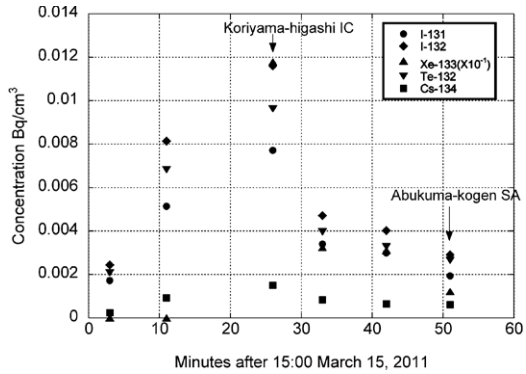


Figure 9 Estimated time variation of concentration 38.3–57.4 km west of the Fukushima Daiichi Nuclear Power Plant between 15:03 and 15:51 in March 15, 2011

Table 4 Measured peak counts rate at Adatara SA

Nuclide	Measured peak count rates (cps)				
	March 15 15:03	March 16 12:18	March 16 17:30	March 17 11:28	April 8 13:13
Te-132	80.1 ± 3.3	400 ± 6	660 ± 7	381 ± 6	0 ± 0
1-131	47.2 ± 2.6	16 ± 4.5	80.9 ± 5.2	12.2 ± 4.2	2.2 ± 0.4
1-132	5.45 ± 0.8	19.1 ± 1.4	31 ± 1.7	17.3 ± 1.4	0 ± 0
Xe-133	0 ± 0	0 ± 0	33.1 ± 6.4	0 ± 0	0 ± 0
Cs-136	1.1 ± 0.5	2.77 ± 0.92	6.7 ± 1	3.97 ± 0.63	1.18 ± 0.13
Cs-134	5.3 ± 0.3	30.3 ± 0.8	41.2 ± 0.9	23.4 ± 0.7	33.1 ± 0.5

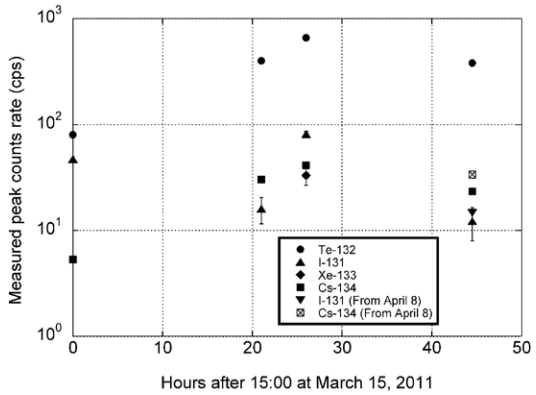


Figure 10 Time variation of measured peak counts rate for Te-132, I-131, Xe-133 and Cs-134 at Adatara SA

that the deposition due to the arrival of the plume on or after March 17 had an influence on this result. Assuming that the result of the measurement performed on March 17 was due to I-131 (which exhibited 100% deposition), the count rate of I-131 due to the deposition at the time of measurement on March 15 was 13.8 cps, which was equivalent to the peak count rate of 29% measured on March 15.

In the case of Miharu PA and Abukuma-Kogen SA on the Ban-Etsu Expressway, only the measurement result obtained on April 8 was available on and after March 15. The peak count rates of I-131 obtained on March 15 after correcting the half-life were 26.3 cps and 16.0 cps, which were 28% and 30% of the peak count rates measured on March 15, respectively. As shown in the case of Adatara SA, it is possible that the contribution of the deposition estimated according to this method contained the deposition due to the plume that arrived on and after March 15. Therefore, it would appear that these values are the maximum values caused by the influence of deposition.

Since the measurements at Abukuma-Kogen SA, which are compared in Figure 8, were conducted starting at 3:51 p.m., it would appear that measurements were conducted more than 25 min after the plume with the maximum concentration had passed. Therefore, it can be estimated that the contribution of the radionuclides deposited on the soil surface reached 30%, as shown above. It is thought that this is the cause of the low energy scattering component of the pulse height distributions reconstructed only by the radionuclide in the plume being larger than the measured ones.

IV. Conclusions

We estimated the concentrations of radionuclides contained in the plumes when they passed over, from the peak count rates corresponding to the radioisotopes of the $LaBr_3$ detector measured on the expressways inside and outside Fukushima Prefecture on March 15, 2011, and the corresponding peak count rates in the plumes containing the radioisotopes calculated with egs5 at a concentration of 1 Bq/cm^3. The pulse height distributions of the $LaBr_3$ detector reconstructed using the estimated radionuclide concentrations reproduced the measured values almost perfectly at places where the contribution of the deposition was considered to be small, thus the present method is adequate for estimation. From the radionuclide concentrations estimated at six places located 41.7–57.4 km west of Fukushima Daiichi Nuclear Power Plant, we estimated the change over time as the plume passed over. Although it is difficult to estimate the influence of deposition because it is affected by the plumes that arrived later as well as the conditions around the places of measurement, we could estimate that the contribution of the deposition of I-131 was up to 30%, as a result of considering the values obtained at three locations where measurement was performed at the same place on and after March 15.

Although in this paper we used $LaBr_3$, whose pulse height distribution data were available, it is also possible to apply the same method to a NaI(Tl) scintillation detector. If the pulse height distribution data obtained at a monitoring post using a NaI(Tl) scintillation detector were to be available, it would be possible to evaluate the influence of deposition accurately from the change over time in the peak count rates corresponding to respective nuclides. It is expected to estimate the concentration of I-131 in the plume immediately after the accident, as well as the information on the change in its concentration over time, by the pulse height distribution data at a monitoring post together with an atmospheric dispersion model and to use obtained results for estimation of internal exposure by I-131.

References

1) H. Matumura, K. Saito, J. Ishioka, Y. Uwamino, “Diffusion of radioactive materials from Fukushima Daiichi Nuclear Power Station obtained by Gamma-ray measurements on expressways,” *Trans. At. Energy Soc. Jpn*, 10, 152-162 (2011). [in Japanese]
2) Y. Namito, H. Nakamura, A, Toyoda, K. Iijima, H. Iwase, S. Ban, H. Hirayama, “Transformation of a system consisting of plane isotropic source and unit sphere detector into a system consisting of point isotropic source and plane detector in Monte Carlo calculation,” *J. Nucl. Sci. Technol*., 49, 167-172 (2012).
3) H. Hirayama, Y. Namito, A. F. Bielajew, S. J. Wilderman, W. R. Nelson, *The EGS5 Code System*, SLAC-R-730 (2005) and KEK Report 2005-8 (2005).
4) K. F. Eckerman, J. C. Ryman, *External Exposure to Radionuclides in Air, Water, and Soil*, Federal Guidance Report No. 12, EPA-402-R-93-081 (Sep. 1993).