Deduction of useful information from dose rate monitoring data under emergency conditions

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The surface concentrations and release rates of radionuclides were estimated from gamma dose rates by using a rigorous modeling approach based on the relation between the dose rates and the concentrations. It was shown by applying this method to non-accidental cases that the surface run-off of deposited radionuclides can be analyzed. The concentrations of deposited radionuclides were in good agreement with the observations, although a difference in estimations among monitoring stations was observed due to radionuclides deposited on trees. The release rates estimated with the dose rates at the boundary of the Fukushima Daiichi Nuclear Power Station were in reasonable agreement with JAEA's estimations. However, sensitivity analyses indicated that these estimations have a large uncertainty.

Key Words: *gamma dose rate, radioactivity concentration, release rate, radon decay products,*

1. Introduction

Environmental gamma dose rates are continuously measured around nuclear power plants in order to identify releases of radionuclides from these plants. The Fukushima Daiichi nuclear power station (FDNPS) accident in Japan triggered by a big earthquake on March 11, 2011 caused the release of a huge amount of radionuclides into the atmosphere and ocean[1)].

Subsequent to this accident, dose rate monitoring has been considerably enhanced, and many monitoring stations and posts (MSs and MPs) have been deployed not only in the vicinity of the nuclear power plants but also at distant locations to reinforce the monitoring capability in case of emergency situations. Although this increase in the number of dose rate monitoring points significantly improves emergency preparedness, it still remains as a substantial weakness of the emergency monitoring that the dose rate is only one out of many other monitoring factors that are indispensable for emergency decision- making. These factors include the concentrations and composition of radionuclides in the air, soil, and environmental media. The measurement of these parameters is time-consuming and needs considerable manpower, because, in most cases, samples collected from the field need to be delivered to the laboratory for measurement. On the other hand, the measurement of the dose rates is conducted in-situ, and hence, it is considered as providing continuous real-time data. In order to exploit the usefulness of dose rate measurements, deducing more information from the measured dose rate would prove of greater assistance in emergency situations.

In addition, release rates of radionuclides also provide important information to evaluate the radiological doses that the public have been exposed to along with the magnitude of the accident. Therefore, many researchers have estimated the release rates of radionuclides from environmental data such as air concentrations from dust sampling and dose rates measured in the regional-scale

area[1),2)]. These estimations are considered to insufficiently describe the release trend primarily due to the limited data available, and they can be supplemented by knowledge of the dose rate at the site boundary.

Thus, the purpose of this study is to explore possibilities of deducing concentrations, composition, and release rates of radionuclides from the dose rate monitoring data by using a rigorous modeling approach based on the relation between the dose rate and the concentrations. In this study, we test this approach for the radon decay product deposition cases, and we apply it to the dose rate monitoring data from the FDNPS accident. Futher, we test a simple method based on a plume model to estimate the release rate from the dose rate monitoring at the site boundary. The limitations of this simple method are also discussed.

2. Method

(1) Gamma dose rate model

The gamma dose rate dD_i (nGy h^{-1}) from a radionuclide i in air or on the ground surface is calculated by integrating the dose contribution from each air/ground segment j as

$$dD_i = \sum_j C_{i,j} \cdot dv \cdot \frac{K}{4\pi r_j^2} \cdot E_i \cdot \mu_{air}(E_i) \cdot S \,, \tag{1}$$

with

$$S = \prod_m^N \left(\exp\left(-\mu_m(E_i) \cdot r_{m,j}\right) \cdot B_m\right). \tag{2}$$

The term C denotes the air/surface radioactivity concentration (Bq m^{-3} or Bq m^{-2}), dv the segment size (m^3 or m^2), E the gamma-ray energy (eV), K the conversion factor of the unit of energy from eV for E to nGy h^{-1} for dD, r_j the distance of the gamma-ray path (m), μ the linear attenuation coefficient (m^{-1}), S the term representing attenuation and build-up effects in air and materials, and B the build-up factor. The subscript m indicates air or materials such as soil or concrete. This model can explicitly express the geometry of source distribution around each MS and the height of the NaI(Tl) scintillation detector on the basis of field surveys. This model has been tested for non-accidental and no-rain cases to show that it can reasonably reproduce the background dose rate from the radioactivity in the natural environment and in building materials[3)].

(2) Estimation of surface concentration

The radionuclides are assumed to remain on the ground without being affected by surface run-off, inflow, or infiltration. Therefore, the temporal changes in the surface concentration C of the radionuclides are governed by the following radioactive decay equation:

$$\frac{dC_{i_2}}{dt} = -\lambda_{i_2} C_{i_2} + \lambda_{i_1} C_{i_1} \,, \tag{3}$$

where λ denotes the decay constant (s^{-1}) and i_2 denotes the progeny nuclide of i_1. The conversion factor Γ ((nGy h^{-1})/(Bq m^{-2})) from the unit surface concentration to dose rate was calculated using the gamma dose rate model. In this study, the radionuclides are assumed to be distributed uniformly on horizontal surfaces that catch raindrops. The calculation domain is a 60-m square that is divided into 0.1-m square segments. Thus, the increase in dose rate due to the ground shine dD_{cal} (nGy h^{-1}) can be expressed by the surface concentrations and the conversion factors as

$$dD_{cal} = \sum_i C_i \Gamma_i \,. \tag{4}$$

The surface concentrations of radionuclides were determined by using a least-square method in which the square sum of differences in the increment of dose rate due to ground shine between observations and calculations was minimized.

a) Non-accidental case

Dose rate increases in non-accidental cases during rain events are attributed to the deposition of radon decay products, ^{214}Pb and ^{214}Bi, in rain water[4),5)]. Here, the dose rate increase due to rain (DRI_R) was defined as the dose rate increment from the background dose rate contributed by the radionuclides permanently occurring in soil and in building materials. It was reported that the ^{218}Po concentration in raindrops deposited on the ground surface was a few orders of magnitude less than ^{214}Pb and ^{214}Bi concentrations[5)]. Hence, ^{218}Po was assumed not to contribute to the temporal change in the surface concentration.

This estimation method was applied to routine observations of dose rates during the period 2004–2010 made using $3''\phi \times 3'$ NaI(Tl) scintillation detectors at seven near-by MSs around the Hamaoka nuclear power plant in Shizuoka, Japan. Rain events that met both of the following two criteria regarding DRI_R were selected: the difference in rain intensity among MSs is less than 0.5 mm during each 10-min period, and the difference in total

rainfall is less than 10 mm in a rain event. It has been reported that the radon progeny concentrations in raindrops differ depending on rain intensity[4),5)]. Therefore, the purpose of setting these criteria is to select rain events in which ^{214}Pb and ^{214}Bi depositions are considered to be uniform in the area of interest in which MSs are located.

b) Accidental case

A trial was carried out to estimate radionuclide composition from temporal change in the dose rate attributed to deposited radionuclides by fitting decay curves. The major radionuclides of ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs, ^{136}Cs, and ^{137}Cs were considered in the calculation. The dose rate measured with 2″ϕ × 2″ NaI(Tl) scintillation detectors at six near-byMSs (about 115 km from FDNPS) in Ibaraki, Japan (obtained from authorities of the Ibaraki Prefectural Government) was used. The analysis period was chosen as the interval from 13:20 JST on March 15 to 1:20 JST on March 16, when no significant rain was observed and the radioactive plume did not pass by these MSs. Since this 12-hour period is considerably shorter than the half-lives of most of the major radionuclides, the major radionuclides were divided into three groups based on their half-lives: (I) ^{132}I (half-life ≈ 2 hours), (II) ^{132}Te (half-life ≈ 3 days), and (III) ^{131}I, ^{134}Cs, ^{136}Cs, and ^{137}Cs (half-life of each > 1 week). The surface concentrations of the representative radionuclides of each group (i.e., ^{132}I, ^{132}Te, and ^{131}I) were estimated by assuming that E and μ in Eqs. (1) and (2) were the values of the representative radionuclides.

(3) Estimation of release rate

The purpose of this analysis is to demonstrate how accurately the release rate is estimated from dose rate measurements at the site boundary. The release rate was estimated using an atmospheric dispersion model and the gamma dose rate model described above. The atmospheric dispersion model is a particle random-walk model with a Lagrangian particle dispersion coefficient, and it calculates the distribution of radionuclides. The release rate R (Bq h^{-1}) is obtained as the ratio of the observed (dD_{obs}) to the calculated (dD_{cal}) dose rates at the measurement point, as follows:

$$R = R_0 \cdot dD_{obs} / dD_{cal}\,, \tag{5}$$

where dD_{cal} denotes the dose rate calculated with R_0 = 1 Bq h^{-1}. As a test case, the maximum dose rate value of 11.9 mGy h^{-1} measured at the main gate of FDNPS at 9:00 JST on March 15 was used as the observed dose rate dD_{obs}. The release height was inferred to be 50 m above ground by photos obtained using a live camera by Fukuichi[6)]. In the reference case, wind was assumed to directly blow to the main gate, which is 1 km downwind from the release point. The observed wind speed of 1.5 m s^{-1} and neutral atmospheric stability were used. The composition of major radionuclides in air was inferred by calculating the air concentration of each radionuclide from the HPGe dose rate measurements recorded by the Japan Chemical Analysis Center[7)].

3. Results and discussion

(1) Surface concentration in non-accidental case

The relationship between the surface concentrations of the radon decay products averaged for all rain events and the fractional contribution of DRI_R from the radionuclides on the MS roof and paved area is shown in Fig.1. Here, the area over which the radon decay products were deposited was assumed to be composed of MS roofs, paved areas, and soil areas. The rain events subjected to analysis were selected by criteria in which ^{214}Pb and ^{214}Bi depositions were considered to be the same for all the MSs. Nevertheless, the estimated concentrations of ^{214}Bi differ by approximately 15%. In addition, we find a negative correlation between the estimated concentration and dose rate contribution from the MS roofs and paved areas.

The MS roofs are coated with rubber and have a gentle slope, which implies higher efficiency in the surface run-off of rain water than that in the soil area. Thus, the amount of radionuclides existing on MS roofs and paved areas is considered to be less than that in the

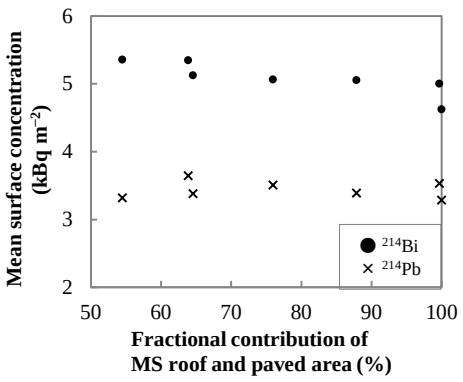

Fig. 1. Relationship between the mean surface concentration and the fractional contribution of dose rate increase from nuclides on MS roof and paved area due to rain.

soil areas, considering that the negative correlation observed for ^{214}Bi is caused by the surface run-off.

On the other hand, such a negative correlation was not found in the case of ^{214}Pb. In soil areas, the attenuation becomes larger due to the infiltration of the radionuclides into the soil. Therefore, the concentration of ^{214}Pb for the MSs with a larger dose rate contribution from soil areas is thought to have been underestimated. It is probable that the zero correlation observed for ^{214}Pb is caused by the competition between the surface run-off and the infiltration.

(2) Surface concentration in accidental case

The estimated surface concentration of the representative radionuclides was 100 kBq m^{-2} in Ibaraki, although the estimated values are different among MSs by a factor of 5 to 7 (see Table 1). If the observed activity ratio (^{131}I:^{134}Cs:^{137}Cs:^{136}Cs= 10:1:1:0.2)[8] in air on March 15 is used, the surface concentration of ^{137}Cs is estimated to be 4 kBq m^{-2}, which is in good agreement with the observation (5 kBq m^{-2}) on September 15.

The increases in dose rates due to cloud shine (DRI_C) and
ground shine (DRI_G) were respectively defined as the difference of the maximum dose rate during a plume passage and the dose rate after the passage with respect to that before the passage (see Fig. 2). The relationship between DRI_C and DRI_G is shown in Fig. 3, where the maximum to minimum ratio of DRI_C is observed to be 1.7, while that of DRI_G is observed to be 5.5. This result indicates that the amount and geometry of the deposition have large MS-to-MS variations even though the air concentration appears to be rather uniform. Therefore, the difference in estimated surface concentrations among MSs can be attributed to the difference in the contributions from the deposited radionuclides. At Ishigami and Toyooka, the DRI_G and surface concentration are larger than those for any other MSs. It has been reported in field investigations that the NaI detectors are overshadowed by trees, while the attenuation due to the building materials around a MS is not very different among MSs. Therefore, additional deposition on trees may have contributed to the large value of DRI_G, thereby resulting in the large concentration estimated in the present method.

Table 1. Estimated surface concentration (kBq m^{-2}) of the representative radionuclides and surface-concentration to dose-rate conversion factors Γ ((nGy h^{-1})/(Bq m^{-2})) for ^{131}I.

MS	^{131}I	^{132}Te	^{132}I	$\Gamma_{\mathrm{I\text{-}131}}$
Ishigami	240	140	220	0.70
Toyooka	130	66	120	0.71
Funaishikawa	45	19	41	0.96
Oshinobe	100	51	97	0.69
Muramatsu	92	36	81	0.69
Mitsubishi	56	26	54	0.68

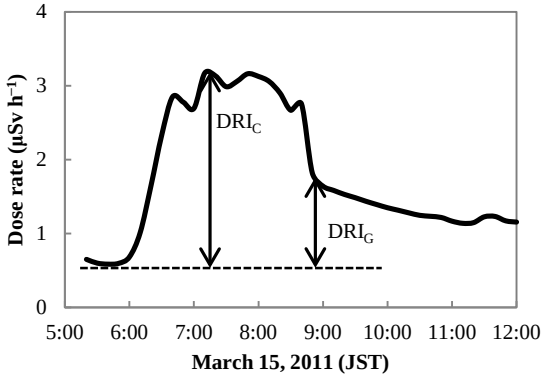

Fig.2 Example of dose rate change (from Ibraki Prefectual Government).

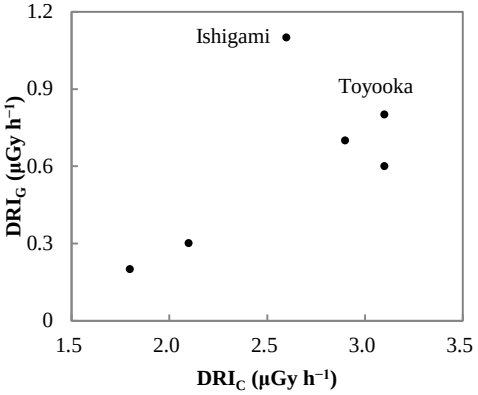

Fig. 3. Relationship between the dose rate increase due to ground shine (DRI_G) and that due to cloud shine (DRI_C).

(3) Release rate

The release rates of ^{133}Xe, ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs, and ^{137}Cs at 9:00 JST on March 15 were estimated to be 98, 1.7, 7.0, 2.0, 0.21, and 0.21 PBq h^{-1}, respectively. Katata et al. (2012) have estimated the release rates of ^{131}I and ^{137}Cs as 3 and 0.3 PBq h^{-1}, respectively[2], and these figures support the present results.

However, it should be pointed out that the input data used, such as the release height, the wind speed/direction, and the atmospheric stability, may have significant

uncertainty. Therefore, sensitive analyses of these parameters were conducted (see Fig. 4). A change in the release height from 20 to 150 m caused changes in the estimated release rate by a factor of 0.68 (20 m) and 4.2 (150 m) when the atmospheric stability was assumed to be neutral; and by factors of 1.0 (20 m) and 1.5 (150 m) for an unstable atmosphere. A change in the stability from neutral to unstable resulted in a increase in the estimated release rate by a factor of 3.2 when the release height was 20 m. The estimation increases by more than 10 times, when the wind direction turns 22.5° clockwise or counter-clockwise. We conclude that the estimation of release rate from the dose rate measurement at a given point suffers from serious error mainly due to the uncertainty in wind direction.

4. Conclusion

A series of experimental trials were performed to deduce as much information as possible from the existing radiation monitoring data. A rigorous gamma dose rate model was applied, and we found that the surface run-off and the infiltration of deposited radionuclides can be analyzed from dose rate data obtained during rain. It is expected that the characteristic of surface run-off and the infiltration obtained by this method are applicable to accidental radiation release cases. The concentrations of the deposited radionuclides were successfully estimated from the dose rate data obtained, although the additional information regarding the radionuclide composition was indispensable to estimate the concentration of each nuclide. The release rates estimated from the dose rate at the site boundary were 7.0 PBq h^{-1} for ^{131}I and 0.21 PBq h^{-1} for ^{137}Cs, which were in reasonable agreement with JAEA's estimations. However, the results of the sensitivity analysis of the release rate suggest that uncertainty in the wind direction would cause a considerable uncertainty in the estimated release rate.

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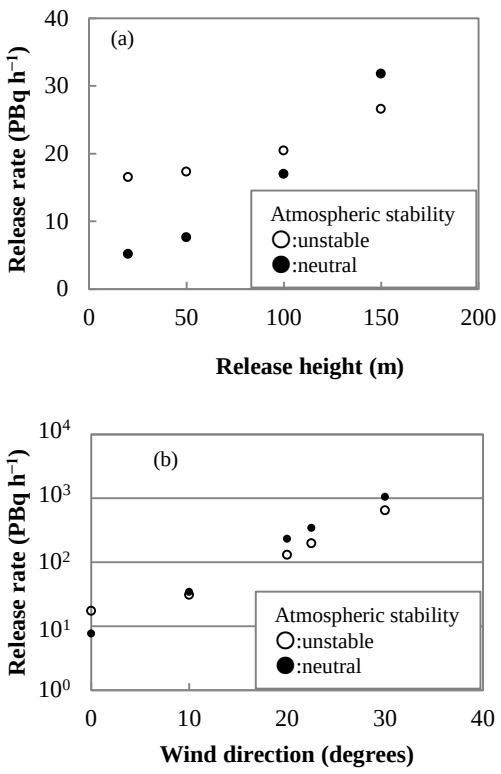

Fig. 4. Sensitive analyses of estimated release rate (a) to release height and (b) to wind direction.

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