Chapter 14
Investigation of Uncertainty in the Release Rates of ^{131}I and ^{137}Cs from Fukushima Dai-ichi NPS Estimated from Environmental Data

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Abstract It is an urgent task to estimate the release rate of airborne radionuclides and its uncertainty for the assessment of internal and external dose to the public from the Fukushima Dai-ichi Nuclear Power Station accident. We estimated the release rates of ^{131}I and ^{137}Cs by using a method commonly used in the previous studies for the source term estimation. This study investigated the sensitivity of the estimated release rate to the deposition parameters. It was found that the dry deposition velocity had only minor significance on the estimated release rate, predominately because of the inherently small contribution to total deposition when wet deposition occurred. The scavenging coefficient, on the other hand, showed a substantial effect on the estimated release rate. The release rate estimated with the small scavenging coefficient could be larger than that with the large scavenging coefficient when the calculated deposition was small because of weak rainfall intensity and the short travel time of the plume in the rainfall area. In contrast, the large scavenging coefficient also resulted in a large estimated release rate when the calculated deposition was small because heavy rainfall caused substantial depletion of the plume before the plume reached sampling sites. Adoption of a scavenging coefficient three times larger could result in increases of 10 and 21 fold in the estimated release rates of ^{131}I and ^{137}Cs, respectively, at maximum.

Keywords Release rate estimation • ^{131}I • ^{137}Cs • Sensitivity analysis • Atmospheric dispersion model • Environmental monitoring data

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14.1 Introduction

During the accident at the Fukushima Daiichi Nuclear Power Station (FDNPS), the released airborne radionuclides spread out over the Kanto and Tohoku regions and consequently caused land surface contamination. Assessment of radiological dose to the public requires information on the spatial and temporal evolution of the atmospheric transport of the radionuclides, land surface contamination, and radioactive composition. The most reliable information is environmental monitoring data. Although in the early phase of the accident atmospheric concentrations were measured, the spatial coverage of the measurements was too coarse to delineate the evolution of the radioactive plume and its radioactive composition.

Airborne measurements were made by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) in collaboration with the U.S. Department of Energy [1]. This work provided the distribution of land surface contamination for the medium- and long-lived radionuclides of ^{134}Cs and ^{137}Cs. The atmospheric dispersion and deposition of short-lived radionuclides, such as ^{131}I, which are important to evaluate radiological doses, still have a large uncertainty. In this situation, atmospheric dispersion simulations can play an important role of evaluating the atmospheric transport of radioactive plumes when source term information, such as release rates of radionuclides, is available.

To estimate the release rates, environmental monitoring data have been used with atmospheric dispersion models in several studies [2–5]. All studies with inverse methods indicate similar temporal variations in the atmospheric release rate of ^{131}I and ^{137}Cs despite of the use of different sets of monitoring data and atmospheric dispersion models. According to these studies, the release began in the morning of 12 March 2011, the largest release occurred on 15 March, and during other periods the releases were one to two orders of magnitude smaller than that on 15 March. Although this similarity implies the reliability of the estimated release rates, the uncertainties in the estimated release rate have not been discussed sufficiently. In our previous study, the uncertainty was preliminary estimated to be approximately a factor of 3 because of errors in the modeling of deposition processes [5]. In the absence of extensive environmental monitoring data, evaluation of the uncertainty of the source term is a difficult task. In these cases, the uncertainty of the source term can typically be evaluated by a sensitivity analysis of parameters that might affect the source term [6]. The purpose of this study is to estimate the release rates of ^{131}I and ^{137}Cs and their uncertainties. We investigated the sensitivity of the estimated release rates to the deposition parameters, that is, the dry deposition velocity and the washout coefficient. The analysis was conducted first with a simplified meteorological condition, and second with an under-realistic meteorological condition during the period of March 2011 that was calculated by a meteorological model.

14.2 Methods

14.2.1 Release Rate Estimation

Estimation of the release rate is based on the principle that the atmospheric dispersion model can calculate a spatial distribution of relative values of deposition rates on the ground although their absolute values are unknown. According to this principle, the ratio of deposition rate to the release rate can be assumed to be the same for both measurements and calculations as follows:

$$\left(\frac{Q_{\mathrm{r}}}{S_{\mathrm{r}}}\right)_{t,i} = \left(\frac{Q_{\mathrm{m}}}{S_{\mathrm{m}}}\right)_{t,i}, \tag{14.1}$$

where S_{m} is the release rate used for model calculation (Bq h^{-1}), Q_{r} is the measured deposition rate (Bq m^{-2} day^{-1}), and Q_{m} is the calculated deposition rate (Bq m^{-2} day^{-1}). The subscripts t and i denote the sampling time and sampling point, respectively. The release rate was estimated with Eq. (14.1) by solving it for S_{r} (Bq h^{-1}). To estimate a release rate at the time when discharged into the atmosphere, a release time was determined by calculating a fractional contribution that represents how much of the release during a unit-time period contributes to each sampling time and sampling point. The contributions were determined by using a series of calculations with an atmospheric dispersion model for every 12-h continuous release. The calculation for each release was carried out until the released radionuclides went out of the calculation domain.

Equation (14.1) does not strictly hold, primarily because of errors in the atmospheric dispersion calculation. For a given time, there might be more than two different release rate values estimated from independent monitoring data. In this case, a geometric mean was applied to estimate a single value for the time.

14.2.2 Atmospheric Dispersion Model

A Lagrangian particle random-walk model (LPRM) [7] coupled with a nonhydrostatic atmospheric dynamic model, MM5 [8], was used to calculate the dispersion of the radioactive plume released from FDNPP. MM5 calculates the three-dimensional wind field and the vertical diffusion coefficient. Radioactive decay, dry deposition, and wet deposition were calculated using LPRM. Iodine-131 and cesium-137 were modeled as passive tracers with half-lives of 8.04 days and 30 years, respectively. Dry and wet depositions were parameterized in terms of a dry deposition velocity and a washout (or scavenging) coefficient, respectively.

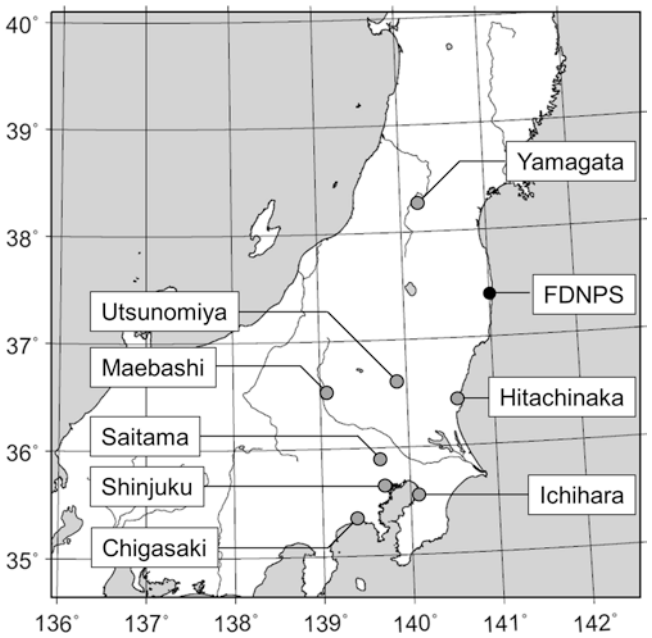

Fig. 14.1 Location of deposition measurement site on calculation domain

As a standard pair of these removal parameters, according to [9], the dry deposition velocity, V_d, was set to be 1.0 mm s^{-1} and the scavenging coefficient, Λ, was expressed as $\Lambda = \alpha(I/I_0)^{\beta}$, where I is the precipitation intensity, $I_0 = 1.0$ mm h^{-1}, $\alpha = 8.0 \times 10^{-5}$ s^{-1}, and $\beta = 0.8$. Dry deposition velocity and α can vary within a range of approximately 0.1–10 mm s^{-1} and 10^{-6}–10^{-3} s^{-1}, respectively, depending on the physicochemical characteristics of the nuclides, gas–particle partitioning, and particle-size distribution of aerosols (see [10, 11] for an overview). The removal parameters were changed by a factor of 3 in the sensitivity analysis.

14.2.3 Environmental Monitoring Data

To estimate the release rate, this study used the daily deposition measured at Chigasaki, Hitachinaka, Ichihara, Maebashi, Saitama, Sinjuku, Utsunomiya, and Yamagata with a 24-h sampling time from 18 March 2011 by MEXT [12]. The sampling sites are plotted in Fig. 14.1. The following criteria were set for data selection to eliminate the influence of resuspended radionuclides: the deposition rate of ^{131}I and ^{137}Cs is greater than 5.0×10^2 and 1.0×10^2 Bq m^{-2} day^{-1}, respectively. Altogether, 56 measured deposition rates of ^{131}I and ^{137}Cs were adopted in this study.

14.2.4 Calculation Condition

The calculation domain of the atmospheric dispersion was a 600-km square with a 6-km depth above the ground to cover most of Tohoku and Kanto Regions. The same physical processes of MM5 as the previous study [5] were used. For initial and boundary conditions and the four-dimensional data assimilation of the meteorological fields, the JRA-25 reanalysis data provided by Japan Meteorological Agency (JMA) and Central Research Institute of Electric Power Industry (CRIEPI) were used. Topography and land-use data were obtained from the United States Geological Survey global database. The radar-AMeDAS precipitation analysis data from JMA were used for the precipitation intensity in the wet deposition calculation. The MM5 calculation was conducted for the period from 09 Japanese Standard Time (JST), 8 March to 00 JST, 1 April. The dispersion of ^{131}I and ^{137}Cs from FDNPS started at 15 JST, 17 March and ended at 00 JST, 1 April. The release height was set to be 15 m above the ground. A constant release rate of 1 TBq h^{-1} was assumed.

14.3 Results and Discussion

14.3.1 Simplified Meteorological Condition

Before carrying out simulations with the calculated meteorological variables from MM5, this study conducted a series of preliminary simulations with a set of simplified meteorological conditions, that is, a uniform and constant wind field of 1 m s^{-1}, horizontally uniform but vertically varying vertical eddy diffusivities based on the calculated values of MM5, and prescribed intensities of uniform precipitation. The LPRM was the same as the main simulations. The purpose of these simulations is to discuss dependency of deposition and air concentration on deposition parameters.

The results are shown in Figs. 14.2 and 14.3. The dependency of dry deposition on the deposition velocity is quite simple. The dry deposition rate from the plume monotonously decreases with travel time, which is the elapsed time from release, mainly from horizontal and vertical diffusion causing air concentration decreases (Fig. 14.2a). The dry deposition rate for the 0.3 mm h^{-1} case is about 11.4 % of that for the 3 mm h^{-1} case at the travel time of 6 h. This ratio is almost same with the ratio of deposition velocities, but slightly modified by the change in the air concentration. The concentration for the 3 mm h^{-1} case is about 88 % of that of the 0.3 mm h^{-1} case (Fig. 14.2b). These results imply that the dry deposition process does not significantly reduce the amount of radioactivity in air within a period of several hours.

The dependency of wet deposition on the scavenging coefficient is somewhat complicated (Fig. 14.3a). A larger scavenging coefficient does not always cause greater deposition. Among the four cases, the largest scavenging coefficient, which corresponds to the rain intensity of 6.6 mm h^{-1}, resulted in the largest wet deposition

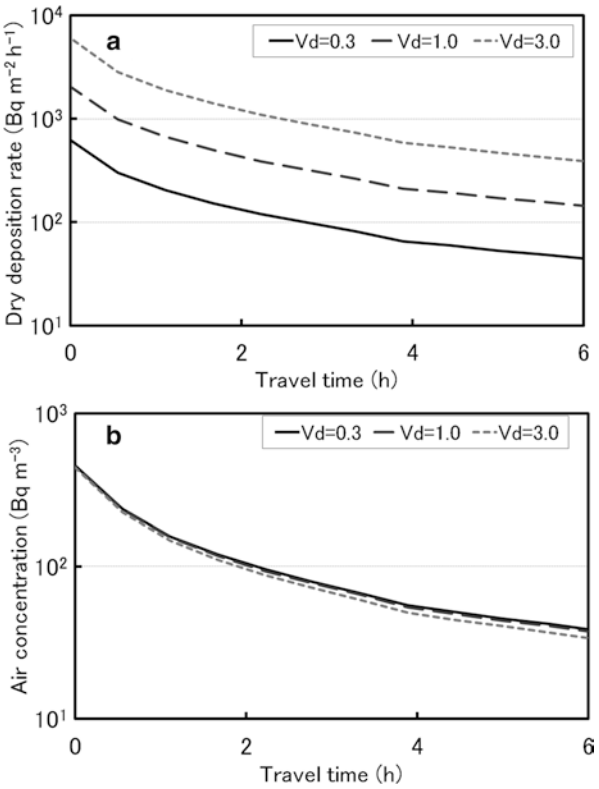

Fig. 14.2 Variation of the calculated values of (**a**) dry deposition rate and (**b**) air concentration with travel time. Dry deposition velocities, V_d, are 0.3, 1.0, and 3.0 mm s^{-1}

only in the first 0.6 h from the release. The wet deposition of this case becomes the smallest after a travel time of 1.7 h. On the other hand, the wet deposition in the case with the smallest scavenging coefficient (rain intensity of 0.37 mm h^{-1}) is the largest among the four cases after the travel time of 3.9 h. This complicated result is caused by the substantial depletion of the plume by scavenging (Fig. 14.3b). In the cases with large scavenging coefficient, the air concentration rapidly decreases with travel time.

14.3.2 Actual Meteorological Condition

14.3.2.1 Release Rate Estimation

The release rates of ^{131}I and ^{137}Cs were estimated for every 12-h time segment during 19 to 23 March and 25 and 29 to 30 March with their geometric standard deviations as shown in Fig. 14.4. Release rate could not be estimated for the period during which there was no measured deposition. The geometric standard deviation could not be calculated for the time segments in which only one monitoring datum was

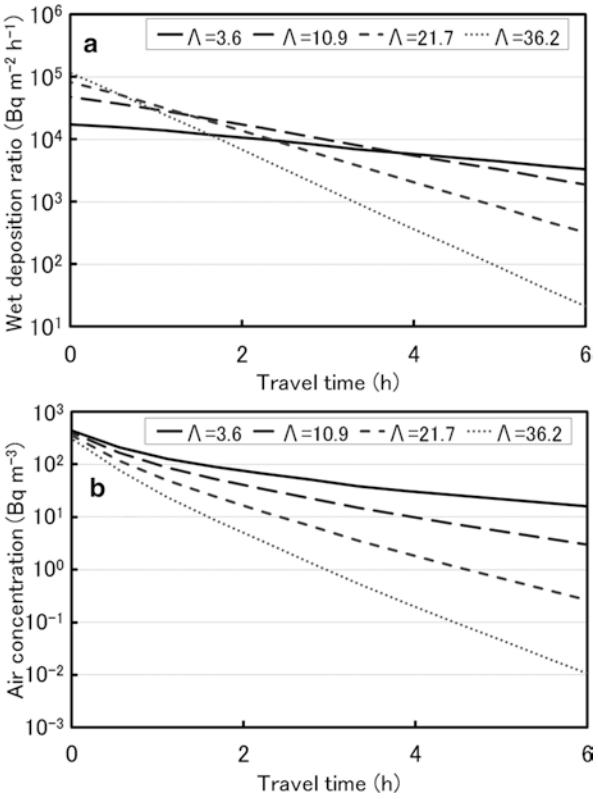

Fig. 14.3 Variation of the calculated values of (**a**) wet deposition rate and (**b**) air concentration with travel time. Scavenging coefficients, Λ, are 3.6, 10.9, 21.7, and 36.2×10^{-5} s^{-1}, which correspond to rain intensities of 0.37, 1.47, 3.48, and 6.60 mm h^{-1}, when the scavenging coefficient is expressed as $\Lambda = 8.0 \times 10^{-5} (I/I_0)^{0.8}$ in s^{-1}

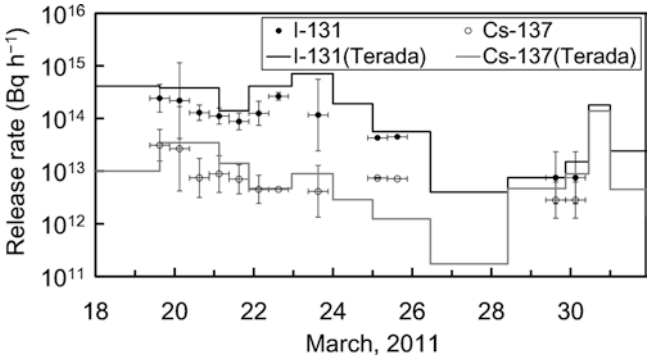

Fig. 14.4 Temporal change in the estimated release rate of ^{131}I and ^{137}Cs. *Lateral bars* on plots show the release duration 6 s of 12 h. *Vertical bars* show geometric standard deviations of the estimated release rate. *Closed* and *open circles* indicate the estimated values. *Black* and *gray lines* indicate the values estimated by Terada et al. [4]

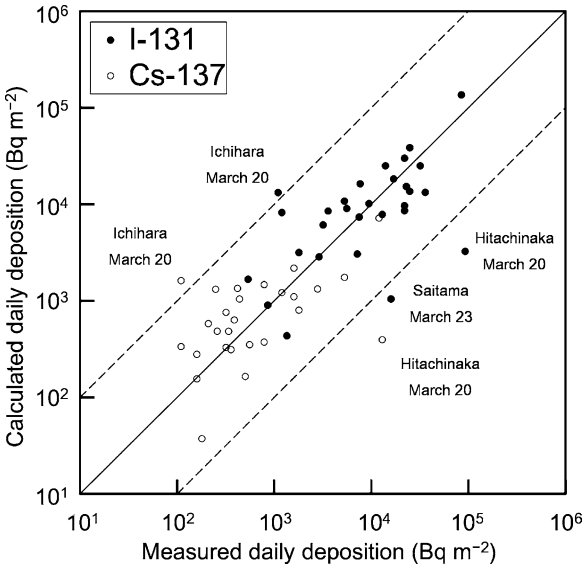

Fig. 14.5 Comparison of measured and calculated daily deposition using estimated release rate of ^{131}I and ^{137}Cs

available. The estimated release rate of ^{131}I was 2.4×10^{14} Bq h^{-1} in the afternoon of 19 March and showed a slight decrease during the following few days. A large increase in release rate was evaluated to have occurred from the afternoon of 22 March to the morning of 23 March to reach the release rate of 2.6×10^{14} Bq h^{-1}. After this day, the release of ^{131}I gradually decreased to the release rate of 7.5×10^{13} Bq h^{-1} on 30 March. For ^{137}Cs, the release rate was 2.7–3.1 $\times 10^{13}$ Bq h^{-1} from the afternoon of 19 March to the morning of 20 March and maintained in the range of 2.8–8.8 $\times 10^{12}$ Bq h^{-1} during the following days. The estimated release rates imply that the release of ^{131}I decreased during March whereas that of ^{137}Cs remained unchanged significantly.

Figure 14.4 also represents the temporal change of the release rate of ^{131}I and ^{137}Cs estimated by Terada et al. [4]. Although the release rates estimated in this study are smaller than the Terada values, the temporal changes are consistent with those of Terada. The difference of our estimated values compared to Terada's values are that the decrease of the release rate in the period from 20 to 21 March starts earlier and the temporal change in the release rate of ^{137}Cs is more moderate.

Scatter diagrams of the daily deposition of ^{131}I and ^{137}Cs of the calculated values using the estimated release rate and of measured values from 18 March to 31 March are shown in Fig. 14.5. For both nuclides, a good correlation was found between the calculated and measured values. The percentages of the calculated values that are within factors of 2 and 5 of the measured values are 55 % and 86 % for ^{131}I and 48 % and 89 % for ^{137}Cs, respectively. The calculated daily depositions showed large underestimation at Hitachinaka on 20 March and Saitama on 23 March and large overestimation at Ichihara on 20 March by a factor of more than 10 of the measured values. The pairs of measured and calculated daily deposition values at these sites resulted in large geometric standard deviations of the release rate in the morning of

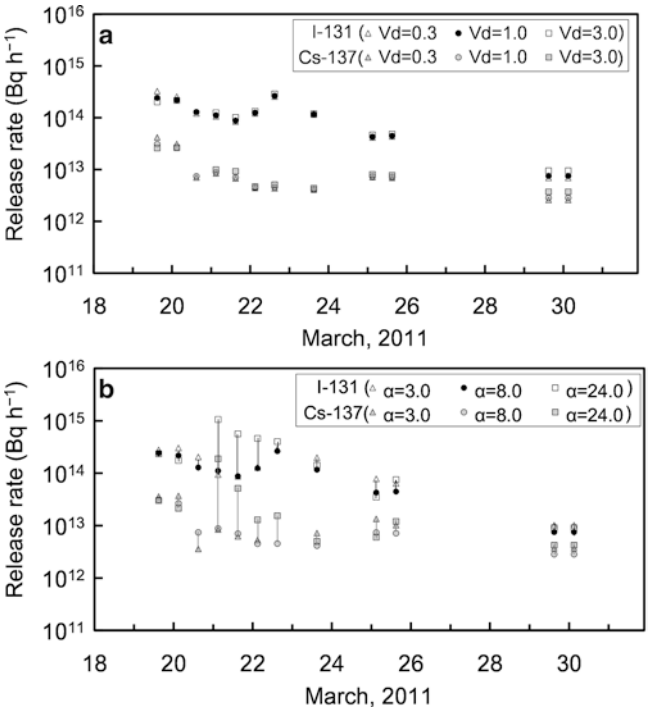

Fig. 14.6 Sensitivity of estimated release rate of ^{131}I and ^{137}Cs to (**a**) dry deposition velocity and (**b**) scavenging coefficient. Dry deposition velocities, V_d, are 0.3, 1.0, and 3.0 mm s^{-1} and α are 3.0, 8.0, and 24.0 × 10^{-5} s^{-1}, when the scavenging coefficient is expressed as $\Lambda = \alpha(I/I_0)^{0.8}$

20 March and 23 March (Fig. 14.4). Errors of the atmospheric dispersion model could lead to a large uncertainty in the calculated deposition. However, the error does not propagate seriously to the estimated release rate because the release rate in one time segment was estimated by several pairs of measured and calculated daily deposition values.

14.3.2.2 Sensitivity Analysis on Deposition Parameters

The changes in the estimated release rates with the change in V_d from the standard value 1.0–0.3 and to 3.0 mm s^{-1}, and in α from the standard value 8.0 × 10^{-5} to 3.0 × 10^{-5} and to 2.4 × 10^{-4} s^{-1}, are shown in Fig. 14.6a and 14.6b, respectively. To investigate the sensitivity to the deposition parameters, we calculated the ratio of the estimated release rate to that for the standard deposition parameters.

The ratios for $V_d = 0.3$ mm s^{-1} varied in the range of 0.9–1.3 for both nuclides. For $V_d = 3.0$ mm s^{-1}, the ratios varied in the range of 0.8–1.3 for both nuclides, indicating that the estimated release rates for ^{131}I and ^{137}Cs vary only by a factor of 1.1–1.3 when the dry deposition velocity is changed by a factor of approximately 3.

Slightly larger release rates were estimated for 19 March and 29 March by using the smaller dry deposition velocity because the plume on this day did not experience rain and dry deposition was dominant. This result is consistent with the discussion for the dry deposition in the previous section. The estimated release rates in the period from 20 to 25 March were not substantially affected by dry deposition velocity because wet deposition processes dominated the deposition.

During the period from 19 to 20 March, the plume was transported to Kanto region after being once transported over the Pacific Ocean. In general, dry deposition velocity over the sea surface is considered to be smaller than that on land surfaces. If horizontally homogeneous dry deposition is assumed in the estimation of release rate, which might be an overestimation of dry deposition over sea surface, and hence underestimation of air concentration and dry deposition at the place of observation, the release rates could be overestimated. However, this effect is not serious, as shown in Fig. 14.2.

In the case of the scavenging coefficient, the ratios for $\alpha = 3.0 \times 10^{-5}$ s^{-1} varied in the range of 0.9–1.8 for ^{131}I and 0.5–1.8 for ^{137}Cs. For $\alpha = 2.4 \times 10^{-4}$ s^{-1}, the ratios varied in the range of 0.8–9.6 for ^{131}I and 0.8–21.3 for ^{137}Cs. A scavenging coefficient three times larger resulted in increases of 10 and 21 fold in the estimated release rate for ^{131}I and ^{137}Cs, respectively, at maximum. Sensitivity to the scavenging coefficient was considerably larger than that to the dry deposition velocity.

For the period from 19 March to the morning of 20 March and the morning of 25 March, the release rates estimated with the small scavenging coefficient were larger than the release rate with the large scavenging coefficient, which is likely because the calculated deposition was small as a result of the weak rainfall intensity and the short travel time in the rainfall area. This tendency is also consistent with the results of the wet deposition for a short travel time discussed in the previous section. In contrast, the release rates in the period from 21 to 22 March showed a high sensitivity to scavenging coefficient because there was heavy rainfall in the Kanto Region where the plume was transported. The large estimated release rate can be attributed to the depletion of plume when it traveled over Kanto Region, causing lessr deposition on the area where the plume passed later. This tendency is depicted in Fig. 14.2, in which the wet deposition rates decrease more rapidly with time if the scavenging coefficient is larger. Except for these periods, the sensitivity analysis showed no systematic tendency.

14.4 Conclusion

The present study estimated the release rate of ^{131}I and ^{137}Cs during March 2011 and showed the results of the sensitivity analysis of the release rate to the deposition parameters. To estimate the release rate, the method applied is based on a simple inverse method by combining the measured daily deposition of ^{131}I and ^{137}Cs with regional range atmospheric dispersion calculations.

The present study showed that the release rates were reasonably estimated by using the measured daily depositions. It was found that the dry deposition velocity

had only minor significance on the estimated release rate, predominately as a result of the inherently small contribution to total deposition when wet deposition occurred. The scavenging coefficient, on the other hand, showed a substantial effect on the release rate estimation. A three times larger scavenging coefficient resulted in increases of 10 and 21 fold in the estimated release rate for ^{131}I and ^{137}Cs, respectively, at maximum. The uncertainties of the release rate estimated by the inverse method might be large when the plume is transported through the rainfall area. It can be suggested that a more realistic parameterization of wet deposition processes is needed to refine the release rate estimation from environmental data.

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