Estimation of uncertainty in the release rate of I-131 and Cs-137 from FDNPS estimated from environmental data

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Sensitivity of the release rate estimated from environmental data to the deposition parameters was evaluated by a method commonly used in previous studies on source term estimation. It was found that the dry deposition velocity had only a minor effect on the estimated release rate, predominately due to the inherently small contribution to total deposition when wet deposition occurred. The scavenging coefficient, however, showed a substantial effect on the release rate estimation. A scavenging coefficient three times larger resulted in increases in the estimated release rate by a factor of six.

Key Words: *Release rate estimation, deposition rate, atmospheric dispersion model*

1. Introduction

To access radiological doses to the public due to the serious accident at the Fukushima Daiichi Nuclear Power Station (FDNPS) in March 2011, it is necessary to evaluate the dispersion of the radioactive plume and deposition of contained radionuclides on the land surfaces. The most reliable information in this regard is the environmental monitoring data. During the early phase of the accident, atmospheric concentrations and deposition rates were measured. However, the spatial coverage of these measurements was too limited to understand the evolution of the plume and its radioactive composition. Airborne measurements were also made by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) in collaboration with the U.S. Department of Energy (DOE). This work provided data on the distribution of land surface contamination of medium- and long-lived radionuclides Cs-134 and Cs-137, respectively. However, uncertainties still remain regarding the atmospheric dispersion and deposition of short-lived radionuclides such as I-131, which are important to evaluate the radiological doses to the public. In such situations, atmospheric dispersion simulations can effectively evaluate transport of the radioactive plumes when the source term information is available.

To estimate the source terms, environmental monitoring data has been used with atmospheric dispersion models in several earlier works[1–4]. All studies with inverse methods indicate similar temporal variations in the atmospheric release rate of I-131 and Cs-137, despite the use of different sets of monitoring data and atmospheric dispersion models. According to these studies, the release began on the morning of 12 March 2011, and the largest release occurred on 15 March. 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 release rate estimated by the inverse methods, uncertainties in the estimated release rate have not been discussed sufficiently. In our previous study, the uncertainty was estimated to be approximately a factor of three due to errors in modeled deposition processes[4].

The purpose of this study is to estimate uncertainties in the estimated release rates of I-131 and Cs-137. A sensitivity analysis of estimated release rates on the deposition parameters, i.e., the dry deposition velocity and the washout coefficient, was carried out in this study. Data on daily deposition of I-131 and Cs-137 were used. The analysis was conducted first under simplified meteorological conditions and second under actual meteorological conditions during the period of March 2011 using a meteorological model.

2. Methods

(1) Release rate estimation

The estimation of the release rate is based on the principle that the atmospheric dispersion model can calculate a spatial distribution of relative values of the deposition rate on the ground while 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:

$$\left(\frac{Q_r}{S_r}\right)_{t,i} = \left(\frac{Q_m}{S_m}\right)_{t,i}, \qquad (1)$$

where S_m is the release rate used for model calculations, Q_r is the measured deposition rate, and Q_m is the calculated deposition rate. The subscripts t and i denote the sampling time and sampling point, respectively. The release rate was estimated using Eq. 1 by solving for S_r.

Equation 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 rates estimated from independent monitoring data. In this case, a geometrical mean was applied to estimate a single value for the time period.

(2) Atmospheric dispersion model

A Lagrangian particle random-walk model (LPRM)[5] coupled with a nonhydrostatic atmospheric dynamic model MM5[6] 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. I-131 and Cs-137 were modeled as passive tracers with 8.04 days and half lives of 30 years, respectively. Dry and wet depositions were parameterized in terms of a dry deposition velocity and a washout (or scavenging) coefficient, respectively. As a standard pair of these removal parameters, the dry deposition velocity was set as 1.0 mm s^{-1}, and the scavenging coefficient was expressed as 8.0×10^{-5} $(I/I_0)^{0.8}$ s^{-1}, where I is the precipitation intensity and I_0 = 1.0 mm h^{-1}. The dry deposition velocity and scavenging coefficient vary within the range 0.1–10 mm s^{-1} and (10^{-6}–10^{-3}) $(I/I_0)^{0.8}$ s^{-1}, respectively, depending on the physicochemical characteristics of the nuclides, gas-particle partitioning, and particle size distribution of aerosols[7, 8]. In this study, the removal parameters, which changed by a factor of 3, were applied in the sensitivity analysis.

(3) Environmental monitoring data

The data used in the release rate estimation were the daily deposition rates measured at Chigasaki, Hitachinaka, Ichihara, Maebashi, Saitama, Sinzyuku, and Utsunomiya with a 24 h sampling time from 18 March by MEXT[9]. To eliminate the influence of resuspended radionuclides, the following criteria were set for the data selection: the deposition rates of I-131 and Cs-137 were greater than 5.0×10^2 Bq m^{-2} day^{-1} and 5.0×10^1 Bq m^{-2} day^{-1} and, respectively. In all, 58 deposition rates of I-131 and Cs-137 were used in this study.

(4) Calculation conditions

For calculation under actual meteorological conditions, the domain of the atmospheric dispersion was set as 600 km^2 with a 6 km height above the ground to cover most of the Tohoku and Kanto regions. The same physical processes of MM5 as the previous study[4] were used in this work. 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 09:00 JST, 8 March to

00:00 JST, 1 April. The dispersion of I-131 and Cs-137 from FDNPP started at 15:00 JST, 17 March and ended at 00:00 JST, 1 April. The release height was set as 15 m above the ground. A constant release rate of 1 TBq h^{-1} was assumed.

3. Results and discussions

(1) Simplified meteorological conditions

Before carrying out simulations with the meteorological data from MM5, a series of preliminary simulations were conducted under a set of simplified meteorological conditions, i.e., under a uniform and constant wind field of 1 m s^{-1}, horizontally uniform but vertically varying vertical eddy diffusivities with a typical value of 16 m^2 s^{-1} at 100 m a.g.l., and prescribed intensities of uniform precipitation. The LPRM used was the same as in the main simulations. The purpose of these simulations was to evaluate the dependency of deposition and air concentration on deposition parameters.

The results are shown in Figs. 1–2. The dependency of dry deposition on the deposition velocity is quite simple. The dry deposition rate from the plume monotonously decreases with travel time, i.e., the elapsed time from release, mainly due to horizontal and vertical diffusion (Fig. 1a). The ratio of dry deposition rate in the case of 0.3 mm s^{-1} velocity to that in the case of 3.0 mm s^{-1} at a travel time of 6 h is about 11.4%. This ratio is almost the same as the ratio of deposition velocity, but slightly modified by the change in the air concentration. The concentration in the 3.0 mm s^{-1} case is about 88% compared to that in the case of 0.3 mm s^{-1} (Fig. 1b). 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 washout coefficient is somewhat complicated (Fig. 2a). A larger scavenging coefficient does not always cause larger deposition. Among the four cases, the largest scavenging coefficient, which corresponds to a rain intensity of 6.6 mm h^{-1}, resulted in the largest wet deposition only in the first 0.6 h from the release. The wet deposition in this

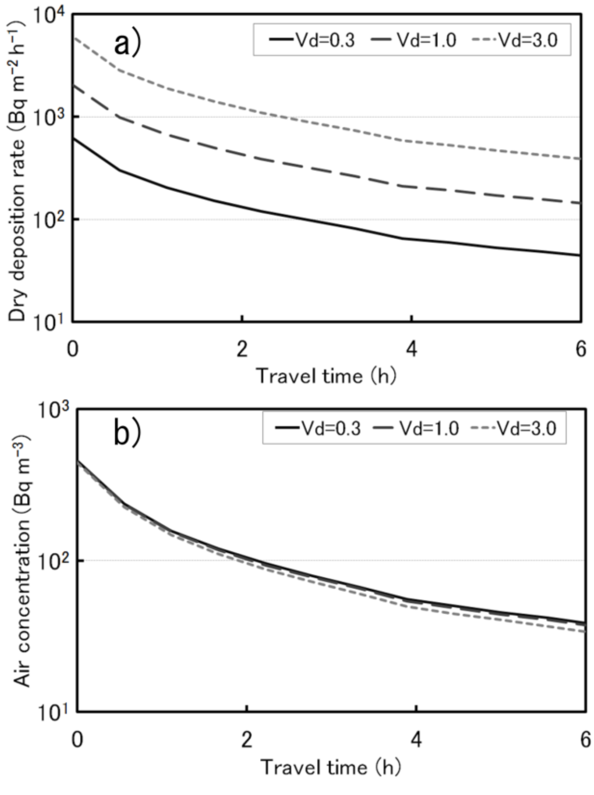

Fig. 1 Variation of calculated values of a) dry deposition rate and b) air concentration with travel time. Dry deposition velocities (mm s^{-1}) are 0.3, 1.0 and 3.0.

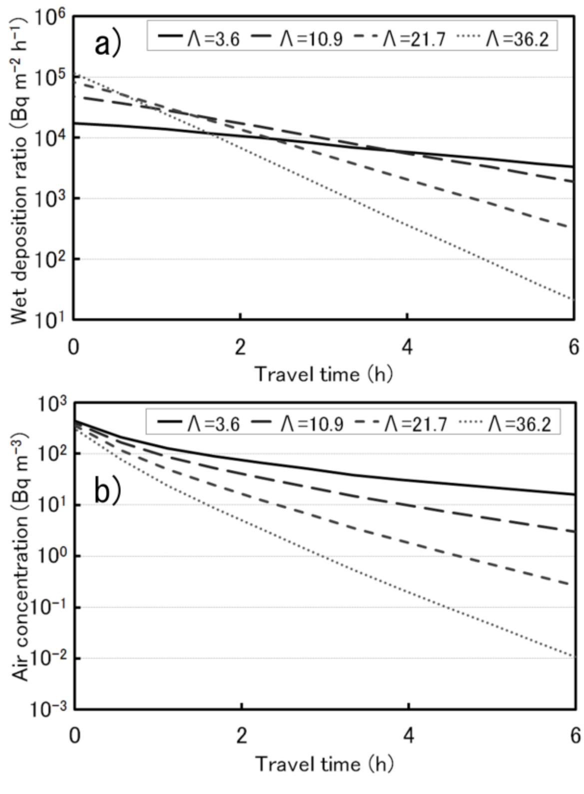

Fig. 2 Variation of the calculated values of a) wet deposition rate and b) air conentration with travel time,. Scavenging coefficients are 3.6, 10.9, 21.7 and 36.2 s^{-1}, which correspond to rain intensities of 0.37, 1.47, 3.48, and 6.60 mm h^{-1}, respectively, when the scavenging coefficient is expressed as $8.0 \times 10^{-5} \, (I/I_0)^{0.8}$ s^{-1}.

case becomes the smallest after the travel time of 1.7 h. However, 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 a travel time of 3.9 h. This complicated result is caused by the substantial depletion of the plume by scavenging as shown in Fig. 2b. In the cases with large scavenging coefficients, the air concentrations rapidly decrease with travel time.

(2) Actual meteorological conditions

The estimated release rates for I-131 and Cs-137 varied only by a factor of approximately 1.3 when the dry deposition velocity was changed by a factor of 3 (Fig. 3). Using the smaller dry deposition velocity, a slightly larger release rate was estimated for 20 March because the plume did not encounter rain and dry deposition processes were dominant on that day. This result is consistent with the discussion on the dry deposition in the previous section. The estimated release rates after 20 March were not substantially affected by dry deposition velocity, because the wet deposition processes dominated during this period.

During the period from 19 to 20 March, the plume was transported to the Kanto region after being transported over the Pacific Ocean. In general, the dry deposition velocity over the sea surface is considered to be smaller than that on land surface. If horizontally homogeneous dry deposition is assumed in the estimation of the release rate, which might be an overestimation of dry deposition over the sea surface and hence an 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. 1.

From the sensitivity analysis of the scavenging coefficient, the release rates of I-131 and Cs-137 varied by a factor of six at the maximum (Fig. 4). The sensitivity to the scavenging coefficient was considerably larger than that to the dry deposition velocity. The release rates in the period from 21 to 22 March showed a high sensitivity to the scavenging coefficient because there was strong rainfall in the Kanto region where the plume was transported. This large estimated release rate can be attributed to the depletion of the plume when it traveled over the Kanto region causing smaller depositions on area passed later by the plume. This tendency is depicted in

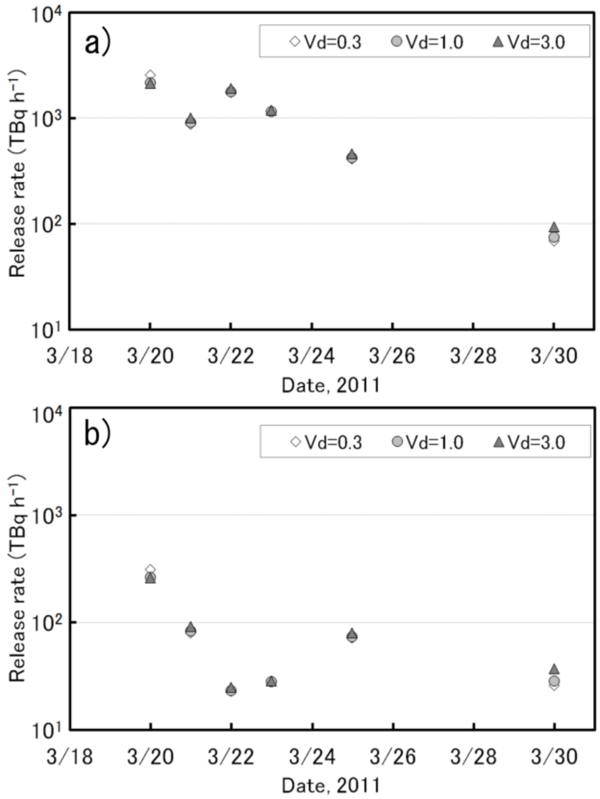

Fig. 3 Sensitivity of estimated release rate to dry deposition velosity, a) I-131 and b) Cs-137. Dry deposition velocities (mm s^{-1}) are 0.3, 1.0 and 3.0.

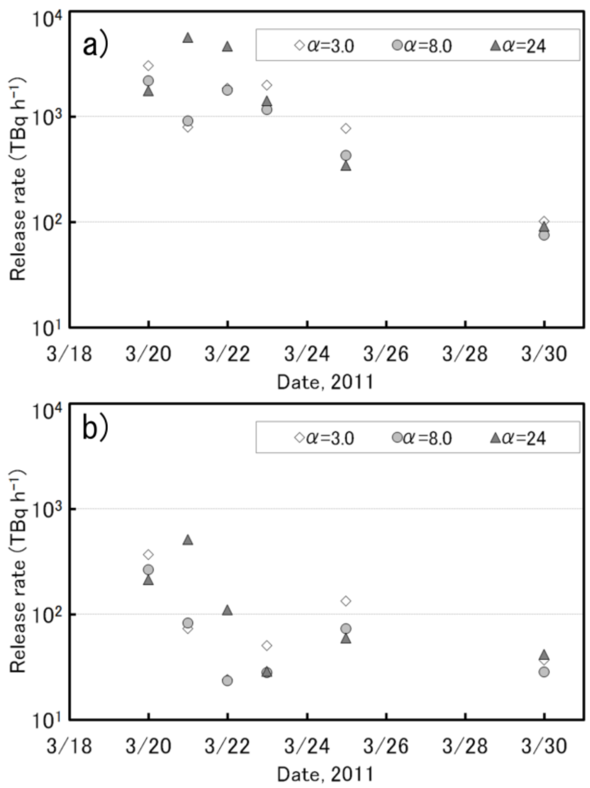

Fig. 4 Sensitivity of estimated release rate to scavenging coefficient, a) I-131 and b) Cs-137. Scavenging coefficients (s^{-1}) are calculated by $\alpha(I/I_0)^{0.8}$, where α (10^{-5} s^{-1}) are 3.0, 8.0, and 24.0.

Fig. 2, in which the wet deposition rate decreases more rapidly with time in the case of a larger scavenging coefficient. Except for this period, the release rates estimated using a small scavenging coefficient were larger than the release rates estimated using a large scavenging coefficient. This might have occurred because the calculated deposition was small owing to the weak rainfall intensity and the short travel time in the rainfall area where the plume was near measurement points. This is also consistent with the results for wet deposition for a short travel time discussed in the previous section.

4. Conclusions

In this study, the sensitivity analysis of the release rate estimated from environmental monitoring data on the deposition parameters was carried out. The applied release rate estimation is based on a simple inverse method by combining the environmental monitoring data and regional range atmospheric dispersion calculations. It was found that the dry deposition velocity had only a minor effect on the estimated release rate, predominately due to the inherently small contribution to total deposition when wet deposition occurred. The scavenging coefficient, however, showed a substantial effect on the release rate estimation. A coefficient three times larger resulted in increases in the estimated release rate by a factor of six. The uncertainties in the release rate estimated by the inverse method might be enhanced when the plume is transported through a rainfall area. It can be concluded that a more realistic parameterization of the wet deposition processes would be needed to refine the release rate estimated from environmental data.

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References

1) M. Chino, H. Nakayama, H. Nagai, H. Terada, G. Katata and H. Yamazawa, “Preliminary Estimation of Release Amounts of ^{131}I and ^{137}Cs Accidentally Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere,” *J. Nucl. Sci. Technol.*, **48**, pp. 1129–1134, 2011.
2) A. Stohl, P. Seibert, G. Wotawa, D. Arnold, J. F. Burkhart, S. Eckhardt, C. Tapia, A. Vargas and T. J. Yasunari, “Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition,” *Atmos. Chem. Phys.*, **12**, pp. 2313–2343, 2012.
3) H. Terada, G. Katata, M. Chino and H. Nagai, “Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power Plant accident. Part II: verification of the source term and analysis of regional-scale atmospheric dispersion,” *J. Environ. Radioact.*, **112**, pp. 141–154, 2012.
4) S. Hirao, H. Yamazawa and T. Nagae, “Estimation of release rate of iodine-131 and cesium-137 from the Fukushima Daiichi nuclear power plant,” (submitted for publication)
5) S. Hirao and H. Yamazawa, “Release Rate Estimation of Radioactive Noble Gases in the Criticality Accident at Tokai-Mura from Off-Site Monitoring Data,” *J. Nucl. Sci. Technol.*, **47**, pp. 20–30, 2010.
6) G. A. Grell, J. Dudhida, D. R. Stauffer, “*A description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5)*,” NCAR Tech. Note NCAR/TN-389+STR, National Center for Atmospheric Research (NCAR), **117**, 1994.
7) A. Baklanov, J.H. Sørensen, “Parameterisation of radionuclide deposition in atmospheric long-range transport modelling,” *Phys. Chem. Earth (B)*, **26**, pp. 787–799, 2001.
8) B. Sportisse, “A review of parameterizations for modelling drydeposition and scavenging of radionuclides,” *Atmos. Environ.*, **41**, pp. 2683–2698, 2007.
9) MEXT, http://radioactivity.mext.go.jp/en/contents/4000/3156/24/115_hp_0912_18.pdf, 2012 (accessed on 15.10.12).