Atmospheric Modeling of Radioactive Materials from the Fukushima Daiichi Nuclear Power Plant Accident

Yu MORINO [1*] and Toshimasa OHARA [2]

[1] *Center for Regional Environmental Research, National Institute for Environmental Studies*
16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan
[2] *Fukushima Branch, National Institute for Environmental Studies*
10-2 Fukasaku, Miharu, Fukushima 963-7700, Japan
**email: morino.yu@nies.go.jp*

Abstract

Atmospheric simulation models have played key roles in elucidating the atmospheric behaviors and deposition patterns of radioactive materials emitted from the Fukushima Daiichi Nuclear Power Plant after the nuclear accident in March 2011. In this review article, we summarize atmospheric modeling studies conducted after that accident. In particular, atmospheric models were used for source term estimates and analysis of atmospheric behaviors of radionuclides. In addition, atmospheric models provided results utilized in estimating health effects, forecasting, assessing long-term mitigation, and other tasks. Performance of atmospheric models for those purposes is evaluated in detail. Many problems remain, however, and future studies will be required for gaining a better understanding of radionuclides' environmental impact.

Key words: atmospheric simulation, Fukushima Daiichi Nuclear Power Plant, radiocesium, radioiodine, source term, SPEEDI

1. Introduction

After the accident at the Fukushima Daiichi Nuclear Power Plant in March 2011 (FDNPP; the Fukushima accident), many studies using atmospheric simulation models were conducted, helping to grasp the atmospheric behaviors, source terms and deposition patterns of radioactive materials released by the accident. In this review article, we summarize atmospheric modeling studies conducted after the Fukushima accident. First, source term estimates using atmospheric simulation models are introduced (Section 2). Then, research on atmospheric model evaluation is described (Section 3). We also introduce modelling studies on analysis of radionuclides' atmospheric behaviors (Section 4) and application of atmospheric simulation models (Section 5) Finally, the future direction of modelling studies is discussed (Section 6).

2. Source Term Estimates

For atmospheric simulation of radionuclides, source terms are one of the most critical input parameters. Source terms can be estimated using bottom-up methods by simulating the accident sequence using severe nuclear accident simulation codes like MELCOR (Methods for Estimation of Leakages and Consequences of Releases, Gauntt *et al*., 2000), or by reverse or inverse modeling using atmospheric transport models and environmental monitoring data (Stohl *et al*., 2012). No bottom-up estimates from a stack monitor or reactor analysis were available after the Fukushima accident, so reverse or inverse analysis has mostly been applied to date.

The first source term estimate of the Fukushima accident was released by researchers at the Japan Atomic Energy Agency (JAEA) (Chino *et al*., 2011). Researchers at JAEA had gained experience in estimating source terms from the Chernobyl accident (Chino *et al*., 1986) and the JCO accident (Hirao & Yamazawa, 2010) using their simulation models: System for Prediction of Environmental Emergency Dose Information (SPEEDI) and the worldwide version of SPEEDI (W-SPEEDI). At first, they made a preliminary estimate of the source terms of radioiodine and radiocesium using a reverse estimation method involving comparison of a regional model with monitoring data for air dose rates and air concentrations in Japan during March 12 – April 6 (Chino *et al*., 2011). This preliminary estimate was updated in subsequent papers by JAEA researchers. Katata *et al*. (2012a) and Katata *et al*. (2012b) revised their estimates of release rates during March 12– 14 and March 15, respectively, using the same model. Terada *et al*. (2012) summarized those source terms for March 12 – April 30 and evaluated the model results using daily and monthly surface deposition data over eastern Japan. Kobayashi *et al*. (2013) updated the source terms by combining atmospheric and oceanic dispersion simulations with observed Cs-134 in seawater collected from the Pacific Ocean. Katata *et al.* (2015) updated the source terms during March 12 – April 30 using atmospheric and oceanic transport, dispersion and deposition models. They updated schemes of dry and fog-water depositions, cloud condensation nuclei activation, and subsequent wet scavenging through mixed-phase cloud microphysics.

Stohl *et al*. (2012) and Winiarek *et al*. (2012) estimated source terms using inverse models at a global scale and atmospheric activity concentrations measured by the global International Monitoring System operated by the Comprehensive Nuclear Test Ban Treaty Organization. The first guesses of Stohl *et al*. (2012) were based on fuel inventories and documented accident events at the site, and these first guesses were successfully improved through inverse modeling by combining it with measurement data.

Deposition data were also used for estimating Cs-137 source terms. Winiarek *et al*. (2014) developed an inversion method using total cumulative deposition and air activity concentration simultaneously. Yumimoto *et al*. (2016) also developed an inversion method to estimate Cs-137 emissions using cumulative deposition data from airborne monitoring, and improved their inventory during the period of inland flow. Using these emissions data, they improved the deposition simulation results for the central region of Fukushima Prefecture and of atmospheric concentrations on March 15. Both these studies, however, found improvement to be limited because of inaccuracy in meteorological model simulations of wind fields and precipitation.

The latest source term results are summarized in Fig. 1 (Katata *et al*., 2015). Most of the estimates have agreed within an order during periods when the plumes flowed over land during March 14 – 15 and March 20 – 21. On March 15, however, the highest Cs-137 release rates were found in the evening and nighttime by Katata *et al*. (2015), though this behavior did not agree with other estimates. We should note that the results of Katata *et al*. (2015) agree with those of Stohl *et al*. (2012) even during periods when the plumes flowed toward the Pacific Ocean. Katata *et al*. (2015) used Cs-134 sea surface concentration data, while Stohl *et al*. (2012) used mainly air concentrations of Cs-137 worldwide. This comparison should indicate that these estimates are somewhat reasonable.

Recently, Chino *et al*. (2016) identified FDNPP reactor units which generated large amounts of airborne matter during March 12–21, 2011 from a combination of measured Cs-134/Cs-137 depositions on ground surfaces and atmospheric simulations. The major source reactor units were Unit 1 in the afternoon of March 12, 2011, Unit 2 during the period from the late night of March 14

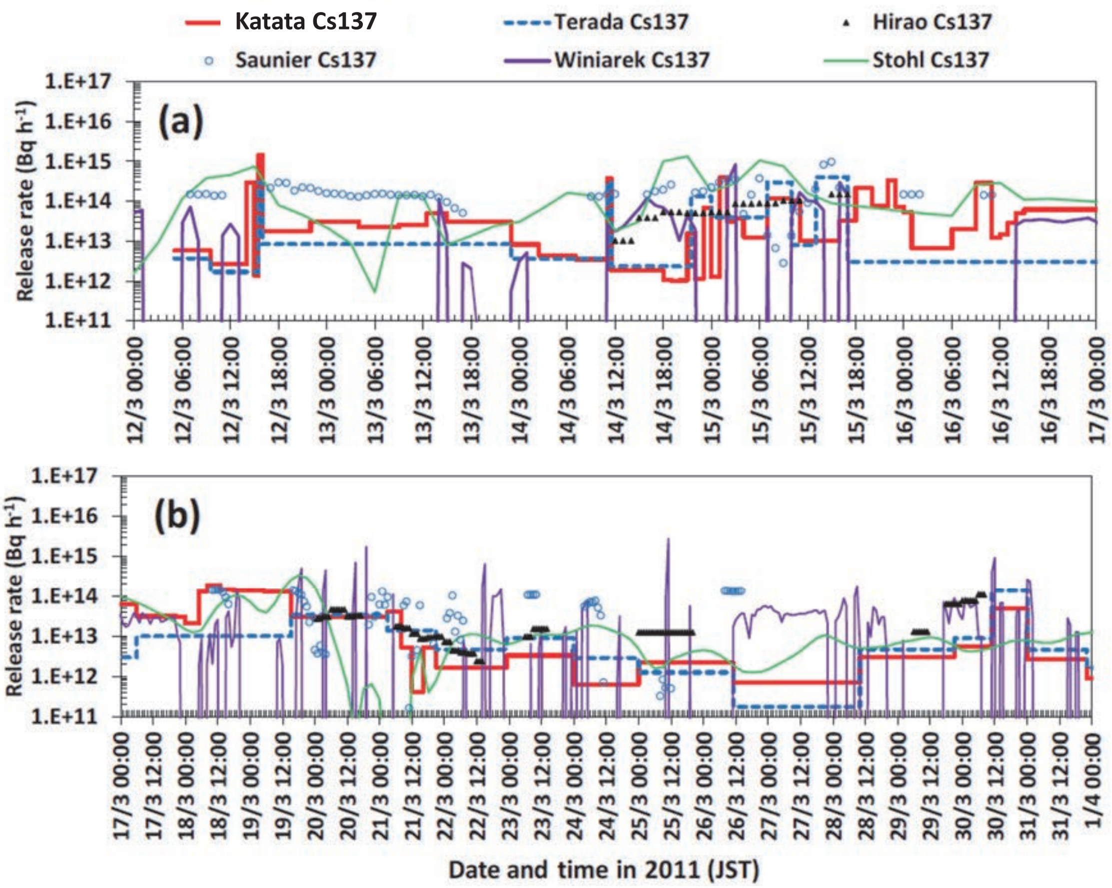

Fig. 1. Comparison of Cs-137 release rates from March 12 to March 17, 2011 (a) and March 17 to April 1, 2011 (b) among six studies (Katata *et al*., 2015).

to the morning of March 15, Units 2 and 3 from the evening of March 15, and Unit 2 on March 20.

3. Model Evaluation

3.1 Inter-comparison of Models

Model results usually include uncertainties, because of problems with their input data (e.g., meteorological and emissions data), the parameterization of each process, and missing science elements. To evaluate the validity and variability of model results, inter-comparisons of models have provided valuable information.

The Science Council of Japan (Science Council of Japan, 2014) compared the simulation results of nine regional atmospheric models and six global atmospheric models. Meteorological fields obviously play an important role in simulating radionuclide deposition. All the models showed that the wet deposition process had a strong impact on accumulated deposition over eastern Japan, although there were large variabilities in the horizontal distribution of accumulated deposition (Fig.2). These variabilities may have been caused by differences in the models' treatment of deposition and in the configuration of meteorological models (e.g., their microphysics and convection parameters). They noted that ensemble means were useful for estimating accumulated deposition.

The World Meteorological Organization (WMO) also performed inter-comparisons among five different atmospheric transport and dispersion models (Draxler *et al.*, 2015). They evaluated the model results using observed deposition and air concentrations, and found that the best model for simulating deposition was not always the best model for simulating air concentrations. In addition, they also reported that the ensemble mean among better performing models provided more consistent results for both types of calculations, and this result is consistent with that of the Science Council of Japan (2014).

Nakajima *et al.* (2017) compared the results of two simulation models, and applied a two-model ensemble method using weights determined by deviations of the respective model results from observed air concentrations (Tsuruta *et al.*, 2014). The ensemble model reproduced observed plumes better, though in some cases, the models failed to simulate the exact location and time of arrival of the plumes. These differences suggest a need for improvements to models regarding emissions scenarios, plume heights, wet deposition processes, and plume propagation in the Abukuma Mountain region.

3.2 Uncertainty analyses

In addition to inter-comparison studies of multiple models, uncertainty analyses from sensitivity simulations of a single model also provide useful information about

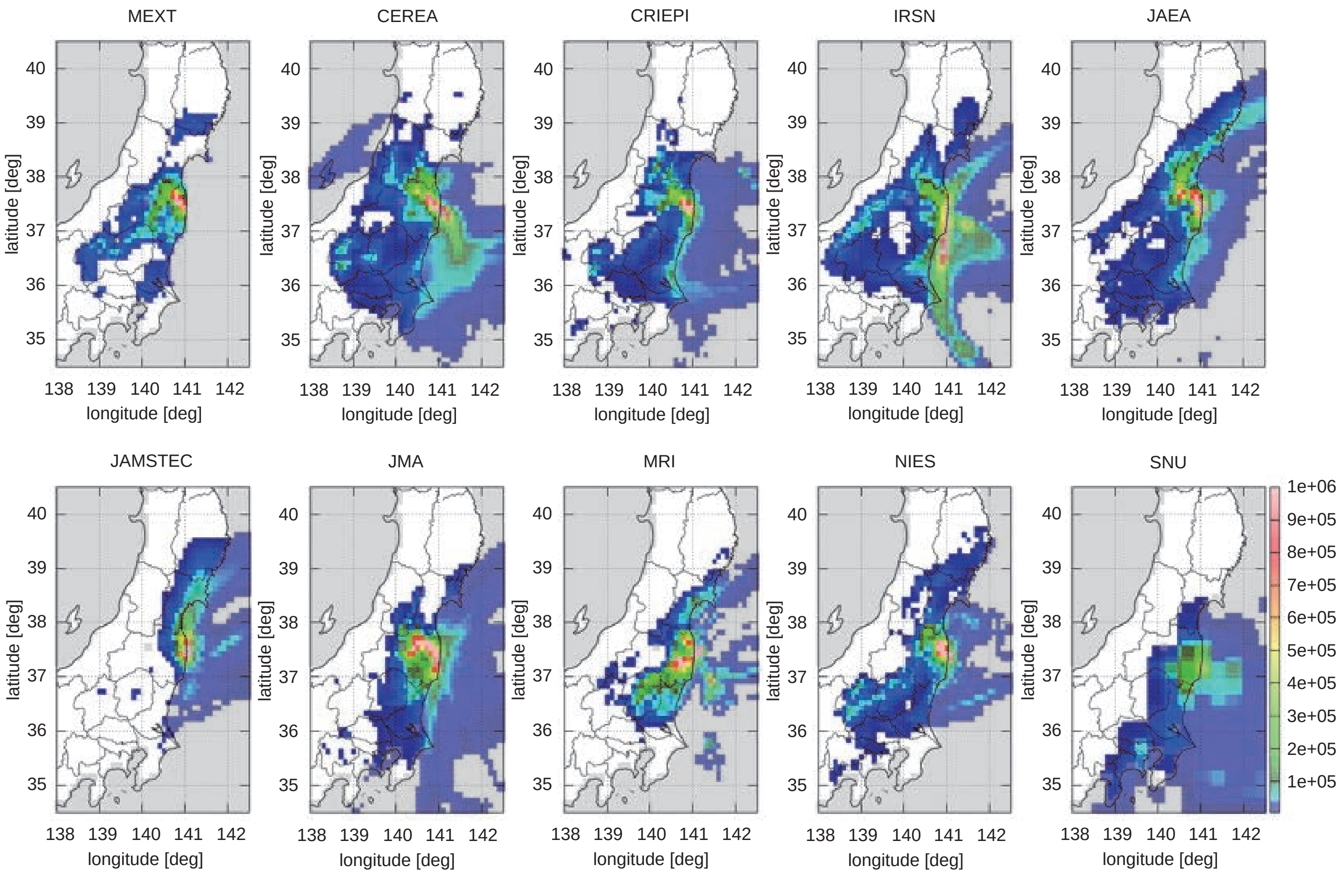

Fig. 2. Observed and simulated spatial distributions of Cs-137 accumulated deposition (Bq m-2) through April 1, 2011 (Science Council of Japan, 2014).

model reproducibility.

Morino *et al*. (2013) conducted ten sensitivity simulations to evaluate the atmospheric model uncertainties associated with key model settings including emissions data and wet deposition modules. They found that simulations using emissions estimated at a regional scales (~ 500 km) model better reproduced the observed Cs-137 deposition pattern in eastern Japan than simulation using emissions estimated at local scales (~ 50 km) or global-scale models. In addition, simulation using a process-based wet deposition module reproduced the observations well, whereas simulation using scavenging coefficients showed large uncertainties associated with empirical parameters.

Girard *et al*. (2014) evaluated uncertainties associated with 19 uncertain input parameters, including meteorological fields and source terms, by conducting simulations with simple perturbations whose magnitudes were estimated from a thorough literature review. They found that wind perturbations and emissions factors for iodine and cesium were the predominantly uncertain parameters, while some inputs, such as the cloud layer thickness, were found to have little influence on most of the outputs considered.

Sekiyama *et al*. (2015) compared the simulation results with low (15-km), medium (3-km), and high (500-m) resolutions. They used the same meteorological analysis from their data assimilation system for consistent comparisons. They concluded that for analyses of the Fukushima accident at a regional scale, low-resolution (15-km grid or greater) atmospheric models should be avoided while models with a grid size of 3 km showed similar results with those having 500-m resolution.

From these analyses, emissions and meteorological fields, including wind and precipitation fields, seem to be the most critical sources of uncertainty. In addition, model results are sensitive to wet deposition modules, which accordingly suggests that the treatment of physical and chemical properties of radionuclides needs to be revealed. In particular, the diameters and hygroscopicity of particles are sensitive parameters in deposition simulations (Adachi *et al*., 2013), and their treatment should be a subject of future studies.

4. Analyses of Atmospheric Behaviors

To understand the atmospheric behaviors of radionuclides, analyses using observational data have been insufficient because of lack of spatial and temporal coverage, and thus atmospheric simulations have been conducted over local, regional and global scales.

At a local scale, Katata *et al*. (2012b) analyzed formation processes of high dose rates using limited environmental data and the nuclear emergency response system, WSPEEDI. They showed that a significant amount of surface deposition was produced in the region northwest of the FDNPP in the evening, when the high-concentration plume discharged in the afternoon was scavenged by rainfall. Wet deposition due to rainfall played an important role in the formation of wide and heterogeneous high dose rate zones. The plume flowed and widely dispersed along a valley leading leeward, expanding the area receiving a large amount of surface deposition.

At a regional scale, Morino *et al*. (2011) showed that during March 15–16 and 20–23, northeasterly, easterly or southeasterly winds associated with a transient cyclone transported radioactive materials from the FDNPP to inland areas. In addition, precipitation was observed during this period, thus radioactive materials were effectively deposited over the land by wet processes. In contrast, on March 17 – 20 when an anticyclone dominated over Fukushima, westerly or northwesterly winds prevailed, and radioactive materials were transported predominantly to the Pacific Ocean. Morino *et al*. (2013) indicated that in their standard simulation, most of the deposition over Japanese land occurred during March 15 – 16 (72.0% of the total deposition during the model period), March 20 – 24 (15.7%), and March 30 – 31 (11.4%), while deposition during other periods was very small (0.9%) (Fig. 3). Nakajima *et al*. (2017) analyzed the simulation results by comparing them with the hourly observed atmospheric Cs-137 surface level concentrations during March 14–23, 2011 at 90 sites of the suspended particulate matter monitoring network. They elucidated the transport routes of the atmospheric plumes (Fig. 4) and the distribution of the surface-level atmospheric Cs-137. Migratory pressure systems periodically brought radioactive materials to the Japanese land area, producing similar plume development patterns. For instance, a group of plumes, P2 and P3, followed by P4 during March 15– 16 and another group of plumes, P7 and P8, followed by P9 during March 20–21, produced two sets of peaks in the time series. The first peaks from P3 in the period of March 15– 16 were caused by a wind field change to northeasterly and later to southeasterly with the progress of a migratory low toward the Japanese islands. The second peaks (P4 and P9) were caused by a northerly wind, after the low-pressure system passed from the Japanese islands into the Pacific area.

At a global scale, Takemura *et al*. (2011) conducted numerical simulations of long-range transport with an aerosol transport model. A large-scale updraft produced by a low-pressure system lifted the particles from the surface layer to the level of the westerly jet stream during March 14 – 15, and the particles were carried across the Pacific within three to four days. Subsequently, those particles were transported across the Atlantic by a poleward-deflected jet stream, first toward Iceland and then southward to continental Europe. Stohl *et al*. (2012) also showed that radioactive clouds reached North America on March 15 and after that, Europe on March 22. They estimated that 18% of the total Cs-137 fallout until April 20 was deposited over land in Japan, while most of the rest fell over the North Pacific Ocean. Only 0.7 PBq, or 1.9% of the total fallout was deposited on land areas other than Japan. Evangeliou *et al*. (2013) also estimated that Cs-137 was deposited mostly in the Pacific and Atlantic Oceans and the Arctic (80%), whereas the rest, falling in the continental areas of North America and Eurasia, contributed slightly to natural background levels on the ground.

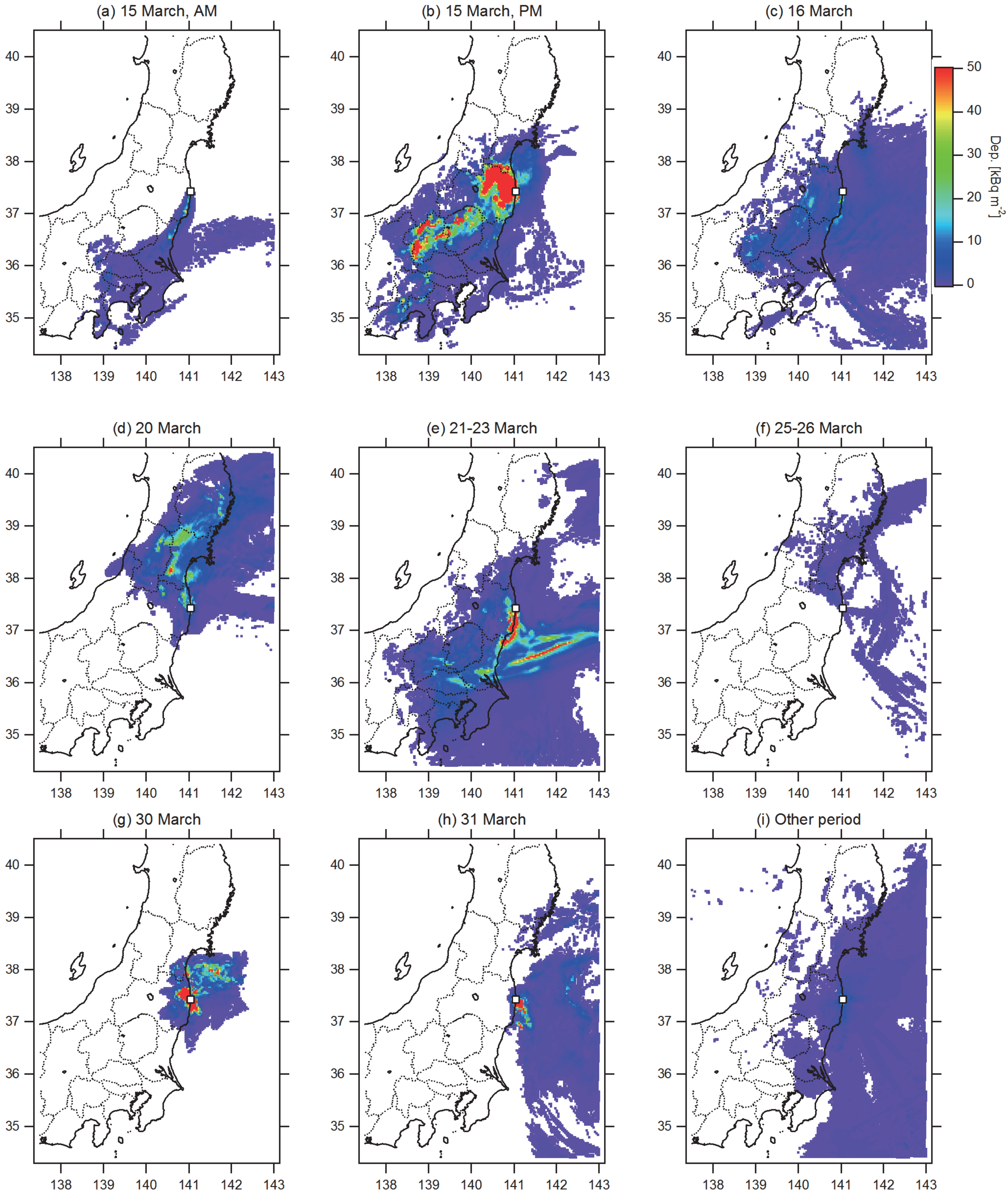

Fig. 3 Simulated Cs-137 deposition during the eight episodes (a-h) and during the periods other than these episodes (i) (Morino *et al*., 2013).

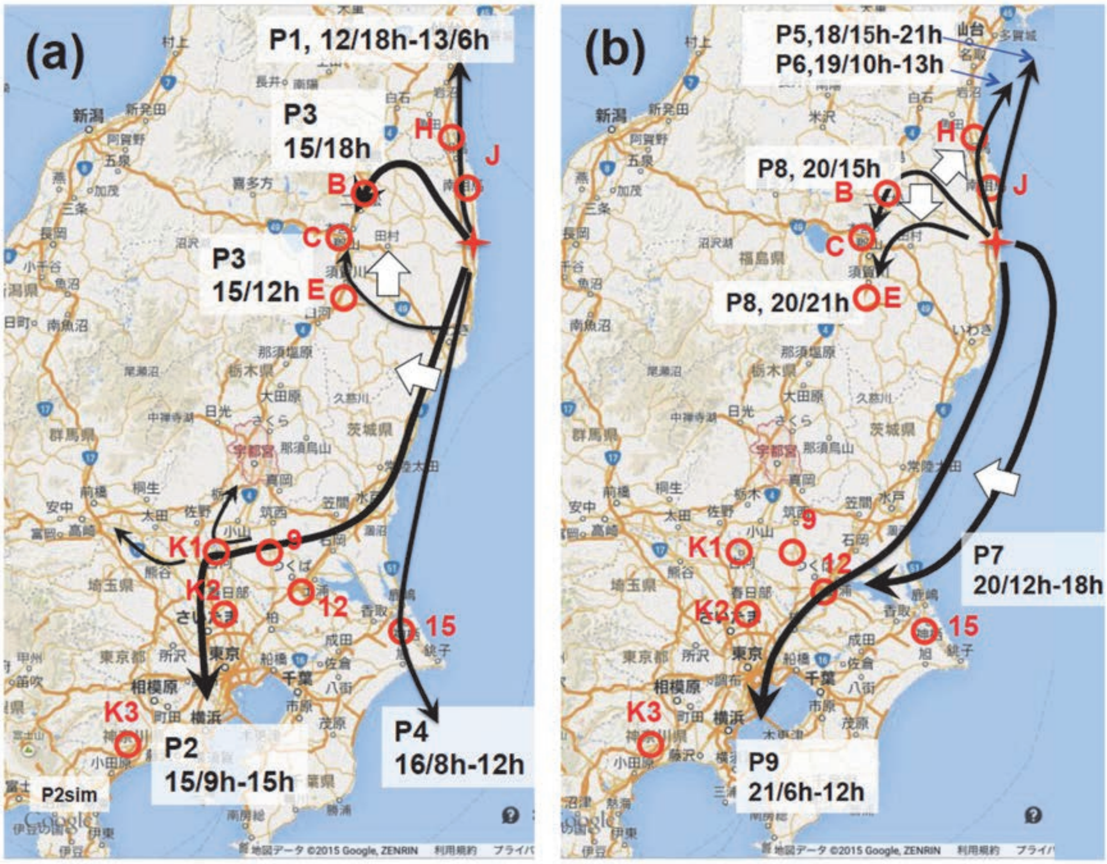

Fig. 4. Schematic diagrams of transport routes of plumes from the Fukushima Daiichi Nuclear Power Plant during March 12–16 (a) and March 18–21 (b) (Nakajima *et al*., 2017). Red circles and characters indicate the locations and codes of measurement stations.

5. Model Applications

5.1 Health Effects

To estimate the health effects arising from the Fukushima accident, dosimetric radiation surveys are critical. After the Fukushima accident, measurement in humans (e.g., activity in the thyroid) was conducted to estimate early internal doses to residents and evacuees (Tokonami *et al*., 2012). No comprehensive dosimetric radiation surveys were conducted, however (Akiba, 2012; Steinhauser *et al*., 2014). Thus dose estimates using atmospheric monitoring and modelling data are required.

Ten Hoeve and Jacobson (2012) quantified health effects with a global atmospheric simulation model. Inhalation exposure and external exposure to radionuclides (I-131, Cs-134, and Cs-137) were estimated using atmospheric simulations, and exposure-dose relationships and dose-response relationships for each radionuclide were determined using Dose and Risk Calculation software (Environmental Protection Agency, 2006). The software provides organ-specific, age-specific, and gender-specific relative risk coefficients for the inhalation pathway. They estimate an additional 130 (15-1100) cancer-related mortalities and 180 (24-1800) cancer-related morbidities incorporating uncertainties associated with the exposure-dose and dose-response models used in their study. It should be noted that the International Commission on Radiological Protection (2007) indicated that the collective effective dose (sum of effective dose to exposed subject) should not be used for risk evaluations of large populations, such as computing cancer deaths. This is because the calculation of collective effective doses includes large biological and statistical uncertainties (e.g., application of linear no-threshold models). In addition, the internal doses evaluated based on atmospheric simulations (inhalation exposure) overestimate those obtained from human measurements by several times (Kim *et al*., 2016). This overestimation might have been caused by the simulations' lack of consideration of possible dose reduction effects, such as by staying indoors or through voluntary evacuation.

5.2 Forecasting Radionuclide Distributions

JAEA developed a forecast model, SPEEDI, to predict environmental doses from radioactive materials and to assist organizations responsible for emergency planning (Imai *et al*., 1985). After the Fukushima accident, though, even though they provided the SPEEDI results to the relevant authorities, the information was reportedly not used in evacuation countermeasures (Chino, 2013).

After the Fukushima accident, SPEEDI simulations were conducted with unit-release rates on a regular basis. In addition, they were conducted for events with presumed radionuclide releases, such as containment venting or hydrogen explosions. Chino (2013) mentioned that regular calculations started for the Fukushima accident at four o'clock p.m. on March 11 and provided qualitative information such as arrival of radionuclide plumes in cities near the FDNPP. No information on source terms, however, was attained from the Emergency Response Support System (ERSS), and emergency monitoring, which could have been used for inverse or reverse analysis of source terms, did not start until March 15. Because of these problems, SPEEDI could not provide sufficient quantitative information for delineating evacuation zones.

Based on these issues, Chino (2013) suggested future usage of SPEEDI before and after the deployment of emergency monitoring. Before monitoring is deployed, SPEEDI could be used for developing emergency monitoring plans, countermeasures and plans for evacuation, and for determining the timing of presumed releases, such as contaminant venting. After it is deployed, SPEEDI could be used to evaluate the accident level and provide a detailed exposure evaluation.

The National Diet of Japan Fukushima Nuclear Accident Independent Investigation (2012) pointed out limitations to using SPEEDI to aid decisions on evacuation. SPEEDI, however, could play an important role regarding the aforementioned purposes. In that respect, specific application of SPEEDI results in decision making should also be discussed.

5.3 Understanding Radoinuclide Behavior Years after the Accident

In addition to understanding radionuclides' behaviors immediately after an accident, learning about their long-term behaviors could be important in assessing migration processes through the atmosphere or inhalation exposure to humans. After the Chernobyl accident, re-suspension from the ground contributed to atmospheric radionuclides to some extent. For example, Garland and Pomeroy (1994) showed from monitoring data at 20 European sites that Cs-137 derived from Chernobyl could be measured in the air up to six years after deposition. In the first year after the accident, the time integral of the deposition flux was a large fraction of the initial deposition. Depending upon the site, between 0.01 and 1.0 times the initial deposit accumulated in the deposition collectors. Wildfires were also a source of the secondary radioactive contamination which spread over Eastern European countries (Evangeliou *et al*., 2016).

Research on radionuclides' behaviors several years after the Fukushima accident has been very limited to date, though there were a few modeling studies on sporadic increases in Cs-137 atmospheric concentration and Cs-137 resuspension years after the Fukushima accident.

Steinhauser *et al*. (2015) analyzed sporadic increases in airborne radiocesium using an atmospheric simulation model, and indicated that debris removal operations conducted at the FDNPP site on August 19, 2013 were likely to be responsible for a late release of radionuclides.

Kajino *et al*. (2016) analyzed the effect of Cs-137 re-suspension from contaminated soil and forests using a numerical simulation model combined with a field experiment on dust emission fluxes and air concentration measurements in a contaminated area during 2013. The estimated re-suspension rate (~10^{-6} day^{-1}) was significantly lower than the decreasing rate of the ambient gamma dose rate in Fukushima Prefecture (10^{-4} – 10^{-3} day^{-1}) as a result of radioactive decay, migration in the soil and biota and decontamination. Consequently, re-suspension contributed negligibly to reducing ground radioactivity. An atmospheric simulation model which only considered dust emissions reproduced the air concentration of Cs-137 in winter, while the model underestimated the air concentration in summer by 1 to 2 orders. When re-suspension from forests was considered, however, the air concentration of Cs-137 and its seasonal variations were reproduced. They still noted that the re-suspension mechanisms, especially through forest ecosystems, remain unknown, and further studies on processes and mechanisms governing the re-suspension would be necessary.

6. Future Directions

More than five years has passed since the Fukushima accident, but there still remain many research tasks with respect to environmental assessment of radionuclides from the FDNPP, as well as model development itself utilizing various data measured after the Fukushima accident. To improve health effects estimates from FDNPP radionuclides, reproduction of Cs-137 atmospheric concentrations (Oura *et al*., 2015; Tsuruta *et al*., 2014) will be necessary. In addition, for accurate thyroid exposure estimates, improvement in I-131 model evaluations is expected when I-131 atmospheric concentration estimates from I-129 analyses become available. Further understanding of the physicochemical properties of radionuclides is also necessary for simulating both human exposures and deposition. Improved estimation of atmospheric deposition of radionuclides could lead to better simulations using oceanic and terrestrial models (Higashi *et al*., 2015; Tsumune *et al*., 2012).

Thanks to the great efforts by observational researchers, after the Fukushima accident, various observational datasets, including atmospheric concentrations, deposition and resuspension fluxes, have become available. Because of these datasets, simulation of the Fukushima accident could also serve as a platform for testing simulation models, including those for source term estimates, meteorological simulations and transport and deposition processes. Improvement of atmospheric simulation models by using datasets would contribute to the advancement of environmental modeling research.

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Yu Morino

Yu Morino is a senior researcher of the Center for Regional Environmental Research at the National Institute for Environmental Studies (NIES), Japan. He received his Ph.D. from the University of Tokyo Department of Earth and Planetary Science in 2007, and has been a member of NIES since then. His field of research is numerical modeling of atmospheric pollutants, including organic aerosols, PM2.5, reactive nitrogen and radioactive materials.

Toshimasa Ohara

Toshimasa Ohara, Ph.D., is a fellow at the National Institute for Environmental Studies (NIES). Immediately after the Fukushima accident, his group embarked on study of the dynamics of radioactive materials in multimedia environments. Currently he is research director at the Fukushima branch of NIES and leads the Environmental Emergency Research Program for contributing to environmental reconstruction and recovery in Fukushima. He is also the president of the Japan Society for Atmospheric Environment.

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