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INVITED REVIEW**Lessons learned from atmospheric modeling studies after the Fukushima nuclear accident: Ensemble simulations, data assimilation, elemental process modeling, and inverse modeling**

MIZUO KAJINO,^{1,2,3*} TSUYOSHI THOMAS SEKIYAMA,¹ ANNE MATHIEU,⁴ IRÈNE KORSAKISSOK,⁴ RAPHAËL PÉRILLAT,^{4,5} DENIS QUÉLO,⁴ ARNAUD QUÉREL,^{4,6} OLIVIER SAUNIER,⁴ KOUJI ADACHI,¹ SYLVAIN GIRARD,^{4,7} TAKASHI MAKI,¹ KEIYA YUMIMOTO,^{8,1} DAMIEN DIDIER,⁴ OLIVIER MASSON⁹ and YASUHITO IGARASHI¹

¹Meteorological Research Institute (MRI), Japan Meteorological Agency (JMA), Tsukuba, Ibaraki 305-0052, Japan

²Faculty of Life and Environmental Sciences, University of Tsukuba, Tsukuba, Ibaraki 305-8577, Japan

³RIKEN Advanced Institute for Computational Science, Kobe, Hyogo 650-0047, Japan

⁴Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-SANTE, SESUC, BMCA, 92262, Fontenay-aux-Roses Cedex, France

⁵Strathom Energie, 75002, Paris, France

⁶Bertin Technologies, 78180, Montigny-le-Bretonneux, France

⁷Phimeca, Engineering, 75012, Paris, France

⁸Research Institute for Applied Mechanics, Kyushu University, Kasuga, Fukuoka 816-8580, Japan

⁹Institut de Radioprotection et de Sûreté Nucléaire (IRSN), BP3, 13115, St Paul lez Durance Cedex, France

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Modeling studies on the atmospheric diffusion and deposition of the radiocesium associated with the Fukushima Dai-ichi Nuclear Power Plant accident is reviewed here, with a focus on a research collaboration between l'Institut de Radioprotection et de Sûreté Nucléaire (IRSN)—the French institute in charge of evaluating the consequences of nuclear accidents and advising authorities in case of a crisis—and the Meteorological Research Institute (MRI) of the Japan Meteorological Agency—an operational weather forecasting center in Japan. While the modelers have come to know that wet deposition is one of the key processes, the size of its influence is unknown. They also know that the simulation results vary, but they do not know exactly why. Under the research collaboration, we aimed to understand the atmospheric processes, especially wet deposition, and to quantify the uncertainties of each component of our simulation using various numerical techniques, such as ensemble simulations, data assimilation, elemental process modeling, and inverse modeling. The outcomes of these collaborative research topics are presented in this paper. We also discuss the future directions of atmospheric modeling studies: data assimilation using the high temporal and spatial resolution surface concentration measurement data, and consideration of aerosol properties such as size and hygroscopicity into wet and dry deposition schemes.

Keywords: ensemble simulation, data assimilation, wet deposition, aerosol properties, inverse modeling

REVIEW ON THE ATMOSPHERIC MODELING STUDIES

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accidentally released nuclear fission products into the atmosphere and surrounding area in March 2011. A great number of studies have been performed and facts have been accumulated after the more than six years that have passed since the event. In terms of the impacts of the nuclear accident on terrestrial animal health,

radiocesium and radioiodine are important due to their relative abundances in the atmosphere, enough long physical half-lives to survive during atmospheric transport, and accumulation in the human body: The total released amount of ¹³¹I and ¹³⁷Cs were estimated as 100–200 PBq and 10–20 PBq, respectively (Katata *et al.*, 2015 and references therein), physical half-lives of ¹³³I, ¹³²Te-¹³²I, ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs are 21.0 h, 3.2 d–2.3 h, 8.0 d, 2.1 y, and 30.0 y, respectively, and I and Cs are accumulated to thyroid and muscle tissues, with bio-logical half-lives of 80 d and 30–150 d, respectively (WHO, 2001).

Radiocesium is particularly important because of its longer physical half-life (compared to that of radioiodine)

*Corresponding author (e-mail: kajino@mri-jma.go.jp)

when habitation is concerned, e.g., when considering sporadic releases (Steinhauser *et al.*, 2015) and resuspension from terrestrial ecosystems (Kajino *et al.*, 2016). There are still evacuation-directed zones in Fukushima Prefecture and their ambient gamma doses are almost entirely attributed to radiocesium.

Atmospheric modeling study comprises two important parts: emission estimation and dispersion modeling. A number of studies have been conducted for the respective parts for emission (e.g. Chino *et al.*, 2011; Stohl *et al.*, 2012; Terada *et al.*, 2012; Winiarek *et al.*, 2012, 2014; Saunier *et al.*, 2013; Achim *et al.*, 2014; IAEA, 2015; Katata *et al.*, 2015; Yumimoto *et al.*, 2016; Liu *et al.*, 2017) and for dispersion modeling (e.g. Morino *et al.*, 2011; Yasunari *et al.*, 2011; Katata *et al.*, 2012a, b; Morino *et al.*, 2013; Hirao *et al.*, 2013; Adachi *et al.*, 2013; Draxler *et al.*, 2015; Groëll *et al.*, 2014; SCJ, 2014; Saito *et al.*, 2015; Sekiyama *et al.*, 2015; Quérel *et al.*, 2015; Kristensen *et al.*, 2016; Nakajima *et al.*, 2017). Validations with observations are particularly indispensable for the both modeling studies. Total depositions of radiocesium and their horizontal distributions have been revealed through systematic aircraft monitoring together with ground-based observations (NRA, 2012)—one of the massive observation data sets to validate the simulation results. Transport pathways and the deposition mechanisms of radionuclides, which cause the spatial distributions of radiocesium deposition, have been qualitatively understood following the substantial efforts of atmospheric modelers (e.g., Morino *et al.*, 2013; Arnold *et al.*, 2015; Katata *et al.*, 2015; Leadbetter *et al.*, 2015; Saito *et al.*, 2015; Nakajima *et al.*, 2017). However, the transport and deposition are not fully understood, as (1) there are still discrepancies between the simulations and observations in some regions and (2) the simulated depositions varied substantially between models (SCJ, 2014). One of the major reasons for the uncertainty in the simulations is the difficulty in wet deposition process modeling. Previous studies have shown that the wet deposition process is the dominant mechanism of radiocesium deposition over Japan (e.g., SCJ, 2014). However, the simulation of wet deposition processes is highly complex; thus, the discrepancies between simulations and observations, as well as differences among the simulation results from different models, are quite large, even for conventional substances, such as sulfate, nitrate, and ammonia (Wang *et al.*, 2008).

More than six years have passed and we have come to know a considerable amount about the behavior of radiocesium in the atmosphere, as mentioned above. However, while we know that wet deposition is one of the key processes, the size of its influence is unknown. We also know that the simulation results vary, but we do not know exactly why. Multi-model intercomparison studies can

show the magnitude of uncertainty but they cannot identify the cause of uncertainty. Therefore, in order to understand the atmospheric processes, especially that of wet deposition, and to quantify the uncertainties of each component of the simulation, a research collaboration was initiated in 2013 between l'Institut de Radioprotection et de Sûreté Nucléaire (IRSN) and the Meteorological Research Institute (MRI). Our collaboration was funded by the Japan-France integrated action program (SAKURA) of the Japan Society for the Promotion of Science (JSPS) and the Ministère des Affaires Étrangères et du Développement International (MAEDI) from September 2014 to August 2016.

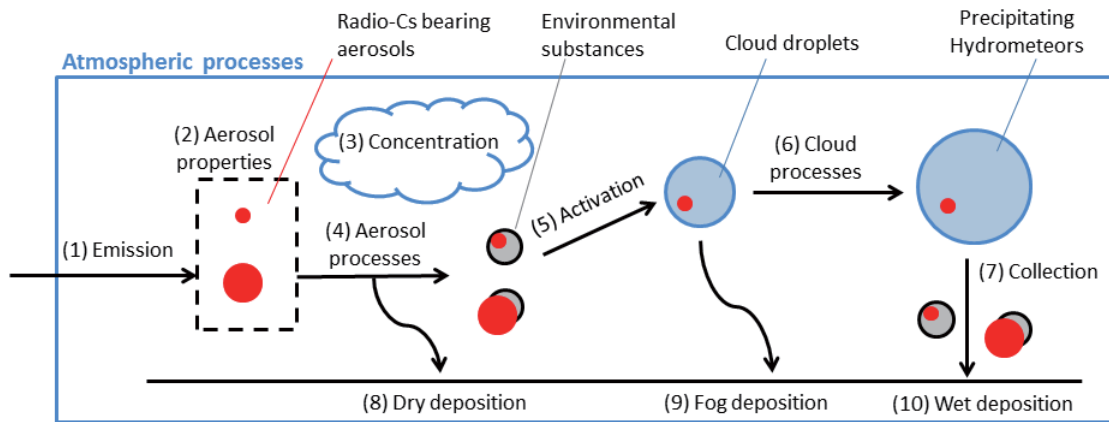
Our work is unique since we applied a state-of-the-art ensemble data assimilation approach (Kunii, 2014; Sekiyama *et al.*, 2015) to the high-resolution weather forecast model operationally developed by the Japanese national weather service (Japan Meteorological Agency; JMA). This ensemble data assimilation system can reproduce meteorological fields with the highest possible reliability at fine horizontal resolutions (up to sub-km scales) using operational observational datasets. This system can simultaneously provide meteorological ensemble members, which are usable for evaluating model uncertainty, called ensemble analysis. The ensemble analysis enables us to quantitatively compare the sensitivity of simulated radiocesium deposition to wet deposition schemes (e.g., Quérel *et al.*, 2015) or aerosol properties (e.g., Adachi *et al.*, 2013) against the difference in simulated radiocesium deposition among meteorological ensemble members. The ensemble analysis also enables us to estimate the uncertainty in the source-term estimations of radionuclides using inverse modeling with meteorological ensemble members.

In the following sections, we presented the numerical methods used under the SAKURA program. Then, the framework of the program and part of the outcomes of these collaborative research topics, including some works which are still ongoing, are presented in the paper. Based on our collaborative activity, we also discuss the future directions for atmospheric modeling studies associated with the accident.

NUMERICAL METHODS

Atmospheric dispersion models

IRSN has two types of operational atmospheric dispersion models for radionuclides for different purposes: a Gaussian puff model, pX (Soulhac and Didier, 2008) and an Eulerian model, ldX. pX is used for the prediction of the dispersion within the area close to a nuclear power plant. ldX (developed based on the chemical transport model Polair3D; Boutahar *et al.*, 2004) is used for the prediction of long-range transport, chemical formation,



		Objectives		Data / Methods		Representative works
Our research background	Estimation	of	(1)	using	(3)	Mathieu et al. (2012) Saunier et al. (2013)
					(8), (9), (10)	Yumimoto et al. (2016)
	Sensitivity	of	(3), (7), (8), (10)	to	(1), (4), (5), (6), (7)	Korsakissok et al. (2013) Groëll et al. (2014) Quérel et al. (2015)
			(4), (5), (6), (7), (8), (9), (10)		(2)	Adachi et al. (2013)
SAKURA program	Uncertainty	in	regional-scale dispersion modeling	using	emulation	Girard et al. (2016)
			local-scale meteorological modeling		NHM-LETKF	Sekiyama et al. (2015)*
	Impacts	on	estimation of (1)	due to	uncertainty in local-scale meteorology*	Saunier et al. (2016)
			(3), (8), (9), (10)			Quérel et al. (2016) Pérrillat et al. (2016)

Fig. 1. Outline of the French-Japanese bilateral collaboration program (SAKURA).

and both dry and wet scavenging processes.

MRI also has two types of atmospheric dispersion models: RATM, which is a Lagrangian model (Shimbori *et al.*, 2009, 2010), and three Eulerian models: NHM-Chem (based on the chemical transport model RAQM2; Kajino *et al.*, 2012), MRI-CCM2 (Deushi and Shibata, 2011), and MASINGAR (Tanaka *et al.*, 2003). These models are used by JMA for the operational prediction of volcanic ash, photochemical oxidants, ultraviolet radiation, and Asian mineral dust. The atmospheric processes associated with radionuclides are incorporated into RATM, NHM-Chem, and MASINGAR. We also utilize other community models, such as WRF, which is a meteorology model (Skamarock *et al.*, 2008); CMAQ, which is an Eulerian chemical transport model (Byun and Schere, 2006); and LM which is a Lagrangian transport model (Kajino *et al.*, 2016), for multi-model/multi-module inter-comparison purposes.

We use different types of models as each of them has pros and cons. The Gaussian puff and Lagrangian models are more efficient and accurate than the Eulerian models when solving point source problems within a limited area (they are computationally expensive when accuracy in

expanded downwind areas is needed). On the other hand, Eulerian models are more efficient and accurate than Gaussian puff and Lagrangian models when solving long-range transport problems (these will be computationally expensive when accuracy near the point source areas is needed). All the details of the model formulations are presented in the references given in this subsection.

Ensemble simulation and data assimilation

Ensemble data assimilation is a powerful tool for reproducing an accurate meteorological field. In this study, we implemented a Local Ensemble Transform Kalman Filter (LETKF; Kunii, 2014 and references therein) in our data assimilation system, coupled with an operational regional weather forecast model called the JMA's Non-Hydrostatic Model (JMA-NHM; Saito *et al.*, 2006). Hereafter, this data assimilation system is called NHM-LETKF. Sekiyama *et al.* (2015) applied the NHM-LETKF to the FDNPP accident analysis, such that they assimilated the JMA's operational meteorological datasets, which are derived from observations at manned observatories, balloons, wind profilers, aircrafts, ships, buoys, and satellites. Sekiyama *et al.* (2015) also assimilated the obser-

vational data of the automated meteorological data acquisition system (AMeDAS) and the Tokyo Electric Power Company (TEPCO) monitoring post data. In fact, the AMeDAS surface wind data assimilation significantly improved the prediction of surface activity concentration of radiocesium (Sekiyama *et al.*, 2017a). The AMeDAS network comprises approximately 1,300 stations throughout Japan, which are laid out at average intervals of 17 km. The TEPCO monitoring posts are placed at FDNPP and the Fukushima Dai-ni Nuclear Power Plant, which is located 12 km south of FDNPP. The ensemble size was set to 20 members and the horizontal model resolution was set to 15 km, 3 km, or 500 m. The technical details of NHM-LETKF are found in Kunii (2014) and the configuration of the data assimilation and the ensemble production method for the FDNPP accident analysis are found in Sekiyama *et al.* (2015).

Inverse modeling

Both IRSN and MRI use inverse modeling tools to estimate the emission fluxes of the source using atmospheric dispersion models, an a priori emission scenario, and the observational datasets of the atmospheric concentrations and depositions. Saunier *et al.* (2013) use the IdX model, a zero-emission a priori scenario, and the gamma dose rate measurements. Winiarek *et al.* (2012) use Polair3D, a zero-emission a priori scenario and the air concentration measurements both in Japan and in North America. Winiarek *et al.* (2014) use Polair3D, a zero-emission a priori scenario, and both the atmospheric concentration and deposition measurements in Japan. Maki (2015) uses the MASINGAR model, the Terada *et al.* (2012)'s source term as a priori emission scenario, and the daily mean observational data of 51 global sites. Yumimoto *et al.* (2016) use the CMAQ model, which was modified by Morino *et al.* (2011), the Terada *et al.* (2012)'s source term as a priori emission scenario, and the airborne measurements of the ¹³⁷Cs deposition field (NRA, 2012). Saunier *et al.* (2013), Winiarek *et al.* (2012, 2014) and Maki (2015) use Bayesian synthesis method. Yumimoto *et al.* (2016) employ Green's function method.

COLLABORATION UNDER THE SAKURA PROGRAM

Figure 1 illustrates the past and the current research collaborations and the links between the two institutes' research topics on the atmospheric processes involved in the dispersion and deposition of radioactive substances. Before the SAKURA program started, a series of works were accomplished by the two groups using their own methods concerning the following topics;

(1) Source-term estimation: Mathieu *et al.* (2012) used pX and IdX, Winiarek *et al.* (2012, 2014) used Polair3D, Saunier *et al.* (2013) used IdX, Maki (2015) used

MASINGAR, and Yumimoto *et al.* (2016) used CMAQ.

(2) Deposition study: Korsakissok *et al.* (2013), Groëll *et al.* (2014) and Quérel *et al.* (2015) used IdX and Adachi *et al.* (2013) and Kajino (2015) used NHM-Chem.

(3) Uncertainty and sensitivity study: Korsakissok *et al.* (2013) used pX, Girard *et al.* (2014) used IdX, Girard *et al.* (2016) used the Gaussian process emulator of IdX, and Sekiyama *et al.* (2015) used ensemble simulations produced by NHM-LETKF. Quérel *et al.* (2015) and Kajino (2015) discussed the uncertainty in simulations due to the selections of wet deposition schemes and aerosol microphysical properties, respectively.

We also reported our simulation results using IdX and NHM-LETKF-Chem to the Science Council of Japan's (SCJ's) model inter-comparison study (SCJ, 2014).

The SAKURA program ended in August 2016, but the collaboration is still ongoing. During the two years, we were supposed to hold two workshops each year. We had a total of five workshops, either in France or in Japan. Through the workshops, we shared our datasets and research experiences and discussed the design of the collaboration based on the abovementioned three topics. During the program, meteorological analysis, the 20 ensemble members produced by Sekiyama *et al.* (2015) and the source term obtained by inversion modeling (Yumimoto *et al.*, 2016) were shared with the MRI and IRSN scientists. Using the meteorological ensemble simulations, we have revisited the three listed topics: source-term estimation, deposition, and uncertainty studies.

Our collaboration was successful in the sense that MRI is a meteorological research institute that belongs to JMA, an operational weather forecasting center in Japan, whereas IRSN is an operational and research center focused on environmental radioactivity during a nuclear crisis. MRI imported technologies on the prediction of radioactivity from IRSN, while IRSN received meteorological ensemble analyses with finer grid resolutions from MRI. We could collaborate and work towards a further understanding and improvement of source-term assessment, atmospheric dispersion, and deposition modeling studies.

ATMOSPHERIC DISPERSION MODELING AND ITS SENSITIVITY TO HORIZONTAL GRID RESOLUTIONS

Sekiyama *et al.* (2015) performed meteorological simulations and dispersion modeling with multiple horizontal grid resolutions, namely, 15 km, 3 km, and 500 m, as shown in Figure 2. Such an analysis is indispensable for model simulations in Japan because the topography in Fukushima and the surrounding prefectures are complex, containing mountainous and hilly regions, and thus, the local wind fields and precipitation patterns are sub-

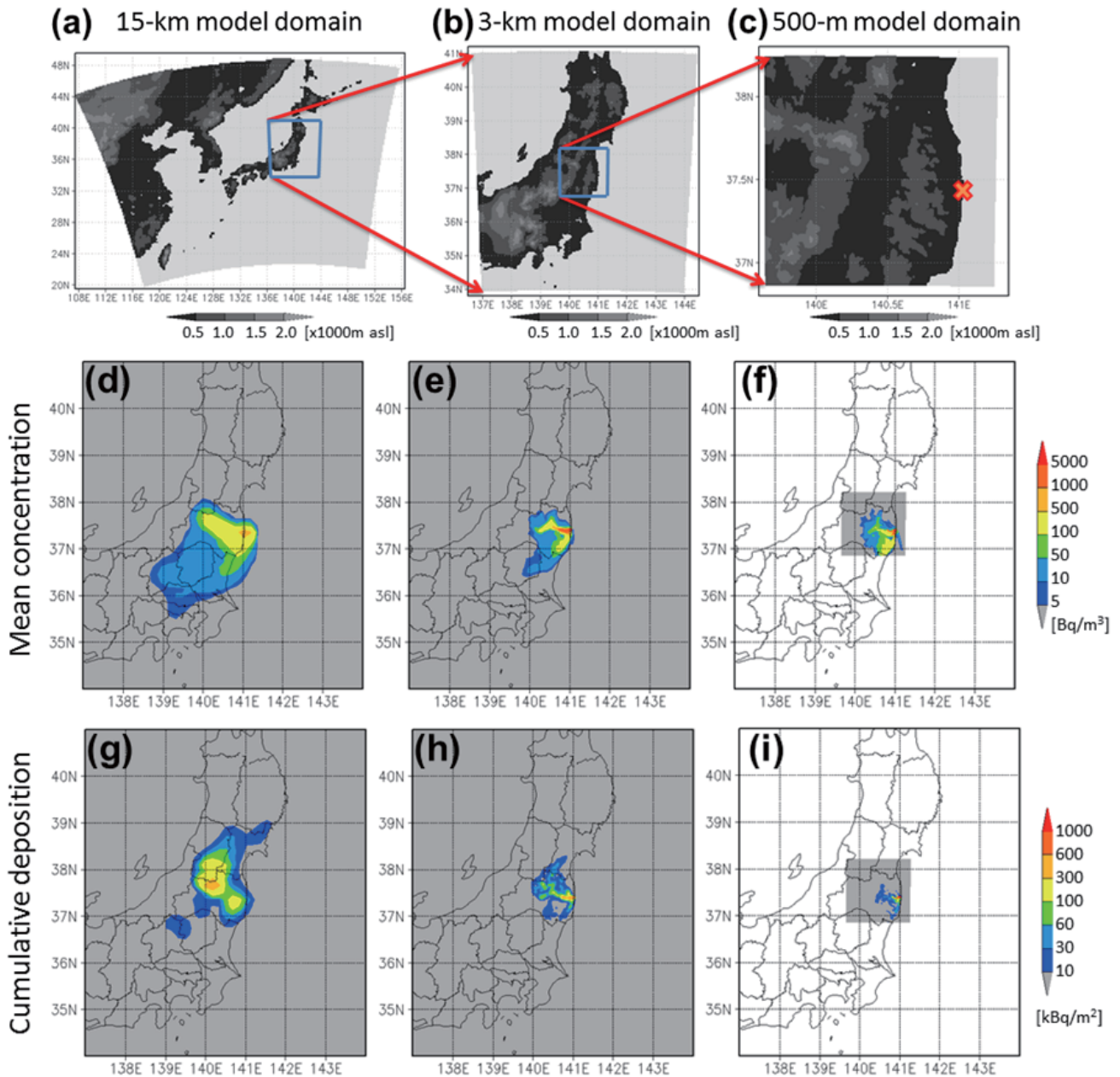


Fig. 2. (a–c) NHM-LETKF model domains of the (a) 15-km, (b) 3-km, and (c) 500-m grid spacing analysis. The cross in panel (c) indicates the location of FDNPP. (d–f) The mean surface ^{137}Cs concentrations from 00 UTC March 15 to 00 UTC March 16, 2011 as simulated by NHM-LETKF-Chem with the three different grid spacing analyses. (g–h) Same as (d–f) but the cumulative deposition of ^{137}Cs (modified figures 2, 5, and 7 of Sekiyama *et al.*, 2015).

stantially affected by the complexity of the topography. Previous multi-model inter-comparison studies, such as SCJ (2014), Draxler *et al.* (2015), and Kristiansen *et al.* (2016), contained models with different grid resolutions, but the differences due solely to the grid resolutions were not extracted because conditions other than the grid resolutions (such as emissions, deposition parameters and

models themselves) were different as well. Sekiyama *et al.* (2015) conducted this analysis with a single meteorological and transport model to identify the simulation differences due solely to the grid resolutions. As summarized in Table 2, the simulated concentrations and deposition fields at the 3-km grid resolution were well correlated with those at the 500-m resolution as a benchmark,

Table 1. List of acronyms

CMAQ	The Models-3 Community Multiscale Air Quality model
FDNPP	Fukushima Dai-ichi Nuclear Power Plant
JSPS	Japan Society for the Promotion of Science
ldX	IRSN's operational long-range Eulerian model
LM	The Lagrangian Model
MAEDI	Ministère des Affaires Étrangères et du Développement International
MASINGAR	MRI's Model of Aerosol Species IN the Global AtmospheRe
MRI-CCM2	MRI's Chemistry Climate Model 2
NHM	JMA's operational Non-Hydrostatic Model
NHM-Chem	Chemical transport model coupled with NHM
NHM-LETKF	Local Ensemble Transform Kalman Filter coupled with NHM
NHM-LETKF-Chem	Chemical transport model coupled with NHM-LETKF
pX	IRSN's operational Gaussian puff model
RATM	JMA's operational regional atmospheric transport model
RAQM2	Regional Air Quality Model 2
SAKURA program	The Japan-France integrated action program between Japan Society for the Promotion of Science (JSPS) and Ministère des Affaires Étrangères et du Développement International (MAEDI)
WRF	Weather Research and Forecasting model

Table 2. Horizontal distribution correlations of the surface concentrations and depositions of ^{137}Cs as simulated by the models (modified from tables 5 and 6 of Sekiyama *et al.*, 2015)

	15-km grid \leftrightarrow 3-km grid	3-km grid \leftrightarrow 500-m grid	500-m grid \leftrightarrow 15-km grid
Concentration	0.75	0.90	0.64
Deposition	0.54	0.84	0.42

whereas the simulation at the 15-km grid resolution was degraded due to the lack of modeled complex topography. The correlation coefficients of the deposition fields between 15 km and 500 m are lower than 0.5.

Another remarkable feature of Sekiyama *et al.*'s (2015) work is that they used LETKF for their meteorological simulation, so that they produced meteorological ensemble members and meteorological analysis simultaneously. They concluded that a grid resolution of 3-km was adequate for simulating the dispersion and deposition of radio-Cs released from FDNPP, and they provided the most realistic meteorological fields by assimilating the observation data with a meteorological ensemble, reflecting the prediction errors. To date, we have worked on source-term estimation (e.g., Saunier *et al.*, 2013), sensitivity studies for wet deposition parameters (e.g., Quérel *et al.*, 2015), and aerosol properties (e.g., Adachi *et al.*, 2013; Kajino, 2015), as presented in the following subsections, but none of the studies had evaluated their results against the uncertainties in the meteorological simulation. In the framework of the SAKURA program, we shared this meteorological analysis and the ensembles, and we are revisiting the above issues. Most of our works are still ongoing, but some have been completed (Saunier *et al.*, 2016; Périllat *et al.*, 2016) and are presented in the following subsections.

INVERSE MODELING FOR SOURCE-TERM ESTIMATION

As shown in Fig. 3, Saunier *et al.* (2013) assessed a source term via inverse modeling. Their technique is to solve the inverse problem by relating atmospheric measurements to emission rates using an atmospheric dispersion model. They used the ambient gamma dose rate measured at the monitoring posts of the Nuclear Regulation Authority (NRA). The gamma dose rate includes both cloud shine (air concentration) and ground shine (deposition) effects. They extracted only the cloud shine effect from the gross rate to assess the potential releases periods. Then, the gross rate is used in the inversion to evaluate the release rates among the periods identified in the previous step. As presented in the previous section, ldX was used for the dispersion modeling. Figures 3(d)–(g) presents the time series of the gamma dose rates at the four monitoring posts. The simulation with the inversed source term (red) is better able to reproduce both the peak height (air concentration) and decay afterwards (deposition) than the related simulation done by Mathieu *et al.* (2012), which used the source term (blue).

Saunier *et al.* (2013) used the European Center for Medium-Range Weather Forecasts (ECMWF)'s analysis (0.125 deg and 3 h resolutions) as the meteorological

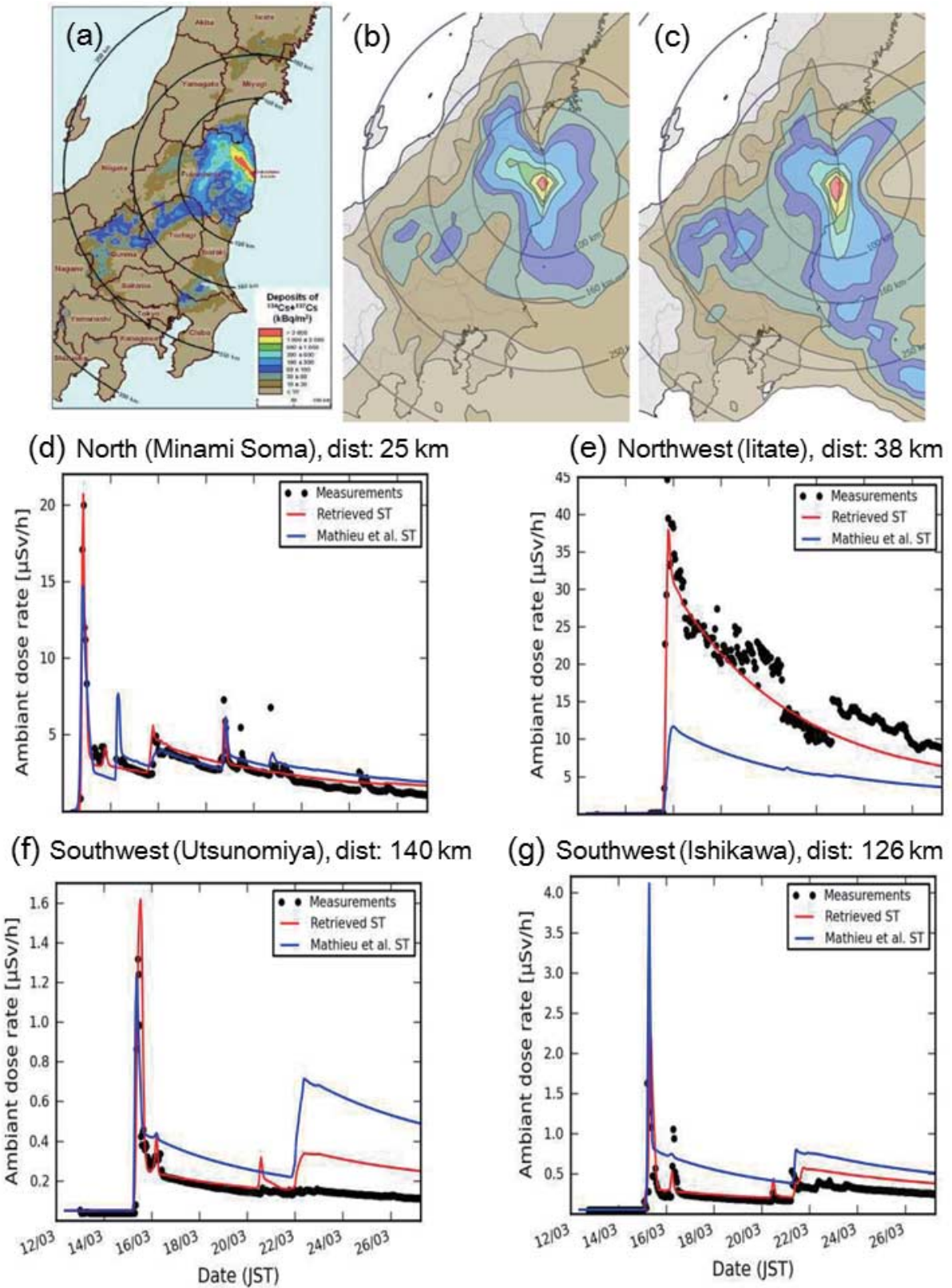


Fig. 3. Cumulative deposition of ^{137}Cs and ^{134}Cs (a) observed, (b) simulated using the retrieved source term, or (c) simulated using a priori source term. (d)–(g) Comparisons of the dose rate observations (black dots) with those simulated using the retrieved source term (red) and using an a priori source term (blue) at the four locations in Japan, showing the directions and distances from FDNPP (modified from figures 5 and 9 of Saunier et al., 2013).

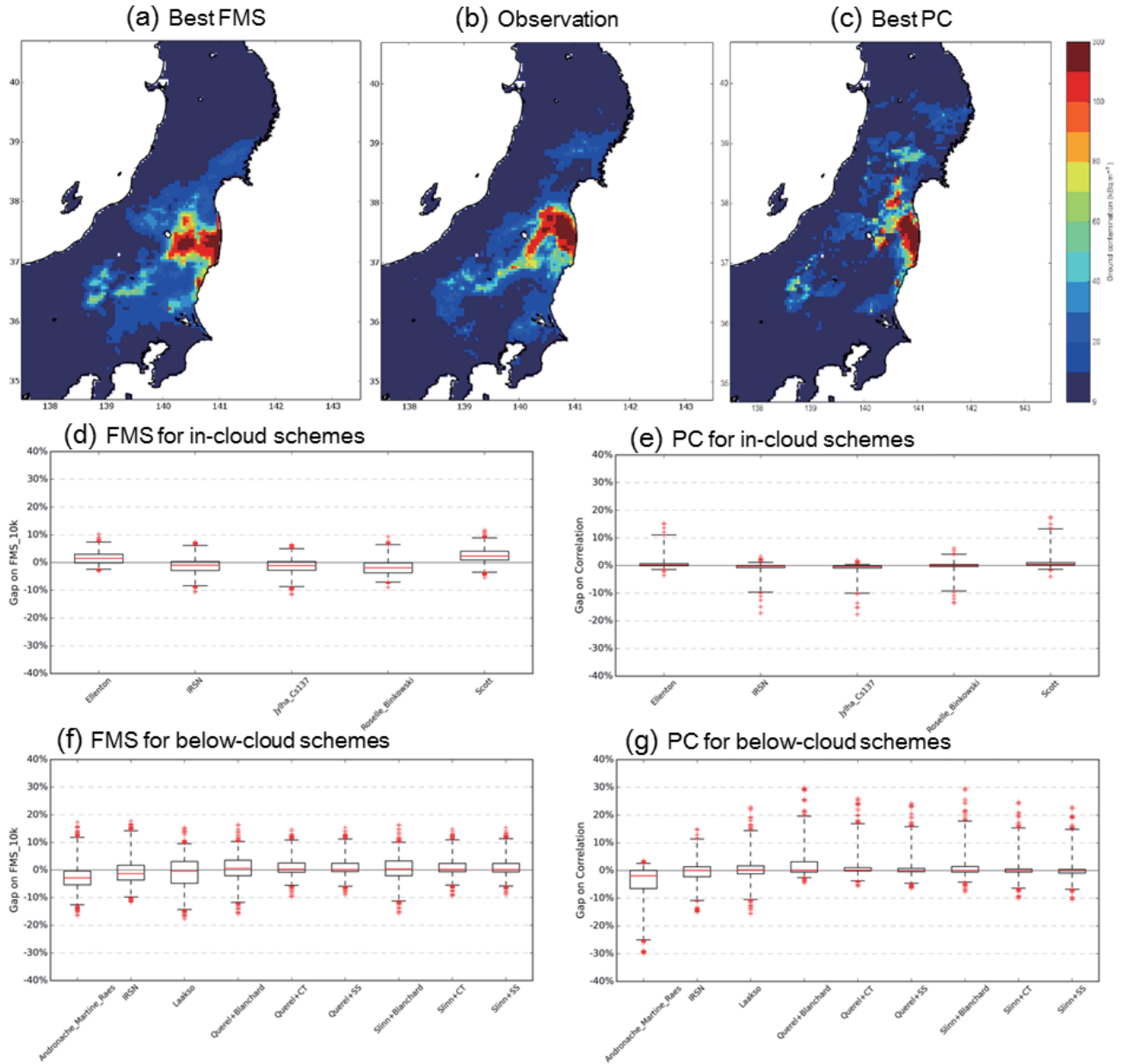


Fig. 4. ^{137}Cs deposition maps of (a) the simulation with the best FMS, (b) observations, and (c) the simulation with the best PC from among the combinations of several in-cloud and below-cloud deposition schemes (modified from figure 1 of Qu  rel *et al.*, 2016). (d)–(g) show the box plots for the (d, f) FMS and (e, g) PC of the simulations of the (d, e) in-cloud and (f, g) below-cloud deposition schemes.

fields. As discussed in Saunier *et al.* (2013) and as shown in Sekiyama *et al.* (2015), the horizontal grid resolution of 0.125 deg may not be adequate to resolve the complexity of the topography, and the 3 h resolution may not be adequate to follow the rapid changes of the wind direction near FDNPP during the release events. The source term is estimated to reproduce the best performance of the dispersion model used for the inversion. In other words, the inverse emission highly depends on the mete-

orological fields, the number and type of measurements, and the dispersion model used. The SAKURA program used the inverse modeling study in two ways: (1) by providing the assimilated meteorological fields at finer grid (3-km) and time (10 min) resolutions and (2) by providing the meteorological ensemble that enables the evaluation of the uncertainty in the estimated source term, as partly presented in Saunier *et al.* (2016).

ATMOSPHERIC DISPERSION MODELING AND ITS SENSITIVITY TO WET DEPOSITION SCHEMES

The selection of an appropriate wet deposition scheme is particularly important for the FDNPP case study mainly for the following two reasons: (1) wet deposition modeling is difficult and highly uncertain and (2) the main contamination of radiocesium occurred via wet deposition (SCJ, 2014). Qu  rel *et al.* (2015) implemented various simple wet deposition schemes in IdX and compared the results with the observations, as presented in Fig. 4. The wet deposition process comprises two distinct processes: in-cloud scavenging (or rainout) and below-cloud scavenging (or washout). The in-cloud process indicates that aerosols are activated as cloud condensation nuclei form cloud droplets and are scavenged via the subsequent cloud microphysical processes. The below-cloud processes indicate that aerosols are collected by settling hydrometeors and are scavenged. There are two ways to solve the wet deposition process in dispersion models: process-based formulations and simple parameterizations. The process-based methods calculate the scavenging rates based on the physical and chemical properties of the aerosols and clouds, sometimes including electrostatic forces (Tinsley *et al.*, 2006) and turbulence (Qu  rel *et al.*, 2014). The simple parameterization methods calculate the rate as simple functions of the precipitation rate (as listed in tables 1 and 2 of Qu  rel *et al.* (2015)). Qu  rel *et al.* (2015) compared simulations of several below-cloud and in-cloud scavenging schemes against the airborne observations of the deposition map of radiocesium in terms of the two statistical indicators: the Pearson correlation coefficient (PC) and the figure of merit in space (FMS). They also included simulations using the radar/rain-gauge precipitation observations of the Japan Meteorological Agency to predict the scavenging rates in the model instead of using the simulated precipitation.

As shown in Fig. 4, the simulated deposition patterns, as well as the simulated amounts, significantly differed among the combinations of the wet deposition schemes. Additionally, the combinations showing the best PC and best FMS scores were different, which indicates that there was no absolute best combination of schemes. Uncertainty in the boundary conditions (i.e., meteorology and source terms) might prevent the successful selection of an accurate deposition scheme, but this could be resolved in the future under the SAKURA program, which raised the accuracy of the meteorological fields and the source term by inverse estimation using accurate meteorology.

ATMOSPHERIC DISPERSION MODELING AND ITS SENSITIVITY TO AEROSOL PROPERTIES

The importance of the meteorological fields, source

term, and deposition schemes was presented in the previous subsection. Another important aspect of the elemental process modeling issue, specifically, the microphysical properties of Cs-carrying aerosols, is addressed in this subsection.

All the models aside from NHM-Chem did not explicitly model the microphysical properties (i.e., size and hygroscopicity) of Cs-carrying aerosols but instead assumed a water-soluble particle with an approximately 1 μm diameter. However, Adachi *et al.* (2013) reported that, in the early stages of the accident, the carrier aerosols of radiocesium were spherical, water-insoluble particles (hereafter, Cs-balls), and they predicted that the atmospheric behavior of these aerosols would be significantly different from those of the water-soluble submicron particles described by Kaneyasu *et al.* (2012). Below-cloud scavenging is not efficient for particles of approximately 1 μm because of their small inertias and slow Brownian motions. The submicron water-soluble particles are efficiently scavenged by in-cloud processes, while the water-insoluble Cs-balls may not be due to the very high supersaturation needed for the water-insoluble aerosols to act as cloud condensation nuclei. Thus, the below-cloud processes are probably the dominant scavenging processes of Cs-balls. Kajino (2015) assessed the uncertainty of the simulated deposition of radiocesium caused by the size and hygroscopicity of its carrier aerosols, and compared it to the uncertainty caused by different meteorological simulations.

Figure 5 shows a comparison of the depositions simulated of the three meteorological fields (NHM and WRF with two cloud microphysics schemes, Morrison *et al.* (2009) and WDM6 (Lim and Hong, 2010), named WRF-MORR and WRF-WDM6, respectively) for the water-soluble and water-insoluble particles (a–f) and with aircraft observational data (g–l). Note that, following Morino *et al.* (2013) and Katata *et al.* (2015), the correlation coefficient R and mean bias (MB) were calculated only when the observed values were larger than 10 kBq/m².

The simulated results were significantly different between the particles assumed to be 100% water soluble (a–c, g–i) and those assumed to be 100% water insoluble (d–f, j–l), because the rainout of the Cs-balls was not considered, whereas the dry deposition velocity of the Cs-balls was approximately four times that of water-soluble submicron aerosols. Adachi *et al.* (2013) detected Cs-balls only in samples collected during the early stages of the accident (14–15 March), but they reported that radioactive Cs was carried by water-soluble aerosols later (20–22 March). Kaneyasu *et al.* (2012), who analyzed samples collected from 28 April to May 12 and during 12–26 May, also reported radiocesium carried by water-soluble aerosols. Doi *et al.* (2013) reported the size distribution of radiocesium for April 4–11 and 14–21, between the

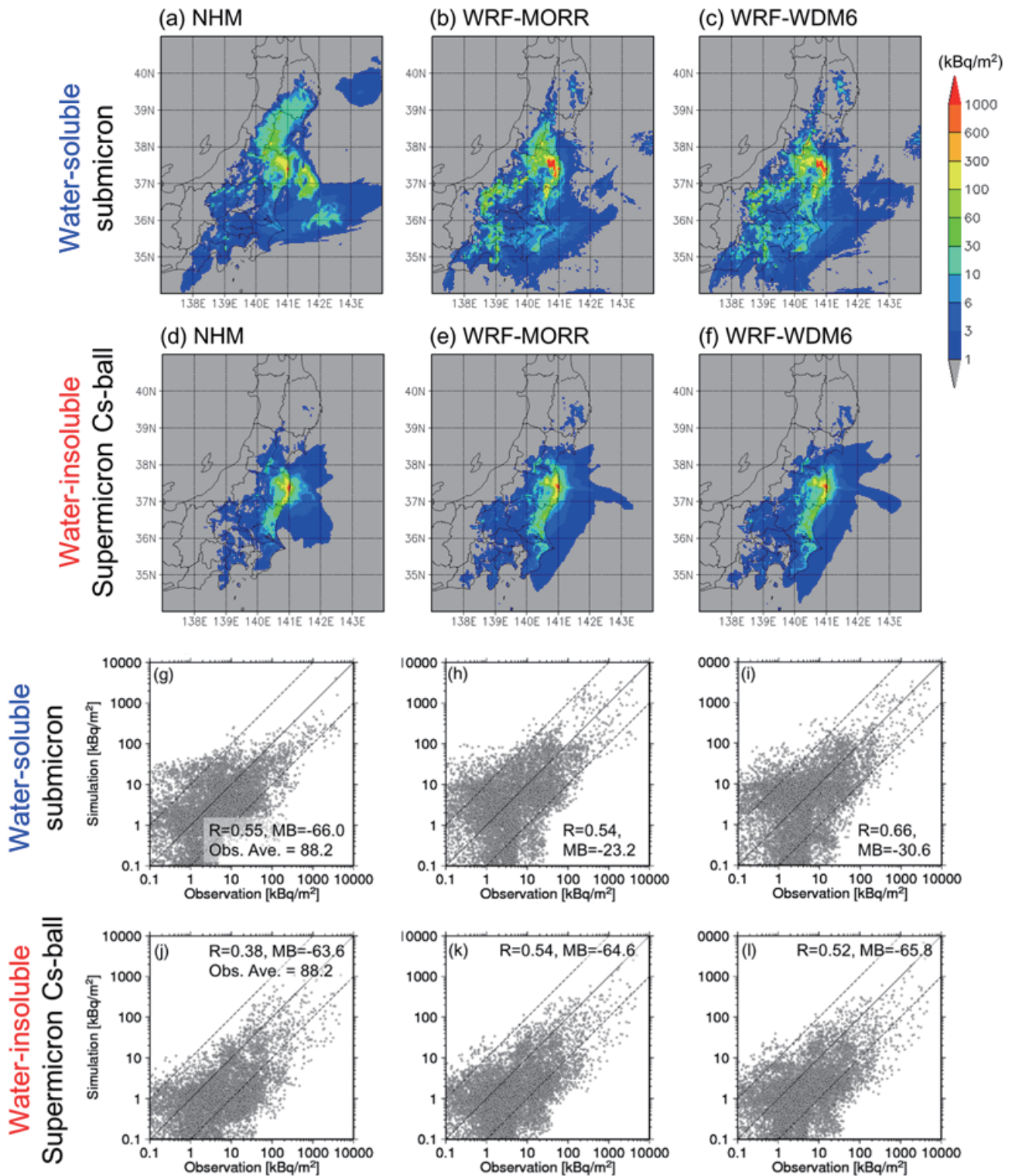


Fig. 5. (a–f) Cumulative depositions simulated using the meteorological fields of (a, d) NHM, (b, e) WRF-MORR, and (c, f) WRF-WDM6 under the assumption that 100% of the ^{137}Cs was carried by (a–c) water soluble or (d–f) water-insoluble particles. (g–l) Scattergrams of the simulations and the aircraft observations. The simulations of (g)–(l) correspond to those of (a)–(f). The statistics R and MB in the panels were calculated on a linear-linear basis.

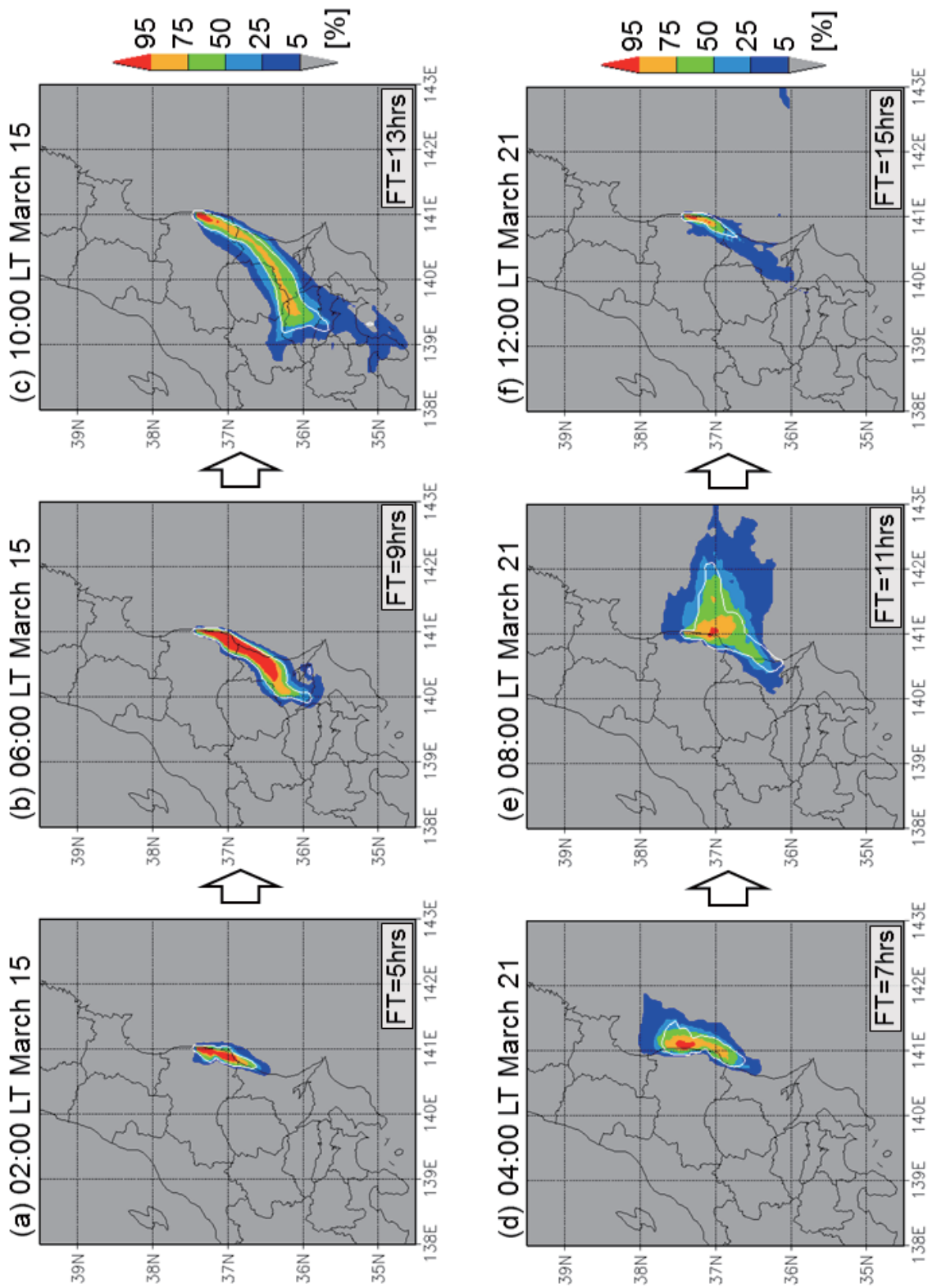


Fig. 6. Percentile distributions of the 20 ensemble forecast members, where the surface ^{137}Cs concentrations exceed the threshold ($=15 \text{ Bq/m}^3$), as predicted by NHM-LETKF-Chem. The white contour lines indicate the 15 Bq/m^3 concentrations of the deterministic forecast. The local times of the panels (a)–(c) are 2:00, 6:00, and 10:00 on March 15, and the forecast durations are 5, 9, and 13 hours, respectively. The local times of (d)–(f) are 4:00, 8:00, and 12:00 on March 21, and the forecast durations are 7, 11, and 15 hours, respectively.

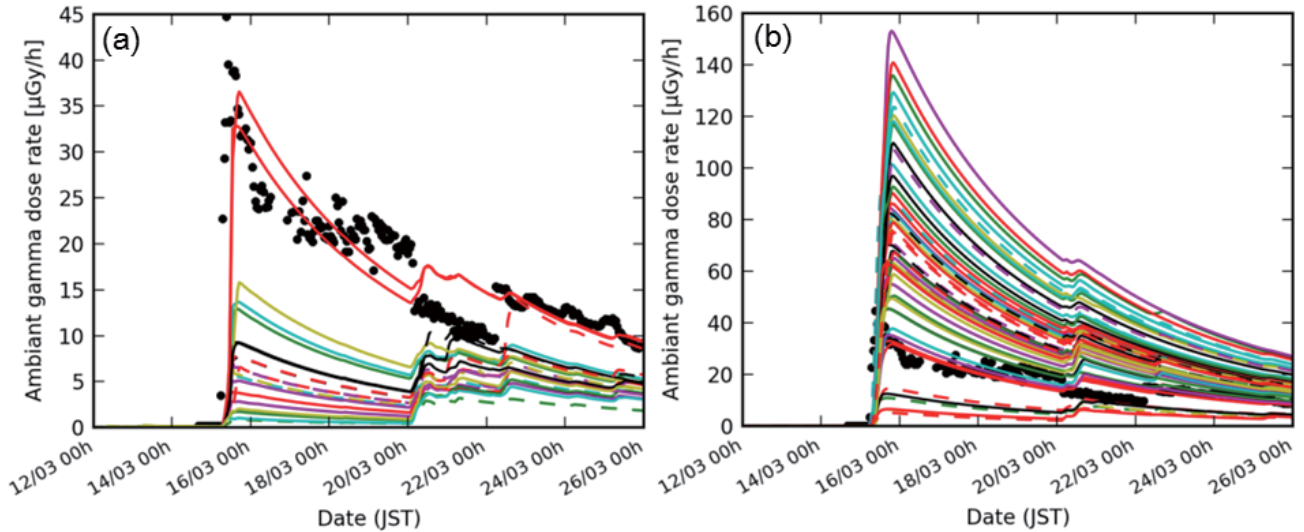


Fig. 7. Observed ambient gamma dose rates (dots) and ensemble members (lines) using the NHM-LETKF meteorological ensemble with the pX model: (a) source terms from Terada *et al.* (2012), and (b) source terms from Mathieu *et al.* (2012).

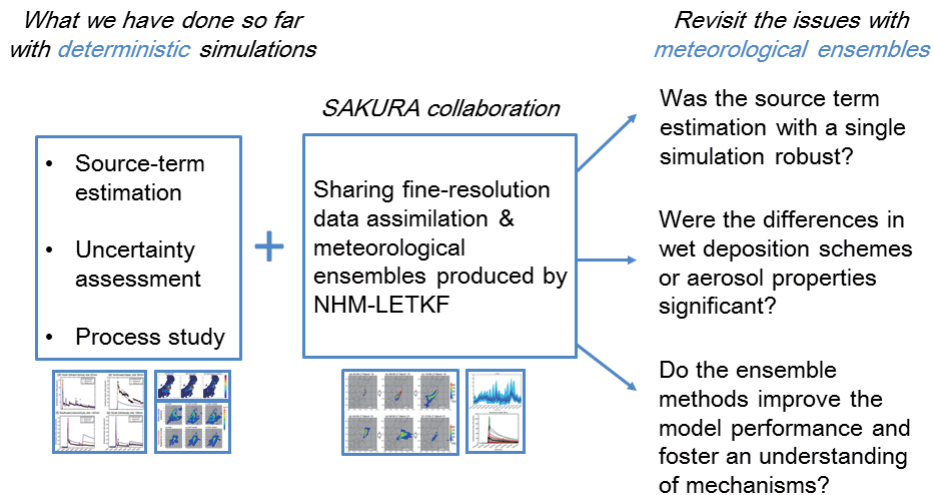


Fig. 8. Perspectives of the post-SAKURA program.

periods investigated by Adachi *et al.* (2013) and Kaneyasu *et al.* (2012). They did not measure hygroscopicity but, interestingly, the activity median aerodynamic diameters of the two periods are 1.8 and 1.0 μm , respectively, which are larger than those reported by Kaneyasu *et al.* (2012) (0.5–0.6 μm). Therefore, the assumption of 100% water-insoluble or water-soluble particles was not realistic; the actual distribution must lie somewhere in between. Even though the simulated amounts and patterns of the depositions are different for the different assumed aerosol properties, a quantitative assessment was not possible due to lack of findings pertaining to the aerosol microphysical properties of the source term.

APPLICATION OF METEOROLOGICAL ENSEMBLES

As previously discussed, ensemble modeling is a powerful tool for predicting the chaotic nature of meteorological simulations, and thus it can provide useful information for environmental modeling in various ways. Some of examples are presented in this subsection.

Figure 6 shows an example of the probabilistic forecast of the radioactive plumes predicted by NHM-LETKF-Chem (Sekiyama *et al.*, 2017b) for (a–c) March 15 and (d–f) 21, 2011, respectively, when the radioactive plume was transported from FDNPP toward the Tokyo Metropolitan Area. The deterministic (or single) run can only

predict the area above or below the threshold concentration (in this case 15 Bq/m^3 , denoted as the white contour lines), whereas the meteorological ensembles can provide the probability of the concentrations exceeding the threshold. This probabilistic forecast provides much more information than the deterministic forecast and is thus helpful for the policymakers and stakeholders in formulating evacuation plans for emergency situations.

Figure 7 presents the time series of the observations of the ambient gamma dose rate at Itate with the NHM-LETKF ensemble using (a) the source term from Terada *et al.* (2012) or (b) the source term from Mathieu *et al.* (2012) (Pérrillat *et al.*, 2016). This figure indicates the success of the ensemble simulations: each member of the ensemble can either overestimate or underestimate the observations, but the observations are well within the range of the ensemble (within the uncertainty of the meteorological simulations). This figure also shows the dependency of the results on the source term used. By combining the meteorological ensembles with inverse modeling, such as was done in Saunier *et al.* (2013), we can obtain a source-term estimation that includes meteorological uncertainties.

CONCLUSION AND FUTURE REMARKS

Modeling studies on the atmospheric diffusion and deposition of the radiocesium associated with the FDNPP accident is reviewed here, with a focus on a research collaboration between IRSN and MRI (the SAKURA program). A part of our research activities has also been reviewed in Mathieu *et al.* (2017). Mathieu *et al.* (2017) dealt with the general feature of radionuclides in atmosphere, whereas the current study focused on technical issues in atmospheric modeling. During this collaboration, we aimed to understand the atmospheric processes using atmospheric dispersion modeling and to quantify the uncertainties of the simulations using various numerical techniques, as summarized in Fig. 8. We had three topics: 1. Reverse/inverse modeling to quantify the source term; 2. Process modeling to better understand the wet deposition and aerosol dynamical processes; and 3. Ensemble simulations and data assimilation to quantify the accuracies and uncertainties of meteorological simulations. Our findings are summarized as follows, corresponding to each topic:

1. Inverse modeling is a useful tool for source-term estimation. Its accuracy highly depends on the quality of the observations (and their quantities), meteorological fields, and transport models.

2. As for the quality of the transport models, the wet deposition schemes and aerosol properties are assessed. Among the currently available deposition schemes, no ideal scheme was found due probably to uncertainty in

the boundary conditions such as meteorology and source terms. Deposition was found to be sensitive to the hygroscopicity of Cs-carrying aerosols but was also found to be uncertain due to our poor understanding of the aerosol properties of the source term.

3. The ensemble data assimilation is a powerful tool to provide meteorological analysis fields with the highest possible reliability at high temporal and spatial resolutions, together with meteorological ensemble members, which are usable for evaluating model uncertainty. The ensemble method is useful for inverse modeling, in that it allows for increased accuracy and better estimations of the uncertainty of the source term, as well as being useful for studies evaluating the significance and importance of atmospheric processes.

The current status of atmospheric modeling associated with the FDNPP accident case study and their near-future perspectives are summarized as follows:

1. There are two big streams of research that could instigate a paradigm shift in the study of the FDNPP accident in the near future: 1. Hourly observational data at a hundred stations (Tsuruta *et al.*, 2014; Oura *et al.*, 2015) are now available and have been shared with the simulation community (some simulations and comparisons have already been done; Saunier *et al.*, 2016; Nakajima *et al.*, 2017). 2. There are extensive studies investigating particles, including Cs-balls, in the regional environment, and since Adachi *et al.* (2013), new findings have been continuously published (Abe *et al.*, 2014; Yamaguchi *et al.*, 2016; Satou, 2016; Satou *et al.*, 2016; Higaki *et al.*, 2017). These will certainly foster the understanding of atmospheric processes through an improvement of the process modeling in the coming ten years.

2. We know the gross amounts of the emissions and depositions of radiocesium. However, the temporal variations of the source term are uncertain. These variations will become clearer as we have more accurate meteorological fields. The simulated depositions are highly variable among the models, but the reasons for these variations have not been identified. These additional variations will become clearer as further model intra-comparisons (i.e., sensitivity runs) and model inter-comparisons, with partially common configurations (e.g., different dispersion models but common domain and meteorology, as prescribed in the 2nd phase of SCJ's inter-comparison project) are completed. The 2nd SCJ project is ongoing, and Sekiyama *et al.* (2015) is used as a common meteorological field in this project.

3. The simulated deposition near FDNPP and to the northwest of the plant is substantially inaccurate. It is certainly difficult for Eulerian models to perform well due to the numerical diffusion at the point source, but the diffusion is readily resolved as the spatial resolution becomes higher. There are other critical reasons for the dif-

ficuity: the deposition occurred via light precipitation (<0.5 mm/h) and could include particles with diameters larger than 10 μm (e.g., Satou, 2016). Light precipitation is usually difficult for meteorological models to simulate, as their target has traditionally been heavy precipitation. When a particle is larger than 10 μm in diameter, the gravitational settling and below-cloud scavenging become significantly different from those assumed by all the previous models, i.e., particles of approximately 1 μm in diameter.

4. Studies on the mechanisms of radioactive aerosol formation will certainly improve the ability of atmospheric dispersion models. As the aerosol formation mechanisms are clearer, so are the aerosol properties. Relating to the items #1 and #3, a series of the Cs-ball studies will foster the understanding of its formation mechanisms and consequently improve the deposition modeling. There are also experimental studies specifically focusing on aerosol formation mechanisms (not Cs-ball formation for the cases): attachment of fission products to pre-existing aerosols (Takamiya *et al.*, 2016a, b) and formation of radioactive aerosols from an experimental reactor, Phébus (Haste *et al.*, 2013). Information on the aerosol properties could be useful for the decontamination, too.

5. The vertical distribution of radionuclides is unknown. Accuracy in the vertical distribution of radionuclides is critically important, especially for in-cloud scavenging. This accuracy is also important because the surface concentration depends on vertical diffusion. For example, even though the simulated surface concentration matches that observed, the amount of emissions in the simulation is underestimated when the simulated concentration in the upper atmosphere is underestimated. The vertical distribution cannot be revealed through measurements, because there are no remaining signatures of the earlier distributions. Still, there are a few available datasets, such as the radioactive soundings of Fukushima University, but these data have not been adequately used by the modeling community. A detailed study on the vertical distribution, using the available data, will be necessary.

6. An understanding of radioiodine will also be fostered. However, it is much more difficult to model than radiocesium because radioiodine was also partitioned into a gas phase, and even included inorganic and organic species in its gas phase. The environmental behaviors of the aerosol-phase, inorganic gaseous, and organic gaseous radioiodine are different from each other, but their partitioning in the atmosphere is difficult to model because our knowledge of their partitioning during their emission is limited.

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REFERENCES

- Abe, Y., Iizawa, Y., Terada, Y., Adachi, K., Igarashi, Y. and Nakai, I. (2014) Detection of uranium and chemical state analysis of individual radioactive microparticles emitted from the Fukushima nuclear accident using multiple synchrotron radiation X-ray analyses. *Anal. Chem.* **86**, 8521–8525.
- Achim, P., Monfort, M., Le Petit, G., Gross, P., Douysset, G., Taffary, T., Blanchard, X. and Moulin, C. (2014) Analysis of radionuclide releases from the Fukushima Dai-ichi Nuclear Power Plant Accident Part II. *Pure Appl. Geophys.* **171**, 645–667, doi:10.1007/s00024-012-0578-1.
- Adachi, K., Kajino, M., Zaizen, Y. and Igarashi, Y. (2013) Emission of spherical cesium-bearing particles from an early stage of the Fukushima nuclear accident. *Sci. Rep.* **3**, 2554, doi:10.1038/srep02554.
- Arnold, D., Maurer, C., Wotawa, G., Draxler, R., Saito, K. and Seibert, P. (2015) Influence of the meteorological input on the atmospheric transport modelling with FLEXPART of radionuclides from the Fukushima Daiichi nuclear accident. *J. Environ. Radioact.* **139**, 212–225, doi:10.1016/j.jenvrad.2014.02.013.
- Boutahar, J., Lacour, S., Mallet, V., Quélo, D., Roustan, Y. and Sportisse, B. (2004) Development and validation of a fully modular platform for numerical modeling of air pollution: POLAIR. *Int. J. Environ. Pollut.* **22**, 17–28.
- Byun, D. and Schere, K. L. (2006) Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality

- (CMAQ) modeling system. *Appl. Mechanics Rev.* **59**, 51–77.
- Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G. and Yamazawa, H. (2011) Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from the Fukushima Daiichi nuclear power plant into atmosphere. *J. Nucl. Sci. Technol.* **48**, 1129–1134.
- Deushi, M. and Shibata, K. (2011) Development of an MRI Chemistry–Climate Model ver.2 for the study of tropospheric and stratospheric chemistry. *Pap. Met. Geophys.* **62**, 1–46.
- Doi, T., Masumoto, K., Toyoda, A., Tanaka, A., Shibata, Y. and Hirose, K. (2013) Anthropogenic radionuclides in the atmosphere observed at Tsukuba: characteristics of the radionuclides derived from Fukushima. *J. Environ. Radioact.* **122**, 55–62.
- Draxler, R., Arnold, D., Chino, M., Galmarini, S., Hort, M., Jones, A., Leadbetter, S., Malo, A., Maurer, C., Rolph, G., Saito, K., Servranckx, R., Shimbori, T., Solazzo, E. and Wotawa, G. (2015) World Meteorological Organization’s model simulations of the radionuclide dispersion and deposition from the Fukushima Daiichi nuclear power plant accident. *J. Environ. Radioact.*, **139**, 172–184.
- Girard, S., Korsakissok, I. and Mallet, V. (2014) Screening sensitivity analysis of a radionuclides atmospheric dispersion model applied to the Fukushima disaster. *Atmos. Environ.* **95**, 490–500, doi:10.1016/j.atmosenv.2014.07.010.
- Girard, S., Mallet, V., Korsakissok, I. and Mathieu, A. (2016) Emulation and Sobol’ sensitivity analysis of an atmospheric dispersion model applied to the Fukushima nuclear accident. *J. Geophys. Res.* **121**, 3484–3496, doi:10.1002/2015JD023993.
- Groëll, J., Quélo, D. and Mathieu, A. (2014) Sensitivity analysis of the modelled deposition of ^{137}Cs on the Japanese land following the Fukushima accident. *Int. J. Environ. Pollut.* **55**, 67–75.
- Haste, T., Payot, F. and Bottomley, P. D. W. (2013) Transport and deposition in the Phébus FP circuit. *Annals of Nuclear Energy* **61**, 102–121.
- Higaki, S., Kurihara, Y., Yoshida, H., Takahashi, Y. and Shinohara, N. (2017) Discovery of non-spherical heterogeneous radiocesium-bearing particles not derived from Unit 1 of the Fukushima Dai-ichi Nuclear Power Plant, in residences five years after the accident. *J. Environ. Radioact.* **177**, 65–70.
- Hirao, S., Yamazawa, H. and Nagae, T. (2013) Estimation of release rate of iodine-131 and cesium-137 from the Fukushima Daiichi nuclear power plant. *J. Nucl. Sci. Technol.* **50**, 139–147, doi:10.1080/00223131.2013.757454.
- IAEA (ed.) (2015) *The Fukushima Daiichi Accident. Volume 4/5 Radiological Consequences*. International Atomic Energy Agency, Vienna.
- Kajino, M. (2015) NHM-Chem: Sensitivity of Cs deposition to the size and hygroscopicity of Cs-bearing aerosols. *Technical Report of the Meteorological Research Institute* **76**, 133–142.
- Kajino, M., Inomata, Y., Sato, K., Ueda, H., Han, Z., An, J., Katata, G., Deushi, M., Oshima, N., Kurokawa, J., Ohara, T., Takami, A. and Hatakeyama, S. (2012) Development of the RAQM2 aerosol chemical transport model and predictions of the Northeast Asian aerosol mass, size, chemistry, and mixing type. *Atmos. Chem. Phys.* **12**, 11833–11856.
- Kajino, M., Ishizuka, M., Igarashi, Y., Kita, K., Yoshikawa, C. and Inatsu, M. (2016) Long-term assessment of airborne radiocesium after the Fukushima nuclear accident: re-suspension from bare soil and forest ecosystems. *Atmos. Chem. Phys.* **16**, 13149–13172.
- Kaneyasu, N., Ohashi, H., Suzuki, F., Okada, T. and Ikemori, F. (2012) Sulfate aerosol as a potential transport medium of radiocesium from the Fukushima nuclear accident. *Environ. Sci. Technol.* **46**, 5720–5726.
- Katata, G., Terada, H., Nagai, H. and Chino, M. (2012a) Numerical reconstruction of high dose rate zones due to the Fukushima Daiichi Nuclear Power Plant accident. *J. Environ. Radioact.* **111**, 2–12.
- Katata, G., Ota, M., Terada, H., Chino, M. and Nagai, H. (2012b) Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power Plant accident, Part I: Source term estimation and local-scale atmospheric dispersion in early phase of the accident. *J. Environ. Radioact.* **109**, 103–113.
- Katata, G., Chino, M., Kobayashi, T., Terada, H., Ota, M., Nagai, H., Kajino, M., Draxler, R., Hort, M. C., Malo, A., Torii, T. and Sanada, Y. (2015) Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of an atmospheric dispersion model with an improved deposition scheme and oceanic dispersion model. *Atmos. Chem. Phys.* **15**, 1029–1070, doi:10.5194/acp-15-1029-2015.
- Korsakissok, I., Mathieu, A. and Didier, D. (2013) Atmospheric dispersion and ground deposition induced by the Fukushima Nuclear Power Plant accident: A local-scale simulation and sensitivity study. *Atmos. Environ.* **70**, 267–279.
- Kristiansen, N. I., Stohl, A., Olivie, D. J. L., Croft, B., Sovde, O. A., Klein, H., Christoudias, T., Kunkel, D., Leadbetter, S. J., Lee, Y. H., Zhang, K., Tsigaridis, K., Bergman, T., Evangeliou, N., Wang, H., Ma, P.-L., Easter, R. C., Rasch, P. J., Liu, X., Pitari, G., Di Genova, G., Zhao, S. Y., Balkanski, Y., Bauer, S. E., Faluvegi, G. S., Kokkola, H., Martin, R. V., Pierce, J. R., Schulz, M., Shindell, D., Tost, H. and Zhang, H. (2016) Evaluation of observed and modelled aerosol lifetimes using radioactive tracers of opportunity and an ensemble of 19 global models. *Atmos. Chem. Phys.*, **16**, 3525–3561, doi:10.5194/acp-16-3525-2016.
- Kunii, M. (2014) Mesoscale data assimilation for a local severe rainfall event with the NHM-LETKF system. *Wea. Forecasting* **29**, 1093–1105, doi:10.1175/WAF-D-13-00032.1.
- Leadbetter, S. J., Hort, M. C., Jones, A. R., Webster, H. N. and Draxler, R. R. (2015) Sensitivity of the modelled deposition of Caesium-137 from the Fukushima Dai-ichi nuclear power plant to the wet deposition parameterization in NAME. *J. Environ. Radioact.* **139**, 200–211, doi:10.1016/j.jenvrad.2014.03.018.
- Lim, K.-S. and Hong, S.-Y. (2010) Development of an effective double-moment cloud microphysics scheme with prognostic cloud condensation nuclei (CCN) for weather and climate models. *Mon. Wea. Rev.* **138**, 1587–1612.
- Liu, Y., Haussaire, J.-M., Bocquet, M., Roustan, Y., Saunier,

- O. and Mathieu, A. (2017) Uncertainty quantification of pollutant source retrieval: comparison of Bayesian methods with application to the Chernobyl and Fukushima Daiichi accidental releases of radionuclides. *Q. J. R. Meteorol. Soc.* **143**, 2886–2901.
- Maki, T. (2015) Emission source estimation by an inverse model. *Technical Report of the Meteorological Research Institute* **76**, 150–153.
- Mathieu, A., Korsakissok, I., Quélo, D., Groëll, J., Tombette, M., Didier, D., Quentric, E., Saunier, O., Benoit, J.-P. and Isnard, O. (2012) Atmospheric dispersion and deposition of radionuclides from the Fukushima Daiichi nuclear power plant accident. *Elements* **8**, 195–200.
- Mathieu, A., Kajino, M., Korsakissok, I., Périllat, R., Quélo, D., Quérel, A., Saunier, O., Sekiyama, T. T., Adachi, K., Igarashi, Y. and Didier, D. (2017) Fukushima Daiichi-derived radionuclides in the atmosphere, transport and deposition in Japan: a review. *Appl. Geochem.* (in review).
- Morino, M., Ohara, T. and Nishizawa, M. (2011) Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi Nuclear Power Plant in March 2011. *Geophys. Res. Lett.* **38**, 7 pp., doi:10.1029/2011GL048689.
- Morino, Y., Ohara, T., Watanabe, S., Hayashi, S. and Nishizawa, M. (2013) Episode analysis of deposition of radiocesium from the Fukushima Daiichi Nuclear Power Plant accident. *Environ. Sci. Technol.* **47**, 2314–2322.
- Morrison, H., Thompson, G. and Tatarskii, V. (2009) Impact of cloud microphysics on the development of trailing stratiform precipitation in a simulated squall line: comparison of one- and two-moment schemes. *Mon. Wea. Rev.* **137**, 991–1007.
- Nakajima, T., Misawa, S., Morino, Y., Tsuruta, H., Goto, D., Uchida, J., Takemura, T., Ohara, T., Oura, T., Ebihara, M. and Satoh, M. (2017) Model depiction of the atmospheric flows of radioactive cesium emitted from the Fukushima Daiichi Nuclear Power Station accident. *Prog. Earth Planet. Sci.* **4**(2), doi:10.1186/s40645-017-0117-x.
- NRA (Nuclear Regulation Authority) (2012) Airborne Monitoring Results in each prefecture, available at: <http://radioactivity.nsr.go.jp/en/list/203/list-1.html> (last access: 26 May 2017).
- Oura, Y., Ebihara, M., Tsuruta, H., Nakajima, T., Ohara, T., Ishimoto, M., Sawahata, H., Katsumura, Y. and Nitta, W. (2015) A database of hourly atmospheric concentration of radiocesium (^{134}Cs and ^{137}Cs) in suspended particulate matter collected in March 2011 at 99 air pollution monitoring stations in Eastern Japan. *J. Nucl. Radiochem. Sci.* **15**(2), 15–26.
- Périllat, R., Korsakissok, I., Mallet, V., Mathieu, A., Sekiyama, T., Kajino, M., Adachi, K., Igarashi, Y., Maki, T. and Didier, D. (2016) Using meteorological ensembles for atmospheric dispersion modeling of the Fukushima nuclear accident. *Proc. International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes (HARMO)* 17th, 5 pp.
- Quérel, A., Lemaitre, P., Monier, M., Porcheron, E., Flossmann, A. I. and Hervo, M. (2014) An experiment to measure rain-drop collection efficiencies: influence of rear capture. *Atmos. Meas. Technol.* **7**, 1321–1330.
- Quérel, A., Roustan, Y., Quélo, D. and Benoit, J.-P. (2015) Hints to discriminate the choice of wet deposition models applied to an accidental radioactive release. *Int. J. Environ. Pollut.* **58**, 268–279.
- Quérel, A., Quélo, D., Roustan, Y., Mathieu, A., Kajino, M., Sekiyama, T., Adachi, K., Didier, D., Igarashi, Y. and Maki, T. (2016) Impact of changing the wet deposition schemes in IdX on Cs-137 atmospheric deposits after the Fukushima accident. *Proc. International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes (HARMO)* 17th, 5 pp.
- Saito, K., Fujita, T., Yamada, Y., Ishida, J., Kumagai, Y., Aranami, K., Ohmori, S., Nagasawa, R., Kumagai, S., Muroi, C., Kato, T., Eito, H. and Yamazaki, Y. (2006) The operational JMA nonhydrostatic mesoscale model. *Mon. Wea. Rev.* **134**, 1266–1298, doi:10.1175/MWR3120.1.
- Saito, K., Shimbori, T. and Draxler, R. (2015) JMA's regional atmospheric transport model calculations for the WMO technical task team on meteorological analyses for Fukushima Daiichi Nuclear Power Plant accident. *J. Environ. Radioact.* **139**, 185–199, doi:10.1016/j.jenvrad.2014.02.007.
- Satou, Y. (2016) Study of relationship between deposition of radioactive materials and radioactive particles in the difficult-to-return zone caused by the Fukushima Dai-ichi Nuclear Power Plant accident. Dr. Sci. Thesis, University of Tsukuba, 128 pp.
- Satou, Y., Sueki, K., Sasa, K., Adachi, K. and Igarashi, Y. (2016) First successful isolation of radioactive particles from soil near the Fukushima Daiichi Nuclear Power Plant. *Anthropocene* **14**, 71–76.
- Saunier, O., Mathieu, A., Didier, D., Tombette, M., Quélo, D., Winiarek, V. and Bocquet, M. (2013) An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations. *Atmos. Chem. Phys.* **13**, 11403–11421, doi:10.5194/acp-13-11403-2013.
- Saunier, O., Mathieu, A., Sekiyama, T., Kajino, M., Adachi, K., Bocquet, M., Igarashi, Y., Maki, T. and Damien, D. (2016) A new perspective on the Fukushima released brought by newly available ^{137}Cs air concentration observations and reliable meteorological fields. *Proc. International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes (HARMO)* 17th, 7 pp.
- SCJ (Science Council of Japan) (2014) A review of the model comparison of transportation and deposition of radioactive materials released to the environment as a result of the Tokyo Electric Power Company's Fukushima Daiichi Nuclear Power Plant accident, available at <http://www.scj.go.jp/ja/info/kohyo/pdf/kohyo-22-h140902-e1.pdf>, 36 pp. (last access: 26 May 2017).
- Sekiyama, T. T., Kunii, M., Kajino, M. and Shimbori, T. (2015) Horizontal resolution dependence of atmospheric simulations of the Fukushima nuclear accident using 15 km, 3 km, and 500 m grid models. *J. Meteor. Soc. Japan* **93**, 49–64.
- Sekiyama, T. T., Kajino, M. and Kunii, M. (2017a) The impact of surface wind data assimilation on the predictability of near-surface plume advection in the case of the Fukushima

- nuclear accident. *J. Meteor. Soc. Japan* **95**, 447–454, doi:10.2151/jmsj.2017-025.
- Sekiyama, T. T., Kajino, M. and Kunii, M. (2017b) Ensemble dispersion simulation of a point-source radioactive aerosol using a square root Kalman filter. *J. Geophys. Res.* (in review).
- Shimbori, T., Aikawa, Y. and Seino, N. (2009) Operational implementation of the tephra fall forecast with the JMA mesoscale tracer transport model. *CAS/JSC WGNE Res. Act. Atmos. Ocean. Model.* **39**, 5.29–5.30.
- Shimbori, T., Aikawa, Y., Fukui, K., Hashimoto, A., Seino, N. and Yamasato, H. (2010) Quantitative tephra fall prediction with the JMA mesoscale tracer transport model for volcanic ash: A case study of the eruption at Asama volcano in 2009. *Pap. Met. Geophys.* **61**, 13–29 (in Japanese with English abstract).
- Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang, W. and Powers, J. G. (2008) A description of the advanced research WRF version 3. *Tech. Note, NCAR/TN~475+STR, Natl. Cent. for Atmos. Res.*, Boulder, Colo., 125 pp.
- Soulhac, L. and Didier, D. (2008) Project pX, note de principe pX 1.0, Note technique IRSN/DEI/SESUC/08-39.
- Steinhauser, G., Niisoe, T., Harada, K. H., Shozugawa, K., Schneider, S., Synal, H.-A., Walther, C., Christl, M., Nanba, K., Ishikawa, H. and Koizumi, A. (2015) Post-Accident sporadic releases of airborne radionuclides from the Fukushima Daiichi nuclear power plant site. *Environ. Sci. Technol.* **49**, 14028–14035, doi:10.1021/acs.est.5b03155.
- Stohl, A., Seibert, P., Arnold, D., Burkhardt, J. F., Eckhardt, S., Tapia, C., Vargass, A. and Yasunari, T. J. (2012) 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**, 2313–2343, doi:10.5194/acp-12-2313-2012.
- Takamiya, K., Tanaka, T., Nitta, S., Itosu, S., Sekimoto, S., Oki, Y. and Ohtsuki, T. (2016a) Attachment behavior of fission products to solution aerosol. *J. Radiat. Prot. Res.* **41**, 350–353, doi:10.14407/jrpr.2016.41.4.350.
- Takamiya, K., Tanaka, T., Nitta, S., Itosu, S., Sekimoto, S., Oki, Y. and Ohtsuki, T. (2016b) Observation of attachment ratio of fission products on solution aerosol. *J. Radioanal. Nucl. Chem.* **307**, 2227–2230, doi:10.1007/s10967-015-4574-7.
- Tanaka, T. Y., Orito, K., Sekiyama, T. T., Shibata, K., Chiba, M. and Tanaka, H. (2003) MASINGAR, a global tropospheric aerosol chemical transport model coupled with MRI/JMA98 GCM: Model description. *Pap. Met. Geophys.* **53**, 119–138, doi:10.2467/mripapers.53.119.
- Terada, H., Katata, G., Chino, M. and Nagai, H. (2012) 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**, 141–154.
- Tinsley, B. A., Zhou, L. and Plemmons, A. (2006) Changes in scavenging of particles by droplets due to weak electrification in clouds. *Atmos. Res.* **79**, 266–295.
- Tsuruta, H., Oura, Y., Ebihara, M., Ohara, T. and Nakajima, T. (2014) First retrieval of hourly atmospheric radionuclides just after the Fukushima accident by analyzing filter-tapes of operational air pollution monitoring stations. *Sci. Rep.* **4**, 6717, doi:10.1038/srep06717.
- Wang, Z., Xie, F., Sakura, T., Ueda, H., Han, Z., Carmichael, G. R., Streets, D., Engardt, M., Holloway, T., Hayami, H., Kajino, M., Thongboonchoo, N., Bennet, C., Park, S. U., Fung, C., Chang, A., Sartelet, K. and Amann, M. (2008) MICS-Asia II: Model inter-comparison and evaluation of acid deposition. *Atmos. Environ.* **42**, 3528–3542.
- WHO (World Health Organization) (2001) *IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Ionizing Radiation, Part 2: Some Internally Deposited Radionuclides*, Vol. 78, IARC Press, 617 pp.
- Winiarek, V., Bocquet, M., Saunier, O. and Mathieu, A. (2012) Estimation of errors in the inverse modeling of accidental release of atmospheric pollutant: Application to the reconstruction of the cesium-137 and iodine-131 source terms from the Fukushima Daiichi power plant. *J. Geophys. Res.* **117**, D05122, doi:10.1029/2011JD016932.
- Winiarek, V., Bocquet, M., Duhanyan, N., Roustan, Y., Saunier, O. and Mathieu, A. (2014) Estimation of the caesium-137 source term from the Fukushima Daiichi nuclear power plant using a consistent joint assimilation of air concentration and deposition observations. *Atmos. Environ.* **82**, 268–279.
- Yamaguchi, N., Mitome, M., Akiyama-Hasegawa, K., Asano, M., Adachi, K. and Kogure, T. (2016) Internal structure of cesium-bearing radioactive microparticles released from Fukushima nuclear power plant. *Sci. Rep.* **6**, 20548, doi:10.1038/srep20548.
- Yasunari, T. J., Stohl, A., Hayano, R. S., Burkhardt, J. F., Eckhardt, S. and Yasunari, T. (2011) Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. *Proc. Natl. Acad. Sci. USA* **108**, 19530–19534.
- Yumimoto, K., Morino, Y., Ohara, T., Oura, Y., Ebihara, M., Tsuruta, H. and Nakajima, T. (2016) Inverse modeling of the ¹³⁷Cs source term of the Fukushima Dai-ichi Nuclear Power Plant accident constrained by a deposition map monitored by aircraft. *J. Environ. Radioact.* **164**, 1–12.