

Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011

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Short title: Radioactive materials from Fukushima

Submitted to GRL on June 27. Last revised on August 2

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Keywords: radioactive materials, chemical transport model, Fukushima nuclear accident

Abstract

To understand the atmospheric behavior of radioactive materials emitted from the Fukushima Daiichi nuclear power plant after the nuclear accident that accompanied the great Tohoku earthquake and tsunami on 11 March 2011, we simulated the transport and deposition of iodine-131 and cesium-137 using a chemical transport model. The model roughly reproduced the observed temporal and spatial variations of deposition rates over 15 Japanese prefectures (60–400 km from the plant), including Tokyo, although there were some discrepancies between the simulated and observed rates. These discrepancies were likely due to uncertainties in the simulation of emission,

transport, and deposition processes in the model. A budget analysis indicated that approximately 13% of iodine-131 and 22% of cesium-137 were deposited over land in Japan, and the rest was deposited over the ocean or transported out of the model domain ($700 \times 700 \text{ km}^2$). Radioactivity budgets are sensitive to temporal emission patterns. Accurate estimation of emissions to the air is important for estimation of the atmospheric behavior of radionuclides and their subsequent behavior in land water, soil, vegetation, and the ocean.

1. Introduction

A nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP) accompanied the great Tohoku earthquake and tsunami on 11 March 2011, and as a result, enormous amounts of radionuclides were emitted into the atmosphere and the ocean [Chino *et al.*, 2011; Butler, 2011; Nuclear Safety Commission of Japan, 2011]. Radionuclides, particularly iodine-131 (I-131) and cesium-137 (Cs-137), adversely affect human health through contamination of air, water, soil, and food [Anspaugh *et al.*, 1988]. Because radioactive contamination of soil and land water is caused mostly by atmospheric deposition, understanding the spatial and temporal distributions of radioactive materials in the atmosphere and their deposition over land masses and oceans is important.

Numerical simulations have played an important role in furthering the understanding of spatiotemporal variations of radioactive materials in the atmosphere. For example, many numerical simulations were conducted after the Chernobyl nuclear accident in 1986 [Albergel *et al.*, 1988; Hass *et al.*, 1990; Wheeler, 1988], and these simulations helped to clarify the atmospheric behavior of the radioactive materials even though

there were large uncertainties in the modeling parameters (see, e.g., [Sportisse, 2007]).

In Japan, the System for Prediction of Environmental Emergency Dose Information (SPEEDI) and the worldwide version of the system (WSPEEDI) were developed by the Japan Atomic Energy Agency to predict environmental doses from radioactive materials accidentally released from a nuclear plant [Imai *et al.*, 1985; Terada and Chino, 2008]. After the FDNPP accident, SPEEDI data were released to the public starting on 23 March 2011, and the data helped the public to understand the behavior of radioactive materials. The target areas of SPEEDI are about $25 \times 25 \text{ km}^2$ and $100 \times 100 \text{ km}^2$ around the FDNPP and does not include the Tokyo metropolitan area (TMA, 120–270 km south-southwest of the FDNPP), which is one of the world's largest megacities (population, 30 million). In some areas, the radioactivity due to I-131 and Cs-137 in water and food (e.g., vegetables and milk) exceeded the provisional standard even in the TMA in March 2011 [e.g., *Ministry of Health, Labour and Welfare*, 2011a; 2011b]. Thus, the development of a simulation model for evaluating the spatiotemporal variations of radioactive materials and the factors that control that variation on a scale that covers both the FDNPP and TMA (100–300 km) is necessary.

In this study, we simulated the transport and deposition of I-131 and Cs-137 using a chemical transport model. First, we compared the simulated atmospheric deposition rates and activity concentrations of I-131 and Cs-137 with observed data, and we evaluated the model performance to reproduce the observed data. Then, we assessed the budget of radioactive materials emitted from the FDNPP. Because there are uncertainties in the model input parameters, we evaluated uncertainties in the model simulation by means of sensitivity analyses.

2. Methodology

We simulated distributions of I-131 and Cs-137 using a three-dimensional chemical transport model, Models-3 Community Multiscale Air Quality (CMAQ) [Byun and Ching, 1999; Byun and Schere, 2006], for 10–30 March 2011. In this simulation, we calculated horizontal and vertical advection, horizontal and vertical diffusion, emission, dry and wet deposition, and radioactive decay. Chemical and aerosol processes were not calculated because they are not well understood for radioactive materials [Sportisse, 2007]. Deposition schemes used in CMAQ are detailed in Byun and Ching [1999] and Byun and Schere [2006]. The dry deposition was simulated using a resistance model. The cloud module of CMAQ includes parameterizations for sub-grid convective precipitating and non-precipitating clouds and grid-scale resolved clouds. We assumed that the gaseous fraction of I-131 was 80% and that all the Cs-137 was in the particulate phase [Sportisse, 2007]. Although there are large variabilities in the diameter of particulate radionuclides, we assumed the diameter of particulate matter to be 1 μm for both I-131 and Cs-137 [Sportisse, 2007; Sparmacher et al., 1993]. The deposition rate for gaseous I-131 was assumed to be the same as that for sulfur dioxide, as in previous studies (e.g., [Baklanov and Sorensen, 2001; Sportisse, 2007]). The radioactive decay of I-131 (half-life 8.02 days) was included in the simulation, whereas the decay of Cs-137 (half-life 30.2 years) was neglected. The model domain covered most of the Tohoku region (including the prefectures labeled 1 – 5 in Figure 1) and the Kanto region (prefectures 6–12, Figure 1) ($700 \times 700 \text{ km}^2$) at a 6-km grid resolution and a 34-layer vertical structure with a surface layer thickness of about 60 m (Figure 1).

We calculated meteorological fields by using the Weather Forecast and Research Model version 3.1 [Skamarock et al., 2008]. Analysis nudging was conducted with the

three-dimensional meteorological fields from the Japan Meteorological Agency Meso-Scale Model datasets available with $5 \times 5 \text{ km}^2$ horizontal resolution for 3-h intervals. Emission data from the FDNPP were taken from *Chino et al.* [2011] (Figure S1). These data have a variable interval of 6–61 h (31 h on average), and we assumed that emission rates were constant in each interval. We assumed that both I-131 and Cs-137 were emitted in the bottom layer of the model. Uncertainties originating from the vertical and temporal emission profiles are evaluated in Section 3.2. Initial and boundary conditions for radioactive materials were set to zero.

Daily deposition rates of I-131 and Cs-137 were monitored over 46 Japanese prefectures starting on 18 March 2011, with bulk samplers [*Ministry of Education, Culture, Sports, Science, and Technology*, 2011]. Each prefecture in Japan has one monitoring station. However, measurement in Miyagi Prefecture (prefecture 4 in Figure 1) was not conducted, owing to damage to an instrument; and measurement at a site in Fukushima Prefecture (5) did not start until 27 March. Bulk samplers were used to collect materials from dry deposition in addition to those from wet deposition, even though the collection efficiency of bulk samplers for trace gases and submicron particles is substantially different from that of a natural landscape [*Aikawa et al.*, 2003; *Staelens et al.*, 2005]. We assumed that deposition rates measured with bulk samplers were between the wet deposition and total deposition (i.e., dry plus wet) rates [*Staelens et al.*, 2005].

In addition, atmospheric activity concentrations of particulate radionuclides were continuously measured at a Tsukuba site (labeled “T” in Figure 1) starting on 15 March [*High Energy Accelerator Research Organization*, 2011]. A high-volume sampler was

used for bulk aerosol sampling. The duration of the sampling periods ranged from 3 to 48 h.

3. Results and Discussion

3.1. Comparison between observed and simulated data

We compared the simulated deposition rates of I-131 and Cs-137 with the observed rates over the monitoring sites in March 2011 (Figures 2 and S2). For this comparison, we selected monitoring sites in 15 of the Japanese prefectures shown in Figure 1. The observed deposition rates of I-131 and Cs-137 were lower than $0.05 \text{ kBq m}^{-2} \text{ day}^{-1}$ over the other 31 prefectures in Japan from March to May 2011. In addition, observed deposition rates of I-131 and Cs-137 were lower than $1 \text{ kBq m}^{-2} \text{ day}^{-1}$ over all the monitoring sites after 31 March 2011. These rates suggest that most of the radioactive materials that were deposited over Japan from March to May 2011 were covered in the target period and area of this study.

Simulated deposition rates of I-131 and Cs-137 increased in Fukushima and the adjacent prefectures on 15–17 and 20–26 March (Figure 2). During the other periods, the deposition rates of I-131 and Cs-137 were low over the most of the observational stations. The simulated rates roughly agreed with the observed rates, although the simulated deposition peak on 15–17 March could not be verified, because of the lack of observation data. The atmospheric activity concentrations of particulate I-131 and Cs-137 measured in Tsukuba behaved similarly to the deposition rates (Figure S3). Note that the model greatly underestimated the observed peak of Cs-137 activity concentration on 21–22 March. Similar behavior was found in deposition rates at a monitoring site in Ibaraki Prefecture (6), in which the Tsukuba site is located (Figure 2).

By contrast, deposition rates of Cs-137 at the Tochigi (7), Gunma (8), and Saitama (9) sites were overestimated by the model. These discrepancies between the observed and simulated data may have been partly due to uncertainties in transport and deposition processes in addition to uncertainties in emissions. Recently, *Kondo et al.* [2011] showed that CMAQ overestimates below-cloud scavenging of aerosols. However, the observed peak of Cs-137 was greatly underestimated even in the sensitivity simulation with wet deposition off (**Figure S3**), and thus, treatment of wet deposition processes in CMAQ was not the only reason for the discrepancy in the atmospheric Cs-137 activity concentration at the Tsukuba site. Overall, despite some discrepancies, both the simulated total rates and the wet deposition rates mostly agreed with the observed deposition rates of I-131 and Cs-137 over the 14 prefectures within one order of magnitude (**Figure 3**). The mean ratios of simulated wet (total) depositions of I-131 and Cs-137 to observed deposition rates were 1.2 (4.0) and 1.8 (2.6), respectively. Although the model performance cannot be quantitatively evaluated because of the unknown collection efficiency of dry-deposited materials by bulk samplers, this agreement suggests that the spatial variations were roughly reproduced by the model. The model performance in this study is comparable to the model performance for the Chernobyl simulations, although the temporal and spatial scales of this simulation and the Chernobyl simulations are different (see, e.g., [*Albergel et al.*, 1988; *Brandt et al.*, 2002; *Davoine and Bocquet*, 2007; *Hass et al.*, 1990]).

During 15–17 and 21–23 March, when deposition rates increased over the areas around Fukushima, northeasterly, easterly, or southeasterly winds associated with a transient cyclone transported radioactive materials from the FDNPP to inland areas (**Figures S4 and S5**). In addition, precipitation was observed on 15–17 and 21–23 March,

when the transient cyclone passed over Japan (Figure 2), and thus, radioactive materials were effectively deposited over the land by wet processes. In contrast, on 17–20 March when an anticyclone dominated over Fukushima, westerly or northwesterly winds prevailed, and radioactive materials were transported predominantly to the Pacific Ocean.

Rates of I-131 deposition were highest in Fukushima Prefecture, followed by Ibaraki Prefecture and other prefectures in the Kanto area. Cs-137 deposition rates were also highest in Fukushima, followed by Miyagi Prefecture. Areas of high deposition of both I-131 and Cs-137 extended from Fukushima Prefecture in northwesterly and southerly directions (Figure 4). This pattern reflected the wind and precipitation patterns during the period of high emissions.

3.2. Budget of radioactive materials

We assessed the I-131 and Cs-137 budgets in the model domain by quantifying the contributions of individual processes, such as advection, diffusion, emission, and deposition, to the atmospheric activity concentrations using Process Analysis, an analytical tool deployed in the CMAQ model [Byun and Schere, 2006] (Figure 5). In the model domain, the predominant loss processes for I-131 were dry and wet deposition on 15 March and advection after 16 March. Outflow of I-131 from the model domain occurred predominantly through the eastern boundary, although outflow from the northern and southern boundaries also occurred. Wet deposition of Cs-137 made large contributions on 15–16 and 20–21 March, and advection dominated the loss processes during the other periods. On average, 13% of I-131 and 22% of Cs-137 emitted from the FDNPP were deposited over the land of Japan in March 2011 (Table 1). About 19% of

I-131 and 10% of Cs-137 were deposited over the ocean in the model domain, and 55–60% of I-131 and Cs-137 emitted from the FDNPP were transported out of the model domain. The deposition amounts in each prefecture are given in [Table S1](#). About 8% of I-131 and 15% of Cs-137 emitted from the FDNPP were deposited over Fukushima Prefecture.

These fractions of budgets do not change even if the total emission amount changes, whereas the fractions would change if the temporal and vertical emission patterns changed. We conducted sensitivity simulations to evaluate the uncertainties of these fractions originating from the vertical and temporal emission profiles ([Table S2 and S3](#)). We found that the simulated fractions of I-131 and Cs-137 deposited over land in the model domain were insensitive to the vertical emission profiles. Temporal profiles of the emission rates had a large impact on the budget of radioactive materials. On 15–17 and 19–23 March, when the transient cyclone passed over Japan, 7–24% of I-131 and 8–41% of Cs-137 emitted from the FDNPP were deposited over land in the model domain, whereas the deposition amounts over land were much smaller when westerly winds prevailed. Thus, radionuclide budget estimated in this study changed substantially when the temporal emission profiles changed. Therefore, accurate estimation of emission amounts and temporal variations is important for the evaluation of the atmospheric behavior of radionuclides.

4. Conclusions

We simulated the spatial and temporal variations of I-131 and Cs-137 around the FDNPP ($700 \times 700 \text{ km}^2$) during 11–30 March, 2011, by using a chemical transport model (CMAQ). The model roughly reproduced the observed spatiotemporal variations

of deposition rates over 15 prefectures in Japan (60–400 km from the FDNPP), although there were some discrepancies between the simulated and observed data, most likely due to uncertainties in the treatment of emission, transport, and deposition processes in the model.

Budget analysis indicated that approximately 13% of I-131 and 22% of Cs-137 were deposited over land in Japan, and the rest was deposited to the ocean or transported out of the model domain. Most of the radioactive materials emitted from the FDNPP were deposited or transported out of the model region within a few days and did not stay in the atmosphere in the model domain.

Considering that the current emission data is from the preliminary assessment and the uncertainty is still large [*Chino et al.* 2011], the accuracy of the model simulation will be improved when more-accurate emission data become available. For example, the inverse modeling [e.g., *Davoine and Bocquet*, 2007] would help improve the accuracy of the emission estimate. Even though there are still large uncertainties in this simulation, agreement between the observed and simulated results indicates the validity of the model, and the budget obtained from this study will be an important tool for understanding the behavior of radionuclides emitted from the FDNPP. It is necessary to understand the behaviors of radioactive materials in land water, soil, vegetation, and ocean. This model results will be an important input to multimedia models, which deals with running water, soils, and oceans around Japan.

Acknowledgments

We thank the staffs of the Ministry of Education, Culture, Sports, Science, and Technology, the High Energy Accelerator Research Organization, and the National

Institute for Environmental Studies (NIES) for carrying out measurements and providing observation datasets. We also thank the staff of the Environmental Information Department of NIES and K. Tanaka for providing computer equipment soon after the earthquakes. We acknowledge R. Ichimiya for providing digitized database of monitoring data on his Web site.

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Figure captions

Figure 1. Model domain used in the CMAQ simulation. Numbered prefectures: 1, Iwate; 2, Akita; 3, Yamagata; 4, Miyagi; 5, Fukushima; 6, Ibaraki; 7, Tochigi; 8, Gunma; 9, Chiba; 10, Saitama; 11, Tokyo; 12, Kanagawa; 13, Shizuoka; 14, Yamanashi; 15, Nagano; 16, Niigata. The white square indicates the site of the Fukushima Daiichi nuclear power plant (FDNPP); the white “T” indicates the Tsukuba site.

Figure 2. Observed and simulated deposition rates of I-131 (left) and Cs-137 (center) and precipitation rates (right) at selected measurement sites shown in [Figure 1](#). Simulated dry and wet deposition rates averaged over each prefecture are indicated by green and blue hatching, respectively.

Figure 3. Observed and simulated deposition rates of I-131 (left) and Cs-137 (right) at measurement sites shown in [Figure 1](#) averaged over the period from 18 to 30 March (i.e., N=13 for each measurement site).

Figure 4. Distributions of average activity concentrations and accumulated deposition rates of I-131 and Cs-137 simulated by CMAQ over 11–29 March 2011. Distribution of accumulated precipitation is also shown.

Figure 5. Budget analysis of I-131 and Cs-137 in the model domain.

Table 1. Simulated Budget of I-131 and Cs-137 (Bq), 11–30 March 2011.

	I-131	Contr. ^a	Cs-137	Contr. ^a
Emission	1.42×10^{17}	100.0%	9.94×10^{15}	100.0%
Advection + diffusion	-8.93×10^{16}	-62.7%	-5.41×10^{15}	-54.5%
Dry deposition	-3.55×10^{16}	-24.9%	-1.41×10^{14}	-1.4%
Wet deposition	-9.73×10^{15}	-6.8%	-3.04×10^{15}	-30.6%
<i>Dry deposition</i>				
land	-1.28×10^{16}	-9.0%	-7.36×10^{13}	-0.7%
ocean	-2.28×10^{16}	-16.0%	-6.73×10^{13}	-0.7%
<i>Wet deposition</i>				
land	-5.55×10^{15}	-3.9%	-2.12×10^{15}	-21.3%
ocean	-4.25×10^{15}	-3.0%	-9.29×10^{14}	-9.3%
<i>Advection</i>				
west	-1.49×10^{12}	0.0%	-1.13×10^{07}	0.0%
south	-1.69×10^{16}	-11.9%	-3.69×10^{14}	-3.7%
north	-9.00×10^{15}	-6.3%	-7.18×10^{14}	-7.2%
east	-6.52×10^{16}	-45.8%	-4.58×10^{15}	-46.1%

^a Contributions of individual processes normalized to emissions from the FDNPP.

Figure 1.

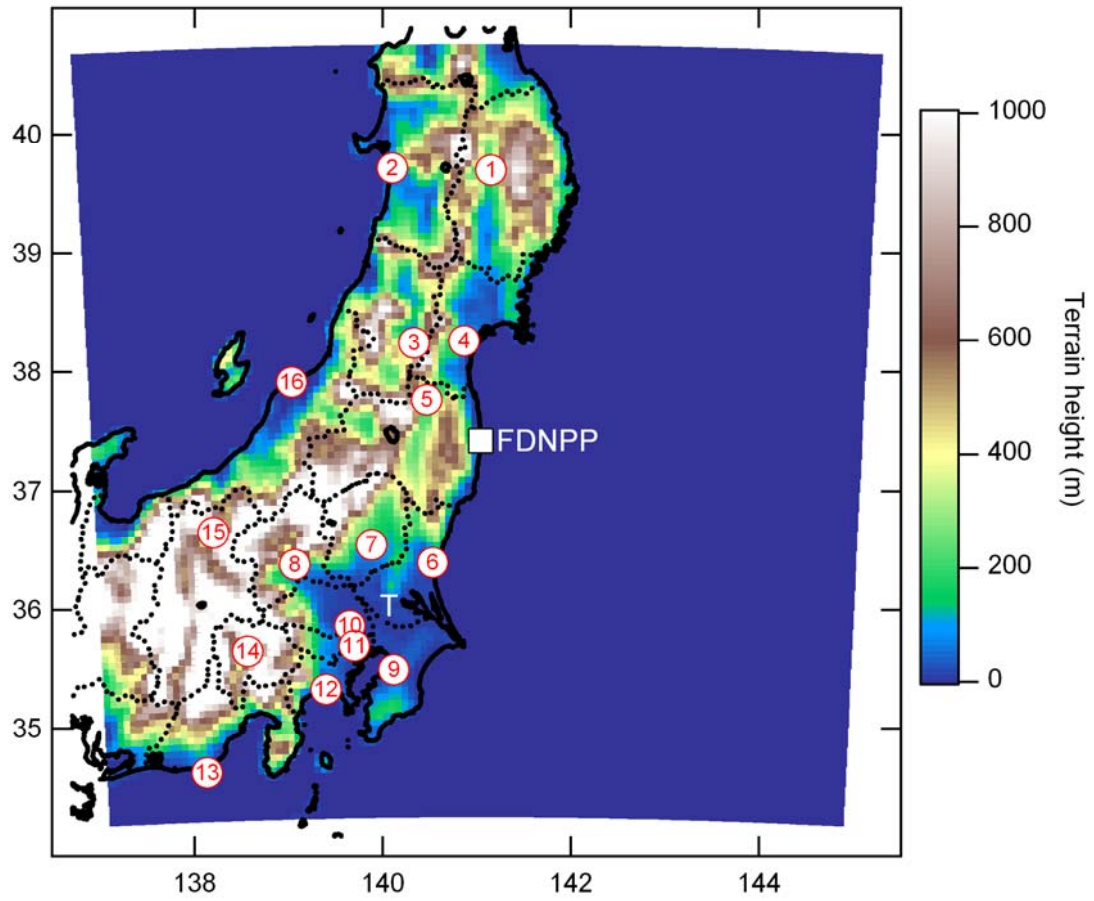


Figure 2.

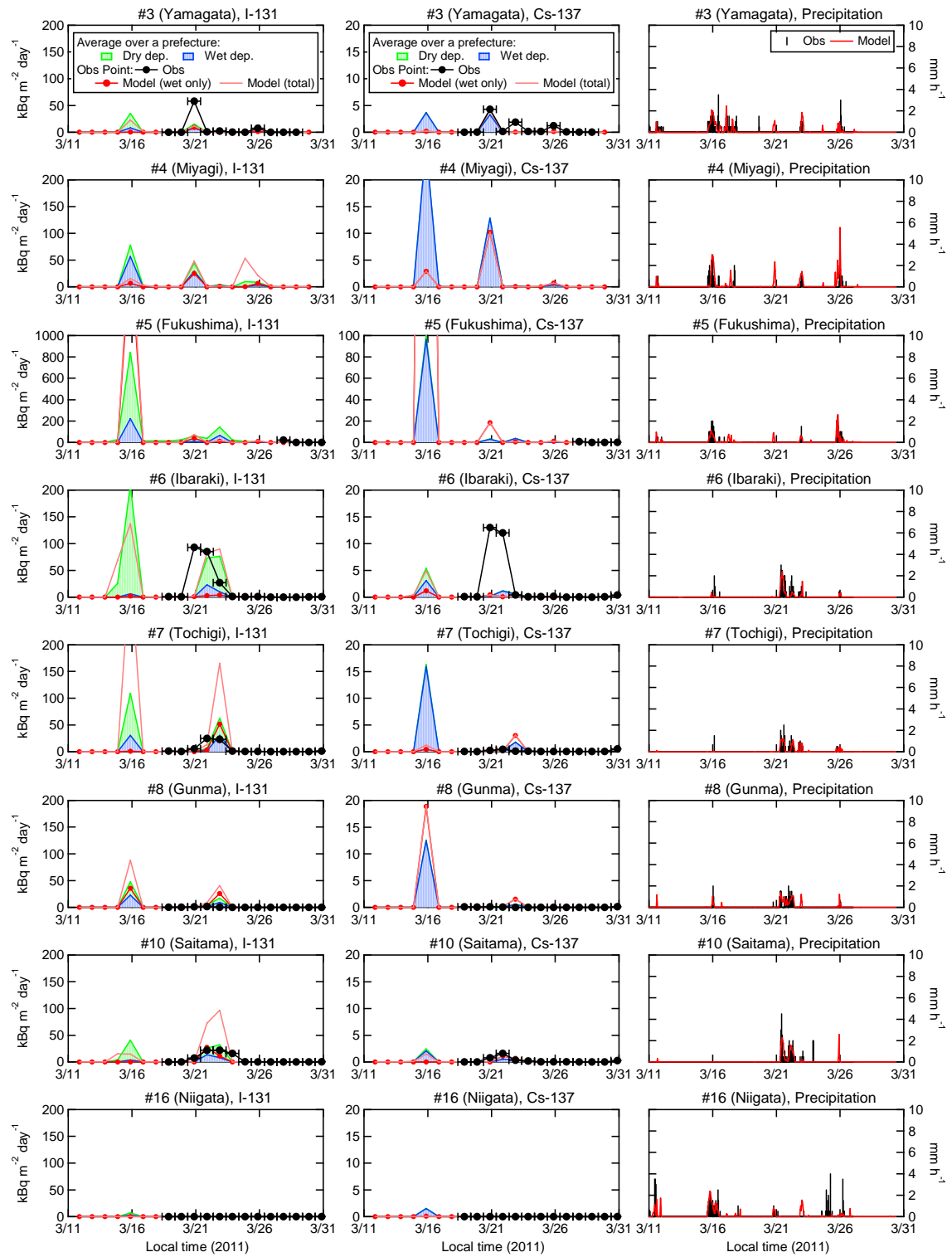


Figure 3.

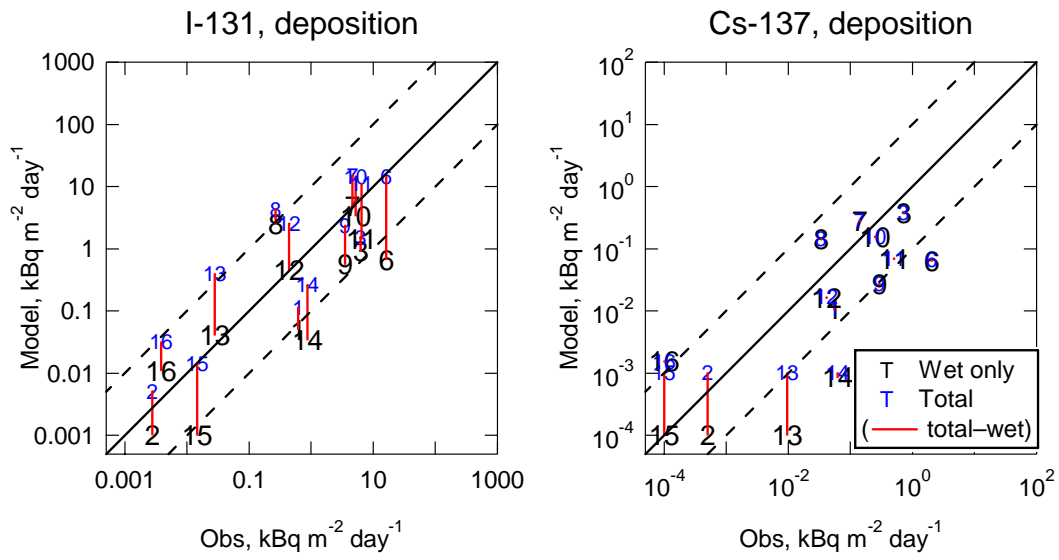


Figure 4.

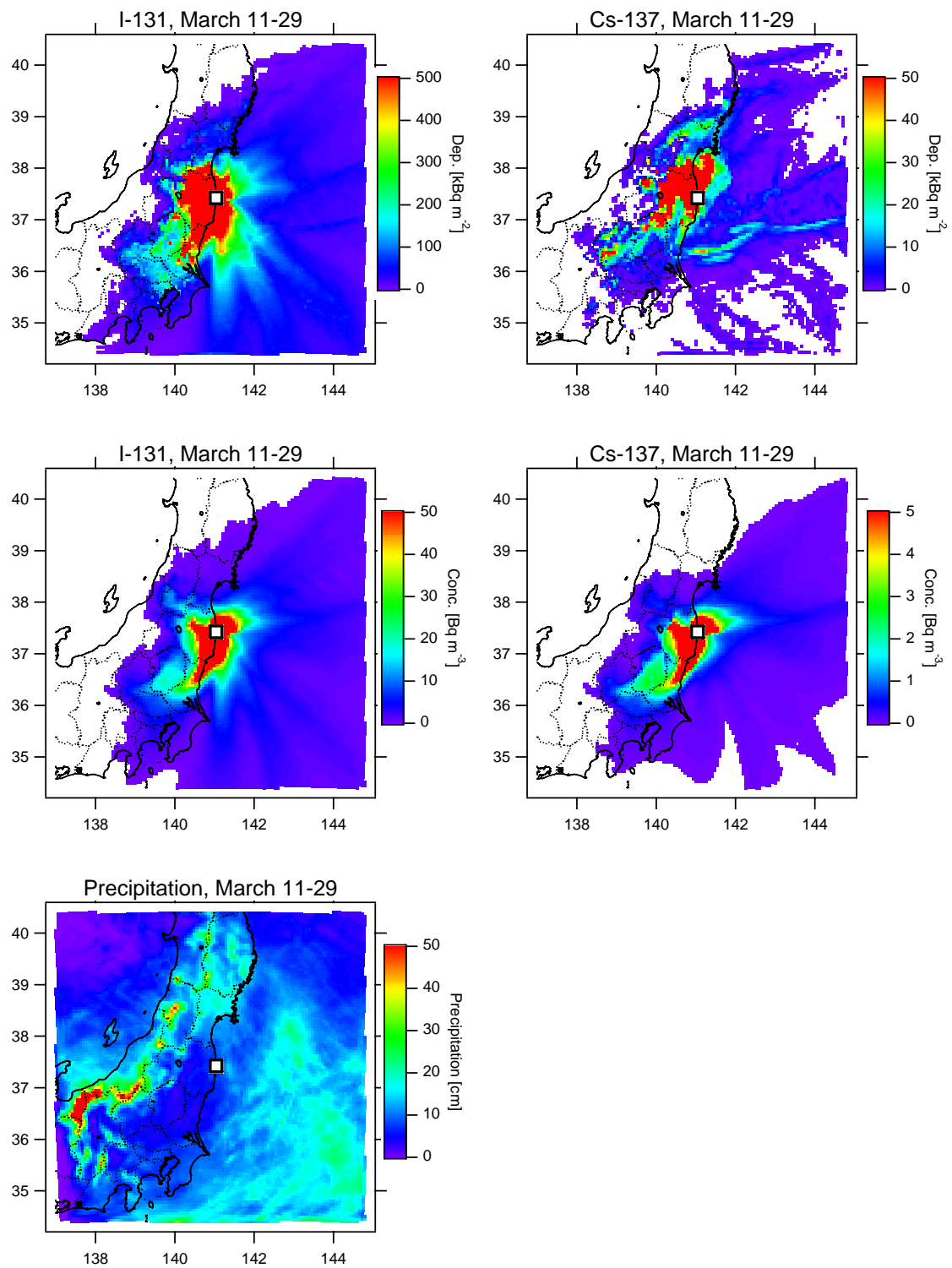
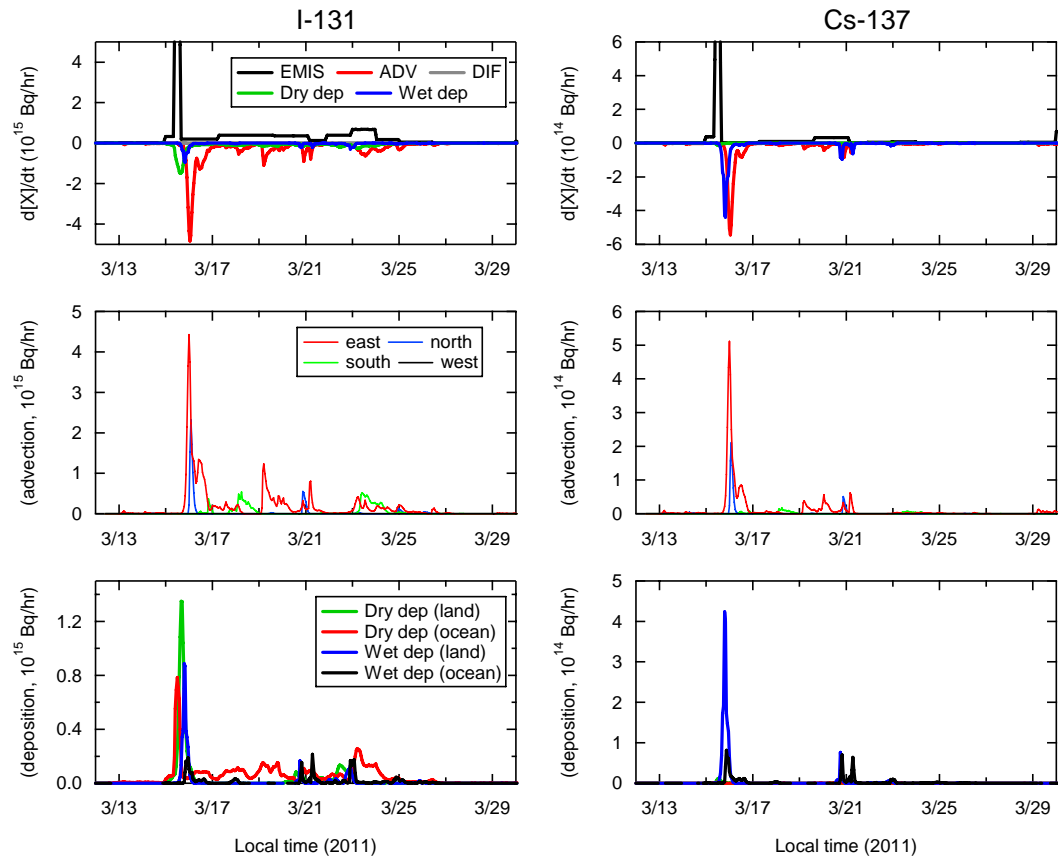


Figure 5.



Auxiliary Materials

Atmospheric behavior and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011

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Number of pages: 8

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Table S1. Simulated Dry and Wet Deposition of I-131 and Cs-137 (Bq) by Prefecture, 10–30 March 2011.

Prefecture	I-131			Cs-137			Area (km ²)
	Dry	Wet	Contr. ^a	Dry	Wet	Contr. ^a	
1 Iwate	7.92×10 ¹³	9.05×10 ¹³	0.1%	5.56×10 ¹⁰	3.50×10 ¹³	0.4%	15,279
2 Akita	1.09×10 ¹³	6.79×10 ¹²	0.0%	3.19×10 ⁹	1.92×10 ¹²	0.0%	11,636
3 Yamagata	3.27×10 ¹⁴	1.33×10 ¹⁴	0.3%	5.46×10 ¹¹	5.69×10 ¹³	0.6%	6,652
4 Miyagi	3.52×10 ¹⁴	6.07×10 ¹⁴	0.7%	3.49×10 ¹¹	2.57×10 ¹⁴	2.6%	6,862
5 Fukushima	8.07×10 ¹⁵	3.60×10 ¹⁵	8.2%	4.77×10 ¹³	1.48×10 ¹⁵	15.4%	13,783
6 Ibaraki	2.12×10 ¹⁵	2.23×10 ¹⁴	1.6%	1.73×10 ¹³	2.67×10 ¹³	0.4%	6,096
7 Tochigi	7.12×10 ¹⁴	4.27×10 ¹⁴	0.8%	2.78×10 ¹²	1.18×10 ¹⁴	1.2%	6,408
8 Gunma	2.18×10 ¹⁴	2.25×10 ¹⁴	0.3%	7.69×10 ¹¹	8.98×10 ¹³	0.9%	6,362
9 Chiba	2.39×10 ¹⁴	3.91×10 ¹³	0.2%	1.07×10 ¹²	2.70×10 ¹²	0.0%	5,082
10 Saitama	2.90×10 ¹⁴	9.51×10 ¹³	0.3%	1.53×10 ¹²	1.07×10 ¹³	0.1%	3,768
11 Tokyo	1.32×10 ¹⁴	2.85×10 ¹³	0.1%	5.90×10 ¹¹	3.75×10 ¹²	0.0%	2,103
12 Kanagawa	1.02×10 ¹⁴	2.11×10 ¹³	0.1%	3.13×10 ¹¹	1.01×10 ¹²	0.0%	2,416
13 Shizuoka	8.52×10 ¹³	8.76×10 ¹²	0.1%	2.52×10 ¹¹	1.92×10 ¹²	0.0%	7,255
14 Yamanashi	4.11×10 ¹³	1.63×10 ¹³	0.0%	7.83×10 ¹⁰	6.78×10 ¹²	0.1%	4,201
15 Nagano	2.49×10 ¹³	1.95×10 ¹³	0.0%	7.70×10 ¹⁰	1.07×10 ¹³	0.1%	13,105
16 Niigata	5.68×10 ¹³	3.07×10 ¹³	0.1%	2.18×10 ¹¹	1.47×10 ¹³	0.2%	10,364

^a Contributions of deposition rates normalized to emissions from the FDNPP.

Table S2. Fractions (Normalized to Emissions from the FDNPP) of I-131 and Cs-137 Deposited over Land in the Model Domain in Each Sensitivity Simulation^a

Vertical layer (height in meters ^b)	I-131 (%)	Cs-137 (%)
1 (50)	12.9	22.0
2 (120)	13.1	22.2
3 (220)	13.3	22.6
4 (340)	13.5	23.1
5 (490)	13.7	23.5
6 (680)	13.2	22.7

^a It was assumed that all radioactive materials were emitted in one vertical layer.

^b Approximate height of the top of each vertical layer. The height of each vertical layer is not constant spatially and temporary as σ_p coordinate is used.

Table S3. Fractions (Normalized to Emissions from the FDNPP) of I-131 and Cs-137 Deposited over Land in the Model Domain in Each Sensitivity Simulation

Date range ^a	I-131 (%)	Cs-137 (%)
11–13 March	0.2	0.0
13–15 March	6.6	8.2
15–17 March	7.5	16.3
17–19 March	0.0	0.0
19–21 March	8.7	13.8
21–23 March	24.3	41.4
23–25 March	3.6	4.8
25–27 March	4.1	6.5

^a From 0900 to 0900 local time; it was assumed that emission of radioactive materials was constant during each 48-h period.

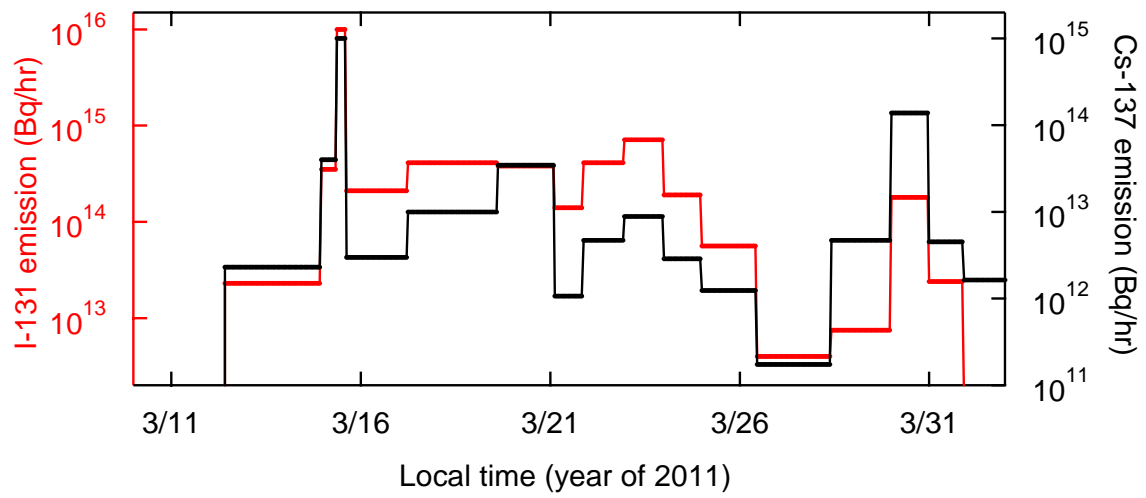


Figure S1. Rates of I-131 and Cs-137 emissions from the Fukushima Daiichi nuclear power plant in March 2011 [*Chino et al.*, 2011].

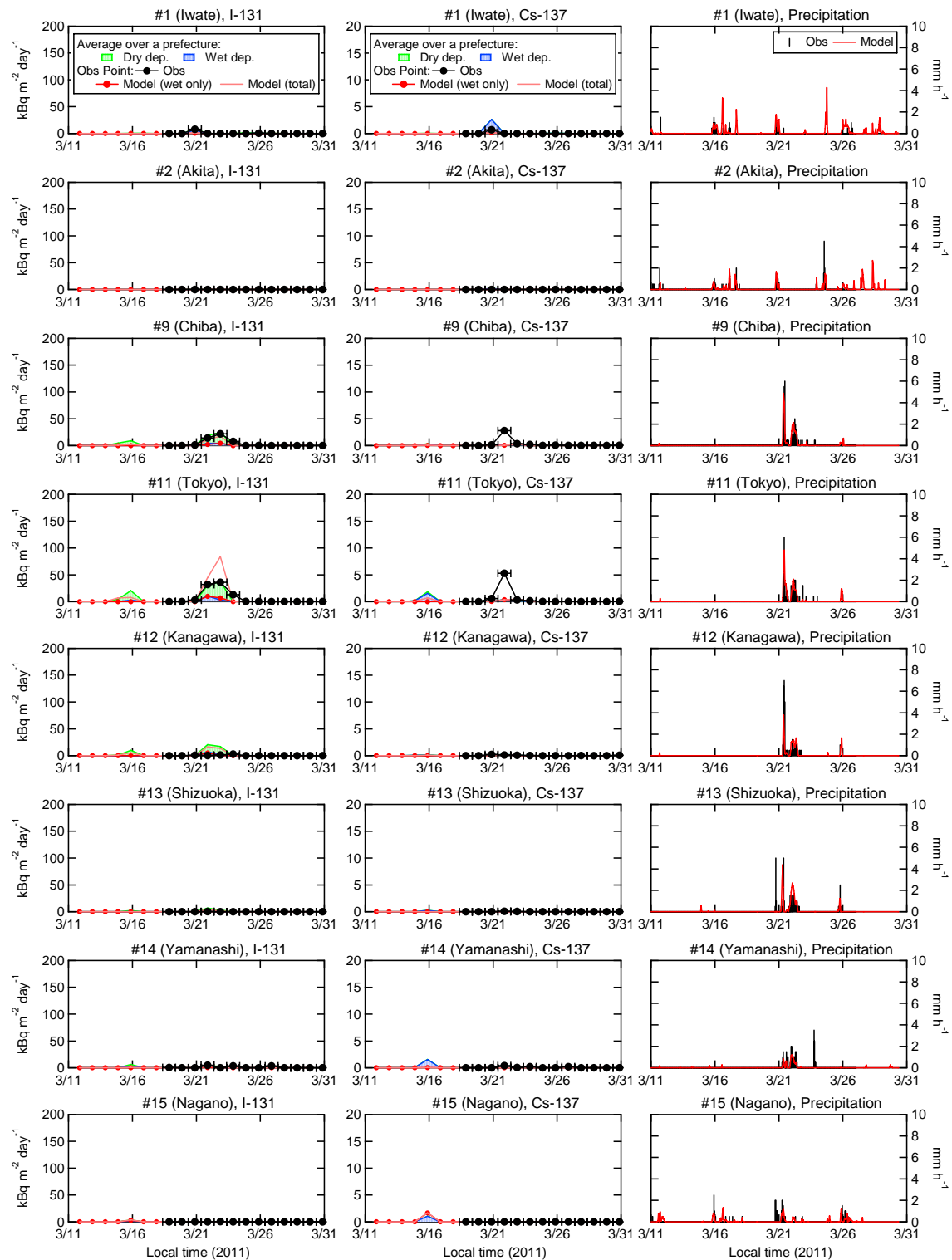


Figure S2. Observed and simulated deposition rates of I-131 (left) and Cs-137 (center) and precipitation rates (right) at selected measurement sites shown in Figure 1. Simulated dry and wet deposition rates averaged over each prefecture are indicated by green and blue hatching, respectively.

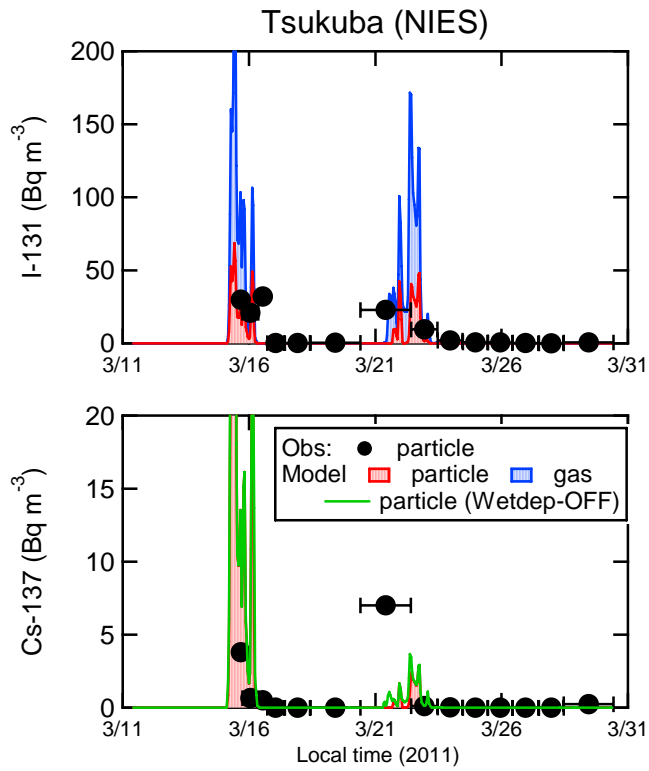


Figure S3. Observed and simulated activity concentrations of I-131 (top) and Cs-137 (bottom) in the particulate phase at the Tsukuba site.

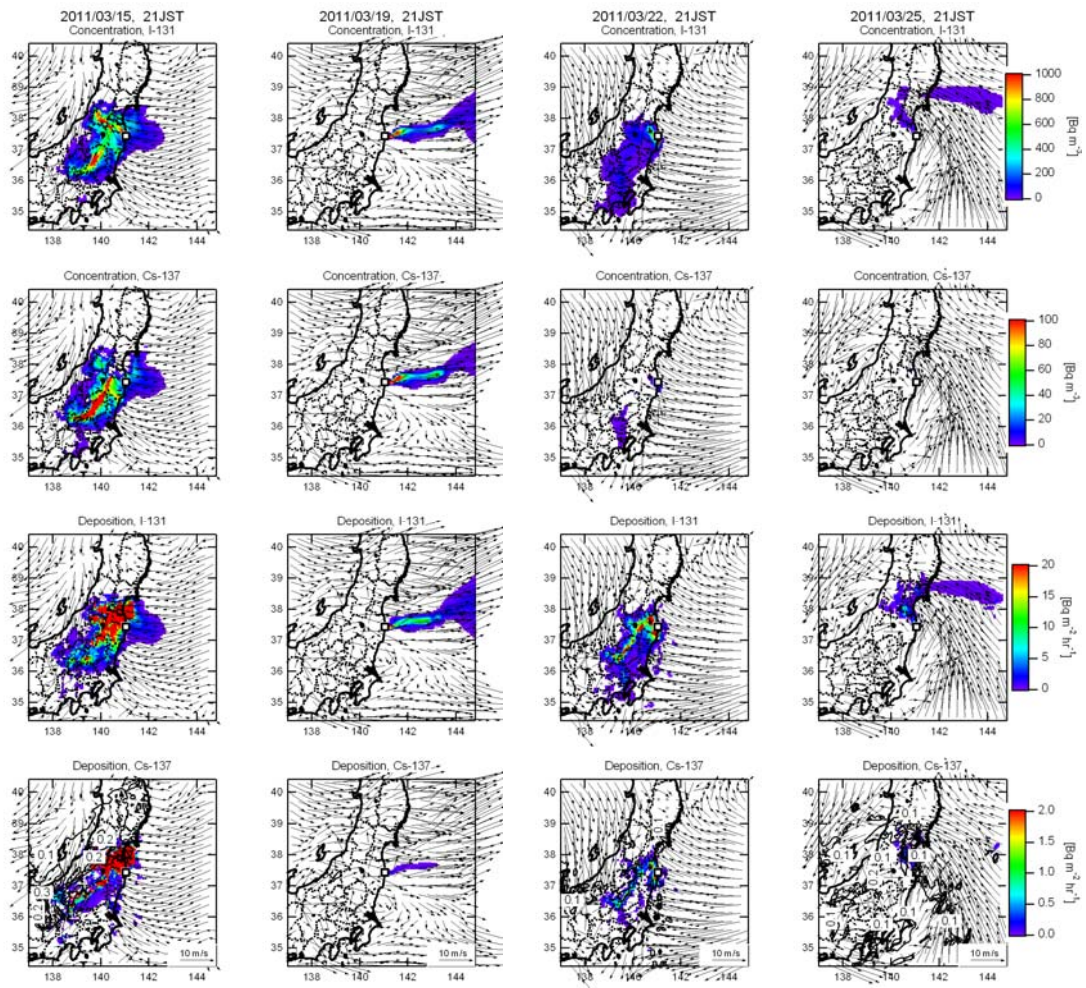


Figure S4. Distributions of activity concentrations and deposition rates of I-131 and Cs-137 simulated by CMAQ at 2100 local time on 15, 19, 22, and 25 March 2011. Simulated wind fields are also shown and simulated precipitation rates (cm h^{-1}) are given in the four bottom figures.

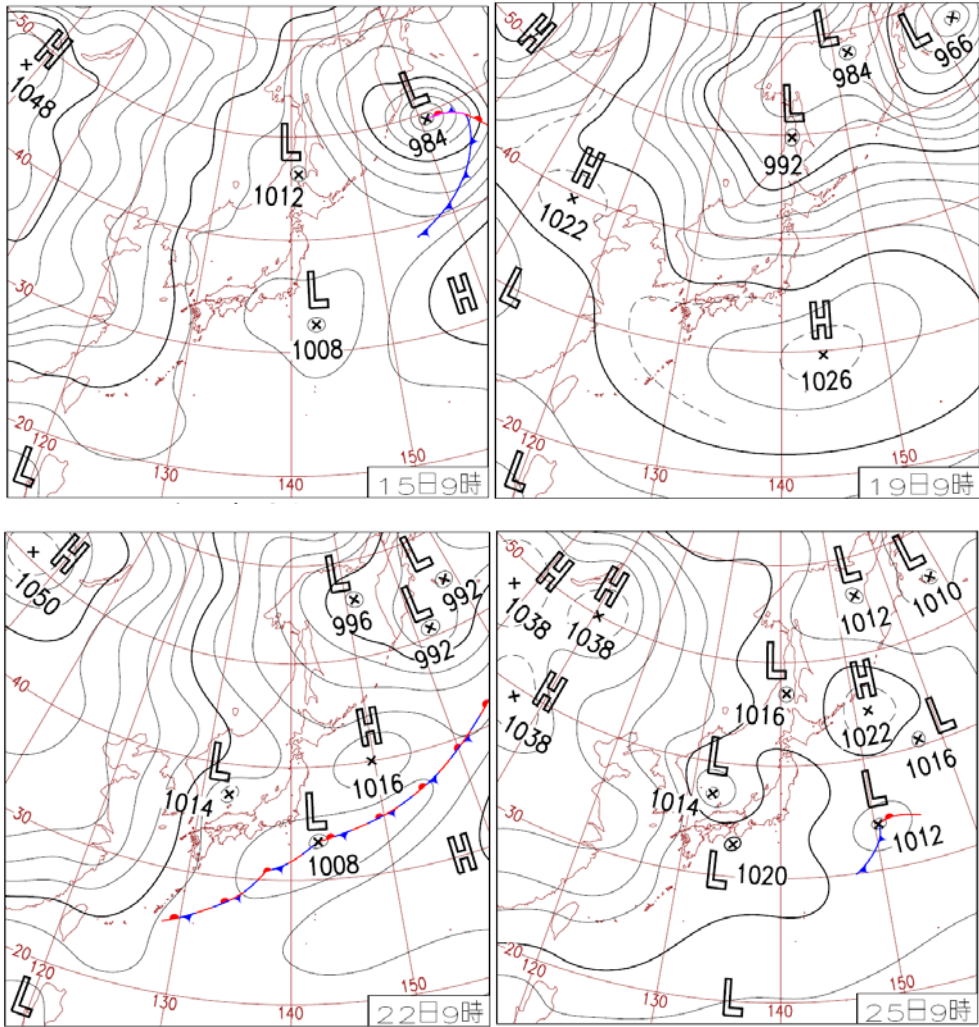


Figure S5. Weather charts at 0900 local time on 15 March (upper left), 19 March (upper right), 22 March (lower left), and 25 March (lower right) 2011.