



Modeling transport
and deposition of
radionuclides

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Modeling and sensitivity analysis of transport and deposition of radionuclides from the Fukushima Daiichi accident

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Abstract

The atmospheric transport and ground deposition of radioactive isotopes ^{131}I and ^{137}Cs during and after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident (March 2011) are investigated using the Weather Research and Forecasting/Chemistry (WRF/Chem) model. The aim is to assess the skill of WRF in simulating these processes and the sensitivity of the model's performance to various parameterizations of unresolved physics. The WRF/Chem model is first upgraded by implementing a radioactive decay term into the advection-diffusion solver and adding three parameterizations for dry deposition and two parameterizations for wet deposition. Different microphysics and horizontal turbulent diffusion schemes are then tested for their ability to reproduce observed meteorological conditions. Subsequently, the influence on the simulated transport and deposition of the characteristics of the emission source, including the emission rate, the gas partitioning of ^{131}I and the size distribution of ^{137}Cs , is examined. The results show that the model can predict the wind fields and rainfall realistically. The ground deposition of the radionuclides can also potentially be captured well but it is very sensitive to the emission characterization. It is found that the total deposition is most influenced by the emission rate for both ^{131}I and ^{137}Cs ; while it is less sensitive to the dry deposition parameterizations. Moreover, for ^{131}I , the deposition is also sensitive to the microphysics schemes, the horizontal diffusion schemes, gas partitioning and wet deposition parameterizations; while for ^{137}Cs , the deposition is very sensitive to the microphysics schemes and wet deposition parameterizations, and it is also sensitive to the horizontal diffusion schemes and the size distribution.

1 Introduction

Large amounts of radionuclides were released into the atmosphere after the nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP) on 11 March 2011. Later, the Japanese government reported that the radioactive materials were detected

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in the food and water supply in Fukushima and adjacent areas (Zakaib, 2011). Radionuclides can significantly jeopardize human health, causing cancer and acute radiation diseases (Till and Grogan, 2008). Understanding the spatial and temporal distributions of radionuclides is key to assessing their health impact; it is thus important to be able to accurately model their atmospheric transport and ground deposition.

Over the past few decades, many numerical models have been developed and applied for studying the transport and deposition of radionuclides (Andronopoulos and Bartzis, 2010; de Sampaio et al., 2008; Lauritzen et al., 2007; Lutman et al., 2004; Terada and Chino, 2008; Leelossy et al., 2011). For this particular accident at Fukushima, the Community Multi-scale Air Quality (CMAQ) model (Morino et al., 2011) and the Lagrangian Particle Dispersion model with meteorological conditions provided by the Weather Research and Forecasting (WRF) model (Srinivas et al., 2012) have been used. These studies, together with many previous studies for other events, have identified a number of meteorological variables that can significantly influence the atmospheric transport and ground deposition of radionuclides, including wind and rainfall (Basit et al., 2008; Mathieu et al., 2012; Takemura et al., 2011; Ten Hoeve and Jacobson, 2012; Yamauchi, 2012). For example, the study of Morino et al. (2011) has shown that during the period from 11 to 30 March 2011, the amounts of ^{131}I and ^{137}Cs transported across the eastern boundary (downwind) of their domain are 6.52×10^{16} Bq and 4.58×10^{15} Bq, respectively; while those across the western boundary (upwind) are only 1.49×10^{12} Bq and 1.13×10^7 Bq, respectively. This confirms that wind direction significantly affects the atmospheric transport of radionuclides. Rainfall is another important factor that can significantly influence the ground deposition of radionuclides. Studies of the Fukushima accident report that the estimated deposition mainly occurred when frontal rain bands passed over Japan on 21 March (Yasunari et al., 2011). Deposition of the radionuclides mainly occurred between 15–17 and 19–21 March, when heavy rainfall was observed, as reported by Srinivas et al. (2012). During these days, wet deposition was significantly higher than dry deposition. Even over longer periods, wet deposition usually still dominates over dry deposition. Given the findings above, it is

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clear that an accurate simulation of meteorological fields is a necessary condition for the accurate simulation of the transport and deposition of the radionuclides. However, most of the previous studies are focused on the behavior of radionuclides with the meteorological conditions simply taken from some numerical weather prediction models or analysis/reanalysis products.

Emission rate is another critical factor that controls the rate of atmospheric transport and ground deposition of radionuclides (Korsakissok et al., 2013; Morino et al., 2011, 2013). For instance, the study of Korsakissok et al. (2013) reported that the total deposition (sum of dry and wet deposition; in this paper, it is used interchangeably with ground deposition) of ^{137}Cs is less than 1.5×10^{15} Bq with the emission rate estimated by (Chino et al., 2011) but more than 5.5×10^{15} Bq with the emission rate estimated by (Stohl et al., 2012). The importance of using accurate estimation of emission source strength has been also demonstrated by the sensitivity analyses of Morino et al. (2013). However, most studies solely focused on the emission rate while other emission characteristics such as the gas partitioning of ^{131}I were not considered. The gaseous fraction of ^{131}I was simply set as a constant such as 80 % (Morino et al., 2011) and 2/3 (Korsakissok et al., 2013). A review (Sportisse, 2007) shows that the gaseous fraction varies among different studies: 65 % to 80 % (Chamberlain, 1991), 50–65 % (Clark and Smith, 1988), and 70–90 % in (Baklanov and Sorensen, 2001). This review also points out that the partitioning between gaseous and particulate phases is crucial for capturing the ground deposition of radionuclides. Apart from the gas partitioning, the size distribution of particles is another important characteristic, which has not been thoroughly studied, since some deposition schemes explicitly take the size distribution into account (Brandt et al., 2002).

In this study, to address some of the research gaps detailed above, we choose to adopt the WRF/Chem framework (Grell et al., 2005), which provides multiple parameterizations for different unresolved physical processes. WRF/Chem directly couples the forecasting of the chemistry and meteorology, allowing the transport simulations to exploit the full spatial and temporal resolutions of the meteorological simulations.

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This is expected to yield better results than offline approaches where pre-computed meteorological fields have to be interpolated to drive the chemical transport module. It is hypothesized that an optimal combination of physical parameterizations yielding the most accurate meteorological fields can be identified and that, by coupling this optimal configuration directly with the modeling of atmospheric transport, the uncertainties in the transport and ground deposition of radionuclides can be reduced.

Meteorological fields are however not the only source of uncertainty that can reduce the accuracy of transport and deposition simulations. Previous studies have also shown that different dry and wet deposition parameterizations can cause different deposition rates and accumulated amounts of radionuclides (see e.g. Brandt et al., 2002). In this study, several dry and wet deposition parameterizations are thus also added to the WRF/Chem model to test and intercompare their performances. Moreover, we consider the emission of ^{131}I with different gaseous fractions and that of ^{137}Cs with different particle size distributions. An aim of this study is therefore to obtain a combination of deposition parameterizations and emission characteristics, including emission rates, which produce the lowest error in the simulated ground deposition. The specific questions to be answered in this study are as follows:

1. What model setup parameters have the largest influence on the simulated meteorological fields and what is the influence of these fields on deposition?
2. What is the relative importance of wet vs. dry deposition, and how sensitive are they to the different parameterizations?
3. How sensitive are the modeled deposition to the imposed emission rates and characteristics, including the gas partitioning of ^{131}I and the size distribution of ^{137}Cs ?
4. How close can model results get to observed deposition given the uncertainties in model physics and inputs, and which of these uncertainties is the most critical?

37.5° N, 141.0° E. Moreover, all ^{137}Cs is assumed to be in particulate phase with different size distributions, while the gaseous fraction of ^{131}I varies in different simulations. Note that particle size distributions and the gas partitioning may change during the transport and deposition processes. However, this is not considered in our simulations.

2.2 Simulation model

In WRF/Chem, advection, turbulent diffusion, emission, radioactive decay and wet deposition are described using the following Eulerian advection-diffusion-reaction equation:

$$\frac{\partial A}{\partial t} + \text{div}(\mathbf{u}A) = \text{div}\left(\rho\mathbf{K}\nabla\left(\frac{A}{\rho}\right)\right) - \Lambda^s A - \lambda A + E \quad (1)$$

where A is the air concentration (Bq m^{-3}), which represents the radioactivity per unit volume equivalent to the number of radionuclides that decay per second in a unit volume. Λ^s is the wet scavenging rate (s^{-1}), λ represents the (first-order) radioactive decay rate (s^{-1}) and E is the point source for the radionuclides. \mathbf{K} is the turbulent diffusivity tensor which includes the effect of dry deposition. Note that WRF is a fully compressible model. The total number of the radionuclides per m^3 at $t = 0$ can be calculated from the following equation

$$N_0 = \frac{A_0}{\lambda} \quad (2)$$

In WRF/Chem, the unit used for transport of gases is ppmv and that for transport of aerosols is $\mu\text{g kg}^{-1}$. However, the unit used in the emission module is Bq m^{-3} , which is consistent with the unit used in the emission files from JAEA and TEPCO. Therefore, in order to use the default units to calculate the atmospheric transport of the radionuclides, a unit conversion is necessary for input of emissions to WRF, and then to convert

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its output into Bq m^{-3} . Based on the Eq. (2):

$$\frac{W_I}{V_m} = \frac{A_I}{\lambda_I \cdot NA} \cdot 10^6 \quad (3)$$

$$\frac{W_{Cs}}{M_{Cs}} = \frac{A_{Cs}}{\lambda_{Cs} \cdot NA \cdot \rho_{\text{air}}} \cdot 10^6 \quad (4)$$

5 where W_I is the air concentration of ^{131}I in ppmv; W_{Cs} is the air concentration of ^{137}Cs in $\mu\text{g kg}^{-1}$; A_I (Bq m^{-3}) is the air concentration of ^{131}I in Bq m^{-3} ; A_{Cs} is the air concentration of ^{137}Cs in Bq m^{-3} ; M_I (g mol^{-1}) is the molar mass of ^{131}I ; M_{Cs} (g mol^{-1}) is the molar mass of ^{137}Cs ; λ_I (s^{-1}) is the radioactive decay rate of ^{131}I ; λ_{Cs} (s^{-1}) is the radioactive decay rate of ^{137}Cs ; NA (mol^{-1}) is the Avogadro constant; V_m ($\text{m}^3 \text{mol}^{-1}$) is the molar volume of the air; and ρ_{air} (kg m^{-3}) is the air density.

10 Applying the ideal gas law to atmospheric air:

$$\rho V_m = RT \quad (5)$$

Thus,

$$V_m = \frac{RT}{\rho} = M_{\text{air}} \cdot \alpha_{\text{air}} \quad (6)$$

15 where M_{air} (kg mol^{-1}) is the molar mass of the air and α_{air} ($\text{m}^3 \text{kg}^{-1}$) is the specific volume of the air.

Based on Eqs. (3) and (4), we obtain the following equations,

$$W_I = \frac{A_I \cdot M_{\text{air}} \cdot \alpha_{\text{air}}}{\lambda_I \cdot NA} \cdot 10^6 \quad (7)$$

$$W_{Cs} = \frac{A_{Cs} \cdot M_{Cs} \cdot \alpha_{\text{air}}}{\lambda_{Cs} \cdot NA} \cdot 10^6 \quad (8)$$

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In the advection-diffusion solver of WRF/Chem, we use W_I and W_{Cs} to calculate the transport of ^{131}I and ^{137}Cs respectively, and subsequently use A_I and A_{Cs} converted based on inversion of Eqs. (7) and (8) for the outputs.

2.3 Parameterizations of removal processes

To simulate the transport and deposition of radionuclides more realistically, we added the radioactive decay process into the advection-diffusion solver. To examine the performance of different parameterizations in capturing the ground deposition of radionuclides, we improved the default resistance method for dry deposition and added two new dry deposition parameterization schemes: (1) the simple method and (2) the constant deposition velocity method. Furthermore, we implement a parameterization based on the relative humidity for wet deposition, in addition to the default WRF/Chem parameterization based on the precipitation rate.

2.3.1 Radioactive decay

The radioactive decay is similar to a first-order chemical reaction. The air concentration of a radioactive material, A , can be described as:

$$A = A_0 e^{-\lambda t} \quad (9)$$

where A_0 represents the air concentration at $t = 0$. The radioactive decay rates are taken from IAEA (2001) (International Atomic Energy Agency): $\lambda_I = 9.98 \times 10^{-7} \text{ (s}^{-1}\text{)}$ and $\lambda_{Cs} = 7.33 \times 10^{-10} \text{ (s}^{-1}\text{)}$. Considering the low radioactive decay rate of ^{137}Cs (equivalent to a half-life of about 30 yr), its decay process is neglected in this study, while the radioactive decay of ^{131}I is retained (half-life of about 8 days).

2.3.2 Dry deposition

As presented in (Seinfeld and Pandis, 2006), we assume that the dry deposition flux is proportional to the local air concentration of the radionuclides at the lowest level of the

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atmospheric model:

$$F = -v_{\text{dep}}A \quad (10)$$

where v_{dep} is the dry deposition velocity. In this study, three different parameterizations of the dry deposition velocity are tested.

5 The resistance method

Based on (Wesely, 1989), the dry deposition velocity for gases is described by three characteristic resistances, as follows:

$$v_{\text{dep}} = \frac{1}{r_a + r_b + r_s} \quad (11)$$

10 where r_a is the aerodynamic resistance, r_b is the quasi-laminar layer (viscous sublayer) resistance and r_s is the surface resistance (describing the resistance of the surface to the uptake/absorption/adsorption of the gas). The parameterizations of these three resistances in our study follows Brandt et al. (2002).

For particles, the surface resistance is neglected while the gravitational settling velocity is considered instead. The deposition velocity for particles can be expressed as (Seinfeld and Pandis, 2006):

$$15 \quad v_{\text{dep}} = u_{\text{grav}} + \frac{1}{r_a + r_b + r_a r_b u_{\text{grav}}}, \quad (12)$$

where u_{grav} is the gravitational settling velocity. According to (Brandt et al., 2002), the gravitational settling velocity can be calculated from the Stokes equation (small particles in the atmosphere experience a creeping flow, Reynolds number $\ll 1$, that appears to change in time due to the larger scale turbulent eddies):

$$20 \quad u_{\text{grav}} = \frac{d_p^2 g (\rho_p - \rho) Cc}{18\nu} \quad (13)$$

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where d_p is the particle diameter, g the acceleration of gravity, ρ_p the particle density (1.88 g cm^{-3} for cesium (Weast, 1988) and 3.5 g cm^{-3} for Iodine (Ristovski, 2006), the units of the particle density are converted to kg m^{-3} for use in WRF/Chem), ρ is the density of air, ν is the kinematic viscosity of air ($1.5 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$) and C_c is the Cunningham correction factor given by (Brandt et al., 2002).

$$C_c = 1 + \frac{\lambda_{\text{air}}}{d_p} \left(2.514 + 0.80 \exp \left(-\frac{0.55 d_p}{\lambda_{\text{air}}} \right) \right) \quad (14)$$

where $\lambda_{\text{air}} = 6.53 \times 10^{-8} \text{ m}$ is the mean free path at standard temperature and pressure.

The simple parameterization

According to Brandt et al. (2002), the dry deposition velocity can be calculated by a simple parameterization based on the friction velocity and the Obukhov length:

$$v_{dep} = \frac{u_*}{a}, \quad L > 0 \quad : \text{ stable conditions} \quad (15)$$

$$v_{dep} = \frac{u_*}{a} \left(1 + \left(\frac{300}{-L} \right)^{2/3} \right), \quad L < 0 \quad : \text{ unstable conditions}$$

where u_* is the friction velocity and L is the Obukhov length (Stull, 1988); a is a constant which for low vegetation is set to 500 and for forests to 100 (Brandt et al., 2002).

The constant deposition velocity method

In this parameterization, the dry deposition velocity is simply a constant. We use some typical values for of ^{131}I and ^{137}Cs that are found in the literature: the dry deposition

velocity of gas-phase ^{131}I is $0.5 \text{ (cm s}^{-1}\text{)}$ (Baklanov and Sorensen, 2001), the dry deposition velocity of particulate ^{131}I is $0.1 \text{ (cm s}^{-1}\text{)}$ (Baklanov and Sorensen, 2001) and the dry deposition velocity of ^{137}Cs is $0.05 \text{ (cm s}^{-1}\text{)}$ (Maryon et al., 1991) (the units of all deposition velocities are converted to m s^{-1} for use in WRF/Chem).

In this study, the accumulated dry deposition at each location is calculated. The decay process of radionuclides after they reach the ground surface follows the same radioactive decay rate. In addition, additional decay can occur due to soil activity; this additional decay can be represented by a constant λ_s , which has the same units as the radioactive decay (s^{-1}). Thus, the accumulated ground deposition can be computed using the following equation,

$$D_{\text{gr}}(t) = \int_{t_0}^t -v_{\text{dep}} \cdot A \cdot e^{-(\lambda+\lambda_s)t} dt \quad (16)$$

where D_{gr} (Bq m^{-2}) is the accumulated ground deposition, t_0 the initial time of deposition, t the duration after deposition, and λ the (first-order) radioactive decay rate (s^{-1}). In this study, the reduction rate due to soil activity λ_s of ^{131}I and ^{137}Cs are specified as 0 and $1.62 \times 10^{-9} \text{ (s}^{-1}\text{)}$ respectively (IAEA, 2001).

2.3.3 Wet deposition

The parameterization based on precipitation rate

Following Sportisse (2007), the wet deposition rate is described as:

$$\Lambda^s = a\rho_0^b \quad (17)$$

where ρ_0 is the rain intensity (mm h^{-1}); a and b are the parameters for specified radionuclides. In this study, we set $a = 4 \times 10^{-5}$ and $b = 0.6$ for gaseous ^{131}I (Sportisse,

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2007), $a = 7 \times 10^{-5}$ and $b = 0.69$ for particulate ^{131}I (Jylha, 1991), and $a = 8 \times 10^{-5}$ and $b = 0.8$ for ^{137}Cs (Baklanov and Sorensen, 2001).

The parameterization based on relative humidity

The parameterization (Pudykiewicz, 1989) based on the relative humidity (RH) is another scheme for calculating the wet deposition rate:

$$\Lambda^s = 0, \quad \text{RH} < \text{RH}_t$$
$$\Lambda^s = 3.5 \times 10^{-5} \left(\frac{\text{RH} - \text{RH}_t}{\text{RH}_s - \text{RH}_t} \right), \quad \text{RH} \geq \text{RH}_t \quad (18)$$

where RH_t (= 80 %) is the threshold value of the relative humidity and RH_s (= 100 %) is the saturation value.

Similar to the accumulated dry deposition, in this study, the accumulated wet deposition is also calculated. The same constants for the increased decay rates due to soil activity of ^{131}I and ^{137}Cs are used for wet and dry deposition. In addition, the wet deposition rate Λ_s is height-dependent in this RH-based model. Following (Seinfeld and Pandis, 2006), the wet ground deposition can be calculated following:

$$W_{\text{gr}}(t) = \int_{t_0}^t \int_0^h \Lambda^s(z) \cdot A(z) \cdot e^{-(\lambda+\lambda_s)t} dz dt, \quad (19)$$

where W_{gr} (Bq m^{-2}) is the wet ground deposition and h is the height of the domain.

2.4 WRF configurations

The simulations are performed using 3 nested domains with horizontal resolutions of 9, 3, and 1 km for domain 1, domain 2, and domain 3, respectively (see Fig. 2). Domain

1 and Domain 2 are centered at 37.5° N, 141.0° E with 160 grid points in both the north–south direction and the east–west direction. Domain 1 nearly covers the whole of Japan and Domain 2 covers most of the Tohoku region and the Kanto region where observational stations are located. The innermost domain has 160 × 160 grids and is centered at 36.9° N, 140.4° E.

The simulation uses 27 vertical levels for all domains, with the highest level at the 10 000 Pa isobaric surface (WRF uses terrain following pressure coordinates in the vertical direction). The emissions are only released at the lowest level. The Global Forecasting System (GFS) reanalysis, with a 0.5° × 0.5° horizontal resolution, is used for initial and boundary conditions. The simulation period starts from 00:00 UTC 11 March and ends at 00:00 UTC 31 March 2011 with 1 h output interval. In this study, we conduct one reference case simulation (REF) and 12 sensitivity simulations as summarized in Table 1. One-way nesting is used for all simulations. Other physics schemes that are not changed include: (1) the Rapid Radioactive Transfer Model scheme for long wave radiation; (2) the Dudhia scheme for short wave radiation; (3) the Yonsei University Scheme for the planetary boundary layer; (4) the Noah Land Surface Model for non-urban land surface physics; (5) the single-layer urban canopy model for urban surface physics; (6) the New Grell scheme for cumulus parameterization (in this study, cumulus parameterization is only used for the domain 1; the other domains have fine resolutions that should allow them to resolve shallow convection).

2.4.1 Reference simulation

Simple aerosol treatment, using an aerosol scheme in which no direct or indirect effects are considered, is used. In the reference case, the 2-D Smagorinsky scheme is used for horizontal diffusion and the WSM 6 scheme is used for the microphysics. The resistance method is used for parameterizing dry deposition and the parameterization based on precipitation rate is used for wet deposition. The partitioning of ¹³¹I at the source is chosen to be 80 % gas as recommended in several studies (Korsakissok et al., 2013; Morino et al., 2011). Moreover, in the reference case, the size distribu-

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tion of particulate radionuclides is not taken into account: all particulate radionuclides have the same size, which is the average value. In this study, the average size of ^{131}I and ^{137}Cs are chosen to be 0.48 and 0.67 μm , respectively (Kaneyasu et al., 2012; Sportisse, 2007).

2.4.2 Sensitivity studies

As shown in Table 1, a variety of sensitivity simulations are carried out to evaluate the impact of different physics/parameterizations on the atmospheric transport and ground deposition of radionuclides. In case 2, the emission rate estimated by TEPCO is used to assess the uncertainty in the emission source term and its impact. In cases 3 and 4, two different microphysics schemes, the Goddard scheme and the Thompson scheme, are used to examine the impact of microphysics schemes on rainfall and on the modeling of transport and deposition of radionuclides. In case 5, the horizontal diffusion scheme is chosen to be the 1.5 order TKE scheme, as compared to the Smagorinsky scheme that is used in the reference case. Cases 6, 7, 8 and 9 are designed to assess the sensitivity of simulated results of ^{131}I to the gas partitioning, with the gaseous fraction of ^{131}I decreasing from 100 % to 0 %. In case 10, the size distribution of ^{137}Cs is considered. The average size remains 0.67 μm but the standard deviation is set to 1.3 μm (Kaneyasu et al., 2012). As compared to the reference case, cases 11 and 12 use the simple parameterization method and the constant deposition velocity method, respectively, to parameterize dry deposition; case 13 uses the parameterization based on relative humidity for wet deposition.

Errors including Percentage Bias (PBIAS), Percentage Root Mean Square Error (PRMSE) and Mean Bias Error (MBE) are used to evaluate the model performance and compare the results from different sensitivity cases. PBIAS and PRMSE are used for wind speed, precipitation and total deposition; while MBE is only used for evaluating the wind direction for which percentage errors are not adequate (e.g. when the observed value is 1° and the modeled value is 359° , the PBIAS is -35800% ; when the

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observed value is 357° and the modeled value is 359°, the PBIAS is -0.57 %; however the absolute errors of the wind direction under these two conditions are both 2°.

PBIAS, PRMSE and MBE are defined as follows:

$$\text{PBIAS} = \frac{\frac{1}{n} \sum_{i=1}^n (O_i - M_i)}{\frac{1}{n} \sum_{i=1}^n O_i} \times 100\% \quad (20)$$

$$\text{PRMSE} = \frac{\sqrt{\frac{1}{n} \sum_{i=1}^n (O_i - M_i)^2}}{\frac{1}{n} \sum_{i=1}^n O_i} \times 100\% \quad (21)$$

$$\text{MBE} = \frac{1}{n} \sum_{i=1}^n (O_i - M_i) \quad (22)$$

where O_i represents the observed value, M_i represents the modeled value.

2.5 Observational datasets

Hourly wind speed, wind direction¹ and rainfall² data are obtained from National Climatic Data Center (NCDC) at stations YAMAGATA, CHIBA, TOKYO, ONAHAMA, NIIGATA, MAEBASHI, SENDAI and ISHINOMAKI. These data are used to assess the WRF-simulated wind and rainfall fields. Daily total deposition of ¹³¹I and ¹³⁷Cs are measured by bulk samplers over 46 stations, which are provided by Ministry of Education, Culture, Sports, Science and Technology (MEXT)³ In this study, we only select 7 of the 46 stations to validate the model since most of the stations do not have available data covering the period from 18 March to 31 March (all of the 46 stations do not have

¹<http://cdo.ncdc.noaa.gov/pls/plclimprod/poemain.cdobystn?dataset=DS3505&StnList=47409099999>

²<http://www.ncdc.noaa.gov/cdo-web/datasets/GHCND/stations/GHCND:JA000047409/detail>

³<http://www.mext.go.jp/english/incident/1307872.htm>

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available data before 18 March). The 7 stations are YAMAGATA, IBARAKI, TOCHIGI, GUNMA, SAITAMA, CHIBA and TOKYO.

3 Results and discussion

This section is organized in the following way: in Sect. 3.1, the simulated wind and rainfall fields are validated, and their impact on the atmospheric transport and ground deposition of radionuclides is detected. Section 3.2 analyzes the contributions of dry and wet deposition to total deposition and examines the sensitivity of ground deposition to different parameterizations of dry and wet deposition. Section 3.3 examines the sensitivity of ground deposition to the different characteristics of the emission rate, the gas partitioning of ^{131}I and the size distribution of ^{137}Cs .

3.1 Meteorological fields and their influence on deposition of radionuclides

This section validates the WRF-simulated wind and rainfall fields using observational data at various locations. The impact of wind and rainfall on the atmospheric transport and ground deposition of radionuclides is also examined. In particular, the sensitivity of deposition to different microphysical parameterizations and horizontal diffusion schemes in WRF is investigated.

3.1.1 Validation of WRF-simulated wind and rainfall fields and their sensitivity to the horizontal diffusion and microphysics schemes

WRF-simulated wind speed and direction at 10 m in the reference case (REF) from domain 2 are compared to observed data over 8 stations in Japan and the results are shown in Figs. 3 and 4. As can be seen, the wind fields simulated by WRF show a good agreement with the observations at most of the stations such as CHIBA, SENDAI and

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ISHINOMAKI. Nevertheless, WRF significantly overestimated the wind speed at YAMAGATA during the whole simulation period.

The biases in the WRF-simulated wind fields are quantified using PBIAS, PRMSE, and MBE as introduced in Sect. 2. As can be seen from Table 2, these statistics are also calculated for the case using the 1.5 order TKE horizontal diffusion scheme (DIF2) in addition to the reference case (REF). The values of PBIAS, PRMSE and MBE are quite close for both cases. However, as shall be seen later, the subtle differences in the wind fields generated by using two different horizontal diffusion schemes can result in significant differences in the ground deposition of radionuclides. It is clear that the PBIAS for wind speed at CHIBA, SENDAI and ISHINOMAKI are lower than 7% and the PRMSE at these 3 stations are also lower than at other stations. The PBIAS and PRMSE for wind speed at YAMAGATA are significantly higher than those at other stations, which is in agreement with Figs. 3 and 4. As for the wind direction, the MBE at CHIBA, NIIGATA and ISHINOMAKI are lower than 30°. Nevertheless, at YAMAGATA, the MBE of wind direction is about 50° for both diffusion schemes. The YAMAGATA station is located in an area surrounded by mountains, thus the large biases in the simulated wind speed and wind direction at YAMAGATA may be due to the coarse grid resolution (3 km) that is unable to resolve the subgrid-scale topography.

The simulated daily precipitation rate in the reference case (REF) is also compared to observational data at the 8 stations and the results are shown in Fig. 5. The WRF-simulated daily precipitation is in good agreement with the observations except at YAMAGATA, SENDAI, and ISHINOMAKI. At YAMAGATA, the WRF-simulated rainfall is a day ahead of the observed rainfall and the maximum rainfall rate is significantly underestimated by WRF. At SENDAI and ISHINOMAKI, the maximum rainfall rate is not well captured by WRF but the timing is almost correct. In order to examine the sensitivity of simulated rainfall to different microphysical parameterizations, the precipitation patterns generated from 3 different microphysics schemes are compared in Fig. 6 (REF with WSM 6; MP2 with Goddard; MP3 with Thompson). The left panels show the daily precipitation on 21 March and the right right panels show the accumulated precipitation from

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11 to 31 March over domain 2. Both the daily precipitation on 21 March and the accumulated precipitations show quite similar patterns overall, but there are differences observed among the three cases with the different microphysical parameterizations. For example, in the left panels, around 35.5–36° N, 138.5–140.5° E where the maximum precipitation occurs, the area with high precipitation values (> 20 mm) in case REF or MP2 is significantly higher than that in case MP3.

The errors associated with the simulated rainfall fields are quantified by PBIAS and PRMSE, as shown in Table 3. It is clear that the PBIAS values at most of the stations are lower than 30 % while the PRMSE values are about 200 %; this indicates that although there are large biases associated with the time series of rainfall, a significant fraction of these biases are related to timing and the averaged or the accumulated rainfall (which cancel the timing errors and are hence more accurately represented by PBIAS) are fairly well captured by WRF. This is not the case at station ISHINOMAKI where the values of PRMSE and PBIAS are extremely high. This is because the observational data at this station is only available after 20 March when the precipitation is significantly overestimated by WRF simulations (see Fig. 5). The comparison among the three microphysical schemes shows that case REF yields the least PBIAS at CHIBA, NIIGATA, MAEBASHI, SENDAI, and ISHINOMAKI. In particular, the PBIAS from case REF are significantly smaller than those from cases MP2 and MP3 at NIIGATA, MAEBASHI, and SENDAI. At TOKYO, the PBIAS from case REF is comparable to that from case MP2; while at ONAHAMA, it is comparable to that from case MP3. Only at YAMAGATA does the REF case produce the largest PBIAS; nonetheless, all of the three microphysical schemes yield very small PBIAS at YAMAGATA. As such, it can be concluded that the WSM6 microphysical scheme used in case REF performs the best among the three schemes examined here, at least for this study.

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3.1.2 Influence of wind and rainfall on the transport and deposition of radionuclides

In order to illustrate the impact of wind fields on the atmospheric transport of radionuclides, the concentration maps of ^{131}I at the lowest level of the atmospheric model at four different times (i.e., 00:00, 06:00, 12:00, 18:00 UTC) on 21 March are illustrated in the upper four panels of Fig. 7, along with wind vector field. At 00:00 UTC, the transport of the radionuclides from FDNPP is driven by northerly winds (towards the south). One can also notice the large concentrations to the east, over the Pacific ocean, and to the north of FDNPP at 00:00 UTC that are probably the remnants of previous north-eastward winds. The transport direction changes with wind and becomes northeasterly (towards the southwest, from the source to the Kanto region) from 12:00 to 18:00 UTC gradually. The bottom panels of Fig. 7 shows the accumulated daily dry and wet deposition on 21 March. As suggested by the concentration maps, the deposition is highest in the Kanto region that lies southwest of FDNPP. Dry depositions is small compared with wet deposition and the two depositions display different spatial patterns. In this REF case, the parameterization of wet deposition is based on precipitation. As shown in Fig. 6, there is a large amount of precipitation in the southwest area and hence the wet deposition is also high over this area. Thus, it is evident that the ground deposition, including dry and wet deposition, is influenced by both wind and rainfall.

The total deposition from cases using different horizontal diffusion schemes and microphysical schemes are compared in Fig. 8, where we show the daily total deposition at stations YAMAGATA and CHIBA as two examples. The results of both ^{131}I and ^{137}Cs indicate that the difference between REF and DIF2 is small at CHIBA but large at YAMAGATA. At YAMAGATA, the total daily deposition of both ^{131}I and ^{137}Cs in REF is only about half of that in DIF2 on 20 March, while it is slightly higher in DIF2 than that in REF on 22 and 25 March. Much larger differences are seen among different microphysics schemes at both CHIBA and YAMAGATA. For example, at YAMAGATA on 20 March, the deposition of ^{137}Cs simulated by REF is about 3.9 kBq m^{-2} and it is

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close to the observed value; while in MP2 and MP3, the amounts of deposition are 2.2 and 7.9 kBq m⁻², respectively. Figure 8 illustrates that the ground deposition of radionuclides are sensitive to both horizontal diffusion schemes and microphysical schemes. This however does not contradict our previous finding that at the 8 stations with measurements of wind speed and wind directions, the biases generated by the two cases using different horizontal diffusion schemes are relatively similar. This is because the daily accumulated ground deposition at one particular location is in fact affected by winds over the upwind fetch as well as turbulence levels at a given location. Despite the fact biases seen in the wind fields over the 8 stations are similar for the two horizontal diffusion schemes, the turbulence and upstream winds in the two cases are not necessarily similar. Furthermore, small differences in wind fields can generate relatively larger differences in precipitation and thus influence wet deposition. As such, subtle differences seen in the wind field in Table 2 might result in significant differences in the ground deposition depending on the sensitivity of ground deposition and precipitation to the wind field.

The PBIAS and PRMSE of total deposition of ¹³¹I and ¹³⁷Cs with different horizontal diffusion schemes and microphysics schemes are presented in Tables 4 and 5, respectively. In order to evaluate the performance of different schemes in a quantitative and consistent way, a ranking system is proposed. At each station, a local rank (LR) is assigned to each scheme. The scheme with the smallest error has rank 1 and the scheme with the second smallest error has rank 2, etc. Then a global rank (GR) is calculated by summing the local rank of each scheme over all stations. Finally, the global rank calculated with PBIAS and that calculated with PRMSE is summed up to yield a summed global rank (SR), which is used to compare the performance of different parameterization schemes. A scheme with the smallest SR performs the best among all the schemes that it is compared against. As shown in Table 4, the errors in case REF are close to those in case DIF2. However, the global ranks inferred from PBIAS and PRMSE are lower in case DIF2 than those in case REF for both ¹³¹I and ¹³⁷Cs, in-

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dicating that, globally, using the 1.5 order TKE scheme predicts the ground deposition better than using the horizontal Smagorinsky scheme.

Table 5 shows the errors in simulated total daily depositions of ^{131}I and ^{137}Cs with different microphysics schemes. It is shown that PBIAS in all 3 cases are below 100% over the 7 stations except that of ^{137}Cs at TOCHIGI and GUNMA. However, most of the PRMSE values of ^{131}I and ^{137}Cs are larger than 100%, especially in TOCHIGI and GUNMA where the PRMSE of ^{137}Cs is over 1200% in case MP3 and at least over 400% in the other two cases, suggesting that the model cannot capture the total daily deposition of ^{137}Cs at these 2 stations. Case REF has the lowest global rank based on both PBIAS and PRMSE for both ^{131}I and ^{137}Cs , which indicates that the microphysics scheme WSM 6 can better predict the total daily deposition than the Goddard scheme and the Thompson scheme. The much higher values of PRMSE compared to PBIAS indicates that a significant component of the errors are due to time shifts in the deposition patterns. Overestimations and underestimations of deposition at various times partially cancel each other in PBIAS, but not in PRMSE. Overall however, since one is interested in total deposition even if the timing is not very accurate, PBIAS might be a better measure of the ability of WRF to simulate the environmental impact of radionuclides deposition from the Fukushima accident.

3.2 Dry and wet deposition

This section examines the contributions of dry and wet deposition to total deposition respectively and examines the sensitivity of ground deposition to different parameterizations of dry and wet deposition.

3.2.1 Contributions of dry and wet deposition to total deposition

Simulated total daily depositions of ^{131}I and ^{137}Cs from 11 to 31 March at various locations indicate that the total daily depositions of ^{131}I and ^{137}Cs are significant at all of the 7 stations during two periods: from 15 to 16 March (not shown) and from 20 to

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23 March. These periods with high total daily depositions correspond to periods with high emission rates from the source (see Fig. 1). Since there is no observational data for ground deposition before 18 March, the following analyses will focus on the period from 18 March to 31 March with maximum total daily depositions occurring from 20 to 23 March.

Comparisons between simulated total daily depositions of ^{131}I and ^{137}Cs and observational data are shown in Fig. 9. During the period from 18 to 31 March, simulated total daily depositions generally follow the pattern observed in the measurements; however, the simulations significantly underestimate the observed deposition peak of ^{131}I around 20 to 22 March at all of the stations. As for ^{137}Cs , the total daily depositions are over-estimated at TOCHIGI, GUNMA and SAITAMA and underestimated at YAMAGATA, IBARAKI and CHIBA. From Fig. 9, it is clear that the total deposition is dominated by wet deposition over all of the stations for both ^{131}I and ^{137}Cs . The exceptions are TOCHIGI where the dry deposition of ^{131}I contributes about half of the total deposition on 21 March and YAMAGATA where the dry deposition of ^{131}I is about 1/3 of the total deposition on 20 March.

Figure 10 examines the spatial distribution of accumulated dry and wet depositions of ^{131}I and ^{137}Cs over domain 2 from 11 March to 31 March. For ^{131}I , the area with dry deposition over 100 kBq m^{-2} is concentrated near the source and is much smaller than the area with wet deposition over 100 kBq m^{-2} . The spatial distribution of the accumulated wet deposition does not exactly follow that of the accumulated precipitation that has been shown in Fig. 6, suggesting that other factors such as wind, concentration fields, and the emission rate also play an important role in determining the distribution of wet deposition. The wet deposition of ^{131}I in the northeast area are much larger than the dry deposition; while along the east coast, the dry depositon is sometimes higher than the wet deposition. This implies that wet deposition does not necessarily dominate over dry deposition at all locations.

As for ^{137}Cs , the pattern of dry deposition is quite different from that of ^{131}I ; and most of the areas have values lower than 5 kBq m^{-2} . The reason for these differences

is that the dry deposition parameterizations depend on resistances are quite different between ^{131}I and ^{137}Cs , and the dry deposition velocity of ^{137}Cs is much smaller than that of gaseous ^{131}I . The wet deposition, on the other hand, shows a similar pattern to that of ^{131}I , but the values are slightly lower than those of ^{131}I in some areas such as north of Fukushima. These comparisons imply that the dry and wet depositions of different radionuclides are affected by wind and rainfall in different ways.

3.2.2 Sensitivity of ground deposition to the parameterizations of dry and wet deposition

To assess the sensitivity of total daily depositions to different dry and wet deposition parameterization schemes, the results from cases REF (with the resistance method for dry deposition; and with the parameterization based on precipitation rate for wet deposition), DRY2 (with the simple parameterization method for dry deposition), DRY3 (with the constant dry deposition velocity) and WET2 (with the parameterization based on relative humidity) are compared. In Fig. 11 the total daily depositions at stations IBARAKI and TOCHIGI are shown as examples. The deposition from case REF and DRY2 are nearly the same during the whole period. Except for ^{131}I at TOCHIGI, the deposition from case DRY3 are also very close to those in case REF and DRY2. The reason that different dry deposition parameterizations do not alter the total daily deposition of ^{131}I at station IBARAKI and those of ^{137}Cs at stations IBARAKI and TOCHIGI significantly is that they are dominated by wet deposition. At TOCHIGI, the dry deposition of ^{131}I contributes nearly the same to the total deposition as wet deposition, as can be seen from Fig. 9. As such, the total deposition of ^{131}I at TOCHIGI is sensitive to the dry deposition parameterizations in WRF. Nevertheless, the results from REF and DRY2 are still very close for ^{131}I at TOCHIGI, suggesting that the resistance method and the simple parameterization yield similar dry daily depositions.

As shown in Fig. 11, the total daily depositions in case WET2 (parameterization based on relative humidity) are significantly lower than those in case REF for both ^{131}I and ^{137}Cs at these two stations. For example, in TOCHIGI, the deposition of ^{137}Cs from

case WET2 is only half of that from case REF. Thus, the total deposition is more sensitive to the choice of the wet deposition scheme than to the choice of the dry deposition scheme, which is due to the fact that the total depositions at these two stations are dominated by wet depositions. However, the TOCHIGI comparison does show that the parameterizations of the two methods of deposition both have comparable influence on the results when their relative contributions are comparable.

The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different dry and wet deposition parameterizations are shown in Tables 6 and 7, respectively. The PBIAS and PRMSE in REF, DRY2 and DRY3 are quite similar, suggesting that the total daily deposition for this accident is not sensitive to the choice of the dry deposition scheme (we reiterate that this can be different for other cases where the dry deposition contributes a larger fraction of the total deposition). This is in agreement with Fig. 11. To select the “best” dry deposition scheme, the sum of global rank (SR) is compared. Case REF has the lowest SR for ^{131}I while case DRY3 has the lowest SR for ^{137}Cs , indicating that the resistance method has the best performance in capturing the total deposition of ^{131}I while the method with constant a dry deposition velocity (0.05 cm s^{-1}) has the best performance in capturing the total deposition of ^{137}Cs , which essentially precludes making any robust inferences or recommendation about the choice of the optimal dry deposition model.

The PBIAS and PRMSE in case WET2 are of the same magnitude over most of stations as those in case REF. As for ^{131}I , case REF has the lowest SR; while for ^{137}Cs , case REF and case WET2 have the same SR. These results suggest that using the wet deposition parameterization based on precipitation rate can better predict the total daily deposition of ^{131}I ; while for capturing the total daily deposition of ^{137}Cs , using the wet deposition parameterizations based on precipitation or relative humidity have a comparable performance.

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3.3 The influence of emission rates and characteristics

This section examines the sensitivity of ground deposition of ^{131}I and ^{137}Cs to the different characteristics of the emission source, including the emission rate, the gas partitioning of ^{131}I , and the size distributions of ^{137}Cs . Figure 12 shows the WRF-simulated and observed daily depositions of ^{131}I and ^{137}Cs at stations GUNMA and TOKYO. As can be seen in the figure, there are significant differences in the daily depositions of ^{131}I and ^{137}Cs at these two stations between the case using the emission rate estimated by JAEA and the case using emission rate estimated by TEPCO. The simulated depositions using the emission rate estimate by JAEA (REF) are lower than those from the case using emission rate by TEPCO (EM2) on 19–23 March when deposition occurs at these two stations. In particular, at GUNMA, the deposition of ^{131}I in EM2 is about 15 times that in REF and the deposition of ^{137}Cs in EM2 is about 4 times of that in REF. Figure 13 depicts the spatial patterns of the accumulated depositions of ^{131}I and ^{137}Cs from REF and EM2 from 11 March to 31. For ^{131}I , the area with accumulated deposition exceeding 100 kBq m^{-2} is much larger in EM2 than that in REF, covering nearly half of domain 2 over the southeast area. For ^{137}Cs , in the west of FDNPP ($37\text{--}38^\circ\text{N}$, $139.5\text{--}140.5^\circ\text{E}$), REF produces higher depositions than EM2. The above results clearly demonstrate that the emission rates, and their temporal distributions, have a major influence on ground deposition of radionuclides. Temporal variability is important since it interacts with changes in wind speed and direction to result in the concentration maps that produce the deposition.

The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different emission rates are shown in Table 8. It is evident that the PBIAS and PRMSE of the total daily depositions of ^{131}I and ^{137}Cs in case EM2 are significantly higher than those in case REF over most of the stations, indicating that case REF better reproduces the observations. This is also reflected by the lower SR value of case REF.

Other than emission rates, the gas partitioning of ^{131}I and the size distribution of ^{137}Cs are the two emission characteristics examined in our study. In the cases REF,

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two cases, which is consistent with Fig. 12. The SR value in case SD2 (= 18) is slightly lower than that in case REF (= 20), which indicates that the case with a log-normal distribution for the size of ^{137}Cs in the emission has a slightly better performance than the case with uniform particle size of ^{137}Cs .

3.4 The assessment of the sensitivity of the total daily deposition to the model physics and inputs

To assess the sensitivity of the total daily deposition to all of the model physics and inputs, the difference between the error in the reference case and that in specific sensitivity cases is calculated and compared. Table 11 shows the averaged absolute value of the difference (AAD) between the error in the reference case and that in different sensitivity cases (e.g. the AAD for PBIAS of ^{131}I between REF and EM2 is 171.63 %). AAD is defined as:

$$\text{AAD} = \left(\sum_{i=1}^n |\text{Error}_{\text{REF}} - \text{Error}_{\text{SENS}}(i)| \right) / n \quad (23)$$

where $\text{Error}_{\text{REF}}$ is the error in the reference case, $\text{Error}_{\text{SENS}}$ is the error in the specific sensitivity case, i is the index of the observational station and n is total number of the stations, here $n = 7$.

It can be seen that the AAD in terms of both PBIAS and PRMSE for both ^{131}I and ^{137}Cs is very large for REF (with emission estimated from JAEA) – EM2 (with emission estimated from TEPCO), thus the total daily deposition is very sensitive to the imposed emission rate. Based on AAD, we can also conclude that for ^{131}I , the total daily deposition is also sensitive to the microphysics schemes, the horizontal diffusion schemes, gas partitioning and wet deposition parameterizations, and the total daily deposition is not sensitive to the dry deposition parameterization; while for ^{137}Cs , the AAD for REF-MP2, REF-MP3 and REF-WET2 also has very large value, so the total daily deposition is also very sensitive to the microphysics schemes and wet deposition

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parameterizations, and it is also sensitive to the horizontal diffusion schemes and the size distribution, but it is not sensitive to the dry deposition parameterization.

4 Conclusions

This paper focuses on the atmospheric transport and ground deposition of radionuclides following the Fukushima Daiichi accident using the WRF/Chem model and observational data. The sensitivity of WRF-simulated results to a variety of parameters, including microphysics schemes, horizontal diffusion schemes, parameterizations for dry deposition and wet deposition, the emission rate, the gas partitioning of ^{131}I , and the size distribution of ^{137}Cs in the emission, is examined. The simulated meteorological fields such as wind speed, wind direction, and precipitation are validated by comparing to observations; the simulated total daily depositions are also compared to measurements. The percent bias (PBIAS) and percent mean square error (PRMSE) are used to assess the errors in the simulated results; the sum of the global rank (SR), which is based on the calculated PBIAS and PRMSE, is then used to identify the schemes that perform the best. The averaged absolute value of the difference (AADE) between the error in the reference case and that in different sensitivity cases is used to compare the sensitivity of the simulated total daily depositions to all model physics and inputs.

The main conclusions, which are linked to questions 1 to 4 that we raise in the introduction, are:

1. the wind fields are overall well reproduced by WRF (wind is usually one of the most challenging parameters to simulate successfully as discussed in (Talbot et al., 2012). The wind speed and wind direction simulated using the Smagorinsky horizontal diffusion scheme (REF) and those using the 1.5 order TKE horizontal diffusion scheme (DIF2) yield similar outputs. However, the subtle differences in the wind fields still result in a significant difference in the ground deposition of radionuclides. Based on SR, simulations using the 1.5 order TKE scheme predicted the

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ground deposition better than those using the horizontal Smagorinsky scheme. The averaged or the accumulated rainfall was fairly well captured by WRF but the maximum rainfall rate was not as accurately predicted in the sensitivity cases with three different microphysics schemes (REF: WSM6; MP2: Goddard; MP3: Thompson). The sensitivity of WRF simulated rain field to microphysics parameterization illustrates the difficulty in reproducing the spatial and temporal precipitation patterns as also concluded in previous studies (e.g. Li et al., 2013). The results demonstrated that the total daily deposition is very sensitive to the microphysics scheme and the WSM6 microphysical scheme performed the best among the three schemes.

2. The simulated total daily depositions generally agreed with the pattern observed in the measurements. But the model did not estimate the observed deposition peaks and magnitudes very well for both ^{131}I and ^{137}Cs . Wet deposition dominated over dry deposition at most of the observation stations, but not at all locations in the simulated domain. Moreover, the dry and wet depositions of different radionuclides are affected by wind and rainfall in different ways. Based on SR, the resistance model for dry deposition yields the best performance in capturing the total deposition of ^{131}I , while the model with constant dry deposition velocity (0.05 cm s^{-1}) has the best performance in capturing the total deposition of ^{137}Cs . This could imply that the resistance model is better for gaseous radionuclides and constant deposition velocity is better for particulates, but more studies are needed to confirm this pattern. Using the wet deposition parameterization based on precipitation rate can better predict the total daily deposition of ^{131}I , while using the wet deposition parameterizations based on precipitation or relative humidity have the same performance for ^{137}Cs . Again these finding could be related to differences between gaseous and particulate species.
3. The results illustrate that the total daily deposition is quite sensitive to the emission rate, whose estimates by two different studies had large discrepancies. At

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some of the stations, the gas partitioning of ^{131}I is also an important parameters that controls the total daily deposition. The total deposition of ^{137}Cs is not very sensitive to the size distribution. Based on SR, case REF (with emission estimated from JAEA) reproduced the observations more accurately than case EM2 (with emission estimated from TEPCO); the cases with gaseous fractions of 30 % or 60 % had comparable performances and can better reproduce the total deposition of ^{131}I for this particular event; the case with a log-normal distribution for the size of ^{137}Cs in the emission has a slightly better performance than the case with uniform particle size of ^{137}Cs . Based on the averaged absolute value of the difference (AAD) between the error in the reference case and that in different sensitivity cases, the total deposition is most sensitive to the emission rate for both ^{131}I and ^{137}Cs ; while it is not sensitive to the dry deposition parameterizations. Moreover, for ^{131}I , the total daily deposition is also sensitive to the microphysics schemes, the horizontal diffusion schemes, gas partitioning and wet deposition parameterizations. For ^{137}Cs , the total daily deposition is also very sensitive to the microphysics schemes and wet deposition parameterizations, and it is also sensitive to the horizontal diffusion schemes and the size distribution.

4. While the analysis allowed us to assess the important physics schemes and inputs that significantly influenced model performance and to provide conclusions about what model options and inputs seem to produce better outputs, general conclusions about the best model configuration are difficult to make due the potential error cancelation between different options and due to fact that for some cases the best configuration or input seem to vary from one station to another. Despite this inherent uncertainty, it is clear that WRF/Chem is generally able to produce realistic deposition patterns and values, and that temporal errors in the deposition partially cancel out as evidenced by the lower values of the PBIAS compared to PRMSE. Moreover, in many cases, simulations with different options bracket the observation. As such, while it seems the uncertainty in inputs and configuration precludes very high accuracy in simulations of ground deposition, ensemble

simulations with different options and a focus on accumulated deposition should prove useful in environmental impact assessments for past or potential accidents.

Finally, the current study has some limitations that the reader needs to bear in mind when using the findings in other studies. First, changes during the transport and deposition processes of the proportion of organic and inorganic forms, the gas partitioning and the particle size distributions were not considered in this study due to the limited knowledge of these processes, though they may strongly affect the transport and deposition of radionuclides. Second, a longer term assessment of the fate and transport of ^{137}Cs is not conducted in this study, but it may be required for assessing the health and ecological impacts of the radionuclides release since ^{137}Cs has a long life span. Future work involving idealized cases to examine in more detail how weather conditions affect the atmospheric transport and ground deposition of radionuclides is needed since our results confirm that slight modifications in the wind fields and precipitation can significantly influence deposition.

Appendix A

The improvement to WRF/Chem

In this paper, WRF/Chem is used to simulate the atmospheric transport and ground deposition of radionuclides. The default WRF/Chem model has no radionuclides; to implement the radionuclides into WRF/Chem, we add a new chemistry package to the registry file registry.chem to include air concentration variables (in the chem array), ground deposition variables (in the misc array) and variables related to the emissions (in the emis_ant array) of ^{131}I and ^{137}Cs . Moreover, several modules in the chem subdirectory are modified to account for new transport and deposition mechanisms. The radioactive decay process is added into the advection-diffusion solver. Dry deposition parameterizations for gaseous species are added into the module_dep_simple; while dry deposition parameterizations for particulate species are added into the

module_gocart_drydep. Wet deposition parameterizations are added into the module_wetscav_driver. The emission rates used by the simulations are imported by the program prep_chem_sources.

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Table 1. Description of WRF simulations. In the column for wet deposition, “precipitation” is short for the parameterization based on precipitation and “RH” is short for the parameterization based on relative humidity.

Simulations	Emissions	Microphysics	Horizontal diffusion scheme	Gaseous fraction of ^{131}I	Size distribution	Dry deposition	Wet deposition
REF	JAEA	WSM 6	Smagorinsky	80 %	Constant size	Resistance	Precipitation
EM2	TEPCO	WSM 6	Smagorinsky	80 %	Constant size	Resistance	Precipitation
MP2	JAEA	Goddard	Smagorinsky	80 %	Constant size	Resistance	Precipitation
MP3	JAEA	Thompson	Smagorinsky	80 %	Constant size	Resistance	Precipitation
DIF2	JAEA	WSM 6	1.5-order TKE	80 %	Constant size	Resistance	Precipitation
GP2	JAEA	WSM 6	Smagorinsky	100 %	Constant size	Resistance	Precipitation
GP3	JAEA	WSM 6	Smagorinsky	60 %	Constant size	Resistance	Precipitation
GP4	JAEA	WSM 6	Smagorinsky	30 %	Constant size	Resistance	Precipitation
GP5	JAEA	WSM 6	Smagorinsky	0 %	Constant size	Resistance	Precipitation
SD2	JAEA	WSM 6	Smagorinsky	80 %	Log-Normal	Resistance	Precipitation
DRY2	JAEA	WSM 6	Smagorinsky	80 %	Constant size	Simple	Precipitation
DRY3	JAEA	WSM 6	Smagorinsky	80 %	Constant size	Constant vd	Precipitation
WET2	JAEA	WSM 6	Smagorinsky	80 %	Constant size	Resistance	RH

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Table 2. The PBIAS and PRMSE of wind speed and MBE of wind direction with different horizontal diffusion schemes. “YA”, “CH”, “TOK”, “ON”, “NI”, “MA”, “SE” and “IS” represent the stations “YAMAGATA”, “CHIBA”, “TOKYO”, “ONAHAMA”, “NIIGATA”, “MAEBASHI”, “SENDAI” and “ISHINOMAKI”, respectively.

Errors	Cases	YA	CH	TOK	ON	NI	MA	SE	IS
PBIAS of wind speed	REF	123.68 %	4.09 %	27.68 %	15.58 %	58.03 %	56.71 %	6.72 %	4.44 %
	DIF2	127.27 %	2.43 %	26.95 %	19.27 %	56.92 %	64.18 %	10.72 %	2.79 %
PRMSE of wind speed	REF	169.87 %	50.69 %	73.11 %	62.47 %	76.42 %	92.91 %	64.65 %	56.36 %
	DIF2	175.85 %	49.64 %	72.64 %	62.65 %	75.86 %	97.78 %	63.92 %	56.57 %
MBE of wind direction	REF	49.183	28.384	32.721	34.344	24.872	36.975	43.121	30.304
	DIF2	49.953	30.888	31.358	33.602	23.465	36.236	42.172	26.655

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Table 3. The PBIAS and PRMSE of precipitation with different microphysics schemes. “YA”, “CH”, “TOK”, “ON”, “NI”, “MA”, “SE” and “IS” represent the stations “YAMAGATA”, “CHIBA”, “TOKYO”, “ONAHAMA”, “NIIGATA”, “MAEBASHI”, “SENDAI” and “ISHINOMAKI”, respectively.

Errors	Cases	YA	CH	TOK	ON	NI	MA	SE	IS
PBIAS	REF	-16.91 %	2.31 %	28.62 %	18.01 %	-19.93 %	14.48 %	-7.17 %	285.47 %
	MP2	4.87 %	35.27 %	29.96 %	12.71 %	-33.15 %	49.84 %	24.39 %	488.86 %
	MP3	-8.51 %	2.64 %	1.27 %	17.82 %	-33.47 %	39.18 %	28.08 %	282.37 %
PRMSE	REF	189.75 %	100.88 %	281.51 %	135.79 %	181.92 %	197.11 %	203.73 %	463.21 %
	MP2	156.27 %	152.74 %	297.02 %	195.70 %	177.18 %	213.27 %	198.46 %	1009.18 %
	MP3	157.29 %	118.27 %	249.68 %	148.86 %	172.26 %	156.30 %	214.75 %	413.62 %

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Table 4. The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different horizontal diffusion schemes. GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	-35.83%	-82.24%	-46.15%	-38.63%	-74.66%	-86.82%	-80.37%	12	24
	DIF2	-28.27%	-80.07%	-54.95%	-54.68%	-71.88%	-84.34%	-78.94%	9	18
PRMSE of ^{131}I	REF	235.25%	186.64%	149.33%	87.21%	151.26%	186.53%	176.16%	12	24
	DIF2	287.67%	183.28%	142.98%	134.36%	149.92%	180.90%	173.93%	9	18
PBIAS of ^{137}Cs	REF	-36.14%	-42.73%	217.64%	157.36%	65.18%	-33.93%	-4.60%	12	23
	DIF2	-51.00%	-42.71%	203.42%	96.29%	79.77%	-28.90%	-1.00%	9	19
PRMSE of ^{137}Cs	REF	55.25%	166.91%	976.58%	451.72%	368.89%	51.63%	44.44%	11	23
	DIF2	97.24%	168.42%	971.79%	316.88%	416.38%	39.23%	50.59%	10	19

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Table 5. The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different microphysics schemes. GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	-35.83 %	-82.24 %	-46.15 %	-38.63 %	-74.66 %	-86.82 %	-80.37 %	12	24
	MP2	-21.86 %	-86.61 %	-66.05 %	-8.12 %	-71.21 %	-89.51 %	-83.50 %	12	25
	MP3	-34.48 %	-89.49 %	-51.00 %	23.18 %	-82.51 %	-93.50 %	-84.56 %	18	35
PRMSE of ^{131}I	REF	235.25 %	186.64 %	149.33 %	87.21 %	151.26 %	186.53 %	176.16 %	12	24
	MP2	333.85 %	194.19 %	145.46 %	78.79 %	149.69 %	191.93 %	180.67 %	13	25
	MP3	104.20 %	199.69 %	145.16 %	172.98 %	158.46 %	200.53 %	182.33 %	17	35
PBIAS of ^{137}Cs	REF	-36.14 %	-42.73 %	217.64 %	157.36 %	65.18 %	-33.93 %	-4.60 %	10	20
	MP2	-47.63 %	-62.12 %	192.42 %	297.30 %	82.75 %	-42.35 %	-26.16 %	16	31
	MP3	7.78 %	-54.72 %	271.50 %	496.27 %	17.05 %	-73.25 %	-28.52 %	16	33
PRMSE of ^{137}Cs	REF	55.25 %	166.91 %	976.58 %	451.72 %	368.89 %	51.63 %	44.44 %	10	20
	MP2	90.12 %	169.03 %	926.31 %	762.99 %	431.13 %	78.24 %	47.61 %	15	31
	MP3	153.82 %	142.47 %	1216.28 %	1207.03 %	226.97 %	176.24 %	49.60 %	17	33

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Table 6. The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different dry deposition schemes. GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	-35.83%	-82.24%	-46.15%	-38.63%	-74.66%	-86.82%	-80.37%	7	17
	DRY2	-35.86%	-82.25%	-46.20%	-38.64%	-74.67%	-86.83%	-80.37%	13	28
	DRY3	-50.17%	-81.68%	-68.93%	-44.84%	-79.09%	-87.00%	-82.07%	21	38
PRMSE of ^{131}I	REF	235.25%	186.64%	149.33%	87.21%	151.26%	186.53%	176.16%	10	17
	DRY2	235.31%	186.66%	149.27%	87.23%	151.27%	186.54%	176.18%	15	28
	DRY3	256.54%	185.74%	147.81%	104.19%	154.73%	186.87%	178.54%	17	38
PBIAS of ^{137}Cs	REF	-36.14%	-42.73%	217.64%	157.36%	65.18%	-33.93%	-4.60%	13	29
	DRY2	-36.23%	-42.81%	216.90%	157.21%	64.95%	-33.97%	-4.68%	14	28
	DRY3	-38.87%	-43.85%	212.04%	153.19%	58.59%	-35.26%	-7.32%	15	26
PRMSE of ^{137}Cs	REF	55.25%	166.91%	976.58%	451.72%	368.89%	51.63%	44.44%	16	29
	DRY2	55.36%	166.91%	974.14%	451.39%	368.29%	51.73%	44.29%	14	28
	DRY3	59.01%	166.39%	962.51%	442.49%	352.90%	54.88%	39.92%	11	26


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Table 7. The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different wet deposition schemes. GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	-35.83%	-82.24%	-46.15%	-38.63%	-74.66%	-86.82%	-80.37%	8	17
	WET2	-24.58%	-88.60%	-63.92%	-57.09%	-75.45%	-90.99%	-81.57%	13	25
PRMSE of ^{131}I	REF	235.25%	186.64%	149.33%	87.21%	151.26%	186.53%	176.16%	9	17
	WET2	218.65%	197.94%	144.14%	154.30%	151.76%	194.80%	177.83%	12	25
PBIAS of ^{137}Cs	REF	-36.14%	-42.73%	217.64%	157.36%	65.18%	-33.93%	-4.60%	11	21
	WET2	-30.62%	-65.19%	64.45%	55.76%	86.87%	-30.18%	2.61%	10	21
PRMSE of ^{137}Cs	REF	55.25%	166.91%	976.58%	451.72%	368.89%	51.63%	44.44%	10	21
	WET2	61.11%	172.03%	445.32%	228.45%	413.59%	46.69%	56.16%	11	21

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Table 8. The PBIAS and PRMSE of total daily depositions of ^{131}I and ^{137}Cs with different emission rates. GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	−35.83 %	−82.24 %	−46.15 %	−38.63 %	−74.66 %	−86.82 %	−80.37 %	10	18
	EM2	−60.91 %	−100.00 %	−37.94 %	919.81 %	45.40 %	−87.09 %	−8.75 %	11	24
PRMSE of ^{131}I	REF	235.25 %	186.64 %	149.33 %	87.21 %	151.26 %	186.53 %	176.16 %	8	18
	EM2	199.24 %	222.73 %	160.52 %	3394.92 %	434.09 %	187.05 %	252.71 %	13	24
PBIAS of ^{137}Cs	REF	−36.14 %	−42.73 %	217.64 %	157.36 %	65.18 %	−33.93 %	−4.60 %	8	15
	EM2	−65.29 %	−80.83 %	203.64 %	775.43 %	128.29 %	−46.92 %	15.33 %	13	27
PRMSE of ^{137}Cs	REF	55.25 %	166.91 %	976.58 %	451.72 %	368.89 %	51.63 %	44.44 %	7	15
	EM2	148.36 %	192.47 %	1002.99 %	1831.08 %	594.65 %	84.75 %	115.05 %	14	27

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Table 9. The PBIAS and PRMSE of total daily depositions of ^{131}I with different gas partitioning of ^{131}I . GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMA-GATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{131}I	REF	-35.83 %	-82.24 %	-46.15 %	-38.63 %	-74.66 %	-86.82 %	-80.37 %	22	39
	GP2	-46.94 %	-93.00 %	-45.47 %	-60.04 %	-86.31 %	-95.82 %	-92.70 %	29	52
	GP3	-25.36 %	-71.83 %	-43.73 %	-17.93 %	-63.44 %	-78.05 %	-68.02 %	14	27
	GP4	7.47 %	-55.73 %	-41.23 %	14.31 %	-44.39 %	-62.40 %	-50.77 %	7	28
	GP5	-63.13 %	-90.98 %	-78.74 %	-68.79 %	-89.65 %	-94.94 %	-92.93 %	33	64
PRMSE of ^{131}I	REF	235.25 %	186.64 %	149.33 %	87.21 %	151.26 %	186.53 %	176.16 %	17	39
	GP2	232.87 %	206.90 %	136.45 %	151.78 %	163.60 %	205.84 %	196.86 %	23	52
	GP3	240.13 %	173.17 %	152.11 %	63.38 %	149.92 %	171.55 %	164.87 %	13	27
	GP4	272.59 %	167.45 %	155.46 %	143.59 %	171.14 %	157.59 %	168.63 %	21	28
	GP5	256.72 %	202.67 %	160.66 %	181.03 %	168.90 %	203.81 %	197.40 %	31	64

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Table 10. The PBIAS and PRMSE of total daily depositions of ^{137}Cs with different size distribution of ^{137}Cs . GR represents the global ranks for PBIAS or PRMSE, and SR represents for the sum of the global ranks. “YA”, “IB”, “TOC”, “GU”, “SA”, “CH” and “TOK” represent the stations “YAMAGATA”, “IBARAKI”, “TOCHIGI”, “GUNMA”, “SAITAMA”, “CHIBA” and “TOKYO”, respectively.

Errors	Cases	YA	IB	TOC	GU	SA	CH	TOK	GR	SR
PBIAS of ^{137}Cs	REF	-36.14 %	-42.73 %	217.64 %	157.36 %	65.18 %	-33.93 %	-4.60 %	12	22
	SD2	-25.92 %	-41.71 %	212.34 %	169.48 %	87.63 %	-17.51 %	2.59 %	9	18
PRMSE of ^{137}Cs	REF	55.25 %	166.91 %	976.58 %	451.72 %	368.89 %	51.63 %	44.44 %	10	22
	SD2	45.39 %	168.09 %	962.72 %	478.61 %	429.77 %	27.96 %	56.49 %	11	18

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Table 11. The averaged absolute value of the difference (AAD) between the error in the reference case and that in different sensitivity cases.

	AAD (PBIAS of ^{131}I)	AAD (PRMSE of ^{131}I)	AAD (PBIAS of ^{137}Cs)	AAD (PRMSE of ^{137}Cs)
REF-EM2	171.63 %	535.84 %	113.62 %	264.85 %
REF-MP2	11.15 %	18.56 %	34.80 %	70.08 %
REF-MP3	13.43 %	37.34 %	80.01 %	198.53 %
REF-DIF2	5.90 %	16.93 %	16.20 %	35.60 %
REF-GP2	10.99 %	21.77 %	–	–
REF-GP3	10.91 %	10.37 %	–	–
REF-GP4	30.28 %	25.06 %	–	–
REF-GP5	19.21 %	28.40 %	–	–
REF-SD2	–	–	10.68 %	21.20 %
REF-DRY2	0.02 %	0.03 %	0.20 %	0.53 %
REF-DRY3	7.17 %	6.69 %	3.46 %	7.33 %
REF-WET2	8.57 %	15.80 %	45.06 %	118.13 %

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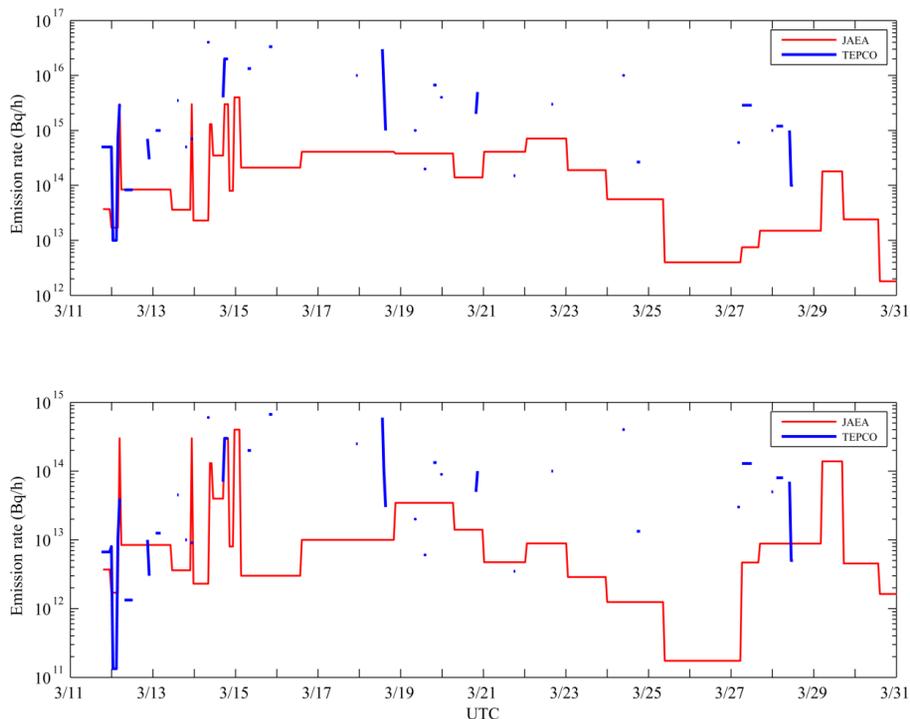


Fig. 1. The estimated emission rates of ^{131}I and ^{137}Cs . Top panel: ^{131}I . Bottom panel: ^{137}Cs . The y axis is the hourly radiological activity (Bq h^{-1}). The emission rate for TEPCO is calculated based on the release amount and duration provided by TEPCO.

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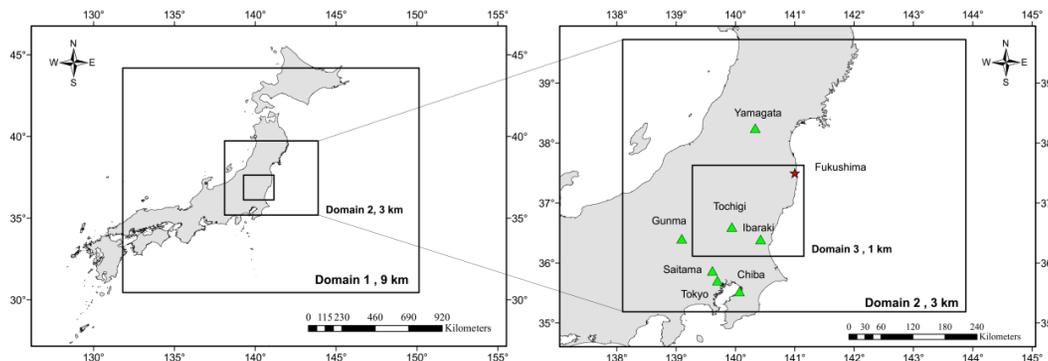


Fig. 2. The WRF domain configurations and observational stations. Left: Domain 1, 2 and 3. Right: Domain 2 and 3. The red star on the right panel represents FNDPP and the green triangles represent observational stations where deposition of radionuclides was measured (other stations are used for validation of the meteorological outputs of WRF).

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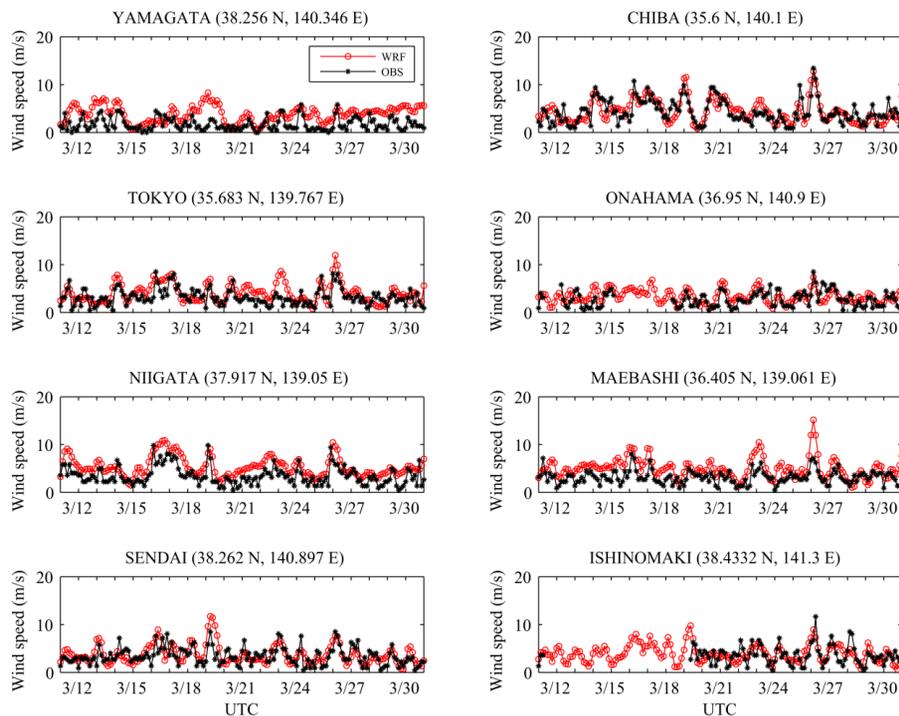


Fig. 3. Simulated and observed surface wind speeds at 8 stations over Japan during the period from 00:00 UTC 11 March to 00:00 UTC 31 March 2011 (case REF). Output from the simulation is collected every 1 h, but we only display on the figure data with 3 h resolution for clarity. Red circles represent the simulated data from WRF and the black asterisks represent the observed data.

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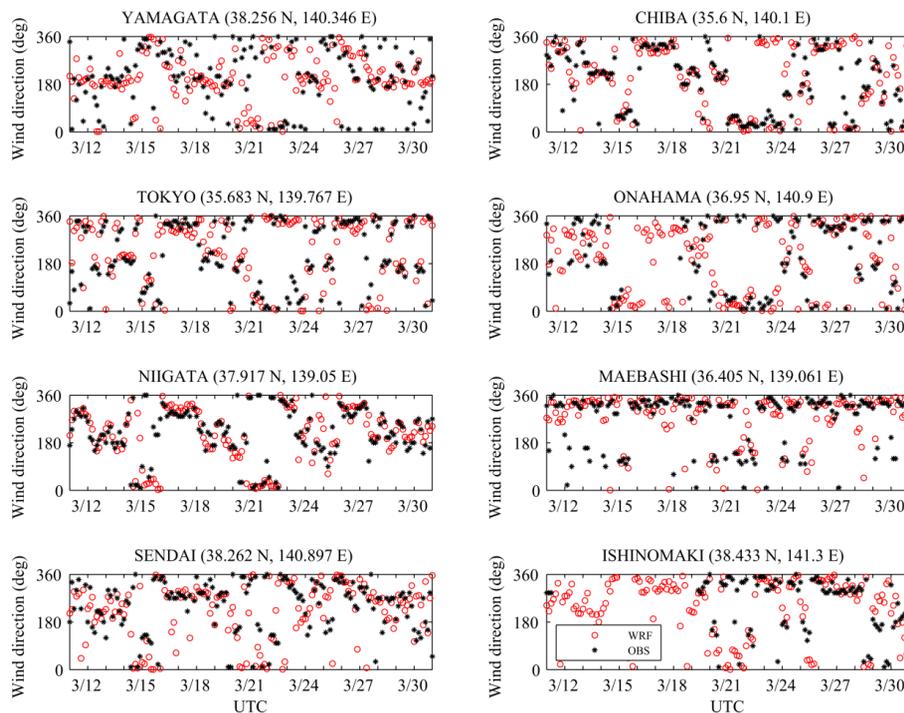


Fig. 4. Simulated and observed surface wind direction at 8 stations over Japan during the period from 00:00 UTC 11 March to 00:00 UTC 31 March 2011 (case REF). Output from the simulation is collected every 1 h, but we only display on the figure data with 3 h resolution for clarity. Red circles represent the simulated data from WRF and the black asterisks represent the observed data.

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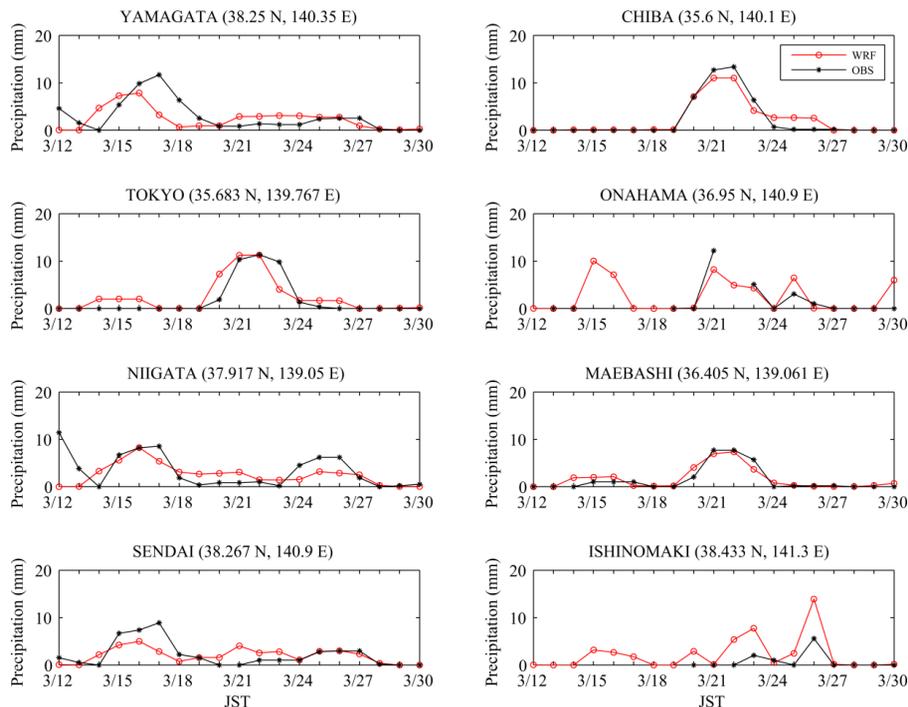


Fig. 5. Simulated and observed daily precipitation at 8 stations over Japan during the period from Japan Standard Time (JST = UTC + 9) 11 to 31 March 2011. Red circles represent the simulated data from WRF and the black asterisks represent the observed data.

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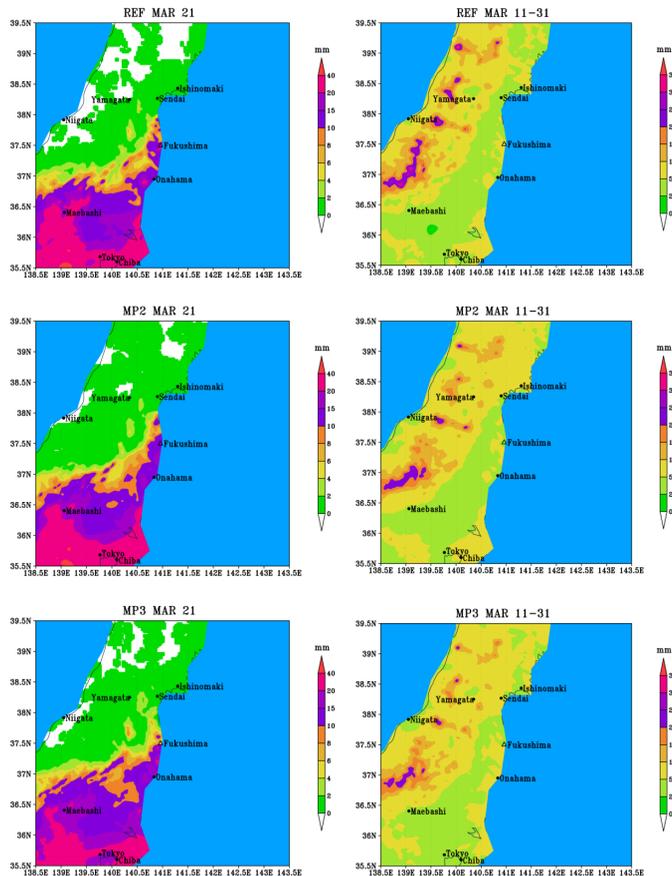


Fig. 6. The simulated precipitation over domain 2 with 3 different microphysics schemes. Microphysics schemes WSM 6, Goddard and Thompson are used in case REF, MP2 and MP3, respectively. The left column shows the daily precipitation on 21 March and the right column shows the accumulated precipitation from 11 to 31 March

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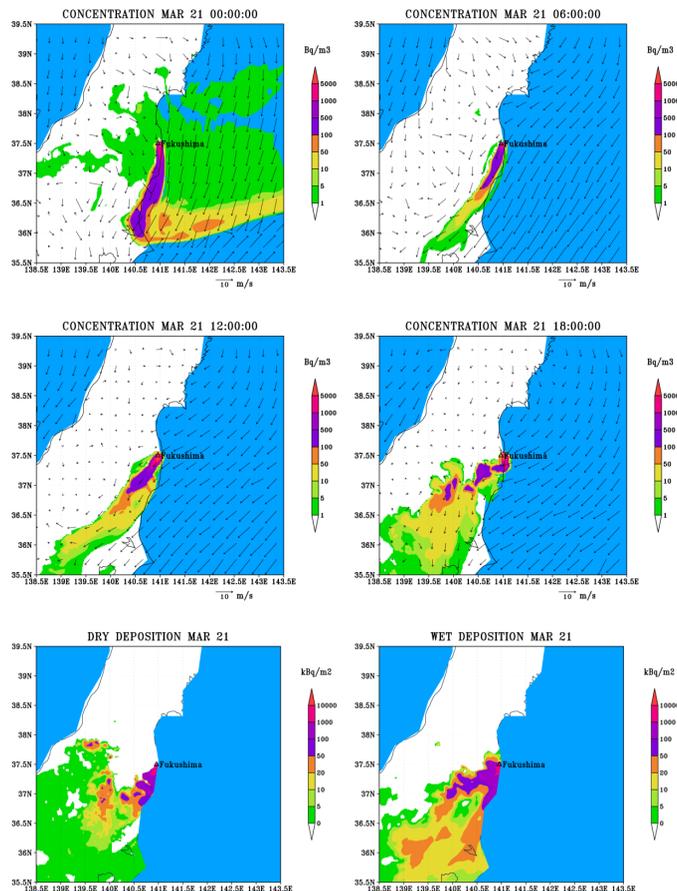


Fig. 7. The near-surface concentration and ground deposition of ^{131}I on 21 March. The near-surface concentration is represented by instantaneous value while the dry deposition and wet deposition are accumulated values on 21 March.

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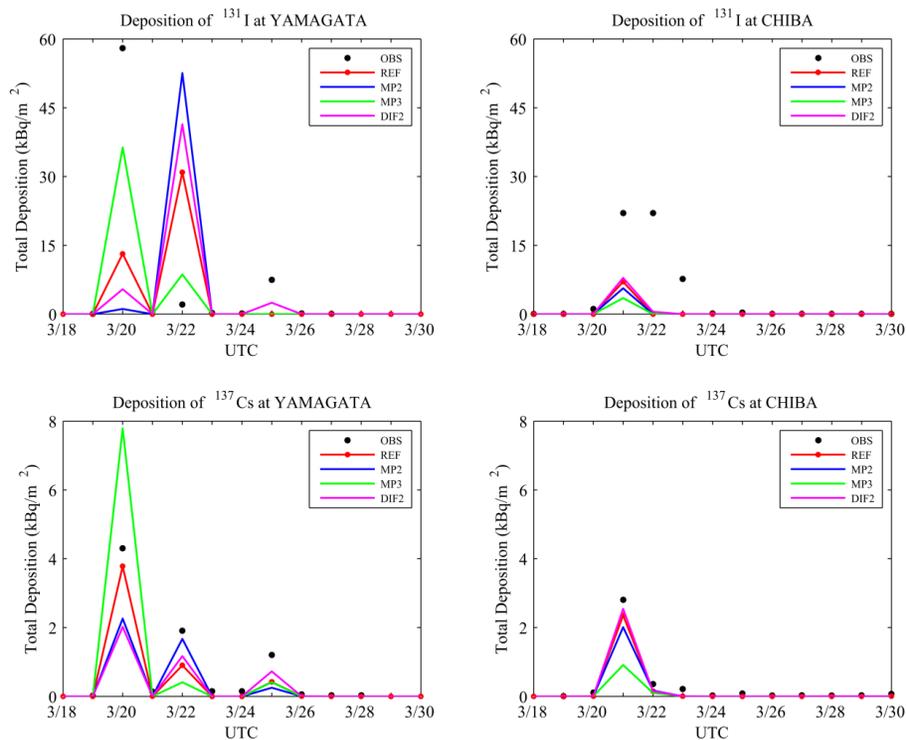


Fig. 8. Daily total depositions in station YAMAGATA and CHIBA with different horizontal diffusion and microphysics schemes.

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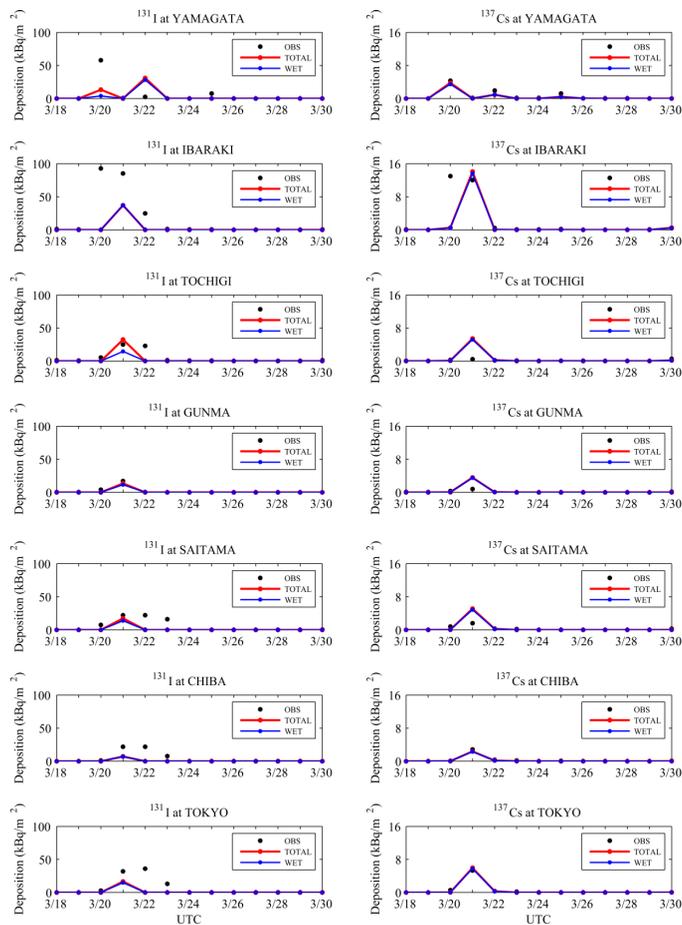


Fig. 9. The comparison between simulated (REF case) total daily depositions and the observed data of ^{131}I and ^{137}Cs .

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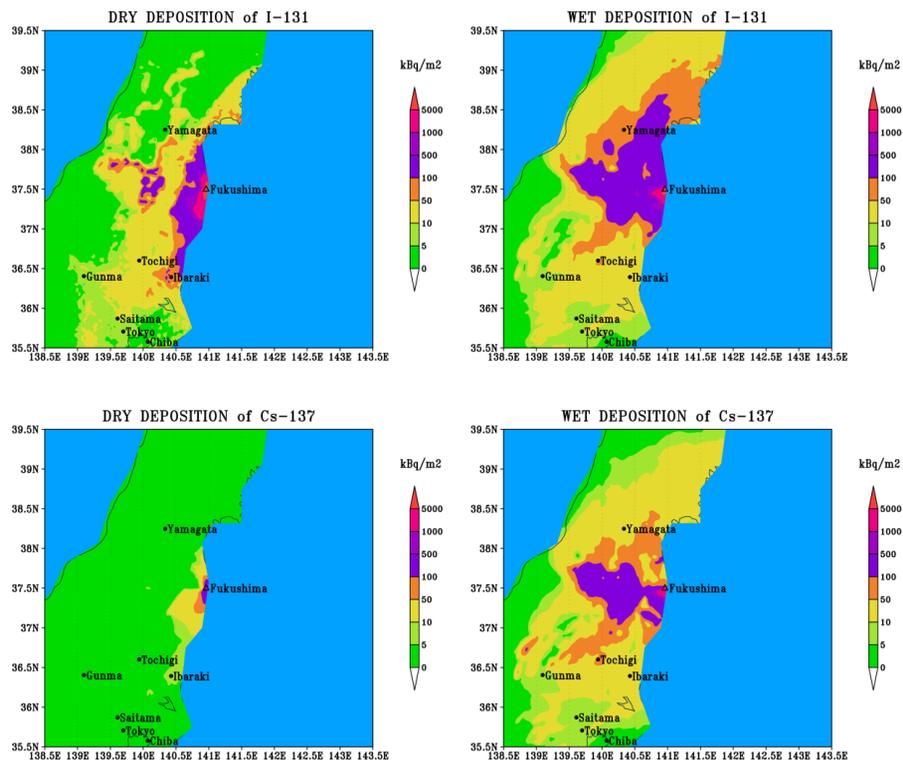


Fig. 10. Distribution of accumulated dry and wet depositions of ^{131}I and ^{137}Cs over domain 2 from 11 March to 31 March.

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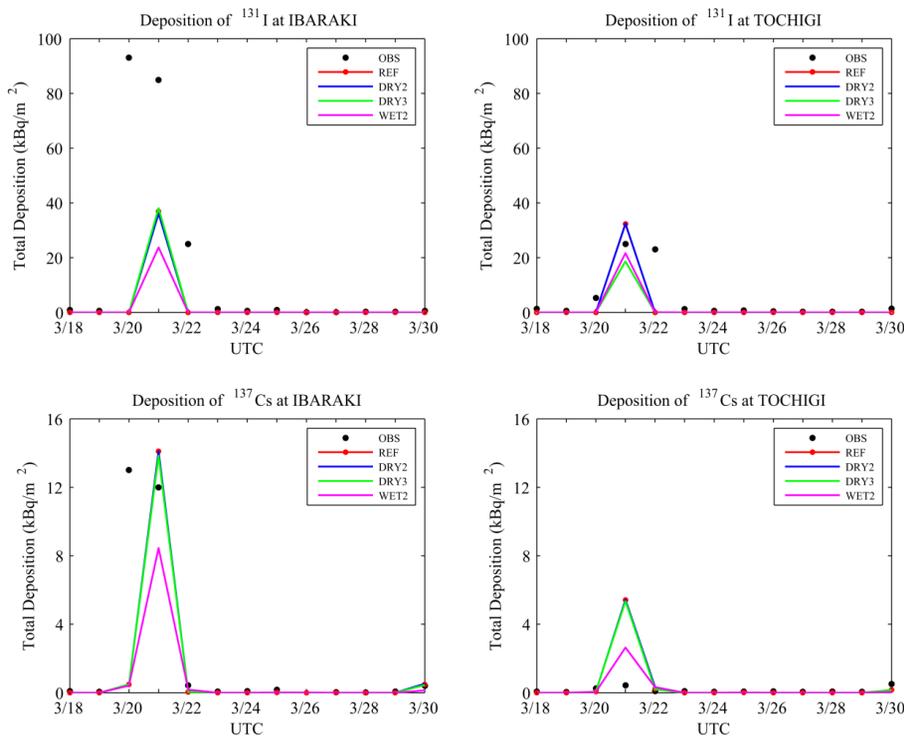


Fig. 11. Daily total depositions in station IBARAKI and TOCHIGI with different dry and wet parameterizations.

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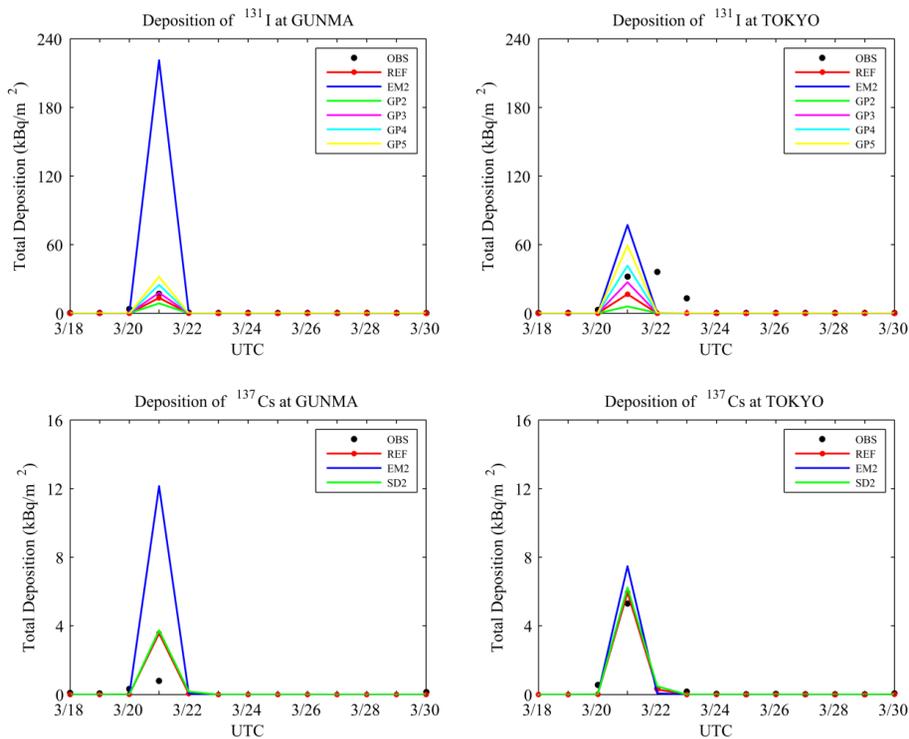


Fig. 12. Daily total depositions in station GUNMA and TOKYO with different emission rates, gas partitioning of ^{131}I and size distribution of ^{137}Cs .

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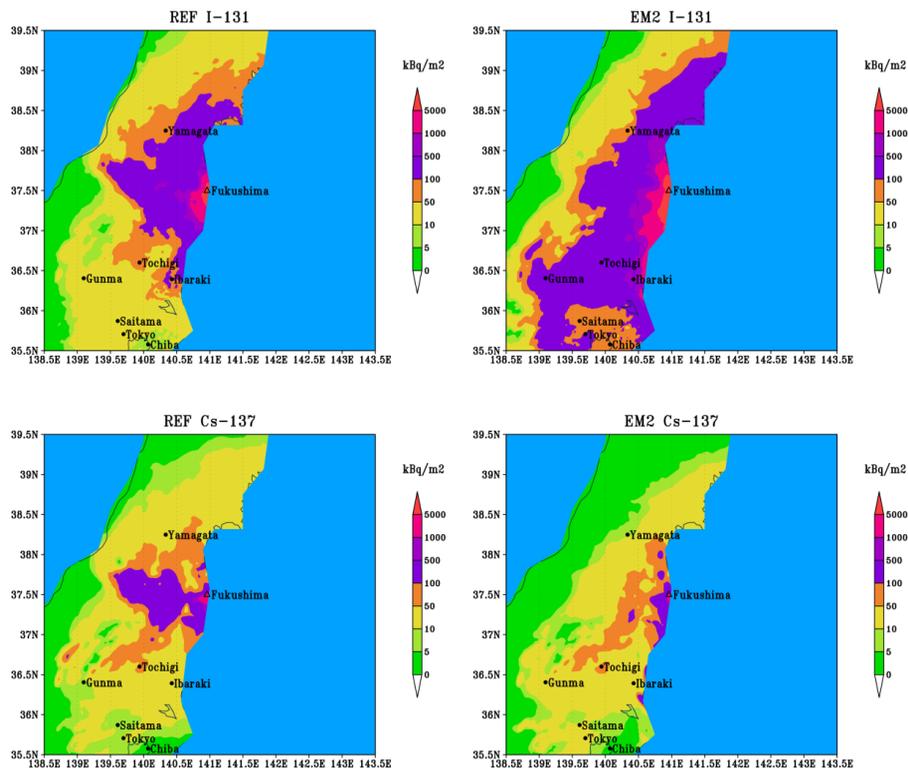


Fig. 13. Distribution of accumulated total depositions of ^{131}I and ^{137}Cs over domain 2 with different emission rates. Emission rates estimated by JAEA are used in case REF and those from TEPCO are used in case EM2.

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