Dear Editors:

As you requested, we have made all necessary changes in our manuscript (acp-2013-910) to address the reviewer's concern and have detailed how the points raised by the referees have been accommodated. From the changes made in the revised manuscript and responses provided below, I hope you are convinced that we have adequately addressed the reviewer's concern and made the paper stronger. If there are any further questions, please feel free to let me know.

Thank you very much.

Sincerely,
Xiaofeng Hu on behalf of all co-authors
2014/05/13

PS. Authors: X. Hu, D. Li, H. Huang, S. Shen, and E. Bou-Zeid

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

Number: acp-2013-910

Answer of the reviewer's questions (RC C436):

General comments:

1. The focus of the manuscript is relevant and appropriate for the Journal.

We thank the reviewer for the overall positive comments and we appreciate that the reviewer carefully reviewed our manuscript.

2. The volume of the work reported in the manuscript is larger than in normal cases.

In this paper, a variety of sensitivity simulations have been carried out to evaluate the impact of different physics/parameterizations on the atmospheric transport and ground deposition of radionuclides. As such, many figures and tables are used to discuss the sensitivity results in addition to the model evaluation results. So the length of the manuscript we believe is appropriate given the scope. Nevertheless, in the revised manuscript, we have reduced the number of tables to make the paper flow more smoothly.

3. An established model (WRF/Chem) is employed to simulate the Fukushima case. A similar model was used by other investigators, and the parameterization procedure was very similar. Detailed comments are given below in the specific comments.

As mentioned by the reviewer, Huh et al. (2012, 2013) used the WRF/Chem tracer model to simulate the transport of radionuclides from Fukushima to Taiwan, so the transport model used in their work is similar to that used in this study. However, there are also some differences with the model used in this paper.

a) The WRF/Chem model used in this paper is improved by implementing a radioactive decay term into the advection-diffusion solver and adding three parameterizations for dry deposition and two parameterizations for wet deposition. So the decay, dry and wet depositions are all considered in the upgraded WRF/Chem model, which is quite different from the tracer model (see the User's Guide of WRF/Chem v3.5).

b) The parameterizations for dry deposition and wet deposition are derived from previous studies. We aimed to examine how sensitive are the simulated ground depositions to the different parameterizations added in the model rather than establish new parameterizations for dry deposition and wet deposition of the radionuclides. So the sensitivity study is another important element of this paper, which also distinguishes it from the previous studies using WRF/Chem.

4. As the authors stated in the conclusions, this manuscript has some limitations. This is because, although they studied the atmospheric transport and deposition of radionuclides, which were highly influenced by their size, the input data of the size distribution used in this study may not be appropriate. Details are given below.

We agree with the reviewer that the size distribution used in this study has some limitations, because the size of radionuclides changes with time and is affected by the transport processes. In order to investigate how sensitive the modeled deposition is to this size distribution of ^{137}Cs, we used two different size distributions (the uniform size distribution and the log normal size distribution), which are widely used in the literature and the sizes of the radionuclides are obtained from previous studies. The input we provided to the model is the best available estimate of the size distribution and we are unaware of any better method to estimate this distribution. More importantly, our study by confirming the sensitivity of the output to size distribution in fact guides future efforts to better characterize this distribution. So we view the fact that we established this “limitation” of our study to be one of the main findings of the paper. Furthermore, changes in the particle size distributions due to transport and deposition processes were not considered in this study due to the limited knowledge of these processes. Although we agree that they may strongly affect the transport and deposition of radionuclides, there is a serious lack of literature to guide us in imposing such changes. For more details, please see the answers to specific comments 3, 4, 5, 7 and 9.

Specific comments:

1. (Abstract) The authors stated that one of the objectives of this manuscript was "to assess the skill of Weather Research and Forecasting/Chemistry (WRF/Chem) model in simulating the atmospheric transport and ground deposition of radioactive isotopes" in contrast to Srinivas et al. (2012) who tried to validate the WRF model in terms of meteorological conditions for their study and performed a statistical analysis similar to the work of the authors. Were the results

of the WRF model obtained by the authors quite different than those of Srinivas et al. (2012)?

In this paper, we use WRF/Chem to simulate the atmospheric transport of radionuclides, which directly couples the forecasting of the chemistry and meteorology. Srinivas et al. (2012) used WRF to drive the Lagrangian transport model HYSPLIT and FLEXPART (this is now clarified in the new manuscript). The configuration of WRF and some of the parameters used in these two studies are also different (e.g. the microphysics). Comparing the simulated meteorological fields from these two studies, we can see that the wind fields simulated in this paper are similar to those from Srinivas et al. (2012) as expected due to the use of the same meteorological model, but the distribution of the daily rainfall shows larger difference between these two studies (e.g. on March 21, the precipitation predicated in the paper is larger than 20 mm in the area around Tokyo, but in Srinivas et al. (2012), the precipitation is lower than 10mm in this area). The reason might be that the schemes that influence the precipitation were used differently, e.g. the cumulus parameterization is used in their study but not in our study.

2. (Abstract/Introduction) As stated, one of the distinguished features of this manuscript is the simulation the Fukushima case using the WRF/Chem model. However, Huh et al.(2012, 2013) already used the WRF/Chem model to verify the transport of radionuclides from Fukushima to Taiwan. It would be nice if the authors could add a discussion of the papers below to their manuscript.

As stated in the responses to the general comments #3, Huh et al.(2012, 2013) used the WRF/Chem tracer model to simulate the transport of radionuclides from Fukushima to Taiwan, which is different from the WRF/Chem model that we have improved. The discussion of these 2 papers is added to the manuscript.

3. (Chapter 2.3.2.c) For the constant deposition velocity method, the authors assumed $v_{\text{particle,I-131}}$ = 0.1 cm/s, corresponding to the dry deposition velocity of SO_4 suggested by Baklanov and Sorensen (2001). Using the mean size of SO_4 measured at the same sites by Kaneyasu et al. (2012), can the authors calculate $v_{\text{particle,I-131}}$ and compare it to the value of 0.1 cm/s?

In this paper, we used three different parameterizations of the dry deposition velocity and the constant deposition velocity method is the simplest one. Based on previous studies, when using the constant deposition velocity method, the dry deposition velocity is fixed as a constant value and does not change with time. Moreover, we also tested two more methods those are the resistance method and the simple parameterization as introduced in Section 2.3.2. With these methods, the dry deposition velocity changes with time and location, moreover, the dry deposition velocity is also a function of particle size as shown in Eq. (12) and (13).

$$v_{\mathrm{dep}} = u_{\mathrm{grav}} + \frac{1}{r_{\mathrm{a}} + r_{\mathrm{b}} + r_{\mathrm{a}} r_{\mathrm{b}} u_{\mathrm{grav}}} \tag{12}$$

$$u_{\mathrm{grav}} = \frac{d_{\mathrm{p}}{}^{2} g(\rho_{\mathrm{p}} - \rho) Cc}{18\nu} \tag{13}$$

Using the mean size of SO_4 measured at the same site (Tsukuba) by Kaneyasu et al. (2012), we calculate $v_{\mathrm{particle,I\text{-}131}}$ with the resistance method. Since the dry deposition velocity changes with time, we take March 11 as an example; the output interval is 1 hour. The comparison is shown in Fig. A1. From Fig. A1, we can see that the dry deposition velocity is about 0.19 cm/s before 6 am or after 9 pm, but below 0.1 cm/s and ranges from about 0.02 to 0.09 cm/s during 7 am to 8 pm. So during March 11, 2011, we can conclude that the dry deposition velocity fluctuate from 0.02 to 0.2 cm/s and the averaged dry deposition velocity of the 24 values is 0.109 cm/s, which is very close to 0.1 cm/s used in the constant deposition velocity method. Although the time variability of the velocity from the resistance method implies that the depositions predicted by the two methods could be different.

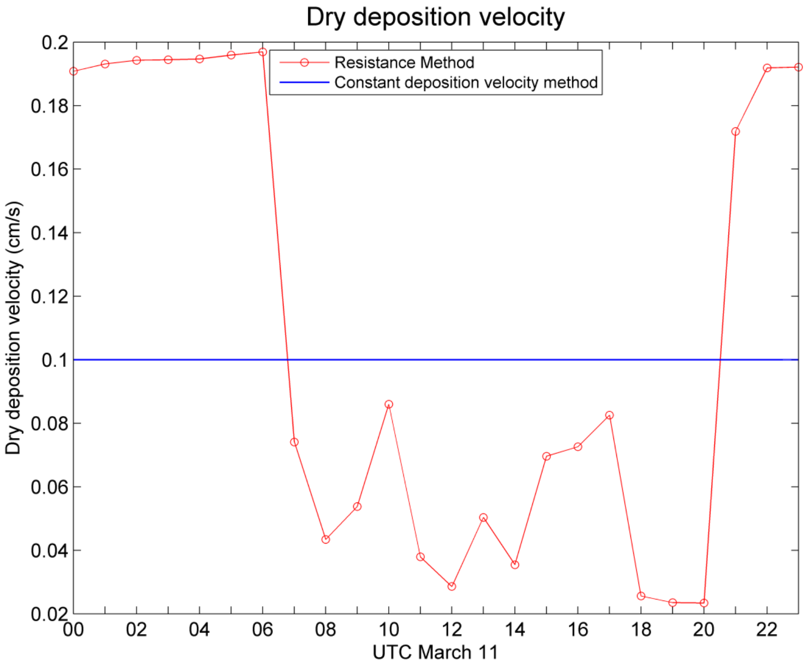

Fig. A1. Dry deposition velocity of particulate ^{131}I. The deposition velocity is calculated by using the mean size of SO_4 measured at Tsukuba with the resistance method on March 11. The red line and circle represents the deposition velocity calculated by the resistance method and the blue line represents the constant dry deposition velocity 0.1cm/s assumed in this paper.

4. (Chapter 2.4.1.) As shown in some equations (e.g., eq 13 in Chapter 2.3.2), size may be an important parameter gauging the contribution of the dry and wet deposition to the total deposition and transport. The average size used in this study was 0.48 µm (for I-131) and 0.67 µm (for Cs-137), respectively. The average size of I-131 represents activity median aerodynamic diameter (AMAD) while that of Cs-137 is the retrieved second mode. Why did the authors choose the value of the retrieved second mode as the average size of Cs-137 although its AMAD (0.53 µm) was available in the same reference?

An aim of this study is to assess how sensitive are the ground deposition to the different size distribution of radionuclides. Thus, different size distributions have been used in this paper: the first one is using the average size and the second is the log-normal size distribution. To compare the difference of the ground deposition with different size distribution, we should take both the average size and the standard deviations into account. So we chose to use 0.67 µm as the average size with the standard deviation as 1.3 µm. If we use the AMAD, no value of standard deviation is available from the reference.

5. (Chapter 3.2.1.) The authors discussed the simulation results of the dry, wet, and total depositions using observational data obtained from 3/18 to 3/30 (e.g., Figures 9, 11, and 12). Despite studying the Fukushima case, the authors used the size distribution obtained probably from the Chernobyl case (for I-131) and the data (for Cs-137) measured by Kaneyasu et al. (2012) after 6 weeks from the Fukushima accident (4/28 to 5/12). Kaneyasu et al. (2012) reported that the size distribution of Cs-137 obtained after 5/12 was different from that before 5/12. This implies that the data, quoted from Kaneyasu et al. (2012) may not represent the initial size distribution of the Fukusima case. In addition, as the authors stated in the introduction, a heavy rainfall event also occurred between 3/15 and 3/17. Could the input data used in this manuscript represent well the Fukushima case?

In order to more accurately reproduce the deposition of ^{131}I and ^{137}Cs, it should be better to use the size distribution during the emission period. However, there is no size distribution information during the period from 3/11 to 4/28 to use as input data for the simulation in this study. So we used the size distribution obtained from the Chernobyl case (for ^{131}I) and the data (for ^{137}Cs) measured by Kaneyasu et al. (2012) after 6 weeks from the Fukushima accident. Kaneyasu et al. (2012) reported that the average size of ^{137}Cs obtained before 5/12 is 0.67 μm and the average size gain after 5/12 is 0.63, which shows that the size of ^{137}Cs was measured differently during different periods but the difference is not big (6% in Kaneyasu et al. (2012)). As is known, the size distribution only affects the dry deposition rather than wet deposition (assumed in this paper), in addition, the wet deposition is dominated in the total deposition as shown in this paper, and thus, the size distribution will not affect the total deposition significantly.

A heavy rainfall event occurred between 3/15 and 3/17 over large areas in Japan. As shown in Fig.5, stations YAMAGATA, NIIGATA and SENDAI observed large precipitation during this period. As is known, it is not easy to reproduce the precipitation patterns as concluded in previous studies (e.g. Li et al., 2013). But we can see from Fig. 5 that the timing of the simulation for precipitation agrees well with the observations. We also need to stress that by investigating the sensitivity of the model to various size characteristics and emission rates, we aim to underline the importance of a better characterization of these parameters in the future.

6. (Chapter 3.2.2.) The authors simulated WET2 to analyze effects of relative humidity (RH) on

the wet deposition. Without considering hygroscopic growth of radioactive particles, is it possible to perform and discuss the sensitivity analysis of the ground deposition to the parameterizations of dry and wet deposition?

The dry and wet depositions of radionuclides depend on several important aspects, e.g. the gas/aerosols partitioning, the aerosol size distribution and the hygroscopic growth of radioactive particles. Without knowledge of these features, it is difficult to simulate the depositions by using detailed microphysical modeling. Due to the limited knowledge concerning these processes and how they affect radionuclides, we deem that adding their influence will introduce great uncertainty and might not necessarily make the model results more accurate. Thus, the changes of the radionuclides characteristics during the transport and deposition were not considered. To investigate the sensitivity in this paper, 3 different dry deposition parameterizations and 2 different wet deposition parameterizations are tested, in which the gas/aerosols partitioning and the aerosol size distribution are initialized as constant values. All of these parameterizations are taken from previous studies and with different dry or wet deposition parameterizations, some important variables may change considerably so that the ground depositions may be strongly influenced (e.g. the wet deposition rate is calculated based on different parameterizations as discussed in Section 2.3.3, and Fig.11 shows that the depositions are significantly different by using these 2 different parameterizations). Thus, it is reasonable to use these different dry and wet parameterizations to investigate the sensitivity of the ground deposition to the deposition parameterizations. To better reproduce the ground deposition, more details about the changes during the transport and deposition processes of the proportion of organic and inorganic forms, the gas partitioning, the particle size distributions and the hygroscopic growth of radioactive particles should be collected in future work, but are missing for Fukushima.

7. (Line 233) The authors used 3.5 g/cm^3 as the density of I-131. Is this a typical value? For example, Baklanov and Sorensen (2001) proposed 4.93 g/cm^3. Does this affect the simulation results of the dry deposition?

We used 3.5 g/cm^3 as the density of ^{131}I in this paper and this value is also a typical value derived for example by Ristovski (2006). As mentioned by the reviewer, Baklanov and Sorensen (2001) proposed 4.93 g/cm^3. Since the ^{131}I has many forms and the methods used to measure the density also vary, the density of ^{131}I can be quite different. In order to examine how this parameter affects the simulation results of the dry deposition, we conducted one more simulation which uses the value 4.93 g/cm^3 as the density of ^{131}I, respectively, and compared the dry deposition velocity of the particulate ^{131}I. As shown in Fig. A2, the dry deposition velocity using the density as 4.93 g/cm^3 is close to that using the density as 3.5 g/cm^3. It can be seen in equation (12) that the dry deposition velocity not only depends on the gravitational settling velocity u_{grav}, but also depends on the aerodynamic resistance r_a and the quasi-laminar layer resistance r_b, all of which are affected by the density. However, when taken together, the density of ^{131}I does not affect the simulation results of the dry deposition considerably. This point is added to Section 2.3.2 Part b. in the new manuscript.

$$v_{\text{dep}} = u_{\text{grav}} + \frac{1}{r_a + r_b + r_a r_b u_{\text{grav}}}, \tag{12}$$

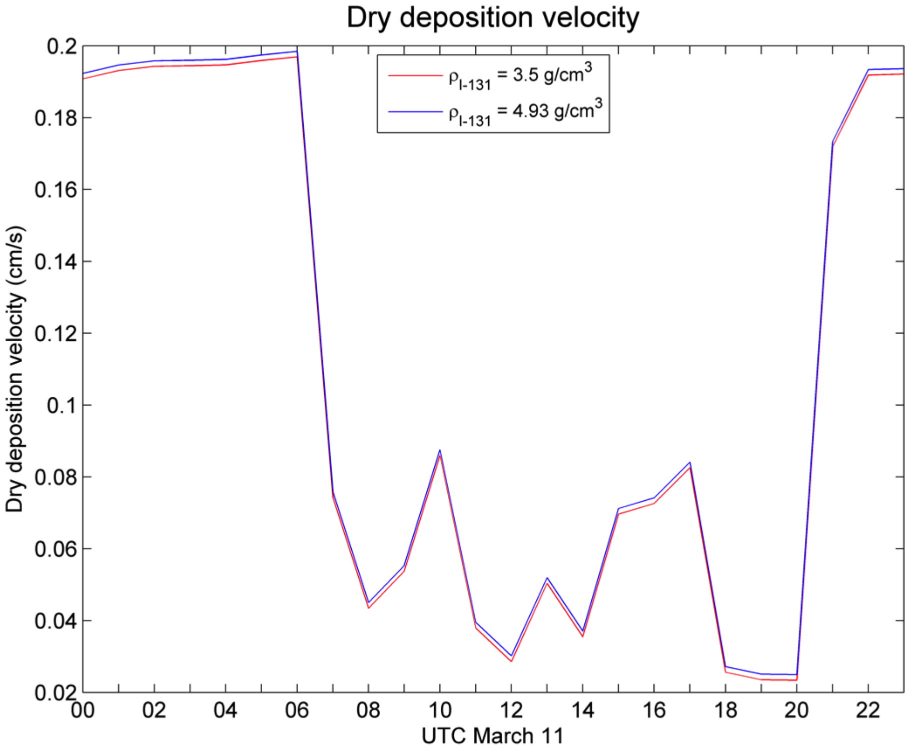

Fig. A2. Dry deposition velocity of particulate ^{131}I by using different typical value of the density. March 11 is taken as an example and the output interval is 1 hour. The red line represents the deposition velocity calculated by using the density as 4.93 g/cm^3 and the blue line represents the deposition velocity calculated by using the density as 3.5 g/cm^3.

8. (Lines 248-249) The authors wrote that "We use some typical values for of ^{131}I and ^{137}Cs that are found in the literature". Remove either "for" or "of".

   The word ‘for’ is removed.

9. (Line 250) The authors used some typical values for $v_{\mathrm{gas,I\text{-}131}}$ = 0.5 cm/s and $v_{\mathrm{particle,Cs\text{-}137}}$ = 0.05 cm/s. However, Sportisse (2007) also reported some values (e.g., $v_{\mathrm{gas,I\text{-}131}}$ = 0.1 to 0.5 cm/s; $v_{\mathrm{particle,Cs\text{-}137}}$ = 0.04 to 0.31 cm/s). It seems that the authors used the maximum value for $v_{\mathrm{gas,I\text{-}131}}$ and a near minimum value for $v_{\mathrm{particle,Cs\text{-}137}}$, which implies that the simulation results may be overestimated or underestimated. The question is how different the results would be for a different set of velocity values? In addition, these values depend on the land use coverage, where radionuclides are deposited, or chemical forms (e.g., elemental iodine or organic iodine). However, it may be hard to find some assumptions about these points. It would be nice if the authors could clearly state their assumptions with justifications.

   a) The typical values for $v_{\mathrm{gas,I\text{-}131}}$, $v_{\mathrm{particle,I\text{-}131}}$ and $v_{\mathrm{particle,Cs\text{-}137}}$ used in the sensitivity case in this study are obtained from previous studies. Sportisse (2007) also reported other values for both ^{131}I and ^{137}Cs as mentioned by the reviewer. But we did not take other values into account in this study, because our purpose is to examine how sensitive of the simulated ground deposition to the dry deposition parameterization rather than to the velocity values that are used in the constant dry deposition velocity method. The constant dry deposition velocity method is only one of the methods used in the sensitivity studies, which is compared to the resistance method and the simple parameterization. So in this study, we did not plan to find an optimal constant value for $v_{\mathrm{gas,I\text{-}131}}$, $v_{\mathrm{particle,I\text{-}131}}$ and $v_{\mathrm{particle,Cs\text{-}137}}$ since using a constant value to represent the dry deposition velocity is not the best approach as concluded from the results in this paper.

   b) Regarding how different the results would be for a different set of velocity values, we can conclude from the paper that the total ground deposition is only slightly affected by the set of velocity values. The reason being that the wet deposition is dominated over most areas in the domain. While for the dry deposition, different methods may have different results. As shown in Fig. A1, the dry deposition velocity using resistance method is not constant, thus for a specific period, the accumulated dry deposition may be different. But since we are mostly interested in the total deposition rather than just the dry deposition, we deem that another set of sensitivity simulations that are dedicated to studying different constant velocity values will not add much to our findings.

c) We agree with the viewpoints of the reviewer that it is hard to find assumptions from previous studies about the chemical forms (e.g., elemental iodine or organic iodine) of the radionuclides in the process of atmospheric transport, especially the changes of the forms with time and special locations. So it will be an important topic in future research.

10. (Line 314) In this manuscript, the abbreviation, "WSM 6" was frequently used but its full definition was not given.

‘WSM6’ represents for ‘WRF Single-Moment 6-class’. The full definition of ‘WSM 6’ is added to the manuscript.

11. (Line 320) Because of the flow of this sentence, the reviewer suggests changing the order of the references: from (Kaneyasu et al., 2012; Sportisse, 2007) to (Sportisse, 2007; Kaneyasu et al., 2012).

It is corrected based on the reviewer’s suggestion.

12. (Lines 392-394) The authors stated that "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". According to the statement, understanding the horizontal diffusion schemes may be required, but some additional explanation is needed for general readers?

In this paper, we compared different diffusion schemes. In the Smagorinsky scheme, the horizontal diffusion coefficient K is diagnosed from horizontal deformation; while in the 1.5 TKE scheme, a prognostic equation for the turbulent kinetic energy (TKE) is included, and K is calculated based on TKE.

We added an explanation to clarify the horizontal diffusion schemes used in this paper.

13. (Lines 574-577) The authors wrote that "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". It would be better if the word "both" was removed.

    The word ‘both’ is removed.

14. (Lines 696-697) In "4. Conclusions", the authors noted the difficulty of simulating the wind field using Talbot et al. (2012) which was cited for the first time in this manuscript. Would it be better if the authors discussed this in "1. Introduction"?

We agree with the viewpoint of the reviewer. We add a sentence in the Introdution Section to discuss it as suggested.

15. (Lines 705-708) This is similar to the above comment. If this part is important, would it be better if this statement and the reference, Li et al. (2013) were moved to the introduction?

We agree with the viewpoint of the reviewer. We also discuss it in the Introdution Section as suggested.