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 reviews' questions:

Reviewer 1:

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 $v_{particle,I\text{-}131}$ = 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 $v_{particle,I\text{-}131}$ 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).

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

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

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.

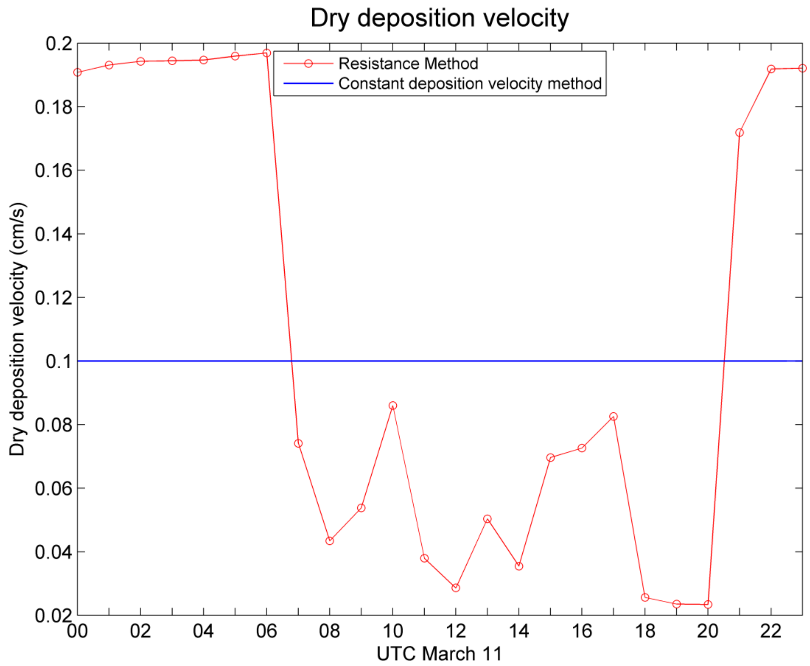

Fig. A1. Dry deposition velocity of particulate ^{131}I. The deposition velocity is calculated by using the mean size of SO_4 measured at Tsukuba with the resistance method on March 11. The red line and circle represents the deposition velocity calculated by the resistance method and the blue line represents 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.

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

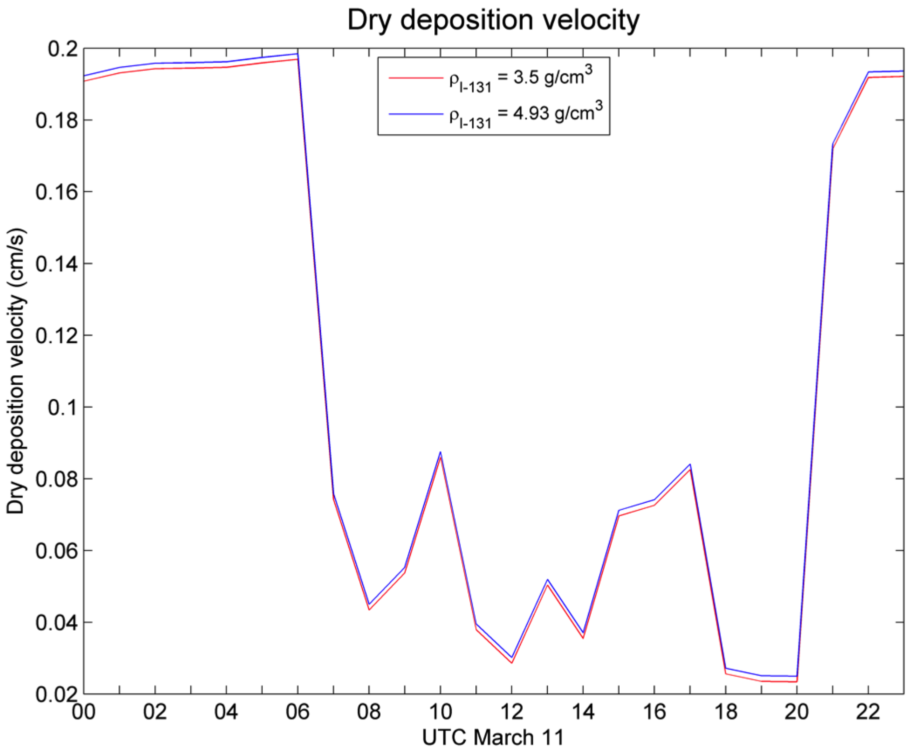

Fig. A2. Dry deposition velocity of particulate ^{131}I by using different typical value of the density. March 11 is taken as an example and the output interval is 1 hour. The red line represents the deposition velocity calculated by using the density as 4.93 g/cm^3 and the blue line represents the deposition 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 $v_{gas,I\text{-}131}$ = 0.5 cm/s and $v_{particle,Cs\text{-}137}$ = 0.05 cm/s. However, Sportisse (2007) also reported some values (e.g., $v_{gas,I\text{-}131}$ = 0.1 to 0.5 cm/s; $v_{particle,Cs\text{-}137}$ = 0.04 to 0.31 cm/s). It seems that the authors used the maximum value for $v_{gas,I\text{-}131}$ and a near minimum value for $v_{particle,Cs\text{-}137}$, 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 $v_{gas,I\text{-}131}$, $v_{particle,I\text{-}131}$ and $v_{particle,Cs\text{-}137}$ 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 $v_{gas,I\text{-}131}$, $v_{particle,I\text{-}131}$ and $v_{particle,Cs\text{-}137}$ 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.

Reviewer 2:

General comments:

1. The introductions state that an aim of the paper is to identify the combination of parameterizations that yield the lowest error in simulated ground deposition. However not all combinations of the available parameterizations are examined under the current methodology.

a) First of all, it is impossible for us to examine 'all' combinations of the available parameterizations in the WRF/Chem model. For example, there are close to 10 microphysical schemes and more than 5 planetary boundary layer schemes that are available in WRF/Chem. As such, only the important processes are tested in the paper and combinations that are constructed based on the extensive experience of co-authors Li and Bou-Zeid with WRF are evaluated, including the meteorological fields, the emission rate, and the dry and wet deposition parameterizations.

b) It was our initial aim to find an optimal set-up for WRF that can best reproduce the meteorological conditions and depositions. However, we found that the large sensitivity of WRF simulated results to a variety of physical parameterizations prevents us from reaching such a conclusion. As such, we decide to reformulate the focus of our study as investigating the sensitivity of WRF simulated results to these physical parameterizations rather than searching for the optimal set-up. By doing so, there is no need to conduct simulations with all possible combinations of physical parameterizations, which is also practically impossible.

2. Additionally, the comparisons with the observations are not clearly and concisely presented (for example, Figures 8, 9, 11 and 12 clearly show the model output for one day, March 21 but deposition values on the other days are not discernibly different from zero for most of the panels).

The simulated daily total deposition is compared with the observed data by using uniform coordinate on the y axis, which clearly shows the peak values of the daily depositions such as on March 21 within most of the panels in Figures 8, 9, 11 and 12. We also deem that the fact that the WRF/Chem model can simulate the low values is as important as the fact that WRF/Chem can simulate the peak values. Only focus on the peak values or low values would bias our analysis.

3. There are numerous tables that give results with 4-5 significant figures, which make the tables difficult to follow (particularly, Tables 2-10). Would 2-3 significant figures be adequate?

We thank the reviewer for this suggestion and we agree with this point of view. To make the tables easy to follow, we give results with 2-3 significant figures in Tables 2 -11.

4. The paper also does not give a well-developed discussion of the present results in the context of previous similar work.

We added a series of discussions about previous and similar studies to compare with the results in this manuscript.

1) In Section 3.2.1, we discuss the deposition rates modeled by Morino et al. (2011) as a comparison to this work: “As for ^{137}Cs, the total daily depositions are overestimated at TOCHIGI, GUNMA and SAITAMA and underestimated at YAMAGATA, IBARAKI and CHIBA, which is consistent with the results reported by Morino et al. (2011) using a CMAQ model coupled with WRF in their study. Morino et al. (2011) indicated that the deposition rates of ^{137}Cs at IBARAKI were underestimated, but those at the TOCHIGI, GUNMA and SAITAMA were overestimated by the model. ”
2) In Section 3.3, we discuss the results reported by Morino et al. (2013) with different emission datasets as a comparison to this work: “Morino et al. (2013) used different emission datasets in their CMAQ model simulation for the same accident and it is also reported that the emission rate estimated by TEPCO generally overestimated the observations, which agrees with the results presented in this paper.”
3) In Section 3.3, we indicate that the optimal gaseous fractions of ^{131}I lies somewhere between 30% or 60% for the model setup in this study. As suggested by the reviewer, we point out that this result is also consistent with the result from the study by Momoshima et al. (2012). So we add the reference Momoshima et al. (2012) to the discussion.
4) In Section 3.3, we indicate that the total deposition of ^{137}Cs is not very sensitive to the size distribution for the model setup in this study, which is consistent with the results from Morino et al.(2013). So we add a sentence in this part: “This is also consistent with the study by Morino et al.(2013), in which the reference case and the sensitivity case nearly have the same errors including FAC2, FAC10 and the Correlation Coefficient.”
5) Huh et al.(2012, 2013) used the WRF/Chem tracer model to simulate the transport of

radionuclides from Fukushima to Taiwan. The discussion of these 2 papers is added to the Introduction.

Specific comments:

1. Abstract: The abstract does not clearly answer each of the research questions presented in Section 1. For example, there is no indication in the abstract about the relative importance of wet versus dry deposition and their respective sensitivity to the parameterizations as outlined in question 2 (only total deposition is mentioned in the abstract). I would also prefer to see quantitative answers as opposed to phases such as 'sensitive' and 'very sensitive'.

   We added the following sentences at the end of the abstract

"The results show that the model can predict the wind fields and rainfall realistically and that the ground deposition of the radionuclides can also be captured reasonably well. The modeled precipitation is largely influenced by the microphysics schemes, while the influence of the horizontal diffusion schemes on the wind fields is subtle. However, the ground deposition of radionuclides is sensitive to both horizontal diffusion schemes and microphysical schemes. Wet deposition dominated over dry deposition at most of the observation stations, but not at all locations in the simulated domain. To assess the sensitivity of the total daily deposition to all of the model physics and inputs, the averaged absolute value of the difference (AAD) is proposed. Based on AAD, the total deposition is mainly influenced by the emission rate for both ^{131}I and ^{137}Cs; while it is not sensitive to the dry deposition parameterizations since the dry deposition is just a minor fraction of the total deposition. Moreover, for ^{131}I, the deposition is moderately sensitive (variations between 10% and 40% between different runs) to the microphysics schemes, the horizontal diffusion schemes, gas partitioning and wet deposition parameterizations. For ^{137}Cs, the deposition is very sensitive (variation exceeding 40% between different runs) to the microphysics schemes and wet deposition parameterizations, but moderately sensitive to the horizontal diffusion schemes and the size distribution."

In order to more quantitatively compare the sensitivity of the total daily deposition to all the model physics and inputs, in Section 3.4 we divide the sensitivity into 3 groups based on AAD. If

AAD > 40%, the sensitivity is defined to 'very sensitive'; if AAD > 10% and < 40%, the sensitivity is defined to 'moderately sensitive'; while if AAD < 10%, the sensitivity is defined to 'not sensitive'. We use 'very sensitive', 'moderately sensitive' and 'not sensitive' to evaluate the sensitivity of the total daily deposition to all the model physics and inputs.

2. Abstract: The total deposition has the lowest sensitivity to dry deposition parameterization. Is this attributable to the finding that the dry deposition is a minor fraction of the total deposition? If so, this should be indicated as opposed to the suggestion that the dry deposition parameterizations are associated with any greater confidence.

   We agree with the viewpoint of the reviewer.

As discussed in the specific comment 8, the dry deposition velocity using the resistance method is close to the one obtained using the simple parameterization, but is quite different from that using the constant dry deposition velocity method. Nonetheless, the total deposition by using different dry deposition parameterizations is quite similar, because total deposition is dominated by wet deposition and the dry deposition is just a minor fraction of the total deposition. We indicate this in the Abstract based on the reviewer's suggestion.

3. Abstract: It is not clear what you mean by "The ground deposition of radionuclides can also potentially be captured. . ."

By 'potentially', what we meant is that the ground deposition of radionuclides can be captured well if some key physical processes are well parameterized or some key parameterization schemes are correctly selected. We changed the sentence to be 'the ground deposition of radionuclides can also be captured reasonably well'.

4. P2116, L3: States that ". . . most of the previous studies. . . meteorological conditions are simply taken from. . . models and analysis/reanalysis products". Use of the word 'most' in this statement leaves me wondering if there were studies that did use an approach similar to this study and how these studies would compare. Again P2116, L14 states "most studies. . . the gaseous partitioning of ^{131}I was not considered". This leaves me wondering if there were studies that considered this partitioning in greater detail and how this compared to the present

study.

We removed the two ‘most’ in these two sentences. However, we still stress that to our knowledge, though some of the previous studies paid attention to the meteorological conditions, the influence of model setup parameters on the simulated meteorological fields and the influence of these fields on deposition have not yet been studied. In terms of the modeling of atmospheric transport of the radionuclides, no study to date used an approach similar to this study to examine the sensitivity of the modeled deposition to the imposed emission rates and characteristics, including the gas partitioning of ^{131}I and the size distribution of ^{137}Cs.

5. P2117, L3 gives the hypothesis that an optimal combination of parameterizations could be identified, which could reduce uncertainties in transport and deposition of radionuclides. It was not clear in the following discussion if this optimal combination was found, particularly since not all of the available combinations were examined (for example, DIF2 as in Table 1, was only ever combined with one of the microphysics parameterizations). The discussion was also not clear about whether the end result of this study was the hypothesized reduction in uncertainty.

It was our initial aim to find an optimal set-up for WRF that can best reproduce the meteorological conditions and depositions. However, we found that the large sensitivity of WRF simulated results to a variety of physical parameterizations prevents us from reaching such a conclusion. As such, we decide to reformulate the focus of our study as investigating the sensitivity of WRF simulated results to these physical parameterizations rather than searching for the optimal set-up. By doing so, there is no need to conduct simulations with all possible combinations of physical parameterizations.

Then the sentence “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” is removed in the Introduction Section.

6. P2117, L14 also states that an aim of the study is to obtain the combination of parameterizations that gives the lowest error, but this is not represented in the abstract. I think that each of the four questions in P2117, L18-26 should be given at least one sentence in the

abstract.

As clarified in the answer to the specific comment 5, we decide to reformulate the focus of our study as investigating the sensitivity of WRF simulated results to these physical parameterizations rather than searching for the optimal set-up.

We agree with the viewpoint of the reviewer that each of the four questions in the introduction section should be given at least one sentence in the abstract. So we add some sentences to the abstract as shown in the answer to the specific comment 1.

7. P2119, L4 notes that the change in the partitioning of ^{131}I between the gas and particle phase may change with time, but that this is not considered in this study. This is an important point. The gas to particle conversion for ^{131}I typically occurs on time scales from 2-3 weeks (Masson et al., 2011). This change in the partitioning over time could have a strong influence on the removal rates. The study tries several fixed partitioning values but neglect of this temporal change in the partitioning could limit how close the agreement can be with the measurements – it might be helpful to note this deficiency in the discussion related to the sensitivity studies in Section 3.3.

We agree with the point of the reviewer. As discussed in Section 3.3, the total deposition of ^{131}I is sensitive to its gas partitioning at the source. Based on that, the gas to particle conversion for ^{131}I over time may also have a strong influence on the removal rates. A sentence is added to Section 3.3 to discuss this point.

8. Section 2.3.2 discusses the dry deposition velocities. How closely do the velocities calculated by the resistance method and simple parameterization agrees with the values used in the constant deposition method?

In this paper, we used three different parameterizations of the dry deposition velocity. With resistance method and simple parameterization, the dry deposition velocity changes with time and location, but with the constant deposition velocity method, the dry deposition velocity is fixed as a constant value and does not change with time. To compare the dry deposition velocity calculated by these 3 methods, we take the particulate ^{131}I as an example. We calculate $v_{particle,I\text{-}131}$ with the resistance method, the simple parameterization and the constant deposition velocity method respectively at Tsukuba on March 11 with an output interval as 1 hour. The comparison is shown in Fig. A3. As shown in Fig. A3, the dry deposition velocity using the resistance method is significantly close to that using the simple parameterization. By using the resistance method and the simple parameterization, the dry deposition velocities fluctuate from 0.02 to 0.2 cm/s and the averaged dry deposition velocities of the 24 values are 0.109 cm/s and 0.105cm/s respectively, those are very close to 0.1 cm/s used in the constant deposition velocity method.

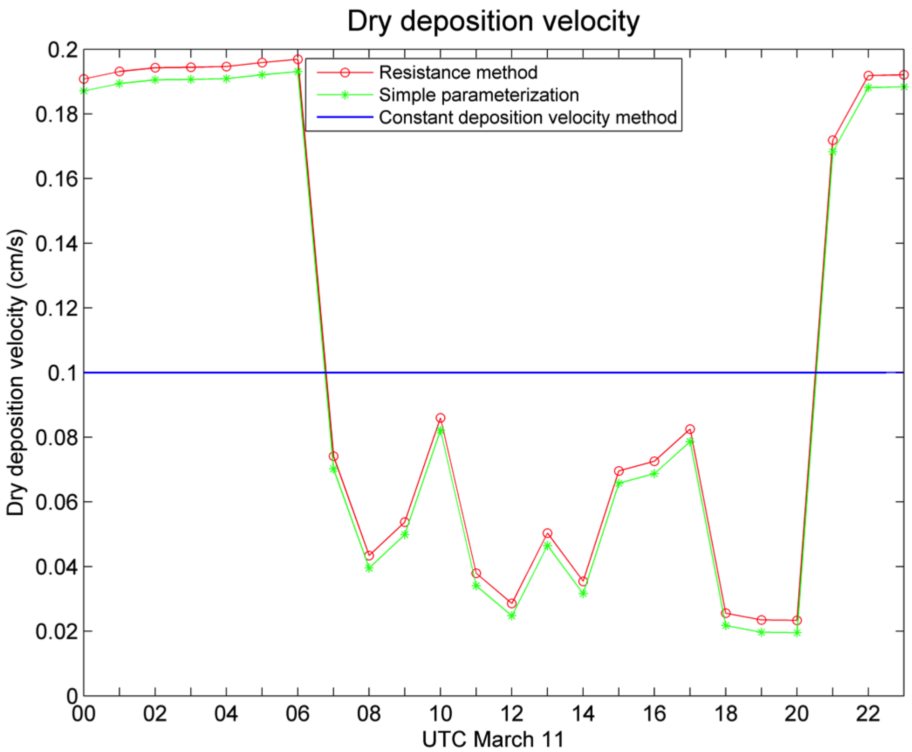

Fig. A3. Dry deposition velocity of particulate ^{131}I. The red line and circle represents the deposition velocity calculated by the resistance method, the green line and star represents the deposition velocity calculated by the simple parameterization and the blue line represents the constant dry deposition velocity 0.1cm/s assumed in this paper.

9. P2125, L10-12: How uncertain are the parameters used to represent the increased decay rates due to soil activity and how does this affect your comparisons with measurements?

In this paper, the decay rate due to soil activity is used based on IAEA (2001). As suggested by IAEA (2001), the decay rate due to soil activity λ_s of ^{131}I is 0; while that of ^{137}Cs is specified as

1.62×10^{-9} (s^{-1}), which has a half-life period as 13.6 years. In terms of the simulation period, the half-life period of the decay rate due to soil activity is significantly long so that the decay rate due to soil activity can even be neglected. Thus, in this paper, the influence of the decay rate due to soil activity on the comparisons with measurements is quite limited.

10. P2126, L8: The emissions are only released at the lowest level. How might this influence your results and conclusions? Indeed there are many confounding factors and this does prevent the authors from making a strong conclusion and creates issues with the possibility of error cancellations yielding a better result but for the wrong reasons. This was acknowledged in the conclusion but could be addressed earlier in the discussion and abstract if an optimal combination of parameterizations were to be presented.

1) As mentioned by Korsakissok et al. (2013), the release height ranges from 20m to 150m. However, in the absence of any accurate data on the vertical distribution of the emissions we opted to place them all at the lowest level. This remains a source of uncertainty that cannot be address in this study.
2) As clarified in the specific comment 5, it was our initial aim to find an optimal combination of parameterizations for WRF/Chem. But we decide to reformulate the focus of our study as investigating the sensitivity of WRF simulated results to these physical parameterizations rather than searching for the optimal set-up due to such varieties of confounding factors. So this was acknowledged in the conclusion and the text was modified to remove the sentence discussing the search for an optimal combination of parameters.

11. P2126, L22: It is not clear what you mean by ‘simple aerosol treatment’. Does the model have a bulk aerosol mass scheme as opposed to size-resolved aerosol simulation?

In this paper, the ‘simple aerosol treatment’ means that no direct or indirect effect is considered. The aerosol scheme used in this paper is similar to the GOCART simple aerosol scheme in WRF/Chem, but the size distribution for the radionuclides is defined by us in an added module in which the size of the particulate radionuclides is set to an averaged value or the log-normal size distribution.

12. P2127, L16: It is not clear what type of size distribution is assumed here. I would guess log-normal.

Yes, the size distribution assumed here is the log-normal size distribution. We add the word "log-normal" to the text here.

13. Section 2.4.2 presents the metrics used for evaluating error, comparing the model and observations. Can you explain why you chose these metrics over others such as mean fractional bias (that allows for error also in the observations) as outlined in Boylan and Russell (2006)?

In this paper, both Percentage Bias (PBIAS) and Percentage Root Mean Square Error (PRMSE) are used to compare the modeled depositions and observations. The Percentage Bias, which is similar to the Mean Fractional Bias, is widely used for evaluating model performance, and it is also reasonable to evaluate the biases of the averaged or the accumulated deposition or rainfall. Since there are usually large biases associated with the time series, PRMSE is used to capture the biases related to timing, and these timing errors are canceled by PBIAS)

14. Section 3.1: I was not clear why in searching for an optimal combination, not all combinations of the various parameterizations were examined. For example, would DIF2 combine with MP2 or MP3 give a better result? And likewise for all the various combinations of wet and dry deposition parameterizations.

As detailed in response to general comment 1, it is practically impossible to examine all combinations of parameterizations so we focus on the important and widely-used ones. The large sensitivity of WRF simulated results to a variety of physical parameterizations prevents us from searching for an optimal combination. So we reformulate the focus of our study as investigating the sensitivity of WRF simulated results to these physical parameterizations rather than searching for the optimal set-up. By doing so, there is no need to conduct simulations with all possible combinations of physical parameterizations.

15. P2133, L10-15: This discussion seems rather hand-waving. Are you able to provide model

results that show that there are differences in the turbulence, upstream winds or precipitation in response to these small differences in the wind fields?

As discussed in this section, small differences in wind fields may generate large differences in precipitation and thus influence wet deposition. The wind speed and wind direction are similar for the two horizontal diffusion schemes, however, the turbulence, upstream winds and the precipitation may be different. We take YAMAGATA as an example. The daily deposition, rainfall and the wind fields with different horizontal diffusion schemes are shown in Fig. A4.

As shown in Fig. A4, the wind fields with the two horizontal diffusion schemes are quite similar to each other, however, the rainfall generated in the two cases are different, especially on the date March 22, 23 and 25. The depositions in the two cases are also quite different since those are considerably influenced by the rainfall as discussed in this paper.

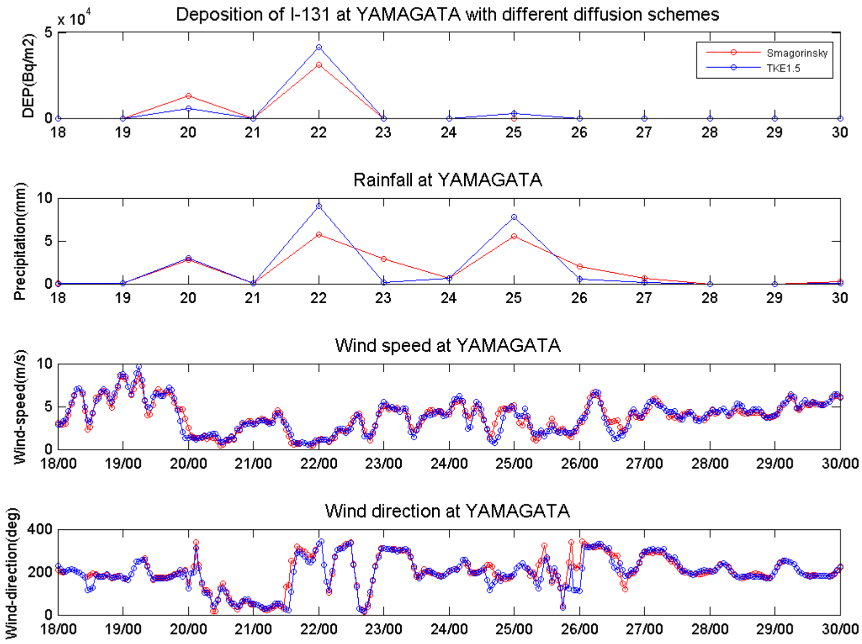

Fig. A4. Daily deposition of ^{131}I, rainfall and wind fields with different horizontal diffusion schemes in the station YAMAGATA. The red line and circle represents the variable with the Smagorinsky scheme; while the blue line and circle represents the variable with the 1.5 order TKE scheme.

16. P2134, L9-10: Case REF is noted to have the lowest global rank and it is suggested that the microphysics scheme WSM6 is superior to the other two schemes. However this is only tested

for one combination of wet and dry deposition parameterizations. Could the result be different with different combinations of deposition parameterizations and microphysics schemes?

As concluded in this paper, the ground depositions are both sensitive to the wet deposition parameterizations and the microphysics schemes. So the result may be different with different combinations of deposition parameterizations and microphysics schemes. However, as answered to the general comment 1, we changed the focus of our study as to investigate the sensitivity of WRF simulated results to these physical parameterizations rather than to search for the optimal set-up. So we did not test all possible combinations of physical parameterizations, which is also practically impossible.

17. Section 3.2.2: How was the one additional wet deposition parameterization chosen and why was this one chosen?

As introduced in Section 2.3.3, we used 2 different wet deposition parameterizations; the one used in the reference case is based on the rain intensity and the additional one used in the sensitivity case WET2 is based on the relative humidity, RH (Pudykiewicz, 1989). As discussed in Pudykiewicz (1989), rain intensity is usually highly uncertain, so the wet deposition based on the rain intensity should take the uncertainty of the rain intensity into account. While the additional wet deposition parameterization is much simpler and only need to use RH to calculate the wet deposition rate. To investigate how sensitive are the modeled ground depositions to different wet deposition parameterizations, the parameterization based on RH is chosen to compare with that based on the precipitation.

18. Would a log scale on the y axis of several of the figures (such as Figures 8, 9, 11, and 12) help with visualization of the deposition for days other than March 21.

As discussed in the general comments part, it is reasonable to see the modeled deposition is extremely close to zero on these days, which is also consistent with the observations. In terms of validation of the model, we deem that the linear y-axis is better since it shows the peak and the low values distinctly.

19. There are a limited number of stations available for measurement data. Did you include all available deposition data for Japan such as in Hirose (2012)?

We agree with that the measurement data is very limited. In this paper, we include most of available deposition data for Japan; though only 7 stations are selected (most of the stations do not have available data covering the period from March 18 to March 31 and all of the 46 stations do not have available data before March 18 as introduced in Hirose (2012).

20. P2139, L20 suggested that the optimal fraction of gaseous ^{131}I is 30-60%. However this is should be clarified as being only for this model setup. Additionally, this fraction is in agreement with the work of Momoshima et al. (2012), which could be added as a reference here.

We agree with the viewpoint of the reviewer and thank him/her for the suggestion on adding the reference. We added a sentence to clarify that the optimal fraction of gaseous ^{131}I is only for this model setup and added the reference Momoshima et al. (2012) to the discussion.

21. P2139, L20. In regard to the log-normal size distribution for ^{137}Cs, it is not clear in the text how the size distributions interact with the deposition parameterizations or other parameterizations to yield changes in the deposition. Please expand on how this is represented in the model.

The log-normal size distribution for ^{137}Cs is assumed in the sensitivity case SD2, in which the average size is 0.67 μm (the same to case REF) and the standard deviation is set to 1.3 μm. The size of the particle influences the dry deposition velocity as described by the equation (12) and (13):

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

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

However, the results show that the difference of total deposition between REF and SD2 is small; moreover, the averaged absolute value of the difference (AAD) for PBIAS of ^{137}Cs between REF and EM2 is only 10.68%. So the total deposition of ^{137}Cs is not very sensitive to the size distribution from the comparisons at these 7 stations. This conclusion is also consistent with the study by Morino et al.(2013), in which the reference case and the sensitivity case nearly have the same errors including FAC2, FAC10 and the Correlation Coefficient.

22. In general, could the results section include more discussion to put this work and these findings in context of previous and similar work as also noted by the first referee?

Yes, we agree with the viewpoint of the reviewer and we add a series of discussions about previous and similar studies to compare with the results in this manuscript. For details, please see the General comment 4.

23. P2141, L25-26. The text uses the words ‘subtle’ and significant’ where more quantitative discussion would be helpful.

At the end of this sentence, to make the discussion more quantitatively, we add an example: the AAD for PRMSE of wind speed between REF and DIF2 is only 1.76% calculated based on Table 2 and Equation 23, however, the AAD for PRMSE of the deposition of ^{131}I is 17% and that of ^{137}Cs is 36% as shown in Table 11.

24. Tables in general: a reduction in the number of significant figures is suggested.

We thank the reviewer for this suggestion and we agree with this point of view. To make the tables easy to follow, we give results with 2-3 significant figures in Tables 2 -11.

25. Figure 6: difference contours for the lower 4 panels might help to display the comparison better.

Based on the reviewer’s suggestion, we add 2 black circles in Fig.6 to show the area in case MP2 and MP3 we compared in the text.

26. Figure 7: Caption should state the simulation presented here and likewise for Fig.10.

The caption of Figure 7 and Figure 10 are rewritten as “Figure 7. The near-surface concentration and ground deposition of ^{131}I on March 21. The upper four panels show the distribution of concentration of ^{131}I at the lowest level of the atmospheric model at four different times (i.e., 00, 06, 12, 18 UTC) on March 21, in which the near-surface concentration is represented by instantaneous value. The bottom panels of Fig. 7 shows the accumulated daily dry and wet deposition on March 21, in which the dry deposition and wet deposition are accumulated values during this day. The results are from the simulation REF." and "Fig. 10. Distribution of accumulated dry and wet depositions of ^{131}I and ^{137}Cs over domain 2 in the reference case (REF) from March 11 to March 31. The upper panels show the accumulated dry and wet deposition of ^{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 lower panels show the accumulated dry and wet deposition of ^{137}Cs, the pattern of dry deposition is quite different from that of ^{131}I; and most of the areas have values lower than 5kBq/m^2.".

Technical Corrections:

1) P2135, L2: remove word 'see' before Fig. 1

The word 'see' before Fig. 1 is removed.

2) P2141, L10: change 'are validated' to 'are evaluated'

The word 'validated' is changed to 'evaluated'.

3) P2141, L15: Change acronym from 'AADE' to 'AAD' to be consistent with previous text

The acronym 'AADE' is changed to 'AAD'.

4) P2141, L22, remove brackets around Talbot et al., 2012

This part is moved to the introduction Section. "As studied by Talbot et al. (2012), wind is usually one of the most challenging parameters to simulate successfully; according to Li et al. (2013), it is also difficult to reproduce the spatial and temporal precipitation patterns by using numerical models."