Title: Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of atmospheric dispersion model with improved deposition scheme and oceanic dispersion model

Authors: G. Katata et al.

MS No.: acp-2014-330

Author response to reviewer comments

Response to Ref.1

The authors present an ambitious article on the analysis of the Fukushima accident in the light of all the work that has been done on the subject. The paper addresses various topics: source term estimation and analysis of the release events; verification of the source term at local, regional and global scales. They partly raise the issue of the sensitivity to the atmospheric dispersion model, to the meteorological data and to the deposition parameterizations. The analysis of the accident (impact of the release events, connection between the release events and the events that occurred in the plant, comparisons with observations) is highly comprehensive. The supplement of the paper is also very useful.

➔ We appreciate your careful reading and giving positive comments on our paper. We tried to respond to suggestions in detail as much as possible, but we sometimes could not bring the whole sentences from the manuscript to this letter because they are too long to cite in the letter. We showed the chapter, section, or subsection number, so please also see the revised manuscript.

The uncertainties associated with the source term estimations due to the assessment method and to the meteorological data need sometimes to be refined and reminded in the text (section 3.1 in particular). The part concerning the importance of improving the modeling of the deposit is less convincing and need to be further developed (without increase the paper size) or addressed more fully in another paper. Given the current state of the study, the authors have to be much less clear-cut on the advantage to use a more sophisticated deposition scheme.

➔ As replied to all of your comments below, we revised the manuscript to solve above concerns of yours within a shorter paper size. We hope the advantage of the new source term and deposition scheme is much clear in the revised version.

The paper is very long. The authors can probably shortening some parts. Some figures may be moved to the supplement of the paper. Some suggestions are given.

➔ On the basis of all reviewer comments, we made the following revisions to reduce the paper length summarized as:

- In Introduction, the sentence after 'First, …' of ln.5/ P.14730 was shorten.
- Chapters 3 and 4 are reconstructed. 3.1.1-3.1.9 which describes the source term and the motion of plume during March was revised and only the periods which were different from the previous paper (Terada et al.

2012) were remained. Related this revision, Figs. 5 and 6 were deleted. As for chapter 4 (Discussion), section 3.2 (Verification of source term) was moved to section 4.1. Section 4.1 (Comparison of source terms) was moved to section 4.2. Finally, section 4.2 (Role in deposition process) was moved to Supplement.

- The section for "Validation using several models" was shortened. Related this revision, (old) Figs. 18, 20, and 22 and Tables 7, 8, and 10 were extracted.
- Conclusion was also shortened based on reviewer's comments.

At the same time, we needed to include more sentences to reflect all reviewers' comments to the manuscript (e.g., Meanwhile, Fig. S3 was moved to the main text as (new) Fig. 12). However, the page numbers and numbers of figures and tables were still reduced compared with our previous paper.

MAJOR COMMENTS

Section 2.2: Reverse estimation method over the land

P14735 What do the authors mean when they explain that they used peaks values from continuous time series of air concentration? Why is it better to do so than to use the full set of data?

➔ The comparison of peak values is carried out for JAEA-Tokai data, because time series data are only available at this place. The air concentration data from JAEA-Tokai is one hour averaged one. When we use the reverse estimation method, "Since the uncertainty of model simulation is the primary cause of the discrepancy in the spatiotemporal distribution of plume between the measurements and simulation results, the above procedures cannot be applied systematically, and the correction of this discrepancy by 'expert judgment' is necessary to reduce the impact of model uncertainty on the source estimation. The process is to check all available measurements to see if the plume is reproduced appropriately or not for comparison with the measurements, and to determine if the discrepancy is caused mainly by errors in the calculated wind direction. If the plume flow direction is clearly different from the measured wind direction, the calculated plume is rotated to match the measured wind direction and Eq. (1) is applied. The use of peak values corrects any discrepancy in the timing of the arrival of the peak air concentrations between the measurement (JAEA-Tokai) and simulation. We assume that the peak values of the measurement and simulation are comparable even though the timing or temporal pattern of the arrival of the peak is different because the central plume axis passes across the sampling point differently between the measurement and simulation" (new subsection 2.1.1).

P14735 With dose rate measurements, only the observed air dose rate from ground shine is used to assess the source term. If so, how the timing of releases (beginning and end) is estimated?

➔ In this study, we divided the release interval and looked for when and where the specific segment of plume increased air dose rate, and then found the appropriate observation data which can be used for the source term estimation for the certain plume. Thus, the timing of releases should be clear. The ground-shine was used for the source estimation on 12 and 15-16 March. For the case of 12 March, the release period for wet venting of Unit 1 was determined from the decrease of pressure of drywell and the release by hydrogen explosion is assumed almost instantaneous. For the case of 15-16 March, the release period was basically divided with every hour and the source term for each plume was estimated by the method as mentioned above. The descriptions how to find the appropriate observation and how to determine the release period of the segment of plume are available in (new) subsection 2.1.1 and 2.3.3.

The isotopic composition is assumed to assess the source term by using observed dose rate from ground shine. Release rate for noble gazes is not assessed and computed dose rate signal does not take into account the contribution of noble gazes to the plume dose rate. Nevertheless Chiba observations showed that for some release events a large part of the total dose rate was due to the noble gazes contribution when the plume was detected. How do the authors interpret the comparisons between total air dose rate (including plume contribution) simulated and observed shown Figures 16 and in the supplements? What is the impact of their assumption on the source term assessment and its evaluation?

➔ In the aspect of source term estimation, we did not consider the effect of noble gases because we used ground-shine for our source estimation from air dose rates. For validation, (new) Fig.11a showed the comparison of air dose rates of 18 March when the plume flowed toward the Pacific Ocean. Hence, the air dose rates over the land should originate from only the ground-shine of deposited materials except for noble gases. The comparison for validation in (new) Figs. 6a and b, and Fig. 12 was done at a slow decrease in the dose rates after the peak (ground-shine). These points are added to sections 3.1 and 3.4, and subsection 4.1.1.

<u>Section 2.3: inverse estimation method over the ocean</u>

Except if I missed something, the method used to assess the source term over the ocean is not based on inverse modelling techniques. To avoid any confusion, the authors should not call their method "inverse estimation method".

➔ According to the reviewer's comment, we changed the description for the method used to assess the source term over the ocean from "inverse" to "reverse". Also, at the first part of (new) chapter 2, we defined the reverse and inverse methods with merit and demerit, and also described why we choose the reverse method for the FNPS1 accident case as "A reverse method evaluates the release rates of radionuclides by comparing measurements of air concentration of a radionuclide or dose rate in the environment with calculated one by atmospheric transport, dispersion and deposition models (ATDM) for a unit release of a radionuclide. The release rate is estimated by the ratio of the measurement to calculation result. The merit of the reverse method is that the comparison can be made with one or more independent data points. For example, the minimum number of data points needed is only one and the measured data used for the estimation can change with time from air concentration to air dose rate and vice versa. The demerit is that this simple comparison without consideration of the uncertainty of the ATDM results may cause the large errors, and, consequently, expert judgment is essential to correct the discrepancy between the measurement and calculation."

This part of the paper has to be improved: the goal and the method have to be clarified.

➔ The goal and method were clarified in (new) subsection 2.1.2. Also, the suffix and equations are revised according to the comments from other reviewers.

Section 3.1 Source term estimation and local-scale dispersion analysis

The reliability and uncertainties of the meteorological data should be given for the various release events.

➔ Although we realize the importance of the evaluation of the reliability and uncertainties of meteorological data for various events, the detailed analysis on this matter needs more space in paper and should be considered in future work. In the revised paper, we partially discussed this issue in subsection 4.1.1, Conclusions, and Supplement.

What monitoring data are used to reconstruct each release events? Their number has to be given (it could be given Table 5). The relevance of the various emissions has to be discussed. This requirement is at least needed for the release events showing the main discrepancies between the actual study and the previous one.

➔ We revised (new) Table 6, in which the monitoring data used for estimation is shown as monitoring locations in (new) Figs. 2 and 3. Also, in sections 3.1-3.5, we described which data are effective to determine the new source term.

What are the specific reasons for the new release assessment (especially on March 15-16)? This point is partially discussed Section 4.1 and need to be completed.

➔ "The events in the reactors (TEPCO, 2011a, 2012; Tanabe, 2012) are also shown in Fig. 5, but it is not clear from the reverse estimation that the events written in Fig. 5 mainly caused the atmospheric releases, particularly after 15 March" (the first paragraph of new chapter 3). Meanwhile, it is clear that the new release assessment on 15-16 March is achieved by both new observation data, e.g., the monitoring post data from Fukushima Prefecture and the improvement of deposition scheme (new section 3.4 and subsection 4.1.1). By this finding, the overestimation of deposition in the south area of Miyagi Prefecture in the previous work was clearly improved when using the new source term (new subsection 4.1.1).

Section 3.2

The main release events that are different from the previous study could be analyzed in more details. Does it give a better agreement by comparing simulations with dose rate measurements and/or air concentrations measurements? Monitoring dose rate comparisons shown in the supplements may be used to explain the impact of the new source term compared to the previous one.

➔ Considering the reviewer's comment, we concentrated on the analysis of release events that are different from the previous study in chapter 3. In particular, for the releases from 15-16 March, we show the comparison in air dose rate at selected monitoring sites affected by large deposition events (Hirono, Kawauchi, Fukushima, Iitate, Kawafusa (NW of FNPS1), and Yamada) in the maintext (new Fig. 12). We hope now the improvement due to new source term is much clear.

Regional deposition

Authors claim that "both improvements resulted from the enhancement of the scavenging coefficient by including in-cloud scavenging in the modified wet deposition scheme" the demonstration is not conclusive and it is difficult to precisely identify what is the specific contribution of the new deposition scheme. The authors should compare simulations done with the new source term and the previous deposition scheme with simulations done with the new configuration and source term. Those simulations could be compared in the various tables.

➔ Considering the comments from all reviewers, we carried out four cases of simulation in terms of deposition distribution of Cs-137 using the combinations of (1) old source term (Terada et al., 2012) and original WSPEEDI-II, (2) new source term and original WSPEEDI-II, (3) old source term and modified WSPEEDI-II and (4) new source term and modified WSPEEDI-II. By comparing the results from (1) and (3), the effect of model improvement on deposition distribution was examined and then, by comparing (3) with (4), the effect of source term improvement to the distribution was investigated. The results were described in subsection 4.1.1.

Moreover, it seems surprising that the in cloud scavenging has a large impact. Indeed, the plume was probably situated in the lower layers of the atmosphere at the regional scale and below cloud scavenging may have been dominant.

➔ We did not consider below-cloud scavenging in the previous manuscript because it was found to have much less significant impact as mentioned in many prior studies (A6 in (old) Appendix). Even though it was less significant, however, we still needed to show that the contribution of below-cloud scavenging to the total wet deposition. In the current simulation, we computed the below-cloud scavenging process and estimated its contribution to the total wet deposition as the difference between the modeled depositions with and without below-cloud scavenging divided by that with below-cloud scavenging (%): $(D_{with\_bl} - D_{wo\_bl}) / D_{with\_bl} \times 100$. The wet deposition amount due to the below-cloud scavenging was highest in the northwest vicinities of the power plant, 10 kBq m^{-2}. We investigated the reason why the contribution of below cloud scavenging was such low in the northwest region by drawing horizontal-vertical cross section of the plume in the night of March 15 when the highest contamination occurred there. In the event, the cloud base height ($0.01 < LWC < 1$ g m^{-3}) was very low near to the ground so that most of radionuclides scavenged by in-cloud processing and the contribution of below-cloud scavenging was relatively low. Amongst the whole regional-scale model domain as well as local-scale domain, the highest contribution of the below-cloud scavenging was smaller than 1%. The model formulation of the below-cloud scavenging process and the quantitative comparisons between the simulated in-cloud and the below-cloud scavenging coefficients were given in (new) Appendix. Further details of the analysis are beyond the scope of this study, which is the estimation of the emission inventory, and so those will be discussed in the future works.

Comparisons done table 6 are not homogeneous. It is not always the same simulations that are compared with the “New-land” one. It has to be more homogeneous.

➔ We completely revised the table for four WSPEEDI calculation cases (combinations of the original/modified WSPEEDI and Terada/new source terms) (new Table 7) so that readers can understand the impact of new source term.

Local air dose rate

A table giving the statistical indicators for air dose rate comparisons should be added.

➔ (Old) Table 6 included the statistics compared with local-scale air dose rates. Furthermore, in (new) Table 6, statistical indicators were provided for four simulation cases (combinations of the original/modified WSPEEDI and Terada/new source terms) so that readers can readily understand the main result in this study.

Section 4.1

Chapter 4.1 could be moved to the beginning of Section 3 in order to better highlight the specificity of the new source term estimation.

➔ (Old) Chapter 4 (Discussion) was completely reorganized to focus on new source term estimation.

The authors claim that the release of March 15-16 is assessed because of the new data set: what data are useful to reconstruct this event?

➔ The advantage of new dataset was described in (new) chapter 3.

I do not believe that the modified wet scavenging scheme could explain the new timing of the release event. It can help to decrease the release rate on March 15 pm but it cannot explain the increase of the release rate in the evening.

➔ The sentence was inappropriate. We corrected the part as the new timing of release events was found out by using new monitoring data in section 4.2 as "These were particularly effective to find this release and determine the timing and release rates."

Section 4.2

As previously said, it is difficult to precisely identify what is the specific contribution of the new deposition scheme and I do not believe that the authors should end their paper with this section. This discussion could be dispatched partly in section 3.1 and partly in section 3.2.

➔ This section was extracted from the maintext (moved to Supplement) so that we focused on the impact of new source term (subsection 4.1.1).

What is the relative contribution of below cloud and in-cloud scavenging (especially at the local scale)?

➔ As replied above, the wet deposition amount due to the below-cloud scavenging was highest in the northwest vicinities of the power plant, 10 kBq m^{-2}. We investigated the reason why the contribution of below cloud scavenging was such low in the northwest region by drawing horizontal-vertical cross section of the plume in the night of March 15 when the highest contamination occurred there. In the event, the cloud base height ($0.01 < LWC < 1$ g m^{-3}) was very low near to the ground so that most of radionuclides scavenged by in-cloud processing and the contribution of below-cloud scavenging was relatively low. Amongst the whole regional-scale model domain as well as local-scale domain, the highest contribution of the below-cloud scavenging was smaller than 1%.

The authors should be less conclusive on the beneficial contribution of the new deposition scheme considering the various uncertainties (meteorological data, iodine speciation…). Moreover Table 7 shows that the model to data comparison may be less sensitive to the MLDPO deposition scheme than to the meteorological conditions (NAME simulations) and to the source term.

➔ We agreed with the reviewer comment, and revised the description of (new) subsection 4.1.1 so that the beneficial contribution of the new deposition scheme is less conclusive considering the several uncertainties.

The authors present the fog deposition scheme as an important improvement. What about the quality of the fog and drizzle simulations with MM5? What about the fog observations? Light rains are not detected with radar observations. Are they with rain gauge?

➔ According to the comments from other reviewers, we excluded (old) section 4.2 about deposition processes (and moved it to Supplement). However, you can find the simulation result of cloud liquid water content (LWC) at the ground station (Fig. S8). MM5 clearly underestimated LWC event which caused the underestimation of fogwater deposition. Yes, the data referred in the previous manuscript was radar data merged with rain gauge data. More detailed analysis should be provided to the future study.

The relevance of the precipitation data should have been discussed before the end of the paper since it has a huge impact on the release assessment. For instance, what is the impact of the over-estimation of the rain data on March 20?

➔ We partially addressed the relevance of the precipitation data and the release assessment in subsection 4.1.1 and Conclusion. Further analysis is beyond the scope, but some discussion of deposition process is available in (new) Supplement.

<u>Appendix: below cloud scavenging</u>

Nucleation scavenging rate is a process to be considered for in cloud scavenging and not below cloud scavenging. What is the point of the authors?

➔ Yes, we reworded in cloud scavenging by nucleation scavenging in the previous manuscript. In 3-D chemical transport models, in cloud scavenging (nucleation scavenging) is dominant to below cloud scavenging for the removal of hygroscopic submicron aerosols. That was what we meant in the previous manuscript.

How the below cloud scavenging is parameterized since you do not consider aerosol-hydrometeor coagulations scavenging? What is the relative contribution of below cloud and in-cloud scavenging at the regional scale?

➔ In the previous manuscript, we did not consider the below-cloud scavenging because it was found much less significant in our simulation. As replied above, the contribution of below cloud scavenging was smaller than 1% using the revised WSPEEDI-II.

At the local scale, the plume may be situated below the cloud. Therefor below-cloud scavenging cannot be neglect compare to in cloud scavenging.

➔ As replied above, we investigated the reason why the contribution of below cloud scavenging was such low in the northwest region by drawing horizontal-vertical cross section of the plume in the night of March 15 when the highest contamination occurred there. In the event, the cloud base height (0.01 < LWC < 1 g m^{-3}) was very low near to the ground so that most of radionuclides scavenged by in-cloud processing and the contribution of below-cloud scavenging was relatively low.

Paper organization

Section 2.4.1: the reverse estimation method is partially described in section 2.4.1 instead of section 2.2. The observations used in the study were partially described in section 2.2.

See suggestions for Section 4.

➔ Considering all reviewer comments, we completely re-organized the contents of all sections.

MINOR COMMENTS

Section 1: Introduction

P14730 the argument developed following « First, the estimation… » has to be clarified. Too many things are discussed.

➔ This part was revised in (new) Introduction.

When explaining the source of discrepancy they need to add the uncertainties in the meteorological data (wind, rain…).

➔ Although we realize the importance of the evaluation of the reliability and uncertainties of meteorological data for various events, the detailed analysis on this matter needs more space in paper and should be considered in future work. In the revised paper, we partially discussed this issue in subsection 4.1.1, Conclusions, and Supplement.

Section 2.2: Reverse estimation method over the land

Assumptions regarding the ratios of I2, CH3I and particulate iodine have to be specified together with the isotopic composition of the release. The authors need to evoke the strong uncertainties due to the isotopic composition of the release and a fortiori of speciation of the iodine, the behavior of iodine into the atmosphere. This discussion can be done section 2.4.4 if more appropriate.

➔ The ratio of gaseous and particulate iodine is determined from the measurement at JAEA-Tokai and the ratio of I_2 and CH_3 is assumed based on reference, RASCAL4.0, because no observed data on the ratio of I_2 and CH_3I. The determination of the ratio of I_2, CH_3I and particulate iodine mentioned above has strong uncertainty. Because the deposition manner of these three types of iodine is different in environment, the estimation of source term for iodine is affected by this uncertainty. This was written in subsection 2.3.3.

Section 2.4.4: Simulation settings

What meteorological data were used and when? The authors should precise the method they used to choose the more appropriate meteorological data for each release event.

Are the meteorological data different from the previous study? What are the differences?

➔ As description was obscure, we revised as “Two sets of meteorological input data, a Grid Point Value (GPV) of the Global Spectral Model for Japan region (GSM) and the Meso-Scale Model (MSM) provided by the Japan Meteorological Agency (JMA) are used for initial and boundary conditions of MM5. MSM which covers over Japan with finer resolution is adopted to the reverse estimation over the land and GSM over the globe to the estimation over the ocean” in subsection 2.3.2.

Section 3.1

Section 3.1 is very interesting but it is sometimes difficult to discriminate between what is known for sure and what is due to the analysis of the results/model outputs. For example, P14745 “the light rain or drizzle”. Is it observed? The text has to be carefully re-read in order to avoid any ambiguity.

➔ We revised words from “the light rain or drizzle bands” to “rain bands” observed by rain gauge at Koriyama (Fig. S1). We also revised the whole text to avoid ambiguity of wording.

Section 3.2

Statistical indicators must always be the same in the various tables and in the text. For instance, p 14752 FA 10 is used; p 14754 FA 5 is used. You should use always FA5 for instance.

➔ To make the paper shortened, we deleted Tables 7-8 for WMO model calculations in the revised version. In addition, Table 6 has been fully revised with the statistics of CC, NMSE, FB, and FA2/FA5/FA10 to demonstrate the difference between four WSPEEDI simulation cases (combinations of the original/modified WSPEEDI and Terada/new source terms) in subsection 4.1.1.

Validation using several models: this part can be shortened

➔ The text is shortened to exclude the discussion using NAME model, which is now present in supplement.

Section 5

Modifications have to be done in accordance with the previous remarks (uncertainties, impact of the new cloud scheme…).

➔ Conclusion was modified to show the potential uncertainties with the new source term.

Tables

A table similar to table 3 could be added for dose rate observations used in the reverse method.

➔The new table (Table 3) for air dose rate was added to the revised version

Table 5: description of the last column is missing. Does it give what monitoring data are used to assess the source term?

➔Thank you for your suggestion. We revised (new) Table 6.

Tables 6-7-8: please give the same list of statistical indicators (add FA2, FA5, FA10 in tables7-8 and NMSE, FB, FMS, KSP, Rank in table 6).

➔ As replied above, we deleted Tables 7-8 for WMO model calculations in the revised version to make the paper shortened. In addition, Table 6 has been fully revised with the statistics of CC, NMSE, FB, and FA2/FA5/FA10 to demonstrate the difference between four WSPEEDI simulation cases (combinations of the original/modified WSPEEDI and Terada/new source terms) in subsection 4.1.1.

Figures

Generally Figures are too small.

➔ All figures were modified so that the readers could readily understand them.

Some figures are not essential and may be removed if the paper is too long for publication. For instance
Figure 1 can be suppressed.
I am not sure that Figures 5-6 and 23 are required.
Figures 8 are too small to be useful.
There is a problem with the blue curve on Figure 11c.

➔ According to your comment, many figures (e.g., old Figs. 5, 6, and 23) were extracted from the main text. As mentioned above, (old) Figs. 8 and 11 were also modified.

Bands within a factor of 10 have to be added on Figures 18-20-21-24.

➔ According to your comment, bands within a factor of 10 are added to (new) Figs. 19 and 21.

Appendix

The authors should give more information on the initiation of the various parameterizations and the rain threshold used.

➔ We tried our best to describe each variable used in the equations of (new) Appendix in the revised manuscript. The rain thresholds were added in Appendix. The in-cloud scavenging is activated in a model grid where cloud water mixing ratio is higher than 10^{-6} (kg kg^{-1}) and the surface precipitation intensity is larger than zero. The below-cloud scavenging is activated in a model grid where each settling hydrometeor mixing ratio is higher than 10^{-9} (kg kg^{-1}) and the surface precipitation intensity is larger than zero.

The authors should talk about Iodine particulate instead of restricting it to particulate I-131.

➔We revised the word "particulate I-131" to the general form "particulate iodine" in Appendix section.

L22 p14770 has to be modified.

➔ We revised the word "particulateI" to "particulate iodine".