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.3

In this paper the authors propose a new estimation of the releases of several radionucleides during the Fukushima accident. For that they use a modified version of the atmospheric dispersion model WSPEEDII – with a new deposition scheme – and the oceanographic dispersion model SEA GEARN-FDM, together with air concentration and surface deposition measurements – augmented with respect to the one used in previous work. Also, a detailed analysis of the recovered source is provided, comparing it with the events that took place during the nuclear accident. A validation of the source is performed by comparing the simulated measurements provided by the new source with the real ones. These simulated measurements are obtained, first, using the WSPEED-II atmospheric dispersion model and then, using other proposed atmospheric dispersion models. The article provides new insights on the releases during the Fukushima accident, and it fits within the scope of Atmospheric Chemistry and Physics. Hence I recommend it for publication after the following comments are addressed.

➔ Thank you for your careful reading and providing critical 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.

MAJOR COMMENTS

In general, the article is too long. It should be written in a much more concrete and concise way, making it easier to understand for the reader.

➔ 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.

Section 2.2 Why, instead of posing the problem as a linear system (Stohl et al., 2012), the authors estimate the source unknowns one by one? Is there any advantages in using the method proposed in the paper with respect to (Stohl et al., 2012)? Using a linear system, the situation explained in p 14735 | 12 would be solved in a more reasonable way. The same applies for the correction of the source in section 2.3

➔ At the first part of chapter 2, we defined the reverse and inverse methods with merit and demerit and also described why we choose the reverse method for the Fukushima 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.”

The explanation of the estimation methods is, in general, confusing. Many details should be clarified:

How is the temporal discretization of the source defined, i.e., starting and ending points of each temporal element? Why is this discretization not regular? This explanation must be included in the manuscript.

➔ 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) subsections 2.1.1 and 2.3.3.

The mathematical notation is, in general, quite confusing and makes the method description unnecessarily difficult to understand.

Eq. (1), (3): Qi, Mi and Cidepend not only on space, but also on time. This must be indicated (Qi(t), for example). Also it is necessary to make clear the difference between time of emission and time of detection. For example, in Eq.1, the time of Qi and time of Mi are different, Qi(t1) , Mi(t2).

➔ We modified the equations and these explanations in subsections 2.1.1 and 2.1.2.

Section 2.2 and 2.3 The measurements and the dilution factors may contain errors. The estimated source may be sensitive to these errors. How do you address this problem?

➔ We realize the importance of the evaluation of the reliability and uncertainties. However, the detailed discussion on this matter needs more space in paper. Thus, we would like to discuss this matter shortly in Conclusion. Detailed discussion will be considered in future paper with discussion of the effect of our deposition model.

p 14735 | 11 Explain why only the peak values are used.

➔ 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).

p 14736 | 25 How do you determine in which periods the plume flows towards the ocean?

➔ This was written in (new) subsection 2.1.2 as "The judgment of whether the plume during each segment directly flowed toward the ocean is done by evaluating the simulation of the modified WSPEEDI-II, observed wind direction, and monitoring data on the land."

Section 2.3 In general, the subindexes of the variables are extremely confusing here, because they mix space and time. To make clearer what is what, the notation must be revised completely. For example in Eq. (4), Cnj [k], instead of Cj,k

➔ The suffix and equations in (new) subsection 2.1.2 are revised according to the comments from each reviewer.

p 14738 | 2 Where does this equality come from? A more detailed explanation should be included.

➔We revised the related sentences in (new) subsection 2.1.2.

p 14760 | 14 In 14759 | 13, you use the events that took place during the accident to assess your source, and thus claim that your source estimation is correct, and previous source estimations in the literature are not correct. But later, you compare again to the same previous estimations in the literature (which are supposedly wrong) and where they agree with your results, you claim that this again confirms the correctness of your results. This is not a consistent argument!

➔ At page 14759 line 13 in our previous paper, it is described that the source term in the afternoon to the night of 15 March in present study is better than previous one, while at page 14760 line 14, the estimations in present and previous studies when the plume flowed toward the ocean are resemble. The evaluated period is different.

Minor comments

p 14735 | 10 What does it mean "if the data show a continuous time series"? The measurements are always discrete.

➔We deleted this sentence in the revised version.

p 14737 | 15 The variable name Cos can be mistaken with a cosine. A different name should be used.

p 14737 | 11 typo "Note that the only the observational. . ."

p 14738 | 2 Equation number missing.

➔ We modified the above mistakes. Thank you for your suggestions.

Section 3.2.1 Does it have sense to validate the source with the same measurements and model that were used to estimated it? Because the source that fits best with these measurements is the one that produces overfitting. If the same model and the same measurements are used for validation, then some kind of cross-validation technique must be used.

➔ As described in section 2.2.3, for validation, we did not use the same measurements used to estimated source term but used surface deposition, air dose rate, and global air concentration data.

Section 4.1 The wet venting at Unit 3 and DW pressure deficits do not directly imply that the major release took place at this time. If it does, you should argument it properly.

➔ As you suggested, it is not sure that the venting at Unit 3 generated the MAJOR release or not. However, we believe that it is reasonable to assume that DW pressure deficits imply the atmospheric releases. In the case of the evening to night of 15 March, we cannot determine the ratio of releases from Units 2 and 3 due to lack of information. Thus, though we assumed both are the source of releases we changed the sentences less conclusive as "As a result, the period of the potential major release is coincident with the wet venting at Unit 3 and/or DW pressure deficits at both Units 2 and 3 reported on 15–16 March (Fig. 9)." in section 4.2.

p 14760 | 1 How do you know that the source changes drastically in this period? How do you asses that?

➔ From environmental monitoring data, it was expected that a large amount of releases occurred from 15-16 March. However, the reason is still not clear. Thus, the source term is estimated in detail which is one of the main objectives of our paper. We added this point in subsection 2.3.3.

A general correction of minor typos through the whole paper is necessary.

➔ We checked and revised typos throughout the manuscript (including English corrections).