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
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Author response to reviewer comments

Response to Ref.2

The manuscript presents the new source term estimation of I-131 and Cs-137 released into the atmosphere from the Fukushima Dai-ichi Nuclear Power Station (FNPS1) in Japan by inversion analysis combining measurement data and offline coupling model of the atmospheric and oceanic dispersion models. Also, the manuscript evaluates the new source term by comparing the simulation using different atmospheric dispersion model with measured atmospheric concentration and surface deposition. At the present time, the multi-media environmental pollution caused by the massive release of radionuclides to the atmosphere from the FNPS1 is still severe natural and social issues, while the total amount of source term and its temporal variation has a large uncertainty. In this situation, the author's work brings very valuable and timely information to the international society. The topic of manuscript certainly is suitable for ACP.

➔Thank you so much for your positive comments on our paper toward the publication.

The new source term was validated by comparing the modified WSPEEDI-II simulation using the new source term and the previous WSPEEDI-II simulation using the previous source term with measurements. As a result, both effects due to improved deposition scheme and improved source term are mixed in the discussion for validation of the new source term. This is a weakness in this manuscript. In the analysis, it is very important to separate two effects of deposition scheme and source term. For example, the authors compare the simulations using the new source term and the previous source term based on the modified WSPEEDI-II model and then analysis the differences between two simulations.

➔As suggested by the reviewer, 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.

Additionally, there are many points which should be clarified. The reviewer recommends publishing this paper with major revisions in response to the following questions and comments.

➔As shown below, we carefully responded to your comments through revision of the manuscript. However, 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

Validation of new source term Most important progress in author's study is to determine the new and detailed source term of I-131 and Cs-137 combining measurement data and offline coupling model of the atmospheric and oceanic dispersion models. Hence, it is important to compare the modeled results of new and previous source terms using the modified WSPEEDI-II model based on measurement data (air concentration, air dose rate, and surface deposition) and then demonstrate the advantage of the new source term quantitatively. From this viewpoint, the authors need to add analysis and discussion. Additionally, for the air concentration, the authors should use the measurement data at not only JAEA-Tokai but also other sites.

➔ As replied above, we made 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.

Concerning the air concentration data, we did not use them for validation using WSPEEDI-II because these are already used for source term estimation. Though the validation by using some WMO models uses only data of JAEA-Tokai, it is because this data only show the temporal variation in detail and suitable for validation. It may be possible to compare other data, but we hesitate to increase the volume of paper.

Validation of new deposition scheme In this paper, a new deposition scheme, which deals with dry and fogwater depositions, CCN activation and subsequent wet scavenging for radioactive iodine gas and other particles, was incorporated into WSPEEDI-II. However, this new deposition scheme wasn't validated objectively based on measurement data, though authors discussed about it in section 4.2. The authors should compare the modeled results with old and new schemes in fixed source term and then demonstrate the advantage of the new scheme.

➔ As replied above, we examined the reliability of deposition model and new source term by carefully comparing above four WSPEEDI simulation cases in the revised paper (subsection 4.1.1).

Ratio of gaseous and particulate I-131 The ratio of gaseous and particulate I-131 is one of critical parameters in the inverse estimation for I-131 source term. It is well known that the ratios of gaseous and particulate I-131 have large variability in time and/or space. It is considered that the ratio depends on the source condition as well as the history of air mass (especially, washout or not). In fact, the ratio of gaseous to total I-131 varies in the range of 0.2 to 0.8 in space and/or time (Tsuruta et al., 2012, 2014). The authors should discuss the impacts of the ratio determined from the data collected at only one site (JAEA-Tokai) on the I-131 source term estimation.

➔ As the concentration data of JAEA-Tokai is used for the source term estimation, we also used the ratio of gaseous and particulate I-131 from JAEA-Tokai data. Of course, we consider the temporal variation of the ratio at Tokai and correct the ratio at Tokai to that at the release point considering the history of air mass by simulation model which can treat the difference of deposition process of gaseous and particulate. However, the reviewers comment is important. Thus, we cited (English) paper (Tsuruta et al., 2012) and noted that the ratio depended on the source condition as well as the history of air mass (new subsection 2.3.3).

Individual comments

1) Page 14735, lines 14-15: Why does the use of peak value reduce the impact of discrepancies in arrival time? If the modeled arrival time differs from the measured time, the modeled peak concentration should be different from the measurement. Additionally, the use of peak values, which is not statistically stable, may cause higher uncertainty in the reverse estimation. Why did the authors use peak value, not average value?

➔ 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).

2) Page 14736, lines 15-16: This part is not clear. The authors should explain more detail.

➔ This part is deleted in the revision to shorten the paper.

3) Page 14737, lines 11-12: The authors should explain how to classify the affected points from the un-affected points.

➔ We revised in subsection 2.1.2 as "Note that by preliminary comparison between measurement points of sea surface concentration of ^{134}Cs and the oceanic dispersion area estimated by simulation using sources of direct release from FNPS1 into the ocean, only observational points that are not affected by the direct release of ^{134}Cs from FNPS1 to the ocean are used for the source term estimation.".

4) Page 14739, line 2: It is better that "Fig.2" is changed to "Fig.2d".

➔ Figure 2d was deleted to reduce the paper length.

5) Page 14740, lines 2-4: It is better that Fig. 6 is moved to section 3.1 because this figure is modeled results. Fig. 6 seems to show the modeled surface concentration of plume emitted in the event shown in captions for each panel. It is better that brief explanation of Fig. 6 is added.

➔ (Old) Fig. 6 was deleted according to other reviewer's comment.

6) Page 14742, line 7: According to Fig. 9a, the model extremely underestimates the peak at 15:00. The authors should add some comments on the reason and effects to source term estimation.

➔ All of source term estimation using air dose rates was carried out by using ground-shine. For example, in (new) Fig. 6a, the timing of peak appearance by the plume arrival, and the values of ground-shine shown as slow-slope after the peak agreed well with observations. Hence, the difference of peak values does not affect the estimation of source term. The usage of ground-shine was addressed in (new) sections 3.1 and 3.4, and subsection 4.1.1.

7) Page 14743, lines 1-3: The modeled narrow deposition band (Fig. 9) shifts around 30 degrees compared with measurement (Fig.10). The authors should add some explanations for the shift.

➔ We added the sentences in section 3.1 as "Figure 6c compares the distribution of air dose rates in day-time of March 13 between the WSPEEDI simulation and observation by portable monitor mentioned above. The calculated result is slightly shifted to the west due to the delay of the wind shift comparing with observed wind shift, but it shows the similar distribution pattern and air dose rates as observed ones."

8) Page 14743, lines 4-5: What is evidence for "deposition area was far from the plant due to the elevated release from the stack"?

➔This sentence was deleted to reduce the paper length. However, the evidence is explained as follows: the radionuclides were released from the stack during the venting event on 12 March. Thus, the downwind distance is required until when the plume disperses and contacted with the ground surface. Since there was no rain event on 12 March, only the dry deposition of the plume formed the contaminated area.

9) Page 14744, line 27: "Fig. 11b" should be changed to "Fig.8b".

➔Thank you for your suggestion. In the revised manuscript, figures are all re-numbered according to organize the contents of the paper.

10) Page 14746, line 9: More detail explanation for "modifications of the deposition scheme" is needed.

➔The sentence was unclear. As explained in (new) subsection 4.1.1, wet deposition of the plume released in the morning of 15 March has been enhanced by high scavenging coefficient of modified deposition scheme. The sentence was now revised in section 3.4: "Due to an increase of wet scavenging coefficient in the modified deposition scheme (Fig. A2), …"

11) Page 14746, lines 10-14: One possible reason for “a large increase in the air dose rates did not appear at Fukushima and Iitate areas” is that most of radionuclides deposited before the air mass arrived at these areas. The authors should analysis and discuss more carefully.

➔As described in section 3.4, we do not think most of radionuclides deposited before the air mass arrived at these areas “because rain bands coming from the northwest during the afternoon of 15 March caused the precipitations started around Iitate area from 16:00, and those were very small about 1 mm h^{-1} (Fig. S1). Moreover, Ohno (4.9 km WSW from the site) had no rain observed until the night (Fig. S1). The fact suggests that the plume discharged in the afternoon should produce less amount of (dry) deposition along the pathway from the FNPS1 to the northwest direction. Therefore, the plume can reach Iitate and increase air dose rate due to wet deposition if a large amount of radionuclides were discharged during the afternoon.”

12) Page 14748, lines 13-23: Both ratio of I-131/Cs-137 and gaseous/particulate iodine on 16 March are higher than those on 15 March. These facts may suggest that particulate Cs-137 and I-131 were deposited by precipitation before arriving at JAEATokai on 16 March while there was no precipitation on 15 March. Hence the authors should analysis and discuss based on the background that the changes in the ratios of I-131/Cs-137 and gaseous/particulate iodine are caused by not only source change but also history of air mass.

➔ We revised the paper in section 3.4 based on your comment. Because the simulation model considers the changes in the ratios of ^{137}Cs/^{131}I and of gaseous/particulate iodine caused by transport history of air mass, the ratios at the measurement point is different from those estimated at the release point in this study.

13) Page 14752, lines 19-22: Which is a larger factor in improvement of regional deposition, enhancement of the scavenging coefficient or revised source term?

➔ To respond your major comment, we made several test simulations using original/revised WSPEEDI and Terada/new source terms and evaluate the impacts of modeled deposition scheme and revised source term to obtain important improvements in simulation results. The discussion is available in subsection 4.1.1.

14) Page 14753, line 22: The authors should show the source term for Te-132 and the evidence that the modification for Te-132 is reasonable.

➔ Since (new) Table 6 has no space, we prepared the supplemental .csv file for the new source term for Te-132, Cs-134, with I-131 and Cs-137. Te-132 (its progeny I-132) and I-131 are major nuclides to contribute the air dose rate due to ground-shine in the early stage (“Local depositions of ^{131}I, and ^{137}Cs, and air dose rate over Fukushima Prefecture“ in subsection 4.1.1). Since the modified model with new source term well reproduced measured air dose rates (Fig. 11a, d and Fig. 16d), we believe that “the modifications to the deposition scheme and source term are reasonable, particularly for ^{132}Te and ^{131}I, which are the major contributors to the ground-shine in the early phases of the accident.” (subsection 4.1.1)

15) Page 14753, line 27: Table 6 shows that the statistical scores of new results are lower than previous results in air dose rate in the north-west area of FNPS1.

➔As replied above, Table 6 was completely modified for four test simulations using original/revised WSPEEDI and Terada/new source terms to demonstrate the impacts of modeled deposition scheme and the new source term.

16) Page 14755, line 6: Why was the measurement data at only one site (JAEA-Tokai) used?

➔ The validation by using some WMO models uses only data of JAEA-Tokai because this is the only data showing the high temporal variation which is suitable for model validation. Although it may be possible to compare other datasets, we did not use those because we needed to reduce the paper size.

17) Page 14755, lines 10-14: The authors should compare the modeled concentration using the new source term with that using the previous source term in Fig.18 and then demonstrate some advantages of the new source term.

➔ Our discussion of WMO model results confused the reviewer. It was not surprising that the performance of each model in (new) subsection 4.1.2 widely varied because these used the source term derived from the different model (i.e., WSPEEDI-II). We should focus on how the new source term works on atmospheric dispersion simulations to some extent. Thus, we deleted those sentences.

18) Page 14755, lines 22-25: The score of FB in Table 7 shows that the modeled deposition using the new source term tends to underestimate and to be worse than the simulation using the previous source term.

➔ Our discussion of WMO model results confused the reviewer. As explained above, we should focus on how the new source term works on atmospheric dispersion simulations to some extent. Thus, we deleted we extracted all of tables related to this work.

19) Page 14757, lines 5-7: In Table 7, The NMSE and FB for I-131 concentration with the new source term was worse than those with the previous source term excluding FB for MLDP0 model. The authors need to make comment on these results.

➔ As replied above, our statistics table confused the reviewer. It was not surprising that the performance of each model in (new) subsection 4.1.2 widely varied because these used the source term derived from the different model (i.e., WSPEEDI-II). We should focus on how the new source term works on atmospheric dispersion simulations to some extent. Thus, we extracted all of tables related to this work, and focused on the overall agreement between calculations and observations when using the new source term in (new) subsection 4.1.2.

20) Page 14758, lines 4-10: The authors should compare the modeled concentration using the new source term with that using the previous one in Figs. 22 and 23 and then demonstrate the advantage of the new source term.

➔ As replied above, it was not surprising that the performance of each model in (new) subsection 4.1.2 widely varied because these used the source term derived from the different model (i.e., WSPEEDI-II). We should focus on how the new source term works on atmospheric dispersion simulations to some extent. Thus, we extracted all of tables and focused on the overall agreement between calculations and observations when using the new source term in (new) subsection 4.1.2.

21) Page 14758, lines 20-24: This part is not clear and more explanation is needed.

➔ Since this part was confused, we deleted in the revised version.

22) Page 14758, lines 25-28: This is true? According to Table 8, the CCs for particulate I-131 and Cs-137 in the new source term case was slightly lower than that in the previous source term. Additionally, the NMSE became worse when the new source term was used. For other scores, the situation was case by case.

➔ Our statistics table confused the reviewer. As replied above, it was not surprising that the performance of each model in (new) subsection 4.1.2 widely varied because these used the source term derived from the different model model (i.e., WSPEEDI-II). We should focus on how the new source term works on atmospheric dispersion simulations to some extent. Thus, we extracted all of tables and focused on the overall agreement between calculations and observations when using the new source term in (new) subsection 4.1.2.

23) Page 14761, lines 3-4: From Fig. 26a, it is needed that the "then particulate iodine, and finally gaseous CH3I" is changed to "then gaseous CH3I, and finally particulate iodine".

➔ This context was deleted because we removed section 4.2 of the original version to shorten the manuscript. However, the calculations using the revised WSPEEDI-II model shown in Supplement (Fig. S5) showed that the DRY deposition of the iodine is larger in the order of: Gaseous I2 > Particle I > Gaseous CH3I, which was consistent to what we mentioned in our previous manuscript.

24) Page 14761, lines 10-11: This is true? Fig. 26 shows the gas species of I-131 contribute to the contamination of East Japan though their contributions are lower than wet deposition of particulate I-131.

➔ This context was also removed because we removed section 4.2 of the original version to shorten the manuscript. However, we still corrected the sentence to emphasize the negligible effect of the gaseous iodine on "wet deposition" not "total deposition" in Supplement.

25) Page 14761, lines 18-20: The authors should show the appropriate reference indicating WRF-CMAQ model overestimated the observed precipitation amount over Tochigi and Gunma Prefectures.

➔ According to reviewer comments, we moved the discussion of this part from the maintext to Supplement. In the revised version, this sentence was completely deleted.

26) References: There are some references in review. These references should be changed to alternative references which readers can access.

➔ We added all available references to References section.

28) Fig. 9: The authors should answer the following questions about Fig. 9c. – The time of calculated air dose rate (12:00) is different from the time of measurement (from 6:00 to 15:00). Why did the authors use the data at different time for comparison? The air dose rate has a peak around 15:00 on 12 March as shown in Figs. 9a and 9b. Why did the authors use data in the daytime on 13 March instead of data around 15:00 on 12 March?

➔ According to the simulation, the plume was transported toward the Pacific Ocean on 13 March and, thus, the measured air dose rates in (old) Fig. 9c are due to deposit radionuclides discharged in the afternoon of 12 March. Thus, we compared the calculated air dose rate at 12:00 with measurements from 6:00 to 15:00 on March 13 to validate the source term by comparing the calculated and observed distributions of air dose rates. We added this point in section 3.1.

27) Fig. 8: The color scale bar is invisible and should be improved. In the title of figure, the "spatial distribution of accumulated surface deposition" may be better.

29) Fig. 13: The color lines in each figure are invisible and should be improved to be visible.

30) Fig. 17: The black and red lines and horizontal axis in each figure are invisible and should be improved to be visible.

31) Fig. 18: The vertical and horizontal axes in each figure are invisible and should be improved to be visible.

➔ We revised as suggested by the reviewer. Thank you so much.