Dear Editor-in-chief and the three anonymous referees,

We very much appreciate your editing and all the constructive comments and intimate investigations of the three anonymous referees. Thanks to their comments, out manuscript has been substantially improved. The major changes from the previous manuscript are listed as follows:

1. According to the Referee #3's comment (RC2), re-calculation has been made for dust emission. Consequently, Figures 7, 8, 9, 10, 11, and 12 were replaced and all the corresponding quantities were changed, accordingly.

2. According to the Referee #2's comment (RC1) and Referee #3's comment (RC2), Appendix C was moved to the main text as Section 5.3.

The letter consists of the following contents:

1. Point-by-point responses to Referee #1 (RC3)
2. Point-by-point responses to Referee #2 (RC1)
3. Point-by-point responses to Referee #3 (RC2)
4. Revised manuscript with strikethrough

Although the manuscript had been sent to a language editing just before the first submission, the substantial grammar corrections were suggested by the referees. However, we did not send it again to the language editing right now, although we had substantial revisions made in the current manuscript. Therefore, we appreciate if you kindly allow us to send the manuscript again to the language editing after accepted. We will pay special attention to prevent changes of meanings at every single line after the editing. Thank you for your consideration.

Best regards,
Mizuo Kajino

Dear anonymous referee #1,

We very much appreciate your constructive comments, useful information and your time for RC3. Especially, introducing articles which we have not been aware and the grammar corrections were really helpful. Thanks to your review, our manuscript has been substantially improved. Point-by-point responses to your comments are written in blue in this letter.

Best regards,
Mizuo Kajino

General comments:

The paper assesses redistribution events of Cesium from different sources in Japan in the following years after the Fukushima accident. It tackles the scientific topic very well from many different directions and I think it would be suitable for publication to ACP.
Thank you for your evaluation.

I have some minor comments / suggestions that I hope they would help the authors to make this publication more comprehensive to the readers.

Specific comments:

Page 1, Line 20 (hereafter it will be P1,L20): Substitute "the contaminated ..." to "a contaminated"
I changed it.

P1,L22: "The assessment period ... 2012". I don't understand what you mean here. The assessment period is 2013, but the field experiments were carried out in December 2012?? Could you please clarify?
The start of field experiment is December 14, 2012 and it has been continued up to now. Our assessment period is for the full year of 2013, after the start of the experiment. I have modified the sentence simultaneously reflecting the Referee #2's comment (RC1) as follows: "In order to assess the long-term effect, the full year of 2013 was selected to study just after the start of the field experiments".

P2,L3: Correct "to reducing" to "in reducing"
I changed it.

P2,L11: Put a full-stop after "remain unknown". Continue the sentence as: "This is the first study that provides a crude estimation ..."
I changed it.

P2,L12: In the next sentence correct as: "Additional research activities on this direc- tion are needed ..."
Referee #2 (RC1) also suggested correction of the sentence. I modified the sentence combining your and his comments as “Additional research activities should investigate the processes/mechanisms governing the re-suspension over the long term. This could be achieved through conducting additional field experiments and numerical simulations”.

P3,L6: Add the recently released reference after Draxler et al.(2015): Kristiansen, N. I., Stohl, A., Olivié, D. J. L., Croft, B., Søvde, O. A., Klein, H., Christoudias, T., Kunkel, D., Leadbetter, S. J., Lee, Y. H., Zhang, K., Tsigaridis, K., Bergman, T., Evangeliou, N., Wang, H., Ma, P.-L., Easter, R. C., Rasch, P. J., Liu, X., Pitari, G., Di Genova, G., Zhao, S. Y., Balkanski, Y., Bauer, S. E., Faluvegi, G. S., Kokkola, H., Martin, R. V., Pierce, J. R., Schulz, M., Shindell, D., Tost, H., and Zhang, H.: Evaluation of observed and modelled aerosol lifetimes using radioactive tracers of opportunity and an ensemble of 19 global models, Atmos. Chem. Phys., 16, 3525-3561, doi:10.5194/acp-16-3525- 2016, 2016.
I added it. Thank you.

P3,L14: "... are of particular concern." Add the reference before the full-stop: Evangeliou, N., Balkanski, Y., Cozic, A., Møller, A. P. (2014), "Global and local cancer risks after the Fukushima Nuclear Power Plant accident as seen from Chernobyl: A modeling study for radiocaesium (134Cs & 137Cs)", Environment International, 64, 17–27.
I added it. Thank you.

P3,L21: Put "Tsukuba" in parentheses instead.
I changed it.

P4,L7: You say that you considered sources (1) and (4) in your study. But source (4) refers to releases from the Chernobyl sarchofagus, which is a different thing comparing to the situation in Fukushima. So, how did you considered this source in your study. I think you must reformulate your sentence here and make it clearer.

I modified the corresponding sentences to “Garger et al. (2012) summarized the re-suspension sources following the Chernobyl accident as (1) dust emission, (2) human activity in fields, and on roads and construction sites, (3) forest fires, and (4) emissions from the power plant (i.e., opening of the Chernobyl sarcophagus). Re-suspension sources (1) dust emission (Ishizuka et al., 2016) and (4) i.e., additional emissions from the reactor buildings of FDNPP (TEPCO, 2012; 2013; 2014a; 2014b; 2015) were considered in the present study.”

P4,L9: Change "low chance of ..." to "low risk of ..."

I changed it.

P4,L22: "The current study is the first but crude estimation of the spatial budget ...". You said the same in a previous line in the ABSTRACT. Please remove or change your sentence.

I removed the sentence and modified the following sentence as “Even though the re-suspension mechanisms still remain unknown, by utilizing observational data …”.

P4,L30: I am not sure if this 4-line paragraph is needed in the beginning of each chapter.

Although this style may not often be found in other manuscripts and may rather often be found in the end of Introduction section, I considered the style of guidance is informative to readers. Thank you for your understanding.

P7,L9: Change "... a maximum estimate of 137Cs ..." to "... the upper boundary of 137Cs ..."

I changed it.

P7,L17: Please move the web reference to the list of references at the end of the manuscript

I couldn’t find any statements of the web reference treatment in the ACP manuscript preparation guideline page. However, judging from previously published ACP papers, the editorial office seemed to allow inserting web references in the body of manuscript. Thank you for your understanding.

P8,L17: There is a mesh with your figures and the way they are placed in the text. First of all, I think that ACP requires format of the full word inside the manuscript, so instead of "Fig." you should change to "Figure" everywhere in the manuscript. Second, Figure 4a is referred in this line, but Figure 3 is referred after!!! This is completely confusing. Either change the sequence of your figures, or reformulate the text. I noticed this may happen more than once, so you need to scan the manuscript carefully.

In the up-to-date manuscript preparation guideline, it says —The abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence, e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that...".

As to the order of figures, I intended to do so. The figures are ordered consistently with the sentences “in general”, but sometimes it is referring a part of figures behind in advance. I tried to remove all the unnecessary reverse to avoid confounding the readers. In case if I assume it necessary, I modified the phrase from “Fig. 4a” to “as shown later in Fig. 4a”, for example.

P8,L30: "Unlike re-suspension from soil, precipitation might not suppress re-suspension from the forest ecosystems since substantial amounts of K-containing particles were observed in the wet season in the Amazon (Pöhlker et al., 2012 and references therein)." Could you please explain this further and link it a little bit more to the paragraph? It does not seem relevant or I am having trouble to understand it.

I removed the sentence as it has no impacts on and rather destroys the context in the paragraph. Also, the sentence is not necessary here because the precipitation effect did not consider either in dust emission nor forest emission equations.

P10,L8: Since you speak about measurements, I think you must be more precise. You only mention that "At both sites,ambient aerosols were collected using a high-volume air sampler and 134Cs and 137Cs concentrations were obtained by γ-ray spectroscopy using a Ge semiconductor detector." YOU MUST MENTION: (1) Relative efficiency of the Ge detector relative to NaI, (2) Resolution energy at the photopeak of 60Co, (3) How the energy calibration was performed, what range of energies was covered and what the resolution per channel was, (4) How the efficiency calibration was performed, what radioactive solution was used, who the provider was, (5) what the geometry was, (6) what the measurement time was, and (7) what the relative statistical error of your measurements was.

In the revised sentence, I removed the instrument parts and just mentioned the elements measured, sampling periods, and sampling intervals.

P11,L13: Change "scattergram" to "scatterplot". In the same sentence change also "the observational and simulation results, respectively" to "the simulation results and observations" and remove "respectively".
I changed it.

P11,L17: Change "employs" to "uses"
I changed it.

P12,L3: Change "a quarter to one third" to percentages
I changed it.

P12,L4: Change "sixty percent" to "60%"
As far as I know, to spell out numbers in the beginning of a sentence is a rule of the English grammar. I couldn't find it in the ACP's guideline but I would like to follow the rule, if any.

P12,L10: "After the screening ...". How did you perform the screening? I am afraid I did not understand that. Could you please explain it better inside the paragraph in order to be clearer?
The screening is to find runs that satisfied the criteria described in the previous sentence. In order to make it clearer, I changed the sentence as "Only one combination (Ec, vd) = (0.04, 0.1 cm/s) satisfied the criteria, and thus …"

P13,L5: "The model successfully reproduced ...". I think you need a more moderate sentence here. Your results are in a logarithmic scale and the discrepancies are rather large to say that your model reproduced the plumes well. I am quite sure that if you perform the same comparison as a scatterplot and not as 2 time series, you will see that there is no correlation at all. But I admit that it is rather difficult to reproduce the plumes much better. Please reformulate though!
I removed the word "successfully".

P13,L16: "Note that resuspension flux ...". This is a bit obscured or I have miss it from the text. Please explain and rationalise your decision to multiply by a factor of 10 here.

This is elaborated in the last paragraph in Sect. 2.2. I changed the sentence to “As discussed in Sect. 2.2, note that the re-suspension flux …”.
There is no rational reason that the result is multiplied by 10 but simply there is evidence that the simulation is underestimated by a factor of 10. As discussed in Sect. 2.2, the dust emission scheme contains parameters obtained at a single location and under a fixed atmospheric condition. It could be one of the reasons for this underestimation. We could have been added the tuning parameter to Eq. (1) to adjust to the observed concentration, but we decided to use the original dust emission module as they were because it was the fact. However, in order to derive the quantitative budget analysis (crude, though), we did the simple multiplication. Certainly, as mentioned in the last sentence of Sect. 2.2 and as listed in Sect. 5.4 (previously Sect. 5.3), the improvement of the dust scheme is one of the key future issues. Also please note that the value has been changed from 10 to five in the revised simulation.

P15,L1: Change "... total deposition amount ..." to "... total deposited amount ...". Please do the same in the next sentence as well.
I changed it.

P15,L7: Please explain the unit %/y2. I do not understand its meaning.
The “2” indicates the footnote number, not a square of year.

P17,L23: Change "Eighty to 90% ..." to "80 to 90%"
Same to the previous comment—As far as I know, to spell out numbers in the beginning of a sentence is a rule of the English grammar. I couldn’t find it in the ACP’s guideline but I would like to follow the rule, if any.

P18,L27: Put the web reference to the list of references.
Same to the previous comment—I couldn’t find any statements of the web reference treatment in the ACP manuscript preparation guideline page. However, judging from previously published ACP papers, the editorial office seems to allow inserting web references in the body of manuscript. Thank you for your understanding.

P18,L31 and everywhere in the text: Change "... toward ..." to "... towards ..."
I changed it.

P19,L7: Change "... from the biota ..." to "... from biota ..."

I changed it.

P20,L7: The conclusion number 2 starts with a sentence that goes on for more than 3 lines. This huge sentence makes the meaning being hard to follow for the reader. Please reformulate this sentence and split it to smaller sentences.
I divided the conclusion #2 into three (#2 – #4). Also, divided the previous conclusion #4 into two (#6 and #7).

P21,L3: "toward" should be changed to "towards"
I changed it.

FIGURES

Figures 1 & 2: Please make them coloured like the rest of the contour plots you have in the manuscript.
I changed it.

Figure 5: Change "scattergram" to "scatterplot" in the caption
I changed it.

Figure 7: Could you please sum up all the resuspension sources (blue, red, green) and compare the result with the observations? It would be good to show this in a scatterplot IN ADDITION to the existing Figure 7. You can plot it below or on the right of Figure 7.
I added the scatterplot on the right of Figure 7 together with a new table (Table 3) showing the statistical measures between them.

Figure 9: Change "Time series of (black) the observed and (colors) the simulated 137Cs ..." to "Time series of observed (black) and simulated (colors) 137Cs ..." in the caption
I changed it.

Figure 11: Change "... were run ..." to "... ran ..." in the caption
I changed it.

Figure C1: You should probably consider putting Figure C1 as Figure 12 or include it to a supplementary chapter together with the Appendix, but this is up to the ACP journal.

According to Referee #2 (RC1) and #3 (RC2), Appendix C has been moved to the main text as Sect. 5.3 and Figure C1 has been moved Figure 14, accordingly.

Dear anonymous referee #2,

We very much appreciate your constructive comments, useful information and your time for RC1. Especially, the intimate investigations, grammar corrections, and introduction of Evrard et al. (2015) were really helpful. Thanks to your review, our manuscript was substantially improved. Point-by-point responses to your comments are written in blue in this letter.

Best regards,
Mizuo Kajino

General comments:

This manuscript quantifies radiocesium resuspension in 2013 in Northeastern Japan following Fukushima accident, based on field observations/experiment and numerical simulations. Overall, the results are well presented (in Tables and Figures) and described in the text. Detailed comments/suggestions are provided in the attached annotated pdf file. Only general comments are provided here:
Thank you for your evaluation. I improved the manuscript according to your general as well as specified comments. Point-by-point responses to your comments in the PDF file are embedded in this letter.

[1] The title does not fully reflect the content of the study, the study period should be provided (rather than mentioning that it is a 'long-term study', which is misleading);
This is a full-year assessment in fact, but the outcomes obtained from the study can have long-term perspectives: Less than 0.1%/y of re-suspension rate will have negligible impacts on reducing the ground radioactivity for a long-term, say ten to several ten years, for example. To avoid the misleading, we defined it in Abstract as "In order to assess the long-term effect, the full year of 2013 was selected to study…". Thank you for your understanding.

[2] Materials and methods section is very clear, and necessary supporting information is provided as Supplementary Material;
Thank you.

[3] In the discussion section (and in the perspectives), hypotheses could usefully be proposed by the authors regarding the mechanisms that may drive the observed/simulated resuspension (in my opinion, it is not sufficient to mention that 'future work should investigate this'. . . The structure/style in which perspectives and conclusions are written could be improved to avoid proving a list of ideas/items;
I modified Sect. 5.4 (Sect. 5.3 in the previous manuscript) excluding the list of items.

[4] It is not clear to me why the so-called 'land surface processes' were removed from the main text and detailed in Appendix C; in my opinion, this could be integrated in the main text.
According to you as well as Referee 3's suggestions, Appendix C has been moved to the main text as Sect. 5.3. Thank you for your suggestion.

Specific comments (embedded in the pdf manuscript):

(P.1) ← page number of your supplement pdf
the accident happened in 2011... I don't know whether we can refer to a long-term study...
As replied to the general comment #1.

why 'ecosystems'?
As discussed in Sect. 2.3, from the previous studies, it is likely that Cs emission exists from forest but the mechanism is totally unknown, whether it is from vegetation, fungi, soil, or litters. That's why we used the word ecosystems to cover everything in the forest.

is it relevant?
Please refer to the reply of the general comment #1.

this statement confirms that it is not a 'long-term' study...
We rephrased the sentence as follows: "In order to assess the long-term effect, the full year of 2013 was selected to study just after the start of the field experiments.".

mainly due to the difference in initial contamination levels at both sites, what do you think?
Basically yes, but the surrounding regions of the sites also affect. Sect. 5.1 together with

Fig. 12 partially answers to your question. For example, 10% of radiocesium concentration at Namie was coming from regions outside the contaminated area (< 300 kBq/m^2) (please see Figure 4a) and 10 – 40 % of radiocesium concentration at Tsukuba was coming from regions inside the contaminated area.

(P.2)

what do you mean?

I modified the phrase as “migration in the soil and biota”. The phrase, migration in the soil, includes migration with soil water and with soil particles (as summarized in Evrard et al., 2015) in the soil.

in negligible proportions?

Referee #1 (RC3) also suggested grammar correction here. The both should work but I took the Referee #1’s suggestion just because it is simpler. Thank you for your understanding.

= reproduce?

I changed it.

= could explain?

I changed it.

within?

This is re-suspension “from” forest.

please rephrase this sentence

>> suggestion: future research should investigate the processes/mechanisms governing this resuspension over the long term?

to avoid repetition, could be as follows:

>> this could be achieved through conducting additional field experiments and numerical simulations

Referee #1 (RC3) also suggested correction of the sentence. I modified the sentence combining the above two of your comments and his comments as “Additional research activities should investigate the processes/mechanisms governing the re-suspension over the long term. This could be achieved through conducting additional field experiments and numerical simulations”

maybe add 'Japan'?
I added it.

the ocean
I changed it.

that occurred
I added it.

replace with 'soil' only
I changed it.

during the months and the first years that followed the accident
Here I meant March 2011 so I modified the sentence as “during the months that followed the accident”.

what type of field observations?
I modified the sentences as “field observations (ground aerosol sampling: Masson et al., 2011, 2013; Kaneyasu et al., 2012; Adachi et al., 2013; Tsuruta et al., 2014; Igarashi et al., 2015; Oura et al., 2015, aircraft measurements: NRA, 2012, and afoot measurements: Hososhima and Kaneyasu, 2015)”.

to simulate emissions....?
I added “of transport and depositions”.

GENERAL REMARK HERE:
I would only provide 3 representative references for each item
I would like to show the significant differences in numbers for the study of air concentrations, during the crisis phase (March 2011) and post-accidental phase (this study select 2013 as an example).

(P.3)
replace 'but' with 'and'
I changed it.

I would make two sentences here, and start the second one with 'However,...'
I changed it.

replace with: 'for unmonitored locations'
I changed it.

(P.4)
Although
I changed it.

remains possible
I changed it.

use 'activities' to avoid repetition of 'concentration'?
I changed it.

please rephrase
I rephrase it to “They found substantial amounts of bioaerosols upon scanning electron microscopy samples collected in the summer, ”

You could stress the fact that with 'Fukushima', you mean 'Fukushima Prefecture' to avoid confusion for the readers
I changed it to “(Namie town, Fukushima prefecture)” and to be consistent changed “(Tsukuba, Ibaraki)” to “(Tsukuba city, Ibaraki prefecture)”. I changed them in the abstract accordingly.

collected?
I changed it to “conducted”, instead.

please rephrase
The current sentence has been deleted according to the Referee #1’s comment (RC3).

these?
The current sentence has been deleted according to the Referee #1’s comment (RC3).

please avoid repetitions with the text above (LL. 18-20)

I changed the sentence to “By utilizing the observational data both inside and outside together with the transport model,”.

>> a robust analysis?
I changed it.

>> the
I added it.

(P.5)
crucial to understand?
I remained the sentence as it was.

low to moderately?
I changed it.
low/large?
I added “large”.

(P.6)
level?
I meant “schoolyard” here. I changed it.

(P.7)
do you mean runoff/erosion?
Yes, runoff, erosion, percolation and all the processes resulting in radiocesium migration in the soil and biota.

Furthermore?
I changed it.

where are they mentioned? I missed this...
I added the phrase as “into the four above-mentioned four categories (i.e., sand, loamy sand, sandy loam, and silt loam)”.

(P.8)
>> an analogue

I remained the word as it was.

= bare ground?
According to the referee #1's comment (RC3), I have deleted the sentence.

it would be useful to add in 1-2 lines how they explain this resuspension in forests?
They didn't explain the mechanisms and just stated the fact. Anyway, according to the referee #1's comment (RC3), I have deleted the sentence.

(P.9)
> the previous versions
I changed it.

However
I changed it.

(P.10)
I changed it.

please provide the max period
I found the max period is 2 days for the analysis period (year 2013), and so the phrase in the parenthesis was removed.

this is rather confusing!
In the revised manuscript, the Tsushima site is referred to as Namie (sometimes as Namie (Tsushima)) and the Omaru site is referred to as Omaru (sometimes as Namie (Omaru)) throughout the manuscript.

remained
I changed it.

(P.11)
aerosols carrying...?
I changed it.

to

I added it.

(P.12)

in agreement...?

I changed it.

valid?

I changed it.

please rephrase

I changed it to “rather give simulation results consistent with the available observations”.

please rephrase >> you mean that it was simulated but not observed?

I rephrased it.

(P.13)

used?

I added it.

in order to adjust?

I changed it.

unclear to me, please rephrase

I rephrased it to “in order to adjust”.

facilitated?

I changed it.

unclear what you mean here

If the tuning parameter varies in time and space, the source of variation in the simulated concentration is hardly identified, whether it is coming from the varying tuning parameters or from the model variation itself (i.e., emission and meteorology). I rephrased it from “by keeping the simulated variation as it was” to “by keeping the simulated variation solely originating from the variation of boundary conditions (i.e., emission and meteorology)”.

remove 'in'

I removed it.

or soil erosion?

Yes, and runoff, percolation and all the processes resulting in radiocesium migration in the soil and biota. The definition of land surface processes are additionally written in the beginning part of the revised manuscript, in Sect. 2.2.

AFTER decontamination, as DURING decontamination works, this flux should increase

Thank you for your suggestion. This is a very important implication and I added the following sentence in the Introduction section: “Although the decontamination-related work could be a potential source of re-suspension, it is not considered in the current simulation as the re-suspension flux has been hardly quantified.” The similar statement was added to Sect 5.4 (previously Sect. 5.3) as “The decontamination may reduce resuspension afterward, whereas the resuspension may occur during decontamination-related work. This effect should be evaluated in the future.”

(P.14)

unclear what you mean here

I changed it to “continuously presenting emissions, such as natural emissions, and not accidental ones.”

unclear what you mean here

I changed it to “from the highly contaminated areas such as within the premises of FDNPP (e.g. debris removal operations) or very close to FDNPP on these days, as indicated later in Sect. 5.2)

please rephrase

I changed it to “inside and outside”.

where to? The Pacific Ocean? Other Japanese regions?

I changed it from “out of the region” to “out of the model domain”. We cannot tell exactly where to but anyway toward out of the model domain, which only covers a part of Japan.

(P.15)

??

It meant “The discrepancy between the observed peak and the dust simulation could be due to underestimation of the simulation but it is less likely because the simulated dust peak reached an intensity of 4 – 5 mBq/m^3 in other days in the winter, which is of the same order of magnitude as that of the observed peak.” But I deleted the sentences because I judged it is not necessary here.

(P.16)

??

In winter in Japan, northwesterly wind prevails due to Siberian high, which is called winter monsoon. On the other hand, southerly wind prevails due to Pacific high, which is called summer monsoon, in Japan.

(P.17)

this is unclear, please rephrase

“around Tsukuba” is added at the end of sentence.

a line of explanation would be welcome here

“due to higher surface wind speed in the cold season” is added at the end of sentence.

> constant emission sources?

I changed it from “constant emission” to “continuous emission sources”.

(P.19)

Not sure whether the journal recommends to structure the perspectives section as a list of 'bullet points'

I modified the section by excluding list of items.

>> could be rephrased and could usefully include suggestions/hyptheses of potential mechanisms driving the observed processes

Based on the approaches used in the study, the authors cannot hypothesize the mechanisms of the emissions. This should be done by other studies on experimental or theoretical basis.

what do you mean exactly here?

I meant “improved”. I modified the sentence as follows “The module needs to be

improved to be applicable to various land use and soil texture conditions".

this
I changed it.

(P.20)
during?
I changed it.

(P.25)
why it is separated from the main text?
I moved the Appendix C in the main text as Sect. 5.3. Thank you for your suggestion.

why
This is just a rule of data sampling. Other rules can be applied for example to pick up stations showing median dose rate at 0:00 a.m., January 1, 2014. No matter how data were sampled, a conclusion we extract here would not be changed: Decreasing rate in gamma dose rates due to re-suspension was two to three orders of magnitude smaller than the gross decreasing rate including radioactive decay, decontamination, and the land surface processes.

(P.27)
Cs might be transported with soil as well, see for instance Evrard et al. (2015); J . Env. Rad; for a review on these processes
Thank you for introducing the very important work. I modified the last paragraph of Sect. 5.3 (previously Appendix C) referring their work as "Evrard et al. (2015) summarized that significant transfer of particulate-bound radiocesium occurs during major rainfall and runoff events (typhoons and spring snowmelt) …".

(P.44) Figure 8.
a ratio is not a %...
I changed from "ratios" to "fractions".

Dear anonymous referee #3,

We very much appreciate your constructive comments, useful information and your time for RC2. Especially, your suggestion on the changing definition of p_{20} from mass fraction to surface area fraction was really helpful. Thanks to your review, our manuscript was substantially improved. Point-by-point responses to your comments are written in blue in this letter.

Best regards,
Mizuo Kajino

General comments:

This paper estimated significance of the resuspension/deposition processes in the Cs-137 budget at the ground surface in a wide area in the northern part of Japan. Although the conclusion that the resuspension is insignificant in changing the contamination distribution is somewhat too obvious, the procedures and discussions that result in this conclusion are pertinent and informative. It is also interesting that the different sources were found to account for the air concentration variations in the different seasons. Since technical comments were already made in the previous reviewing process, the reviewer raises some points for discussion here.

Thank you for your evaluation.

Specific comments:

1. The resuspension scheme in this paper (p.8, line 8-24) assumes that the Cs-137 flux is in proportion with that of the soil mass. This obviously is too crude an assumption to make in contrast to other sophisticated formulations for resuspension since the activity concentration is usually much higher in a fine particle fraction due to its larger specific surface area. This assumption may result in considerable underestimation of Cs-173 resuspension, and is highly probably one of main causes necessitating the unphysical parameter of 10 (p.8, line 31). Discussion on this point must be included in the text.

- I fully agree with your point. We made re-calculation by changing the definition of p_{20} from mass fraction to surface area fraction. Because the dominant soil texture over the contaminated area was sandy loam and the ratio of surface-area-based p_{20} (=0.45) to mass-based p_{20} (=0.09) of sandy loam was approximately five, the re-calculated ^{137}Cs emission from soil increased by a factor of five for the whole region and time. On the other hand, a bug has been found in the previous simulation: $\rho_{b,soil}$ in Eq.(3) was calculated as porosity (0.4) times dust particle density (2650), but correctly, $\rho_{b,soil}$ is 1 minus the porosity (=0.6) times the density (2650), and so the corrected concentration decreased by 0.4/0.6 = 0.6666. Therefore, the final dust concentration is the previous dust concentration times 5 times 0.6666 = 3.3333. Consequently, the previous dust concentration times 10 is equivalent to the final dust concentration times 3. Because the previous concentration times 10 slightly underestimated the observation, the "unphysical parameter" of 5 was chosen for the final revised simulation. Please see that the observed and simulated medians with the parameter of 5 are consistent with each other as shown in Table 3, a new table in the revised manuscript. All the figures (i.e., 7-12) and quantities in the text have been replaced, accordingly.

2. For the same reason, the statement (p.14, line 1) "the flux might be a maximum estimate" seems impertinent. If the authors determined the rule-of-thumb value of 10 for the above-mentioned parameter to have reasonable air concentration values, the flux might not be a maximum estimation.

   As replied previously, we made the re-calculation using a surface-area based fraction. Now the new simulation is a maximum estimation in a sense that it does not consider any suppression effects due to precipitation, snow cover and land surface processes.

3. The results of sensitivity analysis in Table. 2 (the "range" line) is not informative. The reviewer cannot tell what kind of sensitivities exists from the ranges of statistical values.

   Yes, exactly. But the point here is the range of statistical values after optimization becomes generally smaller compared to the ranges after optimization, indicating that the optimization is successful excluding deposition parameters with worse performances. As to the "a kind of sensitivity exists in the ranges of statistical values", the relationship between the input parameters (deposition parameters) and statistical measures was elaborated from line 26 of page 12 in the previous

manuscript as “Generally speaking *R* became higher …”. The author judged that the statement on the relationship was hardly reflected in Table 2, and so we just showed the ranges in Table 2 and described the relationship in the main text, separately. Thank you for your understanding.

4. There are statements that the surface air concentration has positive correlation with the surface wind speed (p.15, line 26 and p.26, line 15). However, there is no evidence of it. At least a statistics (e.g. correlation coefficient) is necessary.
   I added the correlation coefficients to the main text.

5. The discussion in Appendix C should be presented in the main text since it is substantial in discussing the significance of resuspension quantitatively. However, it is recommended that the last part of this part (p.27, Line 11-15) be changed or deleted since research has been done extensively; for example a paper by Kimiaki Saito and Nina Petoussi-Henss, Journal of Nuclear Science and Technology, 2014, Vol. 51, No. 10, 1274–1287, http://dx.doi.org/10.1080/00223131.2014.919885 has discussed the migration-dose rate relation. The group headed by Dr. Kimiaki Saito also conducted extensive field measurements on the dose rate trend and in-soil concentration distribution.
   Thank you for your suggestion and introducing Dr. Saito’s work. I moved Appendix C into the main text as Sect. 5.3 and changed Figure C1 to Figure 14. I deleted the corresponding paragraph and modified the sentence referring their work as well as Evrard et al. (2015) as follows: “Evrard et al. (2015) summarized that significant transfer of particulate-bound radiocesium occurs during major rainfall and runoff events (e.g. typhoons and spring snowmelt). Together with the relaxation depth – dose rate relationship provided by Saito and Petoussi-Hess (2014), the decreasing rate due to land surface processes such as downward migration, runoff, and erosion could be quantified and thus the decontamination effect could be separately extracted.

Long-term assessment of airborne radiocesium after the Fukushima nuclear accident: Re-suspension from bare soil and forest ecosystems

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Abstract

The long-term effect of ^{137}Cs re-suspension from contaminated soil and forests due to the Fukushima nuclear accident has been quantitatively assessed by numerical simulation, a field experiment on dust emission flux in a~~the~~ contaminated area (Namie town, Fukushima prefecture), and air concentration measurements inside (Namie) and outside (Tsukuba city, Ibaraki prefecture) the contaminated area. In order to assess the long-term effect, ~~The assessment period is for~~ the full year of 2013 was selected to study just after the start of the field experiments~~, December 14, 2012~~. The ^{137}Cs concentrations at Namie and Tsukuba were approximately 10^{-1} – 1 and 10^{-2} – 10^{-1} mBq/m^3, respectively. The observed monthly median concentration at Namie was one to two orders of magnitude larger than that at Tsukuba. This observed difference between the two sites was consistent with the simulated difference, indicating successful modeling of ^{137}Cs re-suspension and atmospheric transport. The

estimated re-suspension rate was approximately 10^{-6} /d, which was significantly lower than the decreasing rate of the ambient gamma dose rate in Fukushima prefecture (10^{-4} – 10^{-3} /d) as a result of radioactive decay, migration in the soil and biota~~land surface processes (migration in the soil and biota)~~, and decontamination. Consequently, re-suspension contributed negligibly ~~to~~ in reducing ground radioactivity. The dust emission model could ~~account for~~reproduce the air concentration of ^{137}Cs in winter, whereas the summer air concentration was underestimated by one to two orders of magnitude. Re-suspension from forests at a constant rate of 10^{-7} /h, multiplied by the green area fraction, ~~quantitatively accounted for~~could explain the air concentration of ^{137}Cs at Namie and its seasonal variation. The simulated contribution of dust re-suspension to the air concentration was 0.~~6~~ 7 – 0.~~8~~ 9 in the cold season and 0.~~1~~ 2 – 0.4 in the warm season at both sites; the remainder of the contribution was re-suspension from forest. The re-suspension mechanisms, especially through the forest ecosystems, remain unknown.~~, and thus the current study is the first but~~This is the first study that provides a crude estimation of the long-term assessment of radiocesium re-suspension. ~~Further study will be~~Additional research activities should investigate the processes/mechanisms governing ~~needed to understand~~ the re-suspension ~~mechanisms~~ over the long term. ~~and to accurately assess~~This could be achieved ~~the re-suspension mechanisms~~ through conducting additional field experiments and numerical simulations.

Keywords: Atmospheric radioactivity, Re-suspension, Dust emission, Unknown re-suspension source, Aerosol, Numerical simulation, Budget analysis, Japan

1 Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accidentally released nuclear fission products into the atmosphere and the ocean environment following the catastrophic earthquake and tsunami that occurred in March 2011. The accident caused serious contamination of the ~~ground~~ soil over the Tohoku region (northeastern part of Japan, including Fukushima and Miyagi prefectures) and the Kanto region (eastern part of Japan, including Ibaraki, Tochigi, Gunma, and Chiba prefectures) (NRA, 2012). Since then, a number of studies have been conducted, particularly ~~in the crisis phase of the disaster~~during the months that followed the accident. These assessments include primary emission estimations (Chino et al., 2011; Danielache et al., 2012; Stohl et al., 2012; Terada et al., 2012; Katata et al., 2012a, 2012b; Winiarek et al., 2012, 2014; Hirao et al., 2013; Saunier et al.,

2013; Katata et al., 2015; Yumimoto et al., 2016; Danielache et al., 2016), field observations (ground aerosol sampling: Masson et al., 2011, 2013; ~~NRA, 2012;~~ Kaneyasu et al., 2012; Adachi et al., 2013; Tsuruta et al., 2014; ~~Hososhima and Kaneyasu, 2015;~~ Igarashi et al., 2015; Oura et al., 2015, aircraft measurements: NRA, 2012, and afoot measurements: Hososhima and Kaneyasu, 2015), and numerical simulations of transport and depositions (deterministic simulation: Chino et al., 2011; Morino et al., 2011; Yasunari et al., 2011; Stohl et al., 2012; Terada et al., 2012; Katata et al., 2012a, 2012b; Winiarek et al., 2012, 2014; Hirao et al., 2013; Saunier et al., 2013; Katata et al., 2015; Yumimoto et al., 2016; Danielache et al., 2016, deterministic simulation with sensitivity runs: Morino et al., 2013; Adachi et al., 2013; Groëll et al., 2014; Saito et al., 2015; Sekiyama et al., 2015; Quérel et al., 2016, uncertainty modeling and probabilistic forecast: Girard et al., 2016; Sekiyama et al., 2016, and multi-model inter-comparison and multi-model ensemble analysis: SCJ, 2014; Draxler et al., 2015: Kristiansen et al., 2016). The targeted radionuclides were species with both short and long half-lives: ^{99}Mo-^{99m}Tc (half-life 65.9 – 6 h), ^{129m}Te (33.6 d), ^{131}I (8.02 d), ^{132}Te-^{132}I (3.2 d – 2.3 h), ^{134}Cs (2.07 y), ^{136}Cs (13.2 d), ^{137}Cs (30.1 y), ^{133}Xe (5.2 d), and ^{35}S (87.5 d).

In contrast, there have been few studies on the long-term (more than one year) quantitative assessment of radioactivity in the atmosphere associated with the Fukushima accident (Igarashi et al., 2015; Ishizuka et al., 2016; Kinase et al., 2016). More than 100,000 people were evacuated (METI, 2012), ~~but~~ and most have still not been able to return to their homes and the public remains anxious about the safety of the affected areas. Radionuclides with long half-lives such as ^{134}Cs (2.07 y) and ^{137}Cs (30.1 y) are of particular concern (Evangeliou et al., 2014). Following the Chernobyl accident there were several studies on the re-suspension and long-term assessment of these radionuclides, such as Holländer and Garger (1996), Garger et al. (1998), Hatano and Hatano (2003) and Garger et al. (2012). For example, Garger et al. (2012) estimated the re-suspension “descending trend” as having a half-life of 300 d based on the surface activity concentration of ^{137}Cs. In the case of the Fukushima accident, Igarashi et al. (2015) estimated the half-reduction time by fitting multi-component exponential functions based on the ^{137}Cs concentration at the Meteorological Research Institute~~,~~ (Tsukuba)~~,~~ as 5.9 d, 16 d, and 1.1 y. These estimates were based on the trend in the observed surface air concentrations of ^{137}Cs, and thus the contributions from advection, diffusion, emission and deposition terms were not quantified.

There are thousands of monitoring posts situated in the contaminated area in Fukushima prefecture to measure the ambient gamma dose rate.~~,~~ ~~but~~ Hoever, the data cannot be used for evaluating internal exposure: evaluation of internal exposure requires direct measurement of the surface air activity concentration. There are only a few observation sites that continuously measure the concentration of radiocesium (e.g., Igarashi et al., 2015; Ishizuka et al., 2016; Kinase et al., 2016). To assess the spatial distribution of the internal exposure hazard, 3D numerical simulation is necessary to interpolate values ~~at locations where there are no measurements~~for unmonitored locations. The numerical simulation requires emission flux as a boundary condition. However, the mechanism and thus the radioactivity flux associated with the re-suspension of ^{137}Cs were unknown, despite extensive efforts based on field observations (e.g., Igarashi et al., 2015; Ishizuka et al., 2016; Kinase et al., 2016).

Garger et al. (2012) summarized the re-suspension sources following the Chernobyl accident as (1) dust emission, (2) human activity in fields, and on roads and construction sites, (3) forest fires, and (4) emissions from the power plant (i.e., opening of the Chernobyl sarcophagus). Re-suspension sources (1) dust emission (Ishizuka et al., 2016) and (4) i.e., additional emissions from the reactor buildings of FDNPP (TEPCO, 2012; 2013; 2014a; 2014b; 2015) were considered in the present study. With respect to source (2), since Namie town~~, Fukushima,~~ is located in the evacuation zone, human activity has been extremely limited except for decontamination-related work. Although the decontamination-related work could be a potential source of re-suspension, it is not considered in the current simulation as the re-suspension flux has been hardly quantified. ~~-~~As to source (3), there is a low ~~chance~~ risk of forest fires in Japan given the high humidity ~~but~~ although some open biomass burning ~~is a possibility~~remains possible. Kinase et al. (2016) found no increase in ^{137}Cs ~~concentration~~ activities when the concentration of levoglucosan, a marker of biomass burning, was increased, and thus re-suspension due to biomass burning was not considered in the present study. In addition to the four sources of ^{137}Cs from the Chernobyl accident, re-suspension from terrestrial biota was considered as suggested by Kinase et al. (2016). They found substantial amounts of bioaerosols upon scanning electron microscopy ~~examination of~~ samples collected in the summer, when ^{137}Cs concentration was high.

In the present study, the long-term effect of radiocesium re-suspension from contaminated soil and terrestrial biota was quantitatively assessed using 3D numerical simulation, a field experiment on dust emission flux in a contaminated area (Namie town, Fukushima prefecture),

and air concentration measurements ~~taken~~ conducted inside (Namie) and outside (Tsukuba city, Ibaraki prefecture) the contaminated area. ~~The current study is the first but crude estimation of the spatial budget of radiocesium via re-suspension since the re-suspension mechanisms, especially through forest ecosystems, remain unknown.~~ Even though the re-suspension mechanisms still remain unknown, ~~By~~ by utilizing the observational data ~~collected~~ both inside and outside ~~of the contaminated area~~, together with ~~3D numerical simulation~~the transport model, we aimed to provide as a robust ~~a budget~~ analysis as possible of the re-suspension, transport, and re-deposition of ^{137}Cs over the Tohoku and Kanto regions of Japan.

2 Numerical simulation

A brief description of the numerical method, such as the processes considered in the model and simulation settings, are presented in this section, and detailed model formulations are described in Appendix A. Because the schemes and assumptions regarding the emissions are key to the current study, they are described in detail in the following subsections.

2.1 Lagrangian Model and simulation settings

Figure 1 shows the domain of the Lagrangian Model (LM) with model terrestrial elevations, covering 138 – 143 °E and 34 – 39 °N. The model domain covers the southern part of the Tohoku region (the northern mountainous part of the domain, including Yamagata, Miyagi, and Fukushima prefectures), and includes the FDNPP and highly polluted areas such as the Habitation-Restricted Zone (HRZ) (20 – 50 mSv/y) and Difficult-to-Return Zone (DRZ) (> 50 mSv/y) (METI, 2012). It also covers the Kanto region (or Kanto Plain, the largest plain in Japan, approximately 120 km × 120 km), a highly populated region that includes low to moderately polluted areas such as Tokyo, Gunma, Tochigi, Ibaraki, Saitama, and Chiba prefectures.

LM considers horizontal and vertical diffusion and advection, gravitational settling, dry and wet depositions, and radioactive decay. It uses simple parameterizations for dry and wet deposition schemes, and it can be driven by meteorological analysis data sets so that it does not require a meteorological model to predict detailed meteorological fields and variables. The model was designed to be easily handled and computationally efficient so that non-specialists of numerical simulations can conduct long-term assessments of atmospheric diffusion problems using their desktop or laptop computers. The LM was designed for rough

budget estimates, as presented in the current study, or for sensitivity analyses using a large number of parameters (e.g., Groëll et al., 2014; Girard et al., 2016; Quérel et al., 2016), rather than for process-oriented analysis (e.g., Morino et al., 2013; Katata et al., 2015) or sensitivity analyses of the physical and chemical parameters of aerosols (Adachi et al., 2013). Details of each process and parameter are described in Appendix A. Statistical error of a Lagrangian simulation is inversely proportional to the square of the number of Lagrangian particles (LPs). The statistical accuracy of the current simulation setting is discussed in Appendix B.

The Grid Point Value Meso-Scale Model (GPV-MSM) of the Japan Meteorological Agency (JMA) was used for meteorological analysis to calculate the transport of LPs. It covers 120 – 150 °E and 23 – 47 °N and provides 3 hourly and 16 pressure levels of 3D meteorological variables, from 1000 hPa to 100 hPa, with a horizontal grid resolution of approximately 11 km (Δlongitude = 0.125° and Δlatitude = 0.1°) and surface variables at twice the resolution as that for the 3D variables (Δlongitude = 0.0625° and Δlatitude = 0.05°). In the simulation, the whole model domain where LPs can travel is 138 – 143 °E, 34 – 39 °N and from ground surface to 500 hPa. For output of the model results, LP fields are converted to Eulerian concentration (Bq/m^3) and deposition (Bq/m^2) fields in the same horizontal space as the 3D variables but are vertically allocated from the ground surface to an altitude of 1 km at 100 m intervals. The observed surface air concentration was compared with the simulated mean concentration at 0 – 100 m above ground level (AGL).

2.2 Re-suspension from bare soil

Ishizuka et al. (2016) developed a re-suspension scheme for radiocesium from bare soil based on measurements on the ~~ground~~ schoolyard at Namie High School, Tsushima Campus (denoted as Namie (Tsushima) in Table 1 and Fig. 1) in the DRZ.

$$F_{soil} = p_{20\,\mu m} F_M (1 - f_{forest}) B_{5\,mm}(t), \quad (1)$$

where F_{soil} is the ^{137}Cs dust re-suspension flux from soil (Bq/m^2/s), $p_{20\mu m}$ is the ~~mass~~ surface area fraction of dust smaller than 20 μm in diameter against soil containing a maximum size of 2 mm particles, and varies depending on soil texture (~~2~~1.3×10^{-9} – ~~8~~ for sand, 0.~~03~~ 19 for loamy sand, 0.~~09~~ 45 for sandy loam, and 0.~~32~~ 80 for silt loam), F_M is the total dust mass flux (kg/m^2/s), f_{forest} is the forest area fraction, and $B_{5mm}(t)$ is the specific radioactivity of surface soil (from the surface to a depth of 5 mm) (Bq/kg) as a function of time since March 2011. The formula is based on the assumption that dust particles smaller than 20 μm in diameter

originated from the surface soil and to a depth of 5 mm were suspended and transported through the atmosphere. The surface area fraction was used for $p_{20\mu m}$ based on the assumption that radiocesium is bound to the surface of soil particles (Evrard et al., 2015). F_M is formulated as being proportional to the cube of the friction velocity u_* (m/s) as described by Loosmore and Hunt (2002) and was applied to the dust emission:

$$F_M = 3.6\times10^{-9}u_*^3. \quad (2)$$

Since u_* is not available in GPV-MSM, u_* was estimated using a wind speed at 10 m AGL by assuming neutral stratification conditions.

$B_{5mm}(t)$ was derived from the combination of B_{obs}, the observed horizontal distribution of ^{137}Cs deposition obtained from an airborne radiological survey (NRA, 2012) (Bq/m^2) and r_{5mm}, the surface soil activity ratio of 0 – 5 mm to 0 – 5 cm obtained from a vertical profile measurement of ^{137}Cs in the ground soil at Namie High School (= 0.57 Bq/Bq) as

$$B_{5\,mm}(t) = \frac{B_{obs}r_{5mm}R_{decay}(t)}{5\times10^{-3}\rho_{b,soil}}, \quad (3)$$

where $\rho_{b,soil}$ is the bulk density of soil particles per unit volume in the ground space (kg/m^3) obtained from the porosity (0.4 m^3/m^3) and the density of dust particles (2650 kg/m^3). For R_{decay}, which is the decreasing rate of activity in the ground, only radioactive decay was considered for the re-suspension calculation. The decreasing rate due to other processes such as land surface processes (e.g., runoff, erosion, percolation and all the processes resulting in migration of radiocesium in the soil and biota; Evrard et al., 2015; Matsuda et al., 2015) ~~(or migration in the soil and biota)~~ and decontamination were not considered here. ~~Also~~Furthermore, suppression of dust emission due to soil moisture and snow cover was not considered. Therefore, it should be noted here that F_{soil} in Eq. (1) is considered as the upper boundary ~~a maximum estimate~~ of ^{137}Cs re-suspension flux from surface soil. Effects such as land surface processes, decontamination, and dust emission suppression due to snow cover are extensively discussed in ~~Appendix C~~Sect. 5.3 using ambient gamma dose rate measurements obtained by the monitoring posts in Fukushima prefecture ~~(Table 1, Fig. 1b, and Fig. C1)~~.

Eq. (1) is a function of soil texture. The areal fraction of soil texture of the model grid was obtained from the database of the advanced research Weather Research and Forecasting model version 3 (WRFV3; Skamarock et al., 2008). Sixteen categories of soil texture (Miller and White, 1998) with a 30 arcseconds resolution dataset can be obtained from the web after

subscription at (http://www2.mmm.ucar.edu/wrf/users/download/get_sources_wps_geog.html, last access: ~~12 February~~26 July 2016) and were re-categorized into the above-mentioned four categories (i.e., sand, loamy sand, sandy loam, and silt loam) and interpolated to the LM resolution (~11 km) as shown in Fig. 2a-c. Note that the loamy sand fraction is not presented because it is zero for the entire domain. The parameter f_{forest} (Fig. 2d) was also obtained from the database of WRFV3 and was calculated based on the 24 United States Geological Survey (USGS) Land Use Categories, which are constant over time. The Land Use Category dataset can also be obtained from the above website.

Ishizuka et al. (2016) validated their dust emission module by using a 1D model and observed the surface air concentration of ^{137}Cs at Namie in the winter. After applying the module to our 3D simulation, we found that the air concentration at Namie was underestimated by about ~~one order of magnitude~~factor of five for the same period. The module was formulated based on physical parameters (such as u_*) but contains parameters obtained at a single location (such as r_{5mm} and $\rho_{b,soil}$) and under a fixed atmospheric condition (Ishizuka et al., 2016), whereas ideally parameters in Eqs. (1) – (3) should have considered variations among locations and atmospheric conditions for the 3D simulation. We simply multiplied the dust emission flux by five ~~set the parameter to 10~~ after adjusting the simulation results against the observed concentration of ^{137}Cs at Namie in the winter. This is one of the simplest top-down approaches for adjusting the emission flux according to the air concentration. The module requires improvement in the future as more reliable parameters become available for various conditions and locations.

2.3 Re-suspension from the forest ecosystems

The re-suspension mechanism of radiocesium from land ecosystems remains unknown. Kinase et al. (2016) found substantial amounts of bioaerosols (rather than mineral dust particles) in samples collected for scanning electron microscopy in the summer, when the ^{137}Cs concentration was high. This does not prove that the bioaerosol was carrying radiocesium, but that it could be a potential carrier. The behavior of Cs in the environment can be inferred by analogy with K, a congener of Cs. Potassium is a necessary and abundant element in plants and circulates between land ecosystems. The addition of potassium fertilizer to a rice field in Fukushima significantly reduced the Cs content of the rice (Ohmori et al., 2014). Substantial amounts of K-salt-rich particles, possibly emitted by active biota such as plants and fungi, and coated with secondary organic aerosols, were observed in pristine

Amazonian rainforest (Pöhlker et al., 2012). The major areal fraction of the contaminated area in Fukushima is covered by biota-rich mountain forests ~~(Figs. 1a, 2d, and 4a)~~. Despite the differences in plant species and locations, it is plausible that water-soluble radiocesium circulating in the biota and soil in the forests was somehow re-emitted into the atmosphere and contributed to the surface air concentration. The re-suspension from the forest ecosystem was simply formulated as follows:

$$F_{forest} = f_{forest} f_{green} r_{const} B_{obs} R_{decay}(t), \quad (4)$$

where F_{forest} is the ^{137}Cs re-suspension flux from forest (Bq/m^2/s), f_{green} and r_{const} are the monthly green area fraction and the constant re-suspension coefficient (/s), respectively, and r_{const} is a tunable parameter to adjust the simulated air concentration of ^{137}Cs to that observed. In the current study, r_{const} is set to 10^{-7} /h by adjusting the simulation data using the observed ^{137}Cs concentration at Namie in the summer, when the re-suspension from soil was negligible due to the higher soil moisture content (following considerable rain) and lower wind speed. As with re-suspension from bare soil, only radioactive decay was considered for R_{decay} and the other processes were not considered. ~~Unlike re-suspension from soil, precipitation might not suppress re-suspension from the forest ecosystems since substantial amounts of K-containing particles were observed in the wet season in the Amazon (Pöhlker et al., 2012 and references therein).~~ The parameter f_{green} was obtained from the database of WRFV3 and was originally derived from satellite Advanced Very High Resolution Radiometer (AVHRR)/Normalized Difference Vegetation Index (NDVI) data (Gutman and Ignatov, 1998). Whereas f_{forest} remains constant, the monthly averaged f_{green} was used in order to reflect seasonal changes in the activity of the biota.

2.4 Emission from FDNPP (primary emission, additional emissions from the reactor buildings, and unexpected re-suspension associated with debris removal operations)

Katata et al. (2015), the Japan Atomic Energy Agency (JAEA)'s latest estimate of the primary emission from FDNPP, was applied for the emergency situation of March 2011, to evaluate the performance of the LM model against the horizontal distribution of ^{137}Cs deposition of the airborne radiological survey (NRA, 2012) (as shown later in Fig. 4a) and surface air concentrations measured at Tsukuba (Fig. 1a). We selected this inventory because it is JAEA's most up-to-date version. Based on an integrated understanding of environmental

radioactivity, atmospheric dispersion, and the nuclear reactors, the JAEA team has carefully established a series of inventories for about five years, staring with Chino et al. (2011), followed by Katata et al. (2012a), (2012b), Terada et al. (2012), and finally the current inventory (Katata et al., 2015), which is substantially improved compared to the previous versions~~its predecessors~~.

Ongoing emissions during the study analysis period after the emergency situation, that is, January to December 2013, was obtained from the Tokyo Electric Power Co., Inc. (TEPCO) monthly mean emission flux from the reactor buildings (TEPCO, 2012; 2013; 2014a; 2014b; 2015). Because only the sum of ^{134}Cs and ^{137}Cs was provided, the fractions of these two isotopes were calculated based on their half-lives and the assumption that their activities were equal in March 2011 (e.g., Katata et al., 2015), as shown in Fig. 3. The values range from 10^5 to 10^7 Bq/h, ~~but~~ however for simplicity we set a constant value of 10^6 Bq/h in the current simulation.

In August 2013, unexpected re-suspension associated with debris removal operations was reported by TEPCO (2014c) and NRA (2014) and the gross amount was 10^{10} – 10^{11} Bq of ^{137}Cs (TEPCO, 2014c, NRA, 2014, Steinhauser et al., 2015). The impact of this unexpected re-suspension is briefly discussed in Sect. 5.2 along with an additional finding, but this emission was not considered in the present LM simulation. In this study we focused on the ongoing and continuous emission, mostly from the natural environment, that is difficult to control.

3 Field observations

Details of the surface air activity concentration measurements techniques can be found in Ishizuka et al. (2016) and Kinase et al. (2016) for Namie and Igarashi et al. (2015) for Tsukuba. At the both sites, ~~ambient aerosols were collected using a high-volume air sampler and~~ ^{134}Cs and ^{137}Cs surface air concentrations were measured. ~~obtained by γ-ray spectroscopy using a Ge semiconductor detector.~~ The sampling intervals were 1 – 2 d ~~(sometimes several days)~~ at Namie and 1 w at Tsukuba for the analysis period of this study, the year 2013. The observations at Namie started on December 14, 2012, while those at Tsukuba started on March 31, 2003, before the FDNPP accident. In March 2011, the sampling interval was increased at Tsukuba to 6 h – 1 d and the data for these periods were used for the validation of LM and its parameters, as presented in Sect. 4.1.

The Namie site is located on the ~~ground~~ schoolyard ~~at~~ of Namie High School, Tsushima Campus, in the Tsushima district of Namie town in Fukushima prefecture, as shown in Table 1 and Fig. 1. Namie town extends from the Hamadori coastal area (denoted as C in Fig. 1) to the Abukuma highland area (B in Fig. 1). There are two sites in Namie town used in the study. In order to distinguish the Tsushima Campus site in the highland area from the monitoring post site located in Omaru district in Namie town in the coastal area, the Tsushima site is referred to as Namie (sometimes as Namie (Tsushima)) and the Omaru site ~~are~~ is referred to as Omaru (sometimes as Namie (Omaru)) throughout the manuscript. ~~as Namie (Tsushima) and Namie (Omaru), respectively. Note that, unless specifically referred to as Namie (Omaru), Namie without brackets indicates Namie (Tsushima) throughout this manuscript.~~ The Tsukuba site is located on the premises of the Meteorological Research Institute (Table 1 and Fig. 1a).

Namie (Tsushima) was located in the DRZ (> 50 mSv/y, ~9.55 μSv/h [1]) and the observed ^{137}Cs deposition amount was 2,300 kBq/m^2 (~~Fig. 4a~~NRA, 2012). The ambient gamma dose rate was 11.2 μSv/h on April 1, 2012 at the site and had dropped to 4.8 μSv/h on Feb 16, 2016, and at the HRZ level (20 – 50 mSv/y, 3.85 – 9.55 μSv/h). Tsukuba is located approximately 170 km southwest of FDNPP. The observed ^{137}Cs deposition amount was 21 kBq/m^2 (~~Fig. 4a~~NRA, 2012), two orders of magnitude lower than at Namie and the dose rate has ~~been~~ remained below 0.1 μSv/h since 2012.

4 Results

Section 4.1 presents a validation of the LM model and the optimization of the model deposition parameters by using airborne observations (NRA, 2012) and the emission inventory of Katata et al. (2015) for the emergency situation of March 2011. Using the optimized ranges of model parameters validated in Sect. 4.1, the simulated re-suspension of ^{137}Cs from soil and forest, and emission from FDNPP, is presented in Sect. 4.2, and the budgets for re-suspension, transport, and re-deposition are presented in Sect. 4.3.

[1] Calculated by the following equation: Annual radiation exposure = (ambient dose rate – background dose rate (=0.04 μSv/h)) × (8 h + 0.4 × 16 h) × 365 d.

4.1 Model and parameter validation for the emergency situation (March 2011)

Figures 4 and 5 show the observed and simulated distribution of ^{137}Cs deposition in March 2011, and the scatterplot~~gram~~ comparing the observational and simulation results~~, respectively~~. In the simulation shown in the figures, the "reference" sets used for dry and wet deposition parameters, namely, the collection efficiency of aerosols using hydrometeors E_c (Eq. A2) and the dry deposition velocity over land v_d (Eq. A4), were 0.04 and 0.1 cm/s, respectively.

Since LM ~~employs~~ uses simple parameterizations for dry and wet deposition, as described in Appendix A, sensitivity tests were conducted for model validation, together with optimization of the deposition parameters. Table 2 summarizes the ranges of the deposition parameters for the sensitivity tests and the results of the ^{137}Cs budget and statistical measurements between the observations and the simulation.

The parameter E_c was 0.05 for the JMA dispersion model (JMA, 1998) but the targeted species are different. For example, volcanic ash (particles larger than 1 μm in diameter) used for the JMA model were generally larger than the ~~carrier~~ aerosols ~~of~~ carrying ^{137}Cs (around 1 μm in diameter observed in the downwind area, Tsukuba, Kaneyasu et al. 2012, Adachi et al., 2013). Since the inertia of these smaller ^{137}Cs particles is likely to be smaller than that for volcanic ash, E_c could be smaller. The range of E_c was set as 0.02 – 0.06. The dry deposition velocity v_d was selected as 0.1 cm/s for ^{137}Cs in Furuno et al. (1999). The range for v_d was set as 0.05 to 0.15 in the present study.

The emission inventory of Katata et al. (2015) amounted to 14.1 PBq from March 12 to April 1, 2013. The simulated deposition over the model domain (138 – 143 °E, 34 – 39 °N) ranged from 3.4 – 4.7 PBq, which is approximately 24 – 33 %~~a quarter to one third~~ of the emission from the FDNPP. Sixty percent of the total deposition occurred over land, for a total of 2.0 – 2.8 PBq, which is close to the observed value of 2.68 PBq, and the observed value is within the range of the sensitivity runs. Statistical measures such as the fractional bias *FB*, the correlation coefficient *R* (linear vs. linear), and *FAx* (fraction of the simulated values within a factor of *x*) are listed in Table 2. To find better combinations of (or to optimize) the dry and wet deposition parameters, sensitivity runs were screened based on the criteria $FA10 > 0.9$, $FA5 > 0.7$, $R > 0.75$, and an absolute value of $FB < 10\%$. ~~After the screening, o~~Only one combination (E_c, v_d) = (0.04, 0.1 cm/s) ~~was left~~satisfied the criteria, and thus this is referred to as the "reference" parameters. To evaluate the sensitivity (or uncertainty) of the re-

suspension simulation for 2013 due to the deposition parameters, the range of the combination of parameters was set as (E_c, v_d) = (0.03 – 0.05, 0.05 – 0.1 cm/s) around the reference parameters (referred to as the "optimized range") by excluding the parameters with the worse performances. The ranges of the statistical measures of the optimized runs are listed in Table 2. *FB*, *R*, *FA2*, *FA5*, and *FA10* after the optimization had the ranges -0.18 – -0.036, 0.74 – 0.77, 0.26 – 0.30, 0.68 – 0.74, and 0.91 – 0.92, respectively. These statistical measures were comparable to those reported in previous multi-model comparison studies (*R*: 0.27 – 0.85, *FB*: -0.84 – 0.56, and *FA2*: 0.14 – 0.57, in SCJ, 2014 and Draxler et al., 2015). The current model is thus shown to be sufficiently credible for the budget analysis in this study, despite the simple parameterization and the low resolution in space (~11 km) and time (3 h).

~~Consistent~~ In agreement with many previous studies, the simulated contribution of wet deposition was larger than that of dry deposition: the ratio of the amount of dry to wet deposition ranged from 0.12 – 0.23 for the optimized parameter ranges, indicating that the results were less sensitive to the dry deposition parameter. Generally speaking, *R* became higher as E_c became lower, whereas *FAx* became higher as E_c became higher for the various ranges of the sensitivity tests. Therefore, lower E_c did not meet the criteria of *FAx* and higher E_c did not meet the criteria of *R*. Consequently, after the optimization, the maximum values of the statistical measures were lower but the minimum values became higher, indicating that the optimization was successful in excluding the parameters with the worse performances (rather than selecting the best parameters). It should be noted here that the optimized deposition parameters are not necessarily physically ~~true~~ valid but rather ~~are~~ give simulation results consistent with the available ~~evidence~~observations. The results presented in this section indicate that the current LM simulation with these optimized parameters has the potential to reproduce consistent features of the radiocesium budget over the Tohoku and Kanto regions of Japan.

Figure 6 shows the temporal variation of simulated (red) and observed (black) ^{137}Cs concentrations at Tsukuba in March 2011. The model ~~successfully~~ reproduced the three major plumes arriving at Tsukuba on March 15-16, 20-21, and 28-30; a plume on March 23 was not observed but only appeared in the simulation. The red shaded areas indicate the range of ^{137}Cs concentrations obtained when the simulations were run using the optimized parameter ranges E_c = 0.03 – 0.05 and v_d = 0.05 – 0.1 cm/s. Due to differences in the parameters used, the

surface air concentration could vary by approximately one order of magnitude in transported plumes that experienced wet scavenging along their pathway.

4.2 Re-suspension in 2013

Figure 7 shows the observed and daily mean simulated (with the reference parameters) surface air concentrations at Namie and Tsukuba for the year 2013. The red, green, and blue lines indicate re-suspension from soil, re-suspension from forest, and emission from the FDNPP reactor buildings, respectively. Scatterplots between the observed and the simulated total concentrations are also shown in the figure and the statistical measures between them are listed in Table 3. The correlation coefficients (linear vs. linear) are low at the both sites, but the simulated and the observed medians are close with each other and *FAx* show high values at the both sites. Consequently, the current simulation is enough credible for the quantitative assessment of the annual radiocesium budget.

As discussed in Sect. 2.2, ~~Note~~ note that the re-suspension flux due to the dust emission module (Ishizuka et al., 2016) is multiplied by five~~10~~ in this study ~~by adjusting~~in order to adjust to the observation level at Namie in the cold season (January to March, October to December). Also note that the re-suspension coefficient r_{const} in Eq. (4) was set as 10^{-7}/h in order to adjust~~by adjusting~~ to the observation level at Namie in the warm season (May to September). The emission flux reported by TEPCO varied from 10^5 to 10^7 Bq/h during the study period but we set it to 10^6 Bq/h for simplicity. We did not try to precisely adjust r_{const} to the observation by, for example, using inverse modeling, and instead we simply multiplied by power-of-ten values (except for dust emission, which is five), which are constant in time and space because (1) this ~~aided~~ facilitated straightforward interpretation of the simulation results by keeping the simulated variation ~~as it was~~solely originating from the variation of boundary conditions (i.e., emission and meteorology), and therefore (2) this provided simple but useful hints for understanding the re-suspension mechanisms, which remain unknown.

Using the dust emission module (which has a physical basis), ^{137}Cs flux re-suspended from soil could account for the level of the observed surface air concentration of ^{137}Cs at Namie in the cold season. Under the influence of the northwesterly winter monsoon, the surface wind speed is high over the contaminated area compared to ~~in~~ the summer (~~see~~ as shown later in Fig. 10). Note that the flux might be a maximum estimate since it does not consider land surface processes (such as soil moisture, snow cover, or migration of ^{137}Cs in the soil and

biota) and decontamination, which could reduce the ^{137}Cs re-suspension flux. In contrast, in the warm season, the estimated flux significantly underestimated the observation by one to two orders of magnitude due to the weak surface wind, indicating that the dust emission process may not be the sole process involved in sustaining the air concentration of ^{137}Cs during this period. Introducing the ^{137}Cs re-suspension component from forest with a resuspension coefficient of 10^{-7}/h and a monthly variation in the green area fraction (derived from NDVI) could quantitatively account for the observed air concentration together with its seasonal variation at Namie. Even though both the simulated re-suspension from soil and forests reproduced the quantity and seasonal variation of the background concentration (in other words, concentrations originating from ~~constantly~~ continuously presenting emissions, such as natural emissions, and not accidental ones) at Namie, sporadic peak events, such as the daily mean ^{137}Cs concentration exceeding 10 mBq/m^3 as observed in June and August at Namie, were not simulated. Some specific re-suspension events might occur from the highly contaminated areas such as ~~within~~ the premises of FDNPP (e.g. debris removal operations) or very close to FDNPP~~this area~~ on these days, as indicated later in Sect. 5.2. The simulated ^{137}Cs concentrations due to the monthly mean emission from the reactor buildings (=10^6 Bq/h) significantly underestimated the observed concentration by more than three orders of magnitude at Namie and by two orders of magnitude at Tsukuba. Even the maximum estimate of 10^7 Bq/h does not reach the observed level. The emission from FDNPP may not have been the sole process sustaining the air concentration of ^{137}Cs in 2013, supporting the discussion in Igarashi et al. (2015), which concluded that direct emission from the FDNPP played a minor role in the observed atmospheric radiocesium concentrations over Tsukuba during 2013-2014.

The observed air concentration of ^{137}Cs at Tsukuba was about one to two orders of magnitude lower than that at Namie. The simulated difference between the two sites inside and outside of the contaminated areas was consistent with the observed difference. This finding indicates that the current LM simulation provided consistent features of re-suspension, transport, and re-deposition in the Tohoku and Kanto regions of Japan of ^{137}Cs originating from Fukushima.

4.3 Budget analysis

Figure 8 illustrates the simulated (with the reference parameters) annual amounts of total re-suspension and re-deposition ~~amounts~~ of ^{137}Cs, together with their ratios to the observed deposition ~~amount~~ (Fig. 4a). The simulated areal total re-~~suspension~~ suspended amount was

1.~~01~~ 28 TBq, which was equivalent to 0.~~037~~048% of the total ~~deposition~~ deposited amount, 2.68 PBq. The areal total re-~~deposition~~ deposited amount (with the reference parameters) was 0.22 TBq (0.18 – 0.23 TBq for the optimized range of the deposition parameters), corresponding to approximately ~~21.7~~17.2 (~~17~~14.~~8~~ 1 – ~~22.8~~18.0) % of the re-suspended amount deposited mainly in the Tohoku region, with the remainder being transported out of the ~~region~~model domain. Therefore, the regional mean rate in the decrease of the land surface ^{137}Cs concentration due to re-suspension was estimated to be 0.~~029~~ 040 (0.~~029~~ 039 – 0.~~031~~041) %/y[2], equivalent to ~~7.9~~1.1 (~~7.9~~1.06 – ~~8.2~~1.12) × $10^{-~~7~~ 6}$ /d. The spatial distribution of the re-suspension and re-deposition ratio to the primary deposition ranged from 0.01 – 0.3% and 0.001 – 0.03%, respectively. The spatial distribution of the land surface ^{137}Cs deposition decay due to re-suspension ranged from 2.~~2~~7×10^{-7} – ~~6.6~~8.2×10^{-6} /d. Re-suspension therefore had a negligible effect on reducing land surface radioactive contamination.

5 Discussion

Seasonal variation of the surface activity concentration and its source contributions are extensively discussed in Sect. 5.1. A possible source of the observed sporadic peak events, which could not be reproduced by the simulation, is discussed in Sect. 5.2. The effects of other processes that were not considered in the model, such as land surface processes and decontamination, are discussed based on the dose rate measurements from the monitoring posts in Fukushima in Sect. 5.3. Future issues are summarized in Sect. 5.~~3~~4. ~~The effects of other processes that were not considered in the model, such as land surface processes and decontamination, are discussed based on the dose rate measurements from the monitoring posts in Fukushima in Appendix C.~~

5.1 Seasonal variation and source contribution

The discussion in this section expands on that in Sect. 4.2. Figure 9 shows the same temporal variation as Fig. 7 but for simulated (using the optimized ranges of parameters) results

[2] The amount re-suspended, excluding re-deposition (1.~~01~~ 28 TBq minus 0.22 (0.18 – 0.23) TBq) for the year 2013, divided by the total ~~deposition~~ deposited amount of 2.68 PBq.

for ^{137}Cs from dust and FDNPP in winter (January to March) and from forest and FDNPP in summer (June to August).

In the winter, the simulated trend for dust agreed well with the observed trend (Fig. 9a), and the surface air concentration during this period was positively correlated with the surface wind speed in both the simulation (R = 0.88 on hourly basis) and the observations (R = 0.32 on daily basis). There was a sporadic peak in the observational data of 6.7 mBq/m^3 from the March 17 at 13:00 local time (LT) to March 18 at 13:00 LT that could not be reproduced by the dust module, and this peak coincided with a plume arriving from FDNPP, as shown in Fig. 9c. ~~The discrepancy between the observed peak and the dust simulation is likely due to underestimation of the simulation because the simulated dust peak reached an intensity of 4 – 5 mBq/m^3 in the winter, which is of the same order of magnitude as that of the observed peak.~~ Therefore, ~~The~~ the observed peak could ~~also~~ be accounted for by specific re-suspension events on the order of 10^9 Bq/h (the left axis divided by the right axis multiplied by 10^6 Bq/h in Fig. 9c) if they occurred on the premises of FDNPP or close to the area. There are also two events exceeding 2 mBq/m^3, one in January and another in February. It is unlikely that the two peaks originated from the direct emission from FDNPP and likely that they originated from the dust emission because the observed peaks coincided with the simulated dust peaks (Fig. 9a) and not with the simulated peaks due to the FDNPP emission (Fig. 9c).

In the summer, the simulated quantity as well as the variation in the forest data agreed well with the observed data (Fig. 9b). Because there is only monthly variation in the simulated emission, the simulated daily trend solely originated from variations in the meteorological parameters (wind field, turbulent mixing, and wet scavenging). A significant peak of 60.4 mBq/m^3 is observed from August 14 at 13:00 LT to August 15 at 13:00 LT. This observed level was approximately two orders of magnitude larger than the simulated level and one to two orders of magnitude larger than the observed level for the other days in this period. Therefore, ~~constant~~ continuous emission such as re-suspension from forest is less likely to be the origin of the peak. Because the observed peak and the simulated peak of ^{137}Cs from FDNPP coincided (Fig. 9d), the observed level could be accounted for by specific re-suspension events on the order of 10^{10} Bq/h either on the premises of FDNPP or close to the area. There have been several arguments that the observed peaks in August 2013 were associated with debris removal operations at FDNPP and this is discussed separately in Sect. 5.2.

Figure 10 illustrates the seasonal mean surface wind vector and surface air ^{137}Cs concentration (simulated using the reference parameters) due to (a) dust re-suspension in the winter and (b) forest re-suspension in the summer. Due to the prevailing northwesterly winter monsoon, ^{137}Cs was carried southeastward in the winter. In the summer, under the influence of the Pacific high pressure system, ^{137}Cs was carried inland. The monthly mean wind speed is high in winter and low in summer. The upper panels of Fig. 11 illustrate the observed and simulated (using the optimized ranges of parameters) total (from soil, forest, and FDNPP) ^{137}Cs concentration at Namie and Tsukuba. The time resolutions of the simulation are daily for Namie and weekly for Tsukuba to be consistent with the sampling intervals of the two respective sites. The simulation successfully reproduced the quantity and variation in the observed background concentration at Namie and Tsukuba but could not reproduce the sporadic peak events observed at Namie, as discussed above. The simulation also significantly underestimated the observations at Tsukuba from January to March, 2013. Due to the northwesterly monsoon (Fig. 10a), there was less air mass transported from FDNPP to Tsukuba in the winter ~~(Fig. 12b)~~ and therefore this underestimation is probably due specifically to underestimation of the simulated re-suspension around Tsukuba. The lower panels of Fig. 11 show the relative contributions of ^{137}Cs from soil and forests at Namie and Tsukuba. The contribution from FDNPP was negligible throughout the year. At both sites, the contribution from dust was high (0.7~~6~~ – 0.8~~9~~) in the cold season and low (0.~~1~~ 2 – 0.4) in the warm season due to higher surface wind speed in the cold season.

Figure 12a shows the observed and simulated (with the reference parameters) monthly Namie to Tsukuba ^{137}Cs concentration ratios. The mean concentration ratio exceeded 100 in June and 200 in August due to the sporadic peak events. The monthly median would be relevant for comparing the background observation with the simulation results by considering only continuous ~~constant~~ emission sources~~s~~. The values of the simulated concentration ratio and its seasonal variation agreed fairly well with the observed monthly median ratio: the observed and simulated annual means were 38.9 and ~~30~~31.~~3~~1, respectively. Fig. 12b shows the monthly mean simulated re-suspension source area contributions to the ^{137}Cs air concentration at Namie and Tsukuba. The re-suspension source area is defined as the model grid where the observed deposition amount exceeded 300 kBq/m^2 (Fig. 4a) and includes the Namie grid (2,300 kBq/m^2). Eighty to 90% of the ^{137}Cs air concentration at Namie originated from the source region, and there was no clear seasonal variation in the value. In contrast, Tsukuba is characterized as a downwind region and there was clear seasonal variation in the source

contribution ratio: high in summer and low in winter, due to the summer and winter monsoons, as discussed above. Nonetheless, the highest value at Tsukuba was 0.4 in July, and so more than half of the ^{137}Cs concentration at Tsukuba originated locally or from areas other than the contaminated regions throughout the year. As shown in Figs. 11a and 11b, the variability in the simulated concentration at Tsukuba due to uncertainty in the deposition parameters was much larger than that at Namie. The differences in the variability indicated that the Namie and Tsukuba sites can be characterized as the source area and the downwind area, respectively: as the time required for the plume to move from the emission site to the observation site increases, the variability becomes larger due to the increased chance for the plume to experience dry and wet scavenging.

5.2 Possible source of sporadic peak events

There have been several scientific studies and governmental reports on the unexpected re-suspension from FDNPP in August 2013. The high dose rate alarm was activated on August 19 within the premises of FDNPP associated with the debris removal operation. Matsunami et al. (2016) related the radiocesium contamination of brown rice in Fukushima in 2013 to this operation, whereas MAFF (2015) denied any association. The NRA estimated the ^{137}Cs emission rate during the debris removal operation as 6.7×10^{10} Bq/h and the cumulative amount as 1.1×10^{11} Bq (NRA, 2014). TEPCO (2014c) estimated the emission rate during the operation as 5.8×10^{10} – 1.2×10^{11} Bq/h and the cumulative amount as 1.3 – 2.6×10^{11} Bq. Steinhauser et al. (2015) estimated the gross amount as 2.8×10^{11} Bq using measurements of weekly air filter sampling and monthly deposition, and a numerical simulation. Their estimates are of the similar order of magnitude as our estimate (10^{10} Bq/h, see Sect. 5.1) but the dates are different: our observed peak was earlier than the reported removal operation.

Our daily sampling showed a peak concentration (60.4 mBq/m^3) from August 14 at 13:00 LT to August 15 at 13:00 LT before the reported operation, but did not detect high concentrations in the August 19 (0.33 mBq/h for August 18 at 13:00 LT to August 19 at 13:00 LT and 1.2 mBq/h for August 19 at 13:00 to August 20 at 13:00 LT). Figure 13 shows the forward trajectories predicted by the LM (statistical locations of LPs) starting from FDNPP on August 14 (left) and August 19 (right). The sky-blue lines and red dashed circles indicate areas containing approximately two-thirds of the LPs within 1 km AGL: the extent of the area reflects horizontal and vertical atmospheric diffusion. The highest dose rate peaks were observed from 13:50 LT to 14:10 LT on August 19 at 2.8 – 8.3 km north and north-northwest

of FDNPP on the leeward side, as reported by Fukushima prefecture (https://www.pref.fukushima.lg.jp/download/1/20130827moni.pdf.pdf, last access: ~~March 11~~26 July~~,~~ 2016). The forward trajectories on August 19 indicated that plumes during the debris removal operation traveled north-northwest to north of FDNPP (Figs 13b and 13d), rather than towards the west-northwest where the Namie site is located. On August 14, on the other hand, plumes were transported towards the west (starting at 12 LT, Fig 13a), and then to the north (starting at 15 LT, Fig. 13c) due to fast changes in wind direction, resulting in the simulated peak concentration shown in Fig. 9d during this period. Our simulation and observations together indicated that the same order of magnitude of ^{137}Cs emission occurred on August 14 – 15 and on August 19. Alarm activation was not reported on August 14 – 15 but debris removal operation was also conducted on August 14 and 16 (MAFF, 2015).

5.3 Land surface processes, decontamination, and dust emission suppression due to snow cover

Figure 14 presents the time series of ambient gamma dose rates measured at the monitoring posts in Fukushima prefecture indicated in Fig. 1b. The data were obtained from the Nuclear Regulation Authority (NRA), Japan website (http://radioactivity.nsr.go.jp/map/ja/index.html, last access: 26 July 2016). A total of six, two of the monitoring posts from three geographical areas (Hamadori coastal area, Abukuma highland area, and Nakadori valley area), were selected. There are tens to hundreds of monitoring posts in each municipality (village, town, and city) in Fukushima prefecture. We selected a monitoring post in each municipality by applying the following conditions: a post showing the highest dose rate of all posts in the municipality at the time of downloading (around 11:00 a.m., December 28, 2015), data are available since April 1, 2012, and the instruments are situated 100 cm above the ground.

The government of Japan designated the evacuation-directed zones as a Difficult-to-Return zone (DRZ) (> 50 mSv/y; 9.55 μSv/h), a Habitation-Restricted Zone (HRZ) (20 – 50 mSv/y; 3.85 – 9.55 μSv/h), and a zone being prepared to have the evacuation directive lifted (< 20 mSv/y; 3.85 μSv/h), in April 2012 (METI, 2012). The two sites in the Hamadori area, Okuma and Omaru, have been designated DRZ (13.6 and 11.8 μSv/h on December 28, 2015). The dose rates at the two sites in the Abukuma area, Iitate and Kawamata, dropped below the HRZ level during the analysis period in this study (1.17 and 0.521 μSv/h on December 28, 2015). The dose rates in the Nakadori area are below 1 μSv/h (0.242 and 0.201 μSv/h on December 28, 2015).

The dose rate significantly dropped when the ground was covered with snow, in January and December 2013 in Hamadori and Nakadori, and from January to early March and December 2013 in Abukuma (the elevation of Abukuma is 500 – 1,000 m and higher than Hamadori and Nakadori). Snow cover suppresses re-suspension due to dust emission. Namie (Tsushima) is located in Abukuma and the ground was covered with snow until early March (Ishizuka et al., 2016). The observed air concentration of ^{137}Cs at Namie (Tsushima) was correlated with the wind speed in the winter from January to March, indicating that re-suspension during the period was mechanically induced. In the winter, dust re-suspension from outside Abukuma, such as from Hamadori and Nakadori, or from land surface where the snow cover was partly melted due to solar radiation, might be the dominant source contributing to the ^{137}Cs surface air concentration at Namie (Tsushima) when the ground was covered with snow.

The first order decreasing rates fitted by the least-square approximation for the period without snow cover, May to October 2012 and 2013, are presented in Fig. 14. The rates ranged from 5.2 – 12.1 $\times 10^{-4}$ /d. The monthly mean radioactive decay rates of total radiocesium (^{134}Cs + ^{137}Cs), determined by assuming that the activities of ^{134}Cs and ^{137}Cs were equivalent in March 2011, were 4.2×10^{-4} /d and 3.0×10^{-4} /d in April 2012 and March 2014, respectively, due to the difference in half-life of ^{134}Cs and ^{137}Cs (2.07 y and 30.1 y, respectively). By assuming that the gamma dose rate primarily originated to radiation from the land surface radiocesium, the radioactive decay accounted for 35 – 50% of the decreasing rate of total ground radioactivity; the exception was Shirakawa, where radioactive decay accounted for 55 – 80% of the decrease. In other words, 50 – 65% of the ground radioactivity decrease was likely due to land surface processes, decontamination, and re-suspension to air. As discussed in Sect. 4.3, the estimated decreasing rate due to re-suspension was 2.7×10^{-7} – 8.2×10^{-6} /d, which is two to three orders of magnitude smaller than the decreasing rates due to the other processes (10^{-4} – 10^{-3} /d).

It is difficult to distinguish the contributions of land surface processes and decontamination. By subtracting the radioactive decay rate (3.0 – 4.2×10^{-4} /d) and the decreasing rate due to re-suspension (2.7×10^{-7} – 8.2×10^{-6} /d) from the gross decreasing rate (5.2 – 12.1 $\times 10^{-4}$ /d), the estimated decreasing rates due to land surface processes and decontamination ranged from 1.0 – 7.9×10^{-4} /d. Matsuda et al. (2015) summarized the depth profiles of radiocesium in soil at more than 80 locations in Fukushima, including Hamadori, Abukuma, and Nakadori. They found that the radiocesium levels have been slowly migrating downward with rates ranging

from 1.7 – 9.6 kg/m^2/y (equivalent to 1.1 – 6.0 mm/y for a dust particle density of 2650 kg/m^3 and a porosity of 0.4 m^3/m^3, for example). Evrard et al. (2015) summarized that significant transfer of particulate-bound radiocesium occurs during major rainfall and runoff events (e.g. typhoons and spring snowmelt). Together with the relaxation depth – dose rate relationship provided by Saito and Petoussi-Hess (2014), the decreasing rate due to land surface processes such as downward migration, runoff, and erosion could be quantified and thus the decontamination effect could be separately extracted.

~~5.3~~5.4 Future issues

Issues that remain to be resolved in future research are summarized in the section. ~~as follows:~~First of all,

~~R~~re-suspension from ~~the~~ biota could be predominant in the warm season but the re-suspension sources as well as mechanisms remain essentially unknown. Further study is needed to understand the mechanism based on field experiments and numerical simulations.

The current estimation could account for the measured background concentration (0.1 – 1 mBq/m^3) but could not reproduce the observed sporadic peak concentration (1 – 10 mBq/m^3) at the Namie site. Further study is needed to identify the cause.

The dust flux module has been validated at a single location. The module ~~could~~ needs to be ~~extended~~ improved to be applicable to various land use and soil texture conditions. The decontamination may reduce resuspension afterward, whereas the resuspension may occur during decontamination-related work. This effect should be evaluated in the future.

The current estimation was based on a single model simulation. Variability in multi-model simulations is rather large (SCJ, 2014; Draxler et al., 2015) and therefore multi-model assessment will be indispensable for long-term re-suspension analysis.

~~1.~~

6 Conclusions

The long-term effect of ^{137}Cs re-suspension from contaminated soil and biota due to the Fukushima nuclear accident has been quantitatively assessed using a numerical simulation, a field experiment on dust emission in the contaminated area (Namie, Fukushima), and air

concentration measurements inside (Namie) and outside (Tsukuba, Ibaraki) of the area. The re-suspension mechanism remains unknown. We therefore utilized the observational data obtained both inside and outside the contaminated area, together with a transport model~~3D numerical simulation~~, to provide a robust budget analysis of the re-suspension, transport, and re-deposition of ^{137}Cs in the eastern part (the Tohoku and Kanto regions) of Japan. Our findings are summarized as follows:

1. Optimization of the deposition parameters of the LM for simulating the emergency situation of March 2011, using aircraft observation data (NRA, 2012) and the prescribed emission inventory (Katata et al., 2015), provided 0.1 (0.05 – 0.1) cm/s for a dry deposition velocity over land and 0.04 (0.03 – 0.05) for a hydrometeor collection efficiency for aerosols. The optimized (or validated) ranges of the deposition parameters were applied to long-term re-suspension assessment for the year 2013.

2. Using the dust emission module (Ishizuka et al., 2016), which was developed based on physical parameters, simulated ^{137}Cs re-suspension from soil multiplied by five~~10~~ accounted for the observed ^{137}Cs surface air concentration measured at Namie ~~in~~ during only the cold season; the module underestimated the ^{137}Cs concentration by one to two orders of magnitude in the warm season.

3. Introducing re-suspension from forest using a constant re-suspension coefficient of 10^{-7} /h and monthly green area fraction could quantitatively account for the observed concentration together with its seasonal variation.

~~2.~~4. The contribution from additional emission from the reactor buildings of FDNPP (10^6 Bq/h) was negligible throughout the year and underestimated the observed air concentration by two to three orders of magnitude at both observation sites.

~~3.~~5. At Namie and Tsukuba, the simulated contribution of re-suspension from soil was high (0.7~~6~~ – 0.8~~9~~) in the cold season and low (0.~~1~~ 2 – 0.4) in the warm season; the remaining contribution was from forest and was low in winter and high in summer. The contribution of the re-suspension from the source area (where the aircraft-observed deposition exceeded 300 kBq/m^2) to the air concentration at Namie was 0.8 – 0.9 throughout the year, while that at Tsukuba varied from 0.1 to 0.4, and was high in the summer and low in the winter.

6. The simulated total re-suspended amount for the whole region was 1.~~01~~ 28 TBq, equivalent to 0.~~037~~048% of the aircraft-observed total ~~deposition~~ deposited amount of 2.68 PBq. The total re-deposition was 0.18 – 0.23 TBq, equivalent to ~~17.8~~14.1 – ~~22.8~~18.0% of the total re-suspended amount: the rest of the ^{137}Cs was transported out of the model domain. The spatial distribution of the decreasing rate of land surface ^{137}Cs due to re-suspension ranged from 2.~~2~~7×10^{-7} – ~~6.6~~8.2×10^{-6} /d.

~~4.~~7. The first order decrease rate of the ambient gamma dose rate in Fukushima prefecture ranged from 5.2 – 12.1×10^{-4} /d. By subtracting the radioactive decay rate of 3.0 – 4.2 ×10^{-4} /d, the ground radioactivity decay due to land surface processes, decontamination, and re-suspension was found to range from 1.0 – 7.9×10^{-4} /d. The estimated re-suspension rate was two to three orders of magnitude lower than the decrease in rate due to the other processes, showing that re-suspension contributed negligibly towards reducing ground radioactivity.

Appendix A: Model description

The current study employs a Lagrangian type model for the simulation of emission (either point sources or areal sources), horizontal and vertical diffusion and advection, gravitational settling, dry and wet depositions, and radioactive decay in the air. As described in Sect. 2.1, the current Lagrangian Model (LM) uses simple parameterizations for dry and wet deposition schemes for computational efficiency, so long-term assessment and parameter sweep experiments are easily feasible. The source code for the model is open with the BSD 3-Clause License and is available on the web (https://ebcrpa.jamstec.go.jp/isetr_a01-1/~~http://157.82.240.167/~dl3/~~, in Japanese, last access: 26 July~~12 February~~ 2016).

The coordinate system of the model is horizontal for longitude and latitude and vertical for pressure level, consistent with meteorological analysis data commonly used. The model can be driven only by fundamental meteorological parameters such as temperature, humidity, 3D wind field, geopotential height, and surface precipitation provided by meteorological analysis data such as GPV-MSM. The model does not need to drive meteorological models to predict detailed meteorological variables such as cloud microphysics, turbulence quantities, and surface variables. Since the temporal and spatial resolution of the meteorological analysis is not very high (e.g., 3 h and ~11 km for GPV-MSM, respectively), linear interpolation is conducted in time and space. Alternatively, higher temporal and spatial resolution can be

achieved by using a meteorological model. Furthermore, although currently not implemented, detailed variables predicted by a meteorological model can be used for more accurate predictions of turbulent diffusion, surface flux, and dry and wet deposition.

In the LM model, LPs are released constantly in time but the initial activity of LPs (Bq/LP) differs accordingly to the emission flux (Bq/h). The initial positions of LPs were randomly distributed within a fixed volume (or line) of plume centered at a point emission source such as FDNPP for the primary emission case simulation, or randomly distributed within a horizontal model grid for the areal emission cases (such as re-suspension from soil and forest). LPs do not disappear unless transported across lateral and upper boundaries or if they reach the surface layer due to gravitational settling (technically, gravitational settling velocity in the surface layer is included in the dry deposition velocity). The other processes, such as dry deposition, wet deposition and radioactive decay, do not decrease the number of LPs but do decrease the radioactivity carried by LPs because LPs represent an air mass rather than an actual particle, except in the case of gravitational settling. The lowest level permitted for the position of LPs is set as 2 m AGL and LPs going down across the level due to vertical turbulent motion will rebound at the level and go up. An LP whose radioactivity is smaller than a preset value, i.e., 10^{-10} Bq, due to deposition or radioactive decay will disappear from the computation to maintain computational efficiency, since the cost of the computation is proportional to the number of LPs in the model domain. To output the model results, the LP fields are converted to Eulerian concentration (Bq/m^3) and deposition (Bq/m^2) fields on a prescribed coordinate system of grids. In Lagrangian type models, the spatial resolution of tracer emission, concentration, and deposition fields can be set independent of each other and with the spatial resolution of meteorological fields. In the current implementation of the LM, the coordinate system of meteorological fields and radioactivity fields is horizontal for common (longitude and latitude) but vertical for different (pressure level and meters AGL, respectively).

The horizontal and vertical diffusion calculation followed JMA (2008), using the horizontal diffusion scheme of Uliasz (1990) with a constant horizontal diffusivity of 5.864×10^4 m^2/s and using a vertical diffusivity calculated based on Louis et al. (1982) (see Eqs. 8.1.8 through 8.1.15 of JMA (2008) for details). The incremental change in location of an LP δx (y, z) after a time step δt was defined as

$$\delta x = \frac{\mathrm{d}x}{\mathrm{d}t} G \delta t \,, \tag{A1}$$

where G is the normalized Gaussian random number (average = 0, standard deviation = 1). δt is set large enough for computational efficiency but without violating the Courant-Friedrichs-Lewy (CFL) condition of $\delta t < 0.5\, U/\Delta x$, where U (or $\mathrm{d}x/\mathrm{d}t$) is the typical wind speed and Δx is the grid size in the direction of U. However, the selection of δt is not critical because every time step prior to applying Eq. (A1) time splitting is made so that the split step always satisfies the CFL condition.

The wet scavenging rate Λ_{wet} (/s) is simply parameterized as a function of the surface precipitation rate P (mm/s) as

$$\Lambda_{wet} = \frac{3}{4} \frac{E_c(a_m, r_m)}{a_m} P \,, \tag{A2}$$

where E_{c} is the collection efficiency of aerosols by the hydrometeor, and a_{m} and r_{m} are the mean radii of the hydrometeor and aerosols, respectively (JMA, 2008). Empirically, a_{m} is characterized by P as

$$a_m = 0.35 P^{0.25} \,. \tag{A3}$$

JMA (2008) uses 0.05 for E_{c}. In the current study, instead of explicitly predicting E_{c}, its range was set for the sensitivity tests as listed in Table 2.

Conceptually, Eq. (A2) is the formulation for the washout process, i.e., the collection of aerosols by the settling hydrometeor particles such as rain and snow. a_{m} and E_{c} should differ for rain and snow, but common parameters are used in the current simulation. Also, Eq. (A2) is not applicable for the rainout process, since this process—cloud condensation nuclei or ice nuclei activation and deposition via subsequent cloud microphysical processes— is totally different from the washout process. Because meteorological models were not utilized in this study and thus only relative humidity and surface precipitation rate are available and no cloud microphysical information (such as hydrometeors mixing ratio in each model grid) is available, Eq. (A2) is applied for all the LPs located above the grid with P. In order to partly account for the rainout process, Eq. (A2) is not applied to LPs in a grid, where the relative humidity is lower than the minimum value, set as 95% in the simulation.

The dry deposition velocity v_{d} (m/s) of aerosols (or gases) is conventionally formulated, using an electrical analogy, as an inverse of the summation of resistances (s/m) representing

turbulent diffusion in the surface layer, Brownian diffusion (or molecular diffusion for gases), interaction with the land surface (soil, water and vegetation), and gravitational settling for aerosols (e.g., Wesely and Hicks, 2000). Therefore, v_d is a function of height as well as of turbulent flux and surface conditions. Nevertheless, v_d is set as constant in the simulation, but the height dependency of v_d is considered in the dry scavenging rate Λ_{dry} (/s), following Furuno et al. (1999) as

$$\Lambda_{\mathrm{dry}} = \frac{2}{z_{srf}}\left(1-\frac{z}{z_{srf}}\right)v_d\,, \tag{A4}$$

where z is the height of the LP (m AGL) and z_{srf} is the surface layer height set as 100 m AGL in the study. Instead of explicitly predicting v_d, its range was set at around 0.1 cm/s, a typical speed for a range of aerosols around 1 μm in diameter, for the sensitivity tests, as listed in Table 2. The value of v_d is applied over land, whereas v_d over the ocean is multiplied by 0.1, because v_d over a flat surface is approximately one and two orders of magnitude smaller than v_d over short vegetation such as grass and tall vegetation such as forest, respectively (e.g., Petroff and Zhang, 2010).

Appendix B: Statistical accuracy of the current simulation setting

Because the statistical error of Lagrangian simulation is inversely proportional to the square of the number of LPs, the statistical accuracy of the current simulation setting was evaluated using the following measures (relative errors of quantities of the sensitivity runs to those of the reference run):

$$E_{con}\,(x,y) = \frac{\left|C_{sens}(x,y) - C_{ref}(x,y)\right|}{C_{ref}(x,y)}, \tag{B1}$$

$$E_{dep}\,(x,y) = \frac{\left|D_{sens}(x,y) - D_{ref}(x,y)\right|}{D_{ref}(x,y)}, \tag{B2}$$

where x and y indicate grid points on the longitudinal and latitudinal axes, respectively. C_{sens} and D_{sens} indicate temporal mean surface concentrations (Bq/m^3) and temporal cumulative depositions (Bq/m^2) of the sensitivity runs, respectively. C_{ref} and D_{ref} are the same as C_{sens}

and D_{sens} but for the reference run. E_{con} and E_{dep} were sampled only at grids where $C_{ref}(x,y)$ and $D_{ref}(x,y)$ are greater than their areal mean values, respectively.

B.1 Point source case

The number emission rate of LPs, N_{LP}, was set as 32,000 /h (=$N_{\mathrm{LP\_ref}}$) for a point source emission case such as the primary emission in March 2011 and additional emission from the reactor buildings in 2013. The median values together with the 25th and 75th percentile values of E_{con} and E_{dep} of the sensitivity runs (sensitivity to deposition parameters and sensitivity to N_{LP}) against the reference run are listed on the top half rows of Table B1. Both E_{con} and E_{dep} of $N_{\mathrm{LP\_ref}}$ ×4 were significantly lower than those for the deposition parameters sensitivity run. This result indicates that 32,000 /h for N_{LP} was sufficient to allow a statistically significant simulation for the purpose of this study, as shown in Figs. 4 and 5: the difference in concentration and deposition due to the deposition parameters was much larger than the difference due to model uncertainty in N_{LP}. E_{con} and E_{dep} of $N_{\mathrm{LP\_ref}} \times 0.25$ (which are also smaller than those of the deposition parameters sensitivity run) are larger than those of $N_{\mathrm{LP\_ref}} \times 4$, indicating fairly well accuracy convergence of the LM model.

B.2 Areal emission case

$N_{\mathrm{LP\_ref}}$ was 16 /h/grid for the areal emission case simulating re-suspension from soil and forests in 2013. The lower half of Table B1 is the same as the upper half except for the areal emission case (re-suspension from forest). Both E_{con} and E_{dep} of $N_{\mathrm{LP\_ref}} \times 4$ were much lower than those for the deposition parameters sensitivity run, indicating that 16 /h/grid supports a statistically significant simulation for the purpose of this study, as shown in Figs. 7 - 12. Usually, Eulerian-type models are appropriate for solving areal emission problems—Lagrangian-type models require many more LPs for areal emission cases compared to point source cases and thus become computationally too expensive. In the case of this simulation, especially for the concentration, sensitivity to deposition parameters was much more significant than sensitivity to model uncertainty in N_{LP} using the sufficiently small number of $N_{\mathrm{LP\_ref}} = 16$ /h/grid. E_{con} and E_{dep} of $N_{\mathrm{LP\_ref}} \times 4$ are smaller than those of $N_{\mathrm{LP\_ref}} \times 0.25$, indicating fairly accuracy convergence of the LM model.

Appendix C: Land surface processes, decontamination, and dust emission suppression due to snow cover

Figure C1 presents the time series of ambient gamma dose rates measured at the monitoring posts in Fukushima prefecture indicated in Fig. 1b. The data were obtained from the Nuclear Regulation Authority (NRA), Japan website (http://radioactivity.nsr.go.jp/map/ja/index.html, last access: Feb 16, 2016). A total of six, two of the monitoring posts from three geographical areas (Hamadori coastal area, Abukuma highland area, and Nakadori valley area), were selected. There are tens to hundreds of monitoring posts in each municipality (village, town, and city) in Fukushima prefecture. We selected a monitoring post in each municipality by applying the following conditions: a post showing the highest dose rate of all posts in the municipality at the time of downloading (around 11:00 a.m., December 28, 2015), data are available since April 1, 2012, and the instruments are situated 100 cm above the ground.

The government of Japan designated the evacuation-directed zones as a Difficult-to-Return zone (DRZ) (> 50 mSv/y; 9.55 μSv/h), a Habitation Restricted Zone (HRZ) (20 – 50 mSv/y; 3.85 – 9.55 μSv/h), and a zone being prepared to have the evacuation directive lifted (< 20 mSv/y; 3.85 μSv/h), in April 2012 (METI, 2012). The two sites in the Hamadori area, Okuma and Namie (Omaru), have been designated DRZ (13.6 and 11.8 μSv/h on December 28, 2015). The dose rates at the two sites in the Abukuma area, Iitate and Kawamata, dropped below the HRZ level during the analysis period in this study (1.17 and 0.521 μSv/h on December 28, 2015). The dose rates in the Nakadori area are below 1 μSv/h (0.242 and 0.201 μSv/h on December 28, 2015).

The dose rate significantly dropped when the ground was covered with snow, in January and December 2013 in Hamadori and Nakadori, and from January to early March and December 2013 in Abukuma (the elevation of Abukuma is 500 – 1,000 m and higher than Hamadori and Nakadori). Snow cover suppresses re-suspension due to dust emission. Namie (Tsushima) is located in Abukuma and the ground was covered with snow until early March (Ishizuka et al., 2016). The observed air concentration of ^{137}Cs at Namie (Tsushima) was correlated with the wind speed in the winter from January to March, indicating that re-suspension during the period was mechanically induced. In the winter, dust re-suspension from outside Abukuma, such as from Hamadori and Nakadori, or from land surface where the snow cover was partly melted due to solar radiation, might be the dominant source contributing to the ^{137}Cs surface air concentration at Namie (Tsushima) when the ground was covered with snow.

~~The first order decreasing rates fitted by the least-square approximation for the period without snow cover, May to October 2012 and 2013, are presented in Fig. C1. The rates ranged from 5.2 – 12.1 $\times 10^{-4}$ /d. The monthly mean radioactive decay rates of total radiocesium (^{134}Cs + ^{137}Cs), determined by assuming that the activities of ^{134}Cs and ^{137}Cs were equivalent in March 2011, were 4.2×10^{-4} /d and 3.0×10^{-4} /d in April 2012 and March 2014, respectively, due to the difference in half-life of ^{134}Cs and ^{137}Cs (2.07 y and 30.1 y, respectively). By assuming that the gamma dose rate primarily originated to radiation from the land surface radiocesium, the radioactive decay accounted for 35 – 50% of the decreasing rate of total ground radioactivity; the exception was Shirakawa, where radioactive decay accounted for 55 – 80% of the decrease. In other words, 50 – 65% of the ground radioactivity decrease was likely due to land surface processes, decontamination, and re-suspension to air. As discussed in Sect. 4.3, the estimated decreasing rate due to re-suspension was 2.2×10^{-7} – 6.6×10^{-6} /d, which is two to three orders of magnitude smaller than the decreasing rates due to the other processes (10^{-4} – 10^{-3} /d).~~

~~It is difficult to distinguish the contributions of land surface processes and decontamination. By subtracting the radioactive decay rate (3.0 – 4.2×10^{-4} /d) and the decreasing rate due to re-suspension (2.2×10^{-7} – 6.6×10^{-6} /d) from the gross decreasing rate (5.2 – 12.1 $\times 10^{-4}$ /d), the estimated decreasing rates due to land surface processes and decontamination ranged from 1.0 – 7.9×10^{-4} /d. Matsuda et al. (2015) summarized the depth profiles of radiocesium in soil at more than 80 locations in Fukushima, including Hamadori, Abukuma, and Nakadori. They found that the radiocesium levels have been slowly migrating downward with rates ranging from 1.7 – 9.6 kg/m^2/y (equivalent to 1.1 – 6.0 mm/y for a dust particle density of 2650 kg/m^3 and a porosity of 0.4 m^3/m^3, for example). The downward migration resulted in decreasing the air dose rate due to incremental soil layers blocking radiation, but the downward migration rate has not been quantitatively related to the decrease in the air dose rate. This quantitative relationship needs to be assessed for the quantitative and individual assessment of land surface processes and decontamination effects.~~

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Table 1. The observation sites and monitoring posts used to provide data for this study.

Name	Location	Description
Observation sites		
Namie (Tsushima)	140.7683 °E, 37.5621 °N	Namie High School, Tsushima Campus[1]
Tsukuba	140.1254 °E, 36.0551 °N	Meteorological Research Institute
Monitoring posts		
Okuma	140.9969 °E, 37.4163 °N	Ottozawa 3 Community Center[2]
Namie (Omaru)	140.9296 °E, 37.4665 °N	Omaru Multipurpose Community Center[3]
Iitate	140.7385 °E, 37.6772 °N	Iitate Junior High School[4]
Kawamata	140.6979 °E, 37.5836 °N	Yamakiya Otsu 8 Community Firehouse[5]
Fukushima	140.4765 °E, 37.6870 °N	Fukushima-Minami Fire Department
Shirakawa	140.1904 °E, 37.1241 °N	Takayama-Kita Park

1. Original location (now moved to Nihonmatsu city).
2. Ottozawa San-ku Chiku Shukaijo (in Japanese)
3. Omaru Tamokuteki Shukaijo (in Japanese)
4. Original location (now moved to Fukushima city).
5. Yamakiya Otsu Hachi-ku Community Shoubou Center (in Japanese)

Table 2. ^{137}Cs budget and statistical analysis for the comparison of observed and simulated deposition data for March 2011.

	E_c[a]	v_d[b]	D_{all}[c]	D_{land}[d]	FB[e]	R[f]	$FA2$[g]	$FA5$[h]
	(-)	(cm/s)	(PBq)	(PBq)	(-)	(-)	(-)	(-)
Sensitivity test								
Range	0.02 – 0.06	0.05 – 0.15	3.4 – 4.7	2.0 – 2.8	-0.25 – 0.00050	0.73 – 0.78	0.25 – 0.30	0.63 – 0.77
Optimization used for the re-suspension analysis for 2013								
Reference	0.04	0.10	4.2	2.5	-0.06.2	0.75	0.28	0.74
Optimized range	0.03 – 0.05	0.05 – 0.10	3.7 – 4.3	2.2 – 2.6	-0.18 – -0.036	0.74 – 0.77	0.26 – 0.30	0.68 – 0.74
Reference values								
Observed deposition over land D_{obs} (NRA, 2012)	2.68 PBq							
Emission amount (Katata et al., 2015)	14.1 PBq							

[a] Collection efficiency, see Eq. (A2) [b] Dry deposition velocity over land, see Eq. (A4). [c] Simulated ~~deposition~~ deposited amount over the whole model domain. [d] Simulated ~~deposition~~ deposited amount only over land. [e] Fractional bias between D_{land} and $D_{obs.}$ [f] Correlation coefficient between each grid cell of the observed and simulated deposition (linear vs. linear). [g] Fraction of simulated values within a factor of 2 of the observed values. [h] Fraction of simulated values within a factor of 5 of the observed values. [f, g, h] Compared only at grids where the observed values are greater than 10 kBq/m^2.

Table 3. Statistical analysis for the comparison of observed and simulated ^{137}Cs air concentration at Namie and Tsukuba for 2013.

Site	*N* (-)	*Median, Obs.* (mBq/m^3)	*Median, Sim.* (mBq/m^3)	*R* (-)	*FA2* (-)	*FA5* (-)	*FA10* (-)
Namie	311	0.71	0.69	0.12	0.67	0.94	0.99
Tsukuba	74	0.022	0.024	0.19	0.70	0.95	1.00

Table B1. Statistical measures of temporal mean ^{137}Cs surface concentration (E_{con}) and cumulative deposition (E_{dep}) of the sensitivity runs against the reference run for (top) the point source case and (bottom) the areal emission case.

	Number emission rate of LP, N_{LP} (/h/grid)	E_c [a] (-)	v_d [b] (cm/s)	Median (25th – 75th percentile) of E_{con} [c] (%)	Median (25th – 75th percentile) of E_{dep} [d] (%)
Point source case, March 2011					
Reference run	32,000	0.04	0.01	-	-
Sensitivity runs					
Deposition parameters	32,000	0.03 – 0.05	0.05 – 0.01	2.7 (0.37 – 5.0)	7.5 (3.6 – 13)
$N_{LP\_ref} \times 4$	128,000	0.04	0.01	0.51 (0.21 – 0.96)	0.72 (0.32 – 1.5)
$N_{LP\_ref} \times 0.25$	8,000	0.04	0.01	0.95 (0.39-1.8)	1.6 (0.73 – 2.9)
Areal emission case, 2013 (re-suspension from forest)					
Reference run	16	0.04	0.10	-	-
Sensitivity runs					
Deposition parameters	16	0.03 – 0.05	0.05 – 0.01	7.3 (2.6 – 13)	7.7 (3.6 – 17)
$N_{LP\_ref} \times 4$	64	0.04	0.10	0.39 (0.17 – 0.78)	2.0 (1.3 – 2.6)
$N_{LP\_ref} \times 0.25$	4	0.04	0.10	0.78 (0.34-1.5)	2.3 (1.5 – 3.1)

[a] Collection efficiency, see Eq. (A2). [b] Dry deposition velocity over land, see Eq. (A4). [c] Relative errors of temporal mean surface concentration at each grid cell of the sensitivity run to that of the reference run, see Eq. (B1). [d] same as E_{con} but for cumulative deposition, see Eq. (B2).

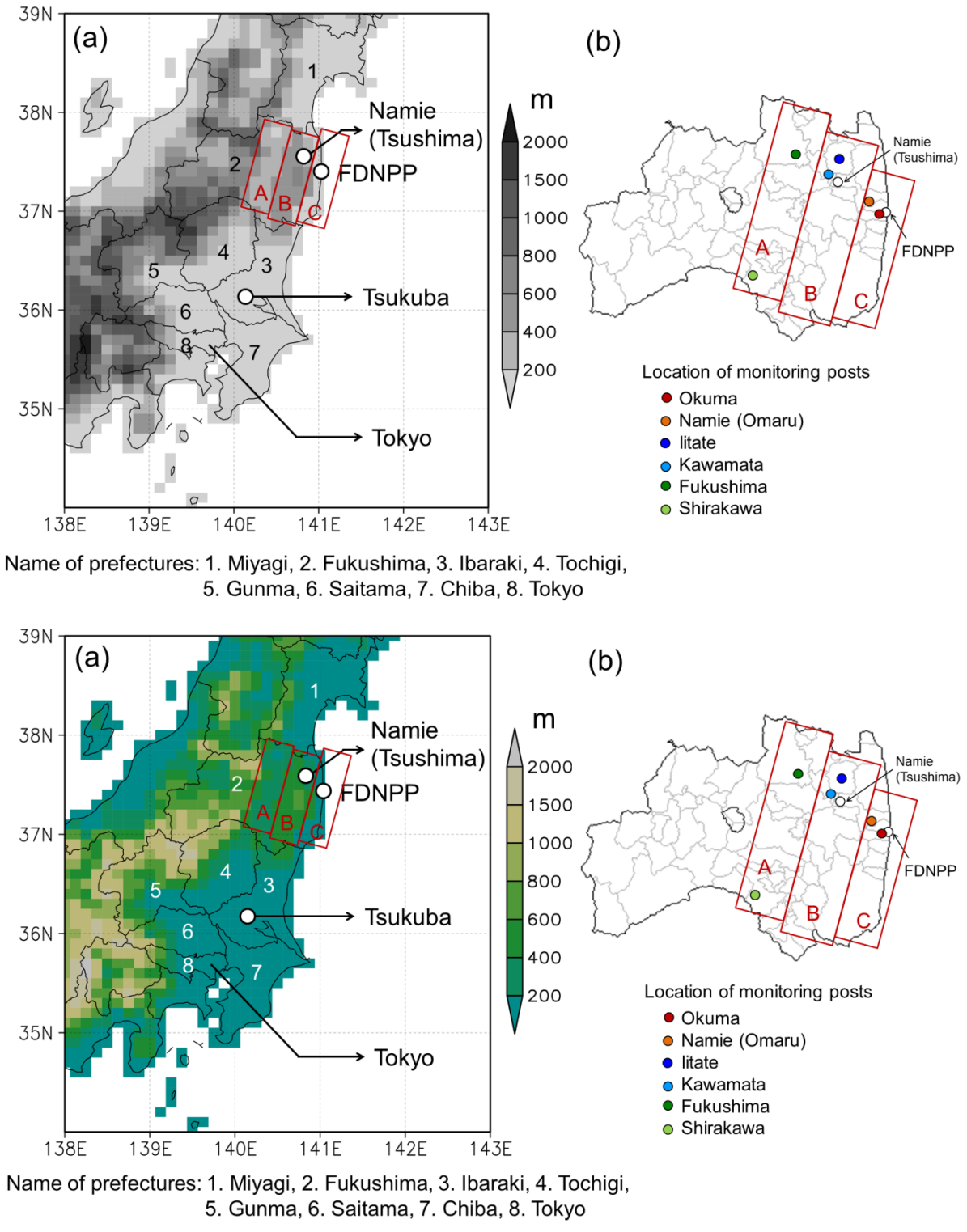
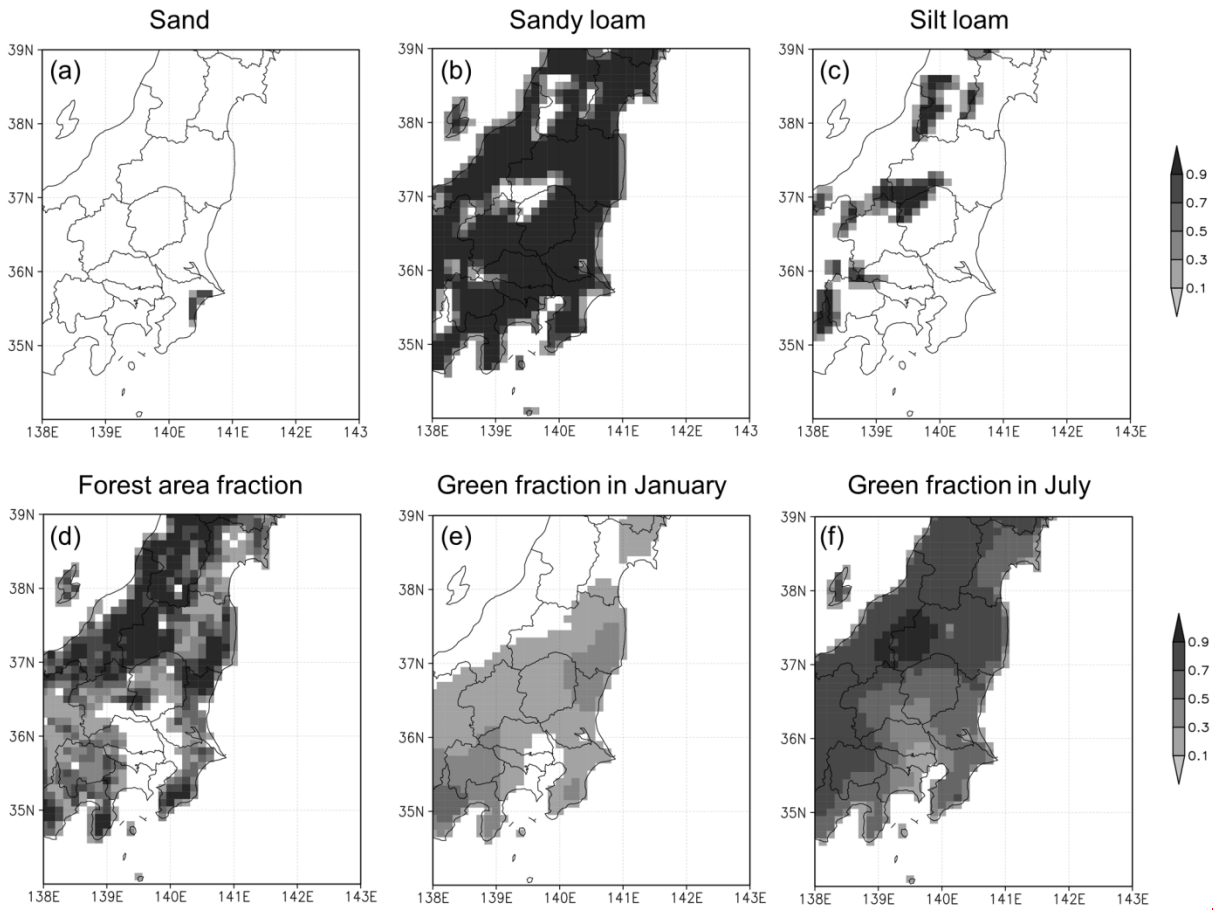


Figure 1. (a) The model domain showing the model terrestrial elevation, observation sites, and other locations described in the study. The linear distances from FDNPP to Namie (Tsushima district, Namie town), Tsukuba and Tokyo are approximately 30 km, 170 km, and 220 km, respectively. The numbers denote prefectures: 1. Miyagi, 2. Fukushima, 3. Ibaraki, 4. Tochigi,

5. Gunma, 6. Saitama, 7. Chiba, and 8. Tokyo. (b) Fukushima prefecture and (colored circles) the locations (village, town, or city name) of monitoring posts used in this study (see Fig. C1). The letters in both (a) and (b) denote the name of the area based on geographical features: A. Nakadori valley, B. Abukuma highland, and C. Hamadori coastal area.


Sand
Sandy loam
Silt loam
(a)
(b)
(c)
Forest area fraction
Green fraction in January
Green fraction in July
(d)
(e)
(f)
39N
38N
37N
36N
35N
138E
139E
140E
141E
142E
143E
0.9
0.7
0.5
0.3
0.1

1

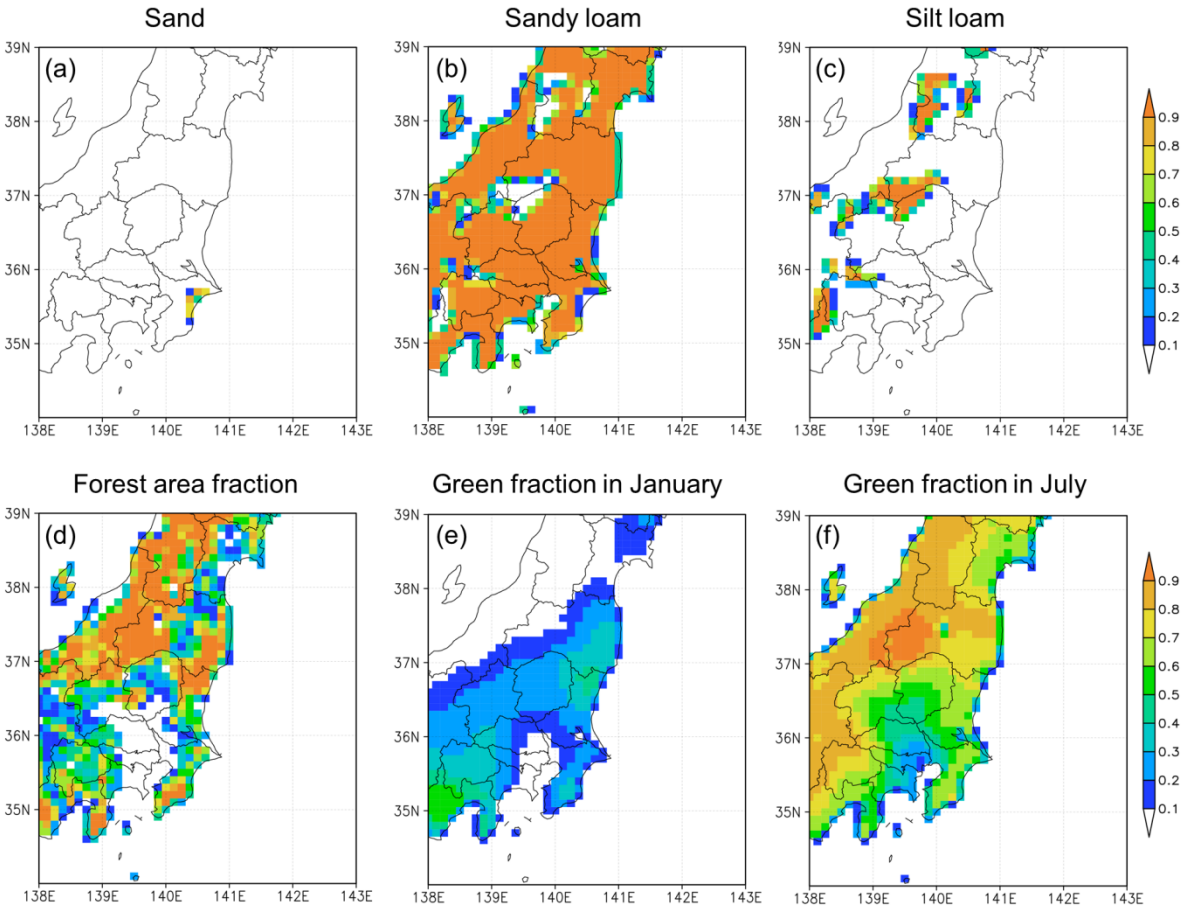


Figure 2. The areal fractions of (a)-(c) soil texture and (d)-(f) land use category used for the boundary conditions of the simulation.

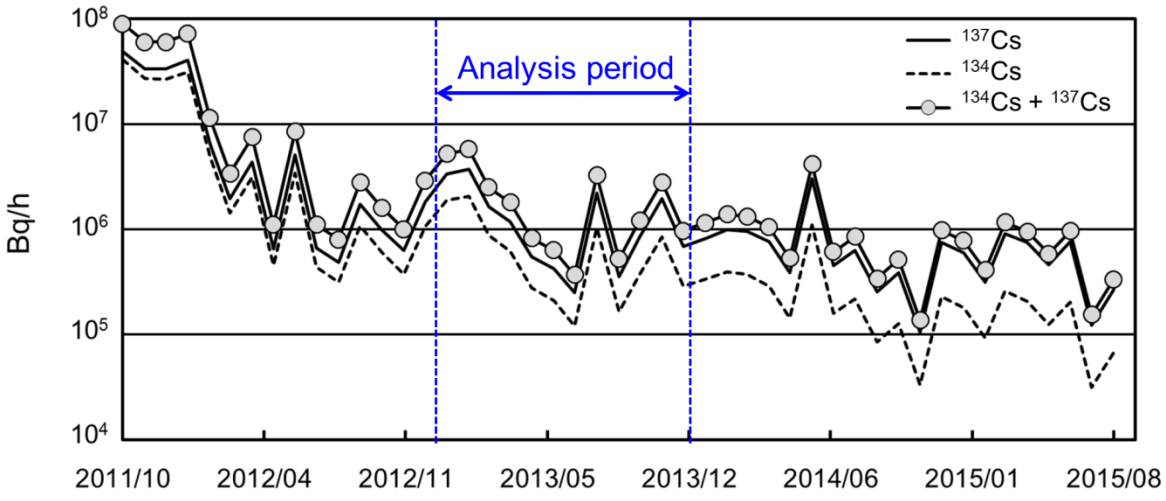


Figure 3. Monthly mean emission flux of radiocesium released from the reactor buildings of FDNPP from October 2011 to August 2015 as estimated by TEPCO (TEPCO, 2012-2015).

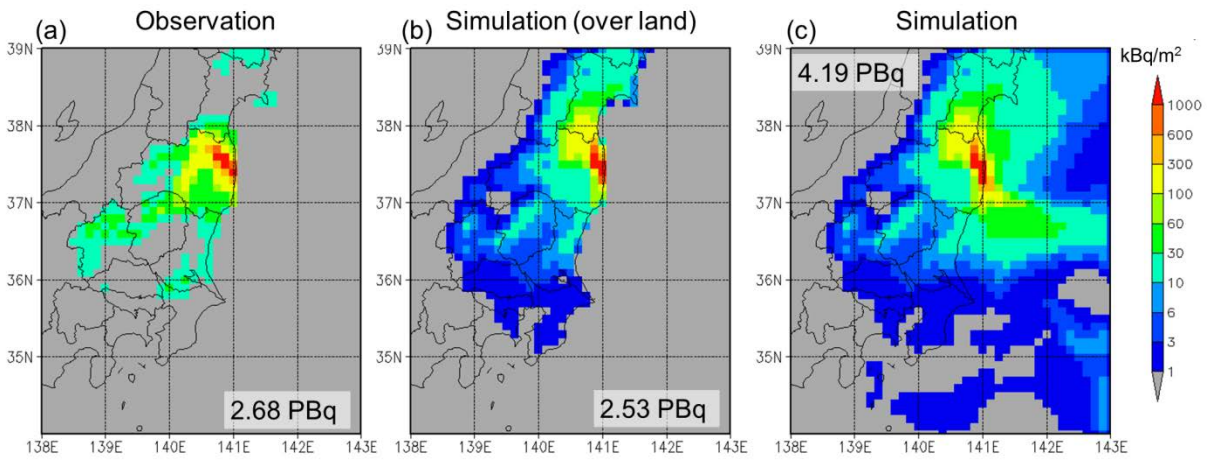


Figure 4. (a) Aircraft observation and (b), (c) simulation of ^{137}Cs deposition depicted (b) only over land and (c) for the whole domain (kBq/m^2). The observation was interpolated to the model grid (Δlongitude = 0.125° and Δlatitude = 0.1°). A decay correction for the observation was made for March - May 2012, varied depending on the region. The simulation provided a cumulative value from March 12 to April 1, 2011. The total activities are presented as numbers. The color scales are the same for (a) – (c) but observed values are not depicted below the detection limit, 10 kBq/m^2. The deposition parameters of the simulation in the figure are E_c = 0.04 and v_d = 0.1 cm/s.

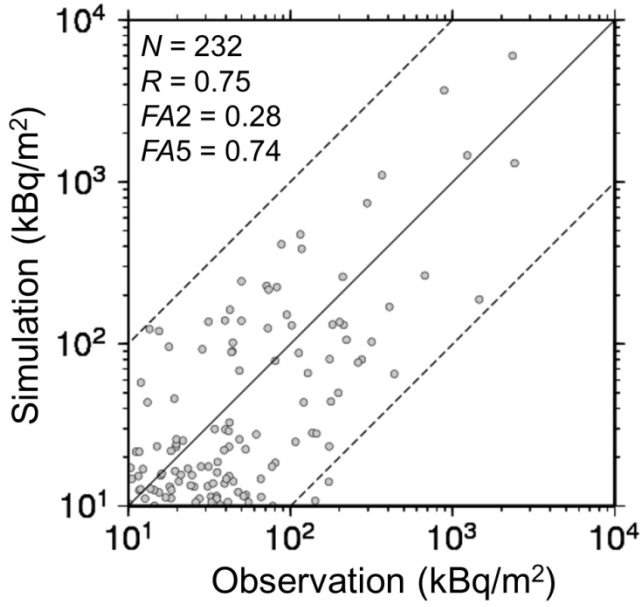


Figure 5. Scatterplotgram between the observational data and the simulation results for ^{137}Cs deposition (kBq/m^2). The deposition parameters of the simulation in the figure are E_c = 0.04 and v_d = 0.1 cm/s. *N* indicates the number of samples, and the statistical measures *R*, *FA2*, and *FA5* are described in Table 2.

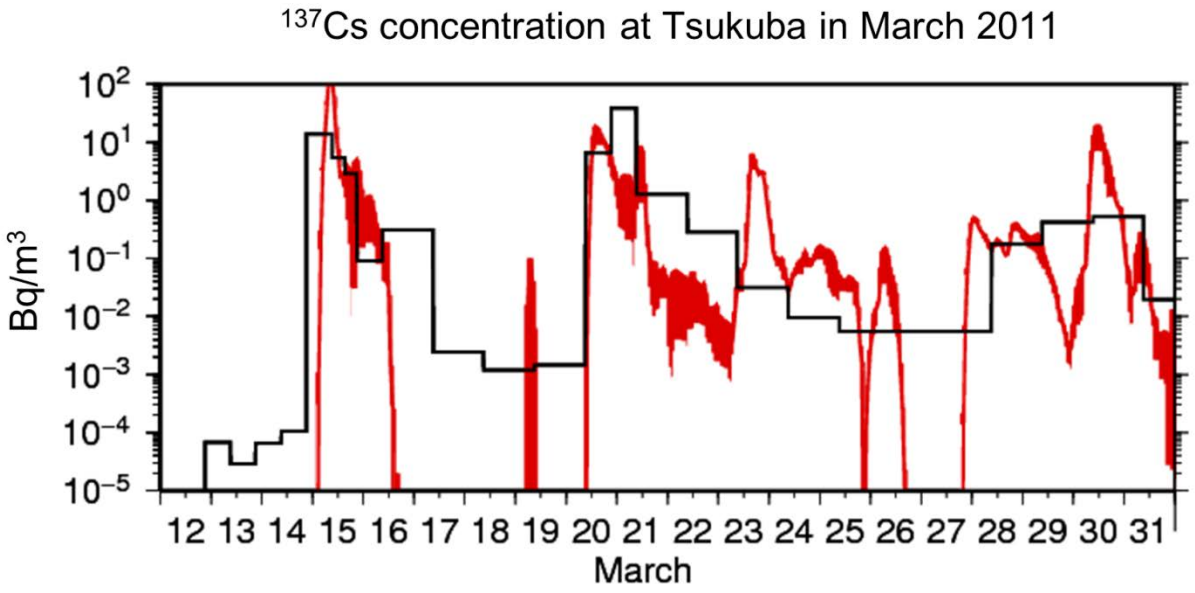
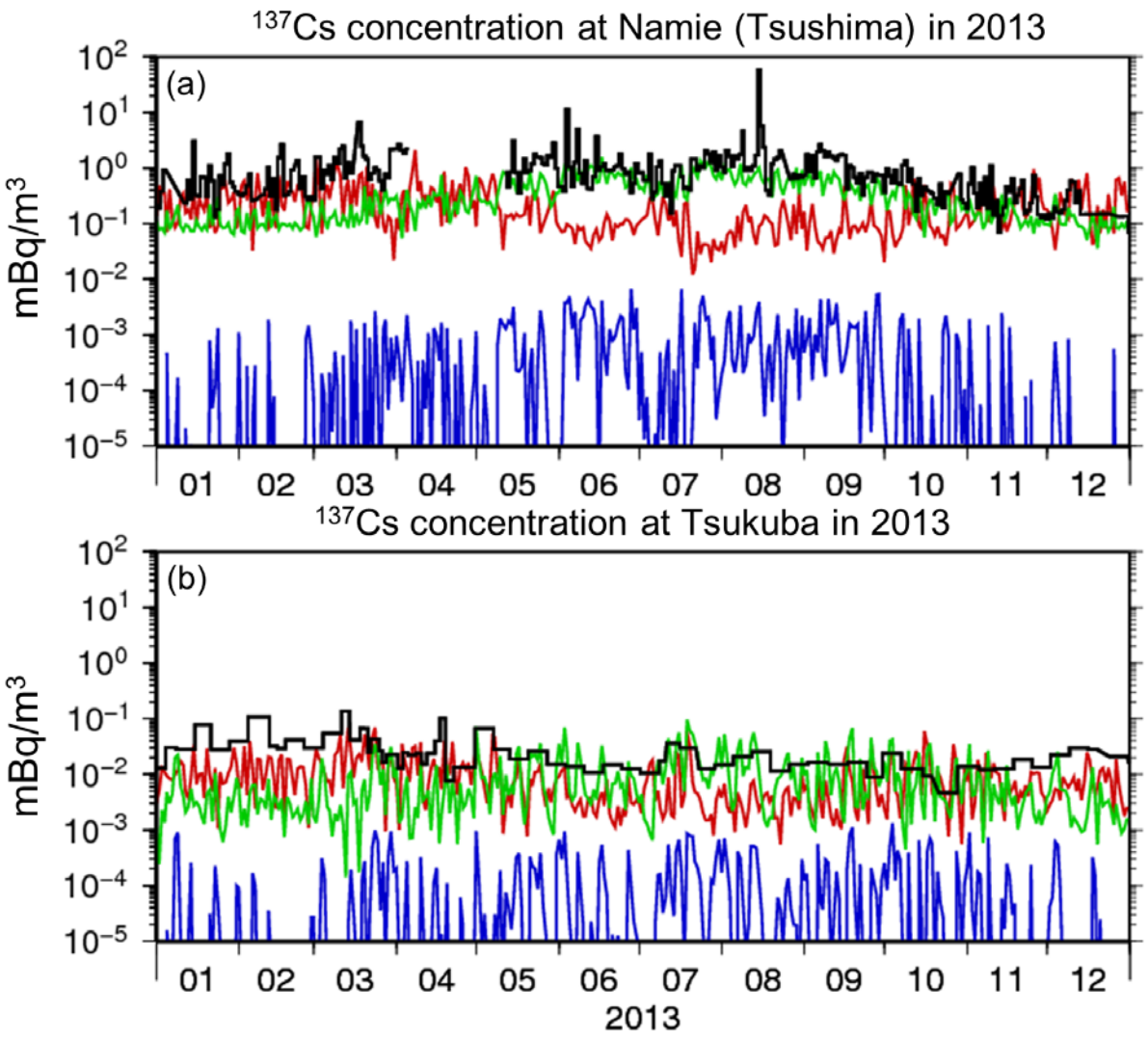


Figure 6. Time series of the surface activity concentration of (black) observed and (red) simulated ^{137}Cs concentrations at Tsukuba from March 12 to April 1, 2011 (Bq/m^3). The red shaded areas indicate the range of ^{137}Cs concentrations obtained when the simulations were run using the optimized parameter ranges E_c = 0.03 – 0.05 and v_d = 0.05 – 0.1 cm/s.


137Cs concentration at Namie (Tsushima) in 2013
(a)
mBq/m3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
01 02 03 04 05 06 07 08 09 10 11 12
137Cs concentration at Tsukuba in 2013
(b)
mBq/m3
01 02 03 04 05 06 07 08 09 10 11 12
2013

1

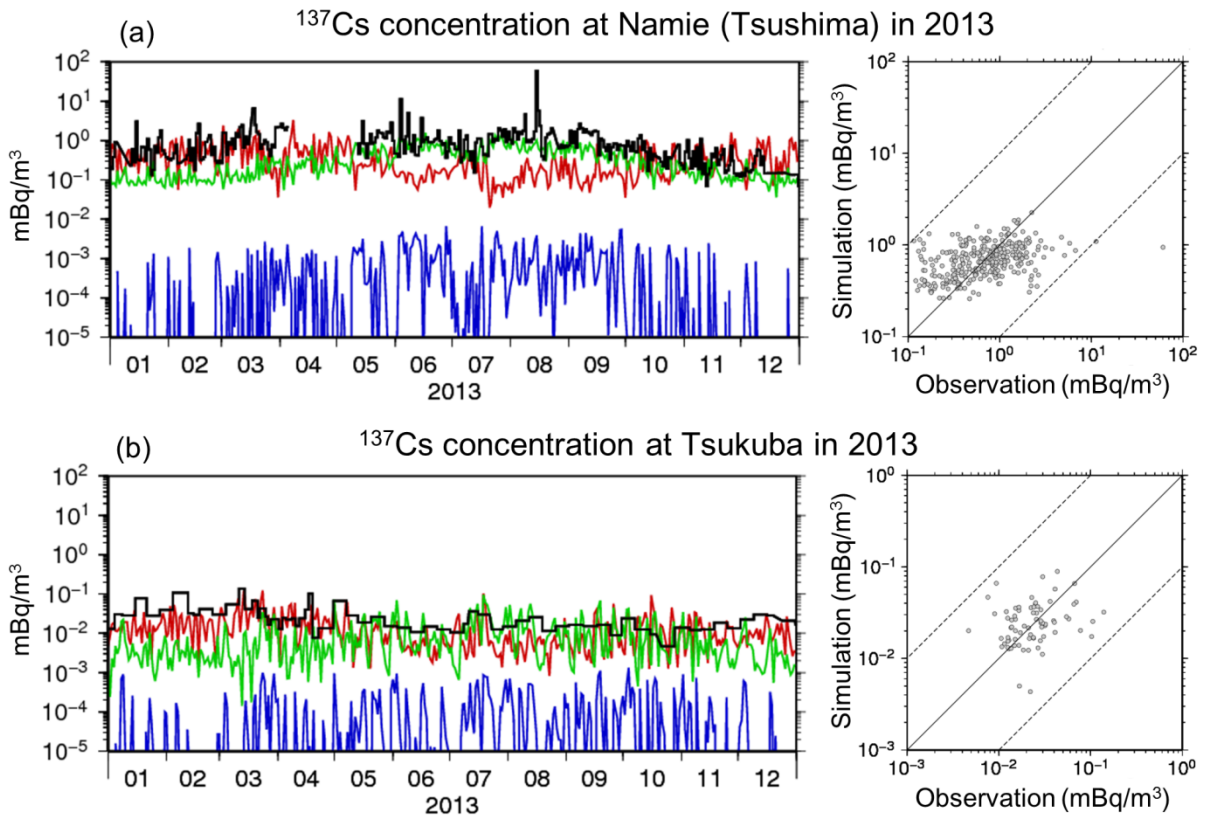
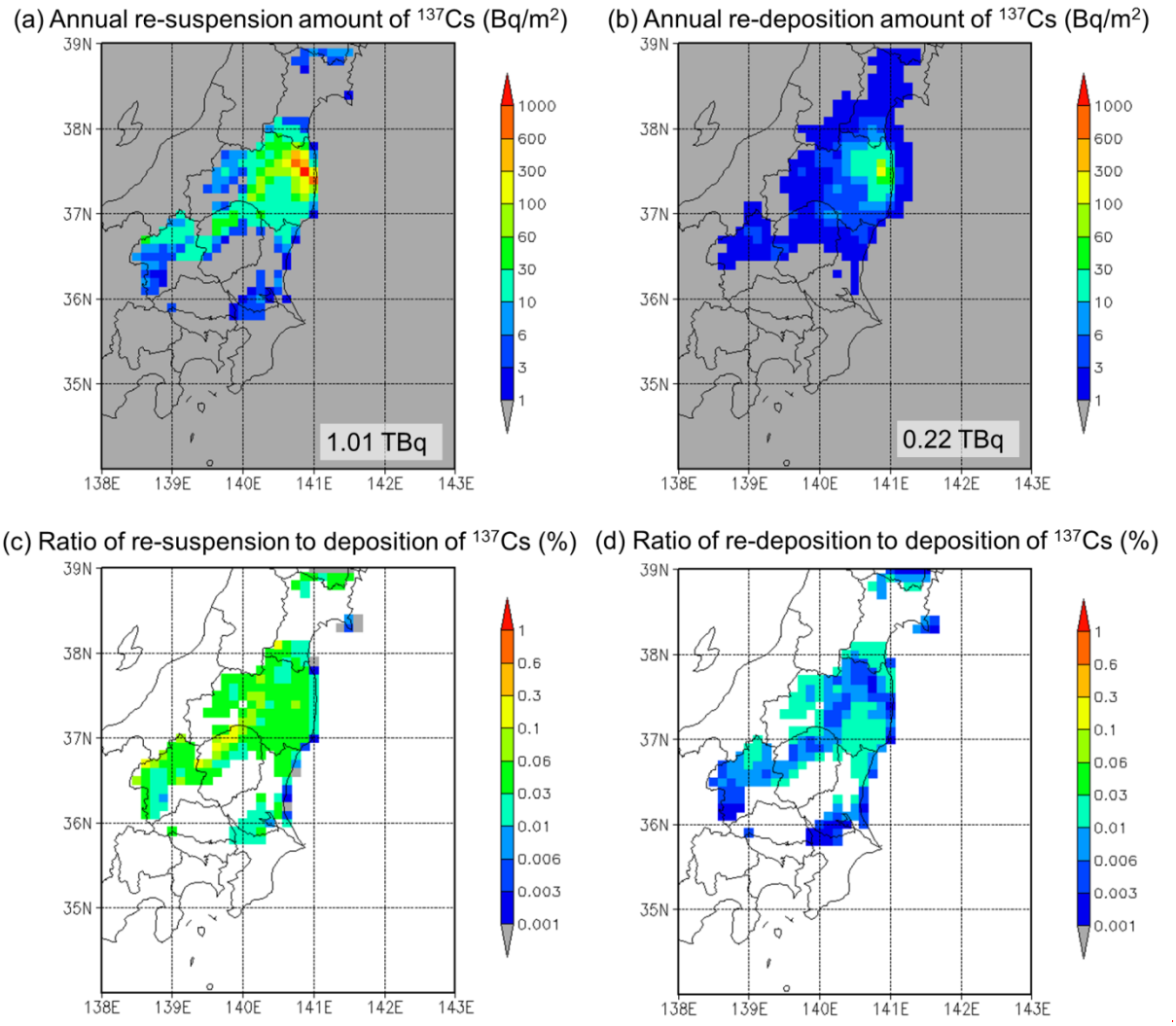


Figure 7. Time series of the surface air concentration of (black) observed ^{137}Cs and (colors) simulated daily ^{137}Cs levels on the left and scatterplots between observed and simulated total (=sum of colors) ^{137}Cs at (a) Namie and (b) Tsukuba. The colored lines indicate simulated ^{137}Cs concentrations due to (red) re-suspension from soil using the scheme given in Ishizuka et al. (2016) (five~~10~~ times), (green) re-suspension from forest with a re-suspension rate of 10^{-7} /h, and (blue) emission from the FDNPP reactor buildings with a constant emission rate of 10^{6} Bq/h. The deposition parameters of the simulation in the figure are E_c = 0.04 and v_d = 0.1 cm/s.


(a) Annual re-suspension amount of 137Cs (Bq/m2)
(b) Annual re-deposition amount of 137Cs (Bq/m2)
(c) Ratio of re-suspension to deposition of 137Cs (%)
(d) Ratio of re-deposition to deposition of 137Cs (%)
1.01 TBq
0.22 TBq
39N
38N
37N
36N
35N
138E
139E
140E
141E
142E
143E
1000
600
300
100
60
30
10
6
3
1
0.6
0.3
0.1
0.06
0.03
0.01
0.006
0.003
0.001

1

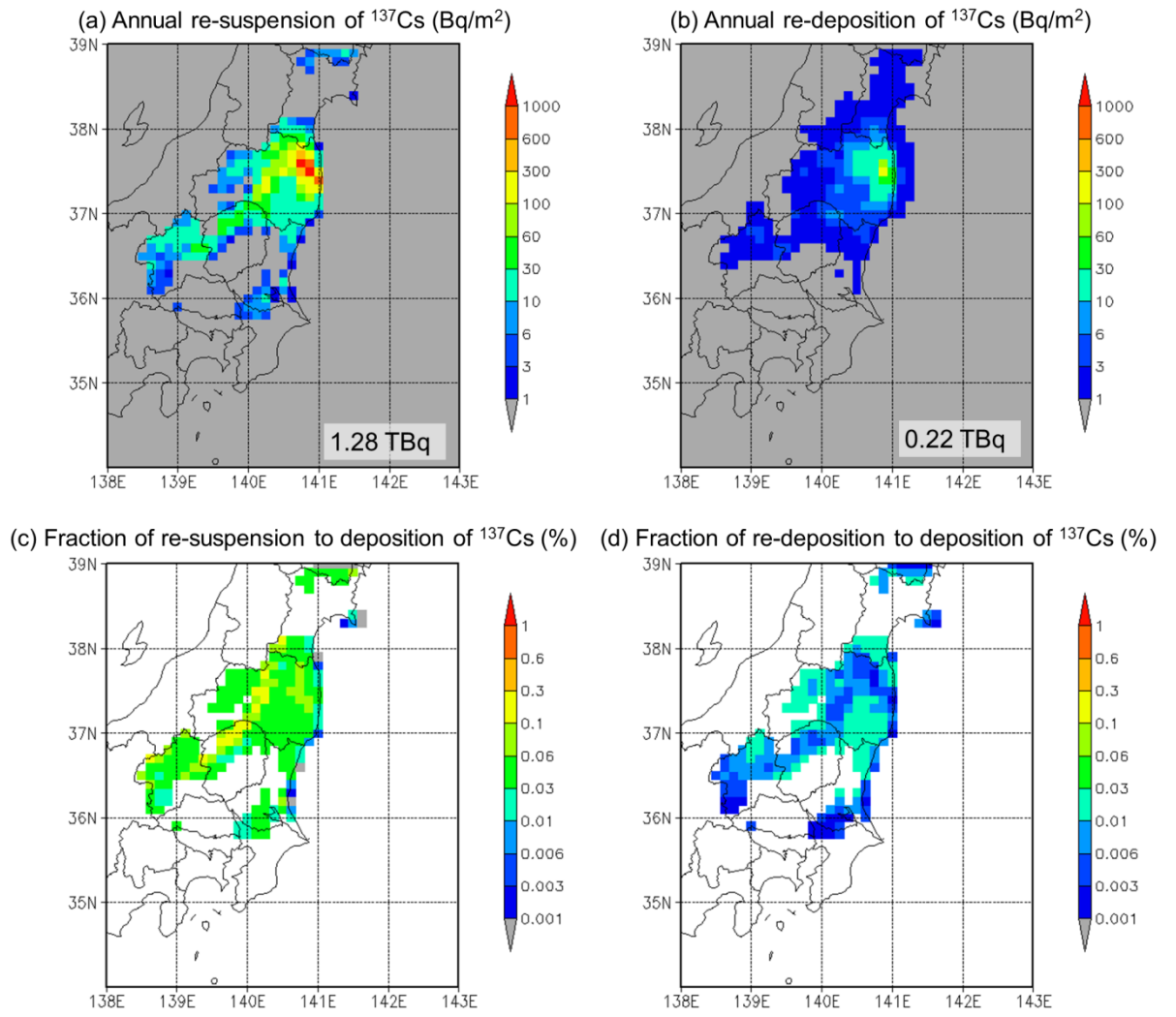


Figure 8. ~~(a)~~ Annual total amounts of (a) the re-suspension ~~amounts of ^{137}Cs~~ and (b) the re-deposition ~~amounts~~of ^{137}Cs. The total activities are presented as numbers. ~~Ratios~~ Fractions of the re-suspension and the re-deposition to the observed ^{137}Cs ~~deposition~~ deposited amounts are also shown in (c) and (d), respectively. The deposition parameters of the simulation in the figure are E_c = 0.04 and V_d = 0.1 cm/s.

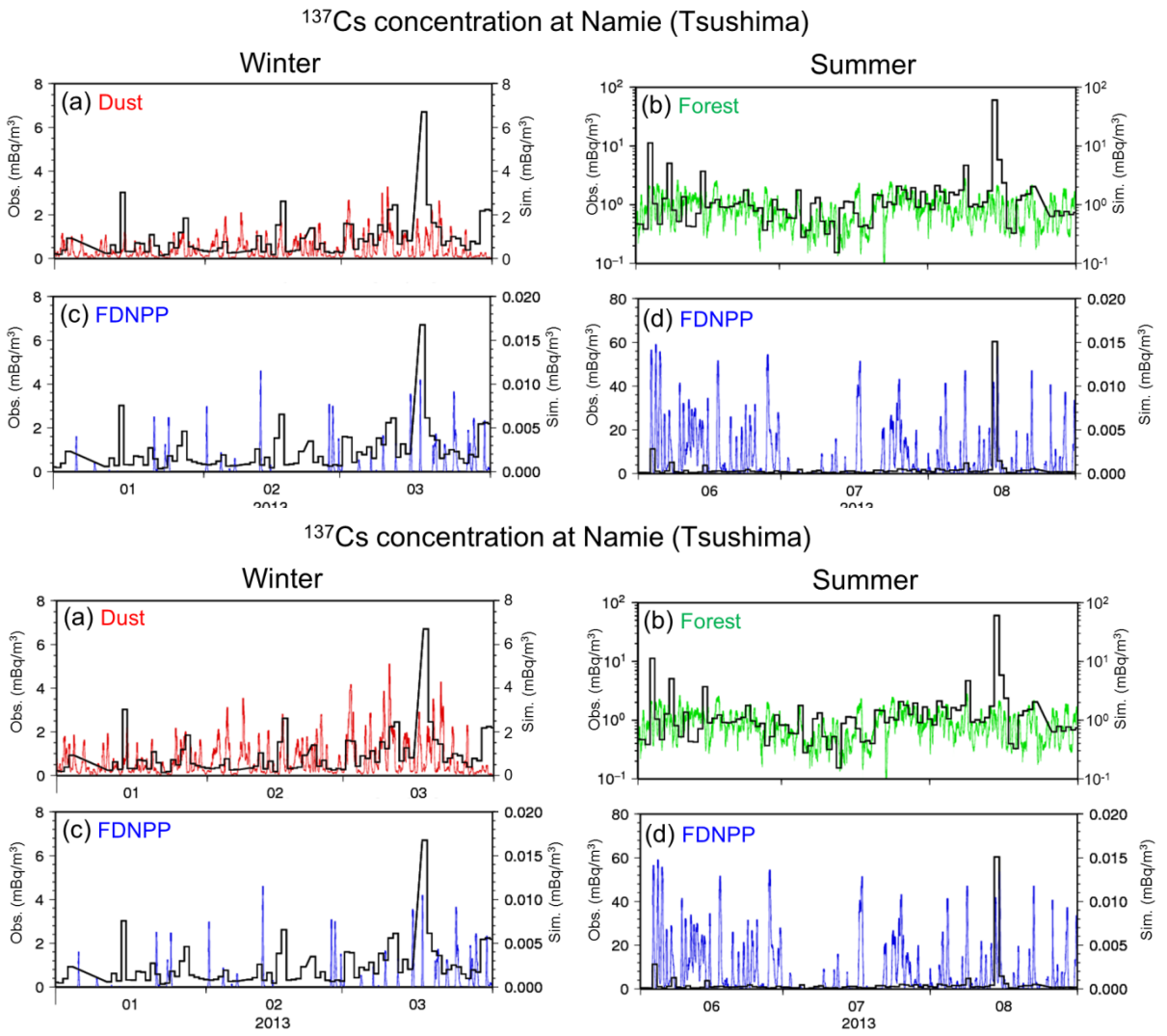


Figure 9. Time series of ~~(black) the~~ observed (black) and ~~(colors) the~~ simulated (colors) ^{137}Cs surface air concentration at Namie (a, c) in winter from January to March and (b, d) in summer from June to August, 2013. The colors of the lines are the same as in Fig. 7 but the time variation is hourly. The shaded areas indicate the range of ^{137}Cs concentrations obtained when the simulations were run using the optimized parameter ranges E_c = 0.03 – 0.05 and v_d = 0.05 – 0.1 cm/s.

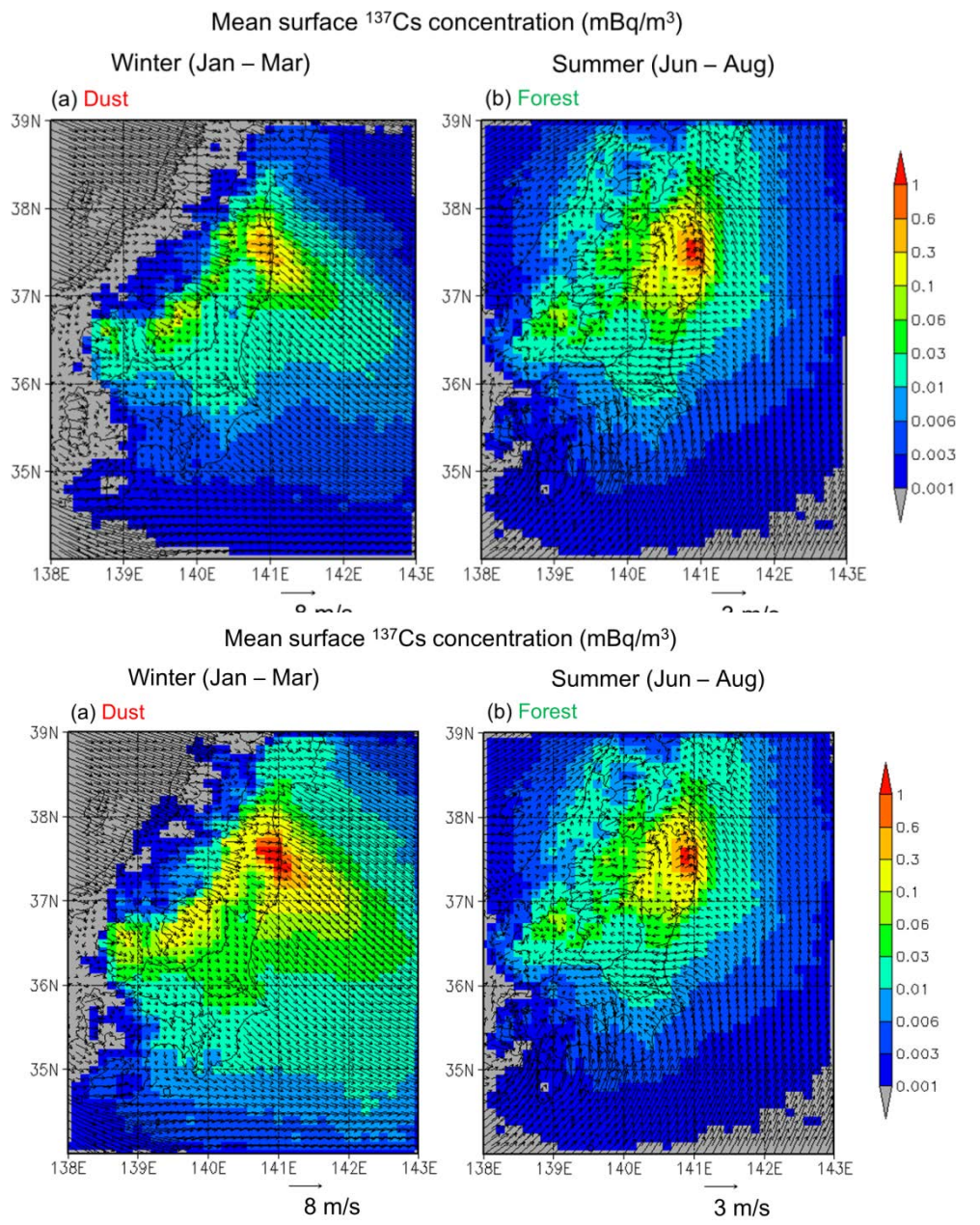


Figure 10. Seasonal mean surface (10 m above ground level) wind vector and ^{137}Cs surface concentration (a) due to dust re-suspension in winter from January to March and (b) due to forest re-suspension in summer from June to August. The deposition parameters of the simulation in the figure are E_c = 0.04 and v_d = 0.1 cm/s.

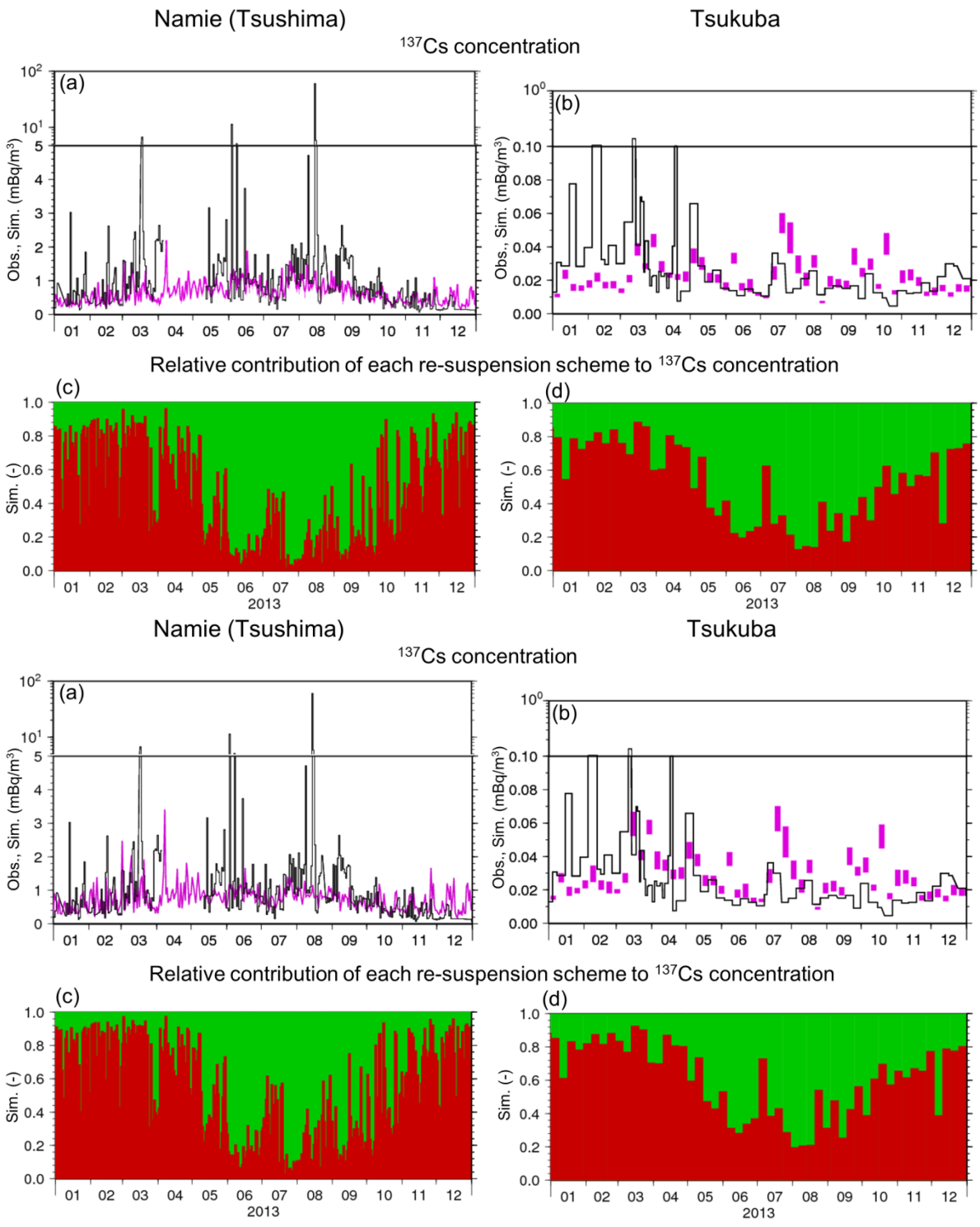


Figure 11. Time series of (a, b) (black) the observed and (purple) the simulated surface ^{137}Cs concentration due to total re-suspension and (c, d) the relative contribution of (red) dust and (green) forest re-suspension to the ^{137}Cs concentration at (a, c) Namie and (b,d) Tsukuba. The shaded areas of (1) and (b) indicate the range of ^{137}Cs concentrations obtained when the

simulations were ran~~run~~ using the optimized parameter ranges E_c = 0.03 – 0.05 and v_d = 0.05 – 0.1 cm/s.

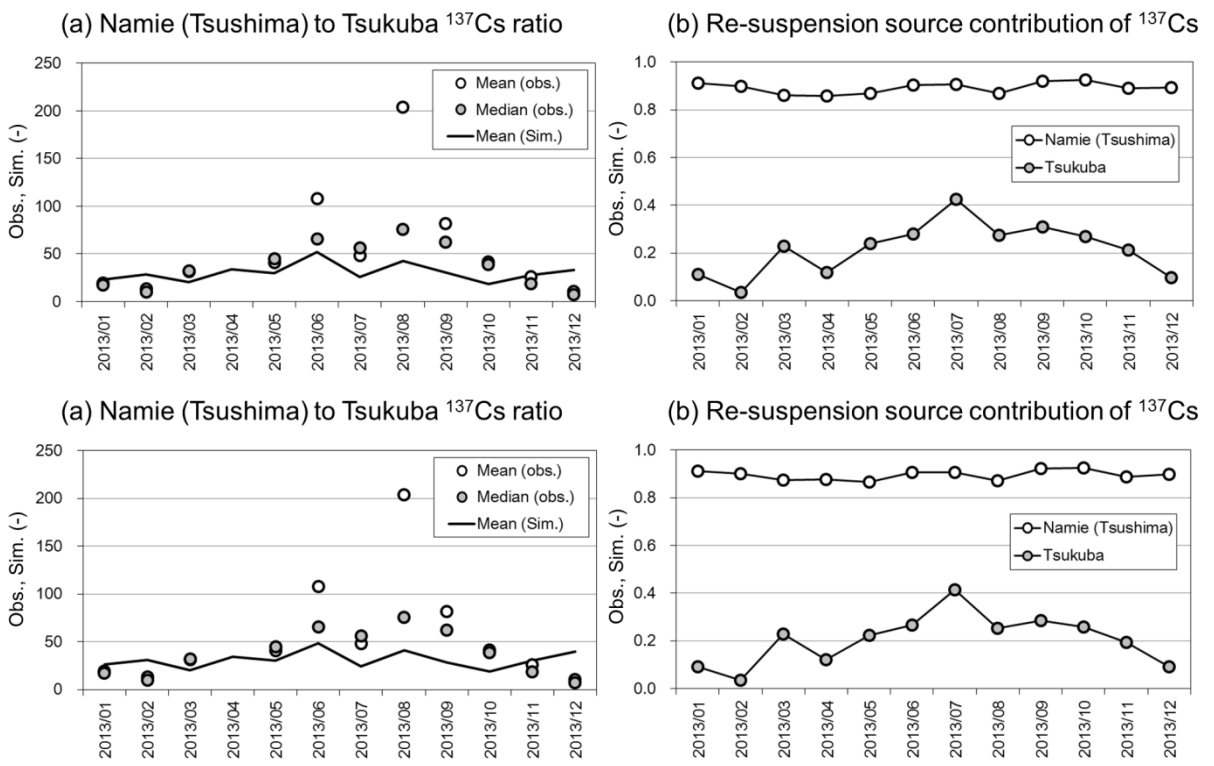


Figure 12. Monthly mean (a) observed mean, observed median and simulated Namie to Tsukuba ^{137}Cs concentration ratio and (b) simulated re-suspension source area (where the observed deposition amount > 300 kBq/m^2) contributions to ^{137}Cs air concentration at Namie and Tsukuba. The deposition parameters of the simulation in the figure are E_c = 0.04 and v_d = 0.1 cm/s.

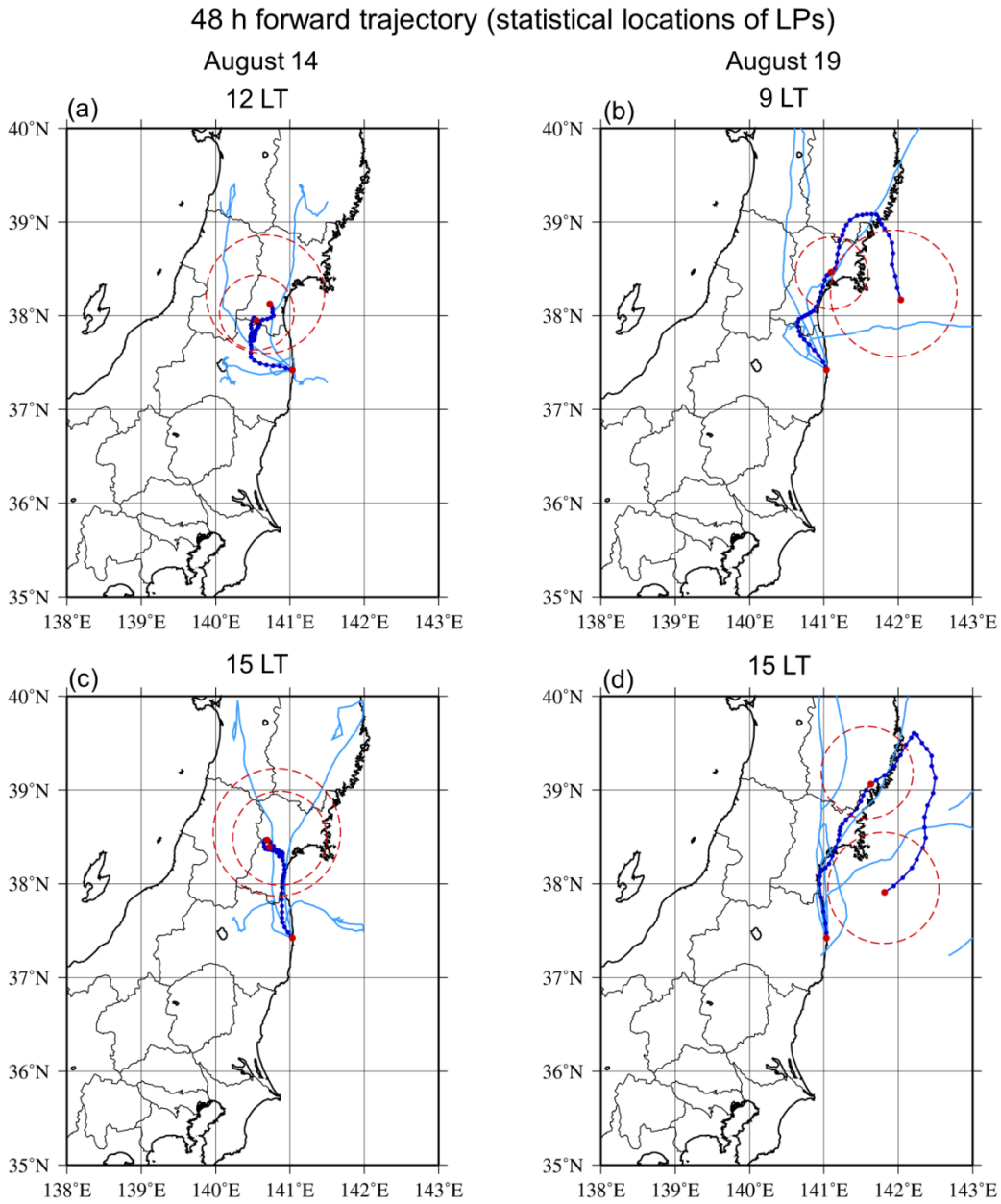
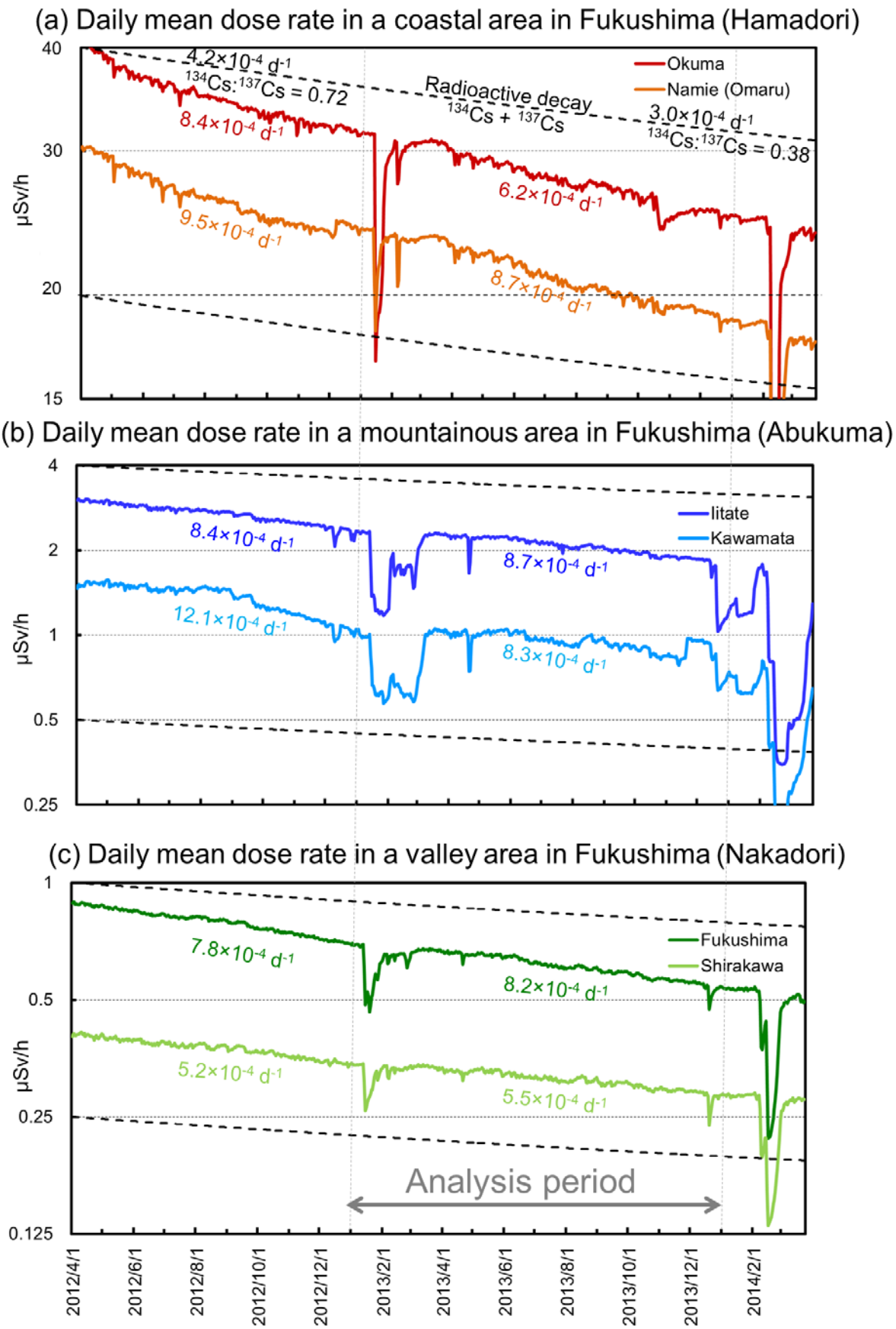


Figure 13. 48 h forward trajectory (statistical locations of LPs within 1 km AGL) predicted by the LM starting at (a) 12 LT and (c) 15 LT of August 14 and (b) 9 LT and (d) 15 LT of August 19. Blue lines indicate median locations of LPs at 1 h and 1 d intervals as blue and red dots, respectively. Sky-blue lines indicate 17th and 83rd percentile locations of LPs and red dashed circles indicate areas containing 66% of the LPs.


(a) Daily mean dose rate in a coastal area in Fukushima (Hamadori)
μSv/h
40
30
20
15
4.2×10-4 d-1
134Cs:137Cs = 0.72
8.4×10-4 d-1
Radioactive decay
134Cs + 137Cs
3.0×10-4 d-1
134Cs:137Cs = 0.38
Okuma
Namie (Omaru)
6.2×10-4 d-1
9.5×10-4 d-1
8.7×10-4 d-1
(b) Daily mean dose rate in a mountainous area in Fukushima (Abukuma)
μSv/h
4
2
1
0.5
0.25
Iitate
Kawamata
8.4×10-4 d-1
8.7×10-4 d-1
12.1×10-4 d-1
8.3×10-4 d-1
(c) Daily mean dose rate in a valley area in Fukushima (Nakadori)
μSv/h
1
0.5
0.25
0.125
Fukushima
Shirakawa
7.8×10-4 d-1
8.2×10-4 d-1
5.2×10-4 d-1
5.5×10-4 d-1
Analysis period
2012/4/1
2012/6/1
2012/8/1
2012/10/1
2012/12/1
2013/2/1
2013/4/1
2013/6/1
2013/8/1
2013/10/1
2013/12/1
2014/2/1

1

Figure 14~~C1~~. Daily mean gamma dose rate at the six monitoring sites: (a) (red) Okuma town and (orange) Namie town (Omaru district) in the coastal area, (b) (blue) Iitate village and (sky blue) Kawamata town in the Abukuma highland area, and (c) (green) Fukushima city and (greenish yellow) Shirakawa city in the Nakadori valley area of Fukushima prefecture as presented in Fig. 1b for two fiscal years (from April 2012 to March 2014). The first-order decreasing rates of the least-square approximation over the period of no snow cover (May to October, 2012 and 2013) is also presented, along with the radioactive decay (^{134}Cs plus ^{137}Cs, by assuming the same amount of activity on March 12, 2011) in April 2012 and March 2013.