Dear anonymous referee #3,

We are very grateful for your time, acceptance of review, and very fine and constructive comments for RC3. Thanks to your review, our manuscript has been substantially improved. Specifically, the comment of removing initial period from the resuspension tendency is very meaningful. We have considered all your comments in the revised manuscript accordingly.

Point-by-point responses to your comments are written in blue in this letter.

With best regards,
Akira WATANABE, Mizuo KAJINO, and Kazuhiko NINOMIYA

[General comments]

[1] This paper observed valuable data with long and steady efforts and showed important and new knowledge (such as change of solubility of Cs containing aerosols in deposition) which can be useful in environmental radioactivity science and various atmospheric science. They aimed to clarify sources of Cs containing aerosol particles, their activities in the environment, and their future estimations. This paper can contribute to the understanding of the environmental cycle of aerosols such as aerosol production, transporting, and deposition by using Cs as a tracer. In addition, this paper rose an important suggestion for the model improvements through improvements of aerosol deposition estimations. Therefore, I think this paper is appropriate for publishing from ACP.
[1] Thank you very much for your evaluation.

[2] However, this paper remains a large uncertainty to the aerosol size measurements. They are making great efforts to evaluate the performance of the 6-stage impactor with the cyclone/impactor instrument. They discussed Cs containing aerosols on the backup filter using a large part of this paper, however, the bouncing effects of large particles had not been denied. Rather, significant bouncing effects on both instruments were shown, but there was no evidence to deny bouncing effects. Therefore, I think a large contribution of bouncing effects will be quietly significant. These results and discussions about the particle sizes make this paper confusing. If authors suggest that the significant contributions of fine aerosol particles, more accumulation of reliable and accurate evidence for this point should be required (such as parallel observations using the same impactors with normal filters and adhesive material applied filter (such as vacuum grease), microscopic observation, and others).
[2] Thank you for raising the very important issue. As for the parallel sampling, we made a new item in the remained issue section of Conclusion for the vertical measurement according to the comments of RC1(#2) and RC2 (#4). In the similar manner, we made an additional item (2) in the remained issue section of Conclusion as follows:

“(2) The rebound issue of the impactor and the cyclone/impactor instruments have not

yet been resolved. Parallel sampling is also required for the size-resolved measurements using normal filters and filters with adhesive materials such as vacuum grease. The additional microscopy of the filters is even more useful."

Other common comments are below.

[3] Please check the significant figures (such as P8 L6) and make the numbers easier to see.
[3] Thank you for your comments. We changed it from 1.158 to 1.16 here. As the significant figures of observation data are from 2 to 3 (please see the supplement excel file), we determined the significant figures of all values derived from statistical analyses not exceeding 3. Thus, we carefully checked all values in other locations and changed them as follows:
"202200 Bq m^{-2}" → "202 10^3 Bq m^{-2}"
"48232$X^{-1.944}$" → Whole sentence removed (Please refer to #7 of RC2)
"0.678" → "0.68" for readability
"7.8376$x^{1.0542}$" → "7.84$x^{1.05}$"
"R^2=0.9965" → "R^2=0.997"

[4] Please clarify the relationship between river sediments and this paper more. This observation does not seem to contribute to the paper significant
[4] In the revised manuscript, the relationship between river sediments and this paper is elaborated in Sect. 2.4 (the methodology section), when it is first appeared in the manuscript as follows:
"River sediments that characterize the surface soils of the Nakadori valley were also measured to assess the composition correlations with the airborne and deposition samples."

[5] "surface air concentration", "atmospheric radioactivity concentration", and "surface concentration" were confused (atmospheric radioactivity concentration?).
[5] Thank you very much for your comment. After considering #5 of RC1 regarding Line 22 of Page 3, we changed "surface activity concentrations" to "airborne surface concentrations". To keep the consistency in the manuscript, we only used the two terms "airborne surface concentrations" and simply "concentrations" throughout the manuscript.

[6] This paper calculated radioactivity decreases using data from 2011 (the early stage after the accident). In the early stage, resuspension is not dominant. It is necessary to distinguish periods of primary stages and resuspension stages. This point will affect the results of future estimations and the rate of Cs discharges.
[6] Thank you very much. It is a very good point. We re-calculated all tendency factors by excluding the initial period of data (March and April 2011 of deposition).

For the concentration measurement (Fig. 2), it started on May 18, 2011, we used all data and so the values have not been changed. For the monthly deposition measurement (Fig. 3), March 2011 data should be excluded, but it is a question of

whether to exclude the April data. Based on Katata et al. (2015), total emission amounts of $^{134}Cs+^{137}Cs$ from FDNPP in April was 0.7 PBq, which accounted for 2.4% of total emission (28.7 PBq). The value “2.4%” seems to be small but with compared to the resuspension rate (less than 1%/yr; Kajino et al., ACP, in press; doi:10.5194/acp-2021-687), it may not be negligible. Thus, we decided to exclude the April data, too.

[7] Please check again for the referencing.
[7] Do you mean the consistency of references in the main text and in the reference list? If so, we changed from “Steinhouse et al., 2015” to “Steinhouser et al., 2015” as found by RC1 and “Kinase et al., 2017” to “Kinase et al., 2018” as found by you (#16 of RC3), but we could not find any further inconsistencies. If it is not the case, kindly please specify the errors. Thank you very much for your time.

[Specific comments]

[8] P6 L15: “The range of particle sizes…” can be shortened as “The 50% cut of particle sizes…”. Then, “(Note…” can be deleted.
[8] We changed it accordingly.

[9] P7 L5: Were carbon filters used for gaseous Cs analysis? Isn't the purpose such as measuring iodine?
[9] Yes, it is. By taking #5 of RC1 (regarding Line 19 of Page 6 and Line 6 of Page 7) into account, we modified the sentences.

[10] P7 L12: Coarse mode samples were collected on the quartz fiber filter. This point is inconsistent with the L8 paragraph.
[10] XRF is applied only for the fine mode fractions which were collected in the glass bottles. The relevant sentence is modified as follows:
“Aerosols larger than 2.5 μm were collected on quartz fiber filters in the system and thus only the fine mode particles in the glass bottles were measured by XRF.”

[11] P13 L14: Also impactor sampling is “time-resolted measurement”.
[11] Yes, it is. We changed it to “all size observation as presented in Fig. 2”.

[12] P13 L24: 2016 and 2017? Or 2015 DJF and 2015 DJF? Also, the opposite trend can be seen in 2013.
[12] I am sorry for the confusion. The sentence is the explanation of only >10.2 μm and not the explanation of comparison between >10.2 μm and backup filter. You mean the opposite trend in 2013 is for >10.2 μm and backup filter, but we meant that the trend of >10.2 μm of 2016 and 2017 (high in JJA) are different (not opposite) from other years (high in DJF and MAM9.

We modified the sentences from
“The seasonal variations in the largest particle fraction, larger than 10.2 μm, are interesting. The trend appears to be synchronized with that of the backup filter particles (high in DJF and MAM), but the opposite trend was observed in 2016 and 2017 (high in

JJA).”

to
“The seasonal variations in the largest particle fraction, larger than 10.2 μm, are interesting: high in DJF and MAM (same as the backup filter) but high in JJA in 2016 and 2017.”

[13] P13 L25: Is the seasonal trend of 1.3-2.1 μm particles significant? It looks quietly stable.
[13] Yes, it is not significant. These parts are discussing the trends of minor size ranges so the following sentences are all REMOVED:
“The contributions of other fractions, i.e., 0.49-4.2 μm, were small in the measured period. Even though the contributions were small, the seasonal trend of 0.39-0.69 μm was similar to that of the backup filter particles, but that of 1.3-2.1 μm was similar to that of 4.2-10.2 μm.”

[14] P15 L3: Too long sampling intervals (recommended operating time is up to 24 hours). Were some parallel observations using the same impacter instruments with and without oil? Were some microscopic checks or any other checks had done?
[14] Unfortunately, no. Since it is the very important and critical point, we itemized the issue in the remained issue part of Conclusion as follows:
“The rebound issue of the impactor and the cyclone/impactor instruments have not yet been resolved. Parallel sampling is also required for the size-resolved measurements using normal filters and filters with adhesive materials such as vacuum grease. The additional microscopy of the filters is even more useful.”

[15] P22 L: Is the wording “difficult -to-return zone” is right? I could find “Areas where it is expected that the residents have difficulties in returning for a long time” in the Japanese governmental report.
[15] Yes, it is found for example in the national report issued by Government of Japan (p.33). (last accessed: December 8, 2021)
https://www.iaea.org/sites/default/files/japan_2nd_em_2012.pdf
The term is currently used in official web pages as well, for example,
https://www.pref.fukushima.lg.jp/site/portal-english/en03-08.html

[16] P22 L26: 2017? 2018?
[16] Thank you. We changed it to 2018.

[17] P24 L17: Authors showed a paper (Okuda et al., 2015) as a reference indicating the rebounds of large particles using the impactor/cyclone instrument with long sampling periods in P7.
[17] We modified the relevant sentence as follows:
“(4) is possible because Okuda et al. (2015) showed that the long-duration impactor/cyclone measurement could be associated with the bouncing effect despite the use of silicone oil.

[18] P27 L13: At the city site, some references showed the same results of seasonal variations (such as Kitayama et al., 2016; Kinase et al., 2019).
[18] We inserted the following underlined sentences in the relevant place:
“these seasonal trends are the same of those observed in the city area (Kitayama et al., 2016) and the opposite of those observed in a contaminated forest area (Ochiai et al., 2016; Kinase et al. 2018)”.

We could not find the paper “Kinase et al. (2019)” of either Dr. Takeshi Kinase and Dr. Sakae Kinase. We suppose it as Kinase et al. (2018), because their measurement was conducted in the forest area, but they also claimed that the trend in the forest differed from that in the other locations. We already cited their paper in the sentence so we additionally cited only Kitayama et al. (2016) for the Fukushima city case.

In case if we misunderstand the reference, kindly please provide the full information of the relevant paper. Thank you for your time.

[19] P28 L3: As mentioned above, these results include high risks of misunderstandings about Cs containing particle sizes.
[19] We agree with you in this point. We removed all size values from the item #3 and modified it as follows:

“(3) The size-resolved measurements revealed that seasonal variations of ^{137}Cs of different sizes are different from each other. Due to the possible bouncing effect of the cascade impactor and long-duration measurement of the impactor/cyclone system, it is hard to quantify the values, but the current measurement indicates that the dominant particles and their sizes may be distinct depending on the season. The XRF analysis showed that biotite may have played a key role in the environmental circulation of particulate forms of resuspended radio-Cs in Fukushima city after September 2014.”