Dear anonymous referee #2,

We are very grateful for your time, acceptance of review, and very fine and constructive comments for RC2. Thanks to your review, our manuscript has been substantially improved, especially for your suggestion on the period of change the in tendencies. 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 has studied and contributed to not only the better understanding for the chemistry of Cs in the atmosphere but also the earlier rebirth from nuclear accident. This study is quite challenging to the non-reproducible event based on the eight-year measurement. Therefore, this study should include many uncertainties. Under such a difficult situation, this paper can give scientists many useful information and knowledge including unsolved agenda. In this meaning, this study should be appropriate for publishing in ACP.
[1] Thank you very much for your evaluation.

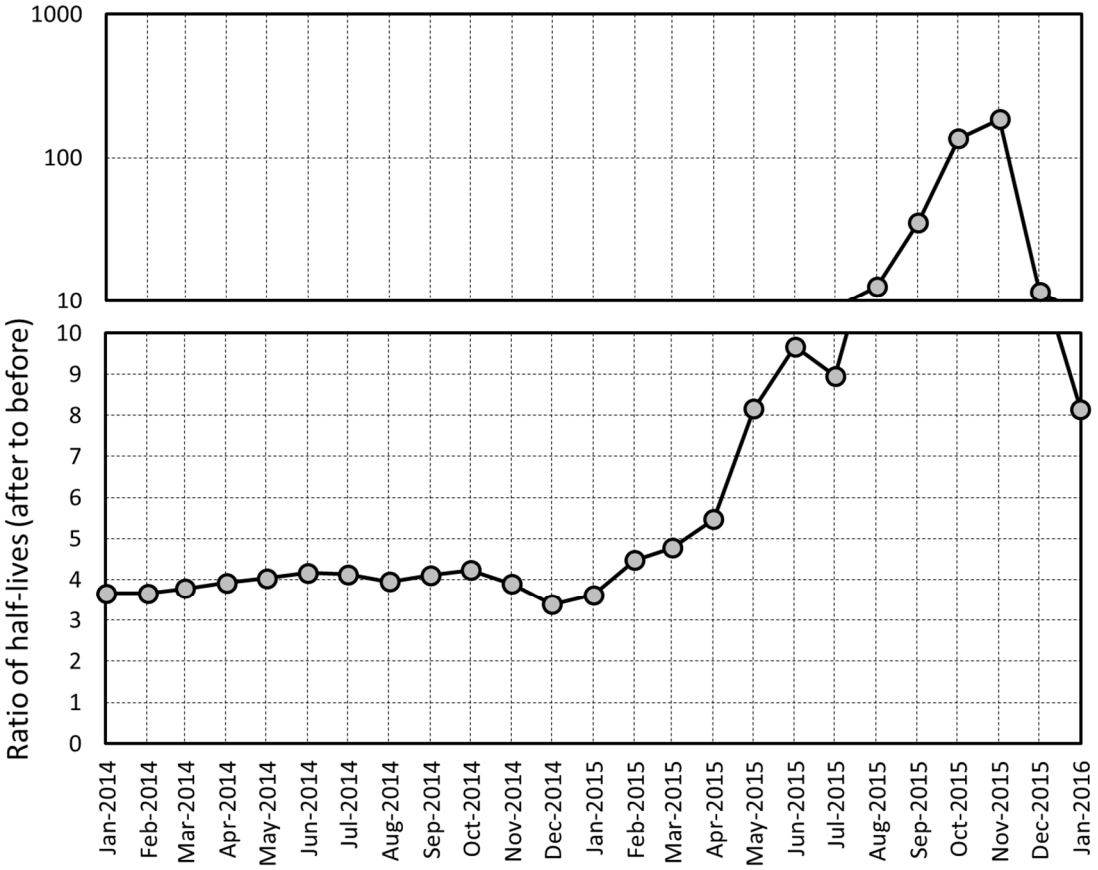
On the other hand, I expect the authors to describe and give suggestions to readers for the points below.

- [2] Give us a clearer scientific (physical, chemical, mathematical or some other) reason why the authors consider that the year of “2015”, neither “2014” nor “2016”, was the turning point in time series, especially for Fig. 3.
- [2] Thanks to your comment, we could find the mathematical reason, indicating that something happened in 2015. In the previous manuscript, it could be either 2014 or 2016 and thus we used the fuzzy term “approximately 2015” which can be from 2014 to 2016. We additionally calculated the half-lives before and after 2014 and 2016, and as you pointed, revealed that there was a significant difference for the 2016 case of the deposition (Fig. 3).

  In addition, after considering the #6 of RC3, we excluded the initial stage data which contains the effects of primary emission in addition to resuspension from the tendency calculation. Thus, please be aware that all the tendency values are changed accordingly.

  The ratios of half-lives before to after January 2014, January 2015, January 2016 are 2.75, 3.06, 2.84, (all around 3.0) respectively for concentrations, but those for depositions are 3.61, 3.64, and 8.12. Some drastic change might occur within the year of 2015.

We made a new figure showing changes in the half-lives of depositions before and after a particular date from 2014 to 2016 as below.



Unfortunately, because half-lives after 2015 varied substantially depending on the start month (it exceeded 100 year for some cases), the ratio after August 2015 is really unstable. Therefore, we gave up to show this figure in the main text but it was inserted in the supplementary data in the excel file (please find tab number #7).

Anyway, this figure indicates that there is not sudden but gradual change started to occur from spring to summer of 2015.

- [3] Why should the fractions in dissolved and particulate change suddenly in 2015? Give us scientific reasons/comments/discussions in detail more.
- [3] We couldn't find any specific reason for this in addition to what we extensively discussed: "dissolved Cs discharged faster from environment than undissolved". Still, however, as shown above (our reply to #2 of your comment RC2), we could find that there was not a sudden change, but gradual changes occurred during 2015.

Consequently, by taking #2 and #3 of RC2 into account, the following changes are made in the manuscript:

(1) A new paragraph is made as the 3rd paragraph of Sect. 3.1 as follows:

"The regression analysis is also performed over different time periods, but the results are not substantially different. The T_h and R_d before December 2013, 2014, and 2015 are 0.670, 0.753, and 0.900 yr, and 103, 92.0, and 77.0 % yr^{-1}, respectively. The T_h and R_d after January 2014, 2015, and 2016 are 2.05, 20.7, and 2.56 yr, and 33.8, 33.5, and 27.1 % yr^{-1}, respectively."

(2) A new paragraph is made as the 4th paragraph of Sect. 3.2 as follows:

"The regression analysis is also applied over different time periods and we found a remarkable change in 2015. The Th before December 2013, 2014, and 2015 are similar 1.09, 1.30, and 1.56 yr, respectively, but Th after January 2014, 2015, and 2016 are 3.98, 4.69, and 12.67 yr, respectively. The ratios of half-lives (after to before) of the three periods are 3.64, 3.61, and 8.12, respectively, indicating that there could be a remarkable change in the tendency between January 2015 and January 2016. Time series of changes in the ratio before and after a particular date from 2014 to 2016 are illustrated in Fig. S1. Due to the lack of data numbers, the half lives after 2015 varied substantially depending on the start month (it exceeded 100 yr for some cases). However, it is obvious that the ratio is stable before January 2015 around four and start to increase from the spring to summer of 2015. We may be able to conclude that the regime change in the physicochemical properties of radio-Cs occurred during the year of 2015."

- [4] When we compare the results between forest sites and current study sites, the sampling height above the ground level might be different. Is there any influences on the measurement results and the subsequent interpretations of the data? (Around p.10, LL.12-14)
- [4] Please also refer to our reply to the comment #2 of RC1. The vertical difference matters for emission source but not in downwind areas. Whether the Fukushima University site is emission or downwind may differ by seasons and relative abundance of carrier aerosols (for example, downwind for forest aerosols and emission source for soil particles (especially for road dust)). These statements are itemized in the remained issue section of Conclusion as follows:

  "The height of our measurement (building roof) is higher than the other measurements referenced in this study (near the ground). When the observation site is characterized as an emission source, there should be a clear vertical difference in concentration, and thus the concentration measured at Fukushima university is not equivalently comparable with the other location data. It may be comparable when the site is characterized as a downwind region, because turbulent mixing during transport may reduce the vertical difference. In the future, parallel sampling near the ground and rooftop will need to be installed to characterize the sampling locations and to quantify the vertical differences at the site."

[Specific comments]

[5] p.3, LL.11-12, Why do we need bracket; (The……2011).
[5] We removed the bracket.

[6] p.6, L.13, Is “Shibata” “Sibata”?
[6] Thank you for the good point. We did not realize it.

[7] p.9, L.14, Why do the authors select power of X, not exponential function?
[7] The function of power of X fitted better than the exponential function, but it was contradictory with the latter statement saying that “This demonstrates that the concentration decreased “exponentially”. We simply removed the sentences because we have already discussed the trend elsewhere in the manuscript. Same for the first paragraph of Sect. 3.2.

[8] p.16, LL.11-13, On the description of “We ……and autumn.”, is the reason for “We can assume” either the present measurement results or other references? If the former is, is the description of “our measurement strongly indicates” better than “we can assume” ?
[8] Thank you. We modified the sentences as follows:
“There was a negative correlation between PM_c and PM_f, which strongly indicates that… and autumn.”

[9] p.17, L.17, Why can the author simply say “the samples have similar origins” although the precipitation is influence by not only below cloud scavenging but also in cloud scavenging?
[9] Yes, it was an overstatement. We changed the whole sentence as follows:
“Composition differences are not very remarkable, the correlation coefficients for the compositions among samples are above 0.9.”

[10] p.20, L.13 and p.27, LL.28-29, Is there any conflicts between two sentences of “it seems that wet deposition plays an important role in the removal of…” and “Therefore, decontamination may play a partial role in explaining the differences…”?
[10] Thank you for your question. It is not contradicting because the former sentence describes the removal mechanism of resuspended ^{137}Cs in the air, whereas the latter sentence describes the reasons in changes in the trend before and after 2015.

To be clearer, the following change is made for the former statement as follows:

Former place:
From “it seems that wet deposition plays an important role in the removal of resuspended ^{137}Cs-bearing atmospheric aerosols”

To “it seems that wet deposition plays an important role in the removal of resuspended ^{137}Cs-bearing particles from the air”