

Dear anonymous referee #1,

We are very grateful for your detailed review and constructive comments and your time for RC1. Thanks to your review, our manuscript has been substantially improved, especially for details and preciseness. We have considered all your comments in the revised manuscript.

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] The authors investigate the role of resuspension in the persistence of airborne radio-cesium in the formerly contaminated city of Fukushima. They focus on dissolved vs particulate fractions of ^{137}Cs as a supplementary reason to explain the change in the effective half-lives of airborne radio-Cs and its seasonal variation. Airborne concentrations, deposition and size distribution analyses are consistent and relevant. This study adds novelty in the fate of airborne radionuclides and their apparent environmental half-lives.

[1] Thank you for your evaluation.

[2] The height at which some of the used aerosol sampler and impactor were installed may be not perfectly propitious to reveal the exact signature of the resuspension process because this height is too high as compared with ground level where the resuspension process originates. The possible bias could have been investigated. The respective contribution of the fine-mode particle on the airborne concentration may suffer from this particular settlement. I suggest to install a sampler at ground level for a period of one year in parallel with the sampler already installed on the roof of the building to check if the location height has a significant influence on the airborne concentration (what is expected given the vertical profile of aerosol usually observed). This could also be mentioned in the remaining issues to investigate.

[2] Thank you for your good suggestion. We fully agree with your point. We inserted the following statement in the first item of the remained issue:

“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 Fukushima University site.”

[3] It is not clear if large particles have indeed being evidenced by microscopy on the backup filter. It is clear on the other hand that a sampling period as long as 3 weeks may favor particle bouncing when using high-volume impactors. Usually, impactor trials last about 2 weeks subject the particle number is very low.

[3] There is no evidence that the rebound occurred in the backup filter in our samples, as written in Sect. 4.2. The electron microscopy revealed evidence of rebound of soil particles and bioaerosols in the same experimental setup (Kinase et al., 2018) as explained in Sect. 2.2.3.

Considering together with the comment #2 of RC3, we added the following statement as item (2) in the remained issue section of Conclusion:

“(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.”

[4] The role and characteristics of biotite are highlighted and the role of the gradual decontamination is scrutinized and show that this sole parameter cannot explain the shift in the half-lives of airborne Cs, thus suggesting the bioavailability of the different chemical forms of Cs in soils as an important factor.

[4] We agree with your point. We inserted the following statement at the beginning of the second item of major findings in Conclusion:

“The bioavailability of different chemical forms of radio-Cs in soils may be an important factor to determine the tendencies of concentrations and deposition at Fukushima University.”

[5] Detailed comments:

[5] Replies are inserted in the tables.

Page	Lines	Comment/Reply	Example
Abstract	Line 4	Use « effective half-lives » instead of « half-lives » We changed it accordingly.	
	Line 5	Convert all durations in year and add respectively after We changed it. Also 756d → 2.07 and 4.69 yr. We also changed them in the figures.	(0.75 and 1.11 yr, respectively)
		Abbreviation for « year » is « yr » We changed them all accordingly.	Change it everywhere in the document
	Line 6	I suggest to cut the explanation given line7 and 8 about the shorter half-lives and paste it just after line 5 We changed it accordingly.	
	Line 11	« an evaluation method » instead of « a method of evaluating » We changed it accordingly.	

Main text Page 3	Line 8	« by precipitation (wet deposition) or during dry weather conditions (dry deposition)» instead of « via precipitation in addition to via dry Deposition » We changed it accordingly.	
	Line 11	Terada et al., 2020). First parenthesis is missing. Remove the period after the final parenthesis We changed it accordingly.	
	Line 13	I suggest to replace « may not be substantial » by « is not expected to be significant » We changed it accordingly.	
	Line 20	since several papers have been published give some references Thank you. The following sentences are the references so we added the phrase « as follows » at the end of the sentence.	
	Line 22	Replace « surface activity concentrations » by « airborne surface concentrations » here and in hereafter in the rest of the document when it refers to concentration of radio-Cs in the atmosphere We changed them all.	Change it everywhere in the document when needed
Page 4	Lines 14, 15	The reason is just because in Kinase et al. (2018) the air mass did not pass over the observational sites. You cannot let this sentence as it is since it could led to a misinterpretation (i.e. a fire cannot re-emit formerly deposited radio-Cs). Numerous researches performed in the Chernobyl environnement give evidence that fire can re-emit radio-Cs Yes, we agree with you. They/we didn't have any evidence that forest fire in Fukushima did not reemit Cs. We changed the relevant sentences as follows : « Certainly, it is not indicated that the forest fire did not reemit radio-Cs, because in fact wildfire played a key role in the migration of radio-Cs in the Chernobyl case »	I would suggest to be very cautious with the role of biomass burning
	Line 20	« (Steinhauser et al., 2015 « instead of « (Steinhouse et al., 2015) » Thank you. We changed it accordingly.	
Page 5	Line 6	You can remove the second « located » We changed it accordingly.	
	Line 7	Put the « m » of mountains in capital We changed it accordingly.	Ou Moutain
	Line 12	Replace « where the peaks are » by « where the summits are » We changed it accordingly.	
Page 6	Line 1	Same remark We changed it accordingly.	

		Replace « at a height of 25 m from» by « at a height of 25 m above» We changed it accordingly.	
	Line 19	« gas-state aerosols » is meaningless. Aerosols are liquid or solid particles. Prefer « volatile or semi- volatile compounds » or « gaseous and volatile or semi-volatile compounds ». I think the exact reason of a charcoal cartridge is not for Cs, may be to track possible ¹²⁹ I revolatilization ? Yes. We changed it to « volatile or semi-volatile compounds ».	
Page 7	Line 6	I have never heard about « gaseous radioactive cesium ». Cs may be volatilized only at temperatures above 650°C but will condense again rapidly as temperature falls. Thus it is considered that it exist only as particle in the atmosphere. Yes, it is already obvious. Thus, the whole sentence was removed.	
Page 8	Line 4	You write « official method ». Is it a national or international method ? Please give a reference It is the national method developed based on international literatures. We inserted the reference as « MEXT, 1976 », which is available at https://www.kankyo-hoshano.go.jp/wp-content/uploads/2020/12/No3.pdf (in Japanese)	
	Line 29	Instead of « which are usually larger than the submicron size range » you can use « which are usually in the supermicron size range » We changed it accordingly.	
Page 9	Line 13	Prefer « the decreasing trend » instead of « the decreasing tendency » We changed it and all relevant places.	
		I suggest « which is much higher than the radioactive hal-life of ¹³⁷ Cs » instead of « thus, the decrease rate was higher than the rate of radioactive decay of ¹³⁷ Cs. » Thank you for the suggestion. Please note that the relevant sentence was removed according to the #7 of RC2.	
Page 10		Because of the numerous data in this plot I strongly suggest to downsize the circles on Figure 2 to see the line between the circles We downsized them by two points, from 6 to 4.	
Page 11	Line 2	202200 should be written 202,200 or 202.2 10 ³ We changed it to 202 10 ³ . (Please also refer to #3 of RC3).	
	Line 9	Convert ($T_h = 275 - 756$ d) in year. We changed it to ($T_h = 0.753 - 2.07$ yr)	

	Line 9	« It is tricky » (or use difficult) instead of « It is hard » We changed it to “tricky”.	
	Line 18	Give a reference for the 72.6% We inserted “(Fig. 3)” here.	
P 16	Line 2	Something seems to be missing in « Thus, compared the cascade impactor and the impactor/cyclone measurement data, as shown in Fig. 5. » We simply changed the whole sentence to « Thus, the cascade impactor and the impactor/cyclone measurement data are compared in Fig. 5. »	
	Lines 15-16	Could you give some evidence of the presence of coarse particles found on the backup filter or explain how you detect them ? We did not find any evidence for this. We suspect the bouncing effect might occur because the cascade impactor with the similar experimental setup (Kinase et al., 2018) showed substantial amounts of soil dust and bioaerosol particles on the backup filter as found by the electron microscopy observation. But, yes, the observation locations are different, so there is no evidence that the bouncing occurred in the Fukushima city case, except the current statistical analysis. Thus, we changed the relevant sentence from « It appears that bouncing effect occurred » to « It appears that bouncing effect might occur ». Please also refered to our reply to your RC1 #3 – item (2) in the remained issue section of Conclusion.	
P17	Line 23	I do not understand what represents « The eight » in « The eight compositional correlation coefficients » We inserted the following phrase (the common 10 species shown in Fig. 7 minus the two, SiO ₂ and Al ₂ O ₃)	Introduce the eight compounds before
P19	Line 14	I suggest « concentrations in Tsushima and Tsukuba (MRI), » instead of « concentrations at Tsushima and the MRI, » We changed it to “Tsushima and Tsukuba”.	
P20	Line 8	« factor < 0.1 %. » instead of « factor for < 0.1 %.» We changed it accordingly.	
	Line 10	« From Fig. 9, the value of <i>b</i> for observations is close » instead of « From Fig. 9, <i>b</i> of observation is close » We changed it accordingly.	
	Line 11	The notion of « climatological deposition velocity » which is not conventional should be explained since it	

		<p>differs from what is consensually used as deposition velocity which refers to dry deposition only.</p> <p>We inserted the following statement just after the relevant sentence as follows :</p> <p>« Note that the concept of climatological deposition velocity differs from that of dry deposition velocity; the dry deposition velocity is defined as the ratio of the mass flux divided by the concentrations, but this climatological deposition velocity is the ratio of total (dry plus wet) deposition amounts divided by concentrations without the concept of mass flux. To account for the wet deposition flux, both in-cloud and below-cloud concentrations are needed, but such vertical measurement data is not available. ».</p>	
P 21	Line 5	<p>« overestimation of simulated airborne ¹³⁷Cs concentration from forests during summer» instead of « overestimation of simulated ¹³⁷Cs from forests in the summer»</p> <p>We changed it accordingly.</p>	
	Line 18	<p>I think you can be more categorical : which demonstrates the efficacy of wet deposition as compared with dry deposition and which plays ... » instead of « but it seems that wet deposition plays... »</p> <p>We changed the whole sentences as follows :</p> <p>« As mentioned above, the magnitude of the instant deposition velocity and our climatological deposition velocity are not directly comparable, but it demonstrates the efficacy of wet deposition as compared with dry deposition. Wet deposition plays an important role in the removal of resuspended ¹³⁷Cs-bearing particles from the air. »</p>	
	Figure 10	<p>It would be better to have the same magnitude for the Y-axis and X-axis scales. Currently, at first glance, one could interpret the figure as if deposition at both sites are equal. Please start from 10⁻¹ to 10⁶ for both axis.</p> <p>We changed it accordingly.</p>	
Page 22	Line 5	<p>Unless I am misunderstood, I dont agree with « The slope of the regression indicates that the ratio of deposition at Fukushima University to that at the MRI did not change significantly from the initial ratio during the eight years, ». This seems contradictory with what can be seen on fig. 10 (right plot) from where it can be conclude that from the first ratio to the last one there is about a factor of 20 based on the regression line</p> <p>We agree with you. There is a slight trend in the right panel of Fig. 1, Fukushima deposition drops somewhat</p>	

		<p>faster than Tusukuba deposition, with strong seasonal variations. We simply removed the relevant sentences and changed the whole sentences as follows :</p> <p>« There was a significant positive correlation between the deposition amounts of ^{137}Cs at the two sites, but the ratios varied substantially over time. The right panels of Fig. 10 indicates that the deposition ratios at the two sites were approximately 10, which is almost the same level as the initial amounts ($202 \cdot 10^3 \text{ Bq m}^{-2}$ at Fukushima University and $17.6 \cdot 10^3 \text{ Bq m}^{-2}$ at the MRI), with a variation of more than one order of magnitude and peaks in winter (especially January) that decreased slightly over time. »</p>	
	Line 7	<p>202200 should be written 202,200 or $202.2 \cdot 10^3$ the same for 23100</p> <p>We changed to the latter, to be consistent with the previous modification. Also, please note that the value 23100 was incorrect (the old undetermined value was used) and it was changed to $17.6 \cdot 10^3 \text{ Bq m}^{-2}$.</p>	
	Line 7	<p>« which is approximately 8-9 times higher at the Fukushima University than at the MRI » instead of « which is approximately 8-9 times »</p> <p>The relevant sentence was removed.</p>	
	Line 9	<p>Could you please add the average ^{137}Cs integrated concentration in soils with depth or at the topsoil layer, at both sites</p> <p>The relevant sentence was removed.</p>	
	Line 11	<p>« January peak is typical at Fukushima city » instead of « January peak is a feature of Fukushima city »</p> <p>We changed it accordingly.</p>	
	Line 16	<p>« the surface air activity concentration of ^{137}Cs has not fallen to the level prior to the » instead of «the surface air activity concentration of ^{137}Cs had not fallen to the level before the»</p> <p>We changed it accordingly.</p>	
	Line 23	<p>« and low from » instead of « and lows from »</p> <p>We changed it accordingly.</p>	
	Line 28	<p>What is « the Pacific high. » ?</p> <p>“pressure system” was inserted at the end.</p>	
P 23	Line 2	<p>« and Fukushima city is downwind of Tsushima, » is already mentioned line 1</p> <p>We removed the latter part.</p>	
	Line 7	<p>I do not see the interest to mention the case of aerosol with a such a high diameter since they are exceptionnally detected or correspond to very specific activities or at coastal sites. Without refering to such</p>	

		<p>extrem value, it could be more interesting to give an example of more « common » aerosol sizes like 20 or 30 μm even if again they remain much less abundant than 10 μm</p> <p>Thank you for your suggestion. We changed the sentence as follows :</p> <p>« the traveling distance of an aerosol with a diameter of several 10 μm is an order of 10 km. »</p>	
	Line 12	<p>Aside from the diameter which is sensitive to gravitational deposition, the efficient deposition onto the ground can be attributed to rain deposition. While dry deposition is almost permanent, this suggest that wet deposition is also more or less regular if not permanent (this cannot be seen based on the precipitation amount which is on a monthly basis)</p> <p>Yes, it is a good point. We changed the relevant sentence as follows :</p> <p>« Consequently, there was a significant enhancement in concentrations in the forests in summer but no enhancement in the downwind urban/rural areas, probably because the carrier aerosols were efficiently deposited onto the ground surface « by wet deposition in addition to dry deposition » before significant amounts of atmospheric ^{137}Cs reached the downwind areas.</p>	
P 24	Line 5	<p>« If the bouncing effect occurred only in the cascade impactor, » The place of this sentence seems strange. Does it already correspond to the second possible explanation ? Isf so the « 2) » should be placed before the sentence</p> <p>Yes, it was awkward. By considering the next comment (on Line 3 to 10) together, we reorganized the whole sentences as follows :</p> <p>(1) If the bouncing effect did not occur in either system, the major sources of radio-Cs in Fukushima city are probably related to combustion (a mass peak below 0.39 μm means that the number peak is approximately 100 nm). (2, 3) If the bouncing effect occurred only in the cascade impactor, (2) the size distributions of soil particles in Fukushima city are smaller, or radio-Cs in the soil exists more within finer particles; or (3) the coarse-mode fraction deposits to the ground surface faster than the fine-mode fraction, such that the proportion of Cs in PMf is larger in Fukushima city. (4) The bouncing effect occurs in both systems, and the origin of radio-Cs is coarse-mode soil particles.</p>	

	Line 3 to 10	The reading is not straightforward and the text would gain to be more intelligible. Please see our reply to your previous comment.	
	Line 13 - 30	The suggestion of an enhanced dust emission during snow period (even if it does have an effect given the short distance between the sampling location and the roads) would worth to be investigated before asserting. May be this idea could be developed in another paper. After line 13, I would suggest to shift to line 30 starting with « Unfortunately, analyses of the surface meteorological observational data for Fukushima City from the JMA, such as temperature, precipitation... » We agree with your suggestion. It is discussed in our follow-up paper and thus the whole sentences are removed here.	In order to keep with what has been measured and what can be interpreted with a relative high confidence. I would skip this snow section because it is too uncertain
P27	Line 15	Convert 456 d in year We changed to 1.25 yr. (and changed to 1.24 yr after recalculation according to #6 of RC3.)	
	Line 18	« changed approximately in 2015 » or « changed around 2015 » instead of « changed in approximately 2015 » We changed it to "around 2015".	
	Line 19-20	Convert 272 d and 825 d in year We changed them to 0.745 and 2.26 yr, respectively. (then to 0.754 and 2.07 after recalculation according to #6 of RC3.)	
	Line 23	In the conclusion, no need to repeat « This is consistent with the findings of Manaka et al. (2016). » We removed the whole sentence.	
P28	Lines 9-12	I would shift this item in the remaining unresolved issues if not deleted (see my previous comment about snow and mud) We shifted this to the #1 of the remaining issue. We are designing a field experiment somehow to prove this effect. Since we removed the « snow and mud » part according to your previous comment, this item is simplified as follows : « The deposition amounts of ¹³⁷Cs in January at Fukushima University were remarkably high compared to the concentrations of ¹³⁷Cs and the deposition amounts of ¹³⁷Cs at the MRI. The reason needs to be investigated in the future. »	