

We would like to thank reviewer#3 for the positive feedback on our paper that helped improve it.

1. *Title: This is a mere suggestion but I find the title rather long. Could the words "aerosol--bound", "during the months" and "power plant" be deleted to shorten it?*

We removed the words “during the months” and “power plant” to make it somewhat shorter. The title now reads: “Atmospheric removal times of the aerosol-bound radionuclides ¹³⁷Cs and ¹³¹I measured after the Fukushima Dai-ichi nuclear accident – a constraint for air quality and climate models.”

2. *Abstract, line 15: Could you mention that the removal time for ¹³¹I is longer due to the aerosol production from gaseous ¹³¹I? It is mentioned in the previous sentence but its influence on removal time not explicitly clarified.*

We included the following two sentences in the abstract: “The removal time of ¹³¹I is longer due to the aerosol production from gaseous ¹³¹I, thus the removal time for ¹³⁷Cs serves as a better estimate for aerosol lifetime. The removal time of ¹³¹I is of interest for semi-volatile species.”

3. *Page 12333, line 4: "key radionuclides of greatest concern", I find this a repetition. Leave the word "key" out.*

We left the word “key” out.

4. *Page 12333, line 20-21: "primarily traced the fate of sulfate aerosol... Once attached, ¹³⁷Cs shares the fate of these aerosols...". A repetition.*

These sentence are now elaborated to read: “..simultaneously measured sulfate aerosols (Kaneyasu et al., 2012). This was found by comparing the measured activity size distributions of ¹³⁷Cs to the mass size distribution of several aerosol components. An overlap of the two size distributions was found for non-sea-salt sulfate aerosols suggesting that ¹³⁷Cs primarily traced the fate of sulfate aerosols. These aerosols grow by coagulation ...”.

5. *Page 12334, line 1-4: I think there are two distinct matters to be discussed: lifetimes of particulate and semi---volatile species. The lifetimes of semi---volatile species can be of interest as such, and not only seen as "upper limits" for particulate species.*

We have tried to distinguish this by changing the sentences to read:

“¹³¹I is less suitable for tracing the fate of non-volatile AM aerosols but can still impose upper limits on the AM aerosol lifetime. In addition, ¹³¹I can be of interest also for considerations of the fate of semi-volatile species in the atmosphere.”

6. *Page 12334, line 16-20: do the models discuss species emitted from the surface only, or also stratospheric species?*

We have used references mainly for model studies of species emitted from the surface since for these species it should be possible to compare reported values to those for ¹³⁷Cs

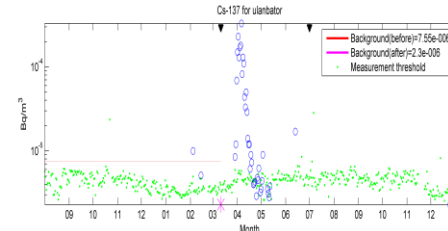
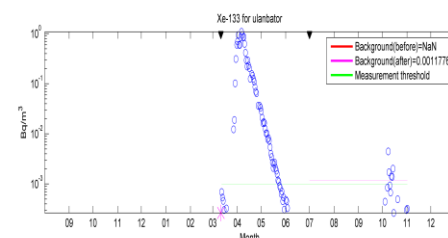
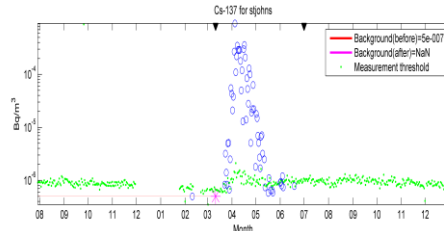
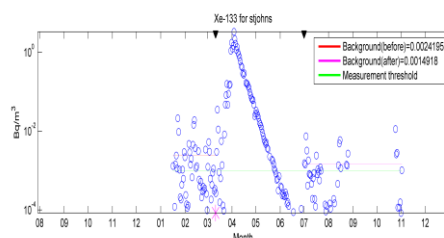
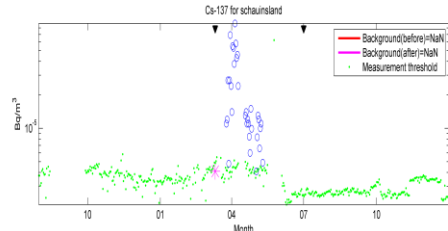
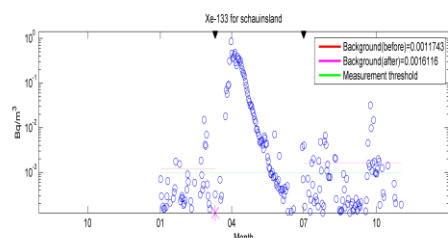
emitted from Fukushima. Species with an upper tropospheric/stratospheric origin typically have much longer residence times as shown by Giorgi and Chameides (1986) (their table 2). To clarify we have added to the sentence so it now reads “Models give global average residence times of AM aerosol in the atmosphere on the order of 3-7 days for species emitted mainly from the surface...”

7. Page 12334, last paragraph: I was wondering if it would be possible to show the data, perhaps as a 2D figure with stations organized by latitude against time? This would give the reader an idea of the variability and differences between stations.

It is not entirely clear how this should be visualized clearly. We could in principle make a contour plot of Cs-137 plotted against latitude and time. But for contouring, some interpolation would be needed and this would likely become very noisy. TABLE 1 provides the results after stations and we think this is the clearest way to show that there is variability between stations.

8. Page 12337, line 15 and wherever latitude bands are discussed: you could easily check the data of stations located in a given latitude band (say Vancouver, St. Johns, Schauinsland, Ulan Bator) to see if the assumption of well mixed latitude bands is valid. Wet deposition, strongly influenced by precipitation, varies largely in space, as you discuss in the beginning of the page 12340.

For the box model one station is valid for only one latitude band. They are not equally distributed. That means some of the latitude bands are very narrow, others are very broad. Also, testing this would only be possible for the mid-latitudes where the network is relatively dense. Below are the 133-Xe and 137-Cs measurements from Schauinsland (47.9N), St. Johns (47.6N) and Ulan-Bator (47.9N). Maximum concentrations are very similar for all three stations, and it also occurs at about the same time. The subsequent decay is also very similar, and this confirms that Xe-133 is reasonably well mixed in this latitude band. However, we do not have a clear criterion for deciding how large differences would have to be to reject the assumption that concentrations are well mixed.



Schauinsland.
Top: Xe-133, bottom: Cs-137

St. Johns
Top: Xe-133, bottom: Cs-137

Ulan-Bator
Top: Xe-133, bottom: Cs-137

9. *Page 12337, line 17: It is not immediately clear for me why the 'suitable time interval' was chosen to be 4 days. If decay---corrected concentrations are used, 4 days sounds like a very short time interval, compared with the obtained lifetimes of >10 days. Or perhaps I'm not understanding your point here. Please clarify.*

We have now added an important reference (Stohl et al 2012b) which describes the box model in more detail. This reference was not available at the time of the initial submission. The averaging time is a compromise between reducing scatter (for longer averaging times) and having more frequent data points (for shorter averaging times). It is to some extent subjective, but our results do not depend on the exact choice of this value.

10. *Page 12339, line 5-7: It does not sound correct to confirm the assumption of well mixed latitude bands (spatial variability) by looking at temporal variability.*

In the presence of horizontal transport, spatial and temporal variability are correlated. For instance, if there is a distinct plume at a given latitude and time that is not appropriately sampled by the network, this plume would subsequently spread to other locations and would cause concentration peaks at other locations. Similarly, if measurements are not representative of zonal background conditions but heavily influenced by the presence of a plume over a certain measurement stations, lower concentrations from other parts of the zonal band would spread and cause concentration declines later on. Thus, given the fast atmospheric transport processes, low temporal variability is only possible if our zonally averaged values are representative for their respective zonal band.

11. *Section 3, Results: Are the total atmospheric burdens estimated by other studies? How well do your results agree with them?*

We are not aware of other independent studies. However, the related study of Stohl et al 2012b which is now included in the reference list gives some more detail for ^{133}Xe and the box model estimations.

12. *Page 12339, line 18, explain τ_b (and other τ s as well).*

It is not clear to us what the reviewer means. If the exact τ_b is unclear or what is meant with "time scale" (e-folding, half-life etc.). For further clarifications we added: (b for "box model") after τ_b , and τ_a (a for "all"). The sentence now reads "By fitting an exponential model to the change of [^{137}Cs] with time (similar to Eq. 3) we find the removal time τ_b (b for "box model") of ^{137}Cs ". We also added before Eq. 3 in Sect. 2 "The time scale of the decrease is referred to as removal time throughout this paper and is based on the e-folding time scale. The removal time is calculated ..."

13. *Page 12339, line 23-24: It would be good to explain here why the τ_b is longer for ^{131}I than ^{137}Cs .*

We added "The τ_b is longer for ^{131}I than for ^{137}Cs due to gas-to-particle conversion of ^{131}I , as discussed further in Sect. 4."

14. Page 12339: last lines: Could you refer to the equations used from the section Data and methods?

No because the equations of that section don't show these calculations. However, the ratio-calculations are illustrated in Figure 2 which we included a reference to. We also re-wrote the sentences to clarify a bit better.

15. Section 4, Discussion. Perhaps "Discussion of uncertainties" or similar would be a more precise title.

Yes we changed the title.

16. Page 12341, line 12, "is obtained" ---> "are obtained"
"is" changed to "are".

17. page 12341, second paragraph: I have trouble following this paragraph. How do the emission times and plume age calculations by FLEXPART relate to this study? Could you provide more details?

We have re-written this paragraph and hope it is clearer now.

18. Page 12342, the first paragraph is very hard to follow. Line 1: "The longer removal time for 131I is expected..." and line 5---6: "The relatively large difference in ...suggests that the lifetime of gaseous 131I must be longer". If you expect a longer removal time for a gaseous 131I in the first place, then this result is trivial. Here, I would suggest clearly distinguishing between particulate and gaseous species and underline their different removal times.

We have re-written this paragraph extensively and hope it is clearer now.

19. Page 12343, line 26-27: "20% of emissions were deposited over land", page 12344, line 4: "Japan received 90% of the emissions over land". How do these two facts relate? Probably the fallout outside of Japan mostly fell over ocean, reducing the fraction over land, but you state that Japan received 90% of the fallout.

From Stohl et al. 2012a: 80% was deposited over the ocean, 20% over land; of the latter, 18% was deposited over Japan, 2% over other land surfaces. That means of the 20% that was deposited over land 90% (18 of those 20%) was over Japan.

20. Page 12345, line 25: what if the models are simply wrong? To my understanding their aerosol scavenging is fairly uncertain. The different sources of aerosols also have to be taken into account.

Our paper points into the direction the reviewer suggests, but we do not want to jump to that conclusion without 100% clear evidence. To obtain this evidence, the models would need to be re-run for the specific Fukushima case. Without further comparison to models simulating the Fukushima case, it is not so straightforward to say that the models are "right" or "wrong". This paper just indicates that there is something that is out of balance. Maybe the models are wrong, or maybe the difference will not be as large if they are run for this particular case study. We appreciate the reviewer's remark and agree that the

aerosol scavenging in models are rather uncertain. That is what this paper can help evaluate.

21. Page 12346, 2nd conclusion: *see previous comments on distinguishing between lifetimes of particulate and gaseous species.*

We have included more on the gas-to-particle exchange time scales and tried to emphasis the lifetime of the particulate species.

22. Figure 3&4: *The labels and units are nearly impossible to read, at least in my copy of the manuscript. Please increase their size.*

For the printed ACPD version the figures are very small, which is a typical problem with the landscape format of ACPD. For the final ACP version, which uses portrait format, this is typically not a problem and we will make sure that the figures are made bigger so that the labels and units are readable.