

Review of "Atmospheric removal times of the aerosol-bound radionuclides ^{137}Cs and ^{131}I during the months after the Fukushima Dai-ichi nuclear power plant accident - a constraint for air quality and climate models" by N. I. Kristiansen et al.

The authors utilise the unique world-wide observations of radionuclides released in the Fukushima nuclear accident to investigate atmospheric removal times of aerosol-bound species. They apply various methods to estimate the removal times and use these to validate their results. I find the manuscript useful and interesting and the presentation mostly clear. It could be further clarified in some parts, discussed in the more detailed comments.

Detailed comments:

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?

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

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

page 12333, line 20-21: "primarily traced the fate of sulfate aerosol... Once attached, ^{137}Cs shares the fate of these aerosols..." A repetition.

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.

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

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

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, Schausland, 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.

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.

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.

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

page 12339, line 18, explain τ_b (and other τ s as well).

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

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

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

page 12341, line 12, "is obtained" -> "are obtained"

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?

page 12342, the first paragraph is very hard to follow. line 1: "The longer removal time for ^{131}I is expected..." and line 5-6: "The relatively large difference in ...suggests that the lifetime of gaseous ^{131}I must be longer". If you expect a longer removal time for a gaseous ^{131}I 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.

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.

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.

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

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