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# Interactive comment on "Interpreting aerosol lifetimes using the GEOS-Chem model and constraints from radionuclide measurements" *by* B. Croft et al.

**B. Croft et al.**

croft@mathstat.dal.ca

The authors wish to thank referee A. Stohl for his constructive comments, which have led to improvements in our manuscript. Below is our point-by-point response to each referee comment (RC).

RC: I recommend the authors to read the paper by Giorgi and Chameides (1986), since it discusses the same issues, for instance by showing modeled aerosol lifetime as a function of altitude. It also reviews why reported aerosol lifetimes differ for radionuclide tracers originating at the surface and such originating from the stratosphere.

AC: We have added a discussion about how our work relates to that of Giorgi and Chameides (1986). Similar to the findings of Giorgi and Chameides (1986), who used an early generation climate model, our study with the GEOS-Chem model shows that mean aerosol lifetimes with respect to wet removal differ by an order of magnitude between the surface layer and about 8 km. Our study also found that mean lifetimes are strongly influenced by the source location, and additionally that e-folding times were relatively insensitive to source location after the radionuclides reached a quasi-steady-state concentration gradient. This discussion can be found in the third paragraph of the revised Section 1 and the second paragraph of Section 4.3. Thank you for pointing out this omission in our original manuscript.

RC: Pg 32393, lines 20-24: Kristiansen et al. (2012) indeed suggested that global models might underestimate aerosol lifetimes, but they also discussed the limitations and admitted that the mismatch may arise from the specific way the observations were made (far from the nuclear accident site) and that the aerosol removal must initially (shortly after release) have occurred much faster. This could be mentioned here, as their suggestion is consistent with the conclusion of the present paper. Kristiansen et al. (2012) did not investigate this quantitatively (although the effect was also seen in FLEXPART simulations), leaving that to a more comprehensive model intercomparison. It is nice to have this explored very systematically and quantitatively with the GEOS-Chem model!

AC: We agree that these suggested additions would make our discussion more complete. The revised text now includes a reference to these suggestions by Kristiansen et al. (2012) about how the mismatch between the measurement e-folding times and simulated aerosol mean lifetimes might arise due to differences in the aerosol removal rates between the boundary layer and the free troposphere and the locations for the measurements. This discussion is found in the second paragraph of Section 1.

RC: Pg 32400, lines 5-7: The authors explain the monotonic increase of instantaneous aerosol lifetime after about a month (Fig. 3) with ongoing mixing in the troposphere.

However, I think this is not the right explanation. Cassiani et al. (2013) report on the modeled fraction of black carbon (BC) residing in the stratosphere as a function of time after emission, both for a passive BC tracer and an aerosol BC tracer. In their Figure 7b, it can be seen that for the aerosol BC tracer, already one month after the emission, a substantial fraction of the aerosol mass remaining in the atmosphere, resides in the stratosphere (because the tropospheric fraction is so effectively removed; this is much less the case for the passive tracer). As in the present study the timescales are even much longer than a month, it is likely that the monotonous increase of lifetime is driven mainly by the increasing stratospheric fraction of the aerosol. Fig. 5 of the present paper suggests that, averaged over four months, the Cs-137 burden is concentrated in the stratosphere in GEOS-Chem as well.

AC: We agree that the text should have explicitly stated that mixing to the stratosphere drives the monotonous increase in the instantaneous lifetimes since this is a region of inefficient removal. We have examined the fraction of the global 137Cs burden in the stratosphere. This fraction is 28%, 42%, 53% and 64% for the months of May, June, July and August, respectively. The text is now revised to include this discussion about how the increasing stratospheric fraction of the global burden drives this increase in lifetime. This is stated in the revised text at the end of the sixth paragraph of Section 4.1 as a part of the discussion of Fig. 3 and also in the first paragraph of Section 4.3 related to Fig. 5.

RC: Pg 32403, lines 5-8: The fact that the e-folding lifetimes do not depend very much on the exact model setup (emission altitude, location and time) has an important implication that the authors do not mention. Radionuclide emissions (both their magnitude and their timing) are not known exactly. If derived lifetimes were sensitive to the uncertainties in the emissions, comparisons between measured (which are affected by the true emissions) and modeled (which are determined by estimated emissions) lifetimes would not be possible. Table 3, however, shows that derived lifetimes are very similar even for the extreme scenarios where all emissions occurred instantaneously on 11

March and at the completely "wrong" location. This means that the e-folding lifetimes derived from the measurements are very robust and indeed can provide a strong constraint on modeled aerosol lifetime, given that the model is sampled at exactly the same locations and times the measurements were taken (i.e., by making an apple-to-apple comparison).

AC: Thank you for pointing this out. We agree that this important implication of our results should be better highlighted in the manuscript. Table 3 does indeed show that e-folding times are relatively insensitive to emissions parameters such as the location, altitude, and time of the emissions. This suggests that measurement-based e-folding times are a very robust constraint on aerosol lifetimes. We have revised the text to include a discussion of this in Section 4.2 (third paragraph) and also in the abstract and conclusion section (fourth paragraph).

RC: Equation (3): The symbol $t_i$ not described. One can certainly guess the meaning but for the sake of clarity, a definition is needed.

AC: The presentation of the equations in Section 2 has been revised in response to the second referee and this symbol is now defined.

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