February 12, 2014

Dear Dr. Nenes,

We are pleased to submit this revised version of our manuscript entitled "Interpreting Aerosol Lifetimes Using the GEOS-Chem Model and Constraints from Radionuclide Measurements" by B. Croft, J. R. Pierce, and R. V. Martin for your consideration for publication in the journal Atmospheric Chemistry and Physics.

This revised manuscript addresses each of the concerns presented by the two referees. The point-by-point responses to each referee are provided as part of the on-line discussion and are also included below. In summary, the primary chanages made to our manuscript are as follows. In response to the second referee's first comment, we revised the formulation of the lifetime definitions in Section 2 and the related text. This revised presentation now more clearly identifies that our mean lifetime calculations are done with respect to the aerosol sink rates following the methodology suggested by the second referee. We also emphasize in response to the second referee's third point that differences between the spatial distributions of the aerosols and the precipitation control the absolute removal (Section 4.1, third paragraph).

The referee A. Stohl noted omissions from our original manuscript that are now corrected. We added a discussion about how our work relates to earlier work by Georgi and Chameides (1986) (Section 1, third paragraph and Section 4.3, second paragraph). We also added an explicit discussion about the fraction of the global burden contained in the stratosphere as the driver of the increase in instantaneous lifetimes shown in Fig. 3 (Section 4.1, paragraph 6 and Section 4.3, paragraph 1). This referee also pointed out one important implication of our results - since the derived e-folding times are insensitive to emissions parameters (location, altitude and time), the e-folding times derived from measurements are a very robust constrant on modeled aerosol lifetimes. This point was added to the abstract (end of second paragraph), to Section 4.2, third paragraph and in the Conclusion (fourth paragraph)

We look forward to your response to our revised manuscript.

Best regards,

Betty Croft

Author response to referee A. Stohl.

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 nonow 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 ti not described. One can certainly guess the

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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.

Author response to anonymous referee 2.

The authors wish to thank the second anonymous referee for the comments and suggestions, which have helped to improve the manuscript. Below is a point-by-point response to indicate how we have addressed each comment.

RC: The authors offered a clear discussion on various definitions of aerosol lifetime and presented corresponding values from GEOS-Chem calculation.However, in many cases including the one discussed here, the equilibrium between source and sink is not established. Thus, a tau bearing specific temporal and spatial scales would have to be derived from the sink term, or [-C/tau]. All the derivations listed in Sect. 2 are mostly approximations better for cases with unknown sinks. However, with a model this would not be a problem. The local sink would be easily calculated for all the corresponding grids, and thus the tau defined with different scales (whether mean or instant) would be obtained (assume dry deposition is negligible). In other words, once the precipitation distribution is known, all the arguments to explain the discrepancy of interest can be offered rather straightforwardly. For instance, the global mean tau commonly used in global models simply reflects an accumulation of all the grid-derived sinks.

AC: The equations presented in Section 2 have been re-formulated to provide a clearer description of our methodology. The approach suggested by this referee was included in our original methodology, but this is now more explicitly indicated with the revisions to all equations in Section 2. The revised text in Section 2 also more clearly indicates our use of the sink term. Additionally, we have modified the text related to Fig. 6 (last paragraph of Section 4.3) to explicitly indicate that the mean lifetimes presented are determined using the sink term. These simulated mean lifetimes are calculated with respect to wet removal.

RC: Page 32398, Line 15: Figure 2 seems showing the CTL integration results from time zero. My understanding is that the CTL should be a cold start run with zero initial concentration of 137Cs. If this is true, please indicate the time interval between the analysis time zero and the actual simulation time zero.

AC: Yes, you are correct. We have allowed a one-month spin up of the model after a cold start with near-zero initial 137Cs concentrations. Figure 2 is intended to only show the time since the onset of the radionuclide emissions. These start-up conditions are now indicated in the revised text of Section 3.1, first paragraph.

RC: Page 32399, Line 3-5, Differences betweenattributed to the efficient aerosol removalclose to the FD-NPP site: The absolute removal should be determined by how the precipitation and tracer distributions match each other. The total sink strength within a given domain divided by the domain-average mole fraction would derive domain-average tau. I assume in the discussion the authors actually was referring to a global domain. So, here are two factors, one is the removal strength, another is the averaged abundance of the tracer (note that

USFC is a pulse run that loads the total emissions once at all grids).

AC: Yes, the referee is correct that the discussion pertains to a global domain. We agree that the absolute removal is determined by how the precipitation and tracer distributions match each other. The revised text in Section 4.1 (third paragraph) now explicitly indicates that the global mean removal strength is different between the two simulations due to the different spatial distribution of the 137Cs over the first few weeks of the simulation relative to the precipitation distribution.

RC: Page 32401, Line 23, we chose days 20-80 after the onset of emissions for the fit this has not been explained clearly or I might miss some statements given in somewhere else. Is this a scale consistent with that of Kristiansen et al. analysis for some reason?

AC: This scale is used to provide a fair comparison of our simulated e-folding times with the measurement-based e-folding times presented by Kristiansen et al. 2012. We now inindicate this clearly in the revised text of Section 4.1 (last paragraph).

RC: Figure 4, the authors have barely touched the issue of model-observation comparison (Table 3 seems indicating a clear discrepancy between the two), only comparisons between different model runs were presented.

AC: We agree that the issue of model-observation comparison is an important one. Indeed radionuclide measurements provide one of the most useful constraints on the wet removal of aerosols. This is highly relevant for global models. A more detailed model-observation comparison than our Table 3 is the subject of an upcoming manuscript that is in preparation with colleagues. The focus of this current manuscript is to explain the reasons behind mean aerosol lifetimes and e-folding time differences and to quantify these differences. We have revised the text to provide an indication of future work and now include an explicit discussion of the value of a more detailed model-measurement comparison. This can be found in the Conclusion section (fourth and also final paragraphs) and in the abstract (second paragraph).