Atmospheric
Chemistry
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Discussions

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

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This paper investigates aerosol lifetimes in the GEOS-Chem model, based on different definitions of the lifetime (mean lifetime, e-folding lifetime) and at the example of aerosol-bound radionuclides released from the Fukushima accident. The paper nicely explains why e-folding lifetimes derived from radionuclide measurements far from the nuclear power plant as reported by Kristiansen et al. (2012), are much longer than the mean lifetimes reported for aerosol models. The difference arises because of the fast removal of aerosols close to the source (i.e., within the boundary layer), followed by much slower removal in the free troposphere where most of the long-range trans-

port occurs. While this explanation was already offered qualitatively by Kristiansen et al. (2012), it is quantitatively explored in this paper with a state-of-the-art model. It is shown that the GEOS-Chem model captures the relatively long "observed" e-folding lifetimes if the model is sampled at the locations of the measurement stations, even though the modeled radionuclide has a very short mean lifetime – a consequence of the rapid aerosol removal in the first few days after the emissions, which is not observed by the measurement network. The paper is very well written and interesting and I congratulate the authors for their very clear and systematic study, which is well worth to be published in ACP. I have a few comments, though, which the authors may wish to consider before submitting the final version of the paper.

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.

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!

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.

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).

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

References:

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