

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

Anonymous Referee #2

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This paper reports lifetimes of ^{137}Cs calculated based on various definitions using the model GEOS-Chem for a time period after the Fukushima Dai-ichi nuclear power plant accident (March 2011). The authors also compared their calculated values with estimates made using ^{137}Cs and ^{131}I surface concentrations measured at different sites by Kristiansen et al. and then explained the reasons behind the discrepancy between the two results. I found the topic rather interesting and believe the result should be informative to the aerosol community and beyond.

Comments.

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1. The authors offered a clear discussion on various definitions of aerosol lifetime and presented corresponding values from GEOS-Chem calculation. It could be more straightforward for them to clearly indicate the scale-dependency of related variables. As atmospheric chemists normally do, the mass budget in any given domain (from air parcel to global) could be expressed as:

$$dC/dt = E - C/\tau$$

Here C is the mole fraction, E emission rate, and τ the removal time scale (e-folding time for $E = 0$). Notably, all the terms in the equation are associate with given spatial and temporal scales. All the formulas used in the paper are more or less from the equilibrium solution of above equation. Estimation for global aerosol or chemistry models is usually derived in this way for a global domain. However, in many cases including the one discussed here, the equilibrium between source and sink is not established. Thus, a τ bearing specific temporal and spatial scales would have to be derived from the sink term, or $[-C/\tau]$. Therefore, the derivation of the e-folding time does not need the equilibrium assumption. 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. GEOS-Chem model likely uses the simple linear wet removal calculation (i.e., Giorgi and Chameides), where the rate of wet removal is simply a multiplication of a given constant (usually with different values for stratiform and convective precipitation) and precipitation rate. The local sink would be easily calculated for all the corresponding grids, and thus the τ 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 τ commonly used in global models simply reflects an accumulation of all the grid-derived sinks. In case of a domain having too many zero-removal grids, the global average would certainly differ substantially with the regional average over an area close to the source.

2. Page 32398, Line 15: Figure 2 seems showing the CTL integration results from

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time zero. My understanding is that the CTL should be a cold start run with zero initial concentration of ^{137}Cs . If this is true, please indicate the time interval between the analysis time zero and the actual simulation time zero.

3. Page 32399, Line 3-5, "Differences between...attributed to the efficient aerosol removal...close 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 τ . 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).

4. 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?

5. 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.

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