Atmos. Chem. Phys. Discuss., 14, C9191–C9195, 2014
www.atmos-chem-phys-discuss.net/14/C9191/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Explaining variance in black carbon's aging timescale" *by* L. Fierce et al.

L. Fierce et al.

lfierce2@illinois.edu

Received and published: 15 November 2014

We thank the reviewer for taking the time to review our paper and for the many insightful suggestions to improve it. We have responded to each comment individual and marked changes throughout the manuscript.

(2.1) The description of the 288 scenarios is unclear. From Table 2 it is difficult to figure out what these 288 exactly entail. I believe what is missing is a Table that identifies the model initial conditions (similar to Table 3 in Zaveri et al., 2008), which of these were held constant, and which were varied, and by how much. Also some motivation should be given for the rationale that led to the sampling of the parameter space and whether all of the combinations are physically reasonable.

We added Tables 1 and 2 to Section 2.1 of the revised manuscript, which

outlines the input parameters for the baseline scenario. We also revised Table 4 in Section 3 of the revised manuscript to clarify which parameters were varied in the sensitivity simulation. The scenarios were designed to represent the range of conditions observed in urban areas, and we have clarified this rationale in Section 3 (lines 334-335).

(2.2) After study of Figure 1, it seems that knowledge of the aging timescale alone is insufficient to constrain the process. Applying the definition of "fresh" and "aged" used in the paper can lead to circumstances where a good fraction of freshly emitted particles are emitted as "aged". Global models need to initialize this fraction correctly, or the applied aging timescale will not be meaningful. A question that arises from this is: if one simply tracks the mass of "fresh" and "aged" BC in two bins (with continuing emissions) and integrates over time with the τ_{aging} function shown in Figure 2, does one accurately predict the split between fresh and aged BC?

The aging timescale represents the effects of changes in per-particle size and composition, which is clarified in Section 2.3 of the revised manuscript, but does not resolve particle-level details. The continuity equations for the number concentration of fresh and aged particles are provided in Riemer et al. (2010), which can be used to estimate fresh and age particles as a function of time.

(2.3) The hygroscopicity parameter for OC seems very low. I presume OC = POA in the model? It suggest that a better description of the aerosol species and terminology is needed. OC could be interpreted as organic carbon, all organic aerosol, or primary organic aerosol. It seems that the hygroscopicity of the condensing material was varied. Was that done by changing κ_{SOA} over its entire dynamic range 0-0.3 or simply allowing for more sulfate? In other words how was κ_{cond} varied and how does it relate to the species in the table? Is composition information retained for each particle? If so, does it imply that multiple SOA/OA species are being explicitly tracked?

The model tracks POA and eight species of SOA for each particle. Everywhere in the paper, OC was changed to POA, and these changes have been marked throughout the document.

The value for κ_{cond} is the volume-weighted average of the hygroscopicity parameter of condensing species for each individual particle. That is, over each time step, we track the change in secondary aerosol species for individual particles and determine the hygroscopicity parameter of that material for each particle and take average across all particles. We clarify how κ_{cond} is computed in Section 5.1 of the revised manuscript (lines 608-609).

(2.4) Since this seems to be the first use of non-parameteric regression to untangle parameter sensitivity in complex atmospheric simulations it would be useful to see some added discussion on the topic. It is exciting to see that the 80-90% of the variance in aging time scale can be explained due to the sensitivity of a few input parameters. However, some additional discussion of the results seems to be warranted. First, how does this approach compare to the emulator approach used by Carslaw and colleagues (Carslaw et at., 2013, Nature), both conceptually and practically? Could the multi-parameter regression parameters for the optimal solution (black lines in Figure 6) be used to construct a plot similar to their Figure 2 with aging time scale replacing their Forcing estimate? On a related note, the regression inputs seem rather poorly defined. Perhaps a specific example is needed in an appendix or supplement to show how a distributed quantity "fresh BC size distribution" can be used in conjunction with Eq. (8) to define the Kernel function.

- *We modified Table 3 of the revised manuscript to make clear how input parameters are varied in the sensitivity simulations.*
- *We clarified the procedure for regression analysis in Section 4, which now includes a schematic of the procedure (Figure 6)*

- *We now show, by example, how particle-level variables are combined with environmental variables in the regression analysis in Section 4.3 (lines 568-590)*
- *The emulator approach was used by Carslaw et al. (2013) to evaluate uncertainty in an output variable, aerosol forcing, associated with model input parameters. The nonparametric regression analysis presented in our study is used to identify the independent variables, which need not be model input parameters, that best explain variance in an output variable, the aging timescale. Although Figure 2 of Carslaw et al. (2013) is similar to Figure 8 in the revised manuscript, the nonparametric regression cannot be used to segment explained variance into different components in this way, but rather evaluates the R^2 for combinations of independent variables. We feel that comparison with the emulator approach is beyond the scope of our paper.*

(2.5) The abstract and text might benefit from a bit more focus. In certain places the manuscript reads like a report. First we did this, then we did that (e.g. "After exploring many combinations of independent variables"). It seems to me that the results from the nonparameteric regression analysis is the main new finding. Those rankings should be better developed and the writing could be directed towards convincing the reader of the implication and utility of the identified parameters in Figure 6.

We thank the reviewer for this critique. We reorganized the manuscript and revised the language to bring out the main findings of study. Changes have been marked in the document. We also added Section 5.2, which demonstrates how these timescales can be used to evaluate the sensitivity of black carbon's aging timescale to key aging conditions.

(2.6) The (BC?) wet diameter is an ill-defined quantity. At minimum a list is needed

(perhaps in a supplement) of all the parameters that are initialized and varied for the scenarios or used in the regression analysis.

We modified the description of the regression analysis in Section 4 of the revised manuscript, clarifying how particle-level variables are included in the regression. We added Table 5 to Section 4, which lists the candidate variables that were investigated in the regression analysis. Further, we now explain in Section 3 that we include size distributions that vary temporally, and show the distribution in particle wet diameter in Figure 4.c. We also revised Table 4 to clarify how input parameters are varied for the sensitivity scenarios.

References

Carslaw, K., Lee, L., Reddington, C., Pringle, K., Rap, A., Forster, P., Mann, G., Spracklen, D., Woodhouse, M., Regayre, L., et al. (2013). Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature*, 503(7474):67–71.

Riemer, N., West, M., Zaveri, R., and Easter, R. (2010). Estimating black carbon aging time-scales with a particle-resolved aerosol model. *Journal of Aerosol Science*, 41(1):143–158.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 18703, 2014.