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

Anonymous Referee #2

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This manuscript presents numerical simulations using the zero-dimensional particle-resolved aerosol box model PartMC-MOSAIC to understand contributions to the variance in the black carbon aging timescale. A total of 288 scenarios were simulated from which the bulk and particle resolved time-scales were computed. The main new contribution is the use of nonparameteric regression to identify the main parameters that contribute to the variability in the aging timescale.

This manuscript is an important attempt to constrain the sources of uncertainty/variability in determining BC aging time scales. Clarification is needed to convince the reader of the utility of the approach, i.e. how the information can be used by others to interpret data, or to develop parameterizations, or how the method can be ported to other types of simulations, or how the results could help guide measurements. In general, more specifics are needed in the description of the setup of the

simulations and derivation of the results. Nonetheless, I believe that the manuscript should be published if the authors can address those points.

Comments:

(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.

(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 tau_aging function shown in Figure 2, does one accurately predict the split between fresh and aged BC?

(3) The hygroscopicity paramter 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 kappa_SOA over its entire dynamic range 0-0.3 or simply allowing for more sulfate? In other words how was kappa_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?

(4) Since this seems to be the first use of nonparameteric regression to untangle parameter sensitivity in complex atmospheric simulations it would be useful to see some

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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.

(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.

(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.

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