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

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We thank the reviewer for their critical assessment of our work. We have responded to each comment individually and have noted changes in the revised comment.

(1.1) What is the goal of this study? To obtain the knowledge for a better parameterization of BC aging processes?

The goal of this study is to identify which variables need to be included in parameterizations of BC aging processes. The timescales in this work were used to explore how independent variables affect the tendency for BC-containing particles to transition from CCN-inactive to CCN-active. Although this CCN-based aging criterion is directly related to the underlying physics that these aging parameterizations are meant to represent, this pa-

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rameterization would be difficult to implement in global models because it depends on the criterion supersaturation level and the size distribution of fresh particles. Using the underlying variables identified in this study, we have developed a different aging parameterization with a different aging criterion for use in global models, which will be introduced in a future paper. We have clarified the goal of the work in Section 1 (lines 112-117) and in Section 7 (lines 822-823) of the revised manuscript. .

(1.2) I want the authors to explain why it is useful and important to understand the key parameters of the variance and how the conclusions obtained in this study are useful for parameterizations and 3D simulations of BC aging processes. I feel that the conclusions obtained in this study are qualitative, not sufficiently quantitative to make a BC aging parameterization. If the goal of this study is to obtain the knowledge for a better parameterization, more quantitative information (e.g., formulation and/or coefficients for each key parameter) is desirable. At the least, the authors should explain how the conclusions will be used in the author's future study.

As discussed in our response to (1.1), we now clarify that the purpose of this study is to identify the governing parameters that need to be included in aging parameterizations through this nonparametric regression analysis. Based on these findings, we will soon introduce a simple expression for black carbon's aging timescale that can applied directly in global models. This new parameterization is beyond the scope of the present study, which focuses on identifying the underlying independent variables that best explain aging.

(1.3) In the sensitivity simulations in section 3, 288 scenarios are chosen. Is the number of scenarios sufficient? Two or three values are used as the range of each parameter in Table 2. Is this sufficient?

For each scenario, the aging conditions and the characteristics of BC-containing particles varied temporally over the course of the 24-hour simulation. We analyzed aging by BC over 143 time steps in each simulation, so all together we consider over 40,000 particle populations, where each population has a different distribution in particle microphysical properties and is exposed to different aging conditions.

For each set of regression variables, we performed the regression analysis multiple times, with randomly chosen testing and training sets. At each time step in all scenarios of the testing set, we computed the exact aging timescale and the expected aging timescale, given different combinations of independent variables. Each time we repeated the analysis, we found similar values for R^2, suggesting that conclusions do not depend not the particular sub-set of scenarios sampled as testing and training sets. We now clarify how we repeated the analysis to ensure that our conclusions are insensitive to the choice of scenarios in Section 4 of the revised manuscript (lines 393-398).

(1.4) Are the conclusions obtained in this study (e.g., the key parameters, the values of R^2) sensitive to the choice of scenarios? Are similar conclusions obtained when the simulation setting in Table 2 is changed (e.g., the range of the value in each parameter is changed)? I hope the conclusions are robust, but the sensitivity to the choice of scenarios should be checked by using some different selections of scenarios.

As discussed in (1.3), although we assign input parameters for 288 different scenarios, many other parameters vary throughout the simulation. We now clarify in Section 3 that the regression was based on aerosol properties that varied temporally (lines 339-341), and we added Figure 4, which shows that the simulations cover a wide range of conditions and aerosol properties.

Further, we construct the regression with a training set, and then evaluate the variance explained by that regression with a different set of simulations, the testing set. The purpose of doing this is to ensure that we are not over-fitting the data and that conclusions do not depend on the choice of testing scenarios. We included Figure 6 to clarify the procedure, and we added further discussion on how we ensured that the conclusions are insensitive to the choice of testing and training sets in Section 4 of the revised manuscript (lines 393-398).

(1.5) There are some other important parameters for the BC aging timescale in addition to the parameters listed in Table 2. For example, the size distribution (median diameter and sigma) and the mixing state of BC emission and their variations will be important, but they are not listed in Table 2 (though the importance of median diameter is discussed in section 4). In addition, the authors do not consider the emissions of BC-free particles and their variations. BC-free particles may be considered as background particles, but the response of microphysical processes may be different by the treatment (whether BC-free particles are given as emissions or background conditions). Please confirm that the conclusions do not change by considering these parameters.

Although the size distribution and mixing state of BC is the same for particles at emission, these properties vary over the course of the 24-hour simulations. This variation in particle properties is shown for the baseline scenario in Figure 1. We added Figure 4, which shows the distribution in aerosol number concentrations and microphysical properties of BC-containing particles across simulations. This figure demonstrates that aerosol number concentrations of all particles and BC-containing particles varies across simulations, as do the the size distribution and the composition of BC-containing particles. Even with the variation in chemical environment and variation in the characteristics of BC-containing particles that

is represented within the ensemble of simulations, we find that most of the variance is explained by our few key parameters. A small portion of variance (10-15%) remains unexplained, which is caused by variation in other independent variables; we clarify this point in Section 5 (lines 613-616).

(1.6) The conclusions in this study can be used for urban air masses only. This limitation should be clarified in Abstract.

We clarified that the conclusions are based on simulations of urban air masses in lines 341-342 of the revised manuscript.

(1.7) P18705, Lines 9-22: There are some other BC aging modeling studies. The authors describe global modeling studies in this paragraph, but there are some regional and box model studies that can calculate BC aging processes (Oshima et al., 2009; Zaveri et al., 2010; Matsui et al., 2013). In addition, some 3D models use a process-based representation of BC aging processes (Jacobson, 2002; Aquila et al., 2011; Matsui et al., 2013), not parameterized one. These studies are important for the modeling of BC aging processes and should be considered in Introduction.

We included reference to these studies in lines 47-48, lines 56-59, and lines 70-75 of the revised manuscript.

(1.8) P18709, Line 18: The simulation setting for the baseline scenario should be described before this paragraph.

We added Tables 1 and 2 to the revised manuscript, which provide input parameters for the baseline scenario.

(1.9) P18710, Line 2: Please give the value of sigma in addition to the mean diameter.

This parameter is now included in the caption of Table 6.

(1.10) P18710, Lines 3-4: I assume that S in this sentence means the supersaturation of ambient atmosphere, not critical supersaturation. Figure 1a shows that some BC particles can activate at $S = 1\%$ just after emissions. $S < 1\%$ should be $S < 0.1\%$?

We clarified that s is the environmental supersaturation in lines 244-245, and corrected the error in line 243.

(1.11) P18711, Line 22: Please show the results for other supersaturations (S=0.1%, S=1%). Are there differences in the aging timescale between supersaturations? For example, the importance of condensation and coagulation processes may be different between supersaturations.

We appreciate this suggestion. We now evaluate how the sensitivity of the aging timescale to aging conditions depends on the environmental super-saturation level in Section 5.2 of the revised manuscript (lines 681-744, including Figures 9 and 10). This analysis reveals that the relative importance of condensation versus coagulation indeed depends on the supersaturation level, as well as the size distribution of fresh BC-containing particles.

The temporal evolution of the overall aging timescale and the condensation aging timescale are shown at various supersaturation levels Riemer et al. (2010), which we now point out in lines 314-316 of the revised manuscript.

(1.12) Sections 2.2 and 2.3: Discussions in these sections are made without the de-scription of simulation setups and model mechanisms. Please describe them before section 2.2 (at least for the baseline simulation). The information of initial condition, emission, the treatment of dilution, SOA scheme, the formation rate of sulfate, nitrate,

and organic aerosol, and number concentrations may be useful for readers. Discussions on temporal variations of mass concentrations of each species and number concentrations in the baseline simulation may also be useful.

We added Tables 1 and 2, which describe all input parameters for the baseline scenario. We also included addition discussion on the SOA scheme (lines 162-163) and treatment of dilution (lines 151-154).

(1.13) Figure 3: Please add a panel for number concentrations. Are number concentrations within realistic range?

Probability density distributions for the overall aerosol number concentration and the number concentration of BC-containing particles are now shown in Figures 4.a and 4.b, respectively.

(1.14) P18713, Lines 16-17, "but also reveals the functional form". Is this advantage used in this study?

We removed this sentence from the revised manuscript.

(1.15) P18715, Lines 5-6: Is it possible to calculate the aging timescale for each data j from the PartMC-MOSAIC simulations? The method to estimate the bulk aging timescale is described in section 2, but is there any description of aging timescale for each data?

We thank the reviewer for this critique. The expected aging timescales computed in this study are, indeed, based on a regression analysis of aging by individual particles, and we have revised the description of the kernel regression in Section 4 to clarify our approach. By computing the expected

probability that an individual particle will age, we are able to include as regression variables per-particle characteristics, which vary between particles at a specific time, as well as environmental conditions, which are the same for all particle at a specific time. We added a schematic detailing our procedure (Figure 6) to show how aging timescales for BC populations are estimated from particle-level aging data.

(1.16) P18716, Line 20: Are the aging timescales calculated by Equations (4) and (12) the same?

Yes, they are the same.

(1.17) P18717, Lines 20-21: just before aging?

We changed the phrase "just before aging" to "when aging timescales are evaluated" or similar everywhere in the paper. The changes have been marked in the revised manuscript.

(1.18) P18718, Lines 1-20: What does the comparison in Figure 6a (black, green, and blue lines) mean? Please explain the usefulness to identify the difference among $N_{CCN,large}$, N_{CCN}, and N_{large}?

The tendency for a particle to age by coagulation depends on the number concentration of CCN-active particles that large enough to be likely coagulation partners small BC-containing particles, where we found "large enough" to be particles with wet diameter $D > 100$ nm. At low supersaturation thresholds, all particles that are CCN-active also have $D > 100$ nm, so knowledge of N_{CCN} is sufficient to explain variance in τ_{coag} at low s. At high supersaturation thresholds, all particles with $D > 100$ nm are also

CCN-active, so N_{large} is sufficient to explain variance in τ_{coag} at high s. Only $N_{CCN,large}$, which refers to the number concentration of particles that are both large ($D > 100$ nm) and CCN-active, is sufficient to explain variance in τ_{coag} at all environmental supersaturation levels. The difference between $N_{CCN,large}$, N_{CCN}, and N_{large} is explained in Section 5 (lines 643-646), which clarifies that, of the three variables, only $N_{CCN,large}$ is sufficient to explain variance for the full range of supersaturation thresholds.

(1.19) P18718, Lines 21-22: In the condensation aging timescale

We corrected the sentence in lines 652-653 of the revised manuscript.

(1.20) P18719, Lines 25-27: How can we use the results for the parameterizations and 3D simulations? Is there no possibility that other parameters could be important in the simulations with other settings of scenarios? (as shown in the major comments (4) and (5))

Although we have shown that most variance is explained by only the few key parameters, 10-15% of variance remains unexplained. This unexplained variance is caused by variance in other parameters. We add further clarification on this point in Section 5 of the revised manuscript (lines 613-616).

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(1.21) P18720, Line 9: Please describe the difference between volume and mass.

Volume-based and mass-based measures of secondary aerosol formation rates yield slightly different results due to differences in density of condensing species. We found R^2 is highest when using volume-based quantities.

(1.22) P18720, Line 13: Is it really possible to apply to any size distribution though there are no sensitivity simulations for size distributions in section 3?

As explained in (1.5), the size-distribution of BC-containing particles varies over time, which is shown by variation in the geometric mean diameter of BC-containing particles in Figure 4.

(1.23) P18720, Line 17: Are these new sensitivity simulations? Or estimated from the 288 scenarios? In the latter case, how can the authors determine the lognormal distributions of 30 nm and 60 nm?

We expanded this analysis in Section 5.2 (lines 681-744). Figure 10 shows how size-resolved aging timescales are estimated for a specific set of aging conditions. Figure 10 shows how the size-resolved aging timescale can be applied to specific size distributions to estimate size-resolved aging rates. Section 5.2 also explains how the overall aging timescale can be estimated from integration across the entire size distribution of rest particles, yielding the bulk timescales in Table 6.

(1.24) Table 1: OC is primary organics?

OC has been changed to POA everywhere in the paper, and these changes have been marked the the revised manuscript.

(1.25) Table 2: I cannot clearly understand the description of "varied precursors for NH4NO3, (NH4)2SO4, both NH4NO3 and (NH4)2SO4, SOA". Please give the values and their ranges. Please clarify the treatment of emissions for other gas-phase species such as SO2, NOx, and VOCs? (G1, G2, G3, ...); (E1, E2, E3);

We modified Table 4 of the revised manuscript to show variation in specific input parameters, and we refer to Tables 1 and 2 for parameters that are held constant.

(1.26) The legend is difficult to understand. Please revise the description. Caption: "(a) condensation and (b) coagulation" should be "(a) coagulation and (b) condensation"

We modified the labeling and corrected the caption for Figure 8 of the revised manuscript.

References

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.

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