Review of “Explaining variance in black carbon’s aging timescale” by L. Fierce, N. Riemer, and T. Bond

In this paper, black carbon (BC) aging processes were simulated with the particle-resolved box model PartMC-MOSAIC. The authors simulated the evolution of gases and aerosols for nearly 300 scenarios of urban air masses and estimated BC aging timescales in terms of CCN activation for condensation and coagulation processes. The authors identified the key parameters driving the aging timescale through a regression analysis and found that most of the variance in the aging timescale was explained by a few microphysical and chemical parameters: two variables, the size distribution of fresh BC and the number concentration of large, CCN-active particles, were important for the coagulation aging timescale, and three variables, the size distribution of fresh BC, the flux of secondary aerosol defined as the volume condensation rate of semi-volatile substances per particle surface area density, and the effective hygroscopicity parameter of secondary aerosol, were needed to explain the variance for the condensation aging timescale.

This paper is generally written well and may be suitable for the publication of this journal if the authors clarify and consider the following points. As some of them may affect the main conclusions of this paper, the authors should give an answer and explanation to them carefully.

Major comments:

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

(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.

(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?

(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.

(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.

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

Other comments:

(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.

(8) P18709, Line 18

The simulation setting for the baseline scenario should be described before this paragraph.

(9) P18710, Line 2

Please give the value of sigma in addition to the mean diameter.

(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%?

(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.

(12) Sections 2.2 and 2.3

Discussions in these sections are made without the description 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.

(13) Figure 3

Please add a panel for number concentrations. Are number concentrations within realistic range?

(14) P18713, Lines 16-17, “but also reveals the functional form”

Is this advantage used in this study?

(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?

(16) P18716, Line 20

Are the aging timescales calculated by Equations (4) and (12) the same?

(17) P18717, Lines 20-21

just before the aging?

(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 Nccn,large, Nccn, and Nlarge?

(19) P18718, Lines 21-22

In the condensation aging timescale

(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))

(21) P18720, Line 9

Please describe the difference between volume and mass.

(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?

(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?

(24) Table 1

OC is primary organics?

(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?

(26) Figure 6

The legend is difficult to understand. Please revise the description.

Caption: “(a) condensation and (b) coagulation” should be “(a) coagulation and (b) condensation”

References:

Aquila, V., J. Hendricks, A. Lauer, N. Riemer, H. Vogel, D. Baumgardner, A. Minikin, A. Petzold, J. P. Schwarz, J. R. Spackman, et al. (2011), MADE-in: A new aerosol microphysics submodel for global simulation of insoluble particles and their mixing state, Geosci. Model Dev., 4, 325–355.

Jacobson, M. Z. (2002), Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, J. Geophys. Res., 107(D19), 4410, doi:10.1029/2001JD001376.

Matsui, H., M. Koike, Y. Kondo, N. Moteki, J. D. Fast, and R. A. Zaveri (2013), Development and validation of a black carbon mixing state resolved three-dimensional model: Aging processes and radiative impact, J. Geophys. Res. Atmos., 118, 2304–2326, doi:10.1029/2012JD018446.

Oshima, N., M. Koike, Y. Zhang, Y. Kondo, N. Moteki, N. Takegawa, and Y. Miyazaki (2009), Aging of black carbon in outflow from anthropogenic sources using a mixing state resolved model: Model development and evaluation, J. Geophys. Res., 114, D06210, doi:10.1029/2008JD010680.

Zaveri, R. A., J. C. Barnard, R. C. Easter, N. Riemer, and M. West (2010), Particle-resolved simulation of aerosol size, composition, mixing state, and the associated optical and cloud condensation nuclei activation properties in an evolving urban plume, J. Geophys. Res., 115, D17210, doi:10.1029/2009JD013616.