Response to reviewer #2

Interactive comment on "Development of a parameterization of black carbon aging for use in general circulation models" by N. Oshima and M. Koike

We thank the reviewer for positive and valuable comments on our paper. We have incorporated them into the revised version. Major revisions made to the manuscript are described first, followed by our point-by-point responses to the comments raised by the reviewer.

Major revisions:

First, we briefly describe the summary of major revisions made to the revised manuscript.

(1) We have estimated the time scale of BC aging due to coagulation for various atmospheric conditions and have estimated the errors included in Eq. (7) in the original manuscript due to neglecting the coagulation effects on BC aging. Details have been described in Appendix A (new appendix) in the revised manuscript.

(2) We have proposed other formulations of parameterization of BC aging including both condensation and coagulation effects. Details have been described in Sect. 6 (new section) in the revised manuscript.

(3) We have emphasized the usefulness of our parameterization using the time scale conversion (τ_{BC}) for many GCMs in the revised manuscript.

General comment:

I agree with the general comments made by Referee 1, but would like to add that while many global models indeed treat mixing state and the aging process explicitly, several global models still exist that use the time scale conversion framework. For those models a parameterization as presented in this paper would be useful, and I think that this paper can be a valuable contribution. Having this said, I do share the concern by Referee 1 regarding the limitations of the parameterization. Given that MADRID-BC does not include coagulation, I assume that it is not possibly at this point to use equation (9) as underlying aging model. However, as already requested by Referee 1, the authors should at least include an error estimate to address this limitation.

Reply:

As described in "Reply 1.1 to General comments 1 for Referee #1" in detail, we estimated the time scale of BC aging due to coagulation and estimated the error included in Eq. (7) in the original manuscript over source and outflow regions due to neglecting the coagulation effects for the BC aging. As a result, the conversion rates shown in Eq. (7) in the original manuscript can give the smaller conversion rates by approximately 50% and 10% due to neglecting the coagulation effects over the polluted source regions and outside of those regions, respectively. These descriptions have been given in detail in Appendix A in the revised manuscript.

To take into account the coagulation effects in our parameterization, we have proposed formulations of the conversion rate from hydrophobic BC to hydrophilic BC that including both condensation and coagulation effects in Sect. 6 in the revised manuscript (please see "Reply 1.2 to General comments 1 for Referee #1").

Specific comments:

p. 1265, l. 3: Description of freshly emitted BC: From single particle analysis (e.g.Toner et al., 2006) it has become evident that even freshly emitted particles usually contain coatings of OC (from lubricating oils), and hence it is misleading to say that they are "bare"

Reply:

Following the reviewer's suggestion, we have removed the word "bare" and have modified the descriptions in the revised manuscript.

Methodology: For the calculation of the critical supersaturation for each 2D grid cell, what diameter is assumed? The mean diameter of the bin?

Reply:

In the 2-D aerosol representation, diameter and BC mass fraction of particles can vary between the low and high edges of the size and BC mass fraction bins, respectively, and the transfer of particle number and mass due to their growth and shrinkage can be simulated simultaneously in both directions (i.e., particle diameter and BC mass fraction) on the basis of the moving center approach (Jacobson, 1997). To clarify this point, we have added this statement in the revised manuscript.

p. 1264, l. 24: Change sentence to: BC has been recognized as one of the most important aerosol type

Reply:

We have revised the manuscript as suggested.

p. 1266, l. 29: treat

Reply:

We have revised the manuscript as suggested.

p.1267, l. 23: add "primary" organic matter for clarification.

Reply:

We would keep the original expression, because MADRID-BC can calculate the SOA formation using the full equilibrium approach, as treated in the original MADRID model (Zhang et al., 2004), although the SOA formation based on the dynamic approach is not included in MADRID-BC. Because the dynamic approach can calculate the condensation/evaporation processes and the time evolution of the entire BC mixing state more accurate than the full equilibrium approach, we used the dynamic approach in this study.

p. 1269, l. 13: supersaturation threshold of 0.1%: The environmental supersaturation in clouds varies widely depending on the underlying aerosol population and the cooling rate, and as such there is no "typical value". Given that there is a strong dependence of the aging time scale on the supersaturation threshold, what is the rationale for choosing this value?

Reply:

As the reviewer pointed out, it is difficult to obviously say that 0.1% is the typical value, because the ambient supersaturation in clouds varies depending on the cloud dynamics (e.g., updraft velocities) and on the aerosol microphysics. The reason for choosing 0.1% is that this value is reported as the median value of the measurements under ambient conditions in the literature (Seinfeld and Pandis, 2006). Although we used 0.1% for the baseline value in the main body of the manuscript, this does not restrict the use of our parameterization, because we showed the method to use other values (between 0.05% and 1%) in Appendix A in the original manuscript (Appendix B in the revised manuscript). To clarify this point, we have modified statements in the revised manuscript.

p. 1269, l. 27: change to “were conducted for one hour”

Reply:

We have revised the manuscript as suggested.

p. 1269, l. 29: The term “increase rates” is awkward. I suggest changing it to “growth rates”.

Reply:

We have revised the manuscript as suggested.

p. 1272, l. 13: “tend to have a hydrophilic nature (i.e. CCN activity)”: This is a strange sentence. I believe the authors mean that for larger particles the Kelvin effect is smaller, and hence they activate at lower environmental supersaturations.

Reply:

Our statement “particles with larger diameters generally tend to have a hydrophilic nature” in the original manuscript might give misleading information. We have modified the statement and have provided the following statement in the revised manuscript. "… and dry particles with larger diameters generally tend to become CCN active according to Köhler theory (i.e., the maximum of the equilibrium saturation ratio is smaller for the larger particles, and hence they can activate at lower environmental supersaturations)."

p. 1272, l. 20: The argumentation regarding the impact of σ is unclear. Please rephrase.

Reply:

Following the reviewer's suggestion, the argumentation regarding the impact of σ has been rewritten clearly. Please see "Reply to Specific comments 13 for Referee #1" for details.

p. 1274, l. 6: treat

Reply:

We have revised the manuscript as suggested.

p. 1277, l. 4: Heterogeneous hydrolysis impact: How is the heterogeneous hydrolysis treated in the model (i.e. what value for the uptake coefficient is used)?

Reply:

The median values of the uptake coefficient (γ) recommended by Jacob (2000) are used in this study (i.e., γ_{HO_2} = 0.2, γ_{NO_2} = 10^{-4}, γ_{NO_3} = 10^{-3}, and $\gamma_{N_2O_5}$ = 0.1). To clarify this point, we have added this statement in the revised manuscript.

Appendix A: The dependence of the parameterization on supersaturation threshold is interesting and I would consider this section integral to this paper. I suggest adding this section to the main body of the manuscript instead of having an appendix.

Reply:

We would keep these descriptions in the appendix (Appendix B in the revised manuscript). As the reviewer pointed out, the dependence of the parameterization on supersaturation threshold is interesting, however if these descriptions are given in the main body of the manuscript, the manuscript will be longer and it will be difficult to understand the key points of this paper.

p. 1280, equation A1: Please supply additional steps how to arrive at this equation.

Reply:

Following the reviewer's suggestion, we have added an approach to obtain Eq. (A1) in the original manuscript (Eq. (B1) in the revised manuscript) in the latter half of the appendix in the revised manuscript.

References

Jacob, D. J.: Heterogeneous chemistry and tropospheric ozone, Atmos. Environ., 34, 2131–2159, 2000.

Jacobson, M. Z.: Development and application of a new air pollution modeling system, part II, Aerosol module structure and design, Atmos. Environ., 31, 131– 144, doi:10.1016/1352-2310(96)00202-6, 1997.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd ed., John Wiley & Sons: New York, USA, 2006.

Zhang, Y., Pun, B., Vijayaraghavan, K., Wu, S. Y., Seigneur, C., Pandis, S. N., Jacobson, M. Z., Nenes, A., and Seinfeld, J. H.: Development and application of the model of aerosol dynamics, reaction, ionization, and dissolution (MADRID), J. Geophy. Res., 109, D01202, doi:10.1029/2003JD003501, 2004.