Response to reviewer #3

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 essential and valuable comments on our paper. We have incorporated these valuable comments 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:

This manuscript presents a new parameterization for time scale τ_{BC} for the aging of black carbon (BC), i.e., the conversion from the hydrophobic to the hydrophilic state. The new parameterization is meant to be used in general circulation models (GCMs) that treat size distribution of hydrophobic and hydrophilic BC separately using lognormal modes. I do not recommend this manuscript for publication in *Atmospheric*

Chemistry and Physics.

I am not convinced of the usefulness of this parameterization for τ_{BC}. τ_{BC} had been used in GCMs that considered only aerosol mass concentrations (i.e., size distributions not treated) as a simple time scale parameter to determine the rate at which BC becomes hydrophilic and thus can be removed from the atmosphere by wet scavenging. Today, as noted by reviewers #1 and #2, a growing number of GCMs explicitly treat aerosol microphysics. The new parameterization for calculating τ_{BC} in this manuscript requires that the GCM treats aerosol microphysics (condensation) using lognormal size distributions. In such GCMs, the aerosol microphysics schemes automatically provide enough information (BC size distribution and coating of other chemical components) that the cloud condensation nuclei (CCN) properties can be derived such that the amount of BC that can undergo wet scavenging can be determined directly without using τ_{BC}. Thus, I don't see how τ_{BC} is needed anymore.

If I am misunderstanding how this parameterization for τ_{BC} can be used in GCMs, then the parameterization needs to be improved. The parameter A in Equation (6) should be re-formulated in terms of mono-dispersed aerosol size distribution instead of lognormal size distribution. Instead of

$$\tau_{BC} = \frac{A(D_m, \sigma)}{V_{BC}}$$

the parameterization should be

$$\tau_{BC} = \frac{A(D_p)}{V_{BC}}$$

where D_p is the particle diameter. This way, the parameterization can be more generally applied to GCMs that use sectional or lognormal modes for modeling aerosol size distributions. Also this way, A can be interpolated with only D_p as an input instead of D_m and σ. In this parameterization, the overall τ_{BC} can be then be determined by integrating $\tau_{BC}(D_p)$ with the modeled BC size distribution.

Reply:

The comments raised by the reviewer are excellent to the point. As the reviewer pointed out, τ_{BC} (the conversion time scale from hydrophobic BC to hydrophilic BC) had been used in GCMs, in which only mass concentrations of aerosol species are predicted with the prescribed aerosol size distributions. Models that explicitly treat aerosol microphysics can calculate the hydrophilic information (e.g., coatings on the BC particles) without using τ_{BC}, and therefore these models do not need our parameterization.

Our parameterization is applicable to models that do not explicitly treat aerosol microphysics (i.e., do not need to calculate size distributions of aerosols in models). Our parameterization requires the separate treatment of hydrophobic BC and hydrophilic BC modes and the calculation of condensation of bulk amounts of aerosols (at least the total amount of sulfate) in models. Although the $A(D_m, \sigma)$ values (Eq. (6) in the original manuscript) and the $k_{coag.,j}$ values (Eq. (12) in the revised manuscript) depend on the D_m and σ values of the lognormal size distributions of aerosols, the D_m and σ values can be chosen arbitrarily on the basis of the size distributions that users want to assume to represent in models, even if the models do not explicitly calculate size distributions of aerosols. The reviewer may have misunderstood this point. To clarify this point, we have modified statements in the revised text.

As also pointed out by Referees #1 and #2, the number of GCMs that explicitly treat aerosol microphysics is growing rapidly. However there are still many GCMs that treat the BC aging processes as a simple time scale conversion from hydrophobic to hydrophilic states using the bulk method, in which only mass concentrations of aerosol species are predicted with the prescribed aerosol size distributions. We believe that our parameterization is useful for those GCMs. To clarify this point, we have modified statements in the revised text. (Please also see "Reply 1.3 to General comments 1 for Referee #1" for details).

The method to represent the parameterization using mono-dispersed aerosol size distribution given by the reviewer is interesting; however, it is difficult to adopt it in this study, because the information of the entire size distribution of aerosols is needed for our estimates of τ_{BC}. Instead, we have proposed formulations of parameterization of BC aging 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").