Response to reviewer #1

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 a very thorough review and constructive comments. We have revised our paper by taking these valuable comments into account. 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 comments:

1. I am not sure how useful the parameterization by Oshima and Koike is. My main concern is the fact that they neglect coagulation. Can the authors give an estimate of the error committed? Coagulation is an important ageing process close to sources (as the authors mention in their manuscript), and what happens in emission regions largely determine the lifetime of BC. The authors need to perform additional tests to prove their

assumptions. I encourage the authors to perform such tests or, if already performed, add them to the manuscript. In my opinion the manuscript is not acceptable for publication without an estimate of the error committed.

I am not sure why the authors did not develop a parameterization based on Eq. 9 instead of Eq. 7. If they were able to show that $A(D_m, \sigma)$ derived from Eq. 7 and Eq. 9 are similar, and the δ term of Eq. 9 is effectively negligible, they could safely assume that neglecting coagulation is reasonable, and this would prove that their parameterization can indeed be used in GCMs.

Furthermore, the number of GCMs that includes an explicit microphysics is growing rapidly (see next general comment), and these GCMs also include coagulation, so why choosing such a simple parameterization as the one that Oshima and Koike here present? This work is interesting, but, instead of aiming to providing a parameterization for GCMs, the authors should consider their results as a upper limit estimate of the ageing time τ_{BC}. These values would work as bases for comparison for GCMs that already calculate BC microphysics and mixing state. If this change of focus is accepted, however, I would consider this manuscript more suitable for ACP than GMD.

Reply:

1.1: The coagulation effects and the error estimates

We estimated the time scale of BC aging due to coagulation for various atmospheric conditions from polluted to clean air. We calculated the instantaneous conversion rate of mass concentration of BC from hydrophobic to hydrophilic states due to coagulation between various combinations of lognormal size distributions of hydrophobic BC and BC-free particles. As a result, we obtained the following relationship:

$$\tau_{BC}^{coag.} = \frac{1}{k_{coag.}\,[BC_{free}]}$$ (Eq. (A3) in the revised manuscript)

where $\tau_{BC}^{coag.}$ is the e-folding time of the conversion due to the coagulation between the size distributions of hydrophobic BC and BC-free particles, $k_{coag.}$ (μg^{-1} m^3 s^{-1}) is a constant value representing the dependences on the lognormal size distributions (D_m and σ) of the hydrophobic BC and BC-free particles, and $[BC_{free}]$ is mass concentrations of BC-free particles. The $k_{coag.}$ values have been summarized in Table 3 in the revised manuscript.

Using Eq. (A3), we estimated the time scale of the BC aging due to coagulation for polluted source regions and outside of those regions. As a result, the $\tau_{BC}^{coag.}$ values were estimated to be 21-107 hours and 17-34 days for the polluted source regions and outside of those regions, respectively, on the basis of the observed values (e.g., mass concentrations and size distributions of aerosols).

We estimated the error included in Eq. (7) in the original manuscript due to neglecting the coagulation effects for the BC aging. For this purpose, we calculated the τ_{BC} values (i.e., the conversion time from hydrophobic BC to hydrophilic BC due to condensation) using the same methods described in Sect. 2 for the source (Beijing) region and the outflow (the ocean around Japan) region on the basis of the observed values. These τ_{BC} values were compared with the corresponding $\tau_{BC}^{coag.}$ values. As a result, the conversion rates shown in Eq. (7) in the original manuscript give smaller values by approximately 50% and 10% for the source regions and outside of those regions, respectively.

These descriptions have been given in detail in Appendix A in the revised manuscript.

1.2: Parameterization of BC aging including the coagulation effects

We proposed two types of formulations of parameterization of BC aging including both condensation and coagulation effects in Sect. 6 in the revised manuscript (i.e., Eq. (11) and Eq. (12) in the revised manuscript). The first representation of the parameterization, which used the constant conversion rate due to coagulation, is expressed as

$$\frac{1}{\tau_{BC}} = \frac{V_{BC}}{A(D_m,\sigma)} + \delta \qquad \text{(Eq. (11) in the revised manuscript)}$$

where the first term of the right-hand side is the same as the right-hand side of Eq. (7) in the original manuscript and δ is the coagulation term (i.e., an e-folding conversion time of 20 days, which approximately represents the time scale due to coagulation for outside of the source regions), and Eq. (11) has to be used concurrently with assuming some fraction of hydrophilic BC for initially emitted BC particles in models (e.g., 80% hydrophobic and 20% hydrophilic), because the time scale due to coagulation over the source regions is significantly shorter than outside of those regions (please see “Reply 1.1 to General comments 1”).

Alternative representation of the parameterization, which includes dependences on the number concentration of aerosols, is expressed as

$$\frac{1}{\tau_{BC}} = \frac{V_{BC}}{A(D_m, \sigma)} + \sum_j k_{coag.,j}\,[M_j] \qquad \text{(Eq. (12) in the revised manuscript)}$$

where the first term of the right-hand side is the same as the right-hand side of Eq. (7) in the original manuscript, $k_{coag.j}$ is the constant coefficient for coagulation between the lognormal size distributions (D_m and σ) of hydrophobic BC and soluble aerosol mode j (e.g., several soluble BC-free modes), and $[M_j]$ is mass concentration of the soluble aerosol mode j. The $k_{coag.j}$ values have been summarized in Table 3 in the revised manuscript.

We would keep our main focus on the condensation effects and treat the coagulation effects additionally in the revised manuscript, because the representations of the condensation effects are more accurate than those of the coagulation effects. Our parameterization due to condensation (e.g., Eq. (7) in the original manuscript) was derived using the results of detailed calculations by the MADRID-BC model, however the coagulation effects (e.g., Eq. (A3) and the second term of the right-hand side of Eq. (12) in the revised manuscript) were simply derived from the instantaneous coagulation rates between the size distributions of hydrophobic BC and BC-free particles, in spite of the non-linearity of coagulation. However, the use of Eq. (11) or Eq. (12) can represent variations of the BC aging rate and they are more accurate representations than the constant conversion rates widely used in previous studies.

These descriptions have been given in detail in Sect. 6 in the revised manuscript.

1.3: Usefulness of the time scale conversion τ_{BC}

As the reviewer pointed out, the number of GCMs that explicitly treat aerosol microphysics with representation of aerosol size distribution by modal or sectional methods 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. For example, among a total of 15 models included in the Aerosol Model Intercomparison Initiative (AeroCom) Phase

II direct aerosol effect experiment (Myhre et al., 2012), more than half of models use the time scale conversion for the BC aging processes (e.g., HadGEM2 (Bellouin et al., 2011) and NCAR-CAM3.5 (Lamarque et al., 2012)) or ignore the BC aging processes (e.g., GISS-ModelE (Koch et al., 2007) and SPRINTARS (Takemura et al., 2005)). Most models participating in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lee et al., 2012) also treat the BC aging processes with the time scale conversion. For climate calculations with long time integration (decades to centuries) and/or with fine spatial resolution by coupled atmosphere-ocean global climate models, a simple parameterization that can capture the essentials of the BC aging processes is still desirable due to limited computational resources, instead of the explicit treating of aerosol microphysics in models which are computationally expensive. To clarify this point, we have modified statements in Sect. 1 (introduction) in the revised manuscript.

Our parameterization is applicable to models that separately treat hydrophobic BC and hydrophilic BC modes and do not explicitly treat aerosol microphysics. We believe that our parameterization is useful for those kinds of GCMs.

2. The introduction is insufficient. It basically states that there is no GCM that includes an explicit simulation of BC ageing, but this is not true. A fair number of GCMs already use parameterization of ageing processes and simulate BC ageing, including also particle coagulation as, for instance, ECHAM-HAM (Stier et al. 2005), ModelE-MATRIX (Bauer et al 2008), WACCM-CARMA (Bardeen et al., 2010), EMAC/MADE-in (Aquila et al., 2011). These are all models that provide a detail representation of ageing processes through condensation and coagulation, and are used for global climate simulations.

Reply:

Following the reviewer's suggestion, we have improved the introduction of our paper. We have included above descriptions raised by the reviewer in the revised manuscript.

3. How do you think your parameterization would change for other kind of insoluble species, as dust or insoluble organics? Would the change be only in the mass of condensed material or would also Eq. [7] change?

Reply:

Our parameterization can be used for other kind of insoluble species (e.g., insoluble organics), as long as their size distributions are within ranges of D_m of 100-200 and σ of 1.5-1.8. In this case, the formula of Eq. (7) in the original manuscript does not change, and the values in Eq. (4) in the original manuscript change accordingly, namely mass concentration of condensed materials onto the insoluble mode (i.e., $M_{cond.}^{BCphob}$) changes and [BC_{phob}] has to be replaced by mass concentration of the insoluble mode. Our parameterization cannot treat dust (coarse) particles, because their sizes are out of the above range. To clarify this point, we have modified statements in the revised text.

4. Have the authors done any study on how the inclusion of SOA would change their results?

Reply:

Our answer to the question is “No”. However, as written in the “Reply to Specific comment 15”, if values of hygroscopic parameters (κ) (e.g., Petters and Kreidenweis, 2007) for water-soluble organic and inorganic species are similar, the CCN activities of those particles are similar. To clarify this point, we have modified a statement in the revised text.

Specific comments:

1. p1264L3: “that” looks like referring to GCMs, you should reformulate this sentence

Reply:

This is our grammatical mistake. We have corrected the sentence in the revised manuscript.

2. p1265L3: …and ARE therefore…

Reply:

We have revised the manuscript as suggested.

3. p1266L4: I would not refer to 2001 as “recently”, it was 11 years ago!

Reply:

Following the reviewer’s suggestion, we have changed the word from “recently” to “previously” in the revised manuscript.

4. p1266L11: Are you sure that the Riemer et al (2004) parameterization is applicable only to polluted regions over land? They used it in that environment, but I am not aware that their parameterization contains limitations that forbid using it on different regions, given the correct boundary conditions. As a matter of fact, Aquila et al. (2011) implemented the MADEsoot model by Riemer et al (2004) into a global climate model.

Reply:

The reviewer may have misunderstood this point. The MADEsoot model itself can be applied to any regions with the correct initial and boundary conditions. However their parameterization, namely the τ_{BC} values (2 hours above 250 m and 8 hours below) during daytime and Eq. (7) given by Riemer et al. (2004) during nighttime, will be only applicable for a polluted region over land where they applied the MADEsoot model. To clarify this point, we have modified a statement in the revised text.

5. p1267L19: you mean “particle diameter ranges”, correct?

Reply:

Our answer to the question is “Yes”, and we have revised the manuscript as suggested.

6. p1268L14: Could you write which Köhler equation you used? You used the one for particles including an insoluble fraction (Seinfeld and Pandis, 2006, section 17.2.5),

correct?

Reply:

Following the reviewer’s suggestion, we have added the Köhler equation, which includes an insoluble fraction, in Eq. (4) and Eq. (5) in the revised manuscript. Our answer to the question is “Yes”.

7. p1268L28: MADRID-BC calculated the mass concentration of condensed material also onto BC-free particles, not only on BC-containing particles, am I right?

Reply:

Our answer to the question is “Yes”. To clarify this point, we have added a statement “and BC-free particles” in the revised text.

8. p1269L5: it would be useful to specify where the conditions of Table 1 are met in the real world.

Reply:

Following the reviewer’s suggestion, we have added a statement regarding the corresponding conditions in the revised manuscript.

9. p1270L10: M_{BCphob} is the mass of condensed material? That is totally counterintuitive! How about $M_{cond.}^{BCphob}$? The use of the partial derivative ∂ implies that it is an infinitesimal increment, but the authors speak about increment per hour. Wouldn’t a discrete increment $\mathit{\Delta}$ be more appropriate?

Reply:

We agree with the reviewer’s statements. The use of $\Delta[M_{cond.}^{BCphob}]/\Delta t$ is more appropriate. We have revised the manuscript as suggested.

10. p1270L11: What do you mean with “initially hydrophobic”? At the beginning of each timestep or of each hour?

Reply:

The calculations were conducted for one hour for each initial condition, and we obtained the increase rate of the total mass concentration of condensed materials onto initially hydrophobic BC particles for each hour simulation. To clarify this point, we have modified a statement in the revised text.

11. p1271L23: “when the BC particles experience ageing processes” as opposed to when they do not? What do you mean?

Reply:

Our sentence in the original manuscript might lead to misunderstanding. We have modified the statement and have provided the following statement in the revised manuscript. “Oshima et al. (2009b) showed that the time evolution of the mass fraction of hydrophilic BC particles with respect to total BC particles due to aging processes depends on the size distribution of initially hydrophobic BC particles.”

12. p1272L14: Is the curvature effect always more important than the fact that particles with larger diameter need more condensed material to be completely coated? Is Eq. 6 also valid for extremely large diameters?

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).” The Köhler equation includes the curvature effect, and therefore Eq. (6) in the original manuscript is also valid for larger particles.

13. p1272L21: why should the left-hand side of the distribution be more important than the right-hand side? When you increase σ, you also increase the number of particles with larger diameter, which, as the authors write, are more prone to become CCN active.

Reply:

As the reviewer pointed out, the greater σ value distributes BC particles over a wider size range and increases the number of smaller and larger particles. Because τ_{BC} is defined as the time required for the hydrophobic BC mass concentration to decay to 1/*e* of its initial value in our calculations and BC-containing particles with greater diameters generally preferentially become CCN active, the particles greater than the minimum CCN particle diameter (D_{min}), which is roughly located where integral of the size distribution from 0 to D_{min} equals to 1/*e* of that of the entire size range and is also located over the left-hand side of the lognormal size distribution (i.e., range with diameters smaller than D_m), become CCN active at τ_{BC} for a given supersaturation due to the BC aging. The greater σ value generally distributes BC particles over a wider size range (please see Fig. 1 of this document) and shifts D_{min} to the smaller diameter (i.e., increase in the number of smaller size BC particles over the left-hand side of the lognormal distribution), and therefore the greater σ case needs longer τ_{BC} for activation of all particles greater than D_{min} for a given supersaturation, at least within a range of D_m of 100-200 nm. To clarify this point, we have added the above descriptions in the revised manuscript.

14. p1273L14: Do you mean that you assume that pure BC particles are hydrophobic, or that BC particles are the only hydrophobic particles in your model?

Reply:

We only treat hydrophobic BC as hydrophobic particles, and POM is not assumed to be hydrophobic in this study. To clarify this point, we have modified a statement in the revised text.

15. p1273L20: Do indeed water-soluble organic and inorganic species play a similar role in the Köhler equation? What do you exactly mean?

Reply:

If values of hygroscopic parameters (κ) (e.g., Petters and Kreidenweis, 2007) for water-soluble organic and inorganic species are similar, the CCN activities of those particles are similar. To clarify this point, we have modified a statement in the revised text.

16. p1279L6: I think you need “whereas” instead of “although”

Reply:

We have revised the manuscript as suggested.

17. p1279L16: what does “ does not take into account the hydrophobic or hydrophilic natures of BC-containing particles themselves” mean?

Reply:

It means that there is not clear threshold value to distinguish between hydrophobic and hydrophilic states of BC particles in models. To clarify this point, we have modified statements in the revised manuscript.

18. p1279L22: “we do not need to...”

Reply:

We have revised the manuscript as suggested.

19. Table 2: You should specify that A is the coefficient of equation 6, because you refer to this table much earlier than where you introduce A. Instead of referring to this table for the size distribution, I would add a figure where you plot the size distribution considered (something like Fig. 1 of this document), with a legend that specifies the parameters used.

Reply:

Following the reviewer’s suggestion, we have added mass size distributions of bare BC particles used in this study in Fig.1 in the revised manuscript (please see Fig. 1 of this document). In addition, we have specified that *A* is the coefficient of Eq. (6) in the original manuscript in the caption of Table 2 in the revised manuscript.

20. Figure 1: the figure is too small. Why not using 3 columns?

Reply:

The too small figure was unexpected for us. Following the reviewer's suggestion, we enlarge the figures in the revised manuscript.

References

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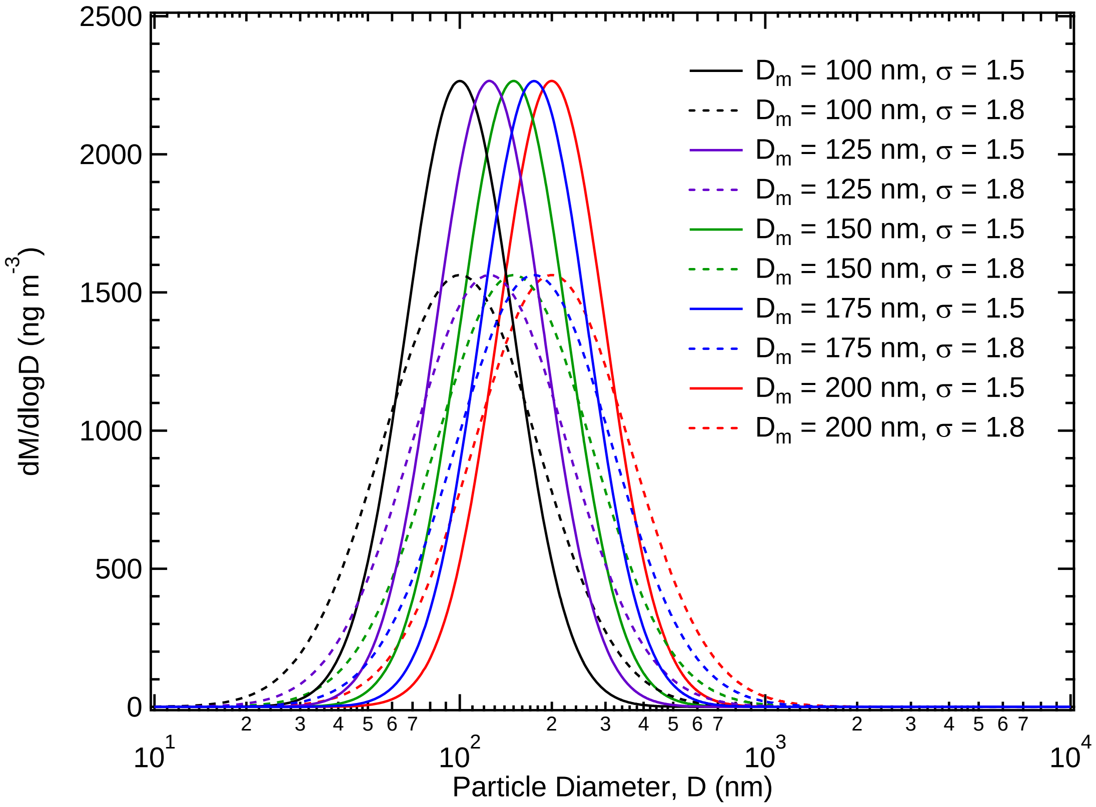

Fig. 1. Mass size distributions of bare BC particles used in this study. We have shown this figure in Fig. 1 in the revised manuscript.