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

Anonymous Referee #1
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The authors present a simple parameterization for the ageing of black carbon (BC) from hydrophobic to hydrophilic. The authors derive their parameterization from many case studies performed with the box-model MADRID-BC, and arrive to a simple formula that describes the characteristic ageing time of BC $\tau_{BC}$ as inversely proportional to the mass condensation rate $V_{BC}$. They fit their model results to derive the coefficient of proportionality between $\tau_{BC}$ and $V_{BC}$. In this parameterization only condensation is considered as an ageing process, and coagulation, cloud processing and photochemistry are neglected.

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 $\delta$ 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 $\tau_{BC}$. 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.

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.

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?

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

Specific comments

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

2. p1265L3: . . . and ARE therefore . . .

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

4. p1266L11: Are you sure that the Riemer et al. (2004) parameterization is appli-
cable 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.

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

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?

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

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

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

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

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

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?

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

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

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

16. p1279L6: I think you need “whereas” instead of “although”
17. p1279L16: what does “does not take into account the hydrophobic or hydrophilic natures of BC-containing particles themselves” mean?

18. p1279L22: “we do not need to...”

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.

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

References


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Fig. 1. Example of what the authors could use to show the BC size distributions assumed in their work.