Interactive comment on “Microphysics-based black carbon aging in a global CTM: constraints from HIPPO observations and implications for global black carbon budget” by C. He et al.

Anonymous Referee #1

Received and published: 4 January 2016

This paper presents a new parameterization of black carbon aging for the use in the global 3D chemical transport model GEOS-Chem. The parameterization accounts for aging due to three processes: coagulation, condensation of secondary aerosol material, and photochemical aging. It consists of a first-order aging time scale that is applied to the BC mass concentration and depends on local environmental conditions, quantifying the transformation of “hydrophobic” BC to “hydrophilic” BC. The parameterization is evaluated using BC measurements from the HIPPO campaign. The authors show that using the new parameterization improves the modeling results considerably, but discrepancies between modeled and simulated values still exist.
This paper represents an important step towards a more process-oriented treatment of BC aging in global models. The analysis is of great interest to the community and the paper fits well into the scope of ACP. A weakness of this paper is the mathematical description of the parameterization and the underlying assumptions. I recommend the paper for publication after the following questions and comments are addressed:

Major comments:

1. In the interest of the reproducibility of the results, the description of the model and the aging scheme need to be improved. In particular, please address the following:

1.1. Before starting with section 2.2.1, please describe briefly the aerosol scheme used in GEOS-Chem more clearly – what are the prognostic variables? From the text that follows it sounds like this is a bulk scheme, and only mass concentrations of different aerosol species are tracked, but it would be useful to mention this early on.

1.2 Equation (3): What expression for the correction factor \( f \) is used? What is the value for \( \alpha \)?

1.3 Equation (4): Please write out the limits for the integration and summation. How is the integral numerically evaluated?

1.4 page 32786, line 23: Please rephrase the explanation for subscript \( i \) (it clearly doesn’t represent various pre-existing particles. Do you mean sub-populations or classes?

1.5 Equation (4) and (5): Please distinguish between the number concentration and the number concentration distribution function \( (dN/dD_p) \). In equation (4) it should be the latter. Usually a lower case \( n \) is used as variable for this.

1.6 Equation (5): \( N_p \) should also have an index \( i \) (for the different classes). What are the values for the particles density?

1.7 Equation (7): Please write out the limits of this summation. Also, this is assum-
ing that the different secondary aerosol species have the same hygroscopicity. Please state this assumption. SOA should actually be less hygroscopic than for example sulfuric acid and ammonium nitrate.

1.8 After shifting the hydrophobic mass of BC into the hydrophilic category, how do you treat the associated secondary aerosol material? Do you track it separately from the mass of sulfate/nitrate/ammonium/SOA that is not mixed with BC. (This comes back to comment 1.1 – what are the prognostic variables?

1.9 Equation (9): This is the coagulation kernel that applies to the coagulation of two particles with radii $R_{BCPO}$ and $R_X$. In the section for condensation you described that you are assuming a log-normal size distribution. How do you reconcile this with assuming specific radii in equation (9)?

1.10: Equation (10): Please write out what the limits are for the summation and rephrase the explanation for index $i$.

1.10 What is the additional computational burden associated with the new parameterization compared to using the fixed aging time scale?

2. Setup of sensitivity studies: A potentially important sensitivity run that is missing is one that investigates the sensitivity to the assumption that 80

3. Given that there are several papers in the literature that use the aging time scale to present their results, it would be helpful to include a figure that shows a map of actual aging time scales and/or a pdf (similar to Figure 4, but instead of BC concentrations, show $\tau$).

4. For the figures that show ratios (such as Figure 10), I suggest to try a blue-to-red color scale, where blue represents values <1 and red represents values > 1.

5. page 32797, line 26: The authors attribute the remaining model-observation discrepancy to BC emissions, wet scavenging and meteorological fields. Is it possible to be more specific which of these three factors contributes most?
6. Page 32801, line 1: How do you reconcile the GMD from the HIPPO observations with the range used for the model simulations?

Minor comments:

Page 32780, Line 2: The parameterization also accounts for photochemical aging, please add this to the abstract.

Page 32781, line 12: Even at emission BC is actually frequently mixed with OC (see for example Willis et al., Atmos. Chem. Phys. Discuss., 15, 33555-33582, 2015, and references therein.)

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 32779, 2015.