Response to Reviewers' comments on "Microphysics-based black carbon aging in a global CTM: constraints from HIPPO observations and implications for global black carbon budget" by He et al.

Referee #2

"The scientific results and the improved model delivered in this study are of interest to the whole aerosol modeling community. I believe the paper merits publication in ACP after addressing the following minor concerns."

Response: We thank the reviewer for kind words. We have provided itemized responses to the following comments.

Minor Comments:

1. *"Considering that BC largely affects aerosol optical depth in the atmosphere, it is interesting to know if such an improvement in BC treatment helps to reduce uncertainty of the simulated AOD in GEOS-Chem compared to satellite measurements."*

Response: We thank the reviewer for this constructive comment. We agree that BC strongly absorbs solar radiation, which could affect AOD (for all aerosols). It is very important to evaluate model simulations of BC from different perspectives, including BC atmospheric concentration and AOD. Previous studies showed that GEOS-Chem overestimates AOD over East Asia (van Donkelaar et al., 2008) but slightly underestimates AOD over North America (Liu et al., 2004). Thus, the smaller global BC column burden resulting from the microphysical BC aging scheme developed in this study could reduce the model bias over East Asia but increase it over North America. The present study focuses on the assessment of simulated BC concentrations instead of AOD, because AOD is largely affected by other aerosols in addition to BC. It would be very useful for future study to compare the simulated AOD with satellite measurements. We have included these discussions in Page 32, Lines 803-805 as follows:
"We note that it is also very important to evaluate BC simulations from other perspectives in addition to atmospheric concentration, such as aerosol optical depth, a subject requiring further investigation."

2. *"Particle density, geometric mean diameter, and geometric standard deviation are given when converting aerosol mass to number concentration. Sensitivity experiments in this study have shown the potential large influence of the aerosol representation on the BC sedimentation and wet removal. Can a two-moment/three-moment aerosol scheme [Li et al., 2008] be a better solution for this problem? Some discussions along this line would be valuable."*

Response: Points are well taken. The two-/three-moment aerosol scheme could be a potential improvement of the aging scheme. We have included additional discussions in Page 30, Lines 748-752 as follows:
"The two-/three-moment aerosol scheme (e.g., Li et al., 2008), which predicts aerosol size distribution from simulated aerosol mass, number, and/or surface area depending on atmospheric conditions, could be a potential improvement to represent and understand BC aging, interaction with cloud, and deposition compared with the microphysical scheme developed in this study."

3. *"The chemical oxidation scheme [Poschl, 2001] used in this study was developed long time ago. It would be better to know the uncertainty with this scheme compared to the recent laboratory and in-situ measurements [Khalizov et al., 2013; Qiu et al., 2012; Wang et al., 2013]."*

Response: We thank the reviewer for this comment and references. We would like to point out that Khalizov et al. (2013), Qiu et al. (2012), and Wang et al. (2013) indicated that the chemical oxidation of volatile organic compounds (VOCs) occurring in the air produces secondary organic aerosols that can condense onto and/or coagulate with BC particles resulting in strong BC aging. These processes are very important but they are related to condensation- and coagulation-induced aging, which differ from the chemical oxidation aging discussed in this study that represents the heterogeneous oxidation process occurring directly on BC particle surface. To comply with this comment, we have incorporated additional discussions and references in Page 15, Lines 326-333 as follows:
"Recent experimental studies also confirmed that BC can be aged through heterogeneous chemical oxidation by O_3 (Decesari et al., 2002; Zuberi et al., 2005) and NO_2 (Khalizov et al., 2010), which results in the formation of soluble organic compounds on BC particle surface. However, none of these recent studies have given explicit quantitative parameterizations for BC chemical aging, which can be applied in modeling studies. Moreover, experimental results presented in these studies vary substantially, suggesting that the BC chemical aging process could involve large uncertainty. For this reason, the parameterization scheme developed by Pöschl et al. (2001) should be used with caution."

References

Li, G., Y. Wang, and R. Zhang, Implementation of a two-moment bulk microphysics scheme to the WRF model to investigate aerosol-cloud interaction, J. Geophys. Res.,113, D15211 (2008)

A. Khalizov, Y. Lin, C. Qiu, S. Guo, D. Collins, and R. Zhang, Role of Oh-Initiated Oxidation of Isoprene in Aging of Combustion Soot, Environmental Science & Technology, 47 (2013), 2254-63.

C. Qiu, A. F. Khalizov, and R. Y. Zhang, Soot Aging from Oh-Initiated Oxidation of Toluene, Environmental Science & Technology, 46 (2012), 9464-72.

Y. Wang, A. Khalizov, M. Levy, R. Zhang, New Directions: Light Absorbing Aerosols and Their Atmospheric Impacts, Atmos. Environ., 81, 713-715 (2013).

Liu, Y., Park, R. J., Jacob, D. J., Li, Q. B., Kilaru, V., and Sarnat, J. A.: Mapping annual mean ground-level PM2.5 concentrations using Multiangle Imaging Spectroradiometer aerosol optical thickness over the contiguous United States, J. Geophys. Res.-Atmos., 109, D22206, doi:10.1029/2004jd005025, 2004.

van Donkelaar, A., Martin, R. V., Leaitch, W. R., Macdonald, A. M., Walker, T. W., Streets, D. G., Zhang, Q., Dunlea, E. J., Jimenez, J. L., Dibb, J. E., Huey, L. G., Weber, R., and Andreae, M. O.: Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada, Atmos. Chem. Phys., 8, 2999-3014, doi:10.5194/acp-8-2999-2008, 2008.