Atmos. Chem. Phys. Discuss., 12, 28993–29023, 2012
www.atmos-chem-phys-discuss.net/12/28993/2012/
doi:10.5194/acpd-12-28993-2012

Atmospheric Chemistry and Physics Discussions

Impact of aging mechanism on model simulated carbonaceous aerosols

Y. Huang[1], **S. Wu**[2], **M. K. Dubey**[3], **and N. H. F. French**[4]

[1]Department of Geological and Mining Engineering and Sciences, Michigan Technological University, Houghton, MI 49931, USA
[2]Atmospheric Science Program, Department of Geological and Mining Engineering and Sciences, Department of Civil and Environmental Engineering, Michigan Technological University, Houghton, MI 49931, USA
[3]Earth System Observations, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[4]Michigan Tech Research Institute, Michigan Technological University, Ann Arbor, MI 48105, USA

Received: 24 October 2012 – Accepted: 5 November 2012 – Published: 9 November 2012

Correspondence to: S. Wu (slwu@mtu.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Carbonaceous aerosols that include organic carbon and black carbon, have significant implications for both climate and air quality. In the current global climate or chemical transport models, a simplified hydrophobic to hydrophilic conversion lifetime for carbonaceous aerosol (τ) is generally assumed, which is usually around 1 day. Based on results from recent chamber studies, we implemented a new detailed aging mechanism for carbonaceous aerosols in a chemical transport model (GEOS-Chem) where τ is affected by local conditions such as O_3 concentration and humidity. The simulated τ exhibits large spatial and temporal variation with the global average calculated to be 4.3 days. The longest τ (up to 40 days for the Amazon forests) are found in the tropical areas, reflecting the low ozone concentration and high humidity there. The conversion lifetime generally decreases with altitude due to increases in ozone concentration and decreases in water vapor concentration. The updated aging mechanism has significant implications for model simulations of carbonaceous aerosols and improves the comparison to observations of carbonaceous aerosols. The strongest effects are found for the tropical regions and upper troposphere where the model simulated concentrations of black carbon and organic carbon increase by up to 0.16 $\mu g\,C\,m^{-3}$ and 0.67 $\mu g\,C\,m^{-3}$, respectively. This updated aging mechanism also leads to increases in model calculated global burden of black carbon and organic carbon by 31 % and 17 %, respectively. In addition, sensitivity studies show that the estimated continental outflow of carbonaceous aerosols would significantly increase with the updated aging mechanism.

1 Introduction

Carbonaceous aerosols, including both elemental carbon (EC) (also called black carbon, BC) and organic carbon (OC) (Chung and Seinfeld, 2002; Park et al., 2003) play important roles in affecting the climate directly and indirectly (Balkanski et al., 2010; Haywood and Boucher, 2000; Lohmann et al., 2000; Schulz et al., 2006). They also

ACPD
12, 28993–29023, 2012

make significant contributions to particulate matter (PM), an important air pollutant in surface air (US Environmental Protection Agency, EPA, 2007).

Major sources of carbonaceous aerosols include fossil fuel combustion, biomass and biofuel burning as well as photochemical oxidation of volatile organic compounds (VOCs). Fresh emitted or newly formed carbonaceous aerosols generally have low solubility (Kondo et al., 2007). However, various physical and chemical processes in the atmosphere can convert the hydrophobic carbonaceous aerosols to hydrophilic ones (Croft et al., 2005; Kanakidou et al., 2005; Liu et al., 2011). Globally it is estimated that wet scavenging provides the dominant sink for carbonaceous aerosols in the atmosphere (Chung and Seinfeld, 2002; Park et al., 2003). Since hydrophobic aerosols are more susceptible to wet scavenging, the conversion of carbonaceous aerosols from hydrophobic to hydrophilic ones can significantly affect the distribution and burden of carbonaceous aerosols in the atmosphere.

Global models for simulations of climate or atmospheric composition have generally assumed a simplified uniform lifetime (τ) for carbonaceous aerosols to convert from hydrophobic to hydrophilic ones, which is usually around 1 day (Chin et al., 2002; Chung and Seinfeld, 2002; Cooke et al., 1999). Previous studies on the regional budget and intercontinental transport of carbonaceous aerosols have also used this simplified assumption (Fu et al., 2009; Heald et al., 2006; Liao et al., 2007; Park et al., 2003; Wang et al., 2011).

On the other hand, recent chamber studies have shown that the aging of carbonaceous aerosols would be affected by local atmospheric environment such as ozone oxidation and water vapor inhibition (Pöschl et al., 2001), which implies that the hydrophobic to hydrophilic conversion rate would vary both spatially and temporally. Maria et al. (2004) reported that the average hydrophobic-to-hydrophilic conversion rate for organic aerosols was at least three times lower than the value widely used in climate models, which would potentially increase the burden of carbonaceous aerosols by 70 % in climate models.

ACPD
12, 28993–29023, 2012

In this work, based on chamber study results from Pöschl et al. (2001), we first implement a new aging mechanism for carbonaceous aerosols' hydrophobic to hydrophilic conversion in a chemical transport model, GEOS-Chem. Model simulation results for atmospheric carbonaceous aerosols with the updated aging mechanism are compared with observations of black carbon and organic carbon to evaluate the improvement in model performance over the standard version. The implications of this updated aging mechanism for model simulated global distribution, budgets, and long-range transport of carbonaceous aerosols are also examined with sensitivity studies.

2 Approach and model descriptions

We use the GEOS-Chem Chemical Transport Model (CTM) (Bey et al., 2001), which is a global three dimensional model driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling Assimilation Office (http://acmg.seas.harvard.edu/geos/index.html). We use GEOS-Chem v8-02-02 with resolution of 4° latitude by 5° longitude, and 47 vertical layers. GEOS-Chem contains fully coupled Ozone-NO_x-VOC-Aerosol chemistry mechanism (Park et al., 2004). We run the model from June 2004 to December 2005, with year 2004 as spin-up runs and 2005 results for final analysis.

Anthropogenic emissions (including emissions from fossil fuel and biofuel combustion) for both BC (4.53 Tg C yr^{-1}) and OC (9.33 Tg C yr^{-1}) are based on Bond et al. (2007) for the year 2000. In the standard version of GEOS-Chem, freshly emitted BC and OC are treated as hydrophobic and hydrophilic components, with 80 % hydrophobic BC and 50 % hydrophobic OC (Chin et al., 2002; Cooke et al., 1999; Park et al., 2003; Wang et al., 2011). For biomass burning emission inventory, we use the Global Fire Emission Database version 2 (GFED2) (van der Werf et al., 2006), with annual BC and OC emissions to be 2.80 Tg C and 21.98 Tg C, respectively. Biogenic emissions of VOCs are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) scheme (Guenther et al., 2006). Following Chin et al. (2002) and Park

ACPD
12, 28993–29023, 2012

et al. (2003), we assume that there is around 10 % of organic carbon aerosol yield (11.46 Tg C yr^{-1}) from terpines.

Dry deposition of aerosols in GEOS-Chem uses a standard resistance-in-series scheme from Wesely's model (Wesely, 1989; Wang et al., 1998). Wet scavenging follows the scheme used by Liu et al. (2001), including scavenging in convective updraft, rainout (in-cloud) and washout (below-cloud) from convective anvils and large-scale precipitation.

To investigate the sensitivity of model simulated carbonaceous aerosols to the aging scheme, we implement a new aging mechanism for carbonaceous aerosol's hygroscopic growth in the GEOS-Chem model based on results from chamber study (Pöschl et al., 2001), accounting for the oxidation of organic compounds by ozone while water vapor serving as inhibitor. The experiment-based formulation for the turn-over rate from hydrophobic to hydrophilic is:

$$\tau = \frac{1 + K_{O_3}[O_3] + K_{H_2O}[H_2O]}{K_\infty K_{O_3}[O_3]} \quad (1)$$

where $[O_3]$ and $[H_2O]$ are the concentrations for O_3 and H_2O in the atmosphere, respectively; τ is the hydrophobic to hydrophilic conversion lifetime for carbonaceous aerosols; K_∞ is pseudo-first-order decay rate coefficient in the limit of high ozone concentrations, which is set to be 0.015 s^{-1} based on experiment data; K_{O_3} is adsorption rate coefficient of O_3 and K_{H_2O} is adsorption rate coefficient of H_2O, with experimentally optimized values of 2.8×10^{-13} cm^3 and 2.1×10^{-17} cm^3, respectively, which are given by,

$$K_i = \frac{S_{0,i}\omega_i}{4[SS]_s}\tau_i \quad (2)$$

where $S_{0,i}$ is the sticking coefficient of O_3 and H_2O, with value 3.3×10^{-3} and 0.4×10^{-3}, respectively; ω_i is the mean thermal velocity; $[SS]_s$ is the surface concentration of

ACPD
12, 28993–29023, 2012

adsorption sites for the studied soot particles and measured to be 5.7×10^{14} site cm^{-2}; τ_i is the mean residence time of O_3 and H_2O on particle surface sites, measured to be 5 s and 3×10^{-3} s, respectively. Following Croft et al. (2005), we apply a physical shielding factor of 0.01 to account for the fact that the oxidized coating materials are not homogenously distributed on the surface of the aerosol.

Ground-based Interagency Monitoring of Protected Visual Environments (IMPROVE) observational data in 2005 (http://views.cira.colostate.edu/web/) is employed to evaluate the global carbonaceous aerosol simulation results from the control run and updated aging scheme simulations. Several studies have frequently applied IMPROVE data to compare with their model results to evaluate and access the effectiveness and efficiency of the model simulations (Chung and Seinfeld, 2002; Drury et al., 2010; Liao et al., 2007; Park et al., 2003). For the purpose of comparison to measurements, we treat BC equivalent to EC in this study although different measurement techniques, such as thermal technique for EC and photo-absorption for BC, could sometimes result in significant mass concentration differences (Jeong et al., 2004).

In order to investigate the impact of the updated aging scheme for carbonaceous aerosols' hydrophobic-to-hydrophilic conversion on intercontinental transport, we set up sensitivity model runs to examine the continental outflow of Asian emissions. For the purpose of this study, we define the Asian region spanning longitudes 63° E to 150° E and latitudes 10° N to 58° N, as shown in Fig. 1.

3 Results

3.1 Improvement in model simulation against observation

Figures 2 and 3 show the simulated BC and OC concentrations in surface air compared to observational data from two tropical sites from the IMPROVE network: the Hawaii Volcanoes National Park (HAVO) and the Haleakala National Park (HALE). All the values are annual means for 2005. Compared to the control run, the simulation

ACPD
12, 28993–29023, 2012

with updated aging scheme increases the annual mean concentration of BC and OC at HAVO by approximately 38 % and 30 %, respectively. This significantly improves the model performance, although it still underestimates the observations by a factor more than 2. Identifying the sources for the remaining model underestimates of carbonaceous aerosols is beyond the scope of this study, but it has been reported that there are around 10 % of sea salt aerosols containing organic compounds during the First Aerosol Characterization Experiment (ACE 1) by analyzing the composition of marine particles under various environmental conditions (Middlebrook et al., 1998). Cavalli et al. (2004) reported even higher fraction of organic compounds in marine aerosols during phytoplankton bloom period at North Atlantic, with 54 % and 4 % of organic compounds in marine aerosols under submicron and supermicron modes, respectively. Global models estimate that the marine OC emission source is around 8–9 $\mathrm{Tg\,C\,yr^{-1}}$ (Meskhidze et al., 2011; Spracklen et al., 2008), which is further confirmed by ship campaigns (9 $\mathrm{Tg\,C\,yr^{-1}}$) reported by Lapina et al. (2011).

3.2 Impacts of the updated aging mechanism on model simulations of carbonaceous aerosol

Figure 4 shows the hydrophobic to hydrophilic conversion lifetime τ for carbonaceous aerosol in surface air calculated with the updated aging mechanism. The longest lifetimes are found in the tropical areas, reflecting low ozone concentration and high humidity there, which is consistent with the simulation output by Tsigaridis and Kanakidou (2003). The global average value of τ in surface air is calculated to be around 5.4 days (Table 1), which is significantly longer than the simplified parameter of one day as commonly used in contemporary atmospheric models (Fu et al., 2009; Heald et al., 2006; Liao et al., 2007; Park et al., 2003; Wang et al., 2011).

Figure 5 shows the zonal mean plot for τ, with the highest value found in the lower tropical troposphere. The lifetimes generally decrease with altitude, due to increases in ozone concentration and decreases in specific humidity with altitude. The global average value for τ is calculated to be approximately 4.3 days. Compared to the control

ACPD
12, 28993–29023, 2012

runs, the decreased hydrophobic to hydrophilic conversion rates imply suppressed wet scavenging and hence increased atmospheric lifetimes of BC and OC with the updated aging mechanism. The global average lifetimes of BC and OC calculated with the new aging mechanism (7.61 days for BC and 5.35 days for OC) are significantly higher than those calculated in the control simulation, by 31 % and 17 %, respectively (Table 1). Consequently, the global burden of BC and OC are also found to be higher with the new aging mechanism (by 31 % and 17 %, respectively). The new aging scheme also puts our calculated lifetime and burden of BC and OC on the high end in comparison to literature studies on carbonaceous aerosols (Table 1).

We examine the impacts of the updated aging scheme on model calculated surface BC concentrations in Fig. 6. With the updated aging scheme, increases in surface BC concentrations by up to 0.05 $\mu g\,C\,m^{-3}$ are observed over South Asia, South America and African areas. These regions have high emissions of BC and also strong precipitation which make the model simulated BC particularly sensitive to the aging mechanism used. The global burden of BC increased by almost 31 % compared with control run (Table 1), indicating that the updated aging scheme could strengthen the BC climate forcings by 31 % (Maria et al., 2004). The ratio plot in Fig. 6 shows that the updated aging mechanism can lead to large increases in model simulated BC concentrations over the tropics and southern oceans with the surface BC concentrations over the tropics more than doubled.

Similar impacts are found on the model simulated OC concentrations with the updated aging mechanism (Fig. 7). In particular, the highest concentration of surface OC concentration is predicted to increase by 0.67 $\mu g\,C\,m^{-3}$ over Asia, with the maximum ratio value up to 2.8 found over the tropic oceans. The global burden of updated OC concentration increases by almost 17 % (Table 1), which implies more cooling effect of OC on climate forcing (Haywood and Boucher, 2000).

Global distributions of model simulated BC and OC are shown in zonal means in Figs. 8 and 9 respectively. For BC (Fig. 8), the highest concentrations are found over the northern mid-latitudes near the surface due to the BC sources from combustion of fossil

ACPD
12, 28993–29023, 2012

fuel, with second peak happening near the tropic, indicating the importance of biomass burning (Chung and Seinfeld, 2002; Ramanathan and Carmichael, 2008). With the updated aging mechanism, the concentrations of BC increase by more than 20 % in the middle and upper atmosphere, with the largest perturbations found in the upper atmosphere (more than double). The strongest perturbations to model simulated OC with the updated aging scheme are found over the tropics (Fig. 9), reflecting the strong sources of OC from biomass burning and biogenic emissions. OC concentrations in the tropical upper troposphere are calculated to increase by a factor of 4 with the updated aging scheme.

3.3 Effect of the updated aging scheme on model simulated intercontinental transport of BC and OC

The large perturbations to the calculated lifetimes of BC and OC with the updated aging scheme imply that the intercontinental transport of BC and OC could be significantly underestimated in previous modeling studies. We examined the potential impacts of the updated aging scheme on model simulated continental outflow from Asia with sensitivity studies where only emissions from the Asian region (as defined in Fig. 1) are turned on in the model. Figure 10 compares the model calculated surface BC concentrations between the control and updated scheme runs. The largest increases, by up to 0.06 $\mu g\,C\,m^{-3}$, are found over the source regions, which is consistent with the sensitivity simulation results discussed in Sect. 3.2.

With the updated aging scheme, the contribution of Asian emission to average surface BC in the United States is about 0.003 $\mu g\,C\,m^{-3}$, which is approximately 1.5 times higher than that from the control run. The simulated contribution of Asian emissions to surface BC in the United States increases by 31 %–63 % with the updated aging scheme. Similar results are found for OC aerosols. The maximum difference between control and updated simulation on surface air is approximately 0.1 $\mu g\,C\,m^{-3}$.

The ratio plots in Figs. 10 and 11 show the largest perturbations to surface BC and OC in remote areas, in particular the Southern Hemisphere. This reflects that the

ACPD
12, 28993–29023, 2012

impacts of the updated aging scheme on simulated BC and OC become more prominent after long-range transport where the BC and OC aerosols experience aging and wet scavenging.

The effects of the updated aging scheme on long-range transport of carbonaceous aerosols can also be examined through the zonal mean plots (Figs. 12 and 13). Compared to the control run, BC and OC concentrations in the upper atmosphere, in particular for the Southern Hemisphere, double with the updated aging scheme. This again reflects that the updated aging scheme has more implications on areas further away from the source regions.

4 Conclusions and discussion

We implement a new aging mechanism for carbonaceous aerosols in GEOS-Chem where the hydrophobic-to-hydrophilic conversion is affected by local conditions including O_3 concentration and humidity based on results from chamber studies (Pöschl et al., 2001). The simulated hydrophobic-to-hydrophilic conversion lifetime of carbonaceous aerosols exhibits large spatial and temporal variation with the global average calculated to be 4.3 days which is much longer than the parameter commonly used in global models. The longest conversion lifetimes are found in the tropical areas such as the Amazon forest, reflecting the low ozone concentration and high humidity there. The conversion lifetime generally decreases with altitude due to increases in ozone concentration and decreases in water vapor concentration.

The updated aging mechanism has significant impacts on the model simulations of carbonaceous aerosols, with increases in model simulated concentrations of BC and OC by up to 0.16 $\mu g\,C\,m^{-3}$ and 0.67 $\mu g\,C\,m^{-3}$, respectively. The largest effects are found for the tropical upper troposphere, where the carbonaceous aerosol concentrations more than double compared to the control simulation. The calculated global burden of BC and OC increases by 31 % and 17 %, respectively, with the updated aging scheme.

ACPD
12, 28993–29023, 2012

Comparisons of the model simulated BC and OC against observational data shows that the updated aging scheme improves model performance, especially in the tropical regions or remote areas, although significant model underestimates still exist. The updated aging scheme also has significant implications for the estimates on carbonaceous aerosols' continental outflow. Our sensitivity model simulations show that the contributions of Asian emissions on background BC and OC in remote regions double with the new aging scheme. The large increases in simulated BC and OC in remote regions with the update aging scheme implies that the intercontinental transport of BC and OC and the anthropogenic influences on remote regions (such as the polar regions) may be underestimated in previous modeling studies.

Besides the chemical aging, there are additional processes (such as coagulation and condensation) that could affect the carbonaceous aerosols' hydrophobic-to-hydrophilic conversion (Capes et al., 2008; Croft et al., 2005; Kanakidou et al., 2005; Liu et al., 2011; Petters et al., 2006; Pöschl, 2005; Riemer et al., 2004; Rudich et al., 2007). Our study here does not account for these coagulation and condensation effects. The strong sensitivities of the model simulations of carbonaceous aerosols to the aging scheme indicate that further studies on the evolution of carbonaceous aerosol are needed to better understand the effects of carbonaceous aerosol on climate, atmospheric composition and air quality.

Acknowledgement. This work was supported by US EPA (grant # R83428601) and NIH (grant #1 RC1 ES018612). We thank Robert Page at Michigan Tech for his help on programming.

References

Balkanski, Y., Myhre, G., Gauss, M., Rädel, G., Highwood, E. J., and Shine, K. P.: Direct radiative effect of aerosols emitted by transport: from road, shipping and aviation, Atmos. Chem. Phys., 10, 4477–4489, doi:10.5194/acp-10-4477-2010, 2010.

Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H., Mickley, L. J., and Schultz, M. G.: Global modeling of tropospheric chemistry with assimilated

ACPD

12, 28993–29023, 2012

meteorology: model description and evaluation, J. Geophys. Res.-Atmos., 106, 23073–23095, 2001.

Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G., and Trautmann, N. M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, Global Biogeochem. Cy., 21, GB2018, doi:10.1029/2006gb002840, 2007.

Capes, G., Johnson, B., McFiggans, G., Williams, P. I., Haywood, J., and Coe, H.: Aging of biomass burning aerosols over West Africa: aircraft measurements of chemical composition, microphysical properties, and emission ratios, J. Geophys. Res.-Atmos., 113, D00C15, doi:10.1029/2008jd009845, 2008.

Cavalli, F., Facchini, M. C., Decesari, S., Mircea, M., Emblico, L., Fuzzi, S., Ceburnis, D., Yoon, Y. J., O'Dowd, C. D., Putaud, J. P., and Dell'Acqua, A.: Advances in characterization of size-resolved organic matter in marine aerosol over the North Atlantic, J. Geophys. Res.-Atmos., 109, D24215, doi:10.1029/2004jd005137, 2004.

Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B. N., Duncan, B. N., Martin, R. V., Logan, J. A., Higurashi, A., and Nakajima, T.: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and Sun photometer measurements, J. Atmos. Sci., 59, 461–483, doi:10.1175/1520-0469(2002)059<0461:taotft>2.0.co;2, 2002

Chung, S. H. and Seinfeld, J. H.: Global distribution and climate forcing of carbonaceous aerosols, J. Geophys. Res.-Atmos., 107, 4407, doi:10.1029/2001jd001397, 2002.

Colarco, P., da Silva, A., Chin, M., and Diehl, T.: Online simulations of global aerosol distributions in the NASA GEOS-4 model and comparisons to satellite and ground-based aerosol optical depth, J. Geophys. Res.-Atmos., 115, D14027, doi:10.1029/2009JD012820, 2010.

Cooke, W. F. and Wilson, J. J. N.: A global black carbon aerosol model, J. Geophys. Res.-Atmos., 101, 19395–19409, doi:10.1029/96JD00671, 1996.

Cooke, W. F., Liousse, C., Cachier, H., and Feichter, J.: Construction of a 1 degrees × 1 degrees fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, J. Geophys. Res.-Atmos., 104, 22137–22162, doi:10.1029/1999JD900187, 1999.

Cooke, W. F., Ramaswamy, V., and Kasibhatla, P.: A general circulation model study of the global carbonaceous aerosol distribution, J. Geophys. Res., 107, 4279, doi:10.1029/2001JD001274, 2002.

ACPD
12, 28993–29023, 2012

Croft, B., Lohmann, U., and von Salzen, K.: Black carbon ageing in the Canadian Centre for Climate modelling and analysis atmospheric general circulation model, Atmos. Chem. Phys., 5, 1931–1949, doi:10.5194/acp-5-1931-2005, 2005.

Drury, E., Jacob, D. J., Spurr, R. J. D., Wang, J., Shinozuka, Y., Anderson, B. E., Clarke, A. D., Dibb, J., McNaughton, C., and Weber, R.: Synthesis of satellite (MODIS), aircraft (ICARTT), and surface (IMPROVE, EPA-AQS, AERONET) aerosol observations over eastern North America to improve MODIS aerosol retrievals and constrain surface aerosol concentrations and sources, J. Geophys. Res.-Atmos., 115, D14204, doi:10.1029/2009jd012629, 2010.

Easter, R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak, H., Leung, L. R., Bian, X. D., and Zaveri, R. A.: MIRAGE: Model description and evaluation of aerosols and trace gases, J. Geophys. Res.-Atmos., 109, D20210, doi:10.1029/2004jd004571, 2004.

Fu, T. M., Jacob, D. J., and Heald, C. L.: Aqueous-phase reactive uptake of dicarbonyls as a source of organic aerosol over eastern North America, Atmos. Environ., 43, 1814–1822, doi:10.1016/j.atmosenv.2008.12.029, 2009.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.

Haywood, J. and Boucher, O.: Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review, Rev. Geophys., 38, 513–543, doi:10.1029/1999RG000078, 2000.

Heald, C. L., Jacob, D. J., Turquety, S., Hudman, R. C., Weber, R. J., Sullivan, A. P., Peltier, R. E., Atlas, E. L., de Gouw, J. A., Warneke, C., Holloway, J. S., Neuman, J. A., Flocke, F. M., and Seinfeld, J. H.: Concentrations and sources of organic carbon aerosols in the free troposphere over North America, J. Geophys. Res.-Atmos., 111, D23S47, doi:10.1029/2006JD007705, 2006.

Jacobson, M. Z.: Short-term effects of controlling fossil-fuel soot, biofuel soot and gases, and methane on climate, Arctic ice, and air pollution health, J. Geophys. Res.-Atmos., 115, D14209, doi:10.1029/2009jd013795, 2010.

Jeong, C. H., Hopke, P. K., Kim, E., and Lee, D. W.: The comparison between thermal-optical transmittance elemental carbon and Aethalometer black carbon measured at multiple monitoring sites, Atmos. Environ., 38, 5193–5204, doi:10.1016/j.atmosenv.2004.02.065, 2004.

ACPD
12, 28993–29023, 2012

Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., and Wilson, J.: Organic aerosol and global climate modelling: a review, Atmos. Chem. Phys., 5, 1053–1123, doi:10.5194/acp-5-1053-2005, 2005.

Koch, D.: Transport and direct radiative forcing of carbonaceous and sulfate aerosols in the GISS GCM, J. Geophys. Res.-Atmos., 106, 20311–20332, doi:10.1029/2001JD900038, 2001.

Koch, D. and Hansen, J.: Distant origins of Arctic black carbon: a Goddard Institute for Space Studies ModelE experiment, J. Geophys. Res.-Atmos., 110, D04204, doi:10.1029/2004jd005296, 2005.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø, Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, doi:10.5194/acp-9-9001-2009, 2009.

Kondo, Y., Miyazaki, Y., Takegawa, N., Miyakawa, T., Weber, R. J., Jimenez, J. L., Zhang, Q., and Worsnop, D. R.: Oxygenated and water-soluble organic aerosols in Tokyo, J. Geophys. Res.-Atmos., 112, D01203, doi:10.1029/2006jd007056, 2007.

Lapina, K., Heald, C. L., Spracklen, D. V., Arnold, S. R., Allan, J. D., Coe, H., McFiggans, G., Zorn, S. R., Drewnick, F., Bates, T. S., Hawkins, L. N., Russell, L. M., Smirnov, A., O'Dowd, C. D., Hind, A. J.: Investigating organic aerosol loading in the remote marine environment, Atmos. Chem. Phys, 11, 8847–8860, doi:10.5194/acp-11-8847-2011, 2011.

Liao, H., Henze, D. K., Seinfeld, J. H., Wu, S. L., and Mickley, L. J.: Biogenic secondary organic aerosol over the United States: comparison of climatological simulations with observations, J. Geophys. Res.-Atmos., 112, D06201, doi:10.1029/2006JD007813, 2007.

Liousse, C., Penner, J. E., Chuang, C., Walton, J. J., Eddleman, H., and Cachier, H.: A global three- dimensional model study of carbonaceous aerosols, J. Geophys. Res.-Atmos., 101, 19411–19432, doi:10.1029/95JD03426, 1996.

ACPD
12, 28993–29023, 2012

Liu, H. Y., Jacob, D. J., Bey, I., and Yantosca, R. M.: Constraints from Pb-210 and Be-7 on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, J. Geophys. Res.-Atmos., 106, 12109–12128, doi:10.1029/2000JD900839, 2001.

Liu, X., Penner, J. E., and Herzog, M.: Global modeling of aerosol dynamics: model description, evaluation, and interactions between sulfate and nonsulfate aerosols, J. Geophys. Res.-Atmos., 110, D18206, doi:10.1029/2004JD005674, 2005.

Liu, J., Mauzerall, D. L., Horowitz, L. W., Ginoux, P., and Fiore, A. M.: Evaluating intercontinental transport of fine aerosols: (1) methodology, global aerosol distribution and optical depth, Atmos. Environ., 43, 4327–4338, doi:10.1016/j.atmosenv.2009.03.054, 2009.

Liu, J., Fan, S., Horowitz, L. W., and Levy, H.: Evaluation of factors controlling long-range transport of black carbon to the Arctic, J. Geophys. Res.-Atmos., 116, D04307, doi:10.1029/2010jd015145, 2011.

Lohmann, U., Feichter, J., Penner, J., and Leaitch, R.: Indirect effect of sulfate and carbonaceous aerosols: a mechanistic treatment, J. Geophys. Res.-Atmos., 105, 12193–12206, doi:10.1029/1999JD901199, 2000.

Mann, G. W., Carslaw, K. S., Ridley, D. A., Spracklen, D. V., Pringle, K. J., Merikanto, J., Korhonen, H., Schwarz, J. P., Lee, L. A., Manktelow, P. T., Woodhouse, M. T., Schmidt, A., Breider, T. J., Emmerson, K. M., Reddington, C. L., Chipperfield, M. P., and Pickering, S. J.: Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, Atmos. Chem. Phys., 12, 4449–4476, doi:10.5194/acp-12-4449-2012, 2012.

Maria, S. F., Russell, L. M., Gilles, M. K., and Myneni, S. C. B.: Organic aerosol growth mechanisms and their climate-forcing implications, Science, 306, 1921–1924, doi:10.1126/science.1103491, 2004.

Meskhidze, N., Xu, J., Gantt, B., Zhang, Y., Nenes, A., Ghan, S. J., Liu, X., Easter, R., and Zaveri, R.: Global distribution and climate forcing of marine organic aerosol: 1. Model improvements and evaluation, Atmos. Chem. Phys., 11, 11689–11705, doi:10.5194/acp-11-11689-2011, 2011.

Middlebrook, A. M., Murphy, D. M., and Thomson, D. S.: Observations of organic material in individual marine particles at Cape Grim during the First Aerosol Characterization Experiment (ACE 1), J. Geophys. Res.-Atmos., 103, 16475–16483, doi:10.1029/97jd03719, 1998.

ACPD
12, 28993–29023, 2012

Park, R. J., Jacob, D. J., Chin, M., and Martin, R. V.: Sources of carbonaceous aerosols over the United States and implications for natural visibility, J. Geophys. Res.-Atmos., 108, 4355, doi:10.1029/2002jd003190, 2003.
Park, R. J., Jacob, D. J., Field, B. D., Yantosca, R. M., and Chin, M.: Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: implications for policy, J. Geophys. Res.-Atmos., 109, D15204, doi:10.1029/2003jd004473, 2004.
Petters, M. D., Prenni, A. J., Kreidenweis, S. M., DeMott, P. J., Matsunaga, A., Lim, Y. B., and Ziemann, P. J.: Chemical aging and the hydrophobic-to-hydrophilic conversion of carbonaceous aerosol, Geophys. Res. Lett., 33, 309–313, 2006.
Pierce, J. R., Chen, K., and Adams, P. J.: Contribution of primary carbonaceous aerosol to cloud condensation nuclei: processes and uncertainties evaluated with a global aerosol microphysics model, Atmos. Chem. Phys., 7, 5447–5466, doi:10.5194/acp-7-5447-2007, 2007.
Pöschl, U.: Atmospheric aerosols: composition, transformation, climate and health effects, Angew. Chem. Int. Ed., 44, 7520–7540, doi:10.1002/anie.200501122, 2005.
Pöschl, U., Letzel, T., Schauer, C., and Niessner, R.: Interaction of ozone and water vapor with spark discharge soot aerosol particles coated with benzo[a]pyrene: O_3 and H_2O adsorption, benzo[a]pyrene degradation, and atmospheric implications, J. Phys. Chem. A, 105, 4029–4041, doi:10.1021/jp004137n, 2001.
Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nature Geosci., 1, 221–227, doi:10.1038/ngeo156, 2008.
Riemer, N., Vogel, H., and Vogel, B.: Soot aging time scales in polluted regions during day and night, Atmos. Chem. Phys., 4, 1885–1893, doi:10.5194/acp-4-1885-2004, 2004.
Rudich, Y., Donahue, N. M., and Mentel, T. F.: Aging of organic aerosol: bridging the gap between laboratory and field studies, Annu. Rev. Phys. Chem., 58, 321–352, doi:10.1146/annurev.physchem.58.032806.104432, 2007.
Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Dentener, F., Guibert, S., Isaksen, I. S. A., Iversen, T., Koch, D., Kirkevag, A., Liu, X., Montanaro, V., Myhre, G., Penner, J. E., Pitari, G., Reddy, S., Seland, O., Stier, P., Takemura, T.: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmos. Chem. Phys, 6, 5225–5246, doi:10.5194/acp-6-5225-2006, 2006.
Spracklen, D. V., Arnold, S. R., Sciare, J., Carslaw, K. S., and Pio, C.: Globally significant oceanic source of organic carbon aerosol, Geophys. Res. Lett., 35, L12811, doi:10.1029/2008gl033359, 2008.

ACPD
12, 28993–29023, 2012

Tsigaridis, K. and Kanakidou, M.: Global modelling of secondary organic aerosol in the troposphere: a sensitivity analysis, Atmos. Chem. Phys., 3, 1849–1869, doi:10.5194/acp-3-1849-2003, 2003.

US Environmental Protection Agency (EPA): Report on air quality in nonattainment areas for 2003–2005 covering ozone, particulate matter, carbon monoxide, sulfur dioxide, nitrogen dioxide, and lead, Washing, D. C., available at: http://www.epa.gov/air/airtrends/pdfs/20070214_aq_na_2003-2005.pdf, 2007.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano Jr., A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004, Atmos. Chem. Phys., 6, 3423–3441, doi:10.5194/acp-6-3423-2006, 2006.

Vignati, E., Karl, M., Krol, M., Wilson, J., Stier, P., and Cavalli, F.: Sources of uncertainties in modelling black carbon at the global scale, Atmos. Chem. Phys., 10, 2595–2611, doi:10.5194/acp-10-2595-2010, 2010.

Wang, Q., Jacob, D. J., Fisher, J. A., Mao, J., Leibensperger, E. M., Carouge, C. C., Le Sager, P., Kondo, Y., Jimenez, J. L., Cubison, M. J., and Doherty, S. J.: Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing, Atmos. Chem. Phys., 11, 12453–12473, doi:10.5194/acp-11-12453-2011, 2011.

Wang, Y. H., Jacob, D. J., and Logan, J. A.: Global simulation of tropospheric O_3-NO_x-hydro-carbon chemistry. 1. Model formulation, J. Geophys. Res.-Atmos., 103, 10713–10725, doi:10.1029/98jd00158, 1998.

Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical-models, Atmos. Environ., 23, 1293–1304, doi:10.1016/0004-6981(89)90153-4, 1989.

ACPD

12, 28993–29023, 2012

Table 1. Model simulated global budgets of carbonaceous aerosols.

Reference	BC lifetime (days)	BC burden (Tg C)	OC lifetime (days)	OC burden (Tg C)
This study, with old aging scheme	5.82	0.117	4.58	0.537
This study, with new aging scheme	7.61	0.153	5.35	0.626
Mann et al. (2012)	4.76	0.100	4.59	0.87
Liu et al. (2011)	9.5	0.20	NA	NA
Colarco et al. (2010)	8.82	0.174	6.9	0.929
Jacobson (2010)	4.7	0.127	NA	NA
Vignati et al. (2010)	6.41	0.12	NA	NA
Koch et al. (2009)	9.2	0.18	NA	NA
Liu et al. (2009)	5.8	0.17	5.3	0.83
Pierce et al. (2007)	8.48	0.19	4.80	0.80
Koch and Hansen (2005)	7.30	0.22	NA	NA
Liu et al. (2005)	3.3	0.12	3.2	0.98
Croft et al. (2005)	9.5	0.209	NA	NA
Easter et al. (2004)	5.9	0.16	5.5	0.99
Cooke et al. (2002)	4.29	0.06	3.39	0.13
Chin et al. (2002)	6.2	0.32	5.1	1.5
Chung and Seinfeld (2002)	6.4	0.22	5.3	1.2
Koch (2001)	4.40	0.15	3.86	0.95
Cooke et al. (1999)	5.29*	0.073*	4.54*	0.087*
Cooke and Wilson (1996)	7.85	0.30	NA	NA
Liousse et al. (1996)	3.88	0.13	NA	NA

* Only accounting for BC and OC from fossil fuel.

ACPD
12, 28993–29023, 2012

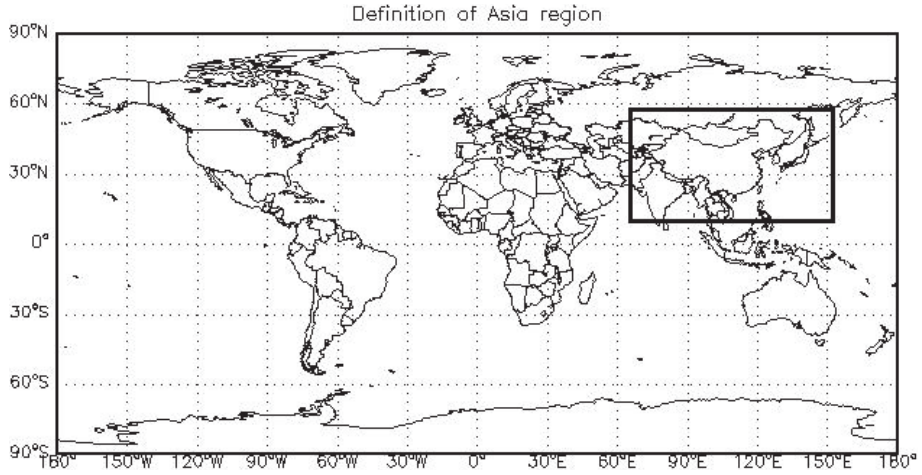

Fig. 1. Definition of Asia region for intercontinental transport study (longitude 63° E–150° E and latitude 10° N–58° N).

ACPD
12, 28993–29023, 2012

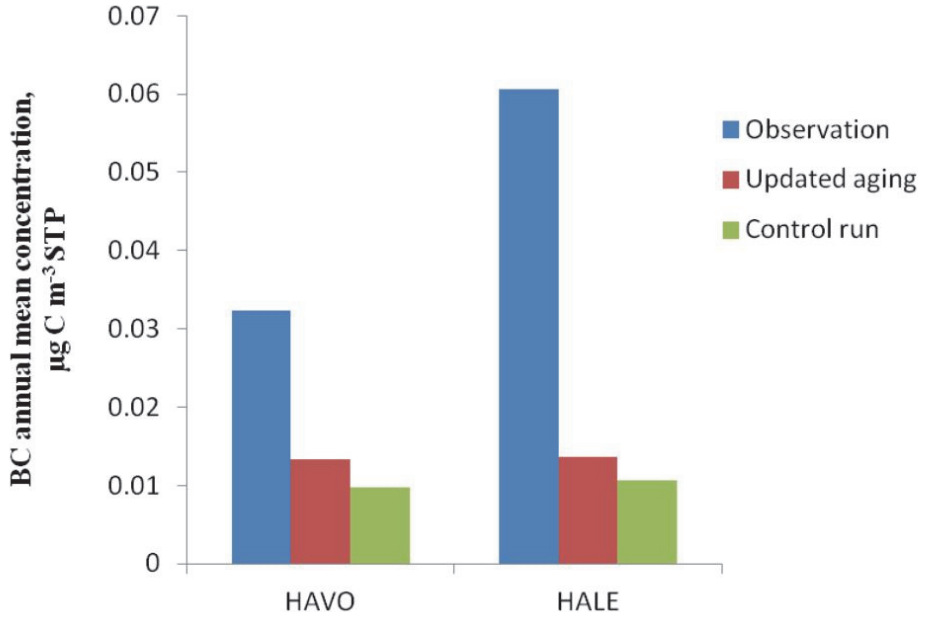

Fig. 2. Annual mean concentrations of BC at HAVO and HALE in 2005. Observational data by IMPROVE is shown in blue, while the updated and standard simulation results by GEOS-Chem are shown as red and green, respectively. STP refers to standard temperature and pressure conditions (273 K, 1 atm). HAVO (19° N, 155° W); HALE (21° N, 156° W).

ACPD
12, 28993–29023, 2012

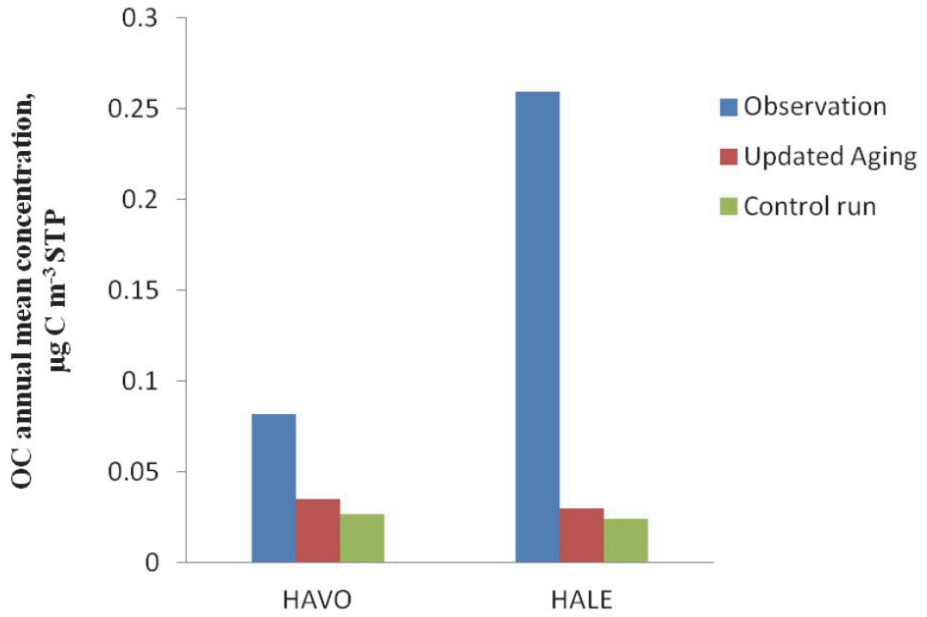

Fig. 3. Same as Fig. 2 but for OC.

ACPD
12, 28993–29023, 2012

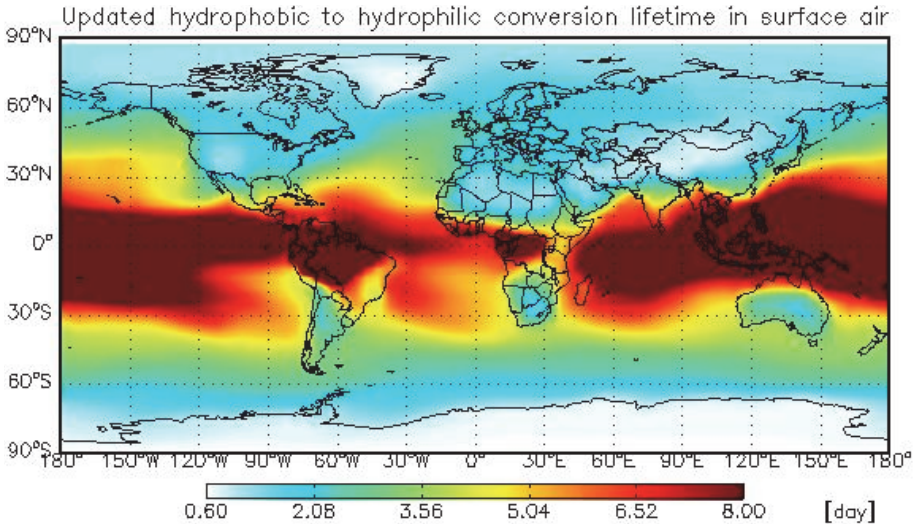

Fig. 4. Model calculated hydrophobic to hydrophilic conversion lifetime for carbonaceous aerosols in surface air.

ACPD
12, 28993–29023, 2012

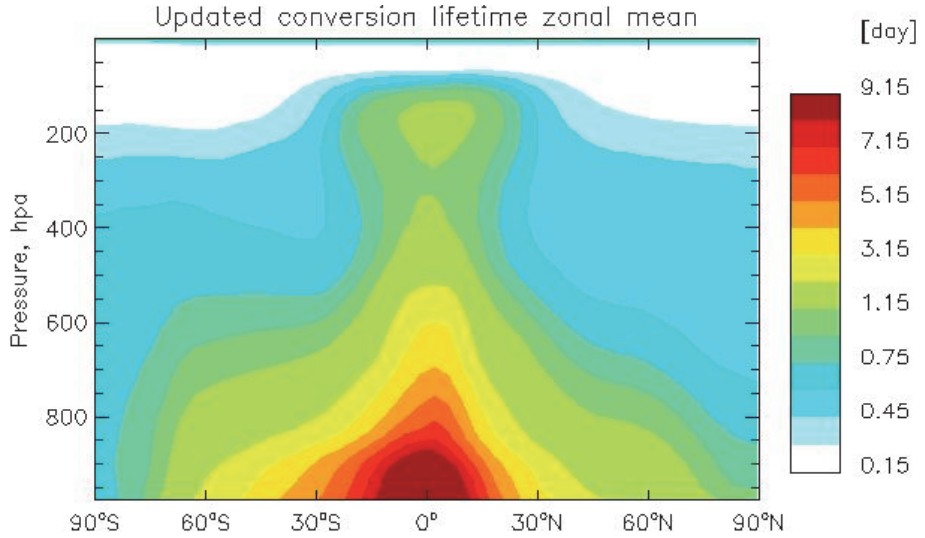

Fig. 5. Zonal mean plot for hydrophobic to hydrophilic conversion lifetime for carbonaceous aerosol.

ACPD
12, 28993–29023, 2012

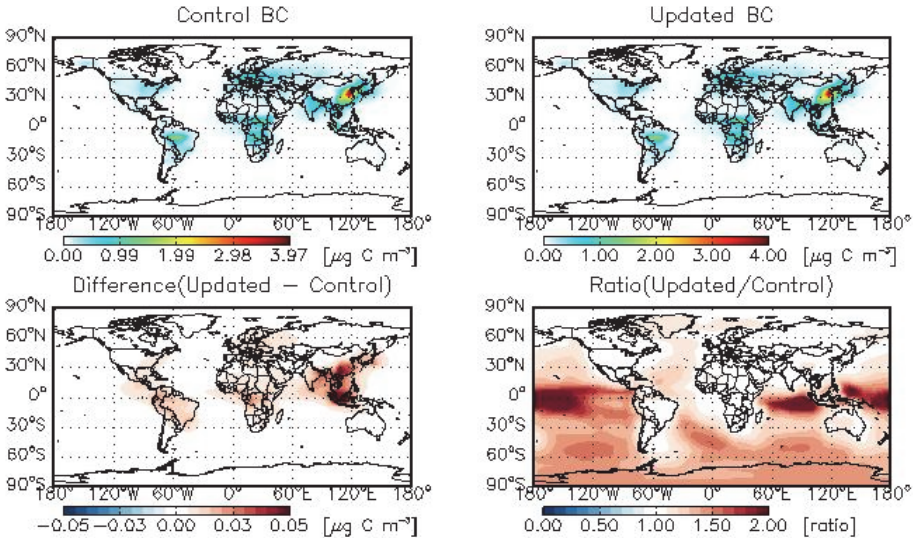

Fig. 6. Model simulated annual mean surface BC concentrations from the sensitivity run versus the control run. (Upper left) BC concentrations from the control run; (upper right) BC concentrations from the sensitivity run using the updated aging scheme; (lower left) differences between the sensitivity run and control run results; (lower right) ratio between sensitivity run and control run results.

ACPD
12, 28993–29023, 2012

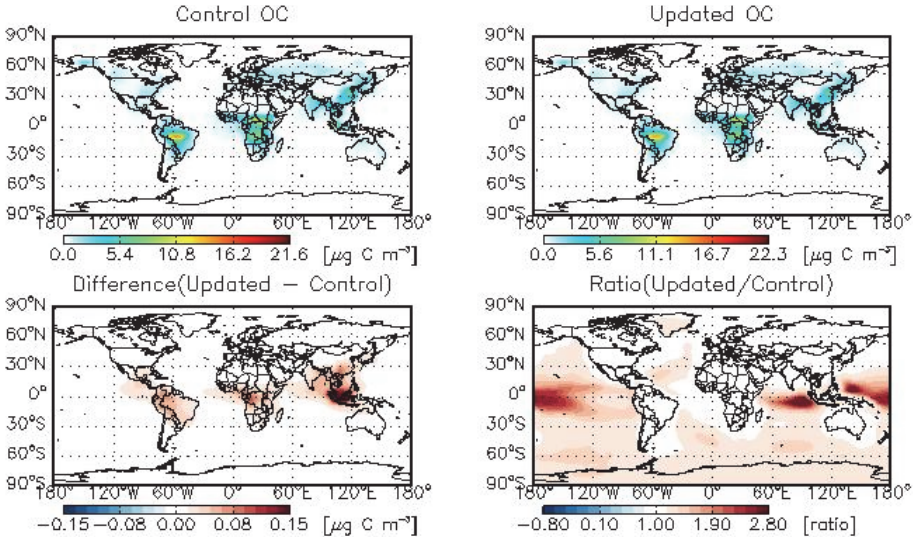

Fig. 7. Same as Fig. 6 but for OC.

ACPD
12, 28993–29023, 2012

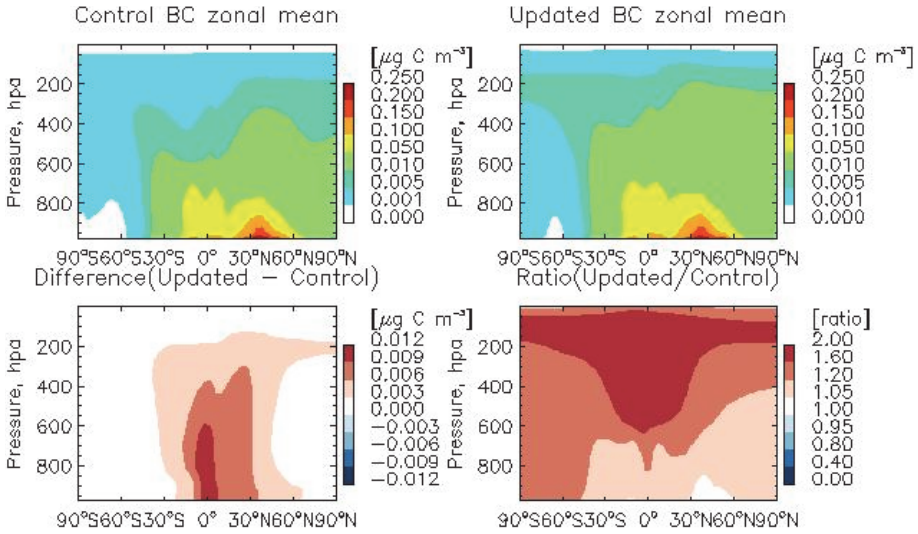

Fig. 8. Same as Fig. 6 but for zonal mean.

ACPD
12, 28993–29023, 2012

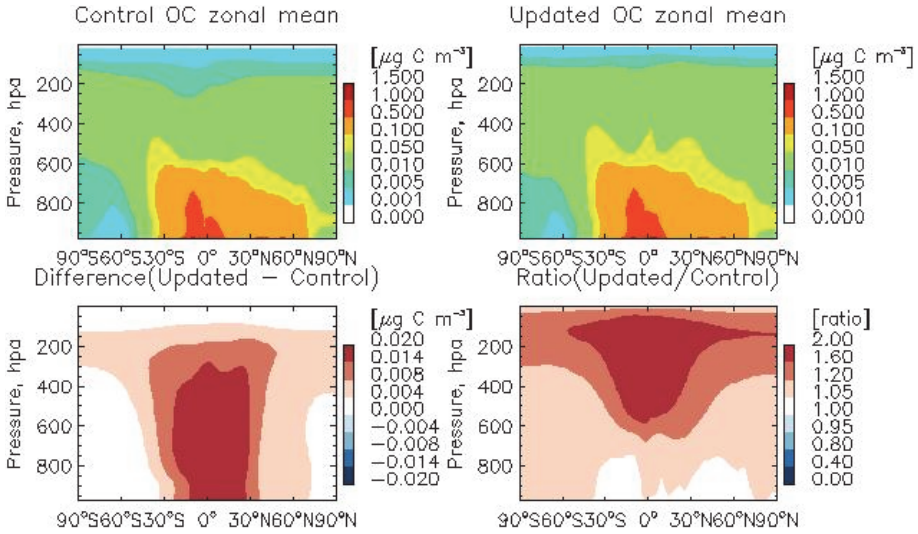

Fig. 9. Same as Fig. 7 but for zonal mean.

ACPD
12, 28993–29023, 2012

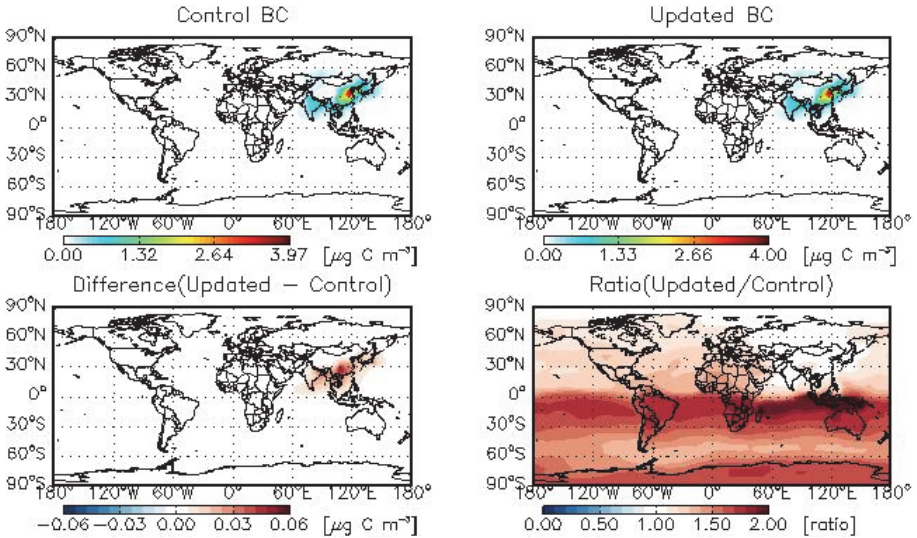

Fig. 10. Impacts of the update aging scheme on model calculated continental outflow of annual mean BC from Asia (the global BC emissions except those over Asia are turned off in the model simulations). (Upper left) surface BC concentrations from the control run; (upper right) surface BC concentrations from the sensitivity run with the updated aging scheme; (lower left) differences between the sensitivity run and control run; (lower right) ratio between the sensitivity run and control run.

ACPD
12, 28993–29023, 2012

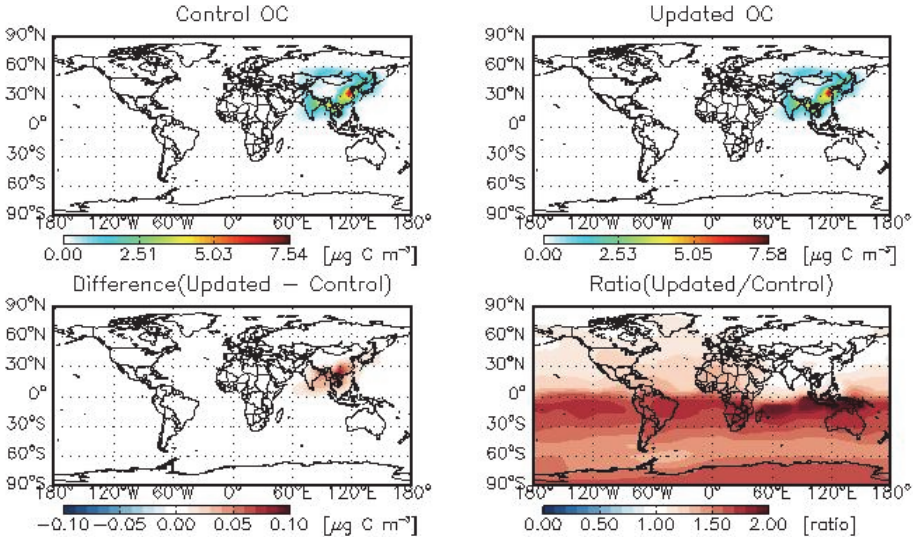

Fig. 11. Same as Fig. 10 but for OC.

ACPD
12, 28993–29023, 2012

Impact of aging mechanism on model simulated aerosols

Y. Huang et al.

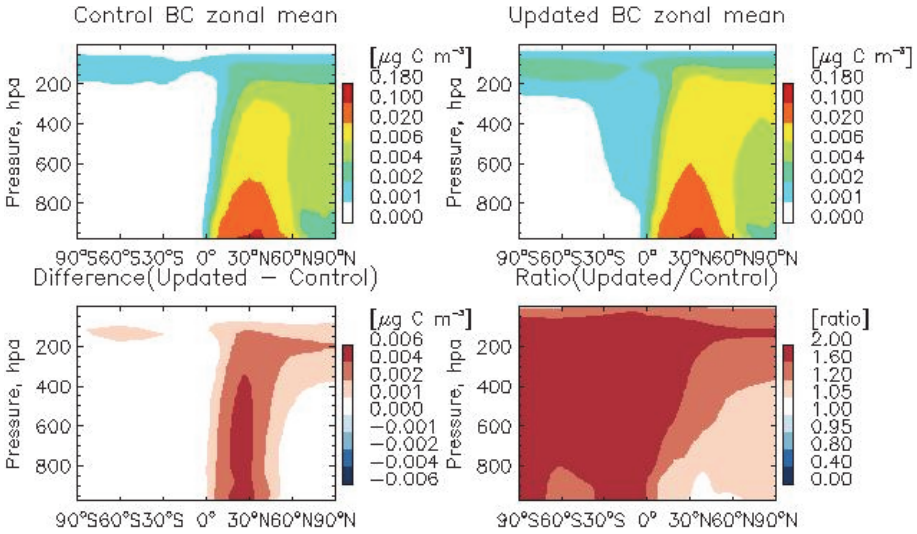

Fig. 12. Same as Fig. 10 but for zonal mean.

ACPD
12, 28993–29023, 2012

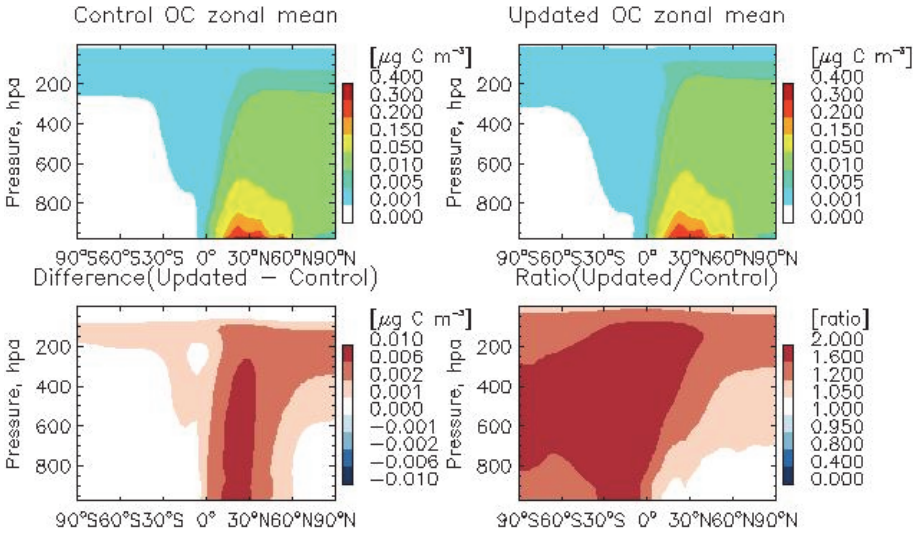

Fig. 13. Same as Fig. 11 but for zonal mean.

ACPD
12, 28993–29023, 2012