Effects of updated aging scheme on the model simulation of global carbonaceous aerosols

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Background

Carbonaceous aerosols in the atmosphere evolve through physical and chemical processes[Capes et al., 2008]. Global models generally apply a simplified uniform lifetime for carbonaceous aerosols to convert from hydrophobic to hydrophilic ones, which is usually around 1 day [*Cooke et al.*, 1999] as is the case for the standard version of GEOS-Chem. However, chamber study has shown that the aging of carbonaceous aerosols should be affected by ozone oxidation and water vapor inhibition[*Pöschl et al.*, 2001], which implies the conversion rate would vary both spatially and temporally. *Maria et al.* [2004] reported that the average conversion rate was at least three times lower than the value that mostly used in climate models, which would potentially increase the burden of carbonaceous particles by 70% in climate models.

Research Method

We use the GEOS-Chem CTM, which is a global 3-D chemical transport model driven by assimilated meteorological observations from the Goddard Earth Observing System(GEOS) of the NASA Global Modeling Assimilation Office. The research is conducted in the following procedures.

(1) Based on data from chamber study [Pöschl et al., 2001], an updated hydrophobic to hydrophilic aging mechanism for carbonaceous aerosol was implemented in GEOS-Chem where the lifetimes of carbonaceous aerosol are calculated online as a function of ozone levels and humidity;

(2) The impacts of the updated aging mechanism on global simulations of carbonaceous aerosols are evaluated with sensitivity studies.

(3) Ground-based measurements from Integency Monitoring of Protected Visual Environments (IMPROVE) and PICO-NARE are employed to compare with simulation results.

Results

1. Updated scheme of hydrophobic to hydrophilic conversion of carbonaceous aerosol in GEOS-Chem

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

[Pöschl et al., 2001; Croft et al., 2005].

τ : conversion lifetime from hydrophobic to hydrophilic for carbonaceous aerosol;
K_{∞} : pseudo-first-order decay rate coefficient in the limit of high ozone concentrations;
K_{O3} : adsorption rate coefficient of O_3;
K_{H2O} : adsorption rate coefficient of H_2O;
$[O_3]$ and $[H_2O]$ are the concentrations for O_3 and H_2O, respectively.

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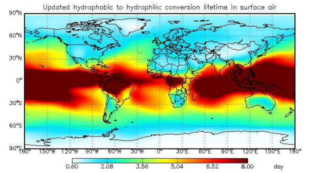
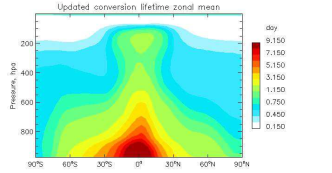
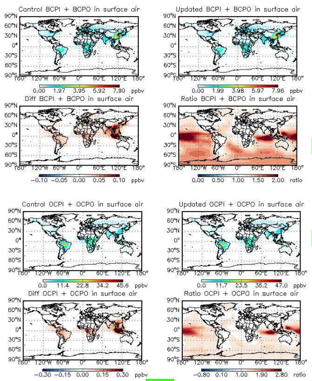


With the updated aging mechanism, the global area-weighted average hydrophobic to hydrophilic conversion lifetime of carbonaceous aerosols in surface air is around 5.4 days, with global volume-weighted average hydrophobic to hydrophilic conversion lifetime approximately 4.3 days.



The highest lifetime of carbonaceous aerosols from hydrophobic to hydrophilic appears at tropical areas. With the increase of altitude, its lifetime decreases accordingly due to high ozone and low water vapor concentration.

2. Impacts of the updated aging mechanism on global simulations of carbonaceous aerosol



Model simulated global budgets of carbonaceous aerosols

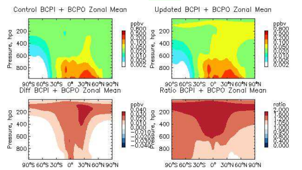
Reference	BC Lifetime (days)	OC Lifetime (days)	BC burden (Tg)	OC burden (Tg)
This study, with old aging scheme	5.82	4.58	0.117	0.537
This study, with new aging scheme	7.61	5.35	0.153	0.626
[Chung and Seinfeld, 2002]	6.4	5.3	0.22	1.2
[Koch, 2001]	4.28-8.42[a]	4.58	0.14-0.29[a]	0.95
[Cooke et al., 1999]	5.29[b]	5.35	0.073[b]	0.11
[Cooke and Wilson, 1996]	6-10	NA	0.28	NA
[Liousse et al., 1996]	4-4.5	NA	0.13	NA

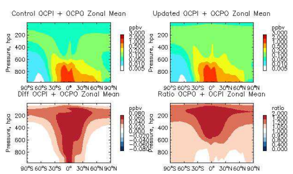
a. The range is due to different wet scavenging schemes
b. Fossil fuel only

(1) Increases in model simulated concentrations of both BC(by up to 0.33 ppb) and OC (by up to 1.4ppb) in surface air are calculated;

(2) The largest impacts on BC/OC's hydrophobic to hydrophilic conversion lifetime are found in the tropical areas;

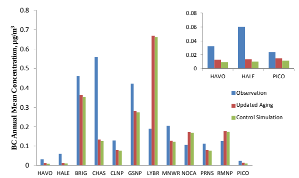
(3) The largest effects on simulated BC/OC concentration are found for tropical upper troposphere, where the carbonaceous aerosol concentrations more than double with the updated aging mechanism.

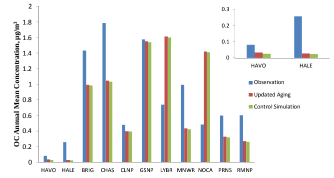




Control : lifetime (1.15 days); Updated: updated aging scheme; BCPI: Black Carbon Hydrophilic; BCPO: Black Carbon Hydrophobic; OCPI: Organic Carbon Hydrophilic; OCPO: Organic Carbon Hydrophobic.
Diff: Updated – Control; Ratio: Updated/Control.

3. Comparison with observations





(1) Ground-based IMPROVE observational data in 2005 and PICO-NARE located at Azores are employed to compare with the global carbonaceous aerosol simulation results from control and updated aging scheme simulation;

(2) Updated aging scheme simulation significantly increases annual mean concentration of BC and OC at tropical regions, such as Hawaii Volcanoes National Park (HAVO) by approximately 38% and 30%, respectively ;

(3) With the updated aging scheme, the remaining low model bias for OC simulations in remote marine sites, such as HAVO and HALE, likely reflect the fact that organic compounds in sea salt aerosols are not accounted in the simulation [*Middlebrook et al.*, 1998; *Cavalli et al.*, 2004].

Conclusions and Discussion

1. New aging mechanism for carbonaceous aerosols in the GEOS-Chem model has been implemented where the hydrophobic to hydrophilic conversion is affected by O_3 concentration and humidity;
2. The updated aging mechanism has significant impacts on the model simulations of carbonaceous aerosols, with the largest effects found for the tropical regions and upper troposphere;
3. The updated aging mechanism leads to increases in simulated global burden of BC and OC by 30.8% and 16.6%, respectively;
4. There still remain low model bias for GEOS-Chem simulated OC over large areas with the updated aging scheme which point to other contributing factors such as emission inventories, missing accounting of organic compounds in sea salt aerosols.

References

Capes, G., et al. (2008). "Aging of biomass burning aerosols over West Africa: Aircraft measurements of chemical composition, microphysical properties, and emission ratios." Journal of Geophysical Research-Atmospheres **113**(D20).

Cooke, W. F., et al. (1999). "Construction of a 1 degrees x 1 degrees fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model." Journal of Geophysical Research-Atmospheres **104(D18): 22137-22162.**

Poschl, U., et al. (2001). "Interaction of ozone and water vapor with spark discharge soot aerosol particles coated with benzo a pyrene: O3 and H2O adsorption, benzo a pyrene degradation, and atmospheric implications." Journal of Physical Chemistry A **105(16): 4029-4041.**

Maria, S. F., et al. (2004). "Organic aerosol growth mechanisms and their climate-forcing implications." Science **306(5703): 1921-1924**

Croft, B., et al. (2005). "Black carbon ageing in the Canadian Centre for Climate modelling and analysis atmospheric general circulation model." Atmospheric Chemistry and Physics **5: 1931-1949.**

Tsigaridis, K. and M. Kanakidou (2003). "Global modelling of secondary organic aerosol in the troposphere: a sensitivity analysis." Atmospheric Chemistry and Physics **3: 1849-1869.**

Middlebrook, A. M., D. M. Murphy, and D. S. Thomson (1998), Observations of organic material in individual marine particles at Cape Grim during the First Aerosol Characterization Experiment (ACE 1), *Journal of Geophysical Research-Atmospheres*, ***103*(D13), 16475-16483**.

Cavalli, F., et al. (2004), Advances in characterization of size-resolved organic matter in marine aerosol over the North Atlantic, *Journal of Geophysical Research-Atmospheres*, ***109*(D24)**.

Slide 1

sw1 It's not a big deal here, but for aerosols we generally use ug/m3 instead of ppb
sw, 11/27/2011