Sensitivity of Atmospheric Transport of Carbonaceous Aerosols to Aging Mechanism

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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 by 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.

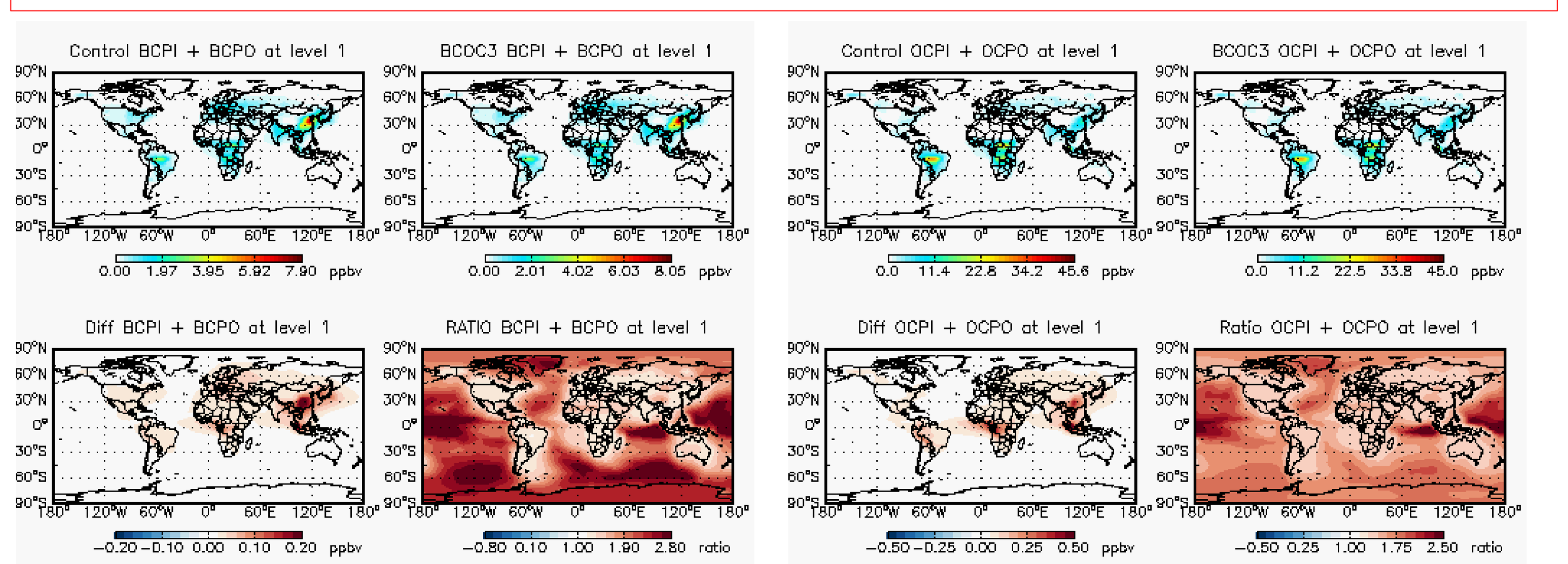
(1) A sensitivity study was carried out with the hydrophobic to hydrophilic conversion lifetime of carbonaceous aerosol increased by a factor of 3 (from 1.15 to 3.45 days) based on Maria et al., [2004]. The simulation results for BC and OC are compared with those from the control run;

(2) 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;

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

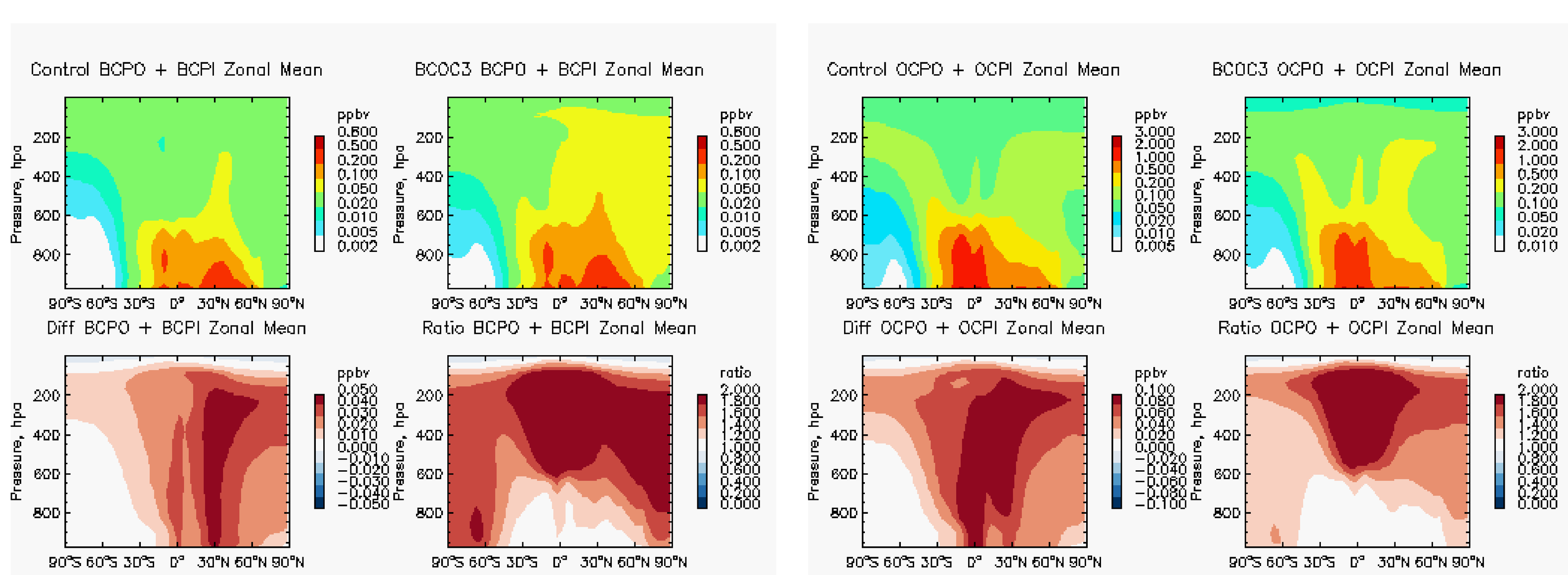
Results

1. Sensitivity study (hydrophobic to hydrophilic conversion lifetime increased by a factor of 3)



Control : lifetime (1.15 days); BCOC3: lifetime (3.45 days); BCPI: Black Carbon Hydrophilic; BCPO: Black Carbon Hydrophobic; OCPI: Organic Carbon Hydrophilic; OCPO: Organic Carbon Hydrophobic.
Diff: BCOC3 – Control; Ratio: BCOC3/Control.

Significant increases in atmospheric concentrations of both black carbon (BCPI + BCPO) and organic carbon (OCPI+OCPO) are calculated with the increased hydrophobic to hydrophilic conversion lifetime.



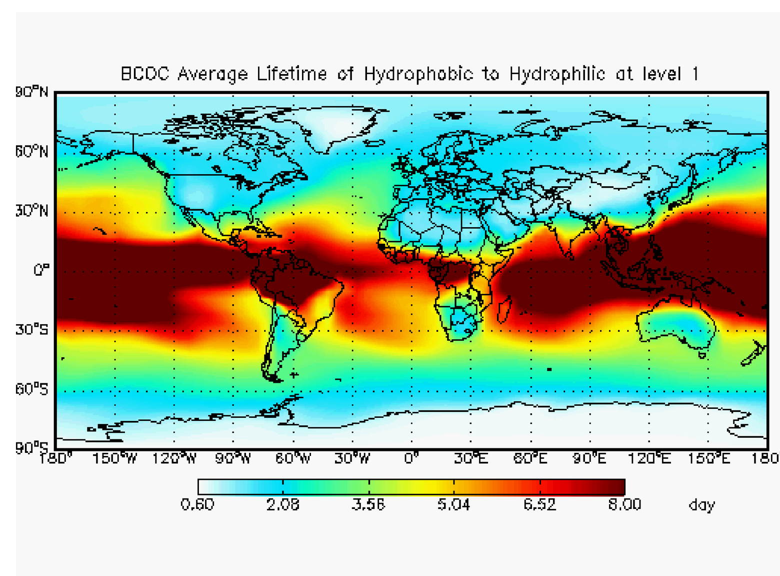
Zonal mean plots of the carbonaceous aerosol concentrations show larger perturbations in the upper troposphere than the lower troposphere.

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

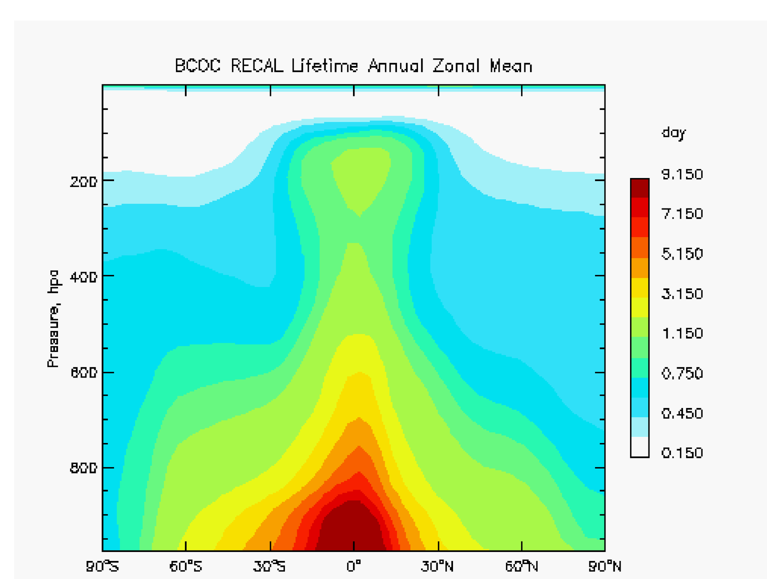
$$\tau = \frac{1 + K_{O_3}[O_3] + K_{H_2O}[H_2O]}{K_\infty K_{O_3}[O_3]}$$

[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.

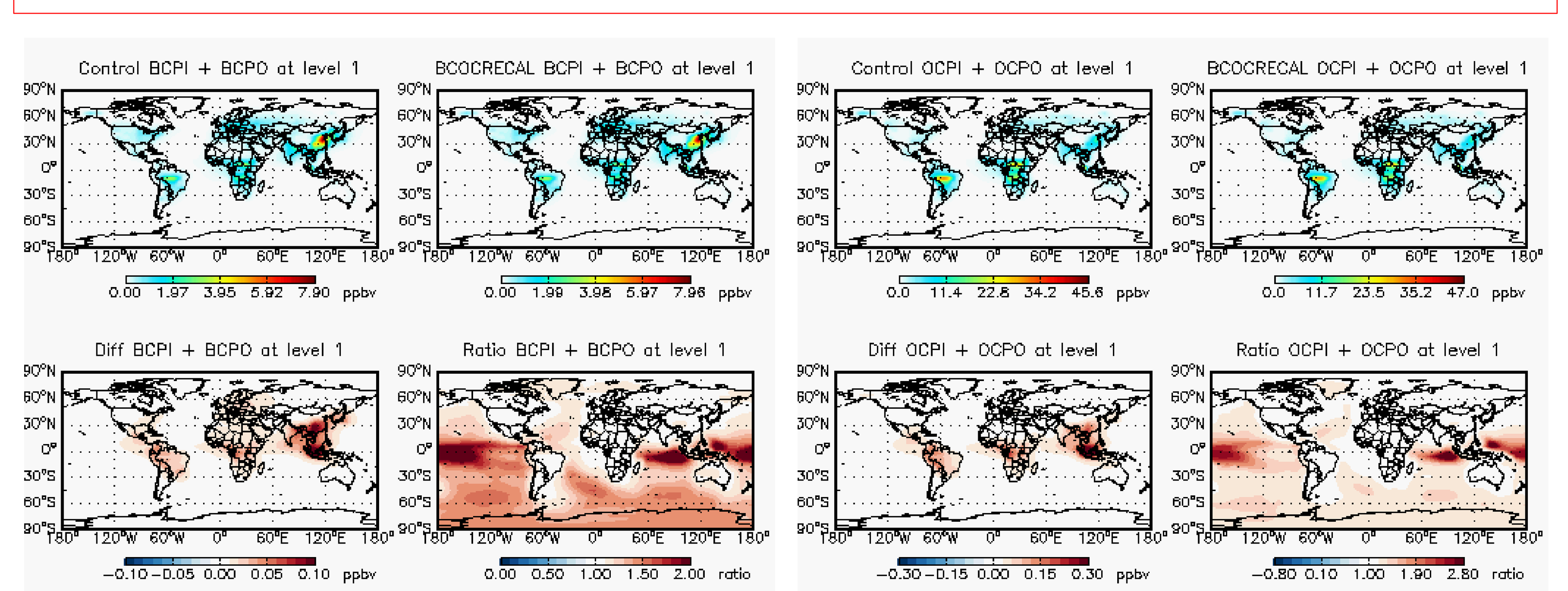


With the updated aging mechanism, the global area-weighted average hydrophobic to hydrophilic conversion lifetime of carbonaceous aerosols in surface air is around 5.4 days, with global volume-weighted average hydrophobic to hydrophilic conversion lifetime approximately 4.3 days. The hot spots in the tropical areas reflect the low ozone concentration and high water vapor there, with the longest lifetime calculated at surface level over the Amazon forest (around 40 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.

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



(1) Increases in model simulated concentrations of both BC(by up to 0.33 ppb) and OC (by up to 1.4ppb) are calculated;
(2) The largest impacts are found for tropical areas.

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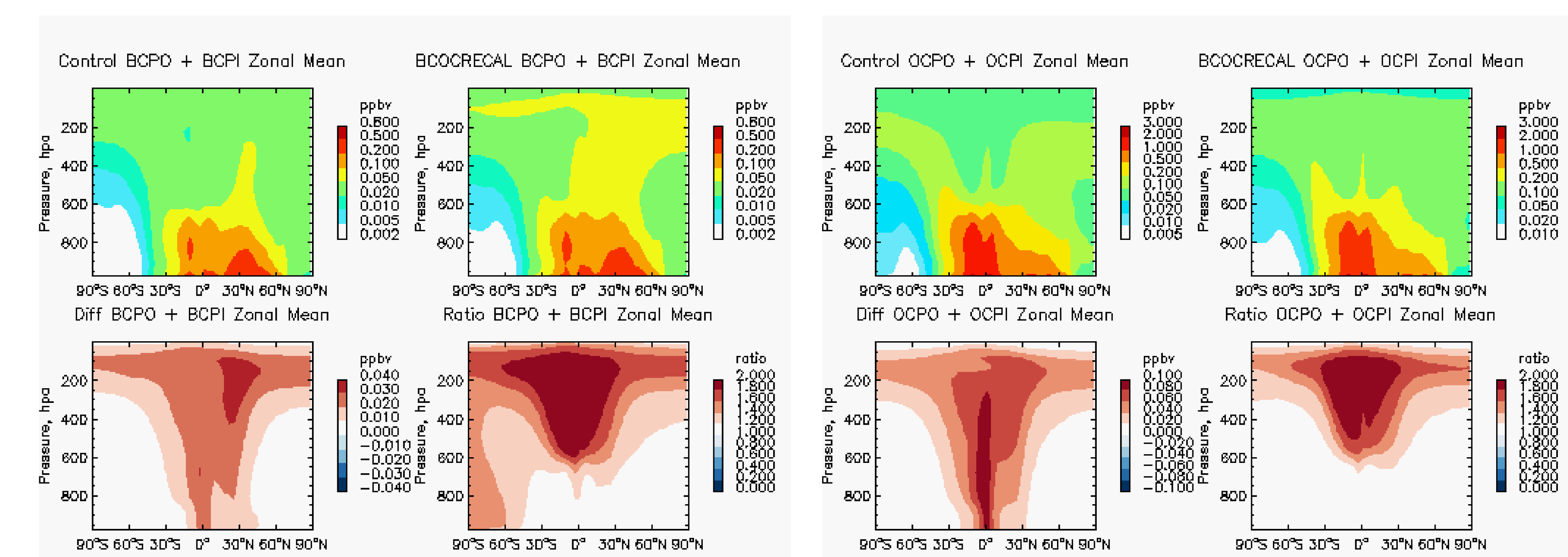

The largest effects are found for tropical upper troposphere, where the carbonaceous aerosol concentrations more than double with the updated aging mechanism.

Conclusions

1. We have implemented a new aging mechanism for carbonaceous aerosols in the GEOS-Chem model where the hydrophobic to hydrophilic conversion is affected by local conditions such as O_3 concentration and humidity;
2. The simulated hydrophobic to hydrophilic conversion of carbonaceous aerosols exhibits large spatial and temporal variation;
3. 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;
4. The updated aging mechanism leads to increases in simulated burden of BC and OC by 30.8% and 16.6%, respectively.

References

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