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.

Results

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

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

\[ \tau = \frac{1}{K_O \cdot \left[ O_3 \right] + K_H \cdot \left[ H_2O \right]} \]

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

 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.

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

With the updated aging mechanism, the global area-weighted average hydrophobic to hydrophilic conversion 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.

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 O3 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.

Acknowledgement: This work was supported by NIH (grant #1 RC1 ES018612). We thank Mark Weis at Michigan Tech for help on programming work.

References


