Improving Black Carbon (BC) Aging in GEOS-Chem Based on Aerosol Microphysics: Constraints from HIPPO Observations

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BC: Second most important human emission in terms of its radiative forcing in the current atmosphere.

atmospheric brown clouds in a mixture with other aerosols, emissions of black carbon are the second strongest contribution to current global warming, after carbon dioxide emissions. In the Himalayan region, solar heating from black carbon at high elevations may be just as important as carbon dioxide in the melting of snowpacks and glaciers. The interception of solar radiation by atmospheric brown clouds leads to dimming at the Earth’s surface with important implications for the hydrological cycle.

End of the Little Ice Age in the Alps forced by industrial black carbon

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Motivations

- We seek to investigate the effects of microphysics on BC aging.

- BC aging timescale significantly affects atmospheric BC concentration, deposition, and thus its radiative forcing [e.g, Liu et al., 2011; Huang et al., 2013; Bond et al., 2013].

- Currently, GEOS-Chem uses a fixed aging e-folding time of 1.15 days globally. This is not realistic and lacks microphysical information associated with BC aging.

\[
\left( \frac{\partial [BC_{phob}]}{\partial t} \right) = -\frac{[BC_{phob}]}{\tau_{BC}} \quad \left( \frac{\partial [BC_{phil}]}{\partial t} \right) = \frac{[BC_{phob}]}{\tau_{BC}} \quad \tau \sim 1 \text{ days}
\]

- However, observations suggested that the e-folding aging time of ~1 day underestimates BC aging rate in urban areas and biomass burning areas [e.g., Moffet and Prather, 2009; Akagi et al., 2012].

- GEOS-Chem overestimated BC concentrations over remote Pacific Oceans compared to HIPPO observations [Wang et al., 2014], which suggested that BC aging may be too slow in the model in addition to the less efficient wet scavenging.
BC aging microphysics and implementation in GEOS-Chem

BC aging process:

\[
\left( \frac{dm_{BCPO}}{dt} \right)_{\text{total}} = \left( \frac{dm_{BCPO}}{dt} \right)_{\text{cond}} + \left( \frac{dm_{BCPO}}{dt} \right)_{\text{coag}} + \left( \frac{dm_{BCPO}}{dt} \right)_{\text{chem}}
\]

Total aging rate  Condensation  Coagulation  Chemical oxidation
1. **Condensation of soluble species** (SO$_4$, NO$_3$, NH$_4$, SOA)

(1) **Condensation rate** ($J$) on single BC particle:  
\[ J_t = 4\pi f(K_n, \alpha)R_{BC}D_A(c_\infty - c_s) \]  
[Seinfeld and Pandis, 2006]

(2) **Implementation in GEOS-Chem:**

1. Condensed mass of soluble species A on hydrophobic BC (BCPO) in unit time:

\[ k_A = \frac{d(m_{BCPO,cond}^A)}{dt} = \frac{N_{BCPO}f_{BCPO}(K_n, \alpha)R_{BCPO}}{\sum N_i f_i(K_n, \alpha)R_i} \sum 4\pi f_i(K_n, \alpha)R_iD_A[A]^g_{\text{total}}M_A \]

Fraction partitioned to BCPO  
(condensation rate, surface area)

Total mass of condensed sulfate, nitrate and SOA  
(newly formed mass at each model timestep)

Aerosol mass conc. -> number conc. :  
(assuming log-normal size distribution)

\[ N_{aer} = \frac{M_{aer}}{\rho_{aer}} \left( \frac{\pi}{6} D^3 \exp \left( \frac{9}{2} \ln^2 \sigma_g \right) \right)^{-1} \]

2. Fraction of BCPO becoming hydrophilic BC (BCPI) in $\Delta t$:

\[ F_{BCPO \rightarrow BCPI} = -\frac{\beta m_{BCPO}}{R_{BCPO}A} \]

Mass fraction of soluble species in coated BC  
required for BCPO -> BCPI  
(e.g., $\beta = 5\%$ [Riemer et al., 2004] -> supersaturation of 0.22% (Kohler theory)).

3. **Condensation equation:**

\[ \left( \frac{dm_{BCPO}}{dt} \right)_{\text{cond}} = -\frac{F_{BCPO \rightarrow BCPI} m_{BCPO}}{\Delta t} = -\frac{k_A}{\beta} \]
2. Coagulation with sulfate, nitrate, OCPI (w/ SOA), BCPI, seasalt

(1) Coagulation rate \( J_{BC,M} \) between BC and particle M: 
\[
J_{BC,M} = \beta K_{BC,M} N_{BC} N_M
\]

Correction factor \_ Coagulation coefficient \_ Number concentration

(2) Implementation in GEOS-Chem:

assuming BCPO-> BCPI once coagulation occurs:

\[
\frac{dN_{BCPO,j}}{dt}_{coag} = -\sum_{i,j=1}^{\infty} \beta K_{BCPO,i} N_i N_{BCPO,j}
\]

3. Chemical oxidation on BC surface

Parameterization from lab experiments

\[
\frac{d[BCPO]}{dt}_{chem} = -k_{chem} \times [BCPO]
\]

\[
k_{chem} = \frac{\gamma K_\infty K_{O_3} [O_3]}{1 + K_{O_3} [O_3] + K_{H_2O} [H_2O]}
\]

[Poschl et al., 2001]
Model simulations and observations

- **GEOS-Chem v9-01-03 full chemistry with SOA simulation**
  Meteorological fields: GEOS-5
  Spatial resolution: $2^\circ \times 2.5^\circ$, 47 vertical layers
  Simulation year: 2009-2011 (corresponding to HIPPO campaigns)

- **Global BC emissions**:
  Anthropogenic: PKU BC inventory for 2008 [R. Wang et al., 2014, PNAS]
  Biomass burning: GFEDv3 inventory w/ small fires [Randerson et al., 2012]

- **Dry deposition**:
  Standard schemes from Wesely [1989] and Q. Wang et al. [2011]

- **Wet deposition**:
  Standard schemes from Liu et al. [2001] and Q. Wang et al. [2011]
  Homogeneous & heterogeneous freezing nucleation for cold clouds following Q. Wang et al. [2014]

- **Observations**:
  HIPPO1-5 aircraft campaigns [Wofsy et al., 2011; Schwarz et al., 2010, 2013]

- **Model-Observation comparison**:
  Model results are sampled along the flight tracks
  Observations are averaged over the GEOS-Chem grid and time step of 15min
Microphysics scheme improves modeled PDF and median of BC concentration, particularly reducing model overestimates at lower values.
BC vertical distributions

- Fixed aging scheme tends to overestimate BC concentrations at all altitudes by a factor of 2-5 within different latitude ranges.
- Microphysics scheme reduces the discrepancies by up to a factor of 2.
• Microphysics scheme reduces the model-observation discrepancies by decreasing BC column concentrations at all latitudes throughout different seasons, particularly over tropical regions.
On-going work

- **Detailed analysis** on model results compared with HIPPO observations

- **Quantifying uncertainties** associated with our BC aging scheme:
  - sensitivity to key microphysical parameters
    (e.g., size distribution, condensation/coagulation coefficients, …)

- Comparison with previous BC aging parameterizations:
  1. Riemer et al. [2004]: \[ \tau = \frac{a}{N} + \frac{b}{N^2} \]
  2. Liu et al. [2011]: \[ k_a = \beta \cdot [OH] + \delta \]
  3. Oshima et al. [2013]: \[ \tau_{BC} = \frac{A(D_m, \sigma)}{V_{BC}} \quad V_{BC} = \frac{1}{[BC_{phob}]} \frac{\Delta[M_{BCphob}]}{\Delta t} \]

- Effects of new BC aging scheme on **global BC burdens, lifetime, and radiative forcing**
THANK YOU!
Observations:

Model - Observation: