Global simulation of brown carbon and its direct radiative forcing

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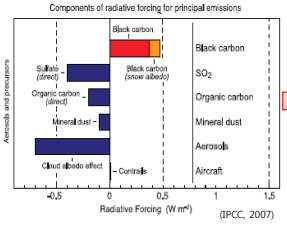
1. MOTIVATION



(IPCC, 2007)

Chemistry models simulate two kinds of carbonaceous aerosols (CAs)

- Black carbon (BC) → Absorption > Scattering → Warming
- Organic carbon (OC) → Absorption < Scattering → Cooling

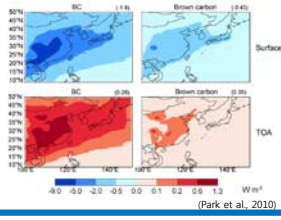
➢ **But recent studies have shown that some OC can also absorb solar radiation – called brown carbon (BrC)**

✓ Kirchstetter and Thatcher (2012) → calculated that BrC would account for 14% of total solar absorption by CA, furthermore, BrC would contribute 49% of solar absorption by CA below 400 nm

✓ Bahadur et al. (2012) → estimated that BrC absorption at 440 nm is about 40% of the BC, whereas at 675 nm it is less than 10% of BC by using aerosol optical properties from AERONET

✓ Chung et al. (2012) → found that OC contributes about 45% to CA absorption at 520 nm by analyzing observation data collected at the Gosan (East Asia)

Previous modeling studies about BrC



(Park et al., 2010)

- Jacobson (2001) assumed that 10% of OC strongly absorb radiation and this assumption results in an increase in the global direct radiative forcing by 0.03-0.05 W m^{-2}.
- Park et al. (2010) estimated BrC concentrations in East Asia using the mass ratio of BrC to BC and resulting direct radiative forcing of BrC was 0.05 W m^{-2}.

★ **No models explicitly simulate BrC yet** ★

2. OBJECTIVES AND MODEL DESCRIPTION

Objectives

- **Explicitly simulate BrC using a chemical transport model**
- **Examine BrC budgets and its contribution to OC**
- **Calculate global direct radiative forcing (DRF) of BrC**

Model description

- GEOS-Chem version 9.1.2 with 2°x2.5° resolution
- Meteological field : MERRA
- Simulation period : 2007, 2010
- Fossil fuel and biofuel OC emissions from Bond et al. (2007)
- Apply annual and monthly scale factors of MACCity inventory for biofuel emissions.
- Biogenic emissions from MEGAN v2.1
- Biomass burning emissions from FINN
- Hydrophilic BrC fraction is assumed to be 50% (identical to the OC simulation)

- Carry 5 Brown carbon species as follows:
  → Hydrophilic BrC from biomass burning
  → Hydrophobic BrC from biomass burning
  → Hydrophilic BrC from biofuel
  → Hydrophobic BrC from biofuel
  → Hydrophilic BrC from secondary organic aerosol

Observations

- One year absorption measurements at 365 nm at 15 sites in the Southern US, (Hecobian et al., 2010)
- Observations in LA and Atlanta (Zhang et al., 2010; Zhang et al., 2013)

3. METHODOLOGY

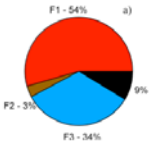
What are the main sources of BrC?

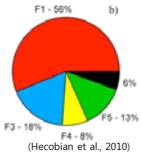
1. Biomass burning → Especially from smoldering combustion (low temperature, incomplete burning)

(Clarke et al., 2007; Lukacs et al., 2007; Favez et al., 2009; Hecobian et al., 2010; Hoffer et al., 2006; Arola et al., 2011; McMeeking, 2008; Kirchstetter et al., 2004; Kirchstetter and Thatcher, 2012; Chen and Bond, 2010)

2. Secondary organic aerosol (SOA) → Especially from aromatic species

(Jaoui et al., 2008; Nakayama et al., 2010; Updyke et al., 2012; Zhang and Jang, 2011, Nakayama et al., 2013; Nguyen et al., 2012; Laskin et al. 2010)





(Hecobian et al., 2010)

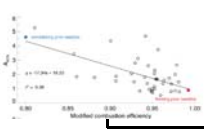
From positive matrix factorization analysis, Hecobian et al. (2010) showed that biomass burning was the most dominant sources of BrC (55%), followed by SOA (26-34%) over the southeastern US in 2007.

F1 → Biomass burning
F3, F4 → Secondary organic aerosols (SOAs)
F5 → Mobile sources

Estimation of BrC from biomass burning

- **There is no emission inventory for BrC → Alternatively, we have to use emission of BC or OC**
- **Possible linkage → We used the relationship between absorption angstrom exponent (AAE) of CA and modified combustion efficiency (MCE) of biomass burning (McMeeking, 2008)**

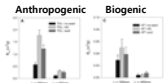
Related with BrC/BC light absorption ratio at specific wavelength ←



→ Can be calculated from emission factors of CO_2 and CO for each vegetation type

Estimation of BrC from SOA

	Brown	Colorless
Source?	Anthropogenic (Aromatic)	Biogenic
Aging?	Aged	Fresh
Seed?	Sulfate seed	No seed



(Zhong and Jang, 2011)

(Jaoui et al., 2008)

For the first step, we consider all anthropogenic (aromatic) SOA as BrC

The relationship between AAE and MCE

Total absorption by carbonaceous aerosol is,

$$\alpha_{\lambda,CA} = \alpha_{\lambda,BrC} + \alpha_{\lambda,BC}$$

Using Angstrom exponent relationship,

$$\alpha_{\lambda_0,CA}\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{CA}} = \alpha_{\lambda_0,BrC}\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BrC}} + \alpha_{\lambda_0,BC}\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BC}}$$

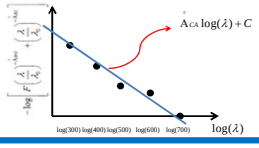
Divide by absorption of BC at λ0

$$(1+F)\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{CA}} = F\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BrC}} + \left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BC}}$$

Take a logarithm on both side

$$\text{Å}_{CA}\log(\lambda) + C = -\log\left[F\left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BrC}} + \left(\frac{\lambda}{\lambda_0}\right)^{-\text{Å}_{BC}}\right]$$

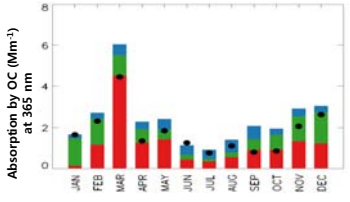
Where $C = \text{Å}_{CA}\log(\lambda_0) - \log(1+F)$



4. EVALUATION

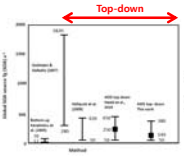
Annual observation – monthly mean data

The southeastern US (2007)

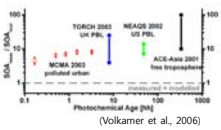


● Observation — From Biofuel — From Biomass Burning — From SOA

Applied mass absorption efficiency (MAE) of 5.3 to mass concentration in the model to compute BrC mass to light absorption



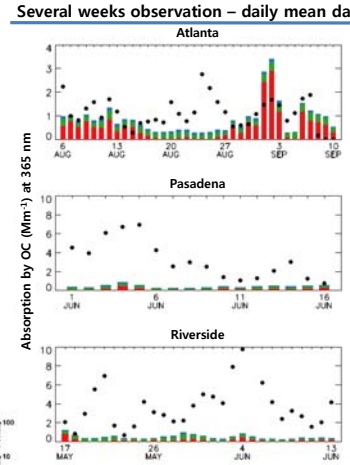
(Spracklen et al., 2011)



(Volkamer et al., 2006)

Chemistry models significantly underestimate SOA concentration (Volkamer et al., 2006; Spracklen et al., 2011)

Several weeks observation – daily mean data

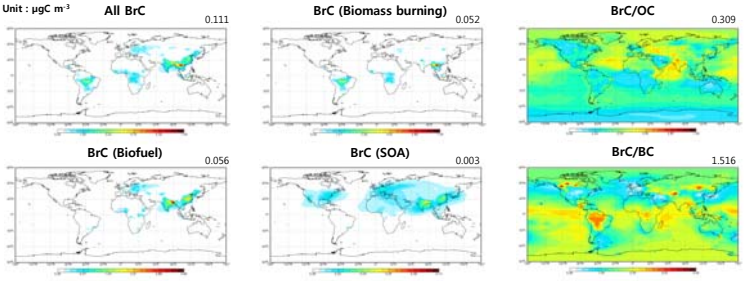


The model captures monthly variation well in the southeastern US, but underestimates the observations in LA basin (Pasadena and Riverside) likely due to the underestimation of BrC from anthropogenic SOA

Main sources of BrC in the Southeastern US and Atlanta → biomass burning (Hecobian et al., 2010)
Main sources of BrC in LA basin → anthropogenic SOA (Zhang et al., 2013)

5. RESULTS

Annual mean BrC concentration and BrC to CA ratios at the surface



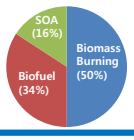
Tropospheric budget of BrC

Emission
11529 GgC/yr
- Biomass : 5676 GgC/yr
- Biofuel : 4333 GgC/yr
- SOA : 1520 GgC/yr

Burden
168 GgC
- Biomass : 87 GgC
- Biofuel : 60 GgC
- SOA : 28 GgC

Deposition
Wet : 9440 GgC/yr
Dry : 2094 GgC/yr
- Biomass : Wet : 4666 GgC/yr, Dry : 1012 GgC/yr
- Biofuel : Wet : 3444 GgC/yr, Dry : 891 GgC/yr
- SOA : Wet : 1330 GgC/yr, Dry : 191 GgC/yr

- **POC emission : 32254 GgC/yr**
- **BrC/BC : 1.63**
- **BrC/OC : 0.20**



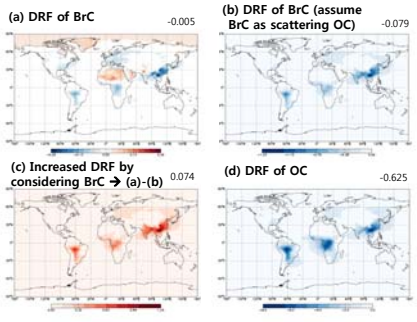
Lifetime
5.5 days
- Biomass : 5.6 days
- Biofuel : 5.0 days
- SOA : 6.6 days

- **Global mean surface concentration of BrC is 0.111 ugC m^{-3} (30% of OC concentration)**
- **Surface BrC concentrations from primary sources (biomass burning and biofuel) are a lot higher than those from secondary sources in the model**
- **BrC concentration peaks occur mainly in heavy biomass burning region**
- **Global tropospheric burden of BrC is 168 GgC, which takes 20% of that of OC**
- **Lifetime of BrC is 5.5 days, which is within the range (4.3-11 days) of OA lifetime of the AeroCom multi model study (Textor et al., 2006)**

6. DRF of BrC

Global DRF of BrC at TOA

Unit : W m^{-2}



- **Mean global DRF of BrC is -0.005 W m^{-2}, close to 0 (imaginary refractive index k = 0.1 at 550 nm is used, from Mcmeeking, 2008)**
- **If we consider BrC effect, then resulting DRF increased by BrC is 0.074 and it is even more important regionally → up to ~1 W m^{-2}**

7.SUMMARY

Conclusions

- This study is the first attempt to simulate BrC explicitly in the model
- BrCs from primary sources (biomass burning and biofuel) are well simulated
- The model significantly underestimates BrC from secondary source (anthropogenic SOA) and it is caused by the underestimation of SOA in current global chemistry models
- Global tropospheric burden of BrC is 168 GgC, which accounts for 20% of OC
- Global mean direct forcing of OC is -0.625 W m^{-2}, and it is increased to 0.551 W m^{-2} when we consider BrC absorption
- Increased DRF by BrC is even more important regionally → up to ~1 W m^{-2}

Suggestions

- We have to obtain chemical structure of BrC in field observations
- We need to know emissions of BrC from various sources
- We have to know the quantitative relationship between BrC and SOA aging

Future Study Plan

- Sensitivity test for examining uncertainties of the model – especially for hydrophilicity and emission

Acknowledgement

- This study was financially supported by National Institute of Environmental Research (NIER)