Exploring the Observational Constraints on the Simulation of Brown Carbon

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Introduction

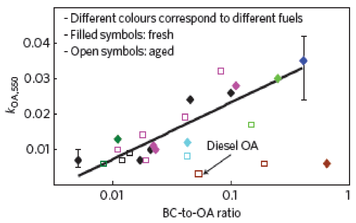
Brown carbon (BrC), is the component of organic aerosols (OA) which strongly absorbs solar radiation in the near-UV range of the spectrum. However the source, evolution, and optical properties of BrC remain highly uncertain.

In this study, we develop a GEOS-Chem simulation of BrC and test it against BrC absorption measurements from two aircraft campaigns in the U.S (SEAC4RS and DC3). To our knowledge, this is the first study to compare simulated BrC absorption with direct, continuous measurements. We explore how assumptions for BrC impact the comparisons with these observational constraints and estimate the resulting global direct radiative effect of BrC.

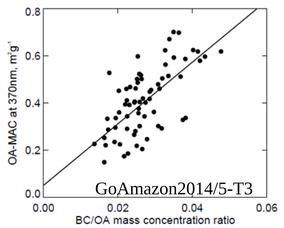
Model Assumptions for BrC

Absorption Properties

We assume biofuel and biomass burning OA are absorbing. The absorption properties are related to emitted BC/OA ratio, which results an MAC of $0.70 m^2 g^{-1}$ for biofuel OA and $0.75 m^2 g^{-1}$ for biomass burning OA. Such relationship is confirmed by both laboratory experiments and field measurements:



(Saleh et al., 2014)



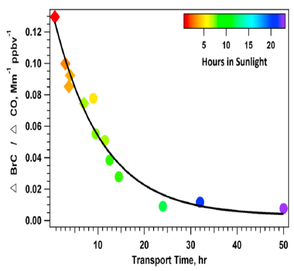
(Wang et al. 2016)

A "Whitening" Scheme

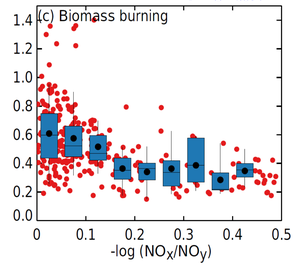
We assume that the absorption of OA decreases at a rate related to OH:

$$Abs_{BrC,\,t+\Delta t} = Abs_{BrC,\,t} \cdot \exp\left(\frac{[OH]\cdot\Delta t}{5\times10^{-5}}\right)$$

Such a "whitening" process is observed in field measurements of biomass burning plumes:



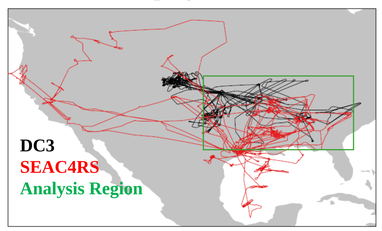
half-life: 9-15 hours
(Forrister et al., 2015)

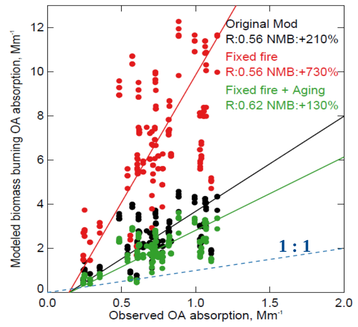


half-life: 9-15 hours
(Wang et al., 2016)

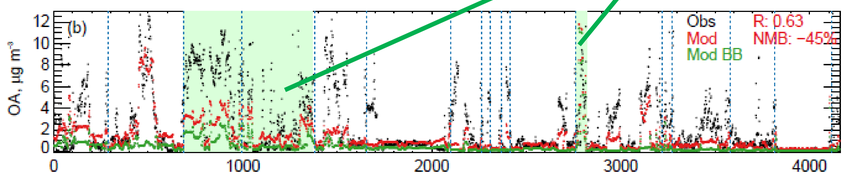
Model vs. Observations

Direct, continuous measurements of OA absorption during DC3 and SEAC4RS campaigns.

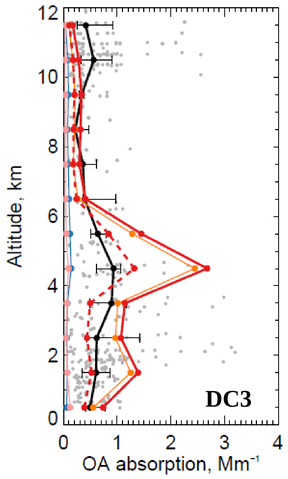


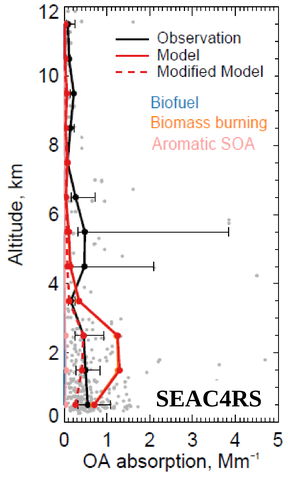


Biomass burning plumes



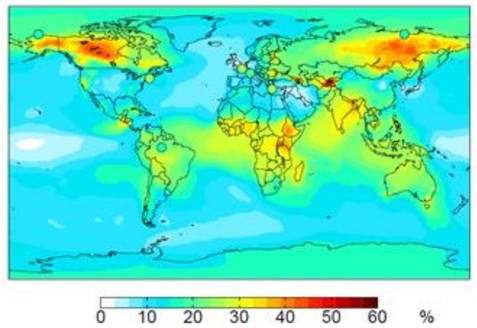
Minute of DC8 flight during DC3





The "whitening" scheme increases the correlation between modeled and observed OA absorption. The observationally constrained initial biomass burning OA-MAC for the model simulation is $0.35 m^2 g^{-1}$ at 365nm, $0.2 m^2 g^{-1}$ at 440nm and $0.09 m^2 g^{-1}$ at 550nm with the "whitening" process.

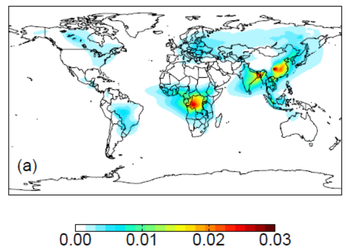
When comparing simulated BrC surface absorption contribution to 8 surface sites with Aethalometer measurements (*Wang et al., 2016*), the model is able to represent the BrC absorption contribution at many sites in other regions though the assumption is constrained by US measurements only.



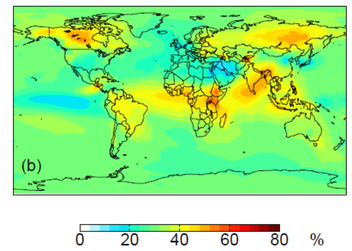
Global distribution of BrC absorption contribution to total aerosol absorption at surface

BrC AAOD and DRE

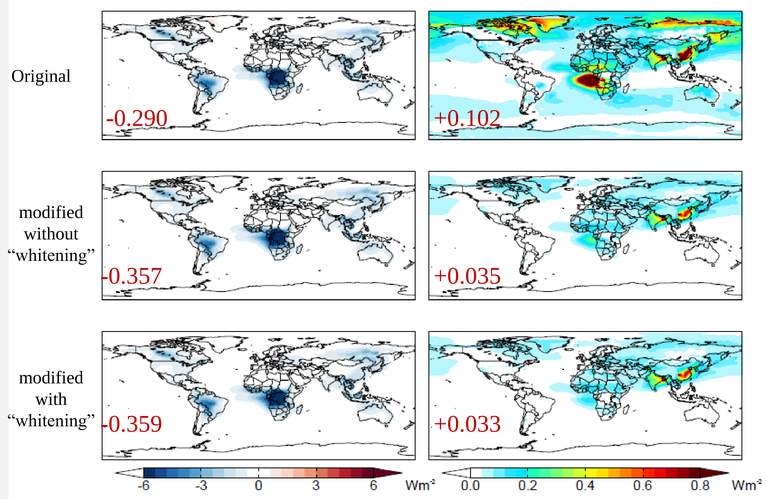
BrC-AAOD at 440nm



Contribution of BrC-AAOD to total AAOD at 440nm



Annual Mean Direct Radiative Effect



Conclusions

- The laboratory-based biomass burning BrC absorption is biased high when compared to aircraft measurements.
- Applying a "whitening" scheme is better able to represent the observed BrC absorption in the model.
- Based on our analysis, we suggest that the initial OA-MAC for biomass burning for model simulation should be $0.35 m^2 g^{-1}$ at 365nm, $0.2 m^2 g^{-1}$ at 440nm and $0.09 m^2 g^{-1}$ at 550nm.
- The global all-sky absorption DRE of BrC is estimated to be $+0.033\ Wm^{-2}$, which is lower than all previous modeling studies (none of which has been evaluated against direct observations).