A global simulation of brown carbon aerosol and its implications

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Recent observations suggest that a certain fraction of organic carbon (OC) aerosol effectively absorbs solar radiation, which is also known as brown carbon (BrC) aerosol. Despite much observational evidence of its presence, very few global modeling studies have been conducted because of poor understanding of global BrC emissions.

We will present our explicit global simulations of BrC in a global 3-D chemical transport model (GEOS-Chem). In order to perform the model simulations, we estimate global primary BrC emissions from open burning and biofuel use based on a reported relationship between absorption Ångström exponent (AAE) and modified combustion efficiency (MCE) (McMeeking, 2008) as shown in Figure 1. Our estimated global BrC emissions are 3.9 ± 1.7 and 3.0 ± 1.3 TgC yr^{-1} from biomass burning and biofuel sources, respectively, and 5.7 TgC yr^{-1} from aromatic oxidation. Table 1 summarizes our estimated MCE, OC and BC emission factors, and BrC/OC ratios for each vegetation type. Using the estimated primary and secondary emissions of BrC, we conducted a full year simulation for 2007 to obtain the global distribution of BrC concentrations.

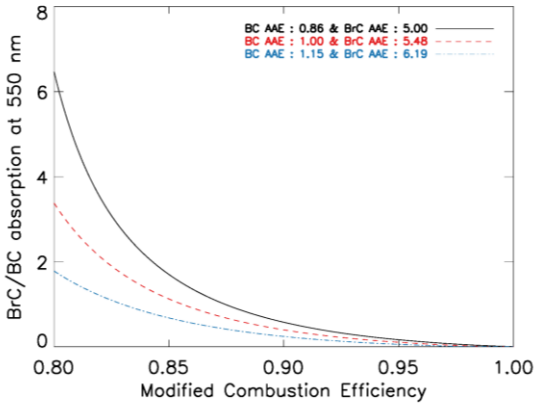

Figure 1. Estimated absorption ratios of BrC to BC at 550 nm as a function of MCE. We assume that the CA absorption is only contributed by BC and BrC absorption. Each line shows a relationship based on different BC and BrC AAE values.

We evaluate the model by comparing the simulated results with observed absorption by OC in surface air in the United States, and with single scattering albedo observations at AERONET sites all over the globe. The model successfully reproduces the observed seasonal variations, but underestimates the magnitudes, especially in regions with high secondary source contributions.

Table 1. Emission factors (EFs) and calculated parameters used for primary BrC emission estimates. Biomass burning emission is classified for six vegetation types based on the FINN inventory. Here BrC/OC is the mass ratio of BrC to OC emitted from biomass burning and biofuel use.

Source Type	**MCE**	**OC EF [g kg^{-1}]**	**BC EF [g kg^{-1}]**	**BrC/OC**
Boreal Forest	0.891	7.8	0.20	0.093
Cropland	0.898	3.3	0.69	0.652
Savanna/Grassland	0.948	2.6	0.37	0.123
Temperate Forest	0.910	9.2	0.56	0.145
Tropical Forest	0.919	4.7	0.52	0.213
Woody Savannah/Shrubland	0.941	6.6	0.50	0.081
Biofuel[1)]				0.452

Our global simulations show that BrC accounts for 21% of the global mean OC concentration, which is typically assumed to be scattering. We find that the global direct radiative effect of BrC is nearly zero at the top of the atmosphere, and consequently decreases the direct radiative cooling effect of OC by 16%. In addition, the BrC absorption leads to a general reduction of NO_2 photolysis rates, whose maximum decreases occur in Asia up to -8% (-17%) on an annual (spring) mean basis. The resulting decreases of annual (spring) mean surface ozone concentrations are up to -6% (-13%) in Asia, indicating a non-negligible effect of BrC on photochemistry in this region.

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McMeeking, G. R.: The Optical, Chemical, And Physical Properties Of Aerosols And Gases Emitted By The Laboratory Combustion Of Wildland Fuels, Dissertation, Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado Fall 2008. Available at: http://chem.atmos.colostate.edu/Thesis/McMeeking%20dissertation.pdf