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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A global simulation of brown carbon: implications for photochemistry and direct radiative effect" *by* D. S. Jo et al.

Anonymous Referee #2

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This manuscript presents a global simulation of primary and secondary brown carbon (BrC) from biomass burning and biofuel emissions. The primary BrC emissions are related to different vegetation types through combustion efficiency based on a fire inventory, while secondary BrC formation is simulated by oxidation of anthropogenic aromatics. The predicted aerosol optical properties are compared with absorption measurements at surface and AERONET data. Direct radiative impact on photochemistry and shortwave radiation is examined with the inclusion of BrC absorption. The content is original and interesting to the community.

I have the following comments that need to be addressed before its consideration for publication:

1. The method that relates BrC absorption to MCE is largely based on McMeeking

(2008), which appears to be a thesis work not published. It would be helpful to provide a link to its electronic or print version for readers' reference since some information about the experiments referred to McMeeking (2008) might be important to understand the limitation and uncertainties of the method presented here. I asked if the Equation 2 from McMeeking (2008) assumes that all the OC from the biomass burning samples are absorbing BrC. The authors responded "No. . .because the colorless OC does not contribute to CAs absorption, the AAE of Equation 2 is only contributed by BrC and BC". I disagree with the second half of the statement. In fact, BC with clear coatings (i.e., non-absorbing OC) could result in AAE (380nm- 750nm) greater than 1.4 due to the lensing effect, as shown in Gyawali et al. (2009) and Lack and Cappa (2010). So can one assume AAE in Equation 2 due to BrC and BC only? If not, how would it affect the method used to drive the BrC emissions? Given the known large uncertainties associated with AAE, MAE for BrC and even BC, error bars representing uncertainties are certainly needed in Figure 1. Discussions on the propagated errors in global budgets of BrC are also needed.

Gyawali, M., Arnott,W. P., Lewis, K., and Moosmueller, H.: In Situ Aerosol Optics in Reno, NV, USA During and After the Summer 2008 California Wildfires and the Influence of Absorbing and Non-Absorbing Organic Coatings on Spectral Light Absorption, Atmos. Chem. Phys., 9, 8007–8017, 2009.

Lack and Cappa, Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmos. Chem. Phys., 10, 4207–4220, 2010.

2. In order to best-match the observed absorption at 365nm, 40% lower MAE values than those used to derive emissions have to be used for BrC optics in aerosol optical comparison and radiative transfer calculations. Are MAE values at other wavelengths lowered similarly? Can you use the low MAE values in obtaining the BrC/BC absorption ratio in emission estimating? Would it increase the BrC/OC fractions and primary BrC emissions estimated? The calculated BrC/OC fractions in Table 1 are much lower than

those used in previous studies except for cropland, but the estimated direct radiative effect due to BrC absorption is similar to others (∼0.12 Wm-2). Can you explain why? How do the model-predicted AAEs compare with the AERONET data, in addition to SSA at 440nm? The AAE comparison would give indications about the simulated BrC/OC or BrC/BC fractions, which presumably is one of the main improvements in this study compared with previous studies.

3. Description for biofuel BrC emission factors (Table S1) is still a bit confusing. How do the biofuel EFs relate to the MCE? based on the Fire inventories for open biomass burning? What is the overall BrC/OC fraction for biomass burning? Is it lower than biofuel? How do they compare with the BrC/OC fractions from SOC? Are there any laboratory studies that could support these estimates? I think that Table S1 should be included in the main text instead of the supplementary as biofuel is as important as biomass burning. In Table S1, the BrC/OC fraction of 0.663 is mistakenly placed in the row for BrC/Biofuel. Then the total burden of BrC from biofuel should be 0.663*6.281=4.16 Tg, not 4.3. Please also check other numbers in Table S1.

Minor comments:

1. Line 1, Page 27807: add "until recently" following ".. as light-scattering aerosols" 2. Line 3, page 27807: replace "climate effect" with "direct radiative forcing" 3. Line 7, page 27807: replace "shortwave" with "ultra-violet" 4. Line 2, page 27808: add "and also strongly absorbing" following "...HULIS" 5. Line 12, page 27810: attenuation means both scattering and absorption. Is it just absorption Angstrom exponent measured? 6. Line 19, page 27810: delete citation: McMeeking, 2008; isn't the equation 2 from McMeeking, 2008? 7. Line 25, page 27813: reference for the secondary BrC MAEs at 365nm and 550nm? 8. Appendix A1: line 17, page 27829: 4.1 for the 350-400nm pairing, while 8.0 for the 550-600nm pairing? Or it's reversed? 9. Appendix A1, lines 3-4, page 27830: this fitting is not clear to me. Because the slope variable (Angstrom for CA) also appears in the residual term C, which is the intercept B, can the obtained fitting function satisfy the slope (A) and the intercept (B) at the same time? It

would be nice to plot an example for the linear regression with one F value. 10. Line 5, page 27830: what wavelengths does the selected Angstrom for BrC correspond to?

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 27805, 2015.