Anonymous Referee #3:

This study applies a global model to study the change in direct radiative effect of including BrC as a special source of OC in the model. The treatment is reasonable, though comparisons with observations are not as good as with other models.

We thank the referee for careful reading and valuable comments.

1. Page 27808, lines 20 -26: you should also summarize results from Lin et al. 2014 (JGR, v. 119, doi:10.1002/2013JD021186) who found absorption from BrC ranging from 0.22 to 0.57 W/m2, though the net TOA forcing for the aerosols containing BrC was negative. This study did not assume a constant ratio of BrC to OC. The fraction depended on the type of OC and the specific emission source.

➔ ***We added and changed the text as follows:***

Lin et al. (2014) calculated the DRF of OC by assuming that all of the biomass burning and the biofuel OC are BrC, and all of the SOC (as a high-absorbing case) as BrC. They estimated the global clear-sky DRF of OC as -0.20 W m^{-2}.

2. Page 27812, lines 4,5: Since BrC absorption is variable, with variable sources, it is of interest to know how much your results would change if you used the MAE for SOC based on the WSOC measurements and the MAE observed by Alexander et al.

➔ ***We added the text in the evaluation section (Section 4.1) as follows:***

For example, applying the MAE value of 3.5 m^2 g^{-1} at 365 nm (a half of the MAE at 365 nm from Alexander et al. (2008)) for secondary BrC yields a similar mean absorption value to the observation over LA basin.

3. Page 27813, line 6,7: some SOCs become whiter when photolyzed (Sareen et al. 2013 Wu et al 2013]

➔ ***Thanks for the constructive comment. We added the text as follows:***

However, we note that some brown SOCs can be bleached when they undergo photodissociation (Zhong and Jang, 2011; Sareen et al., 2013). More detailed treatments of the chemical aging of BrC are needed in future BrC models.

4. Page 27813, line 24-25: the BrC MAE is 5.3 m2/g for primary and 1.5 m2/g for secondary BrC at 365nm. But “primary” is 1.0 at 550nm. For clarity, please specify the wavelength dependent MAE for both types: perhaps a figure would help.

➔ ***We modified the text as follows. We did not add the description of the MAE of secondary***

BrC because it is explained in Section 2.2.

In brief, we use the MAE values of 7.5 m^2 g^{-1} and 1.0 m^2 g^{-1} at 550 nm for primary BC and BrC, respectively. But at a shorter wavelength, higher MAE value was used for primary BrC (e.g., 5.3 m^2 g^{-1} at 365 nm as discussed in Section 4).

5. Page 27816, line 7-9: "Model slightly underestimates": how is this comparison made (difficult to see on scatter plots). Perhaps you could fit a line to these points?

➔ ***We included regression lines in Figure 2.***

6. Page 27816, line 9-11: here you quote annual mean concentrations, but the scatter plot shows values at different locations. How is the annual mean (for the US I assume) computed? i.e. if you use grid box values, do all grid boxes within the US have measurements?

➔ ***We added the text as follows for the clarity:***

We calculate the annual mean concentrations of the model using the simulated values of model grid boxes corresponding to the IMPROVE network sites.

7. Page 27821, line 6-9; Lin et al 2014, also find little bias compared to these simulations. What is your total biomass emissions? How do these compare with other estimates such as used in Lin et al. 2014 (JGR)?

➔ ***Thanks for the constructive comment. We added the text as follows:***

Lin et al. (2014) also reported a small bias in their model compared with AERONET SSA using 4.7 Tg yr^{-1} of global annual biomass burning BC emissions, which is about two times higher than 2.2 Tg yr^{-1} of this study.

8. Page 27824, lines 6-10: seems odd to say you use McMeeking, and then you decrease these values. Need to revise so it does not sound as if you are using 2 values, or clarify that you use 2 values.

➔ ***Thanks for the constructive comment. We conducted additional simulations with three different absorption ratios of BrC to BC as shown in Figure 1 below. We removed our discussion of the use of 40% reduced MAE value, and instead added discussions with new simulations as follows:***

Section 2.1:

Figure 1 shows the estimated BrC/BC absorption ratio at 550 nm as a function of MCE. Different lines indicate different AAEs of BC and BrC according to the Table 1 of Kirchstetter and Thatcher (2012). They calculated BC AAE and BrC AAE using 115 wood smoke samples. For the calculation of BrC AAE, BC AAE had to be decided, and they assumed three different BC AAEs (0.86, 1.00, 1.15) based on their smoke samples and previous studies. Resulting BrC AAEs were 5.00, 5.48, and 6.19.

We conduct three simulations according to the AAE values of Kirchstetter and Thatcher (2012) (Figure 1), as described later in this section.

Section 4.1:

Figure 3 shows monthly mean simulated and observed light absorption coefficients of BrC at 365 nm averaged over all sites in the southeastern United States for 2007. Black circles and colored bars indicate the observed and simulated BrC absorption at 365 nm, respectively, and different colors in the bar show contributions from different sources. Each panel represents different model simulations with each case for AAE selections as shown in Figure 1 and Table 1.

Section 4.1:

The model generally captures the observed seasonal variation with high absorption in the winter, having a peak in March and low absorption in the summer (R of 0.93). On an annual mean basis, we find that the model is too high by 46% for case 1, and is too low by -31% for case 3, relative to the observations, respectively. The model for case 2 is in the best agreement with the observations (4%) on an annual mean basis.

Section 4.1:

The BrC source contribution in the model is similar to the observed source contribution. Hecobian et al. (2010) showed that biomass burning was the main contributor for the winter season, whereas the SOC contribution increased during the summer season. The simulated seasonal variation is consistent with the observation, as shown in Figure 3. The annual mass contribution of SOC to BrC is 38% (in case 2), which is in good agreement with the observed contribution of 32% (Hecobian et al., 2010). Based on the results in Figure 3, the model for the case 2 yields best estimates of BrC emissions.

Section 5.2:

Table 3 summarizes our best estimates of the global tropospheric budgets of BrC, along with BC and OC. The global BrC source is 12.5 ± 3.0 TgC yr^{-1}, which accounts for 27% of OC sources. Although the biofuel emission (6.5 TgC yr^{-1}) is three times lower than the biomass burning emission (22.7 TgC yr^{-1}) for OC, the biofuel emission (3.0 ± 1.3 TgC yr^{-1}) becomes significant for BrC, contributing about 43% of primary sources. The secondary source of BrC is 5.7 TgC yr^{-1}, and is comparable to the primary sources (6.8 ± 3.0 TgC yr^{-1}).

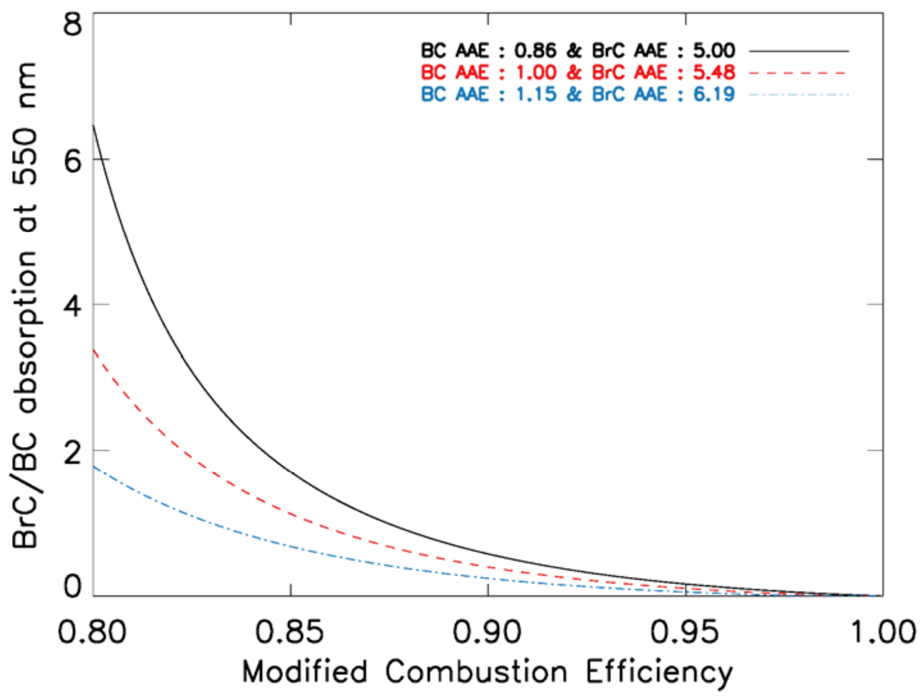

Figure 1. Estimated absorption ratios of BrC to BC at 550 nm as a function of MCE. We assume that the CA absorption is only contributed by BC and BrC absorption. Black solid line indicates case 1, red dashed line represents case 2, and blue dotted line shows case 3.

9. Page 27851, Figure 8 caption: What exactly is panel c? BrC is assumed to be OC and purely scattering minus the normal BrC calculation? Need to rephrase. I understand these positive values must be from a calculation including BrC with absorption minus a calculation assuming all BrC is purely scattering.

➔ ***Yes, you are correct that it is a relative change of BrC DRE from scattering BrC to absorbing BrC. We rewrote the figure caption for clarity. Note that total OC concentration does not change after we simulate BrC (i.e. we estimates BrC sources independently, which should be part of OC sources).***

The DRE increase of OC owing to the absorption of BrC is shown in (c) (i.e. the DRE of OC with absorbing BrC minus the DRE of OC including BrC as scattering OC, which is typically assumed in previous studies).