Referee #1

Thank you for your helpful comments. We revised the manuscript based on your comments and suggestions.

In this study, the authors implement a brown carbon scheme into a global model, evaluate a series of simulations with varying assumptions about photobleaching and convective transport against observations from HIPPO, SEAC4RS and DC3, and then estimate the heating rates and DREs from their simulation which best fits observations. This is certainly an interesting topic which warrants further modeling studies to explore the impacts of large uncertainties in our understanding of the properties and evolution of BrC in the atmosphere. There are however two major issues in this manuscript that should be addressed prior to publication (as well as more minor issues described below):

Author's response: Thank you for your suggestions. The point-by-point responses to your questions and comments are presented below.

1. This modeling study shares significant methodological overlap with previous modeling efforts for BrC, particularly Wang et al, ACP, 2018 and Brown et al., ACP, 2018, the first of which compares to the same BrC observations used here and the second of which uses the same model. Both of these previous model studies explore photobleaching. The primary novelty of this study is therefore the focus on convective transport. The authors should therefore be careful not to overstate the novelty of their work, and acknowledge and contrast to the existing literature throughout (Introduction, Results, Conclusions), particularly how assumptions made in this study might differ from these previous studies, why different assumptions might have been implemented, and how this impacts the comparison with these previous studies.

Author's response: In the revised manuscript, we clarified the differences of our simulations of BrC from previous studies. The implementation of BrC photo-bleaching in Wang et al (2018) and Brown et al (2018) specified a uniform 1-day e-folding time for BrC, and BrC bleaching rate, depending on OH concentrations, until 25% of the original BrC absorption is left. Our approach to BrC photobleaching considers the bleaching effects from different sources. We specify a decay half-life of 12 hours when light is present for primary biomass and biofuel BrC in the daytime until 6% is left and no further photobleaching occurs (Forrister et al., 2015). The half-life for secondary aromatic BrC is specified at 12 hours in daytime (Liu et al., 2016). The revised text is added in Lines 164-177: "Previous modeling of the BrC photo-bleaching effect by Wang et al. (2018) and Brown et al. (2018) applied a 1-day e-folding time for BrC before reaching a threshold of 25% of the original BrC absorption. Our approach to BrC photo-bleaching considers different bleaching effects depending on BrC source. We specify a decay half-life of 12 hours when light is present for primary biomass and biofuel BrC in the daytime until 6% is left and no further photo-bleaching occurs (Forrister et al., 2015) due to stable high molecular weight chromophores (Di Lorenzo and Young, 2015; Di Lorenzo et al., 2017; Wong et al., 2017; Wong et al, 2019). Different components of SOA have different photo-bleaching lifetimes. Aromatic SOA has a half-life of 12-24 hours (Liu et al., 2016; Lee et al., 2014; Zhong and Jang, 2011), limonene SOA has a half-life of <0.5 hours (Lee et al., 2014). Methylglyoxal SOA has a half-life of 90 minutes (Zhao et al., 2015; Wong et al., 2017). Therefore, the half-life for secondary aromatic BrC is specified at 12 hours in daytime until it is completely removed (Liu et al., 2016)."

In Lines 389-393, we added: "The 0.013 W/m^2 DRE in the NCB simulation is lower than previous model studies considering the photo-bleaching effect (Wang et al., 2018; Brown et al., 2018). In the NCB simulation, remote BrC concentrations are mostly affected by the lower threshold for photo-bleaching, which is 6% in this study (Forrister et al. 2015) in comparison to 25% in Wang et al. (2018)

and Brown et al. (2018), causing the difference in the global DRE estimates with photo-bleaching between this work and previous studies.

In the introduction section, Lines 82-83, we added "Model simulation results without considering the differential convective transport and BC and BrC are compared to previous studies."

In the conclusions section, Lines 441-443, we added "Compared to previous studies which did not consider differential convective transport of BrC and BC, the simulated BrC DREs without (NCNB) and with (NCB) photo-bleaching are comparable to previous studies (Feng et al., 2013; Jo et al., 2016; Wang et al., 2018; Brown et al., 2018)."

2. While the heating rate conclusions are the most interesting aspect of the manuscript, the results are substantially overstated. Figure 13 shows that BrC heating rates barely exceed those of BC in the UT in convective regions. Given that uncertainties on the simulation (convective parameterization, removal, optical properties, etc, etc, etc) are large, the authors cannot state with high confidence that the heating rate from BrC exceeds BC. In particular, given that this study does not include any observational evaluation in the tropics, where the authors suggest this effect is most important, this conclusion is unsupported. The authors should temper the discussion of these results. Similarly, the manuscript title should be modified to eliminate overstatement of the results.

Author's response: We agree with the reviewer that the uncertainties of BrC module may lead to biases in the simulated BrC heating over the upper troposphere. We updated the title to "*Modeling global radiative effect of brown carbon: A potentially larger heating source in the tropical free troposphere than black carbon*" and add more discussion in Conclusion section admitting the uncertainties of the BrC parameterization.

In the conclusion section, Lines 461-464, we emphasized the uncertainties: "There are still considerable uncertainties in modeling BrC absorption and its effects in the atmosphere. Parameterizations of emissions, photo-bleaching, and convective transport of BrC all require more field and laboratory observations. The modeling result of stronger atmospheric heating by BrC than BC over the tropical free troposphere in this study are subject to these uncertainties. Field measurements over tropical convective regions during periods of biomass burning are critically needed to further improve our understanding of BrC processes and its climate effects…"

Additional comments

1. Line 70: missing name on reference

Author's response: Thank you for pointing out. Reference added on line 70 (now line 73).

2. Lines 91-95: specify meteorological years simulated

Author's response: Our free-running simulations are based on the climatology of 2010. We updated this in line 101.

3. Section 2.2: Should discuss how many different emission factors (i.e. biomes) are used in the inventory and whether their resulting BrC inventory adequately represents the variability in fuels.

Author's response: We updated the emission factor variability in the page 4, lines 117-119 "The different emission factors for tropical forest, temperate forest, boreal forest, savanna, agriculture waste and peat burning are based on Akagi et al. (2011).” We added “The variability of BrC emission rate among biomes therefore depends on the BC to OA emission ratios in the GFED emission inventory.” in page 7, lines 208-209.

4. Section 2.2: Given that the authors rely on comparisons between BrC and BC later in the text, they should include details on BC aging and optical properties in this section.

Author’s response: We added the information at the end of section 2, lines 133-135.

5. Lines 130 and 133: use the same wavelength for MAEs so that they can be compared

Author’s response: We updated MAEs in the same wavelength in line 144. The MAE value at 550 nm for secondary BrC is 0.19 m^2/g in the model.

6. Line 203-204: specify that this statement applies to the default model

Author’s response: We updated that this statement applies to the default model and all sensitivity runs at lines 241-242.

7. Lines 199-215: discussion of BC removal should also reference and compare to approach of global model study of Q. Wang et al. (JGR, 2014).

Author’s response: Wang et al. (2014) updated the model wet scavenging by scavenging hydrophobic aerosols in convective updrafts and scavenging hydrophilic aerosols from cold clouds. We increased the interstitial BC scavenging by a factor of 5 to increase wet scavenging and reduced stratiform liquid-containing cloud based on model evaluations using HIPPO data. We updated this in Page 8, Lines 240-244.

8. Line 243: requires citation at the end of the sentence.

Author’s response: We moved the citation to the end of the sentence.

9. Lines 245-252: it would be useful to discuss why BrC scavenging isn’t treated similarly to BC scavenging in each simulation

Author’s response: We updated the wet scavenging efficiency of BrC based on the convection outflow/inflow ratio discussed in Zhang et al. (2017).

10. Figure 7: Why don’t the authors compare observed and simulated OA mass and BrC absorption directly?

Author’s response: We updated the comparison of BrC between the model results and observations in the supplement. The change of BrC/BC indicates the different physical chemical properties between BrC and BC, so the ratio of BrC to BC is an important factor when estimating the physical chemical properties of BrC. In figure 7, we compared BrC/BC between the model and the observation.

11. Line 262: “During DC3 experiment, respectively.” Is not a sentence

Author’s response: We changed the typo in the updated manuscript.

12. Line 263: “Both the observations and model simulations show the increase of BrC/BC ratio at in the upper troposphere”; statement is inaccurate, model simulations do not show increase.

Author’s response: The sentence is removed.

13. Lines 261-284: This discussion is a little confusing. It should be clear from the text that the NCB and ICNB simulations are inconsistent with observations, but also that no model simulation captures the

increase in BrC/BC as observed, particularly for SEAC4RS.

Author's response: In section 5.1, lines 319-321, we showed in the text that NCB simulation underestimated the BrC/BC ratio in both DC-3 and SEAC[4]RS, and ICNB overestimated the BrC/BC ratio in the observations. In lines 325-326, we now acknowledged that no model simulation captured the increase in BrC/BC as observed in SEAC4RS.

14. Line 314: I think the authors mean DRE not "radiative forcing" here to be consistent with their earlier discussion.

Author's response: Corrected. Thank you.

Brown, H., Liu, X., Feng, Y., Jiang, Y., Wu, M., Lu, Z., Wu, C., Murphy, S. and Pokhrel, R., 2018. Radiative effect and climate impacts of brown carbon with the Community Atmosphere Model (CAM5). Atmospheric Chemistry and Physics, 18, 17745-17768, https://doi.org/10.5194/acp-18-17745-2018, 2018.

Forrister, H., Liu, J., Scheuer, E., Dibb, J., Ziemba, L., Thornhill, K. L., Anderson, B., Diskin, G., Perring, A. E., and Schwarz, J. P.: Evolution of brown carbon in wildfire plumes, Geophysical Research Letters, 42, 4623-4630, https://doi.org/10.1002/2015GL063897, 2015.

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