Referee #2

Thank you for your helpful comments. We response to your comments and revise the manuscript based on the comments, described in the following content.

This study examines the global direct radiative effect of brown carbon (BrC), using the Community Atmospheric Model version 5 (CAM5). A biomass burning emission inventory and aerosol module for BrC are developed and implemented into CAM5. The direct radiative effect (DRE) due to BrC absorption is estimated, and the results show that the atmospheric heating by BrC is the most significant in the mid and upper troposphere over the tropics, exceeding the BC heating effect. Sensitivity studies are conducted to demonstrate the relative importance of the BrC photo-bleaching effect and convective wet scavenging on the estimated BrC DREs. Overall, this study presents interesting findings about the global distributions of BrC and its radiative effects relative to black carbon (BC). However, there are some major concerns about the modeling approach that need to be addressed before consideration for publication.

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

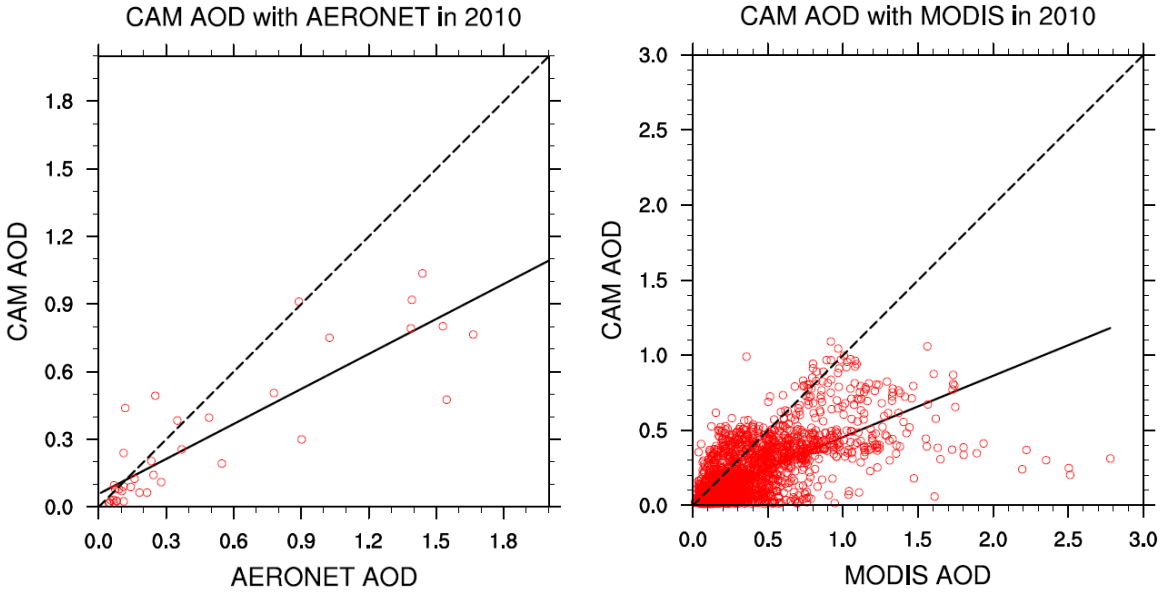
First, the parameterization of BrC needs clarification and justification. The primary BrC is included with explicit emission inventories scaled to the organic carbon emissions. This assumes BrC as an individual aerosol type emitted separately from other non-absorbing organic aerosol (OA) compounds. Are there observational evidence to support the assumption? It is not clarified if the BrC emissions are then excluded from the total organic aerosol (OA) emissions or not. The double-counting of BrC may lead to artificial increases in OA aerosol mass and total AOD. Similarly, is secondary BrC included as part of the SOA formation or additional SOA formed from aromatics? The rest OA should be non-absorbing, scattering only, and what optical properties are used for the non-absorbing OA? Furthermore, how does this approach differ essentially from Brown et al. (2018)? Both of them parameterize the absorption due to all the OAs (absorbing + non-absorbing) following Sahel et al. (2014). The definition of BrC radiative effect is also a bit confusing. If BrC are emitted separately as an aerosol type with explicit emissions, then the radiative effect of BrC refers to both scattering and absorption.

Author's response: We understand the reviewer's concerns. While BrC is part of OC in the atmosphere, we can treat them separately using different tracers in model simulations. One can treat BrC as a fraction of OC (as in Brown et al., 2018) but assumptions have to be made (for example, the BrC/OC fraction does not change during transport including convective transport, which should change based on aircraft observations (Zhang et al., 2017)). Our simulation approach is more flexible. There is no double counting problem as long as scattering and absorption are treated separately (through different tracers) in the model. We did not account for the scattering effect of BrC such that absorption of BrC can be compared directly to BC. We added a paragraph at the end of section 3.1, lines 188-192: "One important finding by Zhang et al. (2017) is that wet scavenging of BrC during convection differs from BC and OC. Therefore, BrC is simulated using a different tracer from OC in this work unlike Browne et al. (2018). The BrC property of interest is absorption and we assume that the tracer's optical property is light absorption only (no scattering). Consequently, there is no double counting of OC scattering. In the following analysis, the DRE from BrC is for light absorption only such that it can be directly compared to BC."

Second, the main focus of this study is on the BrC heating effect, but it lacks of evaluation of the modeled aerosol absorption. The ratio of BrC absorption to BC is compared with the DC3 and SEAC4RS measurements, however, limited only to the North America. There is no model evaluation of aerosol absorption over the tropics where the BrC heating effect is suggested to be important. It would be useful to know how the modeled aerosol total and spectrally-dependent absorption compare with observations, i.e., AERONET data, with vs without BrC parameterizations. This would provide observational constraints for the

calculated heating effects due to BrC and BC. In addition, Figures 5 shows the comparison of modeled AOD with AERONET and MODIS, but it is unclear if the inclusion of BrC improves the simulated AOD or not?

Author's Response: In general, the effect of aerosol absorption on AERONET AOD is small and this is particularly true for BrC. In Figure 5, we compared AOD from our best model (including BrC absorption) to AERONET and MODIS. The following figure shows modelled AOD without BrC, compared with AERONET and MODIS. It is nearly identical to the results with BrC shown in Figure 5. Quantitively, in fire dominate regions (where BC from fire emissions contributes to >50% of the total BC), BrC contributes ~0.37% of the total AOD and 8.5% of the total absorption aerosol optical depth (AAOD). The increase of AAOD from BrC absorption is more significant than AOD. We added in page 9, lines 274-276, "For these data, the effect of BrC absorption on AOD is small; we estimate that BrC absorption contributes 0.37% of the total AOD and 8.5% of the total absorption aerosol optical depth (AAOD)."

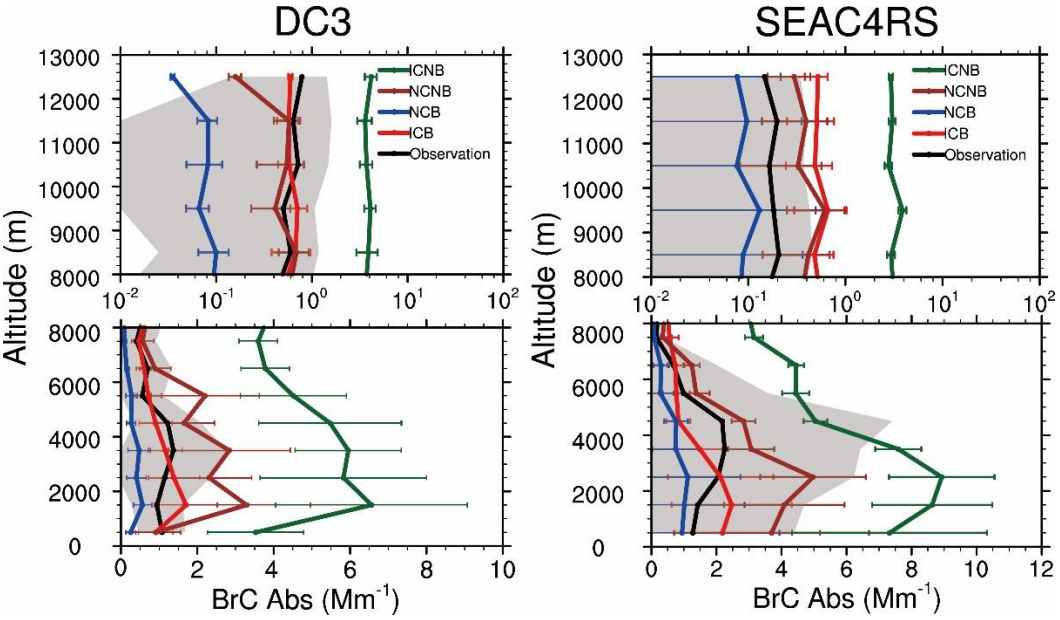


Third, the model sensitivity studies offer limited insights on physical processes. Parameter tuning of the photo-bleaching effects and convective transport of BrC are largely empirical. It is not very convincing, based on Figure 7, to state that the overestimation of BrC/BC ratios in lower troposphere by NCNB (the base mode) and ICNB reflects on the missing of photo-bleaching effect, because it could be due to underestimate of BC. ICB (the best model) that includes the photo-bleaching also overestimates to a similar extent especially when compared with the SEAC4RS data. It is not clear how good or bad these bleaching effects are represented in the model? Or where/when it improves the modeled absorption by BC+BrC? If not, how to improve based on the sensitivity studies conducted? The enhancement of BrC in convective transport is implemented by reducing the wet scavenging efficiency of BrC to match the transported BrC by convective clouds in Zhang et al. (2016). This ignores the impact of uncertainty in model-simulated convective clouds, and possible aqueous formation of secondary BrC in clouds, attributing the BrC changes solely to the scavenging efficiency without justifying it. At least, other factors should be discussed and probably examined in the sensitivity studies.

Author's Response: There are uncertainties in the model simulations. 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 heating by BrC than BC over the tropical free troposphere in this study are subject to the

uncertainties. The results from this study indicate that 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…"

We added the following figure in the supplement (now Figure S2) to show that that BrC in the NCNB model is much higher than the observations in the lower troposphere. Photo-bleaching, which was observed in field observations, is an obvious reason for the overestimated BrC/BC ratio and BrC concentrations in the lower troposphere in the NCNB model.



To further address the reviewer's comments, we added a new paragraph at the end of section 5.1, lines 330-337: "In the ICB simulation, wet scavenging of BrC was reduced relative to BC in order to simulate the observed BrC/BC ratios in DC3 and SEAC^{4}RS. The mechanisms are not yet clear due to a lack of laboratory and field observations. Hydrophobic OC, such as humic-like substances (HULIS), is more likely to have high light-absorption compared to hydrophilic OC (Hoffer et al., 2006). BrC with high molecular weight dominates the aged biomass burning plume (Wong et al., 2017, Wong et al., 2019). Since higher molecular weight compounds have lower hygroscopicity (Dinar et al., 2007), and it is harder to activate hydrophobic OC in cloud, less BrC is removed in deep convection. Another possible mechanism is production of BrC through in-cloud heterogeneous processing of fire plumes (Zhang et al., 2017). However, there is no observation data to implement such a mechanism in a model."

Other than the main concerns, a few minor comments are also included:

1. Page 5, line 136: why do you use different reference wavelengths for primary and secondary BrC?

Author's Response: We kept the reference wavelengths from the experiments of McMeeking, (2008) and Nakayama et al. (2010) for primary and secondary BrC. To make MAE references agree with each other, we have converted secondary BrC MAE to 550 nm (0.19 m2/g) using the AAE = 5.0 in the updated manuscript section 3.1, line 144.

2. Page 5, line 137: AAE is highly variable and the estimated BrC absorption is sensitive to AAE. Uncertainty associated with AAE should be discussed and probably examined in sensitivity studies.

Author's Response: Previous modelling study from Jo et al. (2016) estimated BrC/BC absorption ratio with different AAE of BrC. They found BrC/BC ratio will decrease when BrC AAE increase from 5 to 6.19. We used a BrC AAE of 5.0, which also agrees with the experiment results of Kirchstetter and Thatcher (2012). We discussed the uncertainty of BrC simulation coming from AAE variation in the updated manuscript section 3.1, lines 152-156.

3. Page 5, line 139: this equation is confusing. What is the reference? Where is k_OA, from the second step to the third?

Author's Response: The first step from the equation is to convert the imaginary refractive index of OA (BrC + non-absorbing OA) to the imaginary refractive index of BrC. k_{OA} from the second step to the third is based on the Eq.6 in Liu et al., (2013):

$$k = \frac{\rho\lambda \cdot A(\lambda)}{4\pi \cdot c}$$

where k is the imaginary refractive index, ρ is particle density (g m^{-3}), A(λ) is the light absorption at the wavelength λ, and c is the mass concentration.

4. Page 6, line 165: are you using k_OA from Sahel et al., 2014 for OA, after including explicit BrC emissions?

Author's Response: No. We convert k_{OA} to k_{BrC} using Eq.3, and apply it to the Eq.6 in Liu et al. (2013) to get Eq. 5, and we applied k_{BrC} to the explicit BrC emissions. We added after Eq. (4), lines 200-202, "We computed $k_{OA,550}$ in order to calculate BrC emissions. In the model, the absorption of the OC tracer was specified to be 0. All OC absorption was due to the BrC tracer."

5. Page 10, section 5.3, equations (6) and (7): I think that the parameterizations in Sahel et al., (2014) are for absorption spectral dependence of the total OA including both absorbing and non-absorbing components. But here they are applied to the absorbing components only?

Author's Response: Equations (6) and (7) are the functions for the wavelength dependence of imaginary refractive index for all kinds of particles**,** they obey a power relationship with the wavelength ratio and an exponent w. The parameterizations in Saleh et al., (2014), described in Eq. 4, shows a relationship between BC/OA ratio and the k of total OA including both absorbing and non-absorbing components. We convert k_{OA} to k_{BrC} using Eq. 3 and apply it to the absorbing components.

6. Page 10, line 311: please clarify what two sets of radiative fluxes are used to calculate the BrC DRE

Author's Response: We simulated the clear sky net solar flux at the top of atmosphere with all aerosols (FSNTOAC, the BrC tracers have only absorption and no scattering) and the clear sky net solar flux at the top of atmosphere with all aerosols but no BrC absorption (FSNTOAC_noBrC, the BrC tracers have no absorption or scattering). We calculated BrC DRE by subtracting FSNTOAC and FSNTOAC_noBrC. Lines 366-368 is updated with this information.

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.

Browne, E. C., Zhang, X., Franklin, J. P., Ridley, K. J., Kirchstetter, T. W., Wilson, K. R., Cappa, C. D., and Kroll, J. H.: Effect of heterogeneous oxidative aging on light absorption by biomass-burning organic aerosol, Aerosol Science and Technology, 1-15, https://doi.org/10.1080/02786826.2019.1599321, 2019.

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.

Jo, D. S., Park, R. J., Lee, S., Kim, S.-W., and Zhang, X.: A global simulation of brown carbon: implications for photochemistry and direct radiative effect, Atmospheric Chemistry and Physics, 16, 3413-3432, https://doi.org/10.5194/acp-16-3413-2016, 2016.

Kirchstetter, T., and Thatcher, T.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmospheric Chemistry and Physics, 12, 6067-6072, https://doi.org/10.5194/acp-12-6067-2012, 2012.

Liu, J., Bergin, M., Guo, H., King, L., Kotra, N., Edgerton, E., and Weber, R.: Size-resolved measurements of brown carbon in water and methanol extracts and estimates of their contribution to ambient fine-particle light absorption, Atmospheric Chemistry and Physics, 13, 12389-12404, https://doi.org/10.5194/acp-13-12389-2013, 2013.

McMeeking, G. R.: The optical, chemical, and physical properties of aerosols and gases emitted by the laboratory combustion of wildland fuels, Colorado State University, 2008.

Nakayama, T., Matsumi, Y., Sato, K., Imamura, T., Yamazaki, A., and Uchiyama, A.: Laboratory studies on optical properties of secondary organic aerosols generated during the photooxidation of toluene and the ozonolysis of α-pinene, Journal of Geophysical Research: Atmospheres, 115, https://doi.org/10.1029/2010JD014387, 2010.

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu, S., Aiken, A. C., Sullivan, R. C., Presto, A. A., Dubey, M. K., and Yokelson, R. J.: Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nature Geoscience, 7, 647-650, https://doi.org/10.1038/ngeo2220, 2014.

Wang, X., Heald, C. L., Liu, J., Weber, R. J., Campuzano-Jost, P., Jimenez, J. L., Schwarz, J. P., and Perring, A. E.: Exploring the observational constraints on the simulation of brown carbon, Atmospheric Chemistry and Physics, 18, 635, https://doi.org/10.5194/acp-18-635-2018, 2018.

Zhang, Y., Forrister, H., Liu, J., Dibb, J., Anderson, B., Schwarz, J. P., Perring, A. E., Jimenez, J. L., Campuzano-Jost, P., and Wang, Y.: Top-of-atmosphere radiative forcing affected by brown carbon in the upper troposphere, Nature Geoscience, 10, 486, https://doi.org/10.1038/ngeo2960, 2017.