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Interactive comment on "Modeling global radiative effect of brown carbon: A larger heating source in the tropical free troposphere than black carbon" *by* Aoxing Zhang et al.

Anonymous Referee #2

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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

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Discussion paper

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.

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.

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?

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.

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? 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. 3. Page 5, line 139: this equation is confusing. What is the reference? Where is k_OA,λ from the second step to the third? 4. Page 6, line 165: are you using k_OA from Sahel et al., 2014 for OA, after including explicit BrC emissions? 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? 6. Page 10, line 311: please clarify what two sets of radiative fluxes are used to calculate the BrC DRE

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