

We thank the reviewer for his/her time and comments. We have made efforts to improve the manuscript accordingly, please find response for corresponding points below.

Reviewer #1

Brown carbon (BrC) represents an emerging category of particulate organic compounds that can absorb solar radiation effectively in the spectral range of UV light. Although BrC is increasingly evolved in climate models, its (direct) radiative forcing remains highly uncertain, partly due to underestimation of organic aerosol (OA) mass by chemical transport models and lack of knowledge on optical properties of both primarily emitted and secondarily formed organic aerosols. The manuscript by Wang et al. investigated the observational constraints on the simulation of BrC by GEOS-Chem. The authors found that their modelling results on BrC absorption could be improved through increasing the OA mass associated with biomass burning (BB_OA), decreasing the mass absorption efficiency of BB_OA, and adding an aging scheme for BB_OA. The topic of this manuscript falls well within the scope of ACP. Although the manuscript does not provide much scientific insights into the discrepancy between simulated and observed OA mass, the idea of involving a photo-chemical scheme in chemical transport models to simulate degradation of BrC is interesting. It could be accepted for publication given the authors could address the following concerns.

1. Page 1, line 19-20. An exaggerated statement was made here. Comparison of simulated and observed BrC has been performed by previous studies (e.g., Atmos. Chem. Phys., 16, 3413–3432, 2016). The statement in line 10-11, page 4 is more proper.

Our statement “this is the first study to compare simulated BrC absorption with direct ambient measurements” is correct given that previous studies have only had access to, and compared models with, indirect measurements. However, we have added text similar to that on page 4 to specifically indicate that we are referring to aircraft measurements here.

2. Page 3, line 17-19. These two sentences are questionable since BrC existing as coating materials on BC cores also absorb light. In the case of BrC coating, although the lensing effect is reduced relative to clear coating, this reduction can be overwhelmed by the effect of BrC shell absorption (e.g., Science of the Total Environment, 599–600, 1047–1055, 2017).

This is an interesting point. Given the limited measurements, it is not clear whether BrC coatings on BC cores also absorb light. We have added several sentences to discuss this point (page 3, line 22-24).

3. Page 3, the last paragraph. There have been many ground-based studies in which BrC absorption was directly measured by approaches similar to those used in DC3 and SEAC4RS.

However, these studies were not mentioned and were not used to constrain simulation results on BrC. Although these studies could not provide information on BrC vertical distribution, surface BrC absorption could still be useful for the evaluation of simulated BrC.

We thank the reviewer for this suggestion. However, previous direct observations of surface BrC absorptions are not particularly helpful in constraining model simulations as nearly all of these studies report data at low temporal resolution (e.g. 1-week or 1-month averages) without co-measured species. This makes it difficult to identify the sources of BrC and evaluate the model.

4. Page 5, line 26. Suggest adding “with an OA/OC range of ” before “1.34-1.65”.

Added.

5. Page 7, equation 3. What is the difference between w and the absorption Ångström exponent (AAE)? According to equation 5, they should be the same. Please avoid using different terminologies for the same parameter.

They are different. w refers to the wavelength dependence of imaginary part of refractive index (k); AAE refers to the wavelength dependence of absorption. These two kinds of wavelength dependence are different, since aerosol absorption is not linearly related to k .

6. Page 7, line 10-11. This point, i.e., biomass burning refers to open burning and does not include biofuel, should be clarified when biomass burning/biofuel were used for the first time in section 3.

We move this clarification to Section 3 (page 6, line 3).

7. Page 8, line 20. Why are the observed mass concentrations for externally mixed OA alone? HR-TOF-AMS cannot measure internally and externally mixed OA separately.

We have removed this sentence.

8. Page 10, the first paragraph. I am confused about sources of “the 80% bias”. According to the discussions in line 6-23, it seems that this bias could be primarily attributed to neither the underestimate of POA emission factor nor the underestimate of biomass burning SOA. But then the authors said that this bias was due to “the underestimate of either the POA

emission factor or biomass burning SOA” (line 26-27). These descriptions need to be revised to keep consistent. To my understanding, this bias indicated that the POA emission factor was considerably underestimated for biomass burning, although the actual BC/OA emission ratio would not be as low as 0.027 for biomass burning. In addition, please check “BC/OA emission factor” used in this paragraph. I think all of them should be “BC/OA emission ratio”.

In our discussion on page 10, we conclude that the 80% bias could not be attributed to the underestimate of POA emission factor only nor the underestimate of biomass burning SOA only. This means the bias is likely due to the integrated influence of these 2 factors. We agree with the reviewer that “BC/OA emission ratio” is a better than “BC/OA emission factor” in this paragraph and change the phrasing throughout.

9. Page 13, line 24-25; and page 14, line 7. The authors are required to compare the AAE evolved here (which can be readily derived from the MAC values at different wavelengths) with those directly measured (e.g., Atmos. Chem. Phys., 15, 7841–7858, 2015), to see whether the assumed absorption wavelength dependence was reasonable.

Thank you for the suggestion. We add a short discussion at the end of this paragraph (page 13, line 28-31) to address this point.

10. Page 13, line 25-26. The statement that “we assume that biomass burning SOA is equally absorbing as primary OA from biomass burning” does not agree with the description in section 3.2 that “For SOA, we assume that only aromatic SOA absorbs light”, unless the authors assumed that all of the biomass burning SOA were from aromatic precursors.

We agree that this was unclear and now clarify this on page 7, line 26. The statement of SOA in Section 3.2 does not include biomass burning SOA.

11. Page 15, line 7 and elsewhere in the manuscript. I don’t think the model assumptions applied in “Modified_Age” and “Modified_Simple” were really constrained from US fires observed during SEAC4RS. In fact, SEAC4RS was only used to evaluate or validate these assumptions.

The reviewer is correct. We clarify this point throughout the manuscript.

12. Caption of Figure 7. Wavelength should be clarified.

Added.