

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 #2

General comments

The paper of “Exploring the observational constraints on the simulation of brown Carbon” investigates the optical properties and DRE of BrC using GEOS-Chem model coupled with RRTMG model. They applied a photochemical scheme in the model to address the aging effect of BrC absorption and tested it against BrC absorption measurements from two aircraft campaigns. This study aims to “explore how assumptions for BrC sources, processing, and properties impacts the comparisons with these observational constraints and estimate the resulting global direct radiative effect of BrC under these conditions”. While the authors addresses the topics listed in the paper, it is not immediately clear how significant the results actually are.

First, they need more constraints from observations near sources in addition to the aircraft campaigns used in the study to test the photochemically whitening process for BrC absorption. Detailed comparison between Modified_Age and Modified_Simple should be provided to show the necessity and advantage of this aging scheme.

We agree with the reviewer that it would be helpful to have observational constraints near sources to further evaluate the whitening process. However, unfortunately, to date, there are no such appropriate measurements. Previous direct observations of BrC absorption at the surface have low temporal resolution and have not been accompanied by measurements of other species (e.g. CO, NO, NO_x, etc.) needed to identify the photochemical aging state or transport time. We hope that future measurements (including perhaps during the upcoming FIREX and FireChem campaigns) will enable further evaluation of these schemes.

In Section 4.1, we show that the aging scheme improves the model simulation during the DC3 campaign. Both the Modified_Age and Modified_Simple schemes reflect the influence of aging on absorption, so a comparison between these two would not show the advantage of aging in capturing observations. In Section 5.2 we compare the simulated DRE between Modified_Age and Modified_Simple and find very small difference.

Second, the authors argued “DRE of BrC has been overestimated previously due to the lack of observational constraints from direct measurements and omission of the effects of photochemical whitening”. However, they ignored some studies, which do not include this aging effect but show low DRE of BrC. For example, Hammer et al. (2016) estimated DRE of 0.03 W m² for BrC constrained by OMI UVAI values, which is even lower than the result

in this work (doi:10.5194/acp-16-2507-2016). Comparison with such studies may help to understand the factors contributing to the uncertainties in BrC absorption and to verify this aging scheme.

Thank you for raising this point. We have added a discussion of Hammer et al. 2016 to the end of Section 5. However we emphasize that this study used indirect measurements to constrain the BrC DRE, and uncertainties on these measurements are challenging to estimate.

Finally, the MAC and the subsequently whitening process are strongly affected by the fraction of BrC associated with biomass burning. The authors assumed that the optical properties for biomass burning SOA are the same as those for biomass burning POA. But such assumption contradicts with their earlier statement that SOA is not absorber, at least not a significant absorber. Thus they may overestimate the fraction of BrC and underestimate the MAC for biomass burning OA.

We agree with the reviewer that the text was unclear on this point. We now clarify this on page 7, line 27. The statement regarding SOA absorption in Section 3.2 was not meant to include biomass burning SOA.

In summary, this paper is well written and is easy to follow along. Its topic fits ACP and it is worthy of publication in ACP subject to addressing these and specific comments below.

Specific Comments

p. 1, line 23-24, the AAE is not constrained from absorption measurement

The reviewer is correct. We have removed the sentence about AAE.

p. 2, line 10-11, as stated above, there are also studies with low DRE of BrC

We extend the DRE range here to represent all previous studies.

p.4, line 28-30, the factor converting extract absorption to aerosol absorption is a function of aerosol size distribution. Is the factor of 2 consistent with the model assumption of OA size distribution in this study?

The factor of 2 is related to the size distribution of BrC field measurements at 3 sites (Liu et al., 2013). In these measurements, the mass mean diameter (MMD) of OA is 500nm with standard

deviation (δ) of 1.5 – 2.4. With a standard deviation of 1.8, this MMD can be transferred to a count mean diameter (CMD) of ~180nm. This size distribution is very close to our assumption (CMD = 180nm, $\delta=1.6$). We add a sentence to clarify this point in Section 2, page 4, line 20-31.

p.7, line 15, what is the density of OA used in the model? Will the assumption of the GMD of OA strongly affect its MAC?

The density of OA is assumed to be 1.3 g/m^3 . We add this to the text in Section 3.2, page 7, line 13.

The GMD of OA could affect its MAC strongly but this influence is non-linear. For example, with $\delta=1.6$ and refractive index of BB BrC, a 50% decrease in GMD causes 1% difference in MAC, however, a 50% increase in GMD will increase MAC by 35%. As replied in last comment, we use the same GMD value as measured BrC in filed observations.

p.8, line 16, from biomass burning and biofuel

Changed (typo).

p.9, line 24-25, high CH₃CN and high CH₃CN-OA may be due to the transport of plumes mixed with biomass burning and other sources. More evidence (e.g. enhancement ratio CH₃CN/CO) is needed to support the conclusion of little contribution from sources other than biomass burning.

There is no significant CH₃CN/CO enhancement during the identified BP (the ratio during BP is only ~15% higher than the average during the campaign). However, we find not only high CH₃CN and high CH₃CN-OA correlation but also high OA-BC correlation, high BC and OA concentrations during BP. We think this is enough to identify the biomass burning influenced periods. We have clarified this point in the Section 4.1.

p. 10, line 2-3, the difference between 145% and 36% is _ 110%, not 80%

The difference between 145% and 36% in this context is 80%. After a 36% increase, we still need ~80% increase to get the 145% difference: $(1+0.36) \times 1.8 - 1 = 1.45$.

p.10, line 7-8, although lower than 0.03, the BC/OA of 0.027 should be still within the uncertainty range of biomass burning emission ratios

We agree that this value should be within the uncertainty range of BC/OA emission ratio, but it seems unlikely that all fire emissions fall below the range of emission ratio (0.03-0.06) for biomass burning as given in GFED. Our statement does not preclude this possibility, we simply indicate that it is unlikely.

p. 10, line 29-30, only biomass burning OA is increased in FixBB. This won't affect the estimation of BrC absorption, but overestimates its contribution to total AAOD (the analysis in Sec. 5) as BC mass is still underestimated.

In our analysis of DC3 data, we adjust both the OA and BC biomass burning mass concentrations upwards to match the observations (as indicated in the previous sentence). A further increase to OA is applied to correct for an underestimate in the OA emission factors and/or missing biomass burning related SOA. Therefore biases in biomass burning are addressed for BC and OA and correctly reflected in the AAOD calculations.

p. 14, line 17-18, the peak in the middle troposphere from SEAC4RS is not reproduced

The sentence "... during DC3 and SEAC4RS except for altitudes above 10 km." is changed to "... during DC3 and SEAC4RS at the altitudes with enough data points below 10 km."

p. 15, line 25, any explanation for high BrC absorption contribution in NA and Russia? The discrepancy between model and the observation is large as seen from the figure.

This is an interesting point. The absorption properties and/or BrC contribution to total OA may be different for different fires/biofuel combustion sources. Our model assumptions constrained from the US fires are able to capture the observed BrC absorption contributions in Europe, but have large discrepancy in Alaska and Russia. This suggests that the BrC absorption properties and/or BrC contribution to total OA could be very different between Alaska/Russia and US/Europe. Due to the limitation of measurement data, we are not able to undertake any further analysis of this discrepancy, but we add text acknowledging these discrepancies in page 15, line 17-18.