Responses to Referee's Comments

We thank the referee for careful reading and valuable comments.

Anonymous Referee #2:

The authors have addressed my comments adequately. The manuscript has been significantly improved with the response to all the reviewers' comments.

1. However, the authors still need do a careful read and make sure that their statements are clear and accurate. For instance, just in the abstract, I find two places that need clarification: (1) Line 34-35: in the sentence "The model successfully reproduced…" what model quantity is it referring to, observed absorption? or SSA? (2) Line 36, "BrC accounts for 21% of the global mean OC concentration" – at surface? or the lowest model level?

➔ ***We added and changed the abstract as follows. We further changed several sentences in the text for the clarity.***

Line 34-35 : The model successfully reproduces the seasonal variations of observed light absorption by water-soluble OC, but underestimates the magnitudes, especially in regions with high secondary source contributions.

Line 36 : Our global simulations show that BrC accounts for 21% of the global mean surface OC concentration, which is typically assumed to be scattering.

2. Additionally, some caveats associated with their method and model used were acknowledged by authors in the response to reviewers, and they should also be mentioned in the manuscript in order to put them in the context for interpreting the obtained results. I suggest:

2-1. Add the assumptions made about external mixing of BC and OC, and neglecting the coating effect due to non-absorbing clear species, when describing the method of relating the absorption properties to the MCE.

➔ ***We added and changed the text in Section 2.1 as follows:***

In addition, we are able to obtain the BrC/BC absorption ratio using AAE. In

Appendix A, we present a detailed description of our method for estimating the relationship between the BrC/BC absorption ratio and AAE. Our method assumes external mixing, and this assumption can cause uncertainties when particles are internally mixed (such as coating effect). For uncertainty analysis, we calculate three BrC/BC absorption cases as shown in Figure 1, which shows the estimated BrC/BC absorption ratio at 550 nm as a function of MCE.

2-2. Include the evaluation of the AERONET AAE: Figure 2 and Figure 3 in the response could be placed in the Appendix, but it should be pointed out in the text that the AAE is overestimated by the model (with BrC) compared with AERONET.

➔ ***We added the figures in the supplement and the paragraph in the text as follows:***

We further compare the model against AERONET AAE as shown in Figure S1. We find that the model overestimates the observed AAE after including BrC, in part, because the model underestimates BC emissions as discussed above. However, the simulated AAE will be decreased if we increase BC emissions as suggested by the top-down estimate (Cohen and Wang, 2014). For example, for regions (North America, Central America, South America, Southeast Asia, and Australia) where the difference between our BC emission and the top-down estimate is within a factor of 2, we find that the model with BrC shows a better agreement with the observed AAE (Figure S2) and with the observed SSA (Figure S3).

2-3. Include the response to the question about the fitting method in the Appendix A1, lines 3-4

➔ ***We added the figure in the supplement and the text in the Appendix as follows:***

For example, Figure S4 shows the linear regression case for F=4.0. In this case, R2 is 0.99 and Angstrom exponent of CA is 4.44. Y-intercept of the numerical fitting is -29.81, which is consistent with Y-intercept (-29.64) from Eq. (A5). The difference between two Y-intercept values are always within 1%, which shows the numerical fitting with Eq. (A4) satisfies both the slope (A) and the intercept (B) at the same time within 1% error.