We thank the reviewer for the helpful comments and revisions that have improved the manuscript. This document includes the comments of the reviewer as well as our responses. Reviewer comments are **bolded**, responses are in regular font, and *excerpts of changes to manuscript are in italics, with* <u>*new changes underlined*</u> *if added to an existing sentence*. We include references cited in responses at the end of this document.

**In the revised manuscript, the authors have added substantial basic information on model settings, supplemented the evaluation of the BrC radiative effect, and more thoroughly discussed the model's existing uncertainties and potential reasons for underestimating BrC. In their response to reviewers, the authors noted that this study found the model's OA-to-OC ratio is significantly underestimated compared to aircraft observations, and improving the OA-to-OC ratio expression markedly enhanced the model's performance relative to aircraft observations. This is a scientifically significant conclusion, often overlooked by other models, but the discussion and experimental design related to this conclusion in the current manuscript are not sufficiently clear or detailed, making it easy for readers to overlook the study's importance. Below are listed some comments and suggestions regarding this issue and other minor issues to further improve the manuscript. I recommend this manuscript for publication after these issues are addressed.**

**Because some line numbers are obscured by margins, the following comments refer to page numbers and the last two digits of the line numbers in the revised manuscript marked with modifications.**

**Major issue: Improvements to the OA-to-OC ratio discussion**
**Currently, the discussion of the OA-to-OC ratio is in Section 3.3, covering the motivation, design of sensitivity tests, and results. However, in the model description section (Section 2.2), only ModelE-base is described, with a consistent OA-to-OC ratio of 1.4:1 set. On one hand, this may mislead readers; on the other hand, the setup of these sensitivity tests is critical to the reliability of the conclusions, yet it is described in a scattered manner in Section 3.3, lacking clarity. The current structure also leads to redundant figure information; for example, all information in Figures 6 and 7 of the current version is included in Figures 10 and 11 (please correct me if I misunderstood). Therefore, I suggest clearly describing all changes in the sensitivity tests (the five simulations in Figure 10) and their rationale in a dedicated subsection within Section 2 (Methods).**
The vertical profiles of Figs. 6-7, showing the base case comparison of ModelE to campaign data, are included in Figs. 10-11 for reference when analyzing the impact of each sensitivity test. We have clarified this in Section 3.4 (revised submission L616-618): *"Figures 10 and 11 show the same vertical profile analysis, comparing flight campaign measured and ModelE simulated median BrC-Abs in near-source and remote regions, as Figs. 6 and 7, but with the results of additional sensitivity tests.* <u>*The vertical profiles of the ModelE base case, shown in Figs. 6 and 7, are also included for reference."*</u>

Discussion of Figs. 10-11 does not repeat the discussion of Figs. 6-7. It instead focuses on the observed impact of each sensitivity test, relative to the model base case. As such, we would not characterize this as redundant information: biases were identified with Figs. 6-7, and Figs. 10-11 show the impact of subsequent model changes on those biases. Additionally, the results of Figs.

6-7 are discussed prior to describing sensitivity test changes because it is those results which motivate the changes. The current structure is intentional, as Figs. 6-7 provide the rationale for sensitivity tests.

We do, however, agree that the current discussion of sensitivity tests is scattered. To address this, we've reorganized section 3.3 into sub-sections to clearly identify each potential cause of bias and why that specific sensitivity test may address it (see revised L504-614). This allows for a distinct section to discuss the change to OA-to-OC ratios, allowing this important contribution to stand out. We have also provided further clarification in section 2.2 that the OA-to-OC ratio is varied at a later point (revised L208-209): *This OA-to-OC ratio value is consistent with OA schemes used in other models (Tsigaridis et al., 2014) and is constant across all OAs. In this study, however, we investigate the effect of varying this ratio on model performance relative campaign data (see Sect. 3.3.1).*

**Minor issues:**

**Title: Although this study evaluated GISS ModelE's BrC simulation performance comprehensively, the model used in this study still contain uncertainty that have been justified and improved in the other models (such as BrC in SOA, wet deposition, biomass burning etc.). In this case, the topic is recommended to add information about the model, such as "Exploring biases in brown carbon model representation with in-situ flight observations in the GISS ModelE earth system model" .**

The new title, specifically referencing GISS ModelE, is as follows: *Exploring biases in the GISS ModelE Earth system model brown carbon representation with in-situ flight observations*.

**Page 13, Line 74: The manuscript inconsistently uses "Fig." and "Figure" throughout; this is one example. Please standardize according to the journal's requirements.**

ACP guidelines require "Fig." to be used when it appears in running text, and "Figure" when it comes at the beginning of the sentence. The manuscript has been edited to ensure this requirement is followed.

**Page 34, Line 93: Change "OC-to-OA" to "OA-to-OC" to align with the rest of the manuscript.**

This was a typo that has been corrected.

**Page 12–15, Section 3.1: The model bias in BC, particularly in the free troposphere and upper troposphere, affects the attribution of subsequent BrC bias. A common approach in previous BrC studies is to evaluate the BrC/BC ratio, which can distinguish biases in biomass burning emissions (causing underestimation of both BC and BrC) from biases in BrC aging and deposition. Given the suboptimal performance of your model in simulating BC, an analysis of the BrC/BC ratio is necessary.**

There have been other studies that compared flight campaign and model vertical profiles of BrC/BC ratios, for instance Zhang et al. (2017) evaluated the ratio of BrC and BC absorption coefficients. In that study, BC absorption coefficient was determined from particle soot absorption photometer (PSAP) measurements. We didn't use this as a measurement for comparison with ModelE because it was not available across all flight campaigns—to our knowledge, it was not provided in ATom merge datasets. Another alternative would be to use the mass ratio of BrC to BC. However, this also wasn’t an option in this study, as only measurements of water-soluble organic carbon (WSOC), rather than total BrC, mass was taken in flight campaigns. ModelE does not consider BrC mass to be solely WSOC, so those measurements cannot be taken as BrC mass for model evaluation purposes. Thus, the available data for campaign-model comparison was BrC absorption and BC mass. As the ratio of these two is physically meaningless, we did not use it for vertical profile analysis.

We do, however, utilize the ratio of campaign-to-model BC mass as a “correction factor” for BrC absorption. As discussed in L410-420: *By multiplying model output by this factor, in addition to the CO-scaling factor (see Eq. 1), we can look at BrC scheme performance and remove from consideration biases in model processes that are not unique or specific to BrC and OAs. This does not mean related components, like the model’s ability to capture a fire and transport its plume, are unbiased, but that these biases are not the focus of our study.*

**Figures 3, 4, 6, 7, 10, 11: The font size in these figures is too small, affecting readability. I suggest increasing the font size to at least match that of Figure 5 or larger (closer to the main text’s font size).**

These figures, as well as Appendix Figures A2-7, have been modified with larger font sizes.

**References**

Zhang, Y., Forrister, H., Liu, J., Dibb, J., Anderson, B., Schwarz, J. P., Perring, A. E., Jimenez, J. L., Campuzano-Jost, P., Wang, Y., Nenes, A., and Weber, R. J.: Top-of-atmosphere radiative forcing affected by brown carbon in the upper troposphere, Nature Geosci, 10, 486–489, https://doi.org/10.1038/ngeo2960, 2017.