

We thank both reviewers for the helpful comments and revisions that have improved the manuscript. This document includes the comments of both reviewers as well as our responses. Reviewer comments are **bolded**, responses are in regular font, and *excerpts of changes to manuscript are in italics, with new changes underlined if added to an existing sentence*. We differentiate reviewer 1 and 2 comments with underlined headings. We also include references cited in responses at the end of this document.

Comments from anonymous referee #1:

There is considerable uncertainty in estimating the radiative forcing of brown carbon (BrC) in climate models, and assessing these uncertainties is crucial. Evaluating BrC modules using aircraft observations is particularly important (and currently underrepresented in existing studies), as these observations provide critical information on aged BrC in the upper atmosphere and remote regions—components that contribute significantly to the BrC’s radiative effect. This manuscript aims to address the issue from this perspective, which carries certain scientific merit. However, after carefully reading the current manuscript, one disappointing aspect is that, compared to the authors’ previously published work on the BrC module in the GISS ModelE (Delessio et al., 2024a), the model representation remains outdated in some parts, and shows no clear improvements.

The BrC module in ModelE remains consistent with its initial implementation in 2024 because previous evaluations were either unable to constrain scheme parameters (as was the case in DeLessio et al., 2024) or constrained scheme parameters with AERONET retrieval assumptions, which were not grounded in in-situ BrC chemistry and microphysics (as was the case in DeLessio et al., 2025). This was discussed in submitted manuscript L58-64 and L68-71, included here for reference:

- Unable to constrain properties in DeLessio et al., 2024: “We found we were unable to constrain the BrC scheme with these data, primarily due to a limitation in ModelE’s radiation scheme: ModelE computes radiative properties, including aerosol optical depth (AOD) and absorbing aerosol optical depth (AAOD) over broad radiation bands, rather than distinct wavelengths (Bauer et al., 2010). For the UV-to-visible (UV-VIS) wavelength range—the BrC relevant range—ModelE produces radiative properties over the entire visible band. These properties are indicative of 550 nm, the spectrally-weighted average of light from 300-770 nm. Our results showed that a BrC scheme did not have a discernible effect on ModelE total AOD and AAOD at this wavelength, inhibiting our ability to constrain the scheme’s properties.”
- Constrained in a less “physically correct” way in DeLessio et al., 2025. “In this subsequent study, we were able to constrain our scheme by aligning ModelE’s BrC physical and optical properties as closely as possible to those assumed by the retrieval. However, this speciated AERONET retrieval used simplified parametrizations to represent BrC (Schuster et al., 2016), meaning it was not indicative of in-situ BrC chemistry and microphysics.”

The purpose of this study was to evaluate this initial BrC scheme with flight campaign measurements to address the limitations of previous evaluations.

It is understandable that there are large discrepancies between the current BrC module and observations, but in order to align with aircraft observations, the authors have introduced substantial calibration to the model's aerosol concentrations. These calibrations involve significant uncertainties, rendering the final conclusions unreliable.

The uncertainties introduced by these calibrations are discussed in detail in response to the first reviewer's 7th detailed comment and the second reviewer's 2nd major comment.

As such, the evaluations presented in this manuscript do not provide reliable insights or constructive suggestions for model development. The manuscript lacks sufficient new information to qualify as a publishable journal article and instead reads more like a supplementary validation of the authors' earlier work.

Again, as stated above, the point of this paper was to evaluate and constrain the initial implementation of BrC in ModelE because previous work was unable to do so or was not grounded in in-situ chemistry/microphysics when constraining scheme parameters. We disagree with the reviewer's comment that this paper does not provide reliable insights or constructive suggestions for model development. This study highlighted key physical and chemical processes of BrC and OAs that can impact biases in comparison to observations and identified areas that require further study. We consider that to be an important contribution to the field. Further, this paper shows that varying OC-to-OA ratios by OA species clearly (and substantially) improves model alignment with observations. That is a change that other groups can readily implement in their OA modules to improve aerosol modeling. To emphasize this point, we added an additional sentence to the conclusion of the paper (manuscript with tracked changes, aka "revised" L1183-1188): *"Despite the limitations and uncertainties outlined above, a consistent underestimation bias in ModelE BrC absorption coefficient was observed across five different flight campaigns. The results and subsequent discussion presented here allowed us to highlight important processes of BrC, OAs, and BB aerosols in general, and to identify areas that require additional study. Finally, these results showed that varying OC-to-OA ratios by OA species can greatly improve model alignment to observations. This is a simple but important improvement that should be applied to the representation of OAs in other ESMs."*

Although the paper conducts a series of evaluations—examining the impacts of adjusted emissions, BrC-to-OA ratios, and wet removal—it ultimately does not offer a best estimation for global glyoxal.

Assuming this is referring to global BrC, rather than global glyoxal, this paper doesn't provide a global estimate because its purpose is to explore potential model biases by investigating chemical and microphysical processes. However, to highlight the ultimate goal of BrC/OA-absorption modeling, which is to understand BrC's radiative effect, we have added a section that provides an estimate of global BrC radiative effect: Section 3.5 Estimating global BrC radiative effect (revised L1068-1091).

The methodology overlaps heavily with the previous paper, and the study fails to produce scientifically sound or conclusive findings. The authors need to substantially improve the model based on recent research, studies using other GCMs, and advancements in other aerosol modules of ModelE to make this study an independent scientific work.

The detailed concern and issues are listed below:

Line 199. Aromatics SOA contributed significantly (maybe the most) aged secondary BrC because its bleaching lifetime is long (~12 hours). Please justify how you deal with this underestimation.

The absence of aromatic SOA from ModelE presents an important limitation, likely resulting in an underestimation of absorption, as noted by the reviewer. While this can't be directly addressed, it is partially offset by the SOA that are accounted for: biogenic SOA. Submitted sect. 2.2.2 describes how light absorption is attributed to biogenic SOA in the ModelE scheme. An important limitation of these brown SOA is that their semi-volatile nature makes them incompatible with the BrC aging scheme. As such, brown SOA in ModelE do not bleach, and their contribution to absorption is potentially overestimated. We note the possible balancing effect of these two limitations with the following language in revised 382-388: *"Another limitation is that, as noted in Sect. 2.2.1, aromatic gases and therefore aromatic SOAs are not explicitly represented in ModelE. Since these SOA have been shown to absorb light (Liu et al., 2016; Li et al., 2022), this may cause BrC absorption to be underestimated. However, this is partially balanced by the fact that brown biogenic SOA do not bleach in the current scheme: the BrC aging scheme is incompatible with the two-product model used to parameterize semi-volatile biogenic SOA parameterization in ModelE."*

Line 205-206. In DeLessio et al., 2024a, BrC emissions are parameterized based on BC-to-OA emission ratio, according to lab experiments. In this work, BrC-to-OA emission proportion of 35% is prescribed, without showing a reliable reference. It is confusing why this modification was applied.

This is not a modification from the previous paper: DeLessio et al. (2024) used a parameterization to estimate an average BrC-to-OA emissions ratio. Based on those results, as well as the values used by other BrC modeling studies, a ratio of 0.35 (or proportion of 35%) was used. The current study uses the same value. This was clarified in revised L239-244: *"To determine this proportion, DeLessio et al. (2024) looked at BrC-to-OA emissions ratios used by similar modeling studies and calculated a ratio using a parameterization of BC-to-OA BB emissions ratio from the Community Emissions Data System. On average, other models used a ratio of 0.35 (Feng et al., 2013; Wang et al., 2014; Jo et al., 2016; Zhang et al., 2020), and the global average ratio of the BC-to-OA parameterization was 0.366, so 0.35 was used at the proportion of BB OA emissions attributed to BrC. The same ratio, 0.35 BrC-to-OA, was used in the current study."*

Lines 215-220. The settings of BrC module is essential in this work, so it cannot be simply referred to a reference. On the other hand, there are confusing modifications in Line 205-206, so the authors need to clarify how the BrC module was built in ModelE.

To address this, we have expanded on the description of the BrC module (originally in Sect. 2.2.1) by adding a separate section: Section 2.2.2-Brown carbon scheme (revised L232-390)

Lines 234-235. Are BrC BB emissions prescribed in GFAS? It conflict with previous text.

BrC BB emissions are not currently included in fire emissions inventories, including GFAS. For clarification, this was explicitly stated in the revised L237-239: *"Regarding BB emissions: BB inventories don't currently include BrC. So, to simulate the emissions of primary BrC, a*

proportion of prescribed, BB OA emissions are considered brown, and the remaining portion considered non-absorbing.”

140 **Lines 236-238. There is a strong diurnal cycle of fire, for most of time, it seldom burn at night. Several studies have derived model usable fire diurnal profile to scale, for example that constrained by FIREX (Tang et al., 2022, <https://doi.org/10.1029/2022JD036650>). This potentially leads to a 50% underestimation over the day.**

145 Since ModelE is a global ESM, a global biomass burning emissions inventory at model resolution is needed for prescribed fire emissions. The ModelE 2.1 configuration used in this study does not have the same ability of the MUSICAv0 model used in Tang et al. (2022), as it does not allow for regional refinement down to a resolution of a few kilometers. As such, the diurnal profile used in Tang et al. (2022) is not applicable to this work.

150 We do state in the submitted manuscript that the lack of diurnal cycle in ModelE prescribed fire emissions is a limitation of note (submitted L236-238), but discussion of the potential effect of this limitation (identified by the reviewer) should be included. We add this discussion in revised L398-401: *“Since GFAS provides daily emissions, ModelE divides those by the number of time steps in a UTC-day to generate constant emissions for each 30-minute time step. This must be*
155 *kept in mind in evaluating the model, as real-time fire emissions sampled by flights typically follow a strong diurnal cycle and are not constant. This limitation could result in an underestimation of fire emissions during the day, and an overestimate during the night (Tang et al., 2022).”*

160 We also mention this limitation in the revised manuscript when discussing possible biases in lower troposphere, BC concentrations (revised L529-531): *“Low BB emissions including a possible underestimation due to ModelE’s lack of diurnal emissions, as discussed previously, could bias aerosol mass low in the entire air column.”*

165 **Lines 244-246. The writing style is relatively informal in many places of the manuscript. This sentence is an example.**

Active voice is sometimes used in this manuscript to allow for clear and succinct descriptions of methodology. However, to ensure the language is not too informal, the manuscript has been edited to remove some instances of this.

170 **Lines 252-256. CO is a strong fire tracer, but There is also a strong anthropogenic (like industry and transportation) source. Although high concentrations of CO can be used to identify smoke, the ratio of CO cannot be used to scale fire emissions, especially over the clean region.**

175 This issue is addressed in response to the second major issue identified by the second reviewer (see L245 of this document).

Line 259. Please report the MAE (mass absorption ratio) at a specific wavelength of different types of BrC in this work and compare to previous lab and observation studies.

180 As complex refractive index (RI) is the optical property used to characterize aerosol-light interactions in ModelE, we have added the complex RI of different types of BrC used in this work in revised Table A1. DeLessio et al. (2024) provides a detailed explanation of how these RI

values were obtained (based on lab studies), so that it not described here. However, the imaginary RI of BrC used by other Earth system model (ESM) schemes is provided for reference.

Lines 283-285. The ‘outliers’ in the aircraft observations are fresh plumes. Removing that means the evaluation is not for plumes, then what part of BrC was evaluated in this work? Why 2 times standard deviation is a threshold? The model simulation cannot well fit the fresh plume, so other studies sometimes used hourly mean to compare, or use median + 25-75th quantiles to evaluate, instead of mean and standard deviations.

We had intended to use means and two-times standard deviation because that typically represents 95% of data (above and below the mean), but that relies on the assumption that observations follow a normal distribution. As the reviewer points out, this is likely not the case for observations that sample fresh plumes. To remedy this, all vertical profile comparisons were redone, comparing observation and model medians (rather than means) and presenting variability as the interquartile range (rather than standard deviation). We also removed the filtering of “outliers” from this comparison. Submitted manuscript Figs. 3-12 and appendix Figs. A1-6 have all been updated in this regard. It’s important to note that the key conclusions from observation and model comparisons remain the same, despite this change.

Comments from anonymous referee #2:

This manuscript presents an evaluation of BrC (brown carbon) absorption in the GISS ModelE Earth System Model using aircraft campaign measurements of black carbon (BC), organic aerosol (OA), and BrC. By comparing average vertical profiles, the authors find that the model systematically underestimates BrC absorption, even after correcting for biases in carbon monoxide and BC. To investigate this issue, they explore a range of sensitivity tests, including variations in the OA:OC ratio, BrC aging, the water-soluble fraction of emitted BrC, and assumptions about the relative fraction of emitted/browner/bleached BrC. Despite these efforts, model biases persist in comparison to the in situ observations. While the topic is important and the dataset valuable, several critical issues must be addressed before I can make a recommendation.

Major issues:

1. Model Description of BrC Optical Properties. The paper lacks any discussion of the BrC optical properties implemented in the model, such as the mass absorption coefficient (MAC) values. Since these assumptions directly influence BrC absorption estimates, they should be explicitly described and justified. There is very little discussion on browning and bleaching of BrC. How exactly is it implemented? Is it in agreement with current understanding of the timescale of browning and bleaching?

As noted in response to the first reviewer, an additional section was added to the revised manuscript to more thoroughly describe the BrC scheme (Section 2.2.2, Brown carbon scheme, revised L232-390). This includes a description of the BrC optical properties used in the model, specifically the complex refractive index (RI) used as model input. Values for ModelE BrC complex RI are included in revised Table A1. In response to this comment, a more detailed description of the browning and bleaching of BrC, and how it compares to current

laboratory/observation knowledge of BrC aging, is also included in this new section. A graphic to further demonstrate the aging scheme is provided in revised Figure A1.

Additionally, there is no discussion of how dry and wet deposition processes are handled for BrC species, which is essential for interpreting vertical profile comparisons.

BrC dry and wet deposition are consistent with other OA species in ModelE. As such, in response to this, a description of ModelE OMA aerosol wet and dry deposition processes has been added to Section 2.2.1 (revised L217-223):

“OMA aerosols are removed from the atmosphere through wet and dry deposition. Wet deposition of aerosols can occur either through in-cloud scavenging or rainout. In-cloud scavenging relies on the aerosol fraction considered water soluble as well as cloud properties, while rainout relies only on cloud properties and precipitation (it is independent of aerosol properties; Koch et al., 1999). In terms of dry deposition, the deposition velocity of particles is parameterized in ModelE to represent both gravitational settling and turbulent dry deposition. This deposition velocity, along with particle mass concentrations above the surface and a parameterization of turbulent transfer, are used to determine net surface flux (Clifton et al., 2024).”

2. Emission Corrections. The corrections applied for CO and BC based on wildfire attribution are not well justified. For some of the aircraft campaigns, wildfires are not the only significant sources of BC and CO. Without proper source attribution, applying such corrections could introduce further uncertainties.

Firstly, in response to the first reviewer’s comment, it should be clarified that the ratio of campaign-measured to model simulated CO is not used to scale fire emissions—that ratio is applied to simulated BC mass concentrations, and later to model output of BrC absorption, organic aerosols (OAs), and water-soluble organic carbon (WSOC), along with an additional BC correction for the latter three species. Additionally, it should be noted that there is precedent in the normalization of BrC using CO to account for plume dilution, specifically in analysis of WE-CAN campaign data (Sullivan et al., 2022; Washenfelter et al., 2022). Overall, the goal of such corrections is to remove from consideration biases in model processes that are not unique to OAs/BrC.

In the case of CO, we apply this correction to account for the inherent bias that occurs when plumes or inhomogeneous air masses are diluted over the entire model grid cell (discussed in submitted L248-252). As noted by the reviewers, this does introduce some uncertainty, namely that biomass burning is not the only source of CO, both in-situ and in the model. We try to account for this by filtering for samples with a BB-number fraction greater than 0.5 in DC3 and SEAC⁴RS, meaning the dominant source would be fires. In WE-CAN and FIREX-AQ, measurements are specifically taken in fire plumes, so it is reasonable to assume that the dominant source of CO is, again, fires. We were unable to account for this with ATOm as filtering for BB number fraction above 0.5 removed too much data. However, submitted Fig. A1d and Fig. A2 show that measured and model-simulated CO are typically quite low (<150 ppbV) in ATOm regions; when discussing BrC and CO measurements, Sullivan et al. (2022) assume a CO background concentration of 100 ppbV. Thus, ATOm data is likely more indicative of background conditions, and therefore less influenced by direct CO sources (BB or not).

Language to acknowledge this potential uncertainty, and discuss what was done to limit it, has been added to revised L438-447:

“As fires are a major source of CO, and it has a long atmospheric lifetime, it can be used as a tracer of fires (van der Werf et al., 2017; Zeng et al., 2021; Sullivan et al., 2022; Washenfelder et al., 2022). So, applying such a factor should account for the inability of the model to capture the same magnitude of emissions within the same grid cell. This does not, however, address the potential low bias from emissions or plumes transported in the model falling in an adjacent grid cell and not being sampled. This also introduces some potential uncertainty, as BB is not the only source of CO in the atmosphere. We aim to address this by narrowing analysis to samples influenced by BB whenever possible. This is inherent to samples from WE-CAN and FIREX-AQ, as fire plumes were directly sampled. For DC3 and SEAC⁴RS, only data with a BB number fraction greater than 0.5 is used. The same filtering cannot be applied to ATom campaigns, as it would leave too limited data for analysis. However, this uncertainty is limited in this case as CO concentrations in ATom regions of analysis are relatively lower and indicative of background levels (<150 ppbV), suggesting less influence of CO sources (BB or not).”

When comparing vertical profiles of BrC absorption, WSOC, and OAs, an additional BC correction factor is applied. This is to remove from consideration potential model biases in general aerosol processes in like transport and low/missed BB emissions. Again, this introduces some potential uncertainty because ModelE BC can be produced from industrial combustion in addition to BB. As mentioned previously, we aim to account for this by focusing on data where BB dominates. Additionally, the combined use of CO and BC factors together further limit these uncertainties: both BC and CO are emitted in higher relative quantities in BB, which is more incomplete than anthropogenic combustion processes. Since the two species have distinctly different lifetimes (3 months for CO and approximately one week for BC), an observed peak in both likely corresponds to BB. Thus, when both factors are applied to scale-up BrC absorption or OA concentrations, it is somewhat reasonable to assume this represents a fire event (or scale of fire) not captured by the model. Discussion of this potential uncertainty has been added to revised L577-586:

“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. Like the CO-factor, this BC-scaling factor introduces some uncertainty as ModelE BC can be produced from industrial combustion, in addition to BB. As mentioned previously, we aim to address this by narrowing focus to BB samples whenever possible. The combined use of CO and BC factors together further limit these uncertainties: BC and CO are emitted in higher relative quantities through incomplete BB (compared to anthropogenic combustion), and the two species have distinct lifetimes (approximately 3 months and 1 week for CO and BC, respectively). So, an observed peak in both likely corresponds to BB, and when both factors are applied to scale-up BrC absorption or OA concentrations, it is reasonable to assume this represents a fire event not captured by the model.”

3. Possible Causes of Model Bias. While the authors test several model parameters, their exploration of potential causes for the underestimation of BrC absorption remains somewhat narrow. Other important factors could include the assumed MAC values and

their spectral dependence, photobleaching rates, emission injection heights, numerical diffusion in the model, and general representation errors in model transport and mixing. A broader discussion of these potential factors would strengthen the analysis.

Some of these potential factors are discussed throughout the conclusion of the submitted manuscript, but emphasis on this discussion is needed. There are also remaining factors, highlighted by the reviewer, that were not discussed. Both have been added to the conclusion section (revised L1117-1144):

“Despite implementing physically correct changes based on literature studies, like variable OA-to-OC ratios and more reasonable WS BrC fractions, there are persistent biases between ModelE BrC absorption and in-situ measurements. As such, it’s important to discuss remaining potential factors that could be driving this underestimation. Firstly, there are general biases in model processes, like dilution and numerical diffusion, as well as BB aerosol processes, like emissions and transport, that fall outside the scope of our study. These were removed from consideration to the greatest extent possible by applying both CO and BC scaling factors to BrC and OA model output. Nonetheless, broader model evaluation studies, focusing on these mechanisms, can strengthen more specific model components, like the present BrC scheme, and should be further explored.

There are also remaining biases specific to the BrC scheme. The most straightforward way to address BrC-abs underestimation would be to increase the imaginary RI, and therefore MAE, of BrC species. This would linearly increase absorption at all altitudes. Since the magnitude of BrC biases differed between the lower, mid-, and upper troposphere, we did not focus on this, and instead investigated chemical or physical processes that could explain differences across altitudes. Of course, unilaterally increasing absorption remains an option to reduce BrC-abs underestimation and improve the ModelE scheme. Increasing WBrC fractions seemed to account for some of the potential bias of excess sinks, but further bias due to aging of primary BrC could not be addressed. We can’t conclude this is because it doesn’t cause bias in BrC absorption—the scheme changes made simply had no effect on model output. Since this potential bias can’t be eliminated, future work should implement more effective changes to the primary BrC chemical aging scheme. Possible modifications, like adding a fixed lifetime or limiting aging to non-convective transport, were discussed in Sect. 3.4. We were also unable to investigate missing secondary BrC sources as a cause of low absorption bias. As discussed in Sect. 3.3, recent studies have highlighted the likely importance of BBSOA and in-cloud production of SOA as sources of BrC, but ModelE doesn’t currently simulate either SOA type. Like excess aging, we are unable to rule these out as potential causes of bias. Representation of these two SOAs is a priority for OA scheme development in ModelE. In the meantime, further laboratory and field studies of BBSOA/in-cloud SOA absorption are needed to guide implementation of BrC into these SOA schemes, when model capability allows.”

To specifically address some points brought up by the reviewer:

- Discussion of further exploration needed for the BrC chemical aging scheme is in the submitted manuscript (L627-631).
- The spectral dependence of BrC absorption was not explored because the ModelE radiative scheme is unable to capture spectral dependence within the UV-visible (UV-VIS) wavelength band. This is a limitation of the model that was not emphasized in the submitted manuscript but has been clarified in revised L378-380. However, the AAE used to convert BrC-absorption in the UV-VIS band (indicative of 550 nm) to 365 nm for

comparison with campaign data could be varied for further sensitivity tests. This is already discussed in the submitted manuscript (L660-663).

- As the reviewer highlighted, emission injection heights can influence the vertical profiles of BrC and OAs in ModelE. We do not explore this because injection heights are prescribed by the fire emissions inventory used—GFAS1.2 (see submitted manuscript L233-236). A comparison could be done between model simulations using GFAS1.2 and other inventories, like the global fire emissions database (GFED), which do not provide injection height. In such scenarios, without injection height prescribed, the model would inject all BB emissions at the planetary boundary layer. We did not pursue this, however, as the result would be comparing two different fire emissions inventories; the purpose of this study was not to evaluate fire emissions inventories, but to use flight campaign data to investigate the chemical and physical processes of the ModelE BrC scheme.

Minor issues:

4. The use of dashed lines in the vertical profile figures is confusing. The captions state that dashed lines “indicate altitudes with no data,” yet lines are drawn—implying interpolated or assumed values. This needs to be clarified.

The dashed lines are there to provide linear interpolation between points and help guide the eyes. Not connecting datapoints makes the vertical profiles difficult to interpret, but solid lines are avoided because that strongly implies the existence of data, hence dashed lines are used. We have added a statement to clarify this in the first vertical profile caption: “*Dashed lines provide linear interpolation over altitudes with no data to allow for easier interpretation of results.*” (Revised L439, under revised Fig. 3). This is also referenced in all proceeding vertical profile figure captions.

Overall, the model-observation comparison comes across as rather superficial. The paper presents a potentially useful dataset-model comparison, but it currently lacks sufficient detail in model description and scientific interpretation. I recommend a major revision to address the above issues before the manuscript can be considered for publication.

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