

We thank the reviewer for the helpful comments and suggested minor revisions that have improved the manuscript. This document includes comments from 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 new changes underlined if added to an existing sentence*. Any new references cited in manuscript excerpts have been added to the references section of the manuscript.

The authors have addressed the comments raised by the reviewers and the authors must be commended for developing an improved methodology to represent BrC absorption in climate models. A few minor edits are suggested below; I recommend publication of the manuscript after they have been addressed.

1) Equations 1 and 2 indicate the imaginary refractive index as k_{BrC} as opposed to k_{OA} . This is misleading as the imaginary refractive index calculated by Saleh et al. (2014) is for the OA mass and not the BrC mass. Also, equation 2 only holds if it is k_{OA} !

$E_{BrC} = (4\pi k_{OA} / \rho \lambda) / MAC_{BrC} \times E_{OA}$ (Equation 2)

$E_{BrC} \times MAC_{BrC} = MAC_{OA} \times E_{OA}$ (or the absorption from BrC = absorption from OA).

We specify k_{BrC} in equations 1 and 2, rather than k_{OA} , because we treat all absorption from OA as BrC absorption (as suggested by the reviewer). This treatment wasn't explicitly stated in the manuscript however, which led to the misleading nature of these equations. To resolve this, we have added the following text to revised submission L150: *"In this scheme, we consider total OA to consist of non-absorbing OA and BrC; any organic absorption is attributed to BrC."* We also added clarifying text to revised submission L175 to acknowledge the difference in k specified by Saleh et al. and our study: *"Since all organic absorption in our scheme is attributed to BrC, we use k_{BrC} in place of the k_{OA} specified by Saleh et al. (2014) for equation 1."*

2) The study limitations (section 3.4) include possible missing absorbing species and a limited understanding of the browning and bleaching of BrC. Differences between the optical properties of water-soluble and insoluble fractions may also be included here (Zhang et al., 2020; Liu et al., 2013; Satish et al., 2020; Laskin et al., 2015; Saleh et al., 2020). While the study does a great job of using presently available information on BrC absorption properties, further improvements on these limitations may change the findings of the limited influence of BrC refractive index or browning when considering the global radiative effect of BrC. For example, if water-insoluble darker BrC are included and have a reduced susceptibility to photo-bleaching (of course, only after experimental data becomes available), then their effect may be pronounced even at a global scale. This possibility may be acknowledged in the manuscript.

We agree this limitation should be discussed. We added the following paragraph to section 3.4, starting on revised submission L687: *"Another limitation of this scheme is that water-soluble and water-insoluble fractions of BrC were not differentiated or characterized with different optical properties and aging. This was done to be consistent with pre-existing OA representation in ModelE, as BB OA and biogenic SOA are not differentiated by solubility or hygroscopicity (Koch, 2001), and as mentioned in section 2.2.3, changes to prescribed BrC solubility affect total organic burden and skew estimates BrC radiative effect. Studies have shown, however, that water-insoluble BrC can be more absorbing than water-soluble BrC (Chen and Bond, 2010; Liu*

et al., 2013; Laskin et al., 2015; Satish et al., 2020). It is possible that chemical aging also differs between these two BrC types, for instance darker water-insoluble BrC being more resistant to bleaching, since reactions may proceed faster in aqueous phase (Hems et al., 2021).
50 Accounting for these differences in BrC solubility types could change model sensitivity to refractive index and aging. Further study on aging in water-soluble and water-insoluble conditions could clarify the potential impact of not differentiating BrC by solubility within ModelE.”

55 **3) In the response document (Line 271) it is shown that fixed-time ageing simulations show little variation from the base case that uses an oxidant-concentration-driven ageing scheme in terms of global RF but an oxidant-concentration-driven aging scheme has better spatial accuracies. Is this also not true for other properties like the BrC refractive index?**

In the response document, we identified spatial differences that may be important as they overlap with some biomass burning regions. We can't conclude, however, that these differences are necessarily more accurate. The oxidant-driven aging scheme was characterized as more accurate in the sense that it is more physically accurate because we know BrC aging changes with oxidant concentration—we differ to the scheme that's more reflective of observed chemistry. With regards to the reviewer question about refractive index: the effective refractive of BrC, an evolving value
60 observed in the atmosphere, is represented in ModelE as a mix of multiple BrC tracers. Since BrC absorbs more with greater imaginary RI, we would expect similar spatial differences
65 between fixed-aging and oxidant-driven chemistry for refractive index as we did with radiative effect. Without more observations of BrC RI however, which the recently launched PACE mission will afford us, we can't conclude if these differences are more accurate.

70 **4) From the response document, Line 471: “To briefly summarize, browner BrC (built up overnight) rapidly bleaches during the day. Since BrC can only have a radiative effect when there is insolation, and browner BrC is short-lived during daytime, bleaching is the dominant process with regards to radiative effect.”. This seems important in understanding why browning may not have a significant radiative effect and may be mentioned in the manuscript if it hasn't been mentioned explicitly already.**

A sentence stating this explicitly has been added to revised submission L663, following the statement that BrC radiative effect shows limited change with and without browning: “Such limited sensitivity to browning makes sense: BrC can only have a radiative effect when there is
80 insolation, and browner BrC is short-lived during daytime, so bleaching is the dominant process with regards to radiative effect.”

References:

Zhang et al., 2020: doi:10.5194/acp-20-4889-2020
85 Liu et al., 2013: doi:10.5194/acp-13-12389-2013
Satish et al., 2020: doi:10.1007/s11356-020-09388-7