

We thank the reviewer for constructive suggestions that help improve the manuscript. Below, we provide point-by-point responses, with reviewers' comments presented in black and our responses in blue.

General Comments

The manuscript by Xie et al. updates old assumptions about the reactivity of brown carbon (BrC), an important component of biomass burning emissions, by implementing a parameterization of its lifetime based on atmospheric conditions rather than just oxidant concentrations. They apply this parameterization to reassess BrC lifetime, its direct radiative effect (DRE), and its impacts on OH, O₃, and NO₂ photolysis. They also account for varying plume injection heights, improving upon previous studies (e.g., Schnitzler et al. 2022), which only modeled fast bleaching at low altitudes and no bleaching at high altitudes. Xie et al. use higher spatial resolution (altitude, longitude, and latitude), resulting in simulated BrC concentrations that better match observations. This is a significant advancement in understanding BrC's atmospheric role.

The paper is well written and important, but some statements are unclear, some arguments lack sufficient detail, and the limitations of the study should be discussed more thoroughly.

We thank the reviewer for recognizing the significance of this work.

Specific Comments

The limitations of the study should be discussed more thoroughly:

- The parameterization is based solely on reactions between O₃ and BrC, per Schnitzler et al. Other bleaching mechanisms should be acknowledged.
- The underlying study only considered BrC from smoldering pine wood, which may not represent atmospheric diversity.
- Only the water-soluble fraction of BrC was studied. The water-insoluble fraction, potentially more light-absorbing, was excluded.

Thanks for the suggestions. We have now added a Discussion section (Section 4) to discuss the limitations raised by the reviewer.

Section 4: *“While representing an important step forward, several aspects of the Schnitzler et al. (2022) parameterization merit further investigation. First, the experiment examined only ozone-driven bleaching. The role of other oxidants such as OH and their sensitivity to environmental parameters are yet to be explored. Second, the study focused on water-soluble BrC, though field evidence indicates that water-insoluble BrC accounts for a large fraction of BrC absorption. The impact of temperature and RH on bleaching behaviors of water-insoluble BrC remains uncharacterized. Third, the parameterization derives from BrC produced by smoldering pinewood, while real-world BrC properties may vary considerably across different fuel types and combustion conditions (Sun et al., 2021; Cai et al., 2023). Finally, neither the current study nor Schnitzler's work addresses secondary BrC*

formation pathways, which may involve different bleaching mechanisms. These limitations highlight important directions for future research to further refine the representation of BrC transformation in atmospheric models.”

Line 166–168: What is the reference for total OC, BC, and BrC emissions?

The emissions of OC and BC are based on a series of anthropogenic and natural emission inventories, which are described in Line 144-154 (Section 2.1). We now remove the sentence in Line 166-168 and state clearly in Line 155-158 that total OC and BC emissions are based on the sum of multiple emission inventories.

Line 167–168: “The total brown carbon emissions were 27.46 teragrams, of which biomass burning emissions were 21.17 teragrams.” Since BrC is typically a subset of biomass burning emissions, this needs clarification. Please verify sources and define what is included in each total.

We now clarify in Line 154-158 Section 2.1 that *“These emission inventories yield global OC emissions of 42.74 Tg and BC emissions of 8.24 Tg in 2019. Of the total OC emissions, 21.17 Tg are from biomass burning and 6.29 Tg from biofuel burning. In the simulation, these sources are treated as BrC (see Section 2.2 for details), while the remaining OC emissions are modeled as purely scattering aerosols.”*

Lines 215–231: The aging discussion references Wang et al. (OH) and Schnitzler et al. (O₃). Their lifetimes may not be directly comparable. This should be noted.

We appreciate your comment and agree that the BrC lifetimes associated with OH oxidation and O₃ oxidation cannot be directly compared, since they represent different chemical aging mechanisms and environmental dependencies. In this work, for the purpose of modeling, we use Wang et al. or Schnitzler et al. to parameterize bleaching lifetime for all BrC, without explicitly accounting for chemical mechanisms. Now, following the reviewer’s suggestion, we note both in the method description (Line 221-228 Section 2.2) and the newly added Discussion section (Section 4) that Schnitzler et al. experiment is based on O₃ oxidation and is for water soluble BrC only. We suggest that additional experiments are needed to improve on Schnitzler et al. parameterization.

Section 2.2: *“More recent laboratory experiments by Schnitzler et al. (2022) found that the chemical lifetime of water-soluble BrC, generated from smoldering pine wood in the presence of ozone, increases substantially with enhanced particle viscosity when temperature and RH get lower. Although the experimental setup (i.e., using ozone as the oxidant, generating aerosols from pine wood, and focusing on water-soluble BrC) does not fully represent atmospheric conditions, the finding that τ_{BrC} is highly sensitive to environmental parameters has critical implications for BrC’s radiative and chemical effects.”*

Section 4: *“While representing an important step forward, several aspects of the Schnitzler et al. (2022) parameterization merit further investigation. First, the*

experiment examined only ozone-driven bleaching. The role of other oxidants such as OH and their sensitivity to environmental parameters are yet to be explored. Second, the study focused on water-soluble BrC, though field evidence indicates that water-insoluble BrC accounts for a large fraction of BrC absorption. The impact of temperature and RH on bleaching behaviors of water-insoluble BrC remains uncharacterized. Third, the parameterization derives from BrC produced by smoldering pinewood, while real-world BrC properties may vary considerably across different fuel types and combustion conditions (Sun et al., 2021; Cai et al., 2023). Finally, neither the current study nor Schnitzler's work addresses secondary BrC formation pathways, which may involve different bleaching mechanisms. These limitations highlight important directions for future research to further refine the representation of BrC transformation in atmospheric models."

Line 469: Figure S7 is described as showing the response to wildfire emissions (reactive gases + aerosols), but its caption says it shows only BrC absorption without biomass burning. These statements conflict.

- Clarify what is meant by "BrC absorption without biomass burning emissions."
Is this just biofuel-derived BrC?

Thanks for pointing out this error. The caption now reads "Figure S8. The effect of biomass burning emissions (including all gas and aerosol species) on global surface O₃ (a), and column OH (b), computed as the difference between simulations with and without biomass burning emissions."

Line 472: References Figure S8, which does not exist. Without this figure, lines 473–476 cannot be evaluated.

Correction is made. It should be Figure S7 (Now as Figure S8).

Technical Comments

JNO₂ is used in the abstract without being defined.

The sentence in Abstract has been modified to "Additionally, ...reducing photolysis rate of NO₂ (JNO₂) by up to 7.4%..."

Line 82: Change "BrC around" to "BrC is around."

Thanks. The sentence has been changed as "Studies have shown that the direct forcing effect (DRE) is around +0.1 W m⁻²".

Line 86: "What more" is awkward phrasing. Consider revising.

Now it has been changed to "Moreover, ...".

Line 96: "Tropopause" may be incorrect; did the authors mean "troposphere"?

Thanks. It has been changed to "troposphere".

Line 243–246: Three systems are said to be used, but only two (MC-LWCC and NOAA PILS-LWCC) are listed.

It should be “two systems”. The correction is made.

Line 279: Change “In addition to 2019, but we also” to “In addition to 2019, we also” or simply “We also.”

The sentence is modified as “*In addition to 2019, we also run ...*”.

Figure S4 caption: Correct “ration of BrC” to “ratio of BrC.”

This mistake has been fixed.

Lines 474–476: “The inclusion of BrC light absorption suppresses [...] near-source OH enhancement, but amplifies OH reduction [...]”

- The use of “but” implies a contradiction. Consider revising to: “...suppresses near-source OH enhancement and amplifies OH reduction...”
- Same revision suggested for lines 516–517 in the conclusion.

These two sentences have been revised to “*Comparison of Figure S8 and Figure 8 shows that including BrC light absorption partially offsets the ozone enhancement from wildfire emissions. Moreover, BrC absorption reduces near-source OH enhancement caused by fires while reinforcing OH reduction globally.*” Similar revision has been made for Line 580 in Conclusion.

Lines 187–191: Sentence is unclear. Revise for clarity.

Section 2.2 (Brown carbon simulation) has been re-written for clarity.

Line 203: Reference [50] appears where an author name is expected. Ensure consistent reference formatting.

This mistake has been fixed.

The term “fresh brown carbon” is used but not defined. Clarify that it refers to unbleached BrC if that is the intended meaning.

Thanks for the suggestion. We now clearly define fresh brown carbon and bleached brown carbon explicitly in Section 2.2: “*To capture the change of mass absorption efficiency (MAE) due to chemical processing, organic aerosols from biomass and biofuel burning are modeled as two distinct species: fresh BrC, which is strongly absorbing, and bleached BrC, which is weakly absorbing, following the approach by Wang et al. (2018). All freshly emitted OA from biomass and biofuel burning are specified as fresh BrC. They are subsequently converted to bleached BrC in the atmosphere, with the rate governed by bleaching lifetime τ_{BrC} .*”

Lines 375–378: “The absorption of brown carbon becomes stronger with increasing altitude” is unclear. If referring to mass absorption efficiency, this needs clarification and justification.

This sentence is removed.

Line 435: The citation for Feng et al. appears to have the wrong year. Please verify

and correct.

This mistake has been fixed.