

Referee's comment: *Part of the basis of this study is to build upon the Bian et al. (2017) model. Hodshire et al. (2019) also built upon the Bian et al. (2017) model to test in a similarly theoretical model the effects of fire (plume) size and background aerosol concentration on the near-field aging of aerosol size distributions. This paper may be worth mentioning within the intro and may provide as an appropriate citation for portions of the near-field discussions of EnR.*

We thank the referee for pointing out this omission: unfortunately, we were not aware of the very recent paper by Hodshire et al. prior to completion of our study and submission of our manuscript to ACP. The corresponding reference is provided in the revised manuscript, and the Hodshire et al. (2019) paper is mentioned there several times in various contexts. In particular, we note (in the Introduction) that Hodshire et al. (2019) pointed out a significant impact of background aerosol on near-field BB OA aging processes, and (in Sect. 4) that our findings concerning the impact of the plume size on EnR after a few initial hours of aging are qualitatively consistent with the results of numerical experiments conducted by Bian et al. (2017) and Hodshire et al. (2019). We also tried to make it clear (in Sect 2.3) that the configuration of the numerical experiments in our study is largely similar to that in both Bian et al. (2017) and Hodshire et al. (2019).

Referee's comment: *Page 9: In Table 1 for the S15 scheme, the authors reference “fresh” SOG ( $\beta_{frag} = 0$ ) and reactions involving “aged” SOG ( $\beta_{frag} = 0.85$ ): it's clear where  $\beta_{frag} = 0.85$  come in from the text on pg 9 and equation 11. However, it's not clear where  $\beta_{frag} = 0$  would be in equations 9 or 10, if one to assume that  $\beta_{frag} = 0$  for the fresh SOG. Can the authors include this information within the relevant equations?*

We are sorry for this minor textual inconsistency. To address it, the description of the S15 scheme in Table 1 has been revised: we tried to make clear that a specific value of the fragmentation branching ratio is applicable only to oxidation reactions involving “aged” SOGs, while POGs and “fresh” SOGs are assumed to be not affected by fragmentation at all.

Referee's comment: *Page 10: Can the authors comment in the text on why they chose to only use the FragSVSOA configuration?*

A corresponding comment is provided in the revised manuscript. In particular, we note that the FragSVSOA configuration enables better consistency of the S15 scheme with the other VBS schemes considered in our study, and thus any differences between the simulations performed with the S15 scheme and the other schemes are easier to interpret. We also note that possible formation of NVSOA due to particle-phase reactions is among the factors (discussed more in detail in Sect. 4) that can affect the real BB OA evolution but that were not analyzed in our study, as it is focused on identification of major qualitative nonlinear effects in the BB OA behavior due to gas-phase oxidation reactions in BB plumes.

Referee's comment: *Page 13: “This period is representative of the typical lifetime of BB aerosol in Siberia under conditions without precipitation (Paris et al., 2009)”: can the authors comment on the relative lifetime of BB aerosol in other important fire environments, such as the Amazon, Africa, etc, in order to place the 120 hour designation into a broader context? Also, as the authors are choosing variables representative of Siberia (e.g. diurnal cycle) chose to use  $5 \mu\text{g m}^{-3}$  as their background aerosol concentration, can they comment on how well this is anticipated to represent the Siberian natural background during the fire season? The authors may also consider pointing out that only considering a relatively clean background is a limitation of the study, as entrainment of more polluted backgrounds will change the partitioning and evaporation rates of the plume particles (see e.g. Hodshire et al., 2019).*

In response to the Referee's comments, we have revised the second sentence of Sect. 2.3. We point out that the period of 120 hours has been chosen to be within the range of typical atmospheric lifetimes of submicron aerosol particles emitted from open vegetation fires in the major BB regions worldwide, as indicated, e.g., by a measurement-based estimate (5.1 days) of the lifetime of black carbon (BC) in Siberia (Paris et al., 2009) and global-model estimates (Wang et al., 2016) of the BC lifetimes for open fires in northern Africa (5.6 days) and northern South America (3.1 days).

In respect to our choice of the background OA concentration of  $5 \mu\text{g m}^{-3}$ , we note that the same value was specified in the box model simulations performed by Bian et al. (2017) and that, for comparison, particulate matter ( $\text{PM}_{10}$ ) in a boreal environment of central Siberia under background conditions (that is, without the detectable influence of local or regional pollution sources, including fires) was found by Mikhailov et al. (2017) to have concentrations ranging from about 2 to  $10 \mu\text{g m}^{-3}$  in summer, being composed mostly of organic material. Finally, we also note that specifying a much larger or much smaller value of the background OA concentration would likely result in noticeable quantitative changes of the simulated BB OA behavior, since entrainment of background aerosol affects evaporation rates and gas-particle partitioning in a BB plume (Hodshire et al., 2019).

*Referee's comment: Page 15: The authors could consider using only the 'EnR' or the ' $\gamma_a$ ' notation throughout, as having 2 different variable names for the same thing is a little confusing.*

We have carefully considered the Referee's suggestion. We would like to note that as explained in the reviewed manuscript before Eq. (19), "EnR" is introduced as an abbreviation (rather than a notation) for the "enhancement ratio". A corresponding notation ( $\gamma_a$ ) was introduced in Eq. (19) to allow us to present our quantitative results in a concise way. We hope that using both an abbreviation and a mathematical notation for the same physical characteristic is consistent with the standards of ACP. Nonetheless, to enhance the readability of the text, we tried to avoid (or at least, to minimize) using both  $\gamma_a$  and EnR in the same section of the revised manuscript. More specifically, the use of " $\gamma_a$ " is predominately reserved for presentation of quantitative results of our simulations in Sect. 3, while "EnR" is mostly used to discuss qualitative implications of our findings in Sects. 4 and 5. We hope that in this way a possible confusion between "EnR" and " $\gamma_a$ " has generally been avoided.

*Referee's comment: Section 3.1: Can the authors briefly justify in the text why they chose their given fixed value of initial mass loading for the analysis in Fig 2 and fixed value of plume size for the analysis in Fig. 3?*

A corresponding brief explanation is introduced in the first paragraph of Sect. 3.1 of the revised manuscript. Specifically, we note that the fixed values of  $C_0$  ( $10^3 \mu\text{g m}^{-3}$ ) and  $S_p$  (5 km) in the simulations shown in, respectively, Fig. 2 and Fig. 3 are chosen to approximately represent mid-range values of the corresponding parameters (on a logarithmic scale).

*Referee's comment: Also, why does Figure 4 (and Figure 8) only show results for the T18 and T18f schemes? Perhaps along with a brief justification for this choice the authors could also consider including the results of the other VBS schemes in a supplemental figure. I see that the authors note on lines 9-10 of page 20 "Note that only simulations for the "extreme" values of  $C_0$  and  $S_p$  (among those considered in this study) are shown in Fig 4. Simulations with other (intermediate) parameter values would fall between the brown and blue curves." But does this include all other schemes, that is, that all of the VBS schemes used fall between the brown and blue curves? This should be made clear in the text.*

Following the Referee's suggestion, results of our simulations with the C17 scheme have also been included in Figures 4 and 8 of the revised manuscript. In addition, we explain (both in Sect. 2.3 and Sect. 3.1) that the hygroscopicity parameter was calculated only with the C17, T18 and T18f schemes because the other oxidation schemes (K15, S17 and LIN) considered in our study are not designed to evaluate the O:C ratio. We also tried to make clear that in the text fragment cited by the Referee, we mean the simulations performed using a particular scheme corresponding to each plot in Fig. 4.

*Referee's comment: The authors could help the reader by being more explicit as to why smaller fires have higher hygroscopicity parameter values than larger fires. A brief sentence or 2 would go well on pg 20 (first paragraph) that clearly points out again that smaller fires can undergo more oxidation reactions (more gas phase material available both from initial partitioning and initial evaporation by dilution). Plume-size dependency on e.g. oxidation and partitioning is a complex subject that could aid from simple explanations and reminders such as this throughout.*

Following the Referee's suggestion, a corresponding explanation is introduced in Sect. 3.1 of the revised manuscript.

*Referee's comment: Page 26: Can the authors comment in the text on whether there is information to say how realistic each  $\beta_{frag}$  value is?*

Unfortunately, we are not aware of any strong experimental or theoretical constraints to the  $\beta_{frag}$  values involved in the BB OA oxidation schemes. To address the Referee's comment, we note in the revised manuscript that simulations with the extreme values of  $\beta_{frag}$  (0 and 1) can hardly correspond to any realistic situation and are provided only for reference purposes, but taking into account that the fragmentation branching ratio is a highly uncertain parameter of VBS schemes, neither of the simulations with the other values of  $\beta_{frag}$  is intended to be more or less realistic.

*Referee's comment: It is worth pointing out somewhere within the text (possibly in the methods?) that a limitation of this study is that the largest fires may be have limited oxidation reactions and thus limited SOA formation and/or fragmentation occurring within the dense initial plumes if the plume is dense enough to limit photochemistry, and that this study doesn't try to account for that effect.*

The suggested comment (with an appropriate reference to Konovalov et al. (2016) where the effect mentioned by the Referee was addressed using a 3D model) is included in Sect. 2.3 of the revised manuscript.

*Referee's comment: For each figure that has a horizontal dashed line at  $EnR=1$ , the caption should state that that line is there to indicate where 'no mass enhancement' occurs.*

The suggested explanation is included in figure captions in the revised manuscript.

*Referee's comment: Figure 2: What are the shaded bands on panel d? Presumably this is not a designation of 'nighttime'. They may be present to guide the eye but are confusing and should be explained. I recommend either removing them or making them distinct from the nighttime bands in panels a-c.*

There should have been no shaded bands in panel d. We are sorry for this technical error that has been corrected in the revised manuscript.

Referee's comment: *Figure 6 and 7: It's difficult to see the dashed line for  $C_{tot}/C_0$  within some of the panels (e.g. Fig. 6 panel a); can this line be made more distinctive?*

Visibility of the dashed lines for  $C_{tot}/C_0$  is improved in the revised manuscript by increasing their width.

Referee's comment: *Page 5 line 11: Bian et al., 2017 simulated 4 hours of aging (not 5).*

We thank the Referee for this correction. In the same sentence in the revised manuscript, we mention numerical experiments by Hodshire et al. (2019) along with those by Bian et al. (2017) and refer to the “first few hours” of BB OA evolution.

Referee's comment: *Page 8 line 17: are the SVOCs evenly distributed across the 5 bins? Please clarify.*

Actually, the SVOCs were not distributed evenly across the different volatility bins. To avoid possible confusion, the corresponding sentence has been revised. Instead of saying that the SVOCs were distributed across the 5 bins, we say that all SVOCs are represented using five volatility classes. The volatility distributions used in our experiments are specified in Sect. 2.3.

## References

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