

Referee's comment: *The use of chemistry transport models to simulate a single plume seems to be off-scale. Would the authors comment on the uncertainties associated with this disadvantage?*

We agree with the Referee that chemistry transport models (CTMs) are not designed to simulate a single BB plume and should not normally be used for this purpose. However, in practice, CTMs are generally applied to situations where the actual spatial inhomogeneity of BB OA emissions is not resolved in the simulations. This is what we meant in our remark about simulations of BB OA evolution with CTMs in the introduction (“While three-dimensional chemistry transport models are intended to provide the best possible quantitative representation of the evolution of OA and its gaseous precursors...”). To make our point clearer and to address the Referee’s comment, the following sentence has been included in the Introduction of the revised manuscript: “Note that while the spatial scales representative of isolated BB plumes are typically not resolved by chemistry transport models, simulations of a single BB plume with a box model can provide useful insights into possible uncertainties introduced by neglecting the spatial inhomogeneity of BB OA emissions in chemistry transport models at the sub-grid scales.” The nature of these possible uncertainties is further discussed in Sect. 4.

Referee's comment: *Page 10: Would the authors comment on why they did not include NVSOA formation in MDMOA? Or is it the conventional OA scheme that was mentioned on page 8 line 6?*

The main reason why we disregarded condensed-phase processes and NVSOA formation in this study was briefly explained in the paragraph devoted to the description of the K15 scheme (“In view of the lack of robust knowledge about the condensed-phase processes (see also Section 4) and for consistency with the other numerical experiments performed in the present study, the transformation of SOA into NVSOA has been disregarded in our simulations.”). In the revised manuscript, we provide a similar explanation but concerning the S15 scheme. Our choice of the FragSVSOA configuration for our experiments is further justified in the revised manuscript (specifically, in the description of the S15 scheme). In particular, we note that the FragSVSOA configuration (where NVSOA formation is disregarded) 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 which 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 line 24-26: Would the authors specify in the text each of the size bin's range? Also, which 3 size bins were used for T18 and T18f?*

In the revised manuscript, we specified that in the experiments with the C17, K15, S15, and LIN schemes, the aerosol size distribution included 9 size bins covering the range from 20 nm to 10 μm and following a geometric progression with the common ratio of $500^{1/9}$ (~ 2.0), while the experiments with the T18 and T18f schemes were conducted using only 3 size bins that were defined to cover the same range (from 20 nm to 10 μm) using a geometric progression with the common ratio of $500^{1/3}$ (~ 7.9). We believe that given this information, a reader can easily evaluate each of the size bin’s ranges.

Referee's comment: *Page 14 lines 3-7: “The concentration of OH based on the ambient measurements by Akagi et al. (2012), its value was set to $5 \times 10^6 \text{ cm}^{-3}$ in all our simulations. We also assumed a constant temperature of 298 K... ” Is a plume environment equivalent to ambient conditions? e.g. is it valid to assume a constant temperature of 298K and ambient OH*

concentration in a plume? Would limited photochemistry within a plume reduce OH concentration?

We are sorry for a somewhat misleading word “ambient” which we used to characterize the OH concentration measurements by Akagi et al. (2012). Actually, the OH concentration was indirectly measured by Akagi et al. inside of a BB plume. And indeed, the limited photochemistry within a plume is likely to reduce OH concentration. In the revised manuscript, our assumptions concerning the OH concentration and temperature and corresponding limitations of our study are explained more clearly. In particular, we note that the OH concentration within a plume can be affected by many factors (such as, e.g., the UV flux, the concentrations of nitrogen oxides and VOCs within the plume) which can cause variability of the OH concentration level across different plumes as well as temporal and spatial fluctuations of OH concentration within a given plume. We note further that temperature is also likely to vary, both spatially and temporally, within real-world BB plumes: in particular, it is likely to be lower in the upper part of a plume than near the surface. Finally, we argue that although all possible variability and inhomogeneities of the OH concentration and temperature were disregarded in our simulations, this limitation allowed us to isolate and investigate the internal dynamics of the BB OA system under fixed pre-defined conditions.

Referee's comment: *Page 14 lines 8: “Along with aerosol species, MDMOA has been configured to simulate the evolution of an inert tracer.” Would the authors please clarify the concept and use of an inert tracer (it was initially mentioned in the abstract)? What is its composition and properties?*

The requested clarification concerning the use and properties of an inert tracer is provided in the revised manuscript (Sect. 2.3). In particular, we explain that the tracer is intended to represent the evolution of the BB OA mass concentration in a hypothetical situation where BB aerosol is composed of chemically inert and non-volatile components, and so the tracer was introduced in our model as a chemically inert species which can be affected only by the dilution process (since the dry and wet deposition processes were not considered in our simulations); for definiteness, the molecular weight of the tracer has been set to be the same as of carbon monoxide (CO). We note that the concept of analyzing the evolution of BB aerosol versus the evolution of an inert tracer (usually represented by CO) has been fruitfully exploited in many previous experimental and modeling studies of BB aerosol. The corresponding references are provided in the revised manuscript.

Referee's comment: *Page 19, Figure 4, perhaps setting both figures with the same y-axis scale would be helpful. Also, in the text, it's unclear why only T18 and T18f schemes are shown, but not other schemes.*

Figure 4 is redrawn with the same y-axis scale, as suggested by the Referee. We also provided an additional plot (in the same figure) showing the simulations with the C17 scheme. At the end of Sect. 2.3 of the revised manuscript, we explain 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.

Referee's comment: *Page 24 “This observation indicates that the mass concentration of aged BB OA is likely to be much more strongly affected in the simulations by uncertainties in available representations of the BB OA evolution than the mass concentration of relatively fresh BB OA. One of the reasons is that fragmentation reactions become increasingly important with time when the SOG oxidation level increases, and then the competition between functionalization and fragmentation creates the more complex dependence on the plume parameters.” Would the*

authors please elaborate on or quantify the competition between functionalization and fragmentation? e.g. the branching ratios?

Indeed, the competition between functionalization and fragmentation can be quantified using the fragmentation branching ratio. To address the Referee's comment, we have modified one of the sentences cited by the Referee and included an additional sentence: "As a result of this competition, the outcome of the BB OA evolution becomes strongly dependent on the fragmentation branching ratio associated with a given OA scheme."

Referee's comment: *Page 32 lines 28-29: The authors mentioned that such differences among VBS are under the "typical conditions in summer mid-latitudes", it would certainly be intriguing for future studies to examine how these behaviors vary in different environments.*

We agree with the Referee and hope that future studies can use a similar approach to examine BB OA evolution under a wider range of environmental conditions. A corresponding remark is included in Conclusions.

Referee's comment: *Figures comments: It's unclear what the shaded greys represent in the figures, and they are quite distracting, I suggest that the authors justify them in the caption or remove them.*

The meaning of the shaded grey bands was explained in the caption for Figure 2 ("Shaded bands depict nighttime periods when no oxidation reactions were allowed to occur."). The same explanation is included in the captions for Figures 3, 4, and 10.

Referee's comment: *It may be already sufficient that figures have different colored lines. Adding different symbols are just adding noise to the figures (just as with the grey shades). But it could be a personal preference, just a suggestion.*

We appreciate the Referee's opinion, but we presume that some potential readers of our paper will prefer to print it out on a black and white printer that makes different colored lines indistinguishable. For such readers, we marked the curves with different symbols.

Referee's comment: *Again, it would be helpful to compare different schemes if the axis scales are the same when possible (in the same magnitude range).*

To address the referee's comment, we modified Figures 4, 6 and 7 (except for the panel b) accordingly. Note that not all of the panels have the same scale in other figures because our main intention was to clearly demonstrate the distinctive qualitative (rather than quantitative) features of the simulated BB OA evolution under different values of parameters.

Referee's comment: *For consistency among figures, I would suggest that Figure 4 follow other figures (Fig 3 etc) to include legends in the same locations.*

We thank the Referee for this reasonable suggestion. We have placed one of the figure legends (indicating a scheme) in Figure 4 into the left upper corner, similar to the legends in Figures 2, 3, and 5. Given the large size of the other legend in Figure 4, we preferred to show this legend in a separate panel. Most of the other figures have the legends placed uniformly into the right upper corner, as in many instances the left upper corner is filled with curves. We found that using the same layout for all the figures would not ensure the best visibility for our findings.

Referee's comment: *Figure 6's (and Figure 7's) caption mentions "dashed lines," although at first read (with-out looking at the legend's C_{tot}/C_0), it's unclear if it's the dashed lines with dots and dashes, or dashes alone (which should be C_{tot}/C_0). There two types of dashes, it'd be helpful to distinguish the two.*

We have made the dashed lines for C_{tot}/C_0 and dash-dot lines for "neutral" enhancements more distinguishable in the revised manuscript by increasing and decreasing the lengths of the dashes for the dash-dot and dashed lines, respectively.

Reference

Akagi, S. K., Craven, J. S., Taylor, J. W., McMeeking, G. R., Yokelson, R. J., Burling, I. R., Urbanski, S. P., Wold, C. E., Seinfeld, J. H., Coe, H., Alvarado, M. J., and Weise, D. R.: Evolution of trace gases and particles emitted by a chaparral fire in California, *Atmos. Chem. Phys.*, 12, 1397–1421, <https://doi.org/10.5194/acp-12-1397-2012>, 2012.