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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Insights into the aging of biomass burning aerosol from satellite observations and 3D atmospheric modeling: Evolution of the aerosol optical properties in Siberian wildfire plumes" *by* Igor B. Konovalov et al.

Anonymous Referee #2

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This study analyses changes in optical properties of Siberian biomass burning aerosols during their atmospheric transport using a combination of satellite observations and chemistry-transport model simulations (the CHIMERE model) over Eastern Europe. Similarly to their previous study Konovalov et al., 2017 (https://doi.org/10.5194/acp-17-4513-2017), authors use the enhancement ratios in AOD, AAOD and SSA due to the formation of organic aerosols from BB emissions relative to the corresponding enhancement of an inert aerosol tracer to investigate processes that occur during atmo-

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Discussion paper

spheric aging of BB plumes. I have several major concerns with the assumptions used in this study as well as the interpretation of the results that need to be addressed prior to publication.

Major concerns:

1) The representation of organic aerosol chemistry and processes within the CHIMERE model is expected to play a key role in the interpretation of the satellite observations, and in the conclusions of this paper. My concern is that the parameterizations used in this study are either somewhat outdated i.e. for biogenic and anthropogenic precursors, or have not been previously evaluated i.e. biomass burning precursors. For instance, the VBS parameterization used in this study for BB precursors was derived from the VBS proposed by Ciarelli et al., 2017 that provided a hybrid volatility basis-set model for aging of wood-burning emissions. It seems that organic compounds were lumped over several volatility bins, and given different properties and aging reactions (Table 2), and this was done without any constraint from experimental data. It is critical for this paper to demonstrate that the derived simplified mechanism provides accurate results. Authors should provide a box model simulation comparing their simplified VBS parameterization with the original one for various aging experiments, as well as comparing it with previously published experimental measurements (e.g. total yields) and/or other VBS parameterizations used for BB precursors (e.g. Shrivastava et al. 2017, Majdi et al., 2017).

The term “mechanistic (p4, p13, p30)” should not be used here to refer to the representation of BB organic aerosols in the CHIMERE model given that there is not process level representation of the underlying chemistry and optical properties.

2) The term “BB aerosol photochemical age” is misused in this study. As defined on page 7, this term does not account for the photochemical reactions or the chemical aging of the BB plume. It only accounts for the sunlight exposure of the plume, and should be referred to as “hours of sunlight exposure”. This needs to be corrected

throughout the manuscript and for corresponding figures (e.g. Figure 5).

Also, it is unclear how the transport time from the source region of a given BB plume was determined for the satellite data, and for the model. Please add this explanation to the methods section.

3) This study uses a large number of assumptions, e.g. parameterizations of aging of organic compounds, their optical and absorptive properties, fire emissions, averaging in time and space to match satellite measurements, etc. Please make a table that summarizes all the assumptions used in this manuscript, and quantify the associated sensitivity of the conclusions to this assumption. This is needed to show that the conclusions of this study are robust.

4) Does the proposed method allow separating between the changes in AOD due to oxidation and gas-particle partitioning vs. those due to dry and wet removal of organic gases and particles and subsequent evaporation/condensation. This needs to be clearly explained and justified.

5) Can the CHIMERE model capture the emissions and transport of the smoke plume during the studied period before all the corrections have been applied to the model? In particular, I am concerned about the coarse vertical resolution with only 12 levels up to 200hPa. What is the uncertainty in the transport and vertical distribution of smoke associated with this poor model resolution?

In addition, BB emissions were estimated using the satellite FRPs, and emission factors. What is the total amount of OA, BC, CO emitted by these fires during the period of interest, and how does this emission estimate compares with other publicly available emission inventories e.g. GFED or FINN. By how much were these emissions adjusted to match the satellite AOD data?

6) The analysis performed in this study are following very closely the approach used in the previous study by Konovalov et al., 2017 (except for the estimate of the photochem-

ical age, and the study of a different Siberian fire even). The originality and significance of the present study needs to be well justified with regard to the previous one in the discussion section.

Minor concerns:

The introduction is quite long and dense. Please try to shorten by avoiding the redundancies. Also the description of the modeling approach should be moved into the methodology section (p4 line 19 to p6 line5).

p1 line: 13: please remove “including the Arctic”. p1 line 13: change “Atmospheric evolution” to “changes that occur in”. p3 line 20 remove “recalcitrant” p5 line 1: take a step further instead of forward? p5 line 21 – remove “clockwise, and counterclockwise”. p9 line 21: Remove the parenthesis after tabs. p10 line 17: remove “numerous” p10 line 22: provide a reference for the melchior2 chemical mechanism, and for Fast-JX. p12 line 10: provide a reference for LMDZ-INCA boundary conditions. p17 line 11: What is the model resolution used in this study? And for this regridding? p22 line 5-8: these account for different airmasses?

Figure 4, should these AODs be compared quantitatively given all the adjustments that are applied to the emissions (p19 e.g. equation 9)?

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