Referee's comment: *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 atmospheric aging of BB plumes.*

We regret that the Referee apparently misunderstood the degree of novelty of our study. Contrary to the referee's statement, our previous paper (Konovalov et al., 2017), as evident from its abstract (https://doi.org/10.5194/acp-17-4513-2017), addressed the enhancement ratios only for AOD. This major difference between this study and the previous one is explicitly explained in the introduction of the reviewed manuscript (page 4, lines 6-9, page 5, lines 26, 27). This implies detailed work on the evolution of the optical and absorptive properties of aerosol in this work, by using a Mie code. Another major novel feature of this study is the development of an original analytical framework involving the use of satellite observations as constraints to the adjustable VBS parameterization of the biomass burning (BB) organic aerosol evolution (see the abstract of the discussion paper, and also page 3, lines 30, 31, page 5, lines 4, 11-15, page 29, lines 4-9 in the reviewed manuscript).

Referee's comments: *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 My concern is that the parameterizations used in this study are either somewhat outdated i.e. for biogenic and anthropogenic precursors, or have not been previously evaluated i.e. biomass burning precursors.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.*

We are not aware of any state-of-the-art parameterizations for biogenic and anthropogenic precursors that have been shown to allow improving 3D-model simulations of biogenic and anthropogenic aerosol in Siberia compared to simulations based on relatively simple parameterizations similar to those implemented in the standard version of CHIMERE. At the same time, we believe that our analysis could not be significantly affected by possible uncertainties in the modeled concentrations of anthropogenic and biogenic aerosol. This is specifically indicated by the facts that (1) the AOD level during the analyzed episode exceeded the background AOD level on the average by at least a factor of 6 (see Fig. 4 c, d in the reviewed manuscript) and (2) that the AOD simulations performed without fire emissions fit well the AOD observations in the periods when fire emissions were small (see Fig. S2). The sensitivity of the inferred evolution of the enhancement ratios for both AAOD and AOD to the assumed background conditions was examined in the supplementary section S3 of the reviewed manuscript. Regrettably, we see no evidence that the results of this analysis were taken into account by the referee.

Concerning the modeling of biomass burning precursors of aerosol, we argued in the introduction of the reviewed manuscript (page 3, lines 26-28 and above), that "… the sparse and often contradictory results of field and laboratory studies available so far can hardly provide consistent observational constraints for representations of the effects of atmospheric aging of BB aerosol in chemistry-transport and climate models". We also noted that "using the different VBS oxidation schemes partly constrained by laboratory measurements or atmospheric observations to simulate the multi-day BB aerosol evolution under fixed ambient conditions has been found to result in major quantitative and even qualitative differences between the simulations (Konovalov et al., 2019)" and that "it is not given that any of the available schemes can adequately describe the BB aerosol evolution specifically in Siberia" (page 13, lines 5-8). So, instead of using data of specific laboratory experiments (which are always representative of particular fuel types and a particular range of burning and ambient conditions), we constrained our simulations by satellite observations which are representative of numerous BB plumes in the real atmosphere. These constraints are shown to successfully take the role of the more traditional but sparse and limited constraints provided by "experimental" data from chamber experiments. The BB aerosol evolution simulated with the proposed VBS scheme is demonstrated to be consistent with the satellite observations as evidenced by results shown in Fig. 7 of the reviewed manuscript. Regrettably, we have no evidence that these results were taken into account by the Referee.

Referee's comment: *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).*

Regrettably, we do not see how the suggested comprehensive comparison of box model simulations involving different parameterizations could help us to "demonstrate that the derived simplified mechanism provides accurate results". Indeed, to the best of our knowledge, neither of the alternative VBS parameterizations proposed in the literature so far has been shown to enable an adequate model representation of the atmospheric evolution of BB aerosol in Siberia. Specifically, the original VBS scheme proposed by Ciarelli et al. (2017) was evaluated only against the measurements of organic aerosol (OA) produced from several burns of a particular type of fuel (beech logs) in a residential wood burner. Hence, there is no evidence that the original scheme is applicable to simulations of BB aerosol in Siberia.

In this situation, any difference or resemblance of results obtained with different mechanisms could hardly be indicative of any shortcomings in either of them. Furthermore, a numerical analysis employing a microphysical box model and several VBS schemes of various complexities have already been performed in Konovalov et al. (2019) for a wide range of conditions representative of BB plumes in Siberia. The main conclusions of the previous box model analysis, which are relevant for the present study, are summarized in the introduction and Sect. 2.4 of the reviewed manuscript, and we tried to improve this summary in the revised manuscript.

That said, we agree that a comparison of box model simulations with our simplified parameterization to similar simulations with the original (C17) scheme can be useful as it can provide evidence on whether or not our VBS parameterization enables a realistic representation of the initial stage of the BB aerosol evolution, which is poorly represented in the satellite observations used as constraints for our model. To this end, using the microphysical box model described in Konovalov et al. (2019) with both the VBS scheme used in this study and the original C17 scheme, we simulated the BB OA evolution under the conditions of chamber experiments reported in Ciarelli et al. (2017). The results of these simulations are mentioned in Sect. 2.4 of the revised manuscript and are presented in more detail in the newly introduced supplementary section S2. Briefly, we found that the BB OA concentration initially increases more rapidly in the simulation with our scheme than in the simulation with the original scheme, and so the BB OA concentration predicted by our scheme after about 10 hours of evolution is about 40% larger than the corresponding concentration predicted by the original scheme. Nonetheless, taking into account the range of the experimental variability of BB OA concentrations, the BB OA evolution simulated with our VBS scheme does not look unrealistic. A major qualitative difference between the two simulations is that the original scheme demonstrates a monotonically saturating increase of BB OA concentration, whereas the simplified scheme yields a non-monotonic behavior of BB OA concentration (a rapid increase followed by a gradual decrease due to fragmentation of SOA). Accordingly, a smaller concentration (also by about 40 %) is found in the simulation with our scheme after 110 hours of evolution. These two types of behavior of BB OA were earlier identified and explained in Konovalov et al. (2019).

Referee's comment: *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.*

The meaning of the term "mechanistic model" is somewhat uncertain and varies across different branches of science. Here we specifically meant that our scheme is expected to take into account basic physical "mechanisms" (such as oxidation/evaporation, a decrease or increase of the volatility of semi-volatile organic species, destruction of chromophores) driving the BB aerosol evolution rather than individual chemical processes (such as e.g., the oxidation chain of naphthalene). To avoid misunderstanding, we are not using this term in the revised manuscript, following the Referee's instruction.

Referee's comment: *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).*

We thank the Referee for this concrete comment. Since we do not have any observable photochemical "clock" at our disposal and cannot be sure that the model predicts OH concentration within the plumes accurately, we approximately quantify the exposure of BB aerosol to photochemical processes by assuming that OH concentration in the BB plumes is constant during daytime and is zero during the nighttime. Such an approximate estimate of the BB aerosol photochemical age accounts for the well-known fact that the photochemical processing of organic aerosol is typically much faster during the daytime than during the night time (with exceptions for highly unsaturated forested emissions which are efficiently oxidized also by NO_3 and O_3). In the revised manuscript, the corresponding definition in Sect. 2.1 is corrected accordingly, and an additional caveat is provided. The term "hours of sunlight exposure" is rather cumbersome and is difficult to use throughout the text, although we agree that it is more accurate (as it was noted in the reviewed manuscript).

<u>Referee's comment:</u> *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.*

We presume that the Referee refers to Sect. 3.2 where we identified a strong association between the photochemical age of BB aerosol and the geographical location (specifically, the longitude) of the BB plumes transported westward. This association, which is illustrated in Fig. 6 of the discussion paper, allowed us to estimate the ranges of the photochemical age of BB aerosol in the source and receptor regions. This analysis does not involve any other definitions apart from those already introduced in Sects. 2.1 and 2.3 (the photochemical age or hours of sunlight exposure of BB aerosol) and in Sect. 2.5 (coordinates of the source and receptor regions).

<u>Referee's comment:</u> *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.*

A table summarizing distinctive features and parameterizations of our modeling system is provided in the revised manuscript (Table 3). Furthermore, to address the Referee's comment, we have presented the discussion of the uncertainties in a more focused way in a new section (Sect. 3.4). This new section brings together the relevant content of Sects. 3.2, S3, and 3.3 of the reviewed manuscript, and also it includes some additional discussion.

We dedicated considerable efforts to ensure that the conclusions of our analysis are robust, and these efforts were comprehensively described in the reviewed manuscript. In particular, we quantified the uncertainties in the derived trends of the optical characteristics using a statistical (bootstrapping) method as described in Sect. 2.1. The obtained confidence intervals were shown in all our figures presenting the evolution of the optical characteristics according to both the satellite data and model results. Furthermore, a special section (Sect. S3) was dedicated to the analysis of test cases addressing possible biases associated with the processing of the satellite data in our study. The results of this analysis allowed us to conclude that "the tests overall confirm the robustness of our major findings". Regrettably, we see no evidence that these efforts were noticed and appreciated by the Referee.

While the main results of our analysis of the satellite data are quantitative, our simulations involving the VBS parameterization are mainly used to get insights into qualitative patterns of the BB aerosol transformations, rather than for quantitative characterization of any processes. This feature of our modeling analysis is emphasized in the introduction (page 5, lines 3-5) of the reviewed manuscript and rephrased in the next sections (page 11, lines 1, 2; page 24, lines 12-18; page 28, lines 32, 33). To "quantify" the associated sensitivity of the qualitative conclusions to any assumptions involved in our study is logically not feasible. Furthermore, we emphasized that our interpretation of the detected changes in AAOD, AOD, and SSA is not necessarily unique (page 27, line 30). At the same time, a discussion of several major factors which can potentially affect our conclusions (page 28) allowed us to conclude that "that although our model representation of BB aerosol evolution involves strong assumptions (which yet need to be verified in future research), our qualitative interpretation of the inferred major changes of the optical properties of BB aerosol in Siberia is sufficiently robust and realistic". We regret that this discussion also apparently went unnoticed by the Referee.

<u>Referee's comment:</u> *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.*

As explained in Sect. 2.1 (page 6, lines 14-16) of the reviewed manuscript, "the analysis of EnRs is expected to reveal the differences between the dynamics of AAOD or AOD in the real BB plumes and in a hypothetical simulation in which BB aerosol is assumed to consist of only non-volatile material and SOA formation processes are disregarded". This explanation implies that, by design, our method does not allow separating between the processes indicated by the Referee. We noticed, however, that the simplified formulation of the idea of our analysis in the reviewed manuscript (page 6, lines 11-14) could be somewhat confusing. In the revised manuscript, we clarified that the objective of our analysis was to identify the differences between changes in AAOD or AOD due to aging of BB aerosol in the real BB plumes (including, first of all, the changes associated with oxidation and condensation/evaporation and also the changes that can be indirectly induced by the dry and wet deposition of organic gases and particles) and those in a hypothetical simulation in which the organic fraction of BB aerosol is composed of only non-volatile, inert and hydrophobic material and the SOA formation is negligible.

Referee's comment: *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?*

The performance of our model in capturing the emissions and transport of the smoke plume during the studied period was validated by comparing our simulations and satellite observations of CO columns, but maybe the Referee did not notice this. This comparison was introduced in Sect. 3.1 and was presented in more detail in the supplementary section S2 of the reviewed manuscript (see also Sect. S3 of the revised manuscript). The spatial and temporal distributions of CO are driven predominantly by emissions and transport since the effects of other processes on the time scales considered are small. The spatial distributions of the CO columns during the analysis period according to the IASI observations and the CHIMERE simulations were shown in Fig. S1, while the corresponding temporal variations of CO in the source and receptor regions were compared in Figs. S3 and 4 (e, f) of the reviewed version (see Figs. S3 and S4 in the revised version). The satellite retrievals of CO columns are known to be sensitive to the CO vertical distribution (e.g., George et al., 2009), and this sensitivity has been taken into account in our simulations by applying the averaging kernels (page 17, lines 29, 30 of the reviewed manuscript). Hence, the comparison is indicative of the performance of CHIMERE in capturing not only the spatial distributions of emissions and horizontal transport of BB plumes but also the vertical distribution of CO. As evident in Fig. S1 (in the reviewed version), our model captures the observed spatial distribution of CO columns quite adequately, including the large-scale smoke plume extending from Siberia into the European territory of Russia. Fig. S3 further demonstrates a good quantitative agreement between the temporal variations of CO columns over both source and receptor regions (the correlation coefficient exceeds 0.9 in both cases). Finally, very small differences between the regional averages of observed and modeled CO columns (Fig. 4 e, f) indicate that the overall uncertainty in our simulations of the BB fraction of the CO columns over the receptor region (that is, within the strongly aged BB plume), including its part associated with a limited vertical resolution of our simulations is far less than the relative magnitudes of the variations in AAOD and AOD identified in our analysis. We would like to note that the vertical discretization of the simulations in our study was defined by taking into account not only available computational resources but also probable large random uncertainties associated with the vertical distribution of BB emissions (Sofiev et al., 2012). Given these uncertainties, a higher vertical discretization of our simulations would not necessarily result in smaller errors in the modeled vertical distribution of the smoke. We extended the discussion of our simulations of CO columns in the revised manuscript (specifically, in Sect. 3.1), taking into account the Referee's comment.

Referee's comment: *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?*

Estimation of emissions of aerosol and trace gases from fires is a subject of numerous dedicated studies (including our previous study (Konovalov et al., 2018)) but is not the focus of this one. Nonetheless, to address the Referee's comment, we have provided our top-down estimates for BB emissions of BC and OC in the study region and compared them with the corresponding estimates from the GFED4.1s inventory in Sect. 2.6 of the revised manuscript.

Referee's comment: *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 photochemical 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.*

We regret that the originality and significance of the present study went unnoticed by the Referee. As stated in the introduction of the reviewed manuscript (page 4, lines 6, 7), "this study substantially extends the scope of the previous one by analyzing satellite observations of both absorption and extinction characteristics of BB aerosol". We further noted (page 5, lines 26, 27) that "to the best of our knowledge, this is the first study attempting to constrain simulations of the aging behavior of BB aerosol with satellite observations of both absorption and extinction AODs". Hence, we believe that our study is highly original and that its main original features are clearly explained in the introduction of both the reviewed and revised versions of the manuscript.

The significance of our study is primarily associated with its general objective, such as "to find a way to infer statistically reliable information on the impact of aging processes on the optical properties of BB aerosol from available satellite measurements" (page 3, lines 28-30). We argue that achieving this objective is important because laboratory and field studies do not "provide consistent observational constraints for representations of the effects of atmospheric aging of BB aerosol in chemistry-transport and climate models" (page 3, lines 27, 28). The significance of the study is also emphasized in the last paragraph of the conclusions. Specifically, we note that "the presented analytical framework can be helpful in identifying and interpreting manifestations of the BB aerosol aging processes far beyond the time scales that can currently be addressed in aerosol chamber experiments" and that the proposed methods of the analysis of satellite data are sufficiently general and can be applied to different satellite observations.

The original results of our analysis include an identification of the statistically significant downward parts of the trends of the enhancement ratios for both AOD and AAOD during the multi-day aging of BB plumes as well the simultaneous increase of SSA and the enhancement ratio for AOD during the initial 20-30 hours of the evolution under daytime conditions. These are, to the best of our knowledge, unique results in the literature, and at least highly original. The interpretation of the detected changes using a VBS parameterization is certainly also a major novel point. These new results are discussed in the context of other studies (including our previous ones) in Sect. 3.2 and 3.3. Regrettably, we do not see any indications that the scientific content of these sections was examined and taken into account by the Referee.

Finally, we would like to note that although our idea to analyze satellite observations using modeled tracers was indeed introduced earlier, the concrete methods proposed in the reviewed manuscript are highly original. In particular, unlike Konovalov et al. (2017) we use a general algorithm for the nonlinear trend analysis. Differences between the analysis procedures used in Konovalov et al. (2017) and this study are discussed in the Supplementary Material, Sect. S1. The use of an adaptive VBS parameterization and explicit modeling of the optical properties of aerosol are also important novel methodological features of this study.

Taking into account the Referee's comment, we made an additional effort to emphasize the novel points of our study throughout the revised manuscript. We also tried to better explain the differences between this study and the study by Konovalov et al. (2017) in the introduction and conclusions of the revised manuscript.

Referee's comment: *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).*

The introduction is indeed relatively lengthy, but this is unavoidable because our study addresses not just one but several directions of research (such as modeling of BB organic aerosol, investigations of brown carbon, applications of satellite measurements of atmospheric aerosol, the role of aerosol from Siberian fires for the environment and climate). We tried but could not find any redundancies. However, following the recommendation of the referee, we have removed, for the most part, the description of our modeling approach from the introduction. In this way, the introduction has been noticeably shortened.

Referee's comment: *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"*

All the above textual changes suggested by the referee are done in the revised manuscript.

Referee's comment: *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.*

The requested references are provided in the revised manuscript.

Referee's comment: *p17 line 11: What is the model resolution used in this study? And for this regridding?*

The horizontal resolution of the model grid was 1 by 1 degree (page 11, line 19 of the reviewed manuscript). By saying that satellite data were projected on the model grid we implied that the regridding was done with the same spatial resolution (page 17, line 11), that is, 1 by 1 degree.

Referee's comment: *p22 line 5-8: these account for different airmasses?*

Yes, the conclusions in Sect 3.1 account for different airmasses, since the averaging of the observed characteristics over the big regions and period of several days allows us to suppress the variability associated with individual airmasses. A corresponding remark is added to the first paragraph in Sect. 3.1.

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

As explained in the reviewed manuscript (page 21, lines 3-5), the goal of the comparison presented in Fig. 4 and Sect. 3.1 was to "examine the aging-changes in the optical properties of BB aerosol by considering the satellite and simulated data for the "source" and "receptor" regions. In this context, while the adjustments applied to the BB emissions allowed us to bring the simulations close to observations in the source region, they obviously could not ensure the agreement of the simulations with the observations in the receptor region where the optical properties of BB aerosol could be affected by aging processes. Hence, we consider the discrepancies between the satellite observations and simulated data for the 'bb_trc' scenario in the receptor region as an indication for the BB aerosol aging, and we do not see any reason why these discrepancies should not be evaluated quantitatively. The comparison presented in Fig. 4 was, in our opinion, sufficiently discussed in Sect. 3.1 of the reviewed manuscript.

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