Referee's comment: *1. How is plume rise included in Chimere? What is the maximum vertical height of injection?*

As noted in the reviewed manuscript (page 11, lines 1, 2, 10), the fire emissions in our simulations were specified using a methodology that included calculations of the emission heights using the parameterization proposed by Sofiev et al. (2012) and was detailed in our previous papers. In response to the Referee's comment, a concise description of this parameterization and the information on its validation are provided in Sect. 2.3 of the revised manuscript. In particular, we note that the maximum injection heights of biomass burning (BB) emissions were calculated for each FRP pixel as a function of the observed FRP, the boundary layer height, and the Brunt–Väisälä frequency. In other words, the maximum vertical height of injection was not assumed to be constant but varied both in time and space. We also explain that the emissions for a given model layer were computed proportionally to the weighted number of pixels yielding the maximum injection height corresponding to this layer, with the weight of each pixel evaluated proportionally to the corresponding FRP. The average heights of the BB plumes considered in our analysis are reported in Figs. S8b and S6b in the Supplementary Material for the reviewed and revised versions of our manuscript, respectively.

Referee's comment: *2. A lot of model measurement evaluation is based on satellite data that have their own challenges as described in this study. Are there any in-situ measurements of organic aerosols at source/receptor sites? These would be valuable to constrain surface and vertical profiles of OA simulated by Chimere.*

Indeed, the satellite data which were used in our analysis are certainly not perfect. However, as pointed out in the reviewed manuscript (page 9, lines 6-12), the OMI (OMAERUV) AAOD retrievals were evaluated, both directly and indirectly (through the modeled BB emissions constrained with these retrievals), using AERONET, aircraft, and in situ data in Siberia as part of our previous study (Konovalov et al., 2018) which revealed no serious biases in these retrievals. The uncertainties in the MODIS AOD data are well documented and are relatively small (Levy et al., 2010). Accordingly, we see no evidence that the strong changes in the BB aerosol optical properties, which are identified in our analysis, can be an artifact of biases in the satellite data. Random uncertainties in the satellite data are, of course, quite considerable, but they are taken into account in our estimates of the confidence intervals for the derived tendencies in the analyzed characteristics.

Regrettably, we do not have any measurements of organic aerosols in the study region in the analyzed period at our disposal. But even if such measurements were available, they would hardly be sufficiently informative for our study: as we argue in the introduction, field studies show a very diverse picture of atmospheric transformations of BB aerosol, reflecting big diversity in fuel type and burning conditions, and so the sparse and often contradictory results of these studies can hardly provide consistent observational constraints for model representations of the effects of atmospheric aging of BB aerosol originating from multiple fires from a big region, specifically Siberia.

Taking into account the Referee's comment, we extended the discussion of possible uncertainties in our analysis, including those associated with the analysis of satellite data and the modeling, and provided a caveat concerning potential biases in the satellite data. This discussion is presented in the newly-introduced section (Sect. 3.4) of the revised manuscript.

Referee's comment: *3. Biomass burning emissions inventories like GFED often severely under-predict POA emissions from biomass burning. Can the authors provide any discussions/evaluations of primary BBOA emissions used in the model? Along the same lines, if inventories are missing a large BB-POA source, one could simply increase POA by a factor of 3 to 5, turn off BB-SOA formation and just age POA. This may result in different size evolutions (e.g. SSA, scattering efficiency), AAOD and AOD evolution. Would be interesting to see how this could affect the results and interpretations of this study.*

We thank the Referee for this insightful comment. Indeed, there is evidence (partly based on multiple studies that reported underestimation of AOD by models) that GFED and other inventories strongly underestimate POA (or OC) emissions from biomass burning. At the same time, the underestimation of AOD by a model can, at least partly, be caused by missing SOA sources in the model representation of organic aerosol, as argued, in particular, in one of our previous papers (Konovalov et al., 2015). So, in our understanding, there is a general problem concerning the evaluation of POA (OC) emissions provided by inventories. The results of our study have some implications in the context of this problem, even though the evaluation of the BB emission inventories is not its primary objective.

To address the Referee's comment, we provided a comparison of our "top-down" estimates of OC and BC emissions in the study region in July 2016 (Sect. 2.6 of the revised manuscript) with the bottom-up estimates obtained from the GFED4.1s inventory (van der Werf et al., 2017). While the top-down OC emission estimate for the 'bb_vbs' simulation scenario is found to agree well with the GFED data, the top-down estimate obtained using the 'bb_trc' simulation (which does not take into account the SOA formation) is almost a factor of 3 bigger than the corresponding bottom-up estimate (similar to what was found in our previous study (Konovalov et al., 2018)). As a caveat, we note that the good agreement of the bottom-up and top-down estimates of the OC emissions does not necessarily indicate that the emission inventory is accurate: ideally, the consistency of top-down and bottom-up estimates of the BB OC emissions should be examined by taking into account the partitioning of the measured emissions between gases and particles, but the corresponding data are not provided as part of emission inventories.

Following the referee's instructions, we also performed an additional simulation (referred to as 'bb_poa') in which the SOA formation was turned off but POA emissions were strongly increased (specifically, by a factor of 4.3) to retain the consistency between the simulated and observed AOD in the source region. The results of this simulation are summarized in Sect. 3.3 and presented in more detail in the Supplementary Material (Sect. S5). Consistently with our conclusions about the major role of SOA formation in the evolution of BB aerosol during the analyzed event, the simulations for the 'bb_poa' scenario are not found to exhibit any qualitative nonlinear features of the behavior of SSA and EnR for AOD, which were identified in the analysis of the satellite observations and were reproduced in the simulations for the 'bb_vbs' scenario. We consider this result as evidence that the proposed joint analysis of satellite and model data can be used to distinguish between the two possible reasons for the underestimation of AOD by a model, such as (1) an underestimation of OC emissions in the emission inventory and (2) missing SOA sources in the model.

Referee's comment: *4. Page 18: Its confusing as written: " We selected only those simulations that have the same ranks as the subset of CO observations matching (as described above) the OMI AAOD data". Please elaborate. How did the authors calculate ranks? Did they consider different simulations driven by different renalyses to minimize plume dispersion errors?*

We are sorry that the sentence indicated by the referee was confusing, and we tried to explain our procedure more clearly in the revised manuscript. As it is common in statistics (https://en.wikipedia.org/wiki/Ranking), the ranking of a set of data points was done in our case by assigning the order number to values arranged in ascending order. We'd like to note that while the selection procedure based on the ranking of the CO columns retrieved from satellite observations and simulated with CHIMERE was introduced to ensure more accurate quantitative results of our analysis, our main conclusions concerning the qualitative features of the BB aerosol evolution do not depend on the application of this procedure.

Unfortunately, we could not perform simulations based on different reanalyses in the framework of this study due to computational limitations. We are also not sure that the use of different reanalyses could result in a major reduction of the differences between the simulated and observed data, since these differences appear to be more due to uncertainties in the temporal and spatial variability of fire emissions rather than due to model transport errors.

Referee's comment: *5. Table2: In equation R3 how were coefficients 0.33 and 0.30 determined? Also in R5 and R6 85% of SOA is lost by fragmentation. Is the fragmented SOA recirculated or does it react to yield some MV-SOA/LV-SOA?*

The coefficients 0.33 and 0.3 were defined to retain the same total SOA yields for HV-SOA and LV-SOA from the oxidation of NTVOCs (~ 43 g mol^{-1}) as in Ciarelli et al. (2017) (page 14, lines 16, 17 of the reviewed manuscript), taking into account that the assumed molecular weights of HV-SOA and LV-SOA are 131 and 144 g mol^{-1}. In the revised manuscript (Sect. 2.4), we additionally explain that we took into account the ratio of the molar masses of these species ($131/144 \cong 0.30/0.33$). The products of the fragmentation pathway of reactions (R5) and (R6) were considered as volatile and did not contribute to the SOA formation (page 14, line 27 of the reviewed manuscript).

Referee's comment: *6. Figure 2: Why does Chimere have too high AOD compared to MODIS is South-Eastern part of domain?*

We addressed this question in Sect. 3.1 of the revised manuscript. We suppose that the simulated AOD is biased high outside of the study region in the south-eastern part of the model domain as a result of spatial variations in any parameters determining (or affecting) the relationship between the real biomass burning rate and the calculated aerosol emissions. Such parameters include, in particular, the emission factors, volatility distributions for the POA emissions, and empirical factor relating FRP to the BB rate. The variations in these parameters may, in turn, be due to the inhomogeneity in the spatial distribution of the vegetation species across the study region and model domain. It is noteworthy that the high bias in AOD is not mirrored in the spatial distribution of the simulated CO columns (see Figs. S1 and S2 in the Supplementary Material of the reviewed and revised versions of the manuscript, respectively).

Referee's comment: *7. Page 22 Before section 3.2: Can the authors comment on relative importance of various processes affecting decrease of AOD to AAOD from source to receptor, e.g. fragmentation, evaporation etc.?*

Our in-detail interpretation of the processes affecting the changes in AOD to AAOD during the transport of BB plumes from the source to receptor regions is provided in Sect. 3.3 of both the reviewed and revised versions of the manuscript. In the revised manuscript, we provided an additional remark in Sect. 3.1 that the additional changes (that cannot be explained by dilution) in AOD to AAOD are primarily caused by the losses of the medium-volatility fraction of SOA due to fragmentation.

Referee's comment: *8. Figure 10: Seems we need another case showing BC core with absorbing OA shell and lensing effects.*

Figure 10 shows our computations only for several test cases. The respective results for the base case (where, as suggested by the referee, the BC core is surrounded by the absorbing OA shell and the lensing effects are taken into account) are shown in Fig. 7a. In the revised manuscript, a corresponding explanatory remark is introduced in the caption of Fig. 10.

Referee's comment: *9. How does particle water affect calculated optical properties, especially if water is on the shell?*

Since the relative humidity (RH) characterizing ambient conditions for the BB plumes analyzed in our study typically remained below 70 % (see Figs. S8a and S6a in the reviewed and revised versions of the manuscript, respectively), we did not expect, based on the common knowledge (e.g., Reid et al., 2005), that the water uptake by particle could significantly affect BB aerosol optical properties and their evolution. Indeed, according to our simulations, the water mass fraction stays, on the average, below 15% as long the photochemical age is less than 60 h, eventually increasing up to 26% toward the end of the evolution period considered (see Fig. 8a in the revised manuscript). Possible effects of the water uptake on our simulations are discussed in Sect. 3.3 of the revised manuscript. Specifically, we argue that water and inorganic ions increase the scattering cross-section of BB aerosol particles, thereby increasing AOD and SSA, although the contribution of a unit mass of water to the scattering efficiency is expected to be considerably smaller than that of the organic matter because the real component of the refractive index for water is substantially smaller than for the organic species. We also point out that a substantial increase in the water uptake occurs in our simulations only after 60 h, and therefore it could not contribute significantly to the strong increase in EnR for AOD before 30 h. The water uptake by the organic shell of the particles can also contribute to the lensing effect, thereby increasing AAOD, but the evolution of EnR for AAOD in our case was found to be determined by other factors anyway, as illustrated in Fig. 10. In addition to this discussion, we explicitly estimated the effects of water on AOD, SSA, and the scattering efficiency in the 'bb_poa' test simulation mentioned above. These estimates, which are discussed in Sect. S5 of the revised Supplementary Material and are presented in Fig. S8, confirm that the water uptake of particles was not among the key factors that drove the evolution of the optical properties of BB aerosol during the analyzed episode.

Referee's comment: *10. It would be intuitive to see a map of particle water over the domain, especially given discussion of potential importance of heterogeneous oxidation of biomass burning OA. I would think that even with hygroscopicity of 0.2 water content of BBOA will be large due to its high concentration.*

The calculated mean water content in the BB particles is shown in Fig. 8a of the revised manuscript as a function of the photochemical age. As noted above, it turned out to be relatively small, since the ambient relative humidity in the center of mass of the BB plumes typically did not exceed 70 %. We presumed

that Fig. 8a would be more informative than a map suggested by the referee, because the calculated mean water fraction takes into account the spatial inhomogeneity of the aerosol mass loadings in the atmospheric columns, giving larger weights to the high loadings. As the manuscript and Supplementary Material are already lengthy, we opted to not provide one more figure (which would also entail additional discussion). Note that Fig. 8a provided in the reviewed manuscript was meant to implicitly show the contribution of water among "other" components. However, thanks to the Referee's comment we noticed that the model output data for these components were not processed properly. We apologize for this oversight. A corrected and improved version of Fig. 8a, in which the contribution of water is shown explicitly, is provided in the revised manuscript.

Referee's comment: *11. Can the authors comment on role of photolysis of biomass burning SOA as its loss process?*

A corresponding comment is provided in Sect. 3.4 of the revised manuscript. In particular, we mention the available global-scale estimates (Hodzic et al., 2015) of the SOA lifetime with respect to the in-particle photolysis, and also note that if the in-particle SOA photolysis were a primary driver for the decrease of AOD in our analysis, then the rate of the decrease in EnR_{ext} would not be expected to strongly decelerate after about 70 h, as it does according to Fig. 5b.

Referee's comment: *12. Page 28: If SOA is glassy, it may not mix with POA. How are the authors treating absorptive gas-particle partitioning of POA-SOA mixtures. One could envision treating them as two separate solutions from Raoult's law perspective.*

We made it clear in the revised manuscript (Sect. 2.4) that following Ciarelli et al. (2017) we assumed, for definiteness, that all organic species within particles form a well-mixed liquid and inviscid solution. Possible implications of this assumption – which cannot be validated or invalidated using available observations in Siberia – are briefly discussed in Sect. 3.4 of the revised manuscript. In particular, we note that if POA and SOA species do not mix in reality but form two separate solutions (as assumed, e.g., in Shrivastava et al., 2015), then our model is likely prone to overestimation of their concentrations in particles. It is, however, hard to see how these potential biases can invalidate our qualitative interpretation of the major changes in the BB aerosol optical properties. We plan to address the possible effects of viscosity and non-ideality of organic solutions on the Siberian BB aerosol evolution in our future studies.

Referee's comment: *13. Would the increase of brown carbon with BC to OA ratio imply LV-SOA is more absorbing (brown) than MV-SOA? I recall this was implied in Saleh et al. study referenced by the authors. But this is contrary to the author's hypothesis that LV-SOA is much less absorbing than MV-SOA. This may need some discussions.*

This point is briefly discussed in Sect. 2.4 of the revised manuscript. Specifically, we note that our assumption that LV-SOA is non-absorbing does not contradict the experimental findings indicating high absorptivity of low-volatility organic compounds (Saleh et al., 2014), because the contributions of POA and SOA species to absorption were not isolated in these experiments, and the effect of SOA addition was found there to be comparable to measurement uncertainties.

Referee's comment: *14. In Conclusions, the authors say 100h processing decreases ENRs for AOD and AAOD by 45% but not SSA. Why? One would expected SSA to change as the size distribution shifts with changes in evaporation, fragmentation etc. Would be nice to show change in size distributions simulated by Chimere with different processing times.*

SSA was evaluated in the standard way as 1-AAOD/AOD according to Eq. (6). So, as both AOD and AAOD decrease (due to various processes, including those indicated by the referee) to about the same extent, SSA is not significantly affected. The average size distributions of BB aerosol particles for different processing times are shown in Figs. S9 and S7 of the Supplementary Material of the reviewed and revised versions of the manuscript, respectively. We are sorry if we did not understand this comment of the Referee properly.

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