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Reply on RC1

Igor B. Konovalov et al.

Author comment on "Inferring the absorption properties of organic aerosol in Siberian biomass burning plumes from remote optical observations" by Igor B. Konovalov et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-151-AC2, 2021

We thank the Referee very much for the positive evaluation of our manuscript. We are also grateful to the Referee for the useful comments, which were carefully addressed in the revised manuscript. Our point-by-point responses to the Referee's comments are provided below.

1. Strongly suggest edit title to read "in Siberian biomass burning" given that the methods were developed using Siberian relevant parameter ranges and applied to AERONET observations in this region.

As we tried to convey in the abstract and conclusion, our study focuses on two major points. First, we developed a new and rather general method to infer the absorption parameters of the organic fraction of BB aerosol along with the BC/OA ratio. And second, the capabilities of our method are examined using AERONET observations of BB aerosol in Siberia. In our understanding, the title of the reviewed manuscript more focuses on the first point but does not exclude the second one, since Siberian BB aerosol can be considered as a specific case of BB aerosol. However, we find that the title suggested by the Referee is also not contradictory to any of the above points, although it puts much more emphasis on the second point. Taking this into account along with the fact that possible applications to BB aerosol in other regions of the world will likely require some adjustments of the a priori distributions, we have opted to add the word "Siberian" in the title of the revised manuscript, following the Referee's suggestion.

2. Line 176: How uncertain is the wavelength dependence? how would this impact the results (e.g. if you used the wavelength dependence of McClure et al., 2020 instead)?

First of all, we would like to note that our computations presented in the reviewed manuscript involved the assumptions based on the analysis and data by Lu et al. (2015). In particular, we assumed that probable values of w can vary within a wide range – from 0.5 to 4 and that they tend to be clustered around a specific empirical dependence of w on BC/OA. Note that, within the range of BC/OA ratios relevant for our study, the empirical dependence reported by Lu et al. (2015) is similar to that suggested by Saleh et al. (2014). However, according to the results of a recent lab study by McClure et al. (2020), values of w for fresh BB aerosol in the range of BC/OA ratios relevant for Siberian BB aerosol are typically larger (up to a factor of 2) than those reported by Saleh et al. (2014) and Lu et al. (2015). Taking into account that Lu et al. derived estimates of w from a variety of lab and in situ measurements, we assumed that these estimates are sufficiently representative (as a priori estimates) of BB aerosol in Siberia and that the difference with the values reported by McClure et al. (2020) can mostly be due to specific experimental conditions in McClure et al. (2020). For this reason, we did not consider data from McClure et al. (2020) in our computations presented in the reviewed manuscript.

We would also like to point out that as mentioned in the reviewed manuscript (lines 493, 494), the wavelength dependence cannot be well constrained only by the observations considered in this study. That is, the a posteriori estimates of the wavelength dependence are uncertain and can hardly be of practical use. However, this uncertainty is taken into account in the confidence intervals for the estimates of the inferred properties and does not invalidate our estimates.

To answer the Referee's questions, we demonstrated the wavelength dependence estimates derived from synthetic data (see Fig. S2 in Supplementary Materials for the revised manuscript). The estimates confirm that variability of w cannot be well predicted even when the input data are not affected by the observational error. At the same time, the presented analysis also demonstrates that our algorithm does not introduce any significant bias into *a posteriori* estimates of this parameter.

We also considered a special test case (referred to as test case 3 in the revised manuscript), in which the assumed a priori distribution of w corresponds to McClure et al. (2020) instead of Lu et al. (2015) (as in the base case). Furthermore, to make our a priori estimates for the base case inclusive of the range of values of w according to McClure et al. (2020), we increased the upper bound for the range of a priori estimates of w from 4 to 6. The results for the test case and the base case (see Supplement Figs. S7 and S8) are found to agree within the uncertainty of the a posteriori estimates for either of the cases, while the average values of the inferred absorption parameters are found to be insignificantly (compared to the range of the unconstrained values) smaller than for those for the base case.

Note that our estimates for the base case, which are presented in the revised manuscript, are not the same as our estimates presented in the reviewed manuscript, as the revised computations have been designed to include the effects of the coarse mode, which were mostly disregarded in the reviewed manuscript (please see our response to the comments of Referee #1 for details). The estimates assuming that coarse mode particles are non-absorbing are presented in the revised manuscript as test case 1. However, these estimates are still slightly different from those presented in the reviewed manuscript as a result of the mentioned change in the a priori constraints for w and re-sampling of the look-up table. Please note also that a common exclusion criterion (dependent on the samples in the look-up table) which is explained at the beginning of Sect. 4.3 of the revised manuscript was applied to the estimates presented in both the reviewed and revised versions of the manuscript but was regrettably omitted in the reviewed text.

3. Lines 203-206: Did the authors consider showing plots of the PDFs of parameters? This might be a useful visual to demonstrate adequate sampling of values.

According to our Bayesian algorithm, we did not explicitly derive PDFs of the inferred parameters and characteristics. Instead, we computed directly only integrals of the PDFs by using a Monte Carlo method (according to lines 227-234 in the revised manuscript). More specifically, we calculated the best estimates of the inferred characteristics by integrating PDFs according to Eq. 9, and, to obtain the confidence intervals for the a posteriori estimates (as explained in lines 234-237 in the reviewed manuscript), we implicitly considered corresponding cumulative PDFs.

To address the Referee comments, the examples of cumulative PDFs (CPDFs) for the four parameters were explicitly computed for one data point from the synthetic data set and are shown in Fig. 4 of the revised manuscript. In addition to CPDFs calculated for the base case (when the observation vector included three components: $AAE_{440/870}$, $AAE_{440/870}$, and SSA_{440}), Fig. 4 shows unconstrained CPDFs and those calculated by using only two AAEs or only $AAE_{440/870}$ and SSA_{440} as constraints to the inferred parameters and characteristics. In all the "constrained" cases, the CPDFs are distinctly different from the unconstrained CPDFs. The use of the synthetic data allowed us to demonstrate the adequacy of the sampling, specifically by comparing the confidence intervals determined by the CPDFs with the "true" values of the parameters.

4. Line 184: What fraction of the distribution was removed due to truncation?

In the revised manuscript, we noted that the truncation was mostly (but not always) done at one sigma range (that is, it removed about 32 % of the distributions).

5. Line 220: Could you comment on whether statistical independence is a good assumption for the parameters used here?

The corresponding comment is added to the revised manuscript right after Eq. (6).

6. Lines 258-263: The authors might consider discussing the implication of using only Level 2 data on the general application of this method in the Conclusions, i.e. skewed sampling of high AOD, and whether this would limit BrC estimated using this approach to near-source and perhaps not be appropriate for constraining photochemical aging in Siberia or other regions of the world.

We thank the Referee for this insightful comment. Indeed, constraining photochemical aging in Siberia or other regions of the world with AERONET data is challenging. On the one hand, as the Referee properly indicated, the use of only quality-assured (Level 2) data can result in a skewed sampling of dense BB plumes. In such plumes, BB aerosol composition is, to a significant extent, determined by semi-volatile organic compounds from a medium/high volatility range, whereas low-volatility volatility organic compounds that determine the composition of BB aerosol in highly diluted plumes may feature different absorption properties, as suggested, e.g., by our recent analysis of the evolution of BB plumes from Siberia fires (Konovalov et al., 2021). But on the other hand, even if the Level 1.5 AERONET data were quite reliable, there would be a problem with distinguishing between the absorption associated with BB aerosol and that by background aerosol. Note that in this study, we disregarded the contribution of background aerosol by selecting the AERONET observations with high AOD (AOD_{550}>0.8), in which BB aerosol contribution is presumably predominating: this criterion is typically stronger than that used in the Level 2 AERONET data (AOD_{440}>0.4). To address the Referee comment, a corresponding discussion has been introduced in the Conclusions of the revised manuscript.

7. Line 305: RH values in Figure 1 seem to go up to 80%. Please correct the text with this value or modify phrasing to say that values generally range between 40 and 70%.

We presume that the Referee refers to our phrasing on lines 305 and 306 of the reviewed manuscript: "Values of RH varied between 25 and 70 %, thereby confirming our a priori assumption that occurrences where RH in Siberian BB plumes exceeds 70 % are very rare". This phrasing was intended to be understood in the context of the whole paragraph that begins from line 298 and discusses the "episodes of major enhancements of AOD_{500} over the background fluctuations in 2012". That is, we meant only the range of RH values corresponding to observations of the major BB plumes, whereas the occurrences with high RH values in Fig. 1 correspond to the background conditions. To avoid confusion, we added the words "in the selected episodes" after the words "Values of RH varied between 25 and 70 %" in the revised manuscript.

8. Lines 327-329: Missing definition of sigma3

Indeed, a definition of σ_3 was not provided on lines 327-329, that is, immediately before Eq. (10). However, we strived to define iton lines 319, 320 in the previous paragraph (Accordingly, we used the values of U27 for SSA_{440} as estimates for the standard deviation σ_3). To improve the readability of the text, we defined σ_3 immediately after Eq. (11) (former Eq. 10) of the revised manuscript.

9. Figure 2: The legend or caption should clearly state which BC:OA corresponds to open/filled points.

We have redrawn Fig. 2 to provide legends for each open or filled symbol. In addition, following the recommendations of Copernicus Publications, we avoided the parallel usage of green and red that might cause problems for readers with color blindness.

References

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