Atmospheric Measurement Techniques Discussions

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Reply on RC2

Igor Konovalov

Community 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-CC1, 2021

In this short comment, I would like to briefly address the major points raised by Referee # 1. I appreciate the Referee's comments, even though I respectfully disagree with many of them. I will further appreciate it if the Referee clarifies some of the comments, as I'm not sure that I understood them properly. A comprehensive response (on behalf of all the authors) to the Referee's comments will be published later.

1. First of all, I would like to stress out that we do not attempt to find a unique ("most probable") combination of the aerosol parameters that fit the observed properties (as presumed by the Referee). Instead, using the Bayesian framework, we try to take into account a broad range of the probable parameters' values. It is not expected that each combination of the parameters is representative of the observed aerosol and would enable reproducing the observed radiance field (when values of all other parameters would be taken from the corresponding AERONET retrieval). All these combinations are just a priori guesses (a "cloud" of possibilities) that are specified in a rather arbitrary way. The use of the observational data results in, effectively, eliminating those combinations which are inconsistent with the set of observations (that is, two AAEs and SSA in our case). The "surviving" combinations (samples) represent the uncertainty of the a posteriori estimates of the retrieved parameters. Not all of these combinations are still expected to be representative of the observed aerosol, but this possible inconsistency cannot per se result in biases in our retrievals, as our algorithm integrates over the numerous samples in accordance to Eq. (9). I agree with the Referee that it would be unsound to consider only the optimal combination of the parameters, particularly because such a combination could yield spurious values of the unobserved parameters (including those characterizing the particle size distribution). However, this is not our approach.

Hence, we did not attempt to "re-model" AERONET observations. Rather, our study should be regarded as an extension of multiple studies that attempted to interpret the AERONET data on the wavelength dependence of the imaginary refractive index and other absorption characteristics and to derive the constraints on the BrC absorption. An advantage of our method is that it allows avoiding any subjective assumptions about the wavelength dependence of AAE in the absence of BrC. Even more importantly, unlike the methods used in previous studies, it allows consistent estimation of the uncertainties in the derived absorption characteristics of organic aerosol due to both the observational errors and the lack of knowledge about the microphysical structure of the observed aerosol.

2. I recognize that we should have acknowledged and discussed that AAEs and SSA are computed by AERONET from a model of internally mixed aerosols that have the same wavelength-dependent refractive index for all particle sizes. In my understanding, this limitation means that AAE values derived from the AERONET observations can be affected by the retrieval errors that covariate across the wavelength spectrum, but this is hardly surprising. To my knowledge, these covariances are unknown and have not been addressed in the literature. However, they are hardy significant because the size distribution retrieved by the AERONET algorithm (Dubovik et al., 2000) is highly flexible (the optimization is performed for the 22 size bins), and so it is probable that any covariances in the retrieved values of AAODs can be "broken" due to flexibility of the size distribution. Disregarding the error co-variances in our estimates of δBrC and k_{OA} probably results in overestimation of the uncertainties for these parameters, since the assumption of independent errors in components of the observational vector means more flexibility for components of the parameter vector. This probable overestimation is perhaps a shortcoming of our study. However, I believe that the significance of this shortcoming can be properly evaluated only within a proper scientific context. To this end, it could be pointed out that uncertainties in the AAE-based estimates of the contribution of BrC to absorption have not been estimated (at all!) in most of the previous studies.

3. Our model allows us to calculate intensive optical properties of aerosol but it does not include the radiative transfer code. Therefore, we cannot calculate the radiances and compare them with the observations. I believe that our calculations of the radiances would be pointless anyway. Indeed, the degree of agreement between the measured and calculated radiances would depend on many parameters that are not estimated in our study. Similar to the estimates of the BrC absorption, which were reported in numerous studies previously, our estimates of the absorption parameters cannot be evaluated directly against observations of the Sun and sky radiance measurements.

4. I am not sure that I understood the Referee's comments concerning Eqs. (1) and (2) properly. As noted in the manuscript, these equations explain how we calculated the relative contribution of BrC to the total absorption at 440 nm and MAE of OA. Specifically, Eq. (1) involves the mass absorption efficiencies that were calculated with the OPTSIM software. These efficiencies depend on the aerosol chemical composition and specifically on the BC/OA ratio. For example, in a limiting case, when BC/OA is close to zero, alpha_bc also approaches zero (there is no absorption by BC), and so δBrC approaches 1 (all the absorption is caused by BrC). Similarly, all the terms in Eq. 2 depend on the BC/OA ratio, albeit in a different manner (while the mass absorption efficiencies normally increase with the increase of BC/OA, the mass fraction of the organic fraction decreases with BC/OA). The dependencies of the mass absorption efficiencies on the BC/OA ratio are complex, and, to the best of my knowledge, cannot be described analytically in a general case (although some approximations are possible). Hence, I believe that Eqs. (1) and (2) are correct and meaningful.

5. I am also not sure that I properly understood the Referee's comments about the determination of AAEs and their uncertainties. We use only two wavelengths to determine each AAE instead of considering linear regressions in log-log space at multiple wavelengths (as suggested by the Referee) because it was essential for us not to lose the information about the wavelength dependence of AAE. As properly noticed by the Referee, our study is not the first one to follow this approach, for which I do not see a better alternative. Our ad hoc procedure for estimation of the uncertainty in AAEs is certainly not perfect but I could not find any other similar procedure in the literature.

6. As properly mentioned by the Referee, the particle components in our calculations were indeed distributed among 20 size sections spanning the particle shell diameters from 10 nm to 10 um following the unimodal size distribution. We disregarded the coarse mode (as the second distinctive mode of the size distribution) mainly because we could not provide

well-justifiable a priori constraints to its parameters and composition. However, as also noted in the manuscript, we do not expect that coarse particles can significantly affect our estimates. In this context, an extension of our particle size distribution until 10 um is just an arbitrary choice of no practical importance.

7. In our manuscript, we tried to distinguish between k (the imaginary part of the refractive index of any component), as in Table 1, and k_{OA} (the imaginary part of the refractive index of specifically organic aerosol). For brevity, the latter parameter is also termed absorptivity. In this definition (which is apparently not so common), we followed Saleh et al. (2014).

8. We assumed the power law for k_{OA} following multiple previous studies, some of which refer to Sun et al. (2007). In my opinion, this assumption makes sense because the power law for k_{OA} usually results in an observed power law for the BrC absorption coefficients.

9. As properly noticed by the Referee, Figure 2 is plotted using a set of synthetic (that is, ideal) data. As explained in Sect. 3.1, the analysis illustrated in Fig. 2 aimed to investigate the relationships between the "observable" optical properties of BB aerosol (such as AAE and SSA) and its"unobservable" characteristics (such as δBrC, k_{OA}, and the BC/OA ratio). Therefore, this analysis was by no means intended to examine the possibility of retrieving quantitatively accurate estimates of the parameters by taking into account the observation errors. Application of our algorithm to the "real-world" AERONET data and corresponding uncertainties are discussed in Sect. 4.3 of the manuscript.

10. As properly argued by the Referee, the BC/OA ratio and k_{OA} are two independent intrinsic properties of the aerosol matter. However, Saleh et al. (2014) found that these properties, when measured in multiple samples of carbonaceous aerosol, reveal a significant statistical relationship between each other. I presumed that taking into account our remark that "as argued by Saleh et al. (2014), a larger BC content is associated with a larger fraction of strongly absorbing organic compounds of extremely low volatility", our statement that "it could be expected that k_{OA} is an increasing function of the BC/OA ratio" is sufficiently clear. I am sorry if once again I did not get the proper meaning of the Referee comment.

References

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