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Comment on amt-2021-151

Anonymous Referee #1

Referee 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-RC2, 2021

Review of Inferring the absorption properties of organic aerosol in biomass burning plumes from remote optical observations
by
Igor B. Konovalov, Nikolai A. Golovushkin, Matthias Beekmann, Mikhail V. Panchenko, and Meinrat O. Andreae

This paper uses Bayesian statitistics to "re-model" the AERONET AAE(440-870), AAE(675-870), and SSA(440). That is, the authors use a new set of aerosol size and composition assumptions listed in Table 1 with Bayesian statistics to find the most probable combination of parameters (within the framework of Table 1) that produce the two AAEs and SSA(440). Then they analyze the resulting model and draw conclusions about the Organic Aerosol (OA) MAE and BC/OA ratios at two AERONET sites. This is a fundamentally unsound approach, in my opinion.

The problem is the authors don't acknowledge that AERONET AAEs and SSA are computed from a model of 100% internally mixed aerosols that have the same refractive index for all particle sizes. Thus, replacing the AERONET model with an aerosol system that is not also constrained by the size distribution and is not constrained by the same refractive indices as AERONET (at all particle sizes) will not necessarily produce a system of particles that reproduces the radiance field. The link to the radiance field is crucial, because the information in the radiance field is the information that drives the AERONET absorption retrievals.

To put this another way, it is very likely that AERONET has compensating errors. Thus, we can't pick and choose some AERONET elements (like AAE and SSA) and "re-write" other elements (like size distributions and the distribution of absorption wrt size) and expect to maintain a computational link to the actual measurements (i.e., a link to the radiances and exinctions). Hence, the authors need to demonstrate that their aerosol retrievals are still consistent with AERONET's radiation field in order for this to be a credible approach. That is, they should compute the radiances and extinctions from their Bayesian model and

compare their computed values to the radiance measurements.

MAJOR ISSUES:

The authors claim that "the relative contribution of BrC to the total absorption at 440 nm..." is described by Equation 1. However, Equation 1 is the ratio of mass absorption efficiencies (with units of m2/g):

dBrC = 1 - alpha_bc / alpha_tot

The mass absorption efficiencies are intrinsic parameters that do not depend upon mass; thus, dBrC is also intrinsic, and therefore unaffected by the BC/OA ratio. I don't see how this equation decribes the relative contribution of BrC to the absorption when I can vary BC/OA all that I want without affecting this equation. The authors need to explain this equation so that it makes physical sense to readers.

Likewise, Eq 2 does not make sense.

I am always suspicious of papers that use only two wavelengths to determine AAE (instead of a linear regressions in log-log space at multiple wavelengths), as small uncertainties in AAOD at either wavelength can produce significant changes in AAE. I realize that the authors claim to have derived a "robust" uncertainty estimate for AAE (Eq 10), but they do not explain how they arrive at Eq 10 very well.

Lines 154-158:
Authors state:
"The mass concentrations of the particle components were distributed among 20 size sections spanning the particle shell diameters from 10 nm to 10 um. The particle size distribution was assumed to be lognormal, unimodal, and representative of the accumulation mode. Taking into account that the contribution of coarse particles to the BB aerosol optical properties in the UV and visible wavelength ranges is likely small (Reid et al., 2005b), it was disregarded in our simulations."

This does not make sense... the authors distribute particles up to 10 um in diameter (which clearly includes the coarse mode), and then they disregard they disregard the coarse mode -- why do they include coarse mode sizes (> 1 um dia) in the first place, then?

Line 170:
It looks like the authors have redefined the imaginary refractive index as 'absorptivity' (k_oa). They cite Sun (2007) as the source of this term, but a search for 'absorptivity' in Sun comes up dry. The authors also use 'k' as the symbol for the imag refractive index (IRI) in Table 1, so it seems that they are using IRI and absorptivity interchangeably, which is incorrect.
Furthermore, the authors quote Sun (2007) as the basis for Equation 3, presumably referring to Sun's Eq 7. However, Eq 3 is a
powerlaw for IRI, whereas Sun's Eq 7 is a powerlaw for aerosol absorption (e.g., absorption coefficients or AAODs). Thus, Eq 3 is fundamentally different than the work that the authors cite.

Figure 2b:
Errorbars are needed on both x and y axes in Fig 2. Since the authors estimate the RMS of both AAEs as 0.12, it looks like the errorbars will touch the 1:1 line for most points; therefore, the spectral dependence of AAE for this synthetic data is within the expected noise range for real data. Thus, the two AAEs are essentially the same and one can not hope to obtain information from the curvature of AAE.

Line 550:
"it could be expected that k_OA is an increasing function of the BC/OA ratio..."
I thought that k_OA was the OA IRI? If so, it is an intrinsic property of OA -- Why should it depend upon the BC/OA ratio?

MINOR ISSUES:
Line 301:
I disagree with the authors assertions that SSA(440) = 0.92 is "highly reflective."

Line 304:
Authors state:
"Note that in the situations where the aerosol absorption is determined entirely by BC, AAE440/870 should be expected to be normally smaller than AAE675/870 according to W16."

W16 is another example of how 're-modeling' the AERONET assumptions can lead one to the wrong conclusion... When AAE < 1 in the AERONET database, it usually occurs when the coarse mode dominates the size distribution.

Figure 3:
Since fig 3 is synthetic data, the x-axis should be labeled as such (i.e., the x-axis is not observations, as stated.)

Lines 510-513:
Authors state: "Note also that the required AAE values can be derived not only from the AERONET remote measurements but also from satellite observations. In particular, multi-wavelength retrievals of AAOD are available from MISR observations (Junghenn Noyes et al., 2020)."
This statement is not consistent with Junghenn Noyes (2020), which states in the abstract that the *research algorithm* successully maps *qualitative changes*... I also did a search on AAOD and AAOT in Junghenn Noyes et al., 2020 and came up empty. Thus, I do not believe that the authors of the Junghenn Noyes article would agree with these claims about MISR AAOD.