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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Improved inversion of aerosol components in the atmospheric column from remote sensing data" *by* Ying Zhang et al.

Anonymous Referee #1

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This paper expands upon the authors' previous aerosol-components retrieval (Zhang et al., Atm Env, 2018) by including sodium chloride as a coarse aerosol component. The authors apply their results to about 16 SONET sites all across China. The grammar is clear and for the most part the paper is very well written. This is a good paper that is suitable for publication in ACP after some modifications.

The authors cite Zhang 2018 for their methodology, but I am not exactly sure of their approach. I gather that they use the Zhang 2018 approach to determine separate complex refractive indices (CRI) for the fine and coarse modes from the SONET data. Then for the coarse mode, they use RH to determine the equilibrium mixture ratio of NaCL with water, which has a certain real refractive index (RRI). Once the RRI for the water-NaCl mixture is known, they can iterate the dust mixing ratio until they minimize the χ^2

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of Eq 12. They are using a single “dust," though, so they can not vary the IRI independent of the RRI; thus, they have limited adjustability for the spectral dependence of the CRI. This is all very reasonable, but the use of a single “dust” will sometimes increase their residuals. That is ok, though, as residual values can be monitored and retrievals can be rejected on the basis of residual values when necessary. I am comfortable with their coarse mode methodology.

I am having difficulty understanding the fine mode retrieval methodology, though, which is my biggest reservation about this paper. The authors claim to separate water-soluble organic carbon (WSOC) from ammonium nitrate (AN), but it is not clear to me how this can be accomplished without a specific assumption for the hygroscopicity parameter (kappa) of WSOC. If this is what the authors are doing, they need to specifically state this and provide the reader with the value of kappa that they chose for WSOC (as well as the rationale for using a certain kappa, and some discussion of the repercussions of using the wrong kappa in their retrieval). The authors cite (Zhang 2018), but a brief overview of the Zhang approach for the fine mode in the methodology section would be helpful.

Major Issues

It is not clear to me how the “derived hygroscopic parameter kappa” is obtained (p2, line 59, and Table 2). I believe the authors are deriving the Table 2 values from Equation 4, but that requires the hygroscopicity parameters of the components (κ_i); the authors say that these values can be computed by the component hygroscopic growth factors (lines 144-145 and Eq 5). However, I don’t see how these component growth factors can be derived from their data, so I am assuming that they are obtaining these values from the literature. If this is the case, the authors should provide the reader with the GF_i or κ_i that they use in the retrieval. Otherwise, they should provide additional details about how they obtained the κ_i with the sunphotometer data.

Figure 3:

I was confused by the “non-hygroscopic components” that are a subset of the “water-soluble components” and are the entire basis of the “water-soluble organic matter (WSOM)” âĂŤ I am not a chemist, so I found it odd that water-soluble aerosols could be non-hygroscopic. It would be helpful to some readers (like myself) if the authors spent a sentence or two alerting the reader that water-soluble aerosols are sometimes non-hygroscopic. If they can explain the physics behind this phenomena, that would be even better.

Personally, I am skeptical about separating WSOM from AN using remote sensing techniques. From an optical standpoint, such a mixture would merely be a solution with an effective hygroscopicity parameter (kappa). Knowledge of RH and an assumed kappa allows one to derive the solute mixing ratio (and growth factor) via Eq 2, but I don’t see how one can separate the effects of multiple soluble components with the available remote sensing information (refractive index and RH) without additional assumptions (like the κ_i for each component).

Line 115, authors state:
“For fine mode, the water-insoluble and water-soluble components are identified using an empirical function (Zhang et al., 2018)"

How? The authors need to expand this a little. I checked the Zhang 2018, and I was not able to quickly determine how WI and WS components were separated. At a minimum, the authors should point to the specific section number in Zhang (2018), but it would be best to provide the readers with a brief recapitulation in order to best hold their interest.

Section 3.3:
The forward model is described well in Section 3.2, but the inversion section (3.3) is very light. For instance, the authors cover the relationship of the real refractive index to molar refractivity in Sect 3.2, but none of that shows up in Section 3.3. Presumably the authors are using RH to partition between the soluble components and water and also to assign a RRI to the host solution prior to the minimization procedure described in

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section 3.2 (which requires refractive indices of both the host solution and the insoluble inclusions). None of that is stated here, though, so as a reader I am not sure if I have this correct.

Table 3:
Why is the RRI of NaCl and the coarse water so uncertain? I thought we had some good measurements of these species. Even if we didn't, how do we get 900

Minor Issues

Page 1, line 28, authors state:
"Optical remote sensing techniques do not provide sufficient information for a detailed analysis of chemical composition and therefore refrain to the retrieval of components describing specific properties"

My interpretation of this sentence is that we can not retrieve aerosol composition from remote sensing techniques, but I am sure that is not the authors intent (otherwise, we don't need to read the paper). Consider rephrasing.

Page 2, line 37:
Schuster (2009) is entitled "Remote Sensing of Aerosol Water Uptake," and does not directly address dust.

Page 3, line 82:
Should also reference Dubovik, O., and M. King (2000), A flexible inversion algorithm for retrieval of aerosol optical properties from sun and sky radiance measurements, J. Geophys. Res., 105(D16), 20,673–20,696.

Page 3, lines 87-88.
I don't understand the meaning of these lines:
"Using these data, PVSD and CRI sub-modal parameters of atmospheric aerosols are obtained using the modal decomposition method proposed by

Zhang et al. (2017). Using the sub-modal characteristics data set thus obtained, an aerosol sub-modal model was established for China by Li et al. (2019), but the submodal aerosol components have not been given."

So a sub-model model was established but not given?

Page 6, line 150:
Please replace “refractive index” with “real refractive index.” Page 6, line 156:
Please tell the reader that “n" is the real refractive index.

Equation 10:
Equation 4 uses the symbol epsilon as the component dry volume fraction, whereas here it is the permittivity. Need to change the symbol used for dry volume fraction in Eq 4 and everywhere.

Equation 12:
The numerator should be squared. Otherwise, large negative differences will produce the “best” χ^2.

Table S1:
What is the basis for the numbers in Table S1? That is, which climatology are you using to define WS, BB, and DU?

Page 9, lines 238-241:
Does it make sense to quantitatively discuss BC in the context of Fig 5? BC barely exists in that figure. I recommend adding a table or an additional figure for BC.

Figure 6:
How is the color scale in Fig 6 normalized (range is 0 to 20)? Also, it is odd that some of the “estimated" values are so far off when you are using CRI as a constraint. The χ^2 must be very high in these cases. It would make sense to have a residual requirement (i.e., $\chi^2 <$ some threshold) and to throw out high values of χ^2. This should also improve your statistics (slope, intercept, bias, etc.).

Lines 248-249: Authors state
"As shown in table 3, although the TRE of BC is the lowest, it also causes the largest k_f and $k_{f,440}$ errors."

TRE is total relative error, so how can TRE cause k_f and $k_{f,440}$ errors? Shouldn't cause and effect be the other way around (i.e., k errors cause TRE errors).

Lines 252-253, Authors state:
"This indicates that most inversion results have good optical closure, and the aerosol components retrieved by the remote sensing method used in this study should be reasonable."

This line refers to Fig 6, which is a plot of how well the component-averaged imaginary index compares to the imaginary refractive index that is used as input. Thus, a good comparison just means that you usually have good residuals (i.e., low χ^2). Fig 6 does not assure reasonableness of all components in the retrieval, though, as it only shows the imaginary RI, and most of the components of this retrieval are not sensitive to IRI. The only thing that we can claim via Fig 6 is that the retrieval might be getting BC correct. Additionally, we can't use Fig 6 to argue that the BC mass or volume fractions are correct, as these are sensitive BC refractive index. However, you can use Fig 6 to argue that you are getting the BC AAOD correct; this is because you are using IRI as a constraint, and the IRI that you retrieve will always be the same (as long as BC has a spectrally flat IRI and your other absorbers do not).

Line 275:
I believe that the word "autumn" should be replaced with "spring."

Table 3:
Total relative error is defined with 7 parameters. Presumably this is n_f, k_f, $k_{f,440}$, n_c, k_c, $k_{c,440}$, and RH. Are the (n_f, k_f) averaged from the 675, 870, and 1020 nm wavelengths, then? I don't recall seeing this explicitly stated.

Figure 2:
Caption should describe the timeframe of the boxplots.

Figure 4:
Do the pie charts correspond to both the fine and coarse modes? If so, why isn't there any AWc or SC in the WS and BB pie charts? If not, why does dust dominate over WIOM for those species?

Figure 7:
Throughout the text, authors use SC for sodium chloride. Here, they do not show SC but show SS (sea salt?).

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