Anonymous Referee #1

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 of Eq 12. They are using a single "dust," though, so they cannot 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

1. 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 GFi or κ_i that they use in the retrieval. Otherwise, they should provide additional details about how they obtained the κ_i with the sun photometer data.

Response: Thanks for the reviewer's comment. The values of κ_i are obtained from the literature and listed in table 2. For clarity, we have revised the manuscript as follows:

Line 156-157: "…where κ_i is the hygroscopic parameter of the *i*th component obtained from the literature (table 2), and $f_{dry,i}$ is the dry component volume fraction defined as … "

2. 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)" â˘A˘T 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 _i for each component).

Response: For the non-hygroscopicity of organic components, like vinegar and alcohol, they are water-soluble but more volatile than hygroscopic. In aerosols, the dicarboxylic acids are dominant in the WSOM component, and oxalic acid accounts for more than 50% in dicarboxylic acids, followed by succinic, malonic, maleic, adipic and phthalic acids (Chebbi & Carlier, 1996). Under ambient relative humidity (RH) of 10 - 90%, oxalic acid hardly deliquesce and exhibit low hygroscopicity, and others also have low hygroscopicity except malonic acid as shown in Figure R1 (Ma et al., 2013; Drozd et al., 2014; Jing et al., 2016). Considering the abundance of oxalic acid (more than 50% in dicarboxylic acids) and other low hygroscopic components in the atmospheric aerosols (Sullivan et al., 2009; Fu et al., 2013), WSOM is treated as a non-hygroscopic component in aerosol particles. We explained this point in the manuscript as follows:

Line 124-128: "It should be noted that the water-soluble property of aerosol components is not equivalent to hygroscopicity. Dicarboxylic acids represented by oxalic acid are dominant in the WSOM component but their hygroscopicity is extremely low (Ma et al., 2013; Drozd et al., 2014; Jing et al., 2016). Also other organic compounds in aerosols are less hygroscopic as shown in Zhang et al. (2018) (their figure 1). Hence, the OM components (WSOM and WIOM) are treated as non-hygroscopic components."

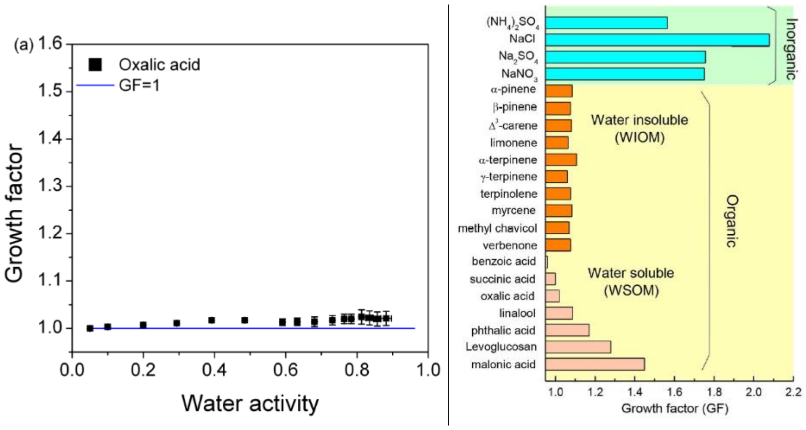

Figure R1. (a) Hygroscopic growth factors of oxalic acid from figure 2 of Jing et al. (2016); (b) Different geometric hygroscopic growth factor between inorganic and organic aerosols with RH around 85% of relative humidity from figure 1 of Zhang et al. (2018).

For the separation of the WSOM and AN components, the refractive index (RI) from remote sensing is used indirectly. From Eq. 2-5, we can get the relationship among the volume fraction of the components in the host. Because the sum of the volume fractions of these components can be equal to 1 in the host, the RI of the host can be calculated using Eq. 6-9. And then we can calculate the RI of the entire particle using the effective medium approximation (Eq.

10-12). Adjusting this fraction and comparing with the RI from the SONET observations, the volume fraction for all components can be obtained. The description and flowchart of the inversion procedure are added in section 3.4.

Ma Q., He H., Liu C. (2013). Hygroscopic properties of oxalic acid and atmospherically relevant oxalates, Atmospheric Environment, 69, 281-288.

Drozd G., Woo J., Hakkinen S.A.K., Nenes A., and McNeill V.F. (2014). Inorganic salts interact with oxalic acid in submicron particles to form material with low hygroscopicity and volatility, Atmos. Chem. Phys., 14, 5205–5215.

Chebbi A. & Carlier P. (1996). Carboxylic acids the troposphere, occurrences, sources, and sinks: a review, Atmospheric Environment, 30(24), 4233-4249.

Jing B., Tong S., Liu Q., Li K., Wang W., Zhang Y. and Ge M. (2016). Hygroscopic behavior of multicomponent organic aerosols and their internal mixtures with ammonium sulfate, Atmos. Chem. Phys., 16, 4101–4118.

Fu, P., Kawamura, K., Usukura, K., and Miura, K.: Dicarboxylic acids, ketocarboxylic acids and glyoxal in the marine aerosols collected during a round-the-world cruise, Mar. Chem., 148, 22–32, 2013.

Sullivan, R. C., Moore, M. J. K., Petters, M. D., Kreidenweis, S. M., Roberts, G. C., and Prather, K. A.: Timescale for hygroscopic conversion of calcite mineral particles through heterogeneous reaction with nitric acid, Phys. Chem. Chem. Phys., 11, 7826–7837, 2009.

3. 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.

Response: Thanks for the reviewer’s comment. We added an explanation of the empirical function in the manuscript and add details to the supplementary as follows:

Line 115-118: “For the fine mode fraction, the water-insoluble and water-soluble components are identified using an empirical function (see section 2.2.2 in Zhang et al., 2018), which describes the ratio of the water-soluble to the water-insoluble volume fractions determined by RH, together with the parameterization of aerosol soluble volume fractions by Kandler and Schutz (2007).”

4. 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 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.

Response: Thanks for the reviewer’s comment. We adjusted the structure of the methods section and added a description of the inversion procedure in both manuscript and supplementary as follows:

Line 179-216:

"3.3 Effective medium approximation

To determine the complex refractive index of a particle, i.e. including both the multi-component liquid system and water-insoluble matter, the complex refractive index (m = n - ik) at wavelength λ is expressed in terms of the permittivity, ε (λ):

$$m(\lambda) = \sqrt{\frac{|\varepsilon(\lambda)| + Re\big(\varepsilon(\lambda)\big)}{2}} + i\sqrt{\frac{|\varepsilon(\lambda)| - Re\big(\varepsilon(\lambda)\big)}{2}} \tag{10}$$

The permittivity of the multi-component liquid system can then be calculated using equations (8) - (10). Considering the water-insoluble matter in a particle as inclusion and the water-soluble matter as the environment, the permittivity of the entire aerosol particle can be obtained by the Maxwell Garnett effective medium approximation (Schuster et al., 2005).

$$\varepsilon_{eff}(\lambda) = \varepsilon_e + 3\varepsilon_e \left[\frac{\sum_j \frac{\varepsilon_j(\lambda) - \varepsilon_e(\lambda)}{\varepsilon_j(\lambda) + 2\varepsilon_e(\lambda)} f_j}{1 - \sum_j \frac{\varepsilon_j(\lambda) - \varepsilon_e(\lambda)}{\varepsilon_j(\lambda) + 2\varepsilon_e(\lambda)} f_j} \right] \tag{11}$$

where, j is the number of water insoluble components and. $\varepsilon_j(\lambda)$ and $\varepsilon_e(\lambda)$ are the permittivities of the inclusion and its environment. The complex refractive index of the entire aerosol is estimated by aerosol component fraction using equation (10).

3.4 Inversion procedure

The flow chart for the inversion of the aerosol components is shown in figure 4. In the fine mode, the ratio of WS and WI matter is estimated using RH as described in section 2.2.2 in Zhang et al. (2018). The initial value of the host refractive index and the extreme value for the BC component are set by the calculation modules of the complex refractive index in the multicomponent liquid system (see section 3.2) and the effective medium approximation (see section 3.3), respectively. In the loop to determine the BC component, two constraints are applied to separate BC from other components. The WSOM/WIOM ratio constraint was developed by Zhang et al. (2018) based on considerations published in the literature (Chalbot et al., 2016; Bougiatioti et al., 2013; Wozniak et al., 2013; Mayol-Bracero et al., 2002; Krivácsy et al., 2001; Zappoli et al., 1999):

$$\begin{cases} f_{WSOM} \cong \alpha f_{WIOM} \\ \alpha = \dfrac{\beta \rho_{WSOM}^{-1}}{1 - \beta \rho_{WSOM}^{-1}} \end{cases} \qquad \beta \in [44\%, 77\%\,] \tag{12}$$

For more detail, see section 2.3.1 in Zhang et al. (2018). The volume normalization of the aerosol components in both the fine and coarse modes is used to constrain the volume fraction of the aerosol components to a reasonable range (similar as section 2.3.2 in Zhang et al., 2018)

$$\begin{cases} f_{fine} + f_{coarse} = 1.0 \\ f_{fine} = f_{BC} + f_{AN} + f_{WSOM} + f_{WIOM} + f_{AW_f} \\ f_{coarse} = f_{DU} + f_{SC} + f_{AW_c} \end{cases} \tag{13}$$

Then the inner loop of WSOM computes the CRIs of the fine mode at different BCs, and output the aerosol components of minimum χ^2. The inversion procedure for the coarse mode is simpler than that for the fine mode. There is only a loop for DU and the complex refractive index of the host can be directly calculated by equations (2) - (8) with only input of RH. The function Chi-squared (χ^2) as an iterative kernel function is expressed in the sum of the differences between the complex refractive index estimated from the forward model (m) and the retrievals (m_{rtrl}), at multiple wavelengths:

$$\chi^2 = \sum_{\lambda} \frac{\left(m_{rtrl}(\lambda) - m(\lambda)\right)^2}{m_{rtrl}(\lambda)}$$

λ=440, 675, 870 and 1020 nm (14)

The retrieval is completed when the value of χ^2 reaches a minimum. The volume fractions of the aerosol components can be obtained by solving the above equations (10-12). The aerosol mass concentration in the atmospheric column is calculated using the volume and effective density of the aerosol components."

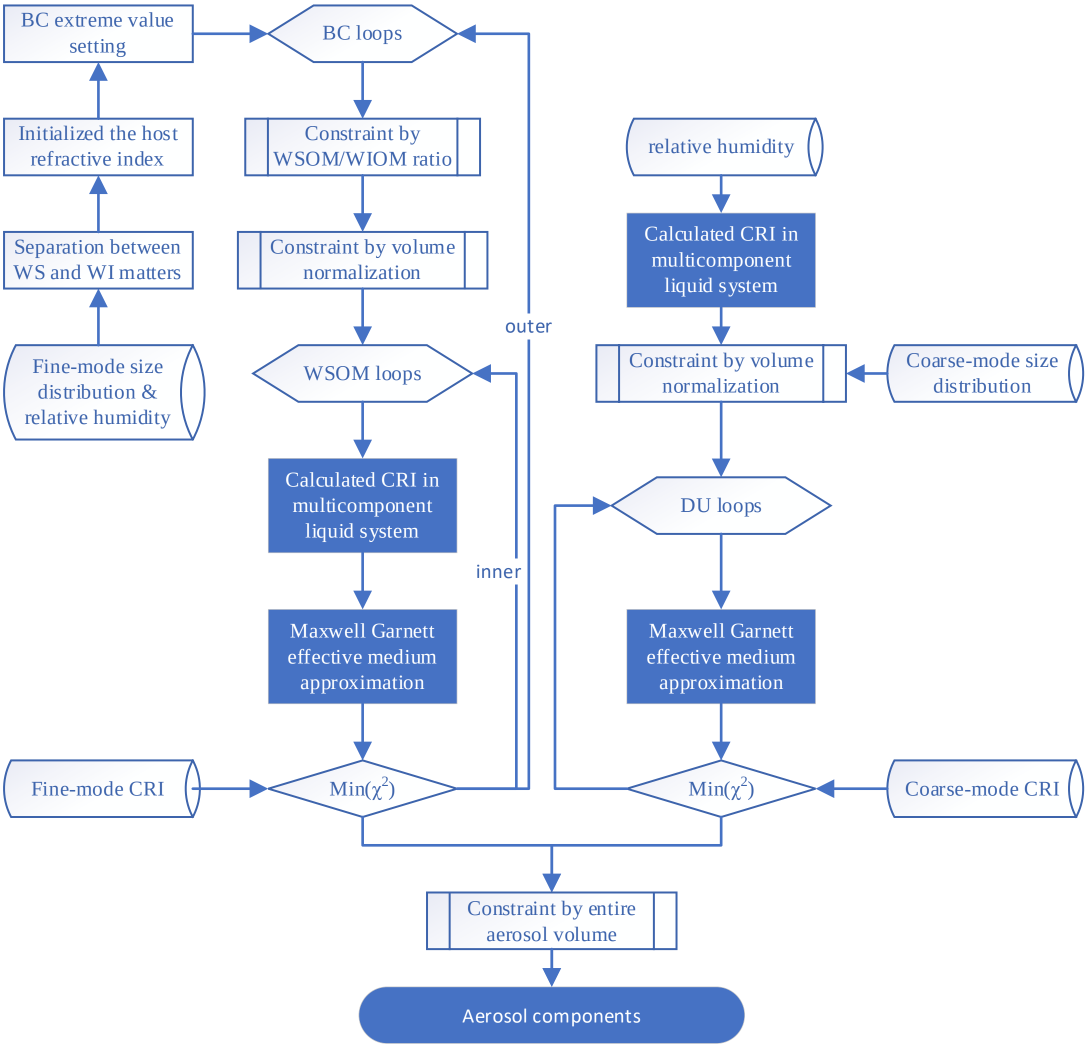

Figure 4. Flowchart of the aerosol component classification inversion algorithm.

5. 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

Response: Thanks for pointing this out. This large error (more than 900%) occurs only when the input error is large. For separation of high scattering components (NaCl and coarse water), the relative humidity (RH) and real part of CRI in coarse mode (n_c) are important dependent parameters and weakly correlated with other parameters. A large error for RH (10%) is applied to estimate the scattering components, which can be reduced because the actual observation error is usually lower than 5%. For the error of n_c summarized from Zhang et al. (2017) listed in table S2, it is large for the WS typical model, which can also affect the scattering component separation. However, if the accuracy of RH is improved, the errors of NaCl and coarse water can be effectively suppressed. Meanwhile, it should be noted that this estimation of uncertainty is for a single measurement. One of important advantages of a remote sensing approach is to perform multiple measurements quickly in a short time period. Thus, the average uncertainty of the aerosol components can be effectively reduced taking account independent errors in each observation. We illustrate this point in the manuscript:

Line 270-275: "Fortunately, the RH observed by ground-based stations is accurate, with an error which is usually less than about 5% (WMO, 2008), which can significantly reduce the uncertainty in the retrieved aerosol scattering components. It should be noted that the uncertainties in table 3 are for single measurements. One important advantage of remote sensing is that multiple measurements can be made during a short period of time. Thus, the average uncertainty of the aerosol components can be effectively reduced by taking into account independent errors in each observation. In addition, the accuracy of the retrieved n_c needs to be improved in order to deal with the aerosol component inversion."

Minor Issues

1. 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.

Response: Thanks for reviewer's comment. This sentence was deleted.

2. Page 2, line 37:

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

Response: Thanks for reviewer's comment. We revised this sentence:

Line 36-41: "In a follow-up study, Schuster et al. (2009) applied a similar procedure to determine the aerosol water fraction by fitting the real part of the refractive index of an internal mixture of water, soluble and insoluble species to observations by minimizing the cost function at all four wavelengths together. In this work the ratio of the dry volume fraction of insoluble to that of soluble aerosols was constrained by using a climatological value and the real refractive index which also prescribes the aerosol hygroscopicity. This constraint also provides a maximum insoluble fraction and the fraction of dust aerosol."

3. 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.

Response: We add this reference in manuscript.

Line 85-86: "Based on the inversion algorithm of Dubovik and King (2000) and Dubovik et al. (2000), the 440, 675, 870 and 1020 nm wavebands are used to … "

4. 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?

Response: We revised this sentence:

Line 91-93: "Using these fine and coarse mode characteristics of the CRI, micro-physical properties of aerosols in each mode were analyzed (Li et al., 2019), but the aerosol chemical components were not determined."

5. 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.

Response: We revised as follows:

Line 162-164: "Firstly, the molar refractivity (A_e) at wavelength λ can be calculated from the real part of the complex refractive index (n_i) and the volume fraction of the individual components"

6. 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.

Response: We change the symbol used for dry volume fraction to $f_{dry,i}$ in Eq 4 and 5. The corresponding descriptions have also been modified.

Line 153-160: "In the multicomponent liquid system, the hygroscopicity parameter κ is given by the simple mixing rule

$$\kappa = \sum_i f_{dry,i}\kappa_i \tag{4}$$

where κ_i is the hygroscopicity parameter of the *i*th component obtained from the literature (table 2), and $f_{dry,\,i}$ is the dry component volume fraction defined as

$$f_{dry,i} = \frac{V_i}{V_s}$$

"

7. Equation 12:

The numerator should be squared. Otherwise, large negative differences will produce the "best" χ^2.

Response: Thanks for pointing this out. We missed the square when wrote the formula, but we used the formula where the numerator is the square.

8. 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?

Response: Considering that the algorithm results from Dubovik et al. (2000) are taken as input in this study, the aerosol model is consistent with Dubovik's. In table S1, the parameters of size distribution are as same as those in table 2 of Dubovik et al. (2000), and the complex refractive index in the fine and coarse modes are further calculated using Dubovik's aerosol model from Zhang et al. (2017). We give more detail on this point in the supplementary.

9. 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.

Response: Thanks for reviewer's comment. All component fractions are listed in the table S3 and the BC fraction is added in figure 7 (Figure 5 from the previous manuscript) as follows:

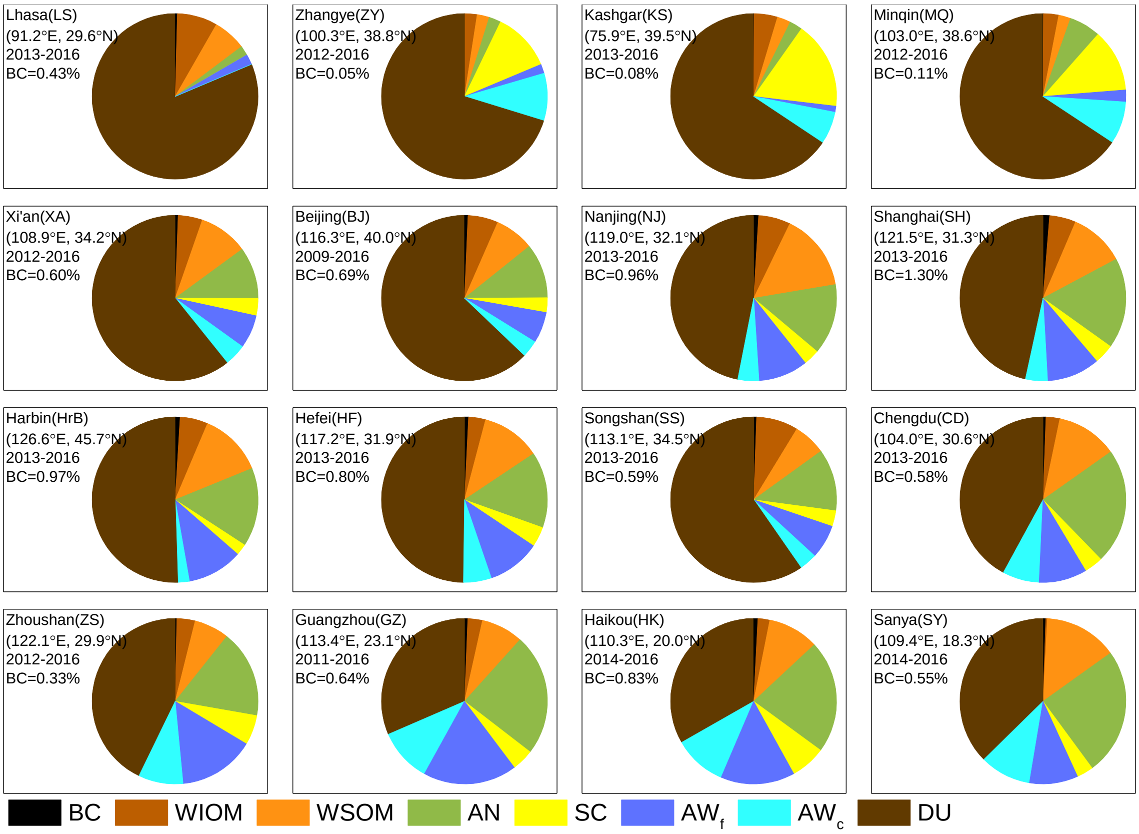

Figure 7. The averaged mass fraction of aerosol components at SONET sites. The site name, location, observation period and BC fraction are marked in each subgraph. The mass fractions of other components are listed in table S3..

10. 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., χ^2 < some threshold) and to throw out high values of χ^2. This should also improve your statistics (slope, intercept, bias, etc.).

Response: Thanks very much for your comments. We set a threshold and filter the results and revised as follows:

Line 303-321: “

The closure of the CRI between instantaneous optical-physical inversion and chemical estimation is examined by the data pair frequency. Figure 8 shows scatter density plots of the chemically estimated and sunphotometer-retrieved imaginary parts of the fine mode at 675 nm (k_f) and 440 nm ($k_{f,440}$) and the real parts of fine mode at 440 nm(n_f). The points are colored by the number of data pairs (Retrieved, Estimated), which are sorted according to ordered pairs in 0.0005 intervals for the imaginary parts of CRI and 0.001 intervals for the real parts. The data pairs of k_f are closely concentrated around the 1:1 line, although with slight underestimation with 94.3% of the estimated values lower than the retrieved values; only 5.3% of the data pairs have a relatively large absolute error (AE > 0.01). The mean bias is not large (-0.003), and the mean absolute value is equal to the mean absolute error (MAE = 0.003). There are two reasons for this slight underestimation in chemical estimation. On the one hand, the imaginary part of the refractive index of BC is much larger than for the other components due to its strong absorption. Thus, the inversion of the BC concentration is very sensitive to the estimation of the refractive index. As shown in table 3, although the TRE of BC is the lowest, the errors caused by k_f and $k_{f,440}$ are larger than for any other component. On the other hand, k_f is not only affected by BC in the inversion process, but also affected by organic components (WSOM & WIOM) with spectral absorption characteristics. Therefore, in most cases, k_f is underestimated in chemical estimation and $k_{f,440}$ is overestimated (Bias = 0.007). The mean relative error (RE) is 27.1%, and 62.8% of the data points are below the average relative error line. This indicates that most inversion results have good optical closure. For the closure of the real part of the fine mode, the data pairs of n_f are also concentrated around the identity line, although 76.5% of the n_f is above the identity line. Underestimation occurs mainly when n_f is larger than 1.56, because the only component with the real part of the CRI larger than 1.56 is BC, but its concentration is mainly determined by the imaginary part. The bias of the estimated n_f (Bias = 0.009) is larger than that of k_f due to the fact that the value and the range of n_f are larger than that of k_f. "

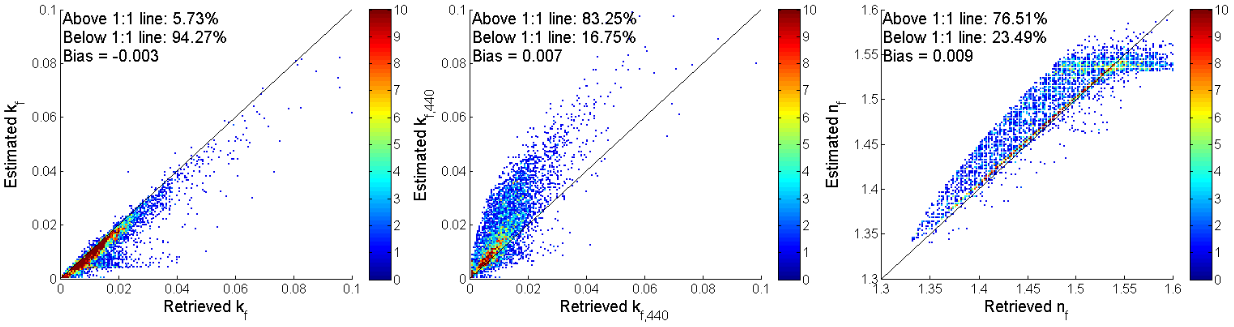

Figure 8. Data pair frequency of instantaneous imaginary parts of the complex refractive index at 675 nm (k_f), 440 nm ($k_{f,440}$), and real part at 440 nm (n_f) which are sorted according to ordered pairs (Retrieved, Estimated) in 0.0005 and 0.001 intervals for imaginary and real parts, respectively. "Retrieved" represents sub-component of CRI from the optical-physical retrievals, and "Estimated" is estimated by retrieved chemical components. The color represents the number of cases (color bar), and the solid black line shows the 1:1 line.

11. Lines 248-249: Authors state

"As shown in table 3, although the TRE of BC is the lowest, it also causes the largest kf **and** kf;440 **errors."**

TRE is total relative error, so how can TRE cause kf and kf;440 errors? Shouldn't cause and effect be the other way around (i.e., k errors cause TRE errors).

Response: We revised this sentence as follows:

Line 312-313: "As shown in table 3, although the TRE of BC is the lowest, the errors caused by k_f and $k_{f,440}$ are larger than for any other component."

12. 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).

Response: Thanks for the reviewer's detailed explanation. We revised this sentence as follows:

Line 317: "This indicates that most inversion results have good optical closure."

13. Line 275:

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

Response: Thanks for pointing out this mistake. We revised as follows:

Line 342-343: "Low concentrations of BC in the other seasons are mainly due to the influence of frequent dust events in the spring and high aerosol hygroscopic growth in the summer."

14. Table 3:

Total relative error is defined with 7 parameters. Presumably this is nf, kf , kf;440, nc, kc, kc;440, and RH. Are the (nf ; kf) averaged from the 675, 870, and 1020 nm wavelengths, then? I don't recall seeing this explicitly stated.

Response: Our method of separating fine and coarse modal CRI parameters (Zhang et al, 2017) can only separate 6 of these parameters (nf, kf , kf;440, nc, kc, kc;440). Thus we assume that the imaginary parts of CRI at other wavebands except 440nm are invariable. We add a statement for CRI sub-modal parameters as follows:

Lien 90-91: "The real parts of the CRI of the fine and coarse modes (n_f and n_c, respectively) are spectrally independent, while the imaginary parts have spectral variation at 440 nm, so they are written as ($k_{f,440}$, k_f) & ($k_{c,440}$, k_c)."

15. Figure 2:

Caption should describe the timeframe of the boxplots.

Response: we revised the caption of figure 2 as follows:

"**Figure 2**. Boxplots of the relative humidities observed near each of the SONET sites. The observation periods for each site are shown in table 1. The line and the diamond represent the median and mean values, respectively, and the box shows the standard deviation (1 σ)."

16. 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?

Response: Yes, this figure shows both the fine and coarse modes. Few studies have given the fraction of water content in coarse mode which SC is closely related to, so we set it very low in the typical model.

17. Figure 7:

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

Response: Thanks for pointing this out. We revised the figure.