Anonymous Referee #3

Article is well in the scope of the ACP journal and discusses a method for retrieval of aerosol chemical composition from remote sensing. Aerosol chemical composition is in a grand demand by a scientific society providing data to evaluate global atmospheric modelling and climatic models. Presented method introduced some of the novelties and meanwhile is based on a well-established and proven techniques. Paper is well referenced at least to my knowledge, although I've found some inaccuracies (see in notes below).

At the same time, I have a feeling that paper doesn't reflect fully the potential of the method and the complexity of the work performed by authors, and definitely doesn't provide enough explication to reproduce the described method. Article is very brief and dry, to the extent it sometimes hard to read and what is more important to understand concepts of the study preformed. Also, I've found out quite a number (for a short paper of 10 pages) of typos, please, consider general grammar revision, some of them I've listed below.

I would recommend this article for publication after major revision, which in my opinion should increase the impact and significance of the presented results.

Below I've highlighted some points where there is a room for improvement.

Major comments:

Formula 12: Possible typo, square is missing in numerator. Either it is missing either it is no chi-squared function. Please, clarify. If no square formula was actually used for the study, I'm not sure the method is legit as opposite sign errors in refractive indices at different wavelengths can compensate each other. Minimisation procedure of chi2 function is not described at all, at least mention how it is done. What is "iterative kernel function", Is it LUT or a version of Newtonian method? is it the same that was used in previous studies? Please clarify or refer.

Whole section 3 is rather confusing, despite of illustrations and formulas it doesn't give a clear understanding of a methodology, just some of its pieces. Also, if this method is an improvement of an existing one, it should be clearly referred and changes introduced highlighted. Please, consider re-formulating this part to make it clearer. Figure 3 and Aerosol classification in section 3.1, it is hard to understand at which part of the retrieval this classification is used. And what parameters it used to classify aerosol in groups? Is it part of forward modelling? Does it use PVD provided by SONET, other part of paper claims only refractive index is used … And section 3.3 refers to refractive index of fine and coarse mode …

Also "combined" aerosol types such as BC, OM, IS and AW are used only in figure 3 and not referenced anywhere else in the paper. Why then there are presented?

Personally, I would prefer here a flowchart of the retrieval algorithm in general, i.e. where it would be easy to understand what parameters enter it, how refractive index is modelled and fitted and what parameters are coming out.

Response: Thank you very much for pointing this out.

(1) Formula 12 is indeed a typo. We have revised it in the manuscript as follows:

Line 211:"

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

λ=440, 675, 870 and 1020 nm (12)"

(2) In order to present our algorithm more clearly, we have added the flowchart in Figure 4 and corresponding descriptions (section 3.4).

Line 192-216:

"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."

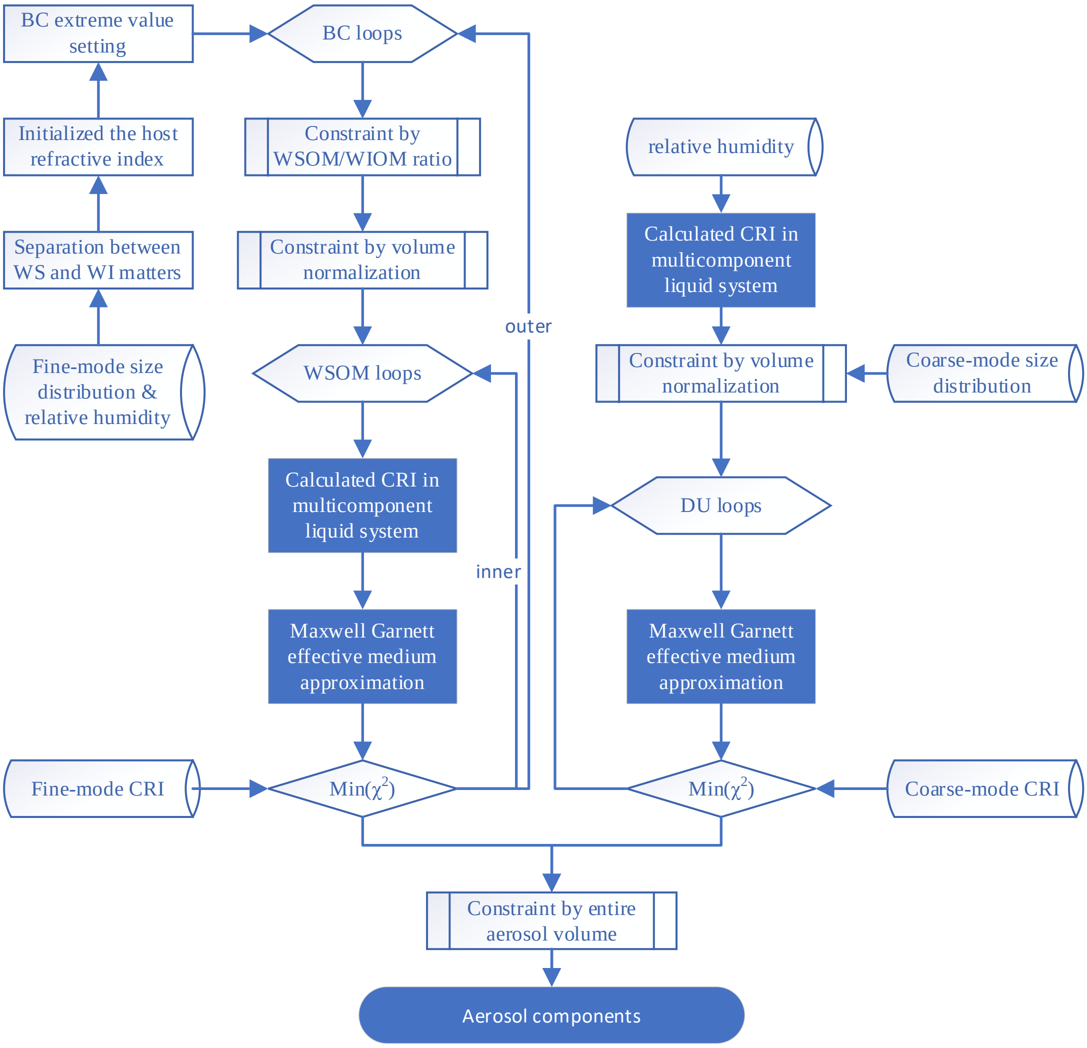

Figure 4. Flowchart of the aerosol component classification inversion algorithm .

(3) The "combined" aerosol types are used in table 3, and subsequent analysis (section 4) was associated with this combined component.

Error analysis: I would like to be convinced that method works, by showing a retrieval without any noise added, proving that it retrieves the exact pre-defined composition, i.e. calculating CRI using the forward model and retrieving it back again. Errors are analysed only from the point of view of data uncertainties, although some of them can emerge from the retrieval itself (i.e. inaccuracy of the forward model) and an obvious fitting bias in figure 6 can be an indication of that.

Response: Thank you for reviewer's comments. A set of retrievals without noise is added in the section 3.5 as follows: Line 226-240: "The uncertainty in the retrieval results was evaluated using synthetic data, both without and with input errors added. For the first case (without input errors), a set of complex refractive indices has been obtained by calculating a set of volume fractions of the aerosol components using the forward chemical model, which was used as input for the retrieval of the aerosol components without any noisy added. For the aerosol components, the volume fraction of BC was constrained between 0.0 to 3.0% with an interval of 0.5%, and corresponding dynamic ranges for the other components with intervals of 10%, in three ambient relative humidity conditions (40%, 60% and 80%). Figure 5 shows the comparison of the aerosol component volume fractions from forward modeling used as input, and their retrieved values. The volume fractions of the retrieved aerosol components are in good agreement with the input values. For the fine mode fraction, most data pairs are located close around the 1:1 line, with the mean absolute error (MAE) of the aerosol component volume fractions of 3.0%. In five samples the difference in the AW_f is more than 20.0%, though the overall MAE for AW_f is only 5.5%. In these five samples, the BC component is low and organic matter contributes substantially to the aerosol light absorption, resulting in underestimation of the AW_f volume fraction at high RH and overestimation for moderate RH. WSOM is overall slightly overestimated and AN is underestimated by only a few percent. The correlation between the input and retrieved aerosol volume fractions in the coarse mode is even better than that in fine mode. The regression coefficient for all samples is 0.99, and the MAE is only 2.0%. These results show the very small uncertainty in the retrieved aerosol component volume fractions."

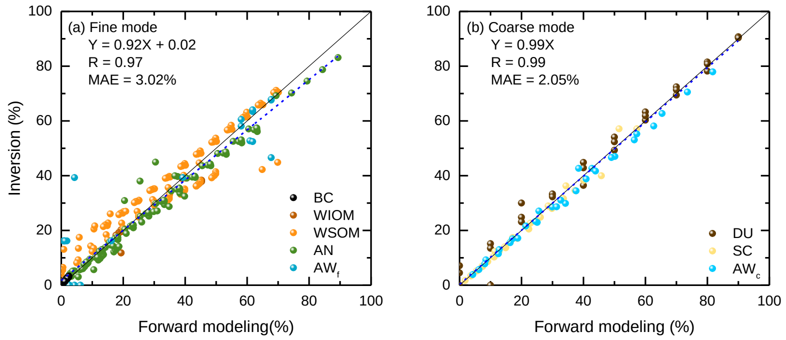

Figure 5. Scatter plots of volume fractions of aerosol components in the fine (left) and coarse (right) modes retrieved using the algorithm described in Chapter 3, versus those used as input calculated with the forward model. The solid line is the 1:1 line, and the dash line is the fitting line.

Data analysis: Again, very dry, some additional analysis (for e.g. splitting sites into several groups and analysing the seasonal averages for the groups) would be appreciated. For instance, there was a statement that higher BC in winter is because of heating in north region, splitting sites into heated/not heated regions and analysing seasonal trend would make this statement much more trustworthy and results obtained more significant. Also, a comparison to a previous method could be presented, if, of course, such comparison could be done. For e.g. comparison with OM from Zhang 2018 could be performed to illustrate improvements (if any), or discuss the similarities/differences observed.

Response: Thank you for the comment. We add the analysis about the seasonal variation of main aerosol compositions in fine mode between north and south China as follows:

Line 346-359: " The seasonal variation of the main aerosol components in the fine mode is discussed on a regional basis (Figure 10). BC concentrations in typical northern regions are higher than in southern regions, because of emissions due to winter heating only in the north. Other BC sources are vehicle emissions and biomass burning. Adverse meteorological conditions in winter result in the accumulation of BC in the atmosphere resulting in high BC values in both the north and the south. The highest BC mass concentrations in the northern region in the winter is 4.3 mg m^{-2}. OM is one of the dominant components in the fine mode, with sources similar to those of BC. The impact of biomass burning in the winter and spring over south China (Chen et al., 2017) is significant, leading to OM concentrations of more than 50.0 mg m^{-2}. In the northern region, much biomass burning occurs in the autumn (Wang et al., 2020). With the influence of heating, the OM level in the north can reach up to 80.1 mg m^{-2}. Therefore, the OM mass concentration in the northern region is only low in the summer (50.8 mg m^{-2}). AN is usually formed by secondary reactions of gaseous precursors in complex air pollution areas. In both the northern and the southern region, AN mass concentration is larger in the summer than in other seasons, and the seasonal variation in the southern region is significantly smaller than that in the north. The mean AN mass concentration in the southern region is 8.7 mg m^{-2} higher than that in the northern region. This suggests that more AN is produced by secondary reactions in the humid climate in the south than in the northern region."

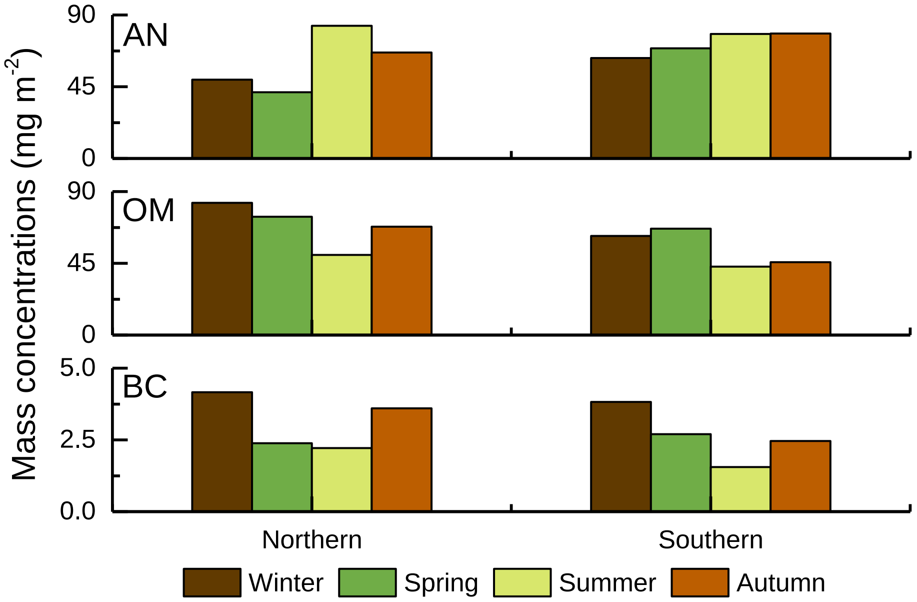

Figure 10. Comparison of aerosol component mass concentrations in northern (Xi'an, Beijing, Harbin, Hefei and Songshan) and southern China (Nanjing, Shanghai, Zhoushan, Guangzhou, Haikou and Sanya).

Compared with Zhang et al. (2018), the main advantage of the new algorithm is the increased flexibility in selecting water-soluble components. The new algorithm gives a reasonable scheme of complex refractive index (CRI) estimation, considering the mixing of multiphase solution. We added an explanation in the revised manuscript as follows:

Line 216-224: "The retrieval algorithm described here is an improvement over that described in Zhang et al. (2018). In that algorithm, the WSOM component was added to the host, but it could only be considered as a non-hygroscopic component. The proportion of solute and solution in the host mixture at different relative humidities should be measured in the laboratory, which limits the choice of aerosol components in the inversion process. Also, the real part of the CRI of the host was calculated by volume averaging, which can introduce a small error. The improved algorithm described here is more suitable for the calculation of the properties of a mixture of multiple water-soluble components as long as the hygroscopic parameter is known, which is not only convenient to measure but also independent of particle size. The hygroscopic parameter of WSOM can be varied according to the choice of mixing components instead of changing the algorithm itself. Similarly, some other water-soluble components (e.g. sulfate) can be introduced into the inversion algorithm without laboratory measurements."

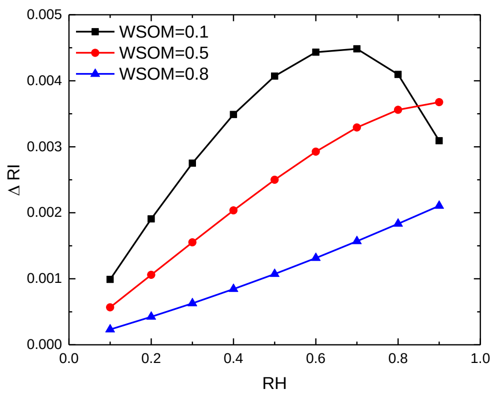

Figure R1. The differences of real part of CRI with relative humidity between the algorithm of Zhang et al (2018) and this study when the WSOM volume fraction is 10%, 50% and 80%.

There is a misfit error available as a result of the retrieval, why not use it to clean up the data a bit? This will provide more trustworthy results. Besides, as I understood estimations of the SONET retrieval errors for complex refractive index are available, why not get rid of the results whose fits are below the error bars? Authors themselves claim that ~40% of the retrieval are above the average error, i.e. these results have questionable quality and could significantly influence the statistics provided.

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. "

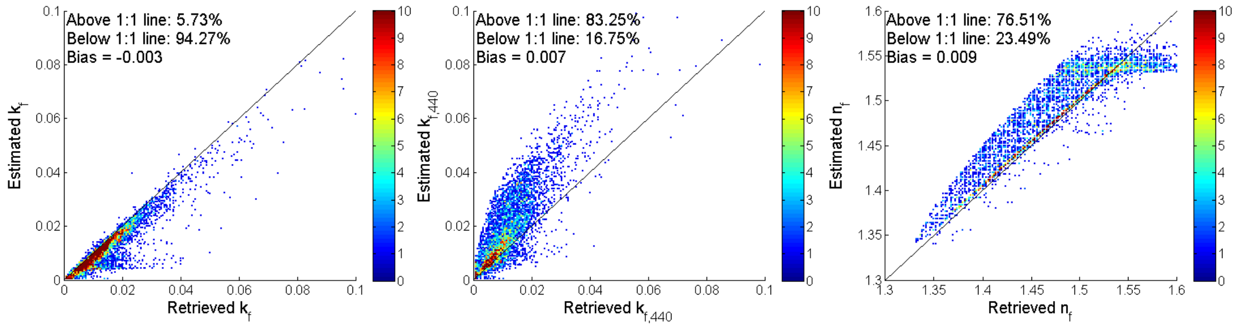

Figure 8. Data pair frequency of instantaneous imaginary parts of the complex refractive index at 675 nm (k_f), 440 nm ($k_{f,440}$), and real part at 440 nm (n_f) which are sorted according to ordered pairs (Retrieved, Estimated) in 0.0005 and 0.001 intervals for imaginary and real parts, respectively. "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.

In addition, I think what the reviewer said "Authors themselves claim that ~40% of the retrieval are above the average error" should refer to "Moreover, the mean relative error (RE) is 29.93%, and 61% of the data points is below the average relative error line." in the manuscript. It is not a constraint on the convergence of the algorithm, but to evaluate whether the convergence condition is reasonable. Therefore, it does not affect our statistical results.

Minor comments:

Line 70: I would suggest replacing "much information" to "sufficient information"

Response: We have revised in the manuscript as follows:

Line 73-74: "These radiation and polarization measurements can provide sufficient information to calculate the columnar aerosol optical depth (AOD) and further retrieve the aerosol microphysical parameters. "

Lines 87-89: "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 sub- modal aerosol components have not been given." I'm not sure what authors mean by this sentence, please, consider rephrasing.

Response: We rephrase this sentence as follows:

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."

Line 94: "... the linear interpolation method is used to match the SONET observations ...", please clarify if interpolation in time or alt/lat/lon or all together were used.

Response: This linear interpolation is only used in timescale. We clarify this point in manuscript:

Line 99: " The CMA stations closest to each SONET site were selected and the meteorological data were collocated in time with the SONET observations by linear interpolation between the nearest observations."

Line 210. It is claimed that error of 50% in AWf is "acceptable". No references or desired thresholds were provided to jump to such conclusion. Consider providing them, or rephrasing the sentence to a milder comparison.

Response: The revised sentence is as follows:

Line 268: "Affected by SC, the TRE of AW_c is also large due to n_c, but the TRE of AW_f is much smaller (50.05%). "

Line 308: "… improved a component inversion algorithm …", please refer to a baseline method. And what improvements authors refer to? No comparison to previous methods shown, is it complexity of the composition? Please clarify.

Response: This study improves the algorithm of Zhang et al. (2018), not only for multiphase solution calculation, but also for adding SC component. We revised this sentence as follows:

Line 389-390: "In the current study, we updated the refractive index calculation in a multi-component liquid system and improved the component inversion algorithm of Zhang et al. (2018) to retrieve atmospheric columnar aerosol components including …"

Line 353: I presume that another Dubovik 2000 paper, "Dubovik, O., and M. D. King, 2000: A flexible inversion algorithm for retrieval of aerosol optical properties from sun and sky radiance measurements. J. Geophys. Res., 105, 20 673¨C20 696." will be more suitable in the context it is referenced in the article.

Response: Thank you for your comment. We add this reference in manuscript.

Figure 5. I would appreciate to have numbers on pie charts too like figure 4, or an additional table with percentage of the components for each site.

Response: Because of the small space in the figure, we only increased the volume ratio of BC in the blank space. The fractions of other components are listed in the table S3 of supplementary as follows:

Table S3. The averaged mass fraction of aerosol components at SONET sites shown in figure 7.

Site	Fine mode					Coarse mode			AW	IS	OM
	BC	WIOM	WSOM	AN	AW_f	DU	SC	AW_c			
Lhasa	0.43%	7.85%	6.47%	1.89%	2.01%	81.34%	0.00%	0.00%	2.01%	1.89%	14.33%
Zhangye	0.05%	2.43%	2.36%	2.31%	1.79%	70.23%	11.53%	9.30%	11.09%	13.84%	4.79%
Kashgar	0.08%	4.57%	2.63%	2.51%	1.19%	65.63%	17.09%	6.30%	7.49%	19.60%	7.20%
Minqin	0.11%	2.97%	2.33%	6.13%	2.33%	65.70%	12.20%	8.23%	10.56%	18.33%	5.30%
Xi'an	0.60%	4.74%	9.62%	10.08%	6.56%	60.75%	3.37%	4.28%	10.84%	13.45%	14.37%
Beijing	0.69%	5.91%	7.67%	10.65%	6.08%	62.86%	2.85%	3.29%	9.38%	13.49%	13.59%
Nanjing	0.96%	6.26%	15.13%	13.79%	9.68%	46.94%	3.14%	4.10%	13.78%	16.93%	21.39%
Shanghai	1.30%	5.14%	10.78%	17.68%	10.33%	46.62%	3.87%	4.28%	14.61%	21.55%	15.92%
Harbin	0.97%	5.43%	12.45%	15.36%	10.80%	50.50%	2.21%	2.26%	13.07%	17.57%	17.88%
Hefei	0.80%	3.33%	11.51%	14.86%	10.34%	49.79%	3.91%	5.46%	15.80%	18.77%	14.84%
Songshan	0.59%	8.14%	6.31%	12.09%	6.63%	59.65%	3.10%	3.50%	10.13%	15.18%	14.45%
Chengdu	0.58%	2.72%	11.90%	22.54%	9.34%	42.10%	3.65%	7.17%	16.51%	26.19%	14.62%
Zhoushan	0.33%	3.55%	6.84%	17.06%	14.86%	42.82%	5.83%	8.70%	23.56%	22.89%	10.40%
Guangzhou	0.64%	2.84%	8.18%	23.80%	18.26%	31.54%	4.28%	10.46%	28.72%	28.08%	11.02%
Haikou	0.83%	2.32%	9.89%	22.03%	14.42%	33.29%	6.90%	10.32%	24.74%	28.93%	12.21%
Sanya	0.55%	0.32%	14.27%	24.78%	9.46%	37.41%	3.21%	10.00%	19.45%	27.99%	14.59%

Figure 6. Why only imaginary for fine and only 670? Fitting statistics is a necessary (but not sufficient) metric to justify that the method works. Please consider showing at least a real part at 670 too, but I would be more convinced to see all of them, all wl and fine/coarse. At least show minimal set, following the structure of Table s2 fine/coarse real/imaginary and 440 too, since they all referenced to have different level of uncertainty. Also, please, mention that it is fine mode in figure caption.

Response: We add the closure figure for the real and imaginary parts of the CRI at 440 nm. Because the spectral changes in the real part are ignored in the CRI inversion, the optical closure at other wavelengths have a similar pattern (We don't show that in the manuscript). The Figure is as follows and its description has been mentioned in the previous response.

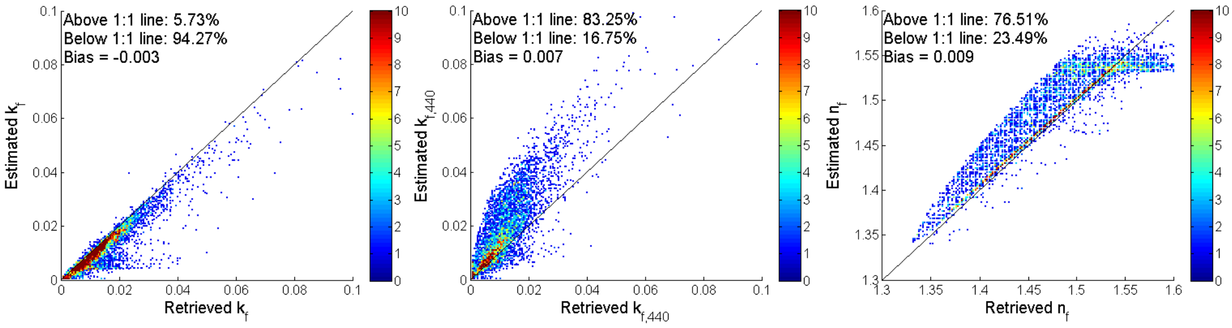

Figure 8. Data pair frequency of instantaneous imaginary parts of the complex refractive index at 675 nm (k_f), 440 nm ($k_{f,440}$), and real part at 440 nm (n_f) which are sorted according to ordered pairs (Retrieved, Estimated) in 0.0005 and 0.001 intervals for imaginary and real parts, respectively. "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.

For the coarse mode, only the real part of CRI (n_c) is used in the inversion because the imaginary part of the CRI for both AW_c and SC are zero. Figure R2 shows the comparison of n_c between retrieved and estimated from chemical components. The residual is tiny when the n_c is less than 1.534. To the left of the 1:1 line, when n_c equals 1.534 (real part of DU), there is a set of points with a large error. That is because the relative humidity is lower than 40% leading to the AW_c is close to 0.0. Hence, the real part of the coarse mode gets the minimum of 1.534. In contrast, the real part of the CRI for the DU component is increased up to the mean value of the retrieved refractive index at all available wavelengths which is higher than the mean value of DU's n_c. The reason for doing that is that DU is a mixture. Even so, when relative humidity is high, some underestimates still occur. Fortunately, 73% of the points are not influenced by the above factors, with the mean error of 0.01. The uncertainty of the retrieved CRI also causes the large residuals.

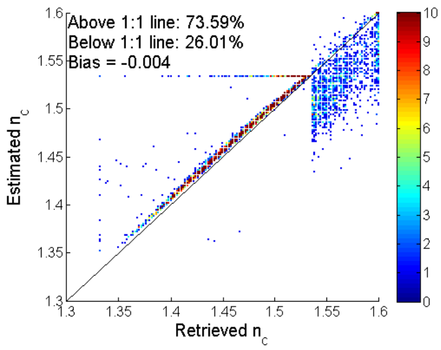

Figure R2. Data pair frequency of instantaneous imaginary parts of the complex refractive index at 440 nm (n_c) which are sorted according to ordered pairs (Retrieved, Estimated) in 0.001 intervals. The color represents the number of cases (color bar), and the solid black line shows the 1:1 line.

Figure 7. Is it possible to re-plot or at least re-paint so the color scheme will be the same as in figs 5 and 6?
Response: We re-plot this figure to match the color scheme of the other figures.

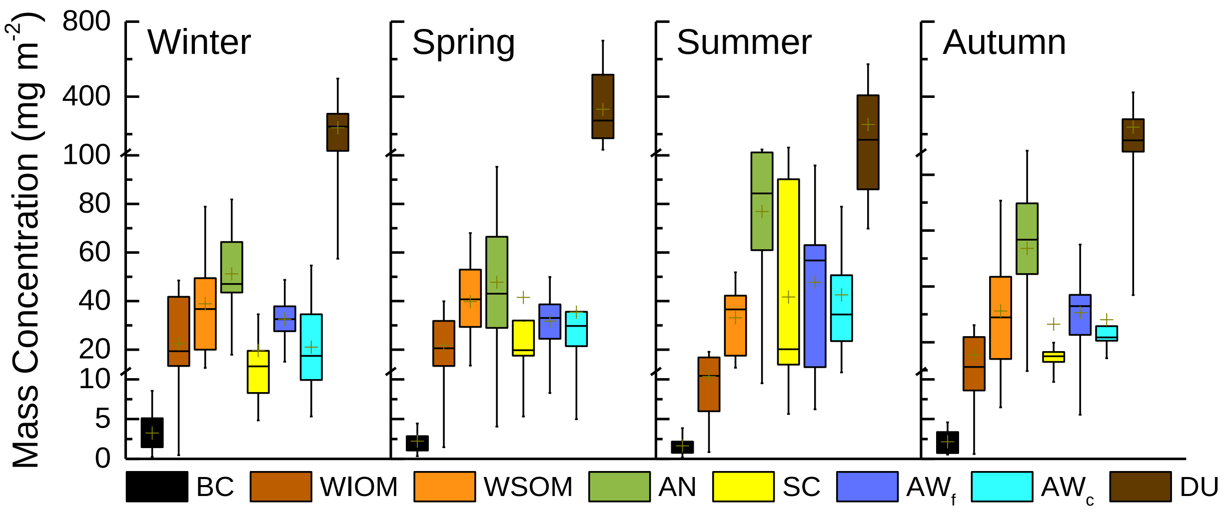

Figure 9. The mass concentrations of aerosol components in four seasons (winter, spring, summer and autumn). For the box-whisker plot, the mean value is indicated by a plus sign (+), and the median value by a short line inside the box (–). The top and bottom edges of each box represent the top and bottom quartiles (Q3 and Q1), and the corresponding whiskers are the outliers (Q3+1.5IQR and Q1-1.5IQR, IQR is interquartile range).

Table 2. Please specify what "No" in the head of the table means. If this is a number of observations, then why for quite different observation periods the numbers are so close? Was observation data filtered? Please, clarify.

Response: "No" is the meteorological station number. We explain it at the bottom of table 2.

Table s2 and s1. They are not a part of a publication provided (.pdf) and I don't see much reason why. I mean they are rather small and multiply referenced from the text, what is the reason to have one external half-page containing them?

Response: These two tables are referenced from other references to provide model input errors. For clarity, we added a description of them to the supplementary (S1).

Technical comments:

Figure 3. Size distribution axis. I believe it is μm3 *μm-2

Response: Thank you for pointing this out. We revised figure as follows:

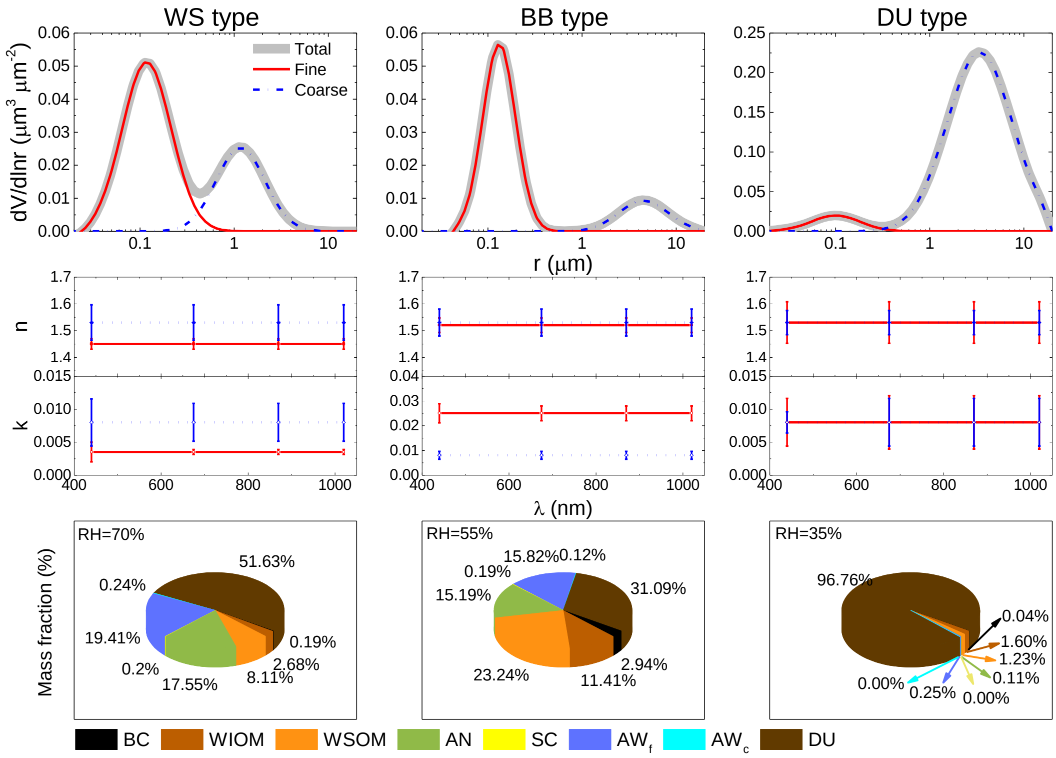

Figure 6. The fine and coarse-mode volume size distribution, complex refractive index and aerosol components of three typical aerosol models (WS: water soluble, BB: biomass burning, DU: dust).

Line 27-28: "… retrieval of the chemical composition …"

Response: Following the comment of another reviewer, we have deleted this sentence.

Line 49: "… simultaneously retrieved …"

Response: We revised this sentence as follows:

Line 52: "Zhang et al. (2018) simultaneously retrieved the WSOM and WIOM components but ignored the error in the refractive index introduced by the aerosol volume averaging method applied to the multicomponent liquid system."

Line 56: "… to solve for the refractive index in a multicomponent liquid system"

Response: We revised this sentence as follows:

Line 59: "hygroscopicity is introduced to solve for the refractive index in a multicomponent liquid system."

Line 57: "… in the algorithm …"

Response: We revised this sentence as follows:

Line 60: "The results are used in the algorithm to retrieve aerosol components … "

Line 70: "… radiation to determine the columnar water vapor"

Response: We revised this sentence as follows:

Line 73: "All bands provide both radiation and polarization measurements, except the 936 nm band which only measures radiation to determine the columnar water vapor."

Line 93-94: Probably author meant "To avoid observation uncertainties, only data from manned weather stations that are maintained regularly were used."

Response: We modified this sentence into the form suggested by the reviewer.

Line 185: "… by the different aerosol size distribution …"

Response: We revised this sentence as follows:

Line 243: "Each type is described by the different aerosol size distribution and refractive index parameters derived from Zhang et al. (2017)."

Line 246: "On the one hand"

Response: We revised this sentence as follows:

Line 310: "On the one hand, the imaginary part of the refractive index of BC is significantly higher than for the other components due to its strong absorption."

Line 247: "… than for the other components …"

Response: We revised this sentence as follows:

Line 310: "On the one hand, the imaginary part of the refractive index of BC is significantly higher than for the other components due to its strong absorption."

Line 252: "… data points are below …"

Response: We revised this sentence as follows:

Line 316: "The mean relative error (RE) is 27.1%, and 62.8% of the data points are below the average relative error line."

Line 260: "… high in spring …"

Line 260: "… to the dust transport from the northwest China …"

Response: We revised this sentence 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."

Line 261: "… seasons indicates that the aerosol at some sites"

Response: We revised this sentence as follows:

Line 337: "The difference between the upper and lower quartiles of AW_f in the summer is larger than in other seasons indicating that in the summer the aerosol at some sites has a low hygroscopicity."

Line 262: "… due to the observational errors …"

Response: We revised this sentence as follows:

Line 373: "The unusually high values at Shanghai in 2016 may be due to the observational errors."

Line 304: "… changes in the meteorological conditions …"

Response: We revised this sentence as follows:

Line 385: " Coarse mode aerosols usually derive from natural sources, and their variations can be associated with changes in the meteorological conditions."