Response to reviewer#1

Thanks for the reviewer’s helpful suggestions! The comments are addressed point-by-point and responses are listed below.

Comments: In this manuscript, the authors introduced a method to retrieve the real part of refractive index (RRI) of ambient aerosols from the measurements of the scattering intensities of size-selected aerosol particles by the combination of the differential mobility analyzer (DMA) and the single particle soot photometer (SP2). The authors pointed out that retrieval of the size-resolved RRI of ambient aerosols is the innovation of this paper in comparison with the measurements of the total equivalent aerosol RRI or aerosol RRI at a given diameter in previous studies. It is a meaningful topic to measure the real part of the complex refractive index of ambient aerosol particles. However, there are still some important issues should be considered before it is publishable.

Reply: We thank the anonymous reviewer’s comments and suggestions.

Comments: 1. The authors pointed out that “there might be significant variations in the aerosol RRI for aerosols of different diameter because the aerosol RRI is highly related to the aerosol density and chemical components…information of the size-resolved aerosol RRI can help to study the chemical information and the aging process of aerosols among different diameters”. However, the results of the size-resolved RRI of the ambient aerosols do not show significant variations among different diameters. The authors should give explanations.

Reply: Thanks for the comments. The ratios of aerosol chemical components are different for different diameters, which might lead to significant variations in aerosols RRI for different diameters. The aerosol chemical component, which is not measured in our study, may not vary significantly for different diameters during the test.

The following discussion would demonstrate that the ambient aerosol RRI can vary significantly among different diameter. The aerosol RRI are estimated by using the measured size-resolved main chemical components of the ambient aerosol from

Liu et al. (2014) in the North China Plain. The measured data is shown in fig. R1. From fig. R1, the ambient aerosols in the North China Plain are mainly composed of NH_4^+, NO_3^-, Ca^{2+}, SO_4^{2-}. These chemical components varies among different diameters. The aerosol RRI were estimated using these measured data and the method of Stelson (1990), and shown in fig. R1(b) in dotted black line. Results show that the aerosols RRI for different diameter change significantly between 1.46 and 1.59.

In our study, the DMA-SP2 system measures the aerosol diameter range between 200 and 450 nm. The content, to which the chemical components may change, is not well known due to the lack of size-resolved aerosol chemical information. More measurements were necessary to study the characteristics of the size-resolved RRI. However, this study mainly focus on the method of measuring the size-resolved RRI.

The reviewer gives a new insight into our future work. We also add some revisions accordingly at section 4.1.

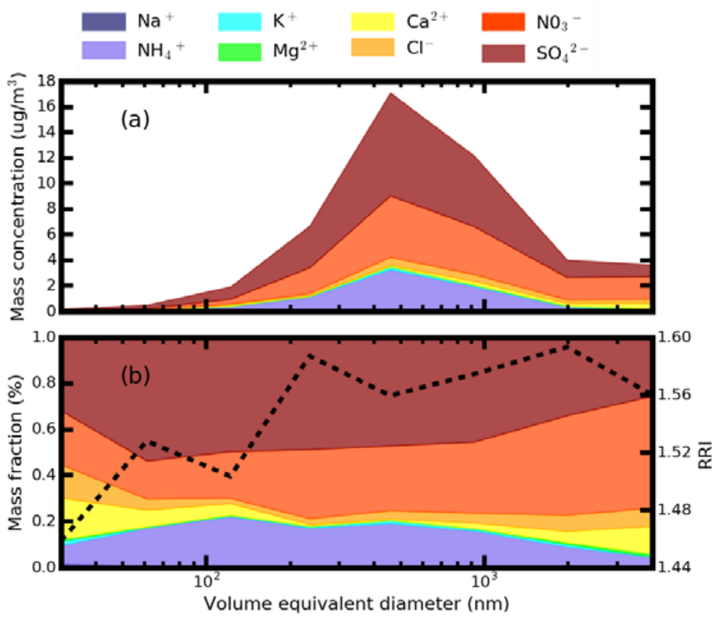

Figure R1. The average size distributions of the particle chemical composition during the a field campaign (Liu et al., 2014). Panel (a) shows the mass concentration of eight main species and panel (b) shows the relative mass fractions in individual impactor stage. The dotted black line in (b) shows the estimated aerosol RRI.

Comments: 2. The size-resolved aerosol RRI is retrieved based on the Mie scattering theory at a given particle diameter. What is the effect of the imaginary part of the complex refractive index on the retrieval?

Reply: Thanks for the comments. In our study, the effects of the imaginary part on the retrieval are not considered because we only select the BC free aerosols for this study.

At the same time, we added some discussions in section 4.2.2 on the uncertainties when the aerosols contain a small amount of BC cores that is below the detection threshold of SP2. Monte Carlo simulations were applied to investigate the influence of the BC core on the retrieved ambient aerosol RRI. We found that these particles can lead to less than 0.02 overestimation of the aerosol RRI for most of the conditions.

There are some organic components that may weakly absorb the light intensity. The imaginary part of the refractive index at a given wavelength λ (k_{λ}) of the BrC can be calculated as:

$$k_{\lambda 1} = k_{\lambda 2} \times (\frac{\lambda 2}{\lambda 1})^{w} \qquad (1),$$

Where w is defined by mass of BC to organic aerosol ratio (R) (Saleh et al., 2015)with:

$$w = \frac{0.21}{R+0.07} \qquad (2).$$

Based on the work of Saleh et al. (2015), the k_{550} can be expressed as:

$$k_{550} = 0.016 \times log_{10}(R) + 0.04 \qquad (3).$$

The values R ranges between 0.09 and 0.35 for different types of aerosols (Saleh et al., 2015). Based on equation (8), (9) and (10), the k_{1024} ranges between 0.01 and 0.024. The maximum value 0.024 is used for further analysis.

The uncertainties of the retrieved RRI when ignoring the effect of BrC are analyzed. Firstly, The scattering light intensity at a given diameter with a refractive index of 1.46 + 0.024*i* is calculated using the Mie model. Then the corresponding RRI are retrieved with given diameter and the calculated light intensity. The retrieved aerosol RRI for different aerosol diameter are shown in fig. 7(b). For the light absorbing particles, their scattering light intensity is smaller than that of the pure scattering particles with the same diameter and RRI. Therefore, the retrieved aerosol RRI is underestimated for most of the conditions. The differences between the given RRI value (1.46) and retrieved RRI value are lower than 0.006 for all of the diameters as shown in fig. 7(b) in the manuscript. The BrC component have little influence on the retrieved aerosol RRI.

The discussions of influence of BrC on the retrieving aerosol RRI are added in section 4.2.3 in the manuscript.

Comments: 3. The impact of non-sphericity of ambient aerosols on the light scattering cannot be neglected, especially for dry particles. The authors should also discuss the uncertainties introduced by the sphericity assumption based on the Mie theory.

Reply: Thanks for the comments. A lot of closure studies between the measured and calculated aerosol optical properties validate the non-sphericity of the ambient continental (Chen et al., 2014; Ma et al., 2014; Ma et al., 2011; Wex et al., 2002). Based on these studies, it is applicable that these particles are spherical for accumulation mode aerosols.

Comments: More specific comments:1. Some details of the method to retrieval real part of the refractive index based on the Mie scattering theory should be added.

Reply: Thanks for the comments. We added some descriptions about Mie scattering theory and the method at section 3.1. The method of retrieving the RRI are summarized as follows: (1) measuring the scattering peak height H values at a given diameter; (2) transferring the H into to the light scattering intensity S as denoted in equation 6 in the manuscript by the established relationship from calibration; (3) calculating the refractive index using equation 6 with the given diameter and S.

Comments: 2. Lines 57-58: the authors pointed out that “Up to now, there is no information in the literature of the size-resolved ambient aerosol RRI over the diameter range between 200nm and 500nm…”. However, the individual particle analysis combining scanning and transmission electron microscopy (SEM and TEM) have been widely used to derive size-resolved information of the complex refractive index of atmospheric aerosol particles (e.g., in the size range from 100 nm up to 50 μm in diameter) (Ebert et al.,2002, 2004; Kandler et al., 2007).

Reply: Thanks for the comments. We revised the manuscript correspondingly. We thank the reviewer for providing us alternative methods to measure the aerosol RRI. The information was summarized and added in the introduction part.

Comments: 3. Section 4.1: The field measurements were carried out at the AERONET BEIJING_PKU station. The results should be compared with the AERONET retrievals considering that the size-resolved RRI of the ambient aerosols doesn’t show significant variation among different diameters.

Reply: Thanks for the comments. The comparison of the results are shown here but not added in the manuscript. The used RRI from AERONET and our proposed method are these results from 9th to 19th, Match in 2018. Results shown that the RRI measured by the two methods is not correlated with each other. The RRI retrieved from AERONET is the results from column averaged value. At the same time, the aerosol optical properties measured by AERONET are at ambient RH. The RH in the mixed layer increases with height, and reaches larger than 90% frequently in the North China Plain (Kuang et al., 2016; Zhao et al., 2017). When the ambient RH is high, the aerosol takes water and then gets hygroscopic growth. The corresponding aerosol RRI should be lower than that of the dried aerosol particle. The measured RRI in our methods are those of the dried aerosol with RH lower than 40%. Therefore, we don’t think it necessary to present the comparison of the RRI measured by our method and the RRI from the AERONET retrievals.

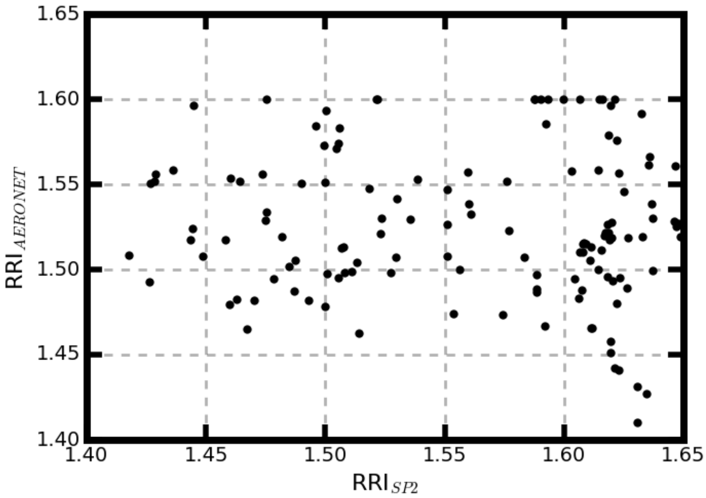

Figure R3. The comparison of the measured RRI by SP2 and AERONET.

Comments: 4. Lines 232-233: "For most ambient aerosols, the RRI ranges from 1.4 to 1.5 …". Some researches have reported the values around 1.53~1.57 for the RRI of most of dry components of atmospheric aerosols, and higher values for RRI of black carbon (BC) component (Xie et al., 2017). The authors should demonstrate their results with other measurements.

Reply: Thanks for the comments. Our proposed method focuses on measuring the RRI of the BC free aerosol. Our results show that the ambient BC free aerosol RRI locates around 1.46. More results in another paper in preparation show that the measured RRI can vary a wide range from 1.36 to 1.54. The measured RRI of ambient aerosol is lower than 1.5 because there are many cases that the RRI of organic matter lower than 1.5 (Moise et al., 2015).

At the same time, our method to measure the aerosol RRI is validated by measuring the RRI of ammonium chloride with the RRI of 1.642 as sample aerosol and the corresponding derived size-resolved RRI is 1.642 ± 0.02.

Comments: Typos/Grammar: 1. Line 12 and some other lines in the text: a space should be placed between the number and the unit.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:2. Lines 27-29 and some other lines in the text: it is better to use the past tense in review of the literature.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments: 3. Line 29: please rewrite the sentence "Valenzuela et al. (2018) also reports an uncertainty of 7% with the uncertainties of RRI of 0.1 in RRI."

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:4. Line 58: "the diameter range between 200nm and 500nm where the aerosol scattering coefficients contributes to…". "contributes" should be "contribute".

Reply: Thanks for the comments. We have changed the paragraph correspondingly.

Comments:5. Line 64 and some other lines in the text: "for aerosol of different diameter" should be "for aerosol of different diameters"

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:6. Line 90: "PNSD" first appears in Section 2.1, but it has not been defined.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:7. Lines 99-100 and some other places in the text: the physical quantities "V" and "Zp" should be set in italic in consistent with the equation.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:8. Line 102: "L" in Eq. (2) has not been defined.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:9. Lines 113 and 140: please distinguish the two "C" in Eqs. (5) and (6).

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:10. Lines 150 and 155: “equation (6)” and “equation 6” should be in a uniform format.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:11. Line 156: “as that described in section 2.2.1”. There is no section 2.2.1 in the manuscript.

Reply: Thanks for the comments. We have changed manuscript correspondingly. The true section should be section 2.1.

Comments:12. Line 177: “PH0” first appears in Section 3.2, but it has not been defined.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:13. Line 184: “” first appears in Section 3.2, but it has not been defined.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:14. Lines 175, 180, 184-185: “fig.2” should be changed into “fig.3”.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

Comments:15. Line 221: “SP” should be “SP2”.

Reply: Thanks for the comments. We have changed manuscript correspondingly.

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Ma, N., Birmili, W., Müller, T., Tuch, T., Cheng, Y.F., Xu, W.Y., Zhao, C.S., Wiedensohler, A. (2014) Tropospheric aerosol scattering and absorption over central Europe: a closure study for the dry particle state. Atmospheric Chemistry and Physics 14, 6241-6259.

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