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:

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

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

$$w = \frac{0.21}{R + 0.07} \tag{2}.$$

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

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

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 + 0.024i 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.

Chen, J., Zhao, C.S., Ma, N., Yan, P. (2014) Aerosol hygroscopicity parameter derived from the light scattering enhancement factor measurements in the North China Plain. Atmos. Chem. Phys. 14, 8105-8118.

Kuang, Y., Zhao, C.S., Tao, J.C., Bian, Y.X., Ma, N. (2016) Impact of aerosol hygroscopic growth on the direct aerosol radiative effect in summer on North China Plain. Atmospheric Environment 147, 224-233.

Liu, H.J., Zhao, C.S., Nekat, B., Ma, N., Wiedensohler, A., van Pinxteren, D., Spindler,

G., Müller, K., Herrmann, H. (2014) Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China Plain. Atmospheric Chemistry and Physics 14, 2525-2539.

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.

Ma, N., Zhao, C.S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y.F., Deng, Z.Z., Liu, P.F., Xu, W.Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildenberger, K., Henning, S., Yu, J., Chen, L.L., Zhou, X.J., Stratmann, F., Wiedensohler, A. (2011) Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study. Atmos. Chem. Phys. 11, 5959-5973.

Moise, T., Flores, J.M., Rudich, Y. (2015) Optical properties of secondary organic aerosols and their changes by chemical processes. Chemical Reviews 115, 4400-4439. Saleh, R., Marks, M., Heo, J., Adams, P.J., Donahue, N.M., Robinson, A.L. (2015) Contribution of brown carbon and lensing to the direct radiative effect of carbonaceous aerosols from biomass and biofuel burning emissions. Journal of Geophysical Research: Atmospheres 120, 10,285-210,296.

Stelson, A.W. (1990) Urban aerosol refractive index prediction by partial molar refraction approach. Environ.sci.technol 24:11, 1676-1679.

Wex, H., Neusüß, C., Wendisch, M., Stratmann, F., Koziar, C., Keil, A., Wiedensohler, A., Ebert, M. (2002) Particle scattering, backscattering, and absorption coefficients: An in situ closure and sensitivity study. Journal of Geophysical Research: Atmospheres 107, LAC 4-1-LAC 4-18.

Zhao, G., Zhao, C., Kuang, Y., Tao, J., Tan, W., Bian, Y., Li, J., Li, C. (2017) Impact of aerosol hygroscopic growth on retrieving aerosol extinction coefficient profiles from elastic-backscatter lidar signals. Atmospheric Chemistry and Physics 17, 12133-12143.

Response to reviewer#2

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

Comment 1: General comments: The present manuscript describes a method for deriving the real part of the refractive index by means of a differential mobility selector (DMA) and scattering intensities measured with a SP2. The derivation of the real part of the refractive index of a quasi-mono disperse aerosol is not completely new. What is new, however, is the application with the use of the SP2, which in a unique way can also determine the mixing state of the aerosol within certain limits. This ensures that errors caused by unknown imaginary parts of the refractive index are minimized. The method shown is limited to non-absorbent particles.

Reply 1: We agree with the anonymous reviewer's comments.

Comment 2: The reviewer thinks that the current limitations and consequences have not been adequately presented. In particular, a consideration of the uncertainties in violation of the restrictions (weakly absorbing particles) is lacking.

Reply 2: Thanks for the comments. The reviewer provides a good view in uncertainties analysis of our proposed method.

There are some brown carbon (BrC) that absorb the light intensity in the near infrared range. The imaginary part of the refractive index at a given wavelength λ (k_{λ}) of the BrC can be calculated as:

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

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

$$w = \frac{0.21}{R + 0.07} \tag{2}.$$

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

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

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.

We added some discussions in section 4.2 on the uncertainties when the aerosols contain a small amount of BC cores that are 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. These particles can lead to less than 0.02 overestimation of the aerosol RRI for most of the conditions. More details are shown in **Reply 4**.

Some corresponding discussions were added in section 4.2.3 and 4.2.4 in the text.

Comment 3: In laboratory experiments, as shown with ammonium sulfate and ammonium chloride, this is easily possible. The application to a complex ambient aerosol, on the other hand, was not treated sufficiently. The example shown in chapter 4.1 shows results of measurements in Beijing, where a complex mixed aerosol is present (that measurement place was characterized as urban roadside; line 96).

Reply 3: We thank the anonymous reviewer's comments and suggestions. In this work, we mainly focus on the method of measuring the aerosol size-resolved real part of the refractive index (RRI). The ammonium sulfate is used for calibration, and the ammonium chloride is used for validation. These studies are easy but power

demonstrations that our proposed is applicable of measuring the BC-free aerosol. More results about field measurements using this method can refers to another work at https://www.atmos-chem-phys-discuss.net/acp-2019-250/.

Comment 4: An error estimation is missing: a) what happens with weakly absorbing organic droplets, b) what happens with internally mixing particles with a small soot core when the incandescence signal is below the detection threshold of SP2. How large are the expected errors in the real part of the refractive index?

Reply 4: We do appreciate the comments. The reviewer provides a good view in uncertainties analysis of our proposed method. We added discussions on this point in section 4.2.

For the organic droplets, the light absorption of these components is ignorable as detailed in reply 2.

There are some particles with a small soot core and the incandescence signals are below the detection threshold of SP2. The derived aerosol RRI should be influenced by small soot core. Uncertainties might be resulted when deriving the RRI for these BC-contained aerosols.

For the BC-contained aerosol, Aquadag soot particles with effective density of 1.8 g/cm³ is used to determine the lower limit of the BC particle diameter when the incandescence signals can be detected by SP2. The calibration procedure is conducted the same as that of the ammonia sulfate in the manuscript. The diameters (Dp) of the aerosol passing through the DMA are manually changed from 60 to 400 nm with a step of 20 nm. The relationships between the measured incandescence signal height and the Dp are shown in fig. R2. From fig. R2, we conclude that our SP2 is not capable of measuring the Aquadag soot particles lower than 80 nm.



Figure R2. The calibrated relationship between the incandescence peak height and the BC diameter for both the incandescence high gain channel and the incandescence low gain channel.

We derived the aerosol equivalent refractive index when the aerosol have BC cores lower than 80 nm with two steps. 1, the scattering strength of the BC-containing aerosols are calculated based on Mie scattering theory with a given core and total diameter. 2, the scattering strength are used to deriving the equivalent refractive index with assuming that the BC-containing aerosols are pure scattering aerosols.

Monte Carlo simulations were applied to investigate the influence of the BC core on the retrieved ambient aerosol RRI. Firstly, the aerosol diameter are first chosen between 200 nm and 500 nm. Then the core diameter are random determined lower than 80 nm. The core diameters flow the log-normal distribution with the mean core diameter of 120 nm (Raatikainen et al., 2017). When calculating the scattering strength, the complex refractive index of the core 1.8+0.54i (Zhao et al., 2018) is used. The complex refractive of the shell adopts the measured mean values (1.46+0i) during the field measurements. The scattering strength can be calculated with the above information and the Mie scattering Model. Then with the calculated scattering strength, the equivalent real part of the refractive index (RRI) can be derived with assuming that the aerosols are pure scattering aerosols. If the core diameter is zero, the derived aerosol equivalent aerosol RRI should be 1.46.

For each aerosol diameter, the Monte Carlo simulations were conducted for

10000 times. Figure. R3 gives the retrieved aerosol equivalent RRI at different diameters. Results show that the retrieved aerosol equivalent RRI are larger than 1.46 for all of the given aerosol diameters. When the aerosols have BC core, the scattering strength are larger than that of pure scattering aerosols with the same aerosol diameter. The derived mean equivalent RRI tend to be closer to 1.46 when the aerosol diameter is larger, where the BC core contributes less and the influence of the BC core are smaller. The derived mean aerosol equivalent RRI is 1.47 and 1.462 at 200 nm and 500 nm respectively. At the same time, the uncertainties associated with the equivalent RRI are larger when the aerosol diameter is smaller. We conclude that the uncertainties associated with BC core are smaller than 0.01 when the aerosol diameter are larger than 250 nm. The maximum of the difference of the derived RRI is 0.02.

The above corresponding discussions were added in the uncertainties analysis part in section 4.2.2 of the manuscript.



Figure R3. The retrieved equivalent aerosol RRI at different aerosol diameter. The filled color represents the 5^{th} and 95^{th} percentiles.

Comment 5: The reviewer believes that this work can make a good step in the optical characterization of sub-micrometer particles using the SP2, and that subsequent work can build on it. Therefore, the reviewer thinks that the manuscript can be published after a major revision.

Reply 5: We thank the reviewer's comments.

Comment: Specific comments: Title: The method shown is very general, but applied to SP2 in the present study. The study is thus adapted to the size range, the size resolution and the optical geometry of the SP2. The author should consider whether the application of the method to SP2 should be mentioned in the title.

Reply: Thanks for the comment. We have changed the title of the manuscript.

Comment: Introduction: Can the author give a first estimate on the accuracy of RRI measurements required to make statements on the chemical composition?

Reply: Traditionally, the ambient aerosol RRI is calculated using some main inorganic aerosol component and the influence of organic is ignored. However, the organic component contributes more than 20% of the total aerosol component in the North China Plain (Hu et al., 2012; Liu et al., 2014). RRI of the organic aerosol changes significantly between 1.36 and 1.66 (Moise et al., 2015). Ignoring the organic component may lead to significantly uncertainties when estimating the aerosol RRI.

In the introduction part, we added some information about the uncertainties of calculating the aerosol RRI using the measured aerosol chemical composition due to aerosol organic component.

We found that the aerosol RRI was determined by aerosol density rather than the aerosol chemical composition, which is discussed in detail in another paper <u>https://www.atmos-chem-phys-discuss.net/acp-2019-250/</u>.

Comment: Line 36: Typo: "Hänel"

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

Comment: Lines 89, 90: The measurements provide the necessary data in five minute intervals. However, no conclusion can yet be drawn that the RRI can be derived with a time resolution of five minutes.

Reply: Thanks for the comments. We have changed the corresponding descriptions.

Comment: Line 121: to be precise, the power is about 1 W/m2 circulating power in cavity

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

Comment: Line 131: what is the unit of the peak height H.

Reply: Thanks for the comments. The avalanche photo-detector measures the light signals and changes the signals series to a light scattering of incandescent intensity time series. There is no unit for these signals. Correspondingly, there is no unit for the peak height H.

Comment: Lines 133,134: How were BC containing particles ruled out for ambient measurements.

Reply: The BC containing particles are ruled out by not counting the scattering signal peak height of these aerosol particles when the incandescent peak height is larger than 500. The manuscript is changed correspondingly.

Comment: Section 3.1: Shouldn't the signals be the same value at 45° and 135° due to the circulating wave in the cavity?

Reply: Yes, the received signals intensity are at 45° and 135° due to the circulating wave in the cavity. When calculating the scattering intensity, the aerosol scattering coefficient at both 45° and 135° in the cavity are considered.

Comment: Line 145: To avoid misunderstandings: The SP2 can determine the scattering signal in a certain scattering angle range. But not the scattering coefficient! **Reply:** Thanks for the comment. We add some sentence to distinguish the light scattering intensity by aerosols and the measured light scattering signals by SP2 at the manuscript.

Comment: Figure 2: Is the scattering strength the same as the scattering intensity S? Please use consistent notations.

Reply: Thanks for the comments. We have changed the manuscript accordingly. All of the scattering strength are changed into scattering intensity S.

Comment: Line 151: monotonously instead of homogeneously.Reply: Thanks for the comment. We have changed the manuscript correspondingly.

Comment: Line 159: Establishing the threshold value at 1000 seems somewhat arbitrary. Is there a justification for this?

Reply: Thanks for the comment. This value is selected based on the analyzing the noise of the incandescence signals. The value 1000 depends on the instrument's stability. When recording the aerosol incandescence signals, the instrument gets the maximum values of 60000. In calibration studies using ammonium sulfate, the fit maximum incandescence signal peak height is 720, which is only 1.2% of the measurement range. This can be caused by signal noise. This value can be different for different instruments. We added some descriptions at the manuscript correspondingly.

Comment: It would be better to read to bring figure S2 und figure 3 together.

Reply: Thanks for the comment. We have changed the figure and manuscript correspondingly.

Comment: Lines 166 and 167: Refer to figure S1

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

Comment: Line 175: Please check that sentence. I can't see different marker for different diameters.

Reply: Thanks for the comment. This sentence is misleading. We changed this sentence into: The H_0 values of corresponding to different elementary charges are labeled with different markers in fig. 3.

Comment: Is fig. 2 the correct figure?

Reply: The figure number is not right. We have changed it into fig. 3 in the manuscript.

Comment: Line 177: What is "PH0"?

Reply: Thanks for the comment. We have changed the PH_0 into H_0 , it is the geometric mean value of H.

Comment: Line 180: Shouldn't it be Fig 3.

Reply: Thanks for the comment. The text in the manuscript has been changed correspondingly.

Comment: Lines 180,181 and Figure 3: Check if the peak height is plotted versus the mobility diameter Zp and not versus the geometric diameter Dp?

Reply: Thanks for the comment. We have checked the result and find the peak height is plotted versus the mobility diameter Dp, but not the electoral mobility diameter.

Comment: Line 184: "Dp superscript tilde" not defined

Reply: Thanks for the comment. We have changed the manuscript correspondingly. The \widetilde{Dp} is the corresponding diameter of the aerosols that share the same Z_p but different charges with those particles that have diameter of Dp with one charge.

Comment: Line 184: There is no dashed line in figure 2

Reply: Thanks for the comment. The figure number is not correctly labeled here and we have changed it into figure 3(a).

Comment: Lines 194 and 200 : Please bring references for the refractive index.Reply: Thanks for the comment. We have changed the manuscript correspondingly.

Comment: Figure 4a: Should be scattering intensity instead of scattering strength

Reply: Thanks for the comment. We have changed the figure.

Comment: Lines 192 – 208, Figure 4: For the reader it is not obvious at first sight which value was calibrated! What is the value of the calibration factor C? The reviewer thinks it is worth giving a short summary list of all steps necessary for deriving RRI. For an absolute calibration, the slope of about unity is more important than the references to the correlation coefficient. The high correlation coefficient is, as written, a good indicator for the potentially high accuracy of this method.

Reply: Thanks for the comment. We changed the figure 4. When comparing the scattering peak height and the scattering intensity, C is assuming to be unity in this study. The values of C and the slope should change proportionally. We agree with the reviewer that the slope is more important than the correlation coefficient. Some discussions about the slope at the end of section 3.2 were added to the manuscript. At the same time, we added a short summary of the step for deriving the RRI at the end of section 3.1.

Comment: Line 211: It would be good to have some additional information, e.g. the mean BC concentration and the number of fractions of internally/externally mixed particles and coated particles provided by SP2. How was it ensured that the purely externally mixed non-absorbent particles were used in the calculation of the RRI? **Reply:** Thanks for the comment. We added the information of the BC mass concentrations integrated by the SP2 in the manuscript. It is 6.31 μ g/m³. The corresponding aerosol scattering coefficient is 385 Mm⁻¹.

The number fractions of internally mixed particles and coated particles retrieved from SP2 were provided here. The measured mean mixing states of the ambient aerosol are shown in fig. R4.

From fig. R4(a), the BC core diameter (Dc) is lower than the Dp for most of the particles when the aerosol Dp is larger than 150 nm. There are a lot of particles with Dc larger than Dp when the aerosol is lower than 150 nm, which reflects the complex morphology of these BC-contained aerosols (Peng et al., 2017; Peng et al., 2016).

There are two mode for the BC-contained aerosol, with one thinly coated or uncoated aerosol mode locating near the Dc equaling Dp line and another thickly coated aerosol mode appearing that the Dc locates between 100 nm and 150 nm, which is in accordance with the previous study of the BC mixing states (Wu et al., 2018). The measured total aerosol PNSD, the measured BC-contained aerosol PNSD, the BC-free aerosol PNSD and the ratio of the BC-contained aerosol to total aerosol PNSD between 80 nm and 560 nm are shown in fig. 4(b). The PNSD for the BC-contained aerosols are calculated from the measurement from SP2. As for the PNSD for the BC-free aerosol, these values between 180 nm and 560 nm come from the measurement of SP2 and these values between 80 nm and 180 nm is calculated by subtracting the total aerosol PNSD to the PNSD of the BC-contained aerosol. Results show that both the total aerosol PNSD and BC-free aerosol PNSD peak around 120 nm. As for the BC-contained aerosol PNSD, it peaks at around 170nm. The ambient aerosol are mainly composed of BC-free aerosol and the BC-contained aerosol contributes less than 0.20 of the total aerosol over the measured size range. The ratio increases from about 0.03 to 0.2 when the aerosol diameter increases from 80 nm to 300 nm. The ratio decreases with the increment of diameter when the diameter is larger than 300 nm. Therefore, the ambient BC-contained aerosol only contributes to less than 20% of the total aerosols. The measured mean PNSD and particle mass size distribution (PMSD) of the rBC is shown in fig. 4(c). The PNSD and PMSD of the rBC peaks at 120 nm and 330 nm respectively. The measured PNSD and PMSD of the rBC is in accordance with many previous studies of the rBC distribution (Schwarz et al., 2006; Sharma et al., 2017).

The above information would help know the surrounding condition of the measurement sites. These results will be presented in another paper in preparation and are not included in this manuscript.



Figure R4. (a) Measured number concentration distributions for different aerosol diameters with different Dc. (b) measured total aerosol PNSD, the BC-free aerosol PNSD, the BC-contained aerosol PNSD and the ratio between the BC-contained aerosol to total aerosol number concentrations are shown in red dashed line marked with hexagon, in green line marked with square, blue dashed line and in dark solid line respectively. (c) the measured number size distribution of the rBC for different Dc is shown in red dashed line and the measured mass size distribution for different Dc is shown in dark solid line.

Comment: Line 216: Can the authors estimate the fraction of the light scattering size distribution that is covered.

Reply: Thanks for the comment. The fraction of the light scattering size distribution in the range between 200 nm and 450 nm is 0.63. We added the estimated result in the manuscript.

Comment: Lines 22ff: The uncertainty of the transfer function is covered by the H fitting, since the transfer function is a system function and relatively stable and also covered by a DMA calibration with size standards. How are other influences taken into account, e.g. uncertainties in sheath air flow or CPC counting efficiency?

Reply: Thanks for the comment. The uncertainties in sheath air flow would influence the transfer function. However, the flow rate of the sheath flow is controlled by a

circulatory system, which is very stable and the uncertainties is lower than 1% to our knowledge. This low fluctuation of sheath flow would have litter influence on the aerosol transfer function when the Q_{sh} in equation 3 changes by 1%. At the same time, the CPC counting efficiency has no impact on the retrieving of aerosol RRI.

Based on equation 6, the aerosol RRI are influenced by the aerosol diameter and the scattering intensity. The uncertainties of the aerosol diameter resulted from the DMA transfer function, which were fully resolved by using the Gaussian distribution to fit the aerosol scattering signal height distribution. Another source of the uncertainties comes from the measured scattering peak height by SP2, which was discussed in detail in section 4.2.

Comment: Line 225: HW not defined.

Reply: Thanks for the comment. The HW is the half width of the transfer function. We have changed the manuscript correspondingly.

Comment: Line 227 to 229: Does this mean that the additional broadening by the H distribution function is 1.073?

Reply: Yes.

Comment: Lines 230,231: Can the authors give more details about the uncertainty analysis?

Reply: Thanks for the comment. More details about the uncertainty analysis are added in the manuscript.

The steps of conducting the uncertainties are as follows. Firstly, the theoretical scattering intensity that can be measured by the SP2 for a given aerosol diameter and RRI are calculated. The scattering intensity are changed by $\pm 6.8\%$ (the uncertainties of the measured scattering intensity by SP2) and the corresponding RRI can be derived using the given aerosol diameter and the changed scattering intensity. Finally, the derived RRI are compared with the initial aerosol RRI. The uncertainties are

analyzed for different aerosol diameter and different RRI.

Comment: Lines 243, 244: How can it be ensured in a mixed aerosol that BC containing particles are excluded and how big would the error be if small amounts of BC affect the measurement?

Reply: Thanks for the comment. This has been addressed in general comment 4. We have added some text in section 4.2 to discuss this.

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- 1 Method to measure the size-resolved real part of aerosol refractive index using
- 2 differential mobility analyzer in tandem with single particle soot photometer
- 3

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Abstract

9 Knowledge on the refractive index of ambient aerosol can help reduce the uncertainties in estimating aerosol radiative forcing. A new method is proposed to retrieve the size-resolved real part 10 11 of **RI**-refractive index (RRI). Main principle of deriving the RRI is measuring the scattering intensity 12 by single particle soot photometer of size-selected aerosol. This method is validated by a series of calibration experiments using the components of known **RRI**. The retrieved size-resolved **RRI** cover a 13 14 wide range from 200 nm to 450 nm with uncertainty less than 0.02. Measurements of the size resolved 15 real part of the aerosol refractive indexRRI can improve the understanding of the aerosol radiative effects. 16

17 **1 Introduction**

Aerosols exert significant influence on the earth energy budget by scattering and absorbing 18 19 radiation (Ramanathan and Carmichael, 2008). There still remain great uncertainties when estimating the aerosol effective radiative forcing (RF) (Ghan and Schwartz, 2007) and an accurate estimation of 20 the aerosol optical properties can help reduce the RF variations. The optical properties of the ambient 21 aerosol particles are determined by their particle size and complex refractive index (RI, m=n+ki) 22 (Bohren and Huffman, 2007;Levoni et al., 1997). Despite that the ambient aerosol particle size 23 24 distribution can be measured with high accuracy (Wiedensohler et al., 2012), an accurate measurement of the ambient aerosol RI remains challenging. The RI is also widely used in remote sensing 25 (Redemann et al., 2000; Dubovik, 2002; Zhao et al., 2017) and atmospheric modelling (Ghan and 26 Schwartz, 2007;Kuang et al., 2015) because the aerosol single scattering albedo (SSA) and aerosol 27 28 scattering phase function are highly related with the RI. At the same time, a small uncertainty in the real part of the RI (RRI) can lead to great uncertainties when estimating the aerosol RF. Zarzana et al. 29

(2014) finds-found that a variation of 0.003 in RRI can lead to uncertainties of 1% in RF for nonabsorbing ammonium sulfate particles. Moise et al. (2015) estimates estimated that the RF will-would increase 12% when the RRI varieds from 1.4 to 1.5. Valenzuela et al. (2018) reported that the uncertainties in RF is estimated to be 7% when the aerosol RRI varied by 0.1.also reports an uncertainty of 7% with the uncertainties of RRI of 0.1 in RRI. Therefore, it is pressing that the uncertainties of the RI be reduced when estimating the RF.

Many methods were proposed to derive the RRI. The RRI can be estimated by linear volume average of the known aerosol chemical components by

38

$$\mathbf{n} = \sum_{i} f_{i} n_{i} \tag{1}$$

where f_i and n_i is the volume fraction and known partial refractive index of *i*th component (Wex et 39 40 al., 2002; Hand and Kreidenweis, 2002; Hänel, 1968; Liu and Daum, 2008). The aerosol RRI can also be calculated by partial molar refraction approach (Stelson, 1990; Hu et al., 2012) which is essentially 41 consistent with the linear volume method (Liu and Daum, 2008). The ambient aerosol RRI can be 42 derived by synthetically using the radiative transfer calculations and the ground-based solar extinction 43 and scattering measurements (Wendisch and Hoyningen-Huene, 1994, 1992). Sorooshian et al. (2008) 44 45 developed a method to measure the aerosol RRI based on the differential mobility analyzer (DMA) 46 and an optical particle counters. The RRI is-could be retrieved from the known particle size from the DMA and the aerosol scattering intensity from the Optical Particle Counter (OPC) for aerosol particles 47 larger than 500 nm. The Scanning Mobility Particle Sizer (SMPS) and OPC is was used in combination 48 49 to derive the RRI by aligning the particle size distributions in the instrument overlap regions (Hand 50 and Kreidenweis, 2002; Vratolis et al., 2018). The aerosol effective RRI is-was also retrieved by applying Mie scattering theory to the aerosol particle number size distribution, aerosol bulk scattering 51 52 coefficient and aerosol absorbing coefficient data (Cai et al., 2011;Liu and Daum, 2000). Spindler et 53 al. (2007) retrieved the aerosol RRI value by using the cavity ring-down spectroscopy to measuring 54 the scattering and absorbing properties of bulk aerosols. Eidhammer et al. (2008)) measured the light 55 scattering at different angles and retrieved the RRI. Similarly, the aerosol RRI is was retrieved by measuring the aerosol phase function (Barkey et al., 2007). Recently, a method by using the single 56 57 particle mass spectrometry is was proposed to measure the aerosol RRI (Zhang et al., 2015). At the 58 same time, aerosol time-of-flight mass spectrometer is-was proved to be capable of measuring the aerosol RRI (Moffet et al., 2008). The aerosol RRI can also be retrieved from the Mie spectroscopy by 59

60 using the optical tweezers in the laboratory (Shepherd et al., 2018).

Up to now, there is no information in the literature of the size-resolved ambient aerosol RRI(Ebert
 et al., 2004;Kandler et al., 2007;Ebert et al., 2002) over the diameter range between 200nm and 500nm
 where the aerosol scattering coefficients contributes to the total scattering coefficients most (Tao et al.,
 2017;Kuang et al., 2018). All the instruments mentioned above can only measure the total equivalent
 aerosol RRI or aerosol RRI at a given diameter. However,

Mmany studies show that aerosol of different diameters shares different properties such as shape 66 67 (Zhang et al., 2016; Peng et al., 2016), density (Qiao et al., 2018), aerosol hygroscopicity (Wang et al., 2017) and most importantly, the chemical components (Liu et al., 2014; Hu et al., 2012). Thus, there 68 69 might be significant variations in the aerosol RRI for aerosols of different diameters because the aerosol RRI is highly related to the aerosol density (Liu and Daum, 2008) and chemical components 70 71 (Stelson, 1990). On the other way round, information of the size-resolved aerosol RRI can help to study the chemical information and the aging process of aerosols among different diameters. Therefore, 72 measurement of the size-resolved aerosol RRI is necessary. 73

Up to now, there are few information in the literature of the size-resolved ambient aerosol RRI 74 75 (Ebert et al., 2004;Kandler et al., 2007;Ebert et al., 2002). Traditionally, the size-resolved ambient aerosols RRI are estimated by measuring the molar fraction or volume fraction of main aerosol 76 chemical compositions. However, the influence of organic component on the aerosol RRI is ignored. 77 The organic component contributes more than 20% of the total aerosol component in the North China 78 79 Plain (Hu et al., 2012;Liu et al., 2014). At the same time, RRI of the organic aerosol changes significantly between 1.36 and 1.66 (Moise et al., 2015). Ignoring the organic component may lead to 80 significantly uncertainties when estimating the aerosol RRI. There were no technique, to our 81 knowledge, that directly measures the size-resolved aerosol optical properties and derives the size-82 83 resolved aerosol RRI.

In this study, a novel method is proposed to measure the size-resolved ambient aerosol RRI by using a DMA in tandem with a single particle soot photometer (SP2). The principle of the system is using the SP2 to measure the scattering properties of size-selected aerosols. Knowing the aerosol diameter and corresponding scattering intensity, the size-resolved aerosol RRI can be retrieved based on the Mie scattering theory. This proposed method can measure the ambient aerosol RRI over a wide size range with high accuracy. The measurement system is employed in a field campaign in the North 90 China Plain and the corresponding results are further discussed.

91 The structure of this manuscript is as follows: section 2 provides the instruments setup and details 92 of the instrument. The method <u>toof retrieving retrieve</u> the size-resolved aerosol RRI is given in section 93 3. Section 4 shows the discussions about the uncertainties of the proposed method and field 94 measurements results of the size-resolved aerosol RRI. Conclusions come at the last part.

95 **2 Instrument**

96 2.1 Instrument Setup

97 The instrument setup is schematically shown in fig. 1(a). Firstly, the dried sample aerosols are guided to a X-ray soft diffusion charger and then lead to a DMA (Model 3081, TSI, USA). The quasi-98 monodisperse aerosols that pass though the DMA at a given diameter are then drawn into a SP2 to 99 measure the aerosol scattering properties with a flow ratio of 0.12 lpm and a condensation particle 100 counter (CPC, Model 3776, TSI, USA) to count the aerosol number concentration with a flow ratio of 101 102 0.28 lpm respectively. Thus, the sample flow (Q_a) of the DMA is 0.4 lpm. Accordingly, the sheath flow (Q_{sh}) of the DMA is 4 lpm. The DMA is set to scan the aerosols diameter from 12.3 to 697 nm 103 104 over a period of 285s and repeats after a pause of 15 s. Thus, tThe combination of DMA, CPC and SP2 105 can provide information of the aerosol PNSD-particle number size distribution (PNSD) and size resolved RRI.and size-resolved RRI every 5 minutes. 106

On 8th, June, 2018, the measurement system was employed at the filed measurement of AERONET station of BEIJING_PKU (N39°59', E116°18') to test the reliability of retrieving the ambient size-resolved RRI. This measurement site locates on the north west of the city of Beijing, China and is about 1.8 km north of the Zhongguancun, Haidian District, which is one of the busiest areas in Beijing. It is surrounded by two main streets: Zhongguancun North Street to the west and Chengfu Road to the south. This site can provide representative information of the urban roadside aerosols (Zhao et al., 2018).

114 **2.2 DMA**

115 When a voltage (*V*) is applied to the DMA, only a narrow size range of aerosol particles, with the 116 same electrical mobility (Z_p) can pass through the DMA (Knutson and Whitby, 1975). The Z_p is 117 expressed as:

(2)

118
$$Z_P = \frac{Q_{sh}}{2\pi VL} ln(\frac{r_1}{r_2})$$

where Q_{sh} is the sheath flow rate; <u>*L* is the length of the DMA;</u> r_1 is the outer radius of annular space and r_2 is the inner radius of the annular space. The transfer function refers to the probability that a particle with a certain electrical mobility can pass through the DMA. For a given *V*, the transfer function is triangular-shaped, with the peaking value of 100% and a half width (HW) of

$$\Delta Z_p = Z_P \frac{Q_a}{Q_{sh}} \tag{3}.$$

124 The aerosol Z_p , which is highly related to the aerosols diameter (D_p) and the number of elementary 125 charges on the particle (n), is defined as:

126
$$Z_p = \frac{neC(D_p)}{3\pi\mu D_p} \tag{4}$$

127 where *e* is the elementary charge; μ is the gas viscosity coefficient, $C(D_p)C(D_p)$ is the Cunningham 128 slip correction that is defined by:

$$C = 1 + \frac{2\tau}{D_p} (1.142 + 0.558e^{-\frac{0.999D_p}{2\tau}})$$
(5)

130 where τ is the gas mean free path.

Based on the discussion above, the aerosols that pass through the DMA with the same Z_p , can have different D_p and different elementary charges.

133 **2.3 SP2**

123

129

134 The SP2 is a widely used instrument that can measure the optical properties of every single particle. The measurement principle and instrumental setup of the SP2 have been discussed in detail previously 135 (Stephens et al., 2003;Schwarz et al., 2006) and will be briefly described here. When the sample 136 137 aerosol particles pass through the continuous Nd:YAG laser beam at 1064_nm with the circulating power about 1 mW/cm² in the cavity, eight sensors distributed at four directions are synchronously 138 detecting the emitted or scattered light by using avalanche photo-detector (APD) at different angles 139 (45° and 135°). For each direction, the two APDs sample the same signal with different sensitivities to 140 get a wider measurement range. The low gain channels are less sensitive to the measured signal and 141 can be used to measure the stronger signal of larger particles. In accordance, the high gain channels 142 are more sensitive to the measured signal, and can be used to measure the weaker signal of smaller 143 particles. The optical head of the SP2 is shown schematically in fig. 1(b). 144

In this study, we utilize signals from four channels of the SP2: two of them measure the scattering signals and another two measure the incandescent light between 350 nm and 800 nm. The peak height (*H*) of the incandescence signals is used to infer whether the sampled aerosol contains the black carbon
(BC). If the *H* of the incandescence signal is larger than 500, the sample aerosol contains the BC and
the scattering signals should deviate from the signals of pure scattering aerosol. Those sample aerosols
are ruled outnot considered when dealing with the aerosol scattering signals. This is achieved by just
studying the signals when the particle are recognized as pure scattering particle.

Despite that some aerosol particles are internally mixed with a small BC core, whose incandescence signal is below the detection threshold of SP2, we will demonstrate that these particles have little influence on the retrieved aerosol RRI. At the same time, there are some weakly absorbing organic components that absorb light intensity in the near infrared range, which were termed as brown carbon (BrC). These BrC components have ignorable influence on the retrieving of aerosol RRI, which will be discussed in detail in section 4.2. Thus, the imaginary part of complex refractive index is set to be zero in the following discussion.

159 **3 Methodology**

164

160 **3.1 Scattering strength** intensity measured by the SP2

From fig. 1(b), the APDs of the SP2 receive signals that were scattered by the sampled aerosols from the directions in a certain small range at 45° and 135° . Thus, the scattering intensity (*S*) measured by the APD can be expressed as:

$$S = C_0 \mathcal{C} \cdot I_0 \cdot \sigma \cdot (PF_{45^o} + PF_{135^o}) \tag{6}$$

where I_0 is the laser's intensity; σ is the scattering coefficient of the sampled aerosol, PF_{45^o} and 165 PF_{135^o} are scattering phase function at 45° and 135° respectively of the sampled aerosols; and $C_0 \in$ 166 is a constant that is determined by the distance from the aerosol to the APD and the area of the APD. 167 168 The scattering intensity of the aerosol is recorded as the H of the scattering signals by in SP2. The 169 following calibration studies show that the scattering intensity S is highly related the H measured by 170 SP2. Therefore, the SP2 can be used as a powerful tool to measure the scattering signals-_of the 171 sampled aerosols, thus determining the corresponding scattering intensity. and the influence of the BC 172 on the aerosol scattering properties can be avoided.

Based on the Mie scattering theory, the scattering coefficient σ can be calculated by integrating the square of scattering intensity function $Q(\theta, x, RRI)$ from 0° to 180°. Angle θ is defined as the angle between the light incident direction and scattering light direction. The size parameter x is defined

as $x = \frac{\pi D_p}{\lambda}$, where λ is the light incident wavelength. The scattering phase function can be directly 176 <u>derived from S(θ , x, m), too. Therefor, the</u> σ , PF_{45^o} and PF_{135^o} in equation 6 are determined by 177 the size <u>Dp</u> and RRI of the aerosol. The amount of scattering signals from the sample aerosol varies 178 179 with the aerosol the diameterDp and RRI-of the aerosol (Bohren and Huffman, 2007). The scattering 180 intensity at different aerosol diameters and RRI is calculated based on equation (6) and shown in fig. 2. The $C_0 \in$ is assumed to be 1 here. From fig. 2, we can see that the aerosol scattering intensity 181 182 increases homogeneously monotonously with the increasing aerosol RRI at a given Dp, which makes 183 it possible to retrieve the aerosol RRI with given when the Dp and the scattering intensity are known.

Bridging the scattering H values measured by the SP2 scattering channel and the scattering 184 185 intensity S defined by equation 6 is achieved by calibrating the SP2 with ammonium sulfate. The 186 instrument setup of the calibration procedure is the same as that described in section 2.12.1. The 187 diameters of the aerosols passing through the DMA are manually changed from 100 to 450_nm with a step of 10 nm. For each diameter, the scattering H value and incandescence signal of every particle are 188 analyzed. When calibrating, there is no aerosol whose incandescence signal exceeds 1000 (This value 189 190 depends on the stability of the instrument and working conditions. It can be different for different 191 instrument), which means that the SP2 works stably and the incandescence signal channel can well 192 distinguish the BC containing aerosols. With the calibration, the relationship between the measured H 193 and theoretically calculated S can be determined.

- After the calibration, the size-resolved RRI can be retrieved with known aerosol diameter selected
 by DMA and the corresponding aerosol scattering H values measured by SP2.
- The procedure of retrieving the RRI are summarized as follows: (1) measuring the scattering *H* values at a given *Dp*; (2) transferring the *H* into to *S* by the established relationship from calibration;
 (3) calculating the refractive index with the given *Dp* and *S* by using equation 6.
- 199 **3.2 Multiple Charging**

Fig. S12 gives the aerosols scattering H probability distribution under different aerosol diameters. For each diameter, the distributions of the scattering H may have more than one mode for both the high gain and low gain channels. The following discussions would give explanation about the multiple mode distributions of *H*.

For each mode, the number of recorded aerosol particles at a given *H* is fit by the log-normal

205 distribution function:

206

$$N(H) = \frac{N_0}{\sqrt{2\pi}\log(\sigma_g)} \cdot exp\left[-\frac{\log(H) - \log(H_0)}{2\log^2(\sigma_g)}\right]$$
(7)

207 Where σ_g is the geometric standard deviation; $\underline{H_0} - \underline{H_0}$ is the geometric standard mean value of H and 208 N_0 is the number concentrations for a peak mode. The geometric standard deviation is highly related 209 to the half width of the transfer function (equation 3). The $\underline{H_0}$ and the $\underline{H_0}$ is discussed below in 210 detailfurther used for discussion in the following part.

The H₀ values of corresponding to different elementary charges each mode at different diameters 211 are labeled with different markers in fig. 23. The σ_g is fitted to be a small range at 1.182 ± 0.02 for 212 different modes and different aerosol diameters. In the following discussion, we conclude that the 213 different PH_0 values in fig. 3 represent that the aerosols are charged with different number of 214 215 elementary charges. Based on the Mie scattering theory (Bohren and Huffman, 2007), the scattering 216 intensity increases with increasing Dp, which imply that the H₀ of the singly charged aerosol should 217 increase with the increment of Dp. Thus, the black square markers in fig.2.3 represent the aerosols 218 that are singly charged. At the same time, the relationships between the H_0 and Dp can be interpolated. Other colored markers represent that the aerosols have more than one charge. We calculated the 219 220 corresponding diameter (\widetilde{Dp}) of the aerosols that share the same Z_p but different charges at the given with those particles that have diameter of -Dp with one charge by the DMA (\widetilde{Dp}). Then the 221 corresponding $\widetilde{H_0}H0$ at \widetilde{Dp} are calculated. Then the relationship between $\widetilde{H_0}$ and Dp is shown in 222 223 dashed line in fig. 23(a). From fig. 23(a), the calculated H_0 shows good consistence with the measured 224 H_0 .

From the discussion above, we conclude that the SP2 <u>can only</u> detect those ammonium sulfate aerosols with the diameter larger than 160_nm. However, the ambient aerosol RRI is always lower than that of ammonium sulfate (Liu and Daum, 2008), thus the lower detecting limit of the ambient scattering aerosols should be larger than 160_nm. The measured H₀ of the SP2 scattering low gain channel signals are shown in fig. <u>3(b)</u>S2. From fig. <u>3(b)</u>S2, the same results can be deduced as those of the high gain channel signals.

Fig. 4(a) gives the relationships between the calculated scattering intensity and the SP2 aerosol scattering $\underline{H_0H}$ at different diameters. When calculating the scattering intensity, the RRI value of ammonium sulfate is set to be $1.521_{(Flores et al., 2009), and the C_0 in equation 6 is set to be unity.$ We can see that tThe aerosol scattering intensity shows good consistence with the peak height(R²=0.9992), which to some extent reflect the high accuracy of our proposed method. When regressingthe scattering intensity on the measured peak height, the value 0.36 were obtained for the slope, whichmeans that the scattering intensity can be calculated by multiplying the peak height with a factor of0.36.

Furthermore, the RRI of the scattering aerosol at a given diameter can be retrieved using the corresponding scattering *H*.

241 **3.3 Validation of the calibration**

Ammonium chloride is used to validate the method of deriving the RRI from SP2. The RRI value of ammonium chloride is $1.642_{(Lide, 2006)}$. The scattering H of the ammonium chloride under different diameters are measured and analyzed. Fig. 4(b) shows the comparison between the measured scattering high gain peak heights and the theoretical peak heights at different aerosols diameters. Results show that the measured peak heights and the calculated ones are well correlated with R²=0.9994, which means that the DMA and SP2 can be used to derived the aerosol RRI with high accuracy.

Fig. <u>S3-S2</u> gives the corresponding results of the scattering low gain channel. In fig. <u>S3S2</u>, the relationship between the aerosol scattering peak height of the low gain channel and the scattering <u>strength-intensity</u> is determined. At the same time, the comparison between the measured peak height and the calculated peak height shows good consistence too.

253 4 Results and Discussion

254 **4.1 Field Measurements**

Figure 5 shows the measured average probability distribution of the ambient size-resolved RRI and the measured mean PNSD<u>over two hours during the measurement</u>. From fig. 5, we can see that the derived RRI is 1.46 ± 0.02 and doesn't vary significantly with diameter between 199 nm and 436 nm.-

The measured aerosol PNSD during the measurement has a maximum of 26400 #/cm³ at 107 nm. The mass concentration of the BC measured by the SP2 is $6.31 \,\mu\text{g/m}^3$. Based on the measured PNSD and the measured RRI, the size distribution of the scattering coefficient is calculated based on the Mie scattering theory. The results in fig. 5 show that the measured RRI diameter range covers most of the aerosol that contributes significantly a fraction of 0.63 to the aerosol scattering properties with. The integrated scattering coefficient at 385 Mm⁻¹. Thus, the derived size-resolved RRI of this range is representative of the ambient aerosols scattering properties.

266 **4.2 Uncertainty analysis**

267 4.2.1 Uncertainties from SP2

268 The factors that influence the accuracy of retrieving RRI include the aerosols scattering H269 measured by SP₂ and the aerosol diameter selected by DMA.

270 The uncertainties of the selected diameter by DMA is are well characterized based on equation 2 271 and 3. The uncertainties from the DMA transfer function can be avoided by fitting the scattering Husing the log-normal distribution function. However, the uncertainties of the measured H from the SP2 272 273 remain unknown. The <u>half width HW ($\Delta Z_p/Z_p$)</u> of the transfer function is 0.1 times the scanning 274 diameter, which means that the geometric standard deviation of the aerosol PNSD selected by the DMA is estimated to be 1.102. At the same time, the measured geometric standard deviation of the 275 276 measured H mode by SP2 is 1.182. Thus, the additional broadening by the H distribution is 1.073, 277 which implies that the geometric standard deviation of the measured H from the SP2 is estimated to be 1.073, whose corresponding uncertainties is 6.8%. 278

279 The uncertainties of the retrieved RRI to the variations in the measured H are analyzed using the Mie scattering theory. Firstly, we calculated the theoretical scattering intensity that can be measured 280 by the SP2 for a given aerosol diameter and RRI. The scattering intensity are changed by $\pm 6.8\%$ and 281 282 the corresponding RRI can be derived using the given aerosol diameter and changed scattering intensity. Finally, the derived RRI are compared with the given aerosol RRI. The uncertainties are analyzed for 283 284 different aerosol diameter and different RRI. and tThe corresponding results are shown in fig. 6. The 285 variations in RRI increase with the increment of RRI but decrease with the increment of the Dp. For 286 most ambient aerosols, the RRI ranges from 1.4 to 1.5 and corresponds to a variation in RRI of 0.015.

Table 1 lists the retrieved ammonium chloride RRI under different diameters. The absolute difference between the retrieved RRI and theoretical values is always smaller than 0.02 regardless of the particle diameter, which means that the measured RRI is in line with the theoretical one. Thus, we conclude that the uncertainty of the retrieved RRI is within 0.02 <u>due to the uncertainties of SP2</u> 291 <u>measurement.-</u>

292 4.2.2 Uncertainties due to BC exists

293 There are some particles with a small soot core and the incandescence signal is below the detection 294 threshold of SP2. The derived aerosol RRI should be influenced by small soot core. Uncertainties 295 might be resulted when deriving the RRI for these BC-contained aerosols. With the calibration of the 296 SP2 with Aquadag soot particles, we concluded that the SP2 can't detect the soot particles lower than 297 80 nm, which is shown in detail in supplementary material in section S3.

298 We derived the aerosol equivalent refractive index when the aerosol have BC cores lower than 80 299 nm with two steps. The scattering strength of the BC-containing aerosols are first calculated based on 300 Mie scattering theory. Then the scattering strength are used to deriving the equivalent refractive index 301 with assuming that the BC-containing aerosols are pure scattering aerosols.

302 Monte Carlo simulations were applied to investigate the influence of the BC core on the retrieved ambient aerosol RRI. Firstly, aerosol with diameter between 200 nm and 500 nm was chosen. Then 303 the core diameter are random determined lower than 80 nm. The core diameters flow the log-normal 304 305 distribution with the mean core diameter of 120 nm (Raatikainen et al., 2017). When calculating the 306 scattering strength, the complex refractive index of the core 1.8+0.54*i* (Zhao et al., 2018) is used. The complex refractive of the shell adopts the measured mean values (1.46+0i) during the field 307 308 measurements. The scattering strength can be calculated with the above information. With the 309 calculated scattering strength, the equivalent real part of the refractive index (RRI) can be derived with assuming that the aerosols are pure scattering aerosols. If the core diameter is 0, then the derived 310 aerosol equivalent aerosol RRI should be 1.46. 311

312 For each aerosol diameter, the Monte Carlo simulations were conducted for 10000 times. Fig. 7(a) gives the retrieved aerosol equivalent RRI at different diameters. Results show that the retrieved 313 314 aerosol equivalent RRI are larger than 1.46 for all of the given aerosol diameters. When the aerosols 315 have BC core, the scattering strength are larger than that of pure scattering aerosols with the same 316 aerosol diameter. The derived mean equivalent RRI tend to be closer to 1.46 when the aerosol diameter 317 are larger, where the BC core contributes less and the influence of the BC core are be smaller. The 318 derived mean aerosol equivalent RRI is 1.47 and 1.462 at 200 nm and 500 nm respectively. At the 319 same time, the uncertainties associated with the equivalent RRI are larger when the aerosol diameter are smaller. We conclude that the uncertainties associated with BC core are smaller than 0.01 when 320

321	the aerosol diameter are larger than 250 nm. The maximum of the difference of the derived RRI is
322	<u>0.02.</u>
323	4.2.3 Uncertainties from BrC
324	There are some BrCs that absorb the light intensity in the near infrared range. The imaginary part
325	of the refractive index at a given wavelength $\lambda_{\lambda}(k_{\lambda})$ of the BrC can be calculated as:
326	$k_{\lambda 1} = k_{\lambda 2} \times (\frac{\lambda 2}{\lambda 1})^w \underline{\qquad} (8)$
327	Where w is defined by mass of BC to organic aerosol ratio (R) (Saleh et al., 2015) with:
328	$w = \frac{0.21}{R + 0.07} \tag{9}$
329	<u>Based on the work of Saleh et al. (2015), the k_{550} can be expressed as:</u>
330	$k_{550} = 0.016 \times \log_{10}(R) + 0.04 $ (10)
331	The values R ranges between 0.09 and 0.35 for different types of aerosols (Saleh et al., 2015).
332	Based on equation (8), (9) and (10), the k_{1024} ranges between 0.01 and 0.024. The maximum value
333	0.024 is used for further analysis.
334	The uncertainties of the retrieved RRI when ignoring the effect of BrC are analyzed. Firstly, The
335	scattering light intensity at a given diameter with a refractive index of $1.46 + 0.024i$ is calculated using
336	the Mie model. Then the corresponding RRI are retrieved using given diameter and the calculated light
337	intensity with assumption that these are pure scattering aerosols. The retrieved aerosol RRI values for
338	different aerosol diameter are shown in fig. 7(b). For the light absorbing particles, their scattering light
339	intensity is smaller than that of the pure scattering particles with the same diameter and RRI. Therefore,
340	the retrieved aerosol RRI is underestimated for most of the conditions. The differences between the
341	given RRI value (1.46) and retrieved RRI value are lower than 0.006 for all of the diameters as shown
342	in fig. 7(b). The BrC component has little influence on the retrieved aerosol RRI.
343	4.2.4 Overall of the uncertainties
344	Monte Carlo simulations were conducted to study the influence of the above three uncertainty
345	sources. Four steps are involved in the Monte Carlo simulations. First, the core diameter of an aerosol
346	particle at a given diameter are randomly given with the core diameter flowing the log-normal
347	distribution with the mean core diameter of 120 nm (Raatikainen et al., 2017). The refractive index of
348	the core is set to be the same as that in section 4.2.2. The RRI of the shell uses the measured mean
349	value 1.46. The imaginary part of the shell is determined randomly with a mean value of 0.023. Second,
the light scattering intensity can be calculated using the Mie model and the information in step one.
Then the light scattering intensity was randomly changed with uncertainties of 6.8%. Finally, the
changed light scattering intensity are used to derive the aerosol RRI with the given diameter and
assumption that the particles are pure scattering particles.

354 The aerosol diameters were changed from 200 nm to 500 nm, and the simulations were conducted 355 for 10000 times for each diameter. The overall uncertainties are shown in fig. 7(c). The uncertainties 356 from SP2 instrument measurement don't lead to bias of the retrieved aerosol RRI. When the aerosol 357 diameter is lower than 300 nm, the influence of the BC core is more important than the influence of BrC. The retrieved RRI tend to be overestimated when the aerosol is lower than 300 nm. When the 358 aerosol diameter is larger than 300 nm, the influence of BrC domains and the retrieved aerosol RRI 359 are underestimated. However, the bias caused by BC and BrC are all the way lower than 0.01. For 360 361 most of the conditions, the retrieved aerosol RRI are within the range of 1.46 ± 0.02 . Thus, we conclude that the uncertainty of the retrieved RRI is 0.02 with considering all of the factors. 362

363 5 Conclusions

Knowledge on the microphysical properties of ambient aerosol is import for better evaluating their radiative forcing. The aerosol RRI is a key factor that determines the aerosol scattering properties. In this study, a new method to measure the ambient aerosol RRI is developed by synthetically using a DMA in tandem with a SP2. This method can continuously measure the size-resolved RRI over a wide range between 198 nm and 426 nm with an accuracy of 0.02. At the same time, it is free from the influence of the BC containing aerosols.

The basic principle of measuring the size-resolved RRI is to select the aerosols at a certain diameter by the DMA and measure the corresponding scattering intensity by the SP2. The relationship between the aerosols scattering intensity and the peak height of the scattering signal channels are determined by calibrating the SP2 using ammonium sulfate (RRI=1.521).

The method is validated by <u>using measuring the size-resolved RRI of</u> the ammonium chloride with the RRI <u>value</u> of 1.642 as sample aerosol and the corresponding derived <u>size-resolved RRI isvalue is</u> 1.642 ± 0.02 ._

There are three factors that influence the accuracy of derived aerosol RRI. The measured scattering intensity by SP2 has an uncertainty of 0.68%, which can lead to the uncertainties of the derived RRI 379 values less than 0.15. There are some particles with a small soot core and the incandescence signal is 380 below the detection threshold of SP2. The light scattering intensity of these particles increases 381 compared with that of the pure scattering particles with the same aerosol diameters. The retrieved 382 aerosol RRI values can be overestimated by up to 0.02. Some BrCs absorb the light intensity in the 383 near infrared range. The corresponding scattering intensity is weaker than that of pure scatter particles 384 for the same diameter and the retrieved aerosol RRI value can be underestimated by up to 0.006. Based 385 on Monte Carlo simulations, the uncertainty of the retrieved RRI is 0.02 with considering all of the 386 factors.

This instrument is employed at a field measurement at the AERONET PKU stating, the sizeresolved RRI of the ambient aerosols is 1.46 and doesn't show significant variation among the diameter. The corresponding aerosol diameter range, which can be detected by SP2 to derive the RRI, covers most of the aerosol scattering. Thus, the derived size-resolved RRI of this range can be used as a good representative of the ambient aerosols scattering properties.

392

393 *Data availability.* The measurement data involved in this study are available upon request to the 394 authors.

395

Author contributions. Gang Zhao and Chunsheng Zhao designed the experiments; Gang Zhao and
 Weilun Zhao conducted the measurements; Chunsheng Zhao and Gang Zhao discussed the results and
 wrote the manuscript.

399

400 *Competing interests.* The authors declare that they have no conflict of interest.

401

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Figure 1. (a) Schematic of the measurement system. (b) Diagram of SP2 Chamber.



Figure 2. The distribution of the aerosols scattering strength-intensity at different Dp and different
RRI.



Figure 3. The geometric mean peak height for different diameters of the high gain channel. The
markers gives the measured values and the dotted line shows the theoretically calculated value.
Different colors represent the different number of elementary charges.



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Figure 4. (a) the relationship between the scattering peak height from the SP2 high gain scattering channel <u>when calibrating by</u> using the ammonia sulfate and (b) the comparison between the measured scattering peak height from SP2 high gain scattering channel using the ammonia chloride and the calculated scattering peak height using the Mie scattering theory. Different colors represents the results at different diameter<u>s</u>.



Figure 5. The measured probability of the size-resolved RRI (the filled color), the measured mean
PNSD (the full line) and the mean scattering size distribution (the dotted line).



Figure 6. The variation in RRI for different kinds of aerosols that have different diameters and different
RRI.



Tabel 1. The retrieved RRI and the absolute difference between the retrieved RRI and the theoretical

605 []]	RRI for	different	ammonia	chloride	diameter <u>s</u> .

Dp(nm)	160	170	180	190	200	210	220	230	240	250	260	270
RRI	1.654	1.650	1.651	1.643	1.656	1.645	1.633	1.626	1.634	1.626	1.624	1.625
Difference	0.012	0.008	0.009	0.001	0.012	0.003	0.009	0.016	0.008	0.016	0.018	0.017

1 Method to measure the size-resolved real part of aerosol refractive index using

2 differential mobility analyzer in tandem with single particle soot photometer

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7 Abstract

8 Knowledge on the refractive index of ambient aerosol can help reduce the uncertainties in 9 estimating aerosol radiative forcing. A new method is proposed to retrieve the size-resolved real part 10 of refractive index (RRI). Main principle of deriving the RRI is measuring the scattering intensity by 11 single particle soot photometer of size-selected aerosol. This method is validated by a series of 12 calibration experiments using the components of known RRI. The retrieved size-resolved RRI cover a 13 wide range from 200 nm to 450 nm with uncertainty less than 0.02. Measurements of the size resolved 14 RRI can improve the understanding of the aerosol radiative effects.

15 **1 Introduction**

Aerosols exert significant influence on the earth energy budget by scattering and absorbing 16 radiation (Ramanathan and Carmichael, 2008). There still remain great uncertainties when estimating 17 the aerosol effective radiative forcing (RF) (Ghan and Schwartz, 2007) and an accurate estimation of 18 the aerosol optical properties can help reduce the RF variations. The optical properties of the ambient 19 20 aerosol particles are determined by their particle size and complex refractive index (RI, m=n+ki) (Bohren and Huffman, 2007;Levoni et al., 1997). Despite that the ambient aerosol particle size 21 distribution can be measured with high accuracy (Wiedensohler et al., 2012), an accurate measurement 22 23 of the ambient aerosol RI remains challenging. The RI is also widely used in remote sensing (Redemann et al., 2000;Dubovik, 2002;Zhao et al., 2017) and atmospheric modelling (Ghan and 24 25 Schwartz, 2007;Kuang et al., 2015) because the aerosol single scattering albedo (SSA) and aerosol scattering phase function are highly related with the RI. At the same time, a small uncertainty in the 26 real part of the RI (RRI) can lead to great uncertainties when estimating the aerosol RF. Zarzana et al. 27 (2014) found that a variation of 0.003 in RRI can lead to uncertainties of 1% in RF for non-absorbing 28 ammonium sulfate particles. Moise et al. (2015) estimated that the RF would increase 12% when the 29

RRI varied from 1.4 to 1.5. Valenzuela et al. (2018) reported that the uncertainties in RF is estimated
to be 7% when the aerosol RRI varied by 0.1. Therefore, it is pressing that the uncertainties of the RI
be reduced when estimating the RF.

- Many methods were proposed to derive the RRI. The RRI can be estimated by linear volume
 average of the known aerosol chemical components by
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$$\mathbf{n} = \sum_{i} f_{i} n_{i} \tag{1}$$

where f_i and n_i is the volume fraction and known partial refractive index of *i*th component (Wex et 36 37 al., 2002; Hand and Kreidenweis, 2002; Hänel, 1968; Liu and Daum, 2008). The aerosol RRI can also be calculated by partial molar refraction approach (Stelson, 1990; Hu et al., 2012) which is essentially 38 consistent with the linear volume method (Liu and Daum, 2008). The ambient aerosol RRI can be 39 derived by synthetically using the radiative transfer calculations and the ground-based solar extinction 40 and scattering measurements (Wendisch and Hoyningen-Huene, 1994, 1992). Sorooshian et al. (2008) 41 developed a method to measure the aerosol RRI based on the differential mobility analyzer (DMA) 42 and an optical particle counters. The RRI could be retrieved from the known particle size from the 43 DMA and the aerosol scattering intensity from the Optical Particle Counter (OPC) for aerosol particles 44 45 larger than 500 nm. The Scanning Mobility Particle Sizer (SMPS) and OPC was used in combination to derive the RRI by aligning the particle size distributions in the instrument overlap regions (Hand 46 and Kreidenweis, 2002; Vratolis et al., 2018). The aerosol effective RRI was also retrieved by applying 47 Mie scattering theory to the aerosol particle number size distribution, aerosol bulk scattering 48 49 coefficient and aerosol absorbing coefficient data (Cai et al., 2011;Liu and Daum, 2000). Spindler et al. (2007) retrieved the aerosol RRI value by using the cavity ring-down spectroscopy to measuring 50 the scattering and absorbing properties of bulk aerosols. Eidhammer et al. (2008)) measured the light 51 scattering at different angles and retrieved the RRI. Similarly, the aerosol RRI was retrieved by 52 measuring the aerosol phase function (Barkey et al., 2007). Recently, a method by using the single 53 54 particle mass spectrometry was proposed to measure the aerosol RRI (Zhang et al., 2015). At the same time, aerosol time-of-flight mass spectrometer was proved to be capable of measuring the aerosol RRI 55 (Moffet et al., 2008). The aerosol RRI can also be retrieved from the Mie spectroscopy by using the 56 57 optical tweezers in the laboratory (Shepherd et al., 2018).

58 Many studies show that aerosol of different diameters share different properties such as shape 59 (Zhang et al., 2016;Peng et al., 2016), density (Qiao et al., 2018), aerosol hygroscopicity (Wang et al., 2017) and most importantly, the chemical components (Liu et al., 2014;Hu et al., 2012). Thus, there might be significant variations in the aerosol RRI for aerosols of different diameters because the aerosol RRI is highly related to the aerosol density (Liu and Daum, 2008) and chemical components (Stelson, 1990). On the other way round, information of the size-resolved aerosol RRI can help to study the chemical information and the aging process of aerosols among different diameters. Therefore, measurement of the size-resolved aerosol RRI is necessary.

Up to now, there are few information in the literature of the size-resolved ambient aerosol RRI 66 67 (Ebert et al., 2004;Kandler et al., 2007;Ebert et al., 2002). Traditionally, the size-resolved ambient aerosols RRI are estimated by measuring the molar fraction or volume fraction of main aerosol 68 chemical compositions. However, the influence of organic component on the aerosol RRI is ignored. 69 The organic component contributes more than 20% of the total aerosol component in the North China 70 71 Plain (Hu et al., 2012;Liu et al., 2014). At the same time, RRI of the organic aerosol changes significantly between 1.36 and 1.66 (Moise et al., 2015). Ignoring the organic component may lead to 72 significantly uncertainties when estimating the aerosol RRI. There were no technique, to our 73 knowledge, that directly measures the size-resolved aerosol optical properties and derives the size-74 75 resolved aerosol RRI.

In this study, a novel method is proposed to measure the size-resolved ambient aerosol RRI by using a DMA in tandem with a single particle soot photometer (SP2). The principle of the system is using the SP2 to measure the scattering properties of size-selected aerosols. Knowing the aerosol diameter and corresponding scattering intensity, the size-resolved aerosol RRI can be retrieved based on the Mie scattering theory. This proposed method can measure the ambient aerosol RRI over a wide size range with high accuracy. The measurement system is employed in a field campaign in the North China Plain and the corresponding results are further discussed.

The structure of this manuscript is as follows: section 2 provides the instruments setup and details of the instrument. The method to retrieve the size-resolved aerosol RRI is given in section 3. Section 4 shows the discussions about the uncertainties of the proposed method and field measurements results of the size-resolved aerosol RRI. Conclusions come at the last part.

87 2 Instrument

88 2.1 Instrument Setup

The instrument setup is schematically shown in fig. 1(a). Firstly, the dried sample aerosols are 89 guided to a X-ray soft diffusion charger and then lead to a DMA (Model 3081, TSI, USA). The quasi-90 monodisperse aerosols that pass though the DMA at a given diameter are then drawn into a SP2 to 91 measure the aerosol scattering properties with a flow ratio of 0.12 lpm and a condensation particle 92 counter (CPC, Model 3776, TSI, USA) to count the aerosol number concentration with a flow ratio of 93 0.28 lpm respectively. Thus, the sample flow (Q_a) of the DMA is 0.4 lpm. Accordingly, the sheath 94 flow (Q_{sh}) of the DMA is 4 lpm. The DMA is set to scan the aerosols diameter from 12.3 to 697 nm 95 96 over a period of 285s and repeats after a pause of 15 s. The combination of DMA, CPC and SP2 can provide information of aerosol particle number size distribution (PNSD) and size resolved RRI. 97

On 8th, June, 2018, the measurement system was employed at the filed measurement of AERONET station of BEIJING_PKU (N39°59', E116°18') to test the reliability of retrieving the ambient size-resolved RRI. This measurement site locates on the north west of the city of Beijing, China and is about 1.8 km north of the Zhongguancun, Haidian District, which is one of the busiest areas in Beijing. It is surrounded by two main streets: Zhongguancun North Street to the west and Chengfu Road to the south. This site can provide representative information of the urban roadside aerosols (Zhao et al., 2018).

105 **2.2 DMA**

When a voltage (*V*) is applied to the DMA, only a narrow size range of aerosol particles, with the same electrical mobility (Z_p) can pass through the DMA (Knutson and Whitby, 1975). The Z_p is expressed as:

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$$Z_P = \frac{Q_{sh}}{2\pi V L} ln(\frac{r_1}{r_2}) \tag{2}$$

110 where Q_{sh} is the sheath flow rate; *L* is the length of the DMA; r_1 is the outer radius of annular space 111 and r_2 is the inner radius of the annular space. The transfer function refers to the probability that a 112 particle with a certain electrical mobility can pass through the DMA. For a given *V*, the transfer 113 function is triangular-shaped, with the peaking value of 100% and a half width (HW) of

114
$$\Delta Z_p = Z_P \frac{Q_a}{Q_{sh}} \tag{3}$$

115 The aerosol Z_p , which is highly related to the aerosols diameter (D_p) and the number of elementary 116 charges on the particle (n), is defined as:

117
$$Z_p = \frac{neC(D_p)}{3\pi\mu D_p}$$
(4)

118 where *e* is the elementary charge; μ is the gas viscosity coefficient, $C(D_p)$ is the Cunningham slip 119 correction that is defined by:

$$C = 1 + \frac{2\tau}{D_p} \left(1.142 + 0.558e^{-\frac{0.999D_p}{2\tau}} \right)$$
(5)

121 where τ is the gas mean free path.

Based on the discussion above, the aerosols that pass through the DMA with the same Z_p , can have different D_p and different elementary charges.

124 **2.3 SP2**

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The SP2 is a widely used instrument that can measure the optical properties of every single particle. 125 The measurement principle and instrumental setup of the SP2 have been discussed in detail previously 126 (Stephens et al., 2003;Schwarz et al., 2006) and will be briefly described here. When the sample 127 aerosol particles pass through the continuous Nd:YAG laser beam at 1064 nm with the circulating 128 power about 1 mW/cm² in the cavity, eight sensors distributed at four directions are synchronously 129 detecting the emitted or scattered light by using avalanche photo-detector (APD) at different angles 130 (45° and 135°). For each direction, the two APDs sample the same signal with different sensitivities to 131 132 get a wider measurement range. The low gain channels are less sensitive to the measured signal and can be used to measure the stronger signal of larger particles. In accordance, the high gain channels 133 134 are more sensitive to the measured signal, and can be used to measure the weaker signal of smaller 135 particles. The optical head of the SP2 is shown schematically in fig. 1(b).

In this study, we utilize signals from four channels of the SP2: two of them measure the scattering signals and another two measure the incandescent light between 350 nm and 800 nm. The peak height (H) of the incandescence signals is used to infer whether the sampled aerosol contains the black carbon (BC). If the *H* of the incandescence signal is larger than 500, the sample aerosol contains the BC and the scattering signals should deviate from the signals of pure scattering aerosol. Those sample aerosols are not considered when dealing with the aerosol scattering signals. This is achieved by just studying the signals when the particle are recognized as pure scattering particle.

143 Despite that some aerosol particles are internally mixed with a small BC core, whose 144 incandescence signal is below the detection threshold of SP2, we will demonstrate that these particles have little influence on the retrieved aerosol RRI. At the same time, there are some weakly absorbing organic components that absorb light intensity in the near infrared range, which were termed as brown carbon (BrC). These BrC components have ignorable influence on the retrieving of aerosol RRI, which will be discussed in detail in section 4.2. Thus, the imaginary part of complex refractive index is set to be zero in the following discussion.

150 **3 Methodology**

151 **3.1 Scattering intensity measured by the SP2**

From fig. 1(b), the APDs of the SP2 receive signals that were scattered by the sampled aerosols in a certain small range at 45° and 135° . Thus, the scattering intensity (*S*) measured by the APD can be expressed as:

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$$\mathbf{S} = \mathbf{C}_0 \cdot \mathbf{I}_0 \cdot \boldsymbol{\sigma} \cdot (PF_{45^o} + PF_{135^o}) \tag{6}$$

where I_0 is the laser's intensity; σ is the scattering coefficient of the sampled aerosol, PF_{45^o} and PF_{135^o} are scattering phase function at 45° and 135° respectively of the sampled aerosols; and C₀ is a constant that is determined by the distance from the aerosol to the APD and the area of the APD. The scattering intensity of the aerosol is recorded as the *H* of the scattering signals in SP2. The following calibration studies show that the scattering intensity S is highly related the H measured by SP2. Therefore, the SP2 can be used as a powerful tool to measure the scattering signals of the sampled aerosols, thus determining the corresponding scattering intensity.

Based on the Mie scattering theory, the scattering coefficient σ can be calculated by integrating 163 the square of scattering intensity function $Q(\theta, x, RRI)$ from 0° to 180°. Angle θ is defined as the 164 angle between the light incident direction and scattering light direction. The size parameter x is defined 165 as $x = \frac{\pi D_p}{\lambda}$, where λ is the light incident wavelength. The scattering phase function can be directly 166 derived from S(θ , x, m), too. Therefor, the σ , $PF_{45^{\circ}}$ and $PF_{135^{\circ}}$ in equation 6 are determined by 167 168 the Dp and RRI of the aerosol. The amount of scattering signals from the sample aerosol varies with the aerosol Dp and RRI (Bohren and Huffman, 2007). The scattering intensity at different aerosol 169 diameters and RRI is calculated based on equation 6 and shown in fig. 2. The C_0 is assumed to be 1 170 here. From fig. 2, we can see that the aerosol scattering intensity increases monotonously with the 171 increasing aerosol RRI at a given Dp, which makes it possible to retrieve the aerosol RRI with given 172 Dp and the scattering intensity. 173

Bridging the scattering H values measured by the SP2 scattering channel and the scattering 174 intensity S defined by equation 6 is achieved by calibrating the SP2 with ammonium sulfate. The 175 instrument setup of the calibration procedure is the same as that described in section 2.1. The diameters 176 of the aerosols passing through the DMA are manually changed from 100 to 450 nm with a step of 10 177 nm. For each diameter, the scattering H value and incandescence signal of every particle are analyzed. 178 When calibrating, there is no aerosol whose incandescence signal exceeds 1000 (This value depends 179 on the stability of the instrument and working conditions. It can be different for different instrument), 180 181 which means that the SP2 works stably and the incandescence signal channel can well distinguish the BC containing aerosols. With the calibration, the relationship between the measured H and 182 theoretically calculated S can be determined. 183

The procedure of retrieving the RRI are summarized as follows: (1) measuring the scattering Hvalues at a given Dp; (2) transferring the H into to S by the established relationship from calibration; (3) calculating the refractive index with the given Dp and S by using equation 6.

187 **3.2 Multiple Charging**

Fig. S1 gives the aerosols scattering H probability distribution under different aerosol diameters. For each diameter, the distributions of the scattering H may have more than one mode for both the high gain and low gain channels. The following discussions would give explanation about the multiple mode distributions of *H*.

For each mode, the number of recorded aerosol particles at a given *H* is fit by the log-normal distribution function:

$$N(H) = \frac{N_0}{\sqrt{2\pi}\log(\sigma_g)} \cdot exp\left[-\frac{\log(H) - \log(H_0)}{2\log^2(\sigma_g)}\right]$$
(7)

195 Where σ_g is the geometric standard deviation; H_0 is the geometric mean value of H and N_0 is the 196 number concentrations for a peak mode. The geometric standard deviation is highly related to the half 197 width of the transfer function (equation 3). The H_0 is further used for discussion in the following part. 198 The H_0 values of corresponding to different elementary charges are labeled with different markers 199 in fig. 3. The σ_g is fitted to be a small range at 1.182 ± 0.02 for different modes and different aerosol 200 diameters. In the following discussion, we conclude that the different H_0 values in fig. 3 represent that 198 the aerosols are charged with different number of elementary charges. Based on the Mie scattering theory (Bohren and Huffman, 2007), the scattering intensity increases with increasing Dp, which imply that the H₀ of the singly charged aerosol should increase with the increment of Dp. Thus, the black square markers in fig. 3 represent the aerosols that are singly charged. At the same time, the relationships between the H_0 and Dp can be interpolated.

Other colored markers represent that the aerosols have more than one charge. We calculated the corresponding diameter (\widetilde{Dp}) of the aerosols that share the same Z_p but different charges with those particles that have diameter of Dp with one charge. Then the corresponding $\widetilde{H_0}$ at \widetilde{Dp} are calculated. Then the relationship between $\widetilde{H_0}$ and Dp is shown in dashed line in fig. 3(a). From fig. 3(a), the calculated H_0 shows good consistence with the measured H_0 .

From the discussion above, we conclude that the SP2 can only detect those ammonium sulfate aerosols with the diameter larger than 160 nm. However, the ambient aerosol RRI is always lower than that of ammonium sulfate (Liu and Daum, 2008), thus the lower detecting limit of the ambient scattering aerosols should be larger than 160 nm. The measured H_0 of the SP2 scattering low gain channel signals are shown in fig. 3(b). From fig. 3(b), the same results can be deduced as those of the high gain channel signals.

Fig. 4(a) gives the relationships between the calculated scattering intensity and the SP2 aerosol scattering H_0 at different diameters. When calculating the scattering intensity, the RRI value of ammonium sulfate is set to be 1.521 (Flores et al., 2009), and the C₀ in equation 6 is set to be unity. The aerosol scattering intensity shows good consistence with the peak height (R²=0.9992), which to some extent reflect the high accuracy of our proposed method. When regressing the scattering intensity on the measured peak height, the value 0.36 were obtained for the slope, which means that the scattering intensity can be calculated by multiplying the peak height with a factor of 0.36.

224 **3.3 Validation of the calibration**

Ammonium chloride is used to validate the method of deriving the RRI from SP2. The RRI value of ammonium chloride is 1.642 (Lide, 2006). The scattering H of the ammonium chloride under different diameters are measured and analyzed. Fig. 4(b) shows the comparison between the measured scattering high gain peak heights and the theoretical peak heights at different aerosols diameters. Results show that the measured peak heights and the calculated ones are well correlated with R^2 =0.9994, which means that the DMA and SP2 can be used to derived the aerosol RRI with high accuracy. Fig. S2 gives the corresponding results of the scattering low gain channel. In fig. S2, the relationship between the aerosol scattering peak height of the low gain channel and the scattering intensity is determined. At the same time, the comparison between the measured peak height and the calculated peak height shows good consistence too.**4 Results and Discussion**

236 4.1 Field Measurements

Figure 5 shows the measured average probability distribution of the ambient size-resolved RRI and the measured mean PNSD over two hours during the measurement. From fig. 5, we can see that the derived RRI is 1.46 ± 0.02 and doesn't vary significantly with diameter between 199 nm and 436 nm.

The measured aerosol PNSD during the measurement has a maximum of 26400 #/cm³ at 107 nm. The mass concentration of the BC measured by the SP2 is $6.31 \ \mu g/m^3$. Based on the measured PNSD and the measured RRI, the size distribution of the scattering coefficient is calculated based on the Mie scattering theory. The results in fig. 5 show that the measured RRI diameter range covers most of the aerosol that contributes a fraction of 0.63 to the aerosol scattering properties withThe integrated scattering coefficient at 385 Mm⁻¹. Thus, the derived size-resolved RRI of this range is representative of the ambient aerosols scattering properties.

248 **4.2 Uncertainty analysis**

249 4.2.1 Uncertainties from SP2

The factors that influence the accuracy of retrieving RRI include the aerosols scattering *H* measured by SP2 and the aerosol diameter selected by DMA.

The uncertainties of the selected diameter by DMA are well characterized based on equation 2 and 252 3. The uncertainties from the DMA transfer function can be avoided by fitting the scattering H using 253 254 the log-normal distribution function. However, the uncertainties of the measured H from the SP2 remain unknown. The half width $(\Delta Z_p/Z_p)$ of the transfer function is 0.1 times the scanning diameter, 255 which means that the geometric standard deviation of the aerosol PNSD selected by the DMA is 256 257 estimated to be 1.102. At the same time, the measured geometric standard deviation of the measured H mode by SP2 is 1.182. Thus, the additional broadening by the H distribution is 1.073, which implies 258 259 that the geometric standard deviation of the measured H from the SP2 is estimated to be 1.073, whose 260 corresponding uncertainties is 6.8%.

261 The uncertainties of the retrieved RRI to the variations in the measured H are analyzed. Firstly, we calculated the theoretical scattering intensity that can be measured by the SP2 for a given aerosol 262 diameter and RRI. The scattering intensity are changed by $\pm 6.8\%$ and the corresponding RRI can be 263 264 derived using the given aerosol diameter and changed scattering intensity. Finally, the derived RRI are 265 compared with the given aerosol RRI. The uncertainties are analyzed for different aerosol diameter and different RRI. The corresponding results are shown in fig. 6. The variations in RRI increase with 266 267 the increment of RRI but decrease with the increment of the Dp. For most ambient aerosols, the RRI 268 ranges from 1.4 to 1.5 and corresponds to a variation in RRI of 0.015.

Table 1 lists the retrieved ammonium chloride RRI under different diameters. The absolute difference between the retrieved RRI and theoretical values is always smaller than 0.02 regardless of the particle diameter, which means that the measured RRI is in line with the theoretical one. Thus, we conclude that the uncertainty of the retrieved RRI is within 0.02 due to the uncertainties of SP2 measurement.

4.2.2 Uncertainties due to BC exists

There are some particles with a small soot core and the incandescence signal is below the detection threshold of SP2. The derived aerosol RRI should be influenced by small soot core. Uncertainties might be resulted when deriving the RRI for these BC-contained aerosols. With the calibration of the SP2 with Aquadag soot particles, we concluded that the SP2 can't detect the soot particles lower than 80 nm, which is shown in detail in supplementary material in section S3.

We derived the aerosol equivalent refractive index when the aerosol have BC cores lower than 80 nm with two steps. The scattering strength of the BC-containing aerosols are first calculated based on Mie scattering theory. Then the scattering strength are used to deriving the equivalent refractive index with assuming that the BC-containing aerosols are pure scattering aerosols.

Monte Carlo simulations were applied to investigate the influence of the BC core on the retrieved ambient aerosol RRI. Firstly, aerosol with diameter between 200 nm and 500 nm was chosen. Then the core diameter are random determined lower than 80 nm. The core diameters flow the log-normal distribution with the mean core diameter of 120 nm (Raatikainen et al., 2017). When calculating the scattering strength, the complex refractive index of the core 1.8+0.54i (Zhao et al., 2018) is used. The complex refractive of the shell adopts the measured mean values (1.46+0i) during the field measurements. The scattering strength can be calculated with the above information. With the calculated scattering strength, the equivalent real part of the refractive index (RRI) can be derived with assuming that the aerosols are pure scattering aerosols. If the core diameter is 0, then the derived aerosol equivalent aerosol RRI should be 1.46.

For each aerosol diameter, the Monte Carlo simulations were conducted for 10000 times. Fig. 7(a) 294 gives the retrieved aerosol equivalent RRI at different diameters. Results show that the retrieved 295 aerosol equivalent RRI are larger than 1.46 for all of the given aerosol diameters. When the aerosols 296 297 have BC core, the scattering strength are larger than that of pure scattering aerosols with the same aerosol diameter. The derived mean equivalent RRI tend to be closer to 1.46 when the aerosol diameter 298 are larger, where the BC core contributes less and the influence of the BC core are be smaller. The 299 derived mean aerosol equivalent RRI is 1.47 and 1.462 at 200 nm and 500 nm respectively. At the 300 same time, the uncertainties associated with the equivalent RRI are larger when the aerosol diameter 301 are smaller. We conclude that the uncertainties associated with BC core are smaller than 0.01 when 302 the aerosol diameter are larger than 250 nm. The maximum of the difference of the derived RRI is 303 0.02. 304

305 4.2.3 Uncertainties from BrC

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There are some BrCs that absorb the light intensity in the near infrared range. The imaginary part of the refractive index at a given wavelength λ (k_{λ}) of the BrC can be calculated as:

(8)

$$k_{\lambda 1} = k_{\lambda 2} imes (rac{\lambda 2}{\lambda 1})^w$$

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

310
$$w = \frac{0.21}{R+0.07}$$
 (9)

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

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

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 + 0.024i is calculated using the Mie model. Then the corresponding RRI are retrieved using given diameter and the calculated light intensity with assumption that these are pure scattering aerosols. The retrieved aerosol RRI values 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). The BrC component has little influence on the retrieved aerosol RRI.

325 **4.2.4 Overall of the uncertainties**

326 Monte Carlo simulations were conducted to study the influence of the above three uncertainty sources. Four steps are involved in the Monte Carlo simulations. First, the core diameter of an aerosol 327 particle at a given diameter are randomly given with the core diameter flowing the log-normal 328 distribution with the mean core diameter of 120 nm (Raatikainen et al., 2017). The refractive index of 329 the core is set to be the same as that in section 4.2.2. The RRI of the shell uses the measured mean 330 value 1.46. The imaginary part of the shell is determined randomly with a mean value of 0.023. Second, 331 the light scattering intensity can be calculated using the Mie model and the information in step one. 332 Then the light scattering intensity was randomly changed with uncertainties of 6.8%. Finally, the 333 334 changed light scattering intensity are used to derive the aerosol RRI with the given diameter and assumption that the particles are pure scattering particles. 335

The aerosol diameters were changed from 200 nm to 500 nm, and the simulations were conducted 336 for 10000 times for each diameter. The overall uncertainties are shown in fig. 7(c). The uncertainties 337 338 from SP2 instrument measurement don't lead to bias of the retrieved aerosol RRI. When the aerosol diameter is lower than 300 nm, the influence of the BC core is more important than the influence of 339 BrC. The retrieved RRI tend to be overestimated when the aerosol is lower than 300 nm. When the 340 aerosol diameter is larger than 300 nm, the influence of BrC domains and the retrieved aerosol RRI 341 are underestimated. However, the bias caused by BC and BrC are all the way lower than 0.01. For 342 most of the conditions, the retrieved aerosol RRI are within the range of 1.46 ± 0.02 . Thus, we conclude 343 that the uncertainty of the retrieved RRI is 0.02 with considering all of the factors. 344

345 **5 Conclusions**

Knowledge on the microphysical properties of ambient aerosol is import for better evaluating their
 radiative forcing. The aerosol RRI is a key factor that determines the aerosol scattering properties. In

this study, a new method to measure the ambient aerosol RRI is developed by synthetically using a DMA in tandem with a SP2. This method can continuously measure the size-resolved RRI over a wide range between 198 nm and 426 nm. At the same time, it is free from the influence of the BC containing aerosols.

The basic principle of measuring the size-resolved RRI is to select the aerosols at a certain diameter by the DMA and measure the corresponding scattering intensity by the SP2. The relationship between the aerosols scattering intensity and the peak height of the scattering signal channels are determined by calibrating the SP2 using ammonium sulfate (RRI=1.521).

The method is validated by measuring the size-resolved RRI of the ammonium chloride with the 356 RRI value of 1.642 as sample aerosol and the corresponding derived value is 1.642 ± 0.02 . There are 357 three factors that influence the accuracy of derived aerosol RRI. The measured scattering intensity by 358 359 SP2 has an uncertainty of 0.68%, which can lead to the uncertainties of the derived RRI values less than 0.15. There are some particles with a small soot core and the incandescence signal is below the 360 detection threshold of SP2. The light scattering intensity of these particles increases compared with 361 that of the pure scattering particles with the same aerosol diameters. The retrieved aerosol RRI values 362 363 can be overestimated by up to 0.02. Some BrCs absorb the light intensity in the near infrared range. 364 The corresponding scattering intensity is weaker than that of pure scatter particles for the same diameter and the retrieved aerosol RRI value can be underestimated by up to 0.006. Based on Monte 365 Carlo simulations, the uncertainty of the retrieved RRI is 0.02 with considering all of the factors. 366

This instrument is employed at a field measurement at the AERONET PKU stating, the sizeresolved RRI of the ambient aerosols is 1.46 and doesn't show significant variation among the diameter. The corresponding aerosol diameter range, which can be detected by SP2 to derive the RRI, covers most of the aerosol scattering. Thus, the derived size-resolved RRI of this range can be used as a good representative of the ambient aerosols scattering properties.

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373 *Data availability.* The measurement data involved in this study are available upon request to the 374 authors.

- Author contributions. Gang Zhao and Chunsheng Zhao designed the experiments; Gang Zhao and
 Weilun Zhao conducted the measurements; Chunsheng Zhao and Gang Zhao discussed the results and
 wrote the manuscript.
- 379
- 380 *Competing interests.* The authors declare that they have no conflict of interest.
- 381
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Figure 1. (a) Schematic of the measurement system. (b) Diagram of SP2 Chamber.


549 Figure 2. The distribution of the aerosols scattering intensity at different Dp and different RRI.550



Figure 3. The geometric mean peak height for different diameters of the high gain channel. The
markers gives the measured values and the dotted line shows the theoretically calculated value.
Different colors represent the different number of elementary charges.



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Figure 4. (a) the relationship between the scattering peak height from the SP2 high gain scattering channel when calibrating by using the ammonia sulfate and (b) the comparison between the measured scattering peak height from SP2 high gain scattering channel using the ammonia chloride and the calculated scattering peak height using the Mie scattering theory. Different colors represents the results at different diameters.



Figure 5. The measured probability of the size-resolved RRI (the filled color), the measured mean
PNSD (the full line) and the mean scattering size distribution (the dotted line).



Figure 6. The variation in RRI for different kinds of aerosols that have different diameters and different
RRI.



Figure 7. The retrieved aerosol RRI at different aerosol diameter. The filled color represents the 5th
and 95th percentiles.

Tabel 1. The retrieved RRI and the absolute difference between the retrieved RRI and the theoretical

577 RRI for different ammonia chloride diameters.

Dp(nm)	160	170	180	190	200	210	220	230	240	250	260	270
RRI	1.654	1.650	1.651	1.643	1.656	1.645	1.633	1.626	1.634	1.626	1.624	1.625
Difference	0.012	0.008	0.009	0.001	0.012	0.003	0.009	0.016	0.008	0.016	0.018	0.017

Method to measure the size-resolved real part of aerosol refractive index using differential mobility analyzer in tandem with single particle soot photometer

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1 The aerosols scattering peak distribution for different diameter









Figure. S1. The measured scattering signal distribution at different diameter using the ammonium sulfate.2 Scattering peak height



Figure S2. (a) the relationship between the scattering peak height from the SP2 low gain scattering channel using the ammonia sulfate and (b) the comparison between the measured scattering peak height from SP2 low gain scattering channel using the ammonia chloride and the calculated scattering peak height using the Mie scattering theory. Different colors represent the results at different diameter.

3. Determine the lower threshold of the SP2 for soot particles

For the BC-contained aerosol, Aquadag soot particles with effective density of 1.8 g/cm³ is used to determine the lower limit of the BC particle diameter when the incandescence signals can be detected by SP2. The calibrating procedure is conducted as that of the ammonia sulfate in the manuscript. The diameters (Dp) of the aerosol passing through the DMA are manually changed from 60 to 400nm with a step of 20nm. The relationships between the measured incandescence signal height and the Dp are shown in fig. S3. From fig. S3, we conclude that our SP2 is not capable of measuring the Aquadag soot particles lower than 80 nm.



Figure S3. The calibrated relationship between the incandescence peak height and the BC diameter for both the incandescence high gain channel and the incandescence low gain channel.