

Response to reviewer#2

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

Comments: The paper 'Method to calculate the aerosol asymmetry factor based on measurements from the humidified nephelometer system' offers a new method for determining the ambient aerosol asymmetry factor. I believe that aerosol asymmetry factor is clearly of real importance. The proposed method has advantages over the traditional methods as it can measure the aerosol asymmetry factor in real time. I am glad to see the results of the effects of aerosol hygroscopic growth on the variation in aerosol asymmetry factor, which is rarely discussed in previous studies. Overall, the paper is clearly written and contains originality. I recommend that the paper be accepted for publication in ACP after some minor work to be done for its improvements.

Reply: Thanks for the comments.

Comments: I have the following suggestions to further improve this work: (1) The authors should highlight the novel and original aspects of the work. More discussions should be added in the text, mostly the introduction section

Reply: Thanks for the comments. We have made some revisions accordingly.

Comments: (2) To my knowledge, the aerosol asymmetry factor is highly related to the aerosol particle number size distribution, the aerosol mixing states, the ambient relative humidity (RH) and the aerosol complex refractive index. The first three parameters are discussed in this work, I suggest that the authors add some work on the sensitivities of the aerosol asymmetry factor on complex refractive index. The uncertainties due to complex refractive index should be well discussed in this paper.

Reply: Thanks for the comments. We have made some revisions accordingly. We have added some discussions at section 3.1 in the manuscript and a section is added in the supporting information to study the influence of the refractive index on g . We

discussed the influence of the real part and the imaginary part of the complex refractive index on g based on the literature studies of the complex refractive index. Results show that the g is slightly influenced by the variation in complex refractive index.

In our study, the complex refractive index of the light absorbing carbonaceous aerosol (LAC) is $1.80+0.54i$, and the corresponding values of the aerosol less absorbing aerosol components (included inorganic salts, acids, and most of the organic compounds) is $1.53+10^{-7}i$. The value of the refractive index for LAC is in accordance with that used in (Kuang et al., 2016; Ma et al., 2012).

In the following parts, the n_{non} and i_{non} refer to the real and imaginary part of the refractive index of less absorbing components and the n_{BC} and i_{BC} refer to the real and imaginary part of the refractive index of LAC.

For LAC, no accurate refractive index is valid in open literatures, and a variety of values for its refractive index have been used in different climate and aerosol optical models (Bond et al., 2013). Table R1 lists some of the values of the refractive index for the LAC.

Table R1. Refractive indices of LAC in open literatures at wavelength of 550nm.

Literature	Refractive index of LAC
(Dalzell and Sarofim, 1969)	$1.56+0.56i$
(Ouimette and Flagan, 1982)	$1.56+0.47i$
(Hasan and Dzubay, 1983)	$1.97+0.65i$
(Sloane, 1984)	$1.90+0.55i$
(Covert et al., 1990)	$1.95+0.66i$
(Hess et al., 1998)	$1.74+0.44i$
(Seinfeld et al., 1998)	$1.96+0.66i$
(Bond and Bergstrom, 2006)	$1.95+0.79i$

For less absorbing components, the refractive indices of different less absorbing aerosol species in the open literature are listed in table R2. The imaginary part of the refractive indices are very small and close to zero. In this study, the i_{non} is assumed to be 10^{-7} . The real part of the refractive indices also don't vary much and the range is

small. The filter-based chemical composition results of Liu et al. (2014) demonstrate that sulfate, nitrate and organic matter dominate the composition of the continental aerosol particles of the accumulation mode. This means that although the chemical composition of less absorbing components varies, the n_{non} will locate within a small range. The n_{non} is assumed to be 1.55 in this research. With this values of refractive index of less absorbing components, good agreement is achieved between calculated and measured scattering coefficients (see section 3 of this supplementary material). It is noteworthy that the magnitude of the imaginary parts for the refractive indices of different less absorbing components shown in table R2 range from 0 to 10^{-3} . To insure the rationality of the usage of i_{non} , we calculate the different g values under different i_{non} conditions at 532nm by using the mean value of the measured aerosol PNSD, aerosol BC mass concentration, aerosol hygroscopicity and the mean mixing state during the observation period of Gucheng. Fig. R1 gives the calculated g values at different RH and the absolute difference between the results of different i_{non} . It can be seen from this figure that even the i_{non} changes significantly, it has negligible impacts on the calculated g . The absolute difference of the g values decrease with the increment of RH because the relative differences of i_{non} of the less absorbing components are smaller at high RH.

Table R2. The refractive indices of different less absorbing species.

Literature	S1 [*]	S2 [*]	S3 [*]	S4 [*]	S5 [*]	S6 [*]	S7 [*]
(D'Almeida et al., 1991)	1.43+10 ⁻⁸ i						
(Morgan et al., 2010)	1.53+0i	1.60+0i				1.63+0.021i	
(Sloane, 1984)	1.53+0i					1.55+0i	1.53+0.0
(Cheng et al., 2008)			1.54+10 ⁻⁷ i	1.54+10 ⁻⁷ i	1.54+10 ⁻⁷ i	1.55+0.001i	1.58+0.0

*S1: Ammonium Sulfate

S2: Ammonium Nitrate

S3: Nitrate

S4: Non-Sea-Salt Sulfate

S5: Sea Salt

S6: Organic Matter

S7: Residue

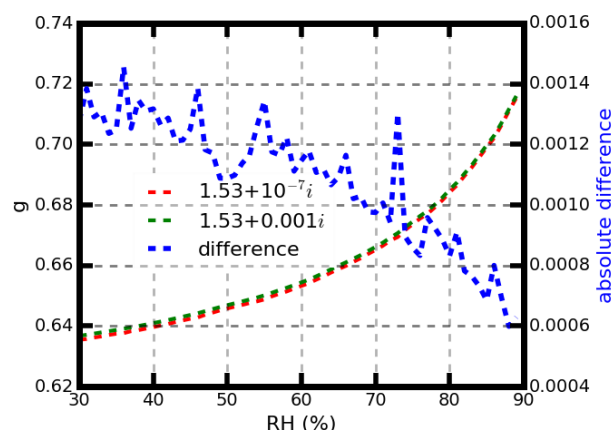


Figure R1. The calculated g under different RH using the complex refractive index of $1.53+10^{-7}i$ (red line) and $1.53+0.001i$ (green line). The blue dotted line shows the absolute difference of the g values under different RH by using the above two refractive index.

Moreover, a Monte Carlo simulation was applied to investigate the influence of the uncertainties of refractive indices of core and shell on calculation of g at different RH by using the Mie theory. The uncertainties of the input parameters for the simulation are listed in Table R3 and those uncertainties are in accordance with that of Cheng et al. (2008) and Ma et al. (2012). The other input of the Mie theory is the mean value of the measured PNSD, aerosol BC mass concentration, aerosol hygroscopicity and the mean mixing state during the observation of Gucheng. The uncertainties of g at the RH range of 30% and 90% with a step of 1% are simulated.

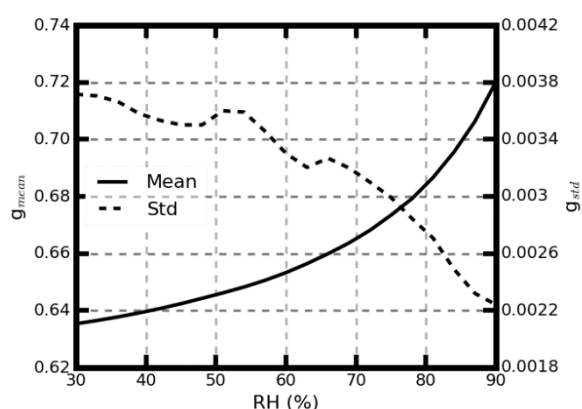


Figure R2. The calculated mean g values and the standard deviation of the g values of the Monte Carlo simulation.

Table R3. Uncertainties of the input parameters for Monte Carlo simulation, giving in

terms one . Uncertainties of the input parameters for Monte Carlo simulation, giving in terms one standard deviation (σ ,%)

Parameter	Standard deviation (σ ,%)
$n_{\text{non}}=1.55$	0.5
$i_{\text{non}}=10^{-7}$	0
$n_{\text{BC}}=1.80$	5
$i_{\text{BC}}=0.54$	6.6

For each RH, the simulation were carried out for 20000 times. The simulated results are shown in fig. R2. The results demonstrate that the uncertainties for g are small and less than 0.004. However, the variation of g resulted from aerosol population are 0.1.

By all accounts, the aerosol refractive indices of less absorbing components and LAC has little influence on the aerosol g .

Comments: (3) The method of training the machine learning model should be reconstructed. There are large uncertainties for measurements of particle number size distribution. I suggest that it would be better if the authors use all of the training data from the calculations of the Mie scattering model with measured particle number size distributions. In this way, the aerosol scattering coefficient, aerosol backscattering coefficient and the aerosol asymmetry factor under different RH can be calculated using the measurements of particle number size distributions, the mass concentration of the black carbon and aerosol hygroscopic growth factor. This can avoid the uncertainties in measurements of the aerosol particle number size distributions.

Reply: Thanks for the comments. We think that the reviewer gives a very good perspective of the work. We have made the revisions. The method of training the machine learning model is reconstructed and the results are changed accordingly.

Comments: (4) In section 3.3, parameterization of the aerosol vertical profiles of the aerosol optical properties should be discussed in detail.

Reply: Thanks for the comments. We have made the revision accordingly. Another section in the supporting information is added to detail the parameterization of the vertical profiles.

Comments: (5) Section 4.4 gives the validation of the random forest machine model, it should be placed after section 4.2.

Reply: Thanks for the comments. We have made the revision accordingly.

Comments: (6) I suggest that figure 6 re-plotted and be presented in a clearer way.

Reply: Thanks for the comments. We have made the revision accordingly. It should be mentioned that the data in the figure is changed because the training of the machine learning model is reconstructed.

Comments: (7) Line 114 : What is the meaning of UCAS? Please describe it.

Reply: Thanks for the comments. We have made the revision accordingly. We add the definition of the UCAS in section 2 and changed some label Huairou to UCAS.

Comments: (8) Line 312: 'to' should be changed to 'in'

Reply: Thanks for the comments. We have made the revision accordingly.

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