Response to reviewer#1

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

Comments: Summary: In this paper, the authors use measurements of dry and humidified aerosol properties to validate a new machine learning algorithm for determining the asymmetry parameter based on routinely measured properties. The asymmetry parameter ($g$) is important in some radiative forcing models that are based on the Henyey-Greenstein approximation of aerosol scattering phase function, a function only of $g$. The algorithm for determining $g$ using dry and humidified nephelometer measurements is validated with the CCD-LADS, a new instrument that directly measures the aerosol scattering phase function. An attempt is then made to connect these results with radiative forcing models.

Reply: We thank the anonymous reviewers’ comments and suggestions.

Comments: General comments: It would be useful to have more information about the different aerosol properties at the three different ground sites. For example, how do the PNSD and BC fraction vary between the different sites, as well as seasonally and diurnally? In particular, examples of the PNSD (dry and ambient) averaged for the different sites would be a useful figure to have since particle size is such an important factor in determining asymmetry.

Reply: Thanks for the comments. We give the characteristics of the measured mean PNSD in section 5 of the supplementary and in section 4.1.1 of the main manuscript.

Fig. S6 gives the measured distribution of the aerosol particle number size distribution (PNSD) at Gucheng and PKU. The red line gives mean results of aerosol PNSD and the red line gives the median aerosol PNSD. At the same time, the aerosol PNSD at the ambient relative humidity are calculated by using the measured aerosol PNSD, the ambient RH and the aerosol hygroscopic growth factor $\kappa$, which is derived from the datasets of the humidified nephelometer (Kuang et al., 2017). At the same time, we calculated the aerosol scattering coefficient distribution under the dry
condition by using the Mie scattering theory and the measured aerosol PNSD. There are total 8613 and 5298 different aerosol PNSDs are measured at Gucheng and PKU respectively.

From fig. S6, the peak diameter of the mean and median PNSD at Gucheng locates around 150nm. However, the peak diameter of the mean and median PNSD at PKU locates at around 100nm. At the same time, there are large partitions of small particles that are lower than 60nm at PKU. However, these particles, which are lower than 100nm, contribute little to the total aerosol scattering. The aerosol PNSD at Gucheng is more dispersed than that of the PKU, which corresponds to a larger variation in the g values. When compared with the dry aerosol PNSD, the calculated ambient aerosol PNSD at Gucheng varies more than that of the PKU, which is resulted from the high relative humidity of the ambient condition.

The size distribution of the aerosol scatter coefficient at around 500nm contributes more to the scatter coefficient at Gucheng than at that of the PKU. Thus these particles with the diameter larger than 500nm contribute less to the aerosol scattering coefficient. As g increases with the aerosol diameter, the aerosol g under dry conditions at Gucheng tends to be smaller than that at PKU.

We are willing to show the measurement results of the aerosol PNSD and mass of the BC concentrations to the reviewers. Fig.R1 and fig.R2 give the measurement results of the Gucheng and PKU respectively.

However, we are preparing the characteristics of the measurement of aerosol PNSD and BC fraction in other works. We decide not to give the detail discussion about these measurements results in the main manuscript, which is not the main scope of this work.
Fig. R1. The measurement data at the station of Gucheng. The time series in the figure represents (a) the wind speed and wind direction, (b) the temperature (in red), the relative humidity (in blue), (c) the scattering coefficient at 550 nm, (d) the BC mass concentrations and (e) the aerosol PNSD.

Fig. R2. The measurement data at the station of PKU. The time series in the figure represents (a) the wind speed and wind direction, (b) the temperature (in red), the relative humidity (in blue), (c) the scattering coefficient at 550 nm, (d) the BC mass concentrations and (e) the aerosol PNSD.
**Comments:** The weakest part of the study is the machine learning algorithm section. The use of the random forest algorithm is not well-justified, nor is there sufficient discussion of the parameterizations used.

**Reply:** Thanks for the comments. We have made some revisions according to the suggestion. Some discussions were added in the introduction in the manuscript. At the same time, section 2 in the supplementary material is added and gives some detail discussion of using the machine learning algorithm.

**Comments:** I find the label “calculated phase function” and “measured phase function”, used extensively in the discussion and throughout the figures, confusing. Please clarify whether referring to theoretical phase functions determined using the PNSD, Mie theory, and assumed refractive index, or phase functions directly measured by CCD-LADS and be consistent with how these are referred to.

**Reply:** Thanks for the comments. We have made some revisions according to the suggestion. Most of the revisions are done in section 4.3.1. At the same time, we also made some revisions about the g values by using different method. The calculated g value by using the Mie theory are labeled as $g_{\text{Mie}}$. The value predicted by using the random forest machine learning model is labeled as $g_{\text{ML}}$. The calculated value by the phase function measured from the CCD-LADS are labeled as $g_{\text{CCD}}$.

**Comments:** I think it would strengthen the discussion to include estimations of the uncertainties of the various measured properties. These can then be used for sensitivity analysis instead of arbitrary deviations.

**Reply:** Thanks for the comments. We think the reviewer gives a good view of conducting the uncertainties test. The method of estimating the uncertainties is reconstructed following the reviewer’s opinion. The uncertainties from the measured properties are analyzed and the corresponding uncertainties are estimated by using the Monte Carlo simulations. The results are mainly discussed in section 4.2.2 of the manuscript.
Comments: The authors should be careful about the input parameters of the random forest algorithm, particularly for their sensitivity analysis. The measured quantities $\sigma_{\text{sca}}$ and $\beta_{\text{sca}}$ for a specific wavelength are related to each other, and therefore I think it is important to question whether they can be treated as independent input variables.

Reply: Thanks for the comments. We agree with the reviewer’s idea and reconstructed the method of conducting the sensitivity analysis. Some discussions were added in section 2 of the supplementary material to determine the input parameters.

Comments: Please ensure that axes labels on figures legible (text size difficult to read on e.g. Figure 7)

Reply: Thanks for the comments. We have made some revisions according to the suggestion.

Comments: Specific comments: P1 L15-16: Does this range only apply to dry aerosol?

Reply: The reported g values apply to the dry aerosol. We made some revisions at the corresponding line. For the g values under different RH values, it can have a wider range as shown in the fig. 2 of the manuscript.

Comments: P1 L16-17: Specify what types of aerosol this enhancement applies to

Reply: The enhancement value 1.2 of the g value applies to the continental aerosol. We made some revisions at the corresponding line.

Comments: P1 L25: Reference desired for this information.

Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P1 L26-28: HG reference
Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P1 L27: “The HG phase function (P_{HG}(\theta)) is...”
Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P2 L34: “P(\theta) is the normalized scattering phase function”
Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P2 L37: More references would be useful here.
Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P2 L39: “few studies” – which ones?
Reply: Thanks for the comments. We have made the revision according to the comment. Pandey and Chakrabarty (2016) measured the phase function of fractal black carbon aerosols and compared the difference of the direct aerosol radiative forcing between using the measured phase function and the HG phase function. Boucher (1998) and Wiscombe and Grams (1976) simulated the difference by simulating the aerosol phase function using the Mie scatter theory. However, there is no study, to our knowledge, that uses the filed measurement of the aerosol optical properties to estimate the difference.

Comments: P2 L49: What is meant by “no available method to measure g directly” – surely a measurement of P(\theta) is a fairly direct measurement technique? (i.e. Bian, Dolgos instruments)
Reply: We agree with the editor’s suggestion and make the revision according to the comments. The corresponding text is changed as ‘There are many methods to derive
the aerosol g for the dry and ambient condition.”

Comments: P2 L55: Define backward hemisphere angles

Reply: The hemispheric backscatter ratio \( b \) is the fraction of the scattered intensity that is redirected into the backward hemisphere of the scattering particle (Marshall et al., 1995), with the definition of

\[
b = \frac{\int_{\theta_b}^{\pi} P(\theta) \sin \theta \, d\theta}{\int_0^{\pi} P(\theta) \sin \theta \, d\theta} \quad (1).
\]

The main advantage of the backscatter ratio is that it can be measured with an integrating nephelometer equipped with a backscatter shutter (Charlson et al., 1974). We also made the revision at the corresponding line.

Comments: P2 L56: It would be appropriate perhaps to also cite Horvath et al, 2016 (J. Aerosol Sci.)

Reply: Thanks for the comment. We have made the revision according to the comments.

Comments: P3 L57-63: How were these values determined – observations (if so, which instruments used?) or models?
Reply: In the previous study, almost all of the aerosol asymmetry factor values are determined by using the Mie scattering theory. We have made the revision according to the comments at the beginning of paragraph 3.

Comments: P3 L64-66: Can you discuss difficulties measuring g under ambient RH vs dry conditions?
Reply: Thanks for the comments. We give some of the difficulties of measuring g under ambient RH vs dry conditions below.

Aerosol g is derived by measuring the aerosol phase function. However, it is very difficult to measure the aerosol phase function accurately.

Figure R3. The aerosol phase functions under different RH conditions. Different colors represent the different RH.

Fig. R3 gives one example of the calculated phase function under different relative humidity by using the Mie scattering theory and the measured mean aerosol PNSD at the measurement site of PKU. Detail of calculating different phase function under different relative humidity can refer to the main manuscript and Zhao et al. (2017). From fig. R3, the aerosol phase function under dry condition can change from 0.2 to 12 at different angle. For the calculated phase function at relative humidity of 90%, the phase function can change from 0.1 to 30. For some other studies, the aerosol phase function can be as low as 0.002 (Horvath et al., 2016). As the aerosol phase function can change significantly at different angle, measurement of aerosol
phase function has very high requirement of the instrument. Up to now, there are few commercial instruments that can measure the aerosol phase function such as nephelometer aurora 4000 (Chamberlain-Ward and Sharp, 2011). However, these instruments are not widely accepted and used. Measurement of the phase function is very difficult.

At the same time, there is no method to measure the aerosol phase function covering all the angles from 0 to 180 degree. The phase function changes significantly at the range of 0~10 degree and 170~180 degree. Many of the studies derive the aerosol phase function by interpolation.

As for measuring the phase function of the ambient RH, the most important requirement is that the RH of the aerosol should not be changed when conducting the measurement. The method developed by Bian et al. (2017) can be used to measure the ambient phase function.

At the same time, the aerosol phase function change significantly with the aerosol morphology, aerosol size, aerosol complex refractive index and the incident light wavelength. Knowledge of the aerosol phase function is very limited. The calculation of the ambient aerosol phase function by using the Mie scattering theory requires more information such as the aerosol PNSD and aerosol hygroscopic growth properties.

Comments: P3 L64-66: It would be appropriate to cite Andrews et al. again here
Reply: Thanks for the comments. We made the revision according to the comments.

Comments: P3 L68: Particles do not absorb water – they can take up water, or water can condense on them
Reply: Thanks for the comments. We made the revision according to the comments.

Comments: P3 L73-76: Define Mwater
Reply: Thanks for the comments. We made the revision according to the comments.
$M_{water}$ is the molecular weight of water.

**Comments:** P3 L79-80: Should specify that Mie theory only applies to spherical particles – g can also vary by morphology

**Reply:** Thanks for the comments. We made the revision according to the comments.

**Comments:** P3 L84: Can you use subscript for f_{RH} to reduce confusion (instead of fRH)?

**Reply:** Thanks for the comments. We have changed all of the fRH to $f_{RH}$ in the manuscript.

**Comments:** P4 L87: Define back-scattering coefficient.

**Reply:** Thanks for the comments. We give the definition of the back-scattering coefficient below. However, back-scattering coefficient is widely used for those who are familiar with the instrument of the nephelometer. We decide to not add the definition of the back-scattering coefficient to the main manuscript.

The back-scattering coefficient is defined as

$$\beta_{sca} = \int_{90^{\circ}}^{180^{\circ}} F_{\lambda}(\theta)d\theta \quad (2),$$

where the angular scattering function of the particle population $F_{\lambda}(\theta)$ is defined as

$$F_{\lambda}(\theta) = \int f_{\lambda}(\theta, m_{\lambda}, d_{p}) \cdot \frac{dn(d_{p})}{dlog(d_{p})} \cdot \pi \cdot \left(\frac{d_{p}}{2}\right)^{2} \cdot dlog(d_{p}) \quad (3),$$

where $f_{\lambda}(\theta, m_{\lambda}, d_{p})$ is the angular scattering function of the individual particles with complex refractive $m_{\lambda}$ and diameter $d_{p}$ and $\frac{dn(d_{p})}{dlog(d_{p})}$ is the aerosol particle number size distribution. More details of the definition of the back-scattering coefficient can refer to Müller et al. (2011).

**Comments:** P4 L106-110: Please provide seasonal information about these field campaigns, as well as whether they covered the full diurnal cycle.

**Reply:** Thanks for the comments. We have made the revision according to the comments at paragraph 4. As shown in fig. R1, theses datasets covered the full diurnal
cycle.

**Comments:** P4 L110-L111: For people who are unfamiliar with Beijing, please describe the location in terms of either distance from the centre or relative population density – some more general metric

**Reply:** Thanks for the comments. We have made the revision according to the comments at paragraph 4 and section 1 in the supplementary material. PKU station is 11km from the center of the Megacity Beijing, which is adjacent to Hebei Province and the megacity Tianjin. In the above three cities, the industrial manufacturing has led to heavy air pollution.
Comments: P4 L110-P5 L116: Can you also describe the general wind patterns (i.e. are the sites downwind or upwind of the urban centres?)

Reply: Thanks for the comments. We have made the revision according to the comments at paragraph 4 and section 1 in the supplementary material. At the same time, the wind patterns can be recognized by fig.R1, fig.R2, and fig.S1.

Comments: P5 L118-119: Type of impactor? Is 10 µm the 50% cut-point?

Reply: Thanks for the comments. We have added the information about the impactor. 10 µm is the 50% cut-point of the ambient aerosol particles.

Comments: P5 L119: How is RH measured?

Reply: Thanks for the comments. At the Gucheng site, the RH of the sample aerosols is measured by the temperature and humidity probe, which is installed after the Nafion drying tube and the RH is all the way lower than 30%. For the RH at PKU, it is measured at the beginning of the field measurement using the same way as Gucheng, and the RH is lower than 30%. One day later, the probe was removed. We believe that the RH at PKU is all the way lower than 30% because the Nafion drying tube works well during the filed measurement and the RH at PKU is significantly lower than that of Gucheng.

Comments: P5 L121-122: Are particles assumed to be spherical for SMPS and APS retrievals of size?

Reply: The SMPS measure the aerosol PNSD by scanning the electrical mobility. The particles are assumed to be spherical when relating the aerosol electrical mobility with the aerosol electrical mobility diameter. The APS 3321 is a time-of-flight spectrometer that measures the velocity of particles in an accelerating air flow through a nozzle. It measures the aerodynamic size. The retrievals of the size of the APS do not require assumption of the particle to be spherical. However, when relating the aerodynamic size and the electrical mobility diameter, the particles are assumed to be spherical.
**Comments:** P5 L119: How low was RH? How sensitive are results to variations in RH at these low levels (presumably it is not sensitive due to low gf in this region but would be useful to say this explicitly)?

**Reply:** The RH may change when the RH and temperature of the ambient aerosol changes. However, we just need to confirm that the measured sample aerosols are lower than 30% (In some fields measurement when the ambient RH is high and the temperature is high, the RH of the sample aerosol may be acceptable to be dried to below 40%). When the RH of the sample RH is lower than 30%, the aerosol physical properties change little because the growth factor is very low. We assume that all of the measurement results measured by the instruments are at dry condition when the RH of the sample aerosol is lower than 30%, which is widely accepted when conducting the measurements.

**Comments:** P5 L119-132: Please provide error/uncertainty margins on key instrumentation (i.e. RH ±1%?)

**Reply:** Thanks for the comments. We have made the revision according to the comments. The uncertainties of the key input parameters are given in section 4.2.2 when doing the sensitivity test.

**Comments:** P5 L129: Provide laser wavelength and polarization characteristics (circular or linear)

**Reply:** Thanks for the comments. We have made the revision according to the comments. The wavelength of the laser is 532nm and a quarter-wave plate was mounted in front of the laser emitter to change the polarization state of the laser from linear to circular.

**Comments:** P5 L126: Please be consistent about f(RH) vs fRH

**Reply:** Thanks for the comments. We have made the revision according to the comments. We have changed all of the f(RH) and fRH to f_{RH}
**Comments:** P5 L127: This is a very long measurement period – can you ignore changes in aerosol population over this timescale?

**Reply:** Thanks for the comments. We agree with the editor’s opinion that the aerosol population over 45 minutes may changes and can’t be ignored. We made some quality control when retrieving the aerosol hygroscopic growth factor κ. The aerosol scattering coefficient and backscattering coefficient are recorded every minute. The scattering coefficient can catch the variation of the aerosol population. The hygroscopic growth factor κ are retrieved when the maximum and the minimum value of the measured scattering coefficient are within the ranges of 1.4 and 0.6 times of the mean measured scattering coefficient. For most of the cases, the scattering coefficients do not have large variations. Fig. R4 gives one example of the measured scattering coefficient at dry and a given relative humidity conditions. We can see that the scattering coefficient is relative stable during the period. Fig. R5 gives part of the measured time series of κ. Form fig. R5, the retrieved κ values can reflect the variation of the aerosol hygroscopicity.

At the same time, some revisions are made accordingly at paragraph 5.

![Figure R4](image)

**Figure R4.** The measured scatter coefficient time series at dry condition and the given relative conditions.
Figure R5. The time series of the measured $\kappa$ during the field measurement at PKU.

Comments: P5 L128: What is time resolution of CCD-LADS?
Reply: Thanks for the comments. We have made the revision at paragraph 5. The time resolution of the CCD-LADS during the UCAS field measurement was set to 5 minutes.

Reply: Thanks for the comments. We have made the revision accordingly at section 3.1.

Comments: P5 L127: Need more details about “self-constructed humidified nephelometer system” – what range/steps of RH? Is the population sufficiently consistent? Is 45 min for range of RH or single RH set point? How was RH altered? How was RH monitored? Is there another reference with more details about this instrument?
Reply: Thanks for the comments. Some revisions are made at paragraph 5. Some revisions are made in the manuscript accordingly. The detail information of the humidified nephelometer is described elsewhere (Kuang et al., 2017) and we give some brief description here. The humidifier is used to control the RH of the sample
aerosol and $\sigma_{sca}$ is measured for each of the controlled RH. The sample aerosol is humidified through a Gore-Tex tube, surrounded by a circulating water layer in a stainless steel tube. The RH is changed by changing the temperature of the circulating water, which is controlled by the water bath and software. For each cycle, the RH points are set to range from about 50% to about 90% over 45 minutes. For most of the cases, the aerosol PNSDs are consistent over the cycle. These cycles of $f_{RH}$ values are abandoned when the measured maximum and the minimum $\sigma_{sca}$ value are beyond the range of 1.4 and 0.6 times of the mean measured scattering coefficient of each cycle.

Two combined RH and temperature sensors (Vaisala HMP110; accuracy of 0.2 and 1.7% for RH ranges from 0 to 90%, respectively, and accuracy of 2.5% for RH ranges from 90 to 100% according to the manufacturer) are placed at the inlet and outlet of the wet Neph, and the measured RHs and temperatures are defined as RH1/T1 and RH2/T2, respectively. The dew points at the inlet and outlet of wet nephelometer were calculated using the measured RH1/T1 and RH2/T2, and the average value was considered as the dew point of the sample air. The sampled RH can be calculated through the derived dew point and the sample temperature, which is measured by the sensor inside the sample cavity of the nephelometer.

**Comments:** P6 L148: Why use volume mixing ratio vs, for example, Maxwell-Burnett or mass?

**Reply:** Thanks for the comments. The method of deriving the refractive index by using the volume mixture ratio is widely used in the previous work (Kuang et al., 2017; Wex et al., 2002; Zhao et al., 2017) and justified by Wex et al. (2002). However, we didn’t measure the complex refractive index during the field measurement and can’t decide whether the method of using the Maxwell-Burnett or mass mixture ratio is better for our study.

**Comments:** P6 L150: If using a core-shell model with BC in the centre, do you assume water only mixes with shell, or with both core and shell?

**Reply:** Thanks for the comments. In our work, we assume the BC to be
non-hygroscopic and the water mixes only with the shell. We make some revision accordingly at section 3.1.

Comments: P6 L147: How is the corresponding ambient aerosol PNSD at given RH computed?

Reply: Thanks for the comments. The ambient aerosol PNSD at given RH were computed by using the measured dry aerosol PNSD, aerosol hygroscopic factor $\kappa$, derived from the dataset of humidified nephelometer, and formula (4) in the manuscript. For each aerosol, the growth factor can be calculated by using $\kappa$ and formula (4). Then the corresponding aerosol diameter of the dry aerosol diameter can be calculated and then the ambient aerosol PNSD at given RH were computed. Fig. R6 gives one example of the calculated aerosol PNSD at different RH.

Figure R6. The changes of aerosol PNSD with RH.

We have made some revisions accordingly in the main manuscript.

Comments: P6 L157-158: Can you provide references going into more detail about random forest model algorithms?

Reply: Thanks for the comments. We made the revision according to the comments. In the manuscript, we added the reference that can be used to know more detail about random forest model algorithms such as (Breiman, 2001; Huttunen et al., 2016;
Pedregosa, 2011). However, the random forest machine learning model is a new technique that can be used for the regression. There are few studies that use the random forest machine learning model.

**Comments:** P6 L158-162: Why is the random forest model appropriate for this specific example?

**Reply:** Thanks for the comments. We have made some revision at section 3.2 and the introduction section. Random forest machine learning model is a powerful technique that can be used for classification and non-linear regression (Breiman, 2001; Hu et al., 2017; Huttunen et al., 2016). This model is a widely used nonparametric machine learning algorithm that has several strengths. First, it involves fewer assumptions regarding the dependence between observations and outcomes when compared with traditional parametric regression models. Second, strict relationships among variables are not needed before implementing the random forest model. The relationship between the aerosol optical properties measured by humidified nephelometer and g is highly non-linear. Thus, the random forest machine learning model is used in this work to study the calculation of g based on the datasets of the humidified nephelometer system.

**Comments:** P6 L164: How was the number of trees determined? What is the sensitivity of the results to npre and ntree?

**Reply:** This comment is replied together with the next comments. Please refer to the next reply of the next comments.

**Comments:** P6 L163-168: Can you provide more justification for your chosen parameterization? Does the connection between measured scatter and backscatter coefficients affect the suitability of the algorithm, or the accuracy of the results?

**Reply:** Thanks for the comments. We ignore that there are many factors that may be influenced by the number of trees. For different number of trees, the accuracy of the random forest machine learning model and the time for running the model would
change. For different numbers of $n_{\text{pre}}$, the accuracy and the time for running the model would change too.

The influence of the $n_{\text{tree}}$ on the accuracy of the model and the times for training the model is studied to determine the best $n_{\text{tree}}$ for the model. The input data of the model come from the dataset of Gucheng and the test data comes from the dataset of PKU. Different values of $n_{\text{tree}}$ are used in the model and then the time for running the model the accuracy of the model is compared. The accuracy of the model is estimated by comparing the calculated $g$ values ($g_{\text{Mie}}$) and the predicted $g$ values by the random forest machine learning model ($g_{\text{ML}}$) of PKU. The correlation coefficient ($R^2$) between $g_{\text{Mie}}$ and $g_{\text{ML}}$, the mean relative differences between $g_{\text{Mie}}$ and $g_{\text{ML}}$, and the standard deviation (Std) of the relative differences between $g_{\text{Mie}}$ and $g_{\text{ML}}$ are studied. The results are shown in fig. R7. From fig. R7, we can see that the $R^2$ increases with the increment of $n_{\text{tree}}$ from 0.934 to 0.95 when $n_{\text{tree}}$ is lower than 32 and changes slightly when $n_{\text{tree}}$ is larger than 32, which means that the random forest machine learning model can increase the accuracy of predicting $g$ with the increment of $n_{\text{tree}}$ when is lower than 32. At the same, the mean relative difference between $g_{\text{Mie}}$ and $g_{\text{ML}}$ is all the time lower than 1%, which means that the $g$ can be unbiased estimated by the random forest machine learning model. The Std fluctuates between 1% and 1.2% which is also a very small value. The time of training the model increases with the increment of $n_{\text{tree}}$ form 0.2s to 1.2s when the $n_{\text{tree}}$ increase from 2 to 100. However, the time for predicting the data is slightly changed with the increment of $n_{\text{tree}}$. Therefore, the time of running the model is not a main concerning when choosing the $n_{\text{tree}}$. With the discussion above, the $n_{\text{tree}}$ is chosen to be 32 with the accuracy and time of running the model taken into consideration.
Different input parameters can result in different behavior of the machine learning model. The number of $n_{\text{pre}}$ is also changed to test the performance of the model by changing the input parameters. From section 3.1.2 of the manuscript, it is obviously that the $\kappa$ and RH is necessary when predicting the $g$. We discussed the accuracy of the model by using different combination of the scattering coefficient and back-scattering coefficient as the input of the random forest machine learning model. There are total six group of tests were carried out. These tests contains (1) all of the three $\sigma_{\text{sca}}$ and three $\beta_{\text{sca}}$; (2) two $\sigma_{\text{sca}}$ and two $\beta_{\text{sca}}$; (3) one $\sigma_{\text{sca}}$ and two $\beta_{\text{sca}}$; (4) three $\sigma_{\text{sca}}$; (5) three $\beta_{\text{sca}}$ and (6) single parameters of the $\sigma_{\text{sca}}$ or $\beta_{\text{sca}}$. The details of the tests and the results are shown in Table. R1. These results include the $R^2$, the mean relative differences and the Std of the relative differences between $g_{\text{Mie}}$ and $g_{\text{ML}}$. If all of the three $\sigma_{\text{sca}}$ and three $\beta_{\text{sca}}$ are used as the input, the model can work
Table R1. The influence of the input parameters on the model performance.

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<td>0.568</td>
<td>1.4±2.90</td>
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<td>0.588</td>
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<td>0.624</td>
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<td>0.355</td>
<td>2.8±3.49</td>
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<td>0.323</td>
<td>2.9±3.58</td>
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<td>True</td>
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<td>0.344</td>
<td>2.8±3.52</td>
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<td>False</td>
<td>False</td>
<td>True</td>
<td>0.325</td>
<td>2.9±3.56</td>
</tr>
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</table>

*1. The mean and the standard deviation of the relative difference between the \( g_{\text{ML}} \) and \( g_{\text{mie}} \).

*2. If the value is True, the corresponding parameter is used as the input parameter of the random forest machine learning model.

well as the \( R^2 \) can reach 0.949 and the relative difference is 0.8±1.28. For those of tests (2), the best results come from the test that using the \( \sigma_{\text{sca}} \) and \( \beta_{\text{sca}} \) at 525 and 635nm with \( R^2=0.962 \) and mean value of 1.3±1.32. Despite the fact that the \( R^2 \) is
slightly higher than that of test (1), both the mean and the standard deviation of the relative difference between $g_{\text{Mie}}$ and $g_{\text{ML}}$ is larger than that of (1). However, the test that uses the $\sigma_{\text{sca}}$ and $\beta_{\text{sca}}$ at 525 only at (3) gets almost the same result. The results of the other tests are much worse than test (1). From test (4) and (5), the corresponding $R^2$ is much lower when the $\sigma_{\text{sca}}$ and $\beta_{\text{sca}}$ are not used as the input at the same time. It is concluded that the backscattering coefficient is very important for estimating the $g$.

For these tests of test (1), the third one in test (2) and the second one in test (3), it is hard to decide which group to be best one as the input parameters. However, it is concluded the test (1) is the most stable when comparing the mean $\pm$ std of the relative difference. Thus, the three $\sigma_{\text{sca}}$, three $\beta_{\text{sca}}$, $\kappa$ and RH are chosen as the input of the model and the $n_{\text{pre}}$ is set to be eight.

**Comments:** P6 L165: scattering coefficients for dry aerosol? Or humidified? If humidified, isn’t there a relationship between $\kappa$ and the scattering coefficients?

**Reply:** Thanks for the comments. The three scattering coefficients and backscattering coefficients are all for dry aerosol. We made the revision accordingly at the manuscript.

**Comments:** P L170-171: Why did you use separate data sets for training vs testing? Why not use a subset of each, or subset of one and test on the other subset?

**Reply:** Thanks for the comments. It does not matter to choose different subset for training and testing. The more amounts of effective data are used as training data, the better the model can be used to predict the result. Thus we picked some data as testing data and all of the rest data as the training data. By using the datasets as training data at Gucheng site and the datasets as testing at PKU site, we can say that the random forest machine learning model can be used to predict the $g$ values for the continental aerosols in the North China Plain.

At the same time, we conducted the random forest machine learning model by using a subset data as the test data and the other part as the training data at the field
measurement of Gucheng. The results are better than the results shown in the main manuscript. However, the aerosol population of the testing subset might be very similar to the aerosol population of the training data as both of the dataset come from the same measurement sites and thus the results using this method might be not that convincing.

**Comments:** P6 L173: Specify humidified vs dry nephelometer

**Reply:** Thanks for the comments. We have made the revision according to the comments. The scattering coefficient and backscattering coefficient are all come from the dry nephelometer.

**Comments:** P7 L189-191: What is ceiling (maximum altitude) of your SBDART model?

**Reply:** For our study, the aerosol distribution in the vertical direction is parameterized based on Liu et al. (2009). There is almost no aerosol above 50km. So the ceiling of the SBDART model in our study is 50km.

**Comments:** P7 L191-192: Assume HG phase functions or Mie phase functions in SBDART?

**Reply:** Thanks for the comments. We have made some revision accordingly to make this sentence more clearly. The profiles of the aerosol extinction coefficient, aerosol single scattering albedo and g are the results from the calculating of the Mie scattering model. The input of the Mie scattering Mie model comes from the parameterization of the aerosol vertical distributions. At the same time, we add some supporting information in section 5 of the supplementary to detail the vertical distributions of the aerosols.

**Comments:** P7 L194-195: Mean results from a specific span of time? Different times of day? Please be more specific.

**Reply:** Thanks for the comments. We have changed the manuscript accordingly.
Atmospheric meteorological parameter profiles come from the results of the intensive radiosonde observations at the Meteorological Bureau of Beijing (39°48’ N, 116°28’ E) at the local time of 13:30 from July to September in 2008. Kuang et al. (2016) studied these measured profiles and found that the vertical distributions of these parameters, which include profiles for water vapor, pressure and temperature, can be used as a good representation of the meteorological parameter profiles in the NCP during the summer. The mean results during the measurements, which is shown in fig. S4, are used in this study.

Comments: P7 L199: Please provide reference for SBDART defaults values

Reply: Thanks for the comments. We added the reference for the SBDART defaults values. At the same time, the default value can found in the manual of the SBDARF model, which can be inquired from the internet https://www.paulschou.com/tools/sbdart/.

Comments: P7 L192-199: I am a bit confused – were radiosonde data from the summer combined with albedo data from the winter? Can you explain?

Reply: Thanks for the comments. We changed the surface albedo value to the mean results of the surface albedo of Beijing from Jul to September in 2008, which is corresponded to the time and the location of the field measurements of the meteorological profiles used in this study.

At the same time, we found the results of our study are scarcely influenced because we are concerning the variation in the DARF due to the change of the aerosol asymmetry but not the absolute values of the DARF.

Comments: P8 L232: Does setting RH and $\kappa$ values give different results from removing them as input variables and setting npre=6?

Reply: Thanks for the comments. The two methods of using the input of the model result in almost the same results. Fig. R8 gives the results of using the method. Fig. R8(a) gives the results of without the RH and $\kappa$ values and fig. R8(b) gives the results
with the RH and $\kappa$ values equaling to zeros. From fig. R8, it’s concluded that using this two methods has almost no different results.

**Figure R8.** Comparison of the calculated $g_{\text{Mie}}$ values and the machine learning predicted $g_{\text{ML}}$ values (a) without the RH and $\kappa$ values and (b) with the RH and $\kappa$ values equaling to zeros.

**Comments:** P9 L238-239: Sensitivity of calculated $g$ values to input RH and $\kappa$?

**Reply:** Thanks for the comments. We just want to show the results of the comparison of the $g_{\text{ML}}$ and $g_{\text{Mie}}$ at different RH. This corresponding paragraph is reconstructed.

**Comments:** P9 L247: Should mention that other empirical relationships have been given between $g$ and $b$ (i.e. Sheridan and Ogren, 1999 (JGR); Moosmüller and Ogren, 2017 (Atmos.); Marshall et al, 1995 (Appl. Opt.)) Perhaps one of the others is more accurate in these cases?

**Reply:** Thanks for the comments. We have add some discussion about the mentioned empirical relationships between $g$ and $b$. The relationship between $g$ and $b$ used in this study is the most widely accepted parameterization scheme. Sheridan and Ogren (1999) used the relationship between $g$ and $b$ and up scatter fraction based on the work of Wiscombe and Grams (1976), which is basically, the same as the one in our discussion. Moosmüller and Ogren (2017) summarized the recent development of
relating the g and b. They give a corrected scheme of Wiscombe and Grams (1976) as:

\[ g = -6.347b^3 + 6.906b^2 - 3.859b + 0.9852 \]  \hspace{1cm} (4).

Another simplified parameterization scheme is also given in this study as:

\[ g = 1 - 2b \]  \hspace{1cm} (5).

The comparison of \( g_{\text{Mie}} \) and the parameterized g using formula (4) and (5) are shown in fig. R9. Results show that neither of the schemes works well for the continental aerosols in the NCP. However, the parameterization scheme by Marshall et al. (1995) depends on the assumption about the aerosol size distribution, which is not suitable in this study.

**Figure R9:** Comparison of the \( g_{\text{Mie}} \) and the parameterized g by using (a) formula (4) and (b) formula (5).
Comments: P9 L256-261: Why random/arbitrary deviations in input values? Can you use the specified/measured uncertainties in the measurements themselves?

Reply: Thanks for the comments. The reviewer gives a good view of doing the sensitivity test. We have made the revision accordingly. We give the uncertainties of the measured parameters, which are used as the input of the machine learning model. The Monte Carlo tests were carried out to study the sensitivity of predicted g to each input parameter. Details of the discussion are shown in section 4.2.2 of the manuscript.

Comments: P10 L269: Based on your own comments earlier in the paragraph isn’t the model simply more sensitive to completely independent input variables, which happen to be RH and κ in this example? What happens if you only input σsca, or specify that σsca and βsca must vary in the same way?

Reply: Thanks for the comments. We have changed the method of conducting the sensitivity test. The Monte Carlo tests were carried out to study the sensitivity of predicted g to the uncertainties of each input parameter. We find that κ is the most important factor that may influence the accuracy of predicting g because the κ has great uncertainties as the input parameter. More details of the discussion are shown in section 4.2.2 of the manuscript.

Comments: P10 L274-276: Can you clarify what you mean by “actual calculated aerosol phase function” – does this imply those measured directly by CCD-LADS, or those calculated using Mie theory? If the latter, can you also compare DARF using directly measured phase functions using CCD-LADS?

Reply: Thanks for the comments. We have made some revision to make the discussion more clear. The ‘actual calculated aerosol phase function’ implies the phase function calculated by using the Mie theory.

We don’t think it is proper to using the measured phase functions by CCD-LADS at this section for study. As shown by Bian et al. (2017), the measured phase function
by CCD-LADS shows good consistence with the calculated phase function, which to some extent means that the calculated phase function by using Mie theory is reliable. However, there still exist some uncertainties of the measured phase function. The phase function calculated by using the Mie theory is a better choice for testing the uncertainties of DARF in the using of HG phase function.

Comments: P10 L289: Suggest citing McComiskey et al, 2008 (JGR)

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

Comments: P10 L289-290: It is unclear how this section ties into the rest of the paper, or what novelty comes from the discussion. As you point out, previous studies have also undertaken to study how modeled DARF varies with input parameters like asymmetry parameter. Can you add anything unique to the literature regarding g and DARF?

Reply: Thanks for the comments. The revisions were made accordingly in section 4.3.2. The logic of this section is briefly described here. In this study, we use the random forest machine learning model to calculate the g. The uncertainties of the g values from the input parameter is estimated to be 1.95% when predicting g and the total variation in running the random forest machine learning model is estimated to be 4.47%. At the same time, the g can varies about 10% for different aerosol PNSD and can be enhanced by 20% with the increment of RH from 30% to 90%. It is very important to know the extent of the variation in DARF corresponding to the uncertainties from g. At the same time, it is very important to know the uncertainties of the DARF when using the random forest machine learning to predict g.

Comments: P10 L290-298: Would it be perhaps more appropriate to vary g according to a typical RH profile? You show that g tends to be higher when RH >90%, conditions which are only likely to occur in specific layers of the atmosphere depending on the local meteorology.

Reply: Thanks for the comments. We have changed the method of estimating the
influence of RH on g and DARF. It is estimated by calculating the DARFs with the g values calculated from the dry parameterized aerosol population profile and g values calculated from the RH and aerosol population profiles.

Comments:P10 L291: Where does 2.3% come from?
Reply: Thanks for the comments. We have made the revision accordingly. We attend to estimate the uncertainty of DARF form the uncertainties of the random forest machine learning model. It is worthy note that the values 2.3% is changed because the method of estimate the uncertainties of predicting g is changed.

Comments:P12 L339: Regarding “actual phase function”, please clarify (see general comments)
Reply: Thanks for the comment. We have made the revision accordingly.

Comments:Figure 1: What about Hauirou? Can you also provide graphs of average PNSDs for each site?
Reply: Thanks for the comments. We have added the information of the Huairou (UCAS) in the figure and corresponding text was changed.

Comments:Figure 3: Clarify which data used for these calculations – which site.
Reply: Thanks for the comments. We have made the revision at section 4.2.1 and the corresponding section.

Comments:Figure 4: Please explain in text a possible justification for the higher dependence on σsca,635 compared to other wavelengths. Also, why is g very dependent on 5% variation in κ, but not significantly affected by greater deviations?
Reply: Thanks for the comments. We reconstructed the method of conducting the sensitivity study. We found some of the result unacceptable in the figure. And the figure is removed from the manuscript.
Comments: Figure 5: Clarify “calculated phase function” (see general comments)
Reply: Thanks for the comments. We have made the revision accordingly.

Comments: Figure 7: Please clarify figure in caption by changing order: gCCD, Cal as a function of gMachine, cal
Reply: Thanks for the comments. We have made the revision accordingly.

Comments: Technical comments
P6 L157: Please correct “tress” to “trees”
Reply: Thanks for the comments. We have changed the manuscript accordingly.


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.
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 (inclued 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 $n_{\text{non}}$ and $i_{\text{non}}$ refer to the real and imaginary part of the refractive index of less absorbing components and the $n_{\text{BC}}$ and $i_{\text{BC}}$ 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.

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

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 $i_{\text{non}}$ 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 $n_{\text{non}}$ will locate within a small range. The $n_{\text{non}}$ 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 of 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 $i_{\text{non}}$, we calculate the different $g$ values under different $i_{\text{non}}$ 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 $i_{\text{non}}$. It can be seen from this figure that even the $i_{\text{non}}$ 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 $i_{\text{non}}$ of the less absorbing components are smaller at high RH.

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

<table>
<thead>
<tr>
<th>Literature</th>
<th>S1*</th>
<th>S2*</th>
<th>S3*</th>
<th>S4*</th>
<th>S5*</th>
<th>S6*</th>
<th>S7*</th>
</tr>
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<tbody>
<tr>
<td>(D’Almeida et al., 1991)</td>
<td>1.43+10^{-8}i</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Morgan et al., 2010)</td>
<td>1.53+0i</td>
<td>1.60+0i</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Sloane, 1984)</td>
<td>1.53+0i</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.55+0i</td>
<td>1.53+0.0</td>
</tr>
<tr>
<td>(Cheng et al., 2008)</td>
<td>1.54+10^{-7}i</td>
<td>1.54+10^{-7}i</td>
<td>1.54+10^{-7}i</td>
<td>1.55+0.001i</td>
<td></td>
<td>1.58+0.0</td>
<td></td>
</tr>
</tbody>
</table>

*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 $1.53+10^{-7}i$ (read 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 ($\sigma,\%$)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Standard deviation ($\sigma,%$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_{\text{non}}=1.55$</td>
<td>0.5</td>
</tr>
<tr>
<td>$i_{\text{non}}=10^{-7}$</td>
<td>0</td>
</tr>
<tr>
<td>$n_{\text{BC}}=1.80$</td>
<td>5</td>
</tr>
<tr>
<td>$i_{\text{BC}}=0.54$</td>
<td>6.6</td>
</tr>
</tbody>
</table>

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.


Method to calculate the aerosol asymmetry factor based on measurements from the humidified nephelometer system

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Abstract

The aerosol asymmetry factor (g) is one of the most important factors for assessing direct aerosol radiative forcing. So far, few studies have focused on the measurements and parameterization of g. The characteristics of g are studied based on field measurements over the North China Plain by using the Mie scattering theory. The results show that calculated g values for the dry aerosol can vary over a wide range (between 0.54 and 0.67). When ambient relative humidity (RH) reaches 90%, g is significantly enhanced by a factor of 1.2 due to aerosol hygroscopic growth of the continental aerosol. For the first time, a novel method to calculate g based on measurements from the humidified nephelometer system is proposed. This method can constrain the uncertainty of g within 2.56% for dry aerosol populations and 4.02% for ambient aerosols, taking into account aerosol hygroscopic growth. Sensitivity studies show that aerosol hygroscopicity is the most important factor that influences the accuracy of predicting g.

I Introduction

In addition to aerosol optical depth and aerosol single-scattering albedo, the aerosol phase function is the most important factor for assessing direct aerosol radiative forcing (DARF) (Andrews et al., 2006; Russell et al., 1997). The Henyey-Greenstein (HG) phase function (PFHG) is a widely used method to parameterize the phase function (Toublanc, 1996; Boucher, 1998; Pandey and Chakrabarty, 2016) because it uses the aerosol asymmetry factor (g) as the only free parameter. The PFHG is expressed as
\[ P_{HG}(\theta) = \frac{1-g^2}{(1+g^2-2g\cos\theta)^{3/2}} \]  

(1)

where \( \theta \) is the angle between the incident light direction and the scattered light direction. In this respect, the free parameter \( g \) can reflect the angular aerosol scattering energy distribution. \( g \) is defined as:

\[ g = \frac{1}{2} \int_0^{\pi} \cos \theta P(\theta) \sin(\theta) \, d\theta \]  

(2)

where \( P(0) \) is the normalized scattering phase function. As a result, \( g \) can be a computationally efficient factor that replaces the phase function to study aerosol radiative transfer properties (Toublanc, 1996; Hansen, 1969; Boucher, 1998). Some researchers have widely accepted the use of \( g \) as a replacement of the phase function (Hansen, 1969; Wiscombe and Grams, 1976; Sagan and Pollack, 1967; Andrews et al., 2006). However, the \( g \)-related \( P_{HG} \) may cause significant bias when estimating photo-dissociation rates (Toublanc, 1996) and aerosol radiative forcing effects (Boucher, 1998). Up to now, there have been few studies that have assessed the bias when replacing the ambient phase function with the \( g \)-related \( P_{HG} \) (Pandey and Chakrabarty, 2016; Boucher, 1998; Wiscombe and Grams, 1976) and there is no study that uses field measurements of aerosol optical properties to estimate the bias. Moreover, variations in \( g \) can influence the evolution of the atmospheric vertical structure through its effects on the atmospheric radiative distribution. Kudo et al. (2016) also found that the vertical profile of the asymmetry factor plays an important role in altering vertical variations in the solar heating rate. Marshall et al. (1995) reported that a 10% overestimation of \( g \) can systematically reduce aerosol climatic forcing by 12% or more. Andrews et al. (2006) found that a 10% reduction in \( g \) would result in a 19% overestimation of atmosphere radiative forcing at the top of atmosphere (TOA). An accurate estimation of \( g \) can help improve the assessment of the aerosol radiative effect.

There are many methods to derive the aerosol \( g \) for the dry and ambient condition. Horvath et al. (2016) measured the phase function of aerosols, calculated the \( g \) of aerosols, and found that the \( g \)-related \( P_{HG} \) can be used as a good approximation of the measured phase function. Many works used the Mic model (Bohren and Huffman, 2007) to calculate the phase function and proved its reliability (Andrews et al., 2006; Marshall et al., 1995; Shettle and Fenn, 1979; Bian et al., 2017). Comprehensive attempts have been made to relate \( g \) with the hemispheric backscatter fraction (\( b \)), where \( b \) is the ratio of light scattered into the backward hemisphere compared to total light scattered in all directions (Wiscombe and Grams, 1976; Andrews et al., 2006; Horvath et al., 2016), with the definition of...
The main advantage of the backscatter ratio is that it can be measured with an integrating nephelometer equipped with a backscatter shutter (Charlson et al., 1974).

The free parameter $g$ varies significantly for different aerosol types and different seasons. In the previous, the $g$ values are studied mainly by using the Mie scattering theory and the measured aerosol particle numbers size distribution (PNSD). D’Almeida et al. (1991) suggested that $g$ at a wavelength of 500 nm ranges from 0.64 to 0.83 depending on the aerosol type and season. Hartley and Hobbs (2001) reported a median $g$ value of 0.7 for aerosols along the east coast of the United States. Formenti et al. (2000) measured Saharan dust aerosol and found that the aerosol $g$ values ranged from 0.72-0.73. Biomass burning aerosols in Brazil had a low $g$ value of 0.54 (Ross et al., 1998).

Some works have studied the impacts of aerosol hygroscopic growth on the parameter $g$ (Hartley and Hobbs, 2001; Kuang et al., 2015; Andrews et al., 2006) and found that variations in $g$ with RH can have significant influences on aerosol radiative effects (Kuang et al., 2015; Kuang et al., 2016; Andrews et al., 2006). A parameterization scheme of $g$, that takes RH and aerosol hygroscopic growth into account, is necessary.

When exposed to the ambient atmosphere, aerosols can grow by taking up water, which causes their corresponding optical properties to considerable change. The $\kappa$-Köhler theory (Petters and Kreidenweis, 2007) is widely used to describe the hygroscopic growth of aerosol particles by using a single aerosol hygroscopic growth parameter ($\kappa$) and the $\kappa$-Köhler equation, which is shown as:

$$\frac{RH}{100} = \frac{g f^{\kappa - 1}}{g f^2 - (1 - \kappa)} \exp \left( \frac{4\alpha_\kappa M_{water}}{RT - D_0 g f^2 \rho_w} \right)$$

where $D_0$ is the dry particle diameter; $g f(RH)$ is the aerosol growth factor, which is defined as the ratio of the aerosol diameter at a given RH and the dry aerosol diameter ($D_{RH}/D_d$); $T$ is the temperature; $\alpha_\kappa$ is the surface tension of the solution; $M_{water}$ is the molecular weight of water. $R$ is the universal gas constant and $\rho_w$ is the density of water. The aerosol hygroscopic growth parameter $\kappa$ can be further used to investigate the influence of aerosol hygroscopic growth on aerosol optical properties (Tao et al., 2014; Kuang et al., 2015; Zhao et al., 2017) and aerosol liquids water contents (Bian et al., 2014).
According to the Mie theory, $g$ is associated with aerosol particle number size distribution, the particle complex refractive index, the aerosol mixing state and ambient RH. At the same time, the aerosol morphology has significant influence on $g$. Datasets from the humidified nephelometer system can partially account for all of these factors. The humidified nephelometer system consists of two nephelometers: one nephelometer measures dry aerosol scattering properties and the other measures aerosol scattering properties under well-controlled RH conditions. This results in the light scattering enhancement factor ($f_{\text{RH}}$), which is defined as $f_{\text{RH}}(\lambda, \text{RH}) = \sigma_{\text{sca}}(\lambda, \text{RH}) / \sigma_{\text{sca,dry}}(\lambda)$ or the ratio of the aerosol scattering coefficient under given RH conditions to that of dry conditions. Each nephelometer can provide a scattering coefficient ($\sigma_{\text{sca}}$) and back-scattering coefficient ($\beta_{\text{sca}}$) at three wavelengths (450, 525, 635nm). $\sigma_{\text{sca}}$ can be used to calculate the aerosol scattering Ångstrom index, which reflects the aerosol PNSD to some extent. In general, a larger value for the Ångstrom index always corresponds to a smaller predominant aerosol size. Variations in $\beta_{\text{sca}}$ and $\sigma_{\text{sca}}$ can be used to deduce the aerosol BC mixing state (Ma et al., 2012). At the same time, datasets from the humidified nephelometer system can also be used alone to measure the aerosol hygroscopicity and provide an overall hygroscopic parameter $\kappa$ (Kuang et al., 2017). All in all, measurements from the humidified nephelometer system might be used for estimating $g$ under the given RH conditions. However, there is no clear relationship between the measured datasets from the humidified nephelometer and $g$. The non-linear influence of the above listed factors on $g$ difficult to parameterize the $g$.

Random forest machine learning model is a powerful technique that can be used for classification and non-linear regression (Huttunen et al., 2016; Breiman, 2001; Hu et al., 2017). This model is a widely used nonparametric machine learning algorithm that has several strengths. First, it involves fewer assumptions regarding the dependence between observations and outcomes when compared with traditional parametric regression models. Second, strict relationships among variables are not needed before implementing the random forest model. Third, this learning model requires much less computing resource than that of the deep learning. Finally, this model has very low risk of overfitting by averaging over an ensemble of decision trees. Thus, the random forest machine learning model is used in this work to study the calculation of $g$ based on the datasets of the humidified nephelometer system.

In this study, the Mie scattering theory and field measurements over the North China Plain (NCP) are used to study the characteristics of $g$. Section 2 describes the related datasets used in this study.
Details of the study on the characteristics of g and impacts of aerosol hygroscopic growth on g are shown in section 3.1. A new method, which is based on a random forest machine learning model, is introduced to calculate g in section 3.2. We also discuss the impacts of g variations on the uncertainties of DARF in section 3.3, and the corresponding results are presented in section 4.3. Section 4.1 gives the calculated characteristics of g and section 4.2 proves the feasibility of using the machine learning model to calculate g. At the same time this method is validated by the ambient aerosol phase function measured with a charge-coupled device-laser aerosol detective system (CCD-LADS). Conclusions are in section 5.

2. Instruments and datasets

Datasets used in this study come from three field campaigns, which were conducted at three different sites in the NCP. The three field measurements are conducted at Gucheng in Hebei Province (Gucheng, 39°09’ N, 115°44’ E) from 15 October to 25 November in 2016, the AERONET BEIJING_PKU station in Beijing (PKU, 39°59’ N, 116°18’ E) from 21 March to 10 April in 2017, and the Yanqi Campus of the University of Chinese Academy of Sciences (UCAS, 40°24’ N, 116°40’ E) in the Huairou district, Beijing from 3 January to 27 January in 2016. Details of these locations are shown in Fig. S1. The PKU station is located at the northwest of Beijing, between the 4th and 5th ring road. Datasets for this location are representative of urban aerosols in the NCP. Gucheng is located between two megacities (120 km from Beijing and 190 km from Shijiazhuang) of NCP and the pollution conditions of Gucheng can be a good representation of the continental background in the NCP. Details for the Gucheng station can be found at Kuang et al. (2017). The UCAS station is 60 km away from the center of Beijing and is at the edge of the NCP, which makes it suitable for measuring the regional pollution properties of the NCP (Ma et al., 2016). More details of the measurement sites can refer to section 1 of the supplementary materials.

Table 1 lists the information for the field campaigns and the datasets used in this study. During the campaigns, sampled aerosols that had an aerodynamic diameter of less than 10 µm are selected by an impactor (Mesa Labs, Model SSI2.5) at the inlet. These aerosols are then dried to below 30% RH with a Nafion drying tube and then lead to each instrument. Aerosol PNSDs ranging from 3 nm to 10 µm are measured by using the scanning mobility particle size spectrometer (SMPS, TSI Inc., model 3936) and an aerodynamic particle sizer (APS, TSI Inc., model 3321) with a temporal resolution of 5 min. Black carbon (BC) mass concentrations are measured by a multi-angle absorption photometer (MAAP
model 5012, Thermo, Inc., Waltham, MA USA) at UCAS and by an Aethalometer 33 (Hansen et al., 1984; Drinovec et al., 2015) at PKU and Gucheng. The aerosol $\sigma_{\text{a}}$ at wavelengths of 450 nm, 525 nm and 635 nm is measured by an Aurora 3000 nephelometer and the corresponding values are recorded every minute (Müller et al., 2011).

The $\sigma_{\text{a}}$ is measured by a self-constructed humidified nephelometer system. The detail information of the humidified nephelometer is described elsewhere (Kuang et al., 2017). Some brief descriptions about the humidified nephelometer is introduced here. The humidifier is used to control the RH of the sample aerosol and $\sigma_{\text{a}}$ is measured for each of the controlled RH. The sample aerosol is humidified through a Gore-Tex tube, which is surrounded by a circulating water layer in a stainless steel tube. The RH is changed by changing the temperature of the circulating water, which is controlled by the water bath and software. For each cycle, the RH points are set to range from about 50% to about 90% over 45 minutes. For most of the cases, the aerosol PNSDs are consistent over the cycle. These cycles of $\sigma_{\text{a}}$ values are abandoned when the measured maximum and the minimum $\sigma_{\text{a}}$ value are beyond the range of 1.4 and 0.6 times of the mean measured scattering coefficient of each cycle.

Ambient aerosol phase function at a time resolution of 5 minutes is measured at UCAS by using a CCD-LADS. This system consists of a continuous laser, two charge-coupled device cameras and the corresponding fish eye lenses. The wavelength of the laser is 532 nm and a quarter-wave plate was mounted in front of the laser emitter to change the polarization state of the laser from linear to circular. The CCD-LADS can measure the ambient aerosol phase function at a wide angular range of 10-170° with a high resolution of 0.1°. More details of the measurement system can be found at (Bian et al., 2017).

3. Methodology

3.1 Calculating characteristics of $g$ based on the Mie scattering theory $g_{\text{Mie}}$

The Mie model (Bohren and Huffman, 2007) is employed to calculate the characteristics of $g_{\text{Mie}}$. When running the Mie model, aerosol PNSD, aerosol complex refractive index, BC mixing state and BC mass concentration are essential. Its results include aerosol phase function, and $g_{\text{Mie}}$ can be calculated by the definition shown in formula 2.

Mixing states of the BC come from the measurements of the field measurements. From the work of Ma et al. (2012), the mixing states of BC in the NCP were presented as both core-shell mixed and
externally mixed. Ma et al. (2012) provides the ratio of BC mass concentrations under an externally mixed state, \( M_{\text{ext, BC}} \), to total BC mass concentration, \( M_{\text{BC}} \), as follows:

\[ r_{\text{ext, BC}} = \frac{M_{\text{ext, BC}}}{M_{\text{BC}}} \quad (5) \]

The mean value of \( r_{\text{ext, BC}} < 0.51 \) (Ma et al., 2012) is used as a representation of the mixing state in this study. The size-resolved distribution of BC mass concentration is the same as that used by Ma et al. (2012). The \( \kappa \)-Köhler theory and the Mie scattering model are employed to calculate \( g_{\text{ext}} \) under different RH conditions. When the aerosol gets hygroscopic growth, the BC is treated as non-hygroscopic and the water are assumed to mix only with the shell. The real time \( \kappa \), which is derived from the measurement of \( f_{\text{dry}} \), is used to account for aerosol hygroscopic growth. For each RH value, the growth factor can be calculated based on formula 3. The corresponding ambient aerosol PNSD at a given RH can be determined too by applying the \( \kappa \) and formula (4). The refractive index \( (\bar{m}) \), which accounts for water content in the particle, is derived as a volume mixture between the dry aerosol and water (Wex et al., 2002a):

\[ \bar{m} = f_{\text{dry}} \bar{m}_{\text{aero, dry}} + (1 - f_{\text{dry}}) \bar{m}_{\text{water}} \quad (6) \]

where \( f_{\text{dry}} \) is the ratio of the dry aerosol volume to the total aerosol volume under a given RH condition; \( \bar{m}_{\text{aero, dry}} \) is the refractive index for dry ambient aerosols and \( \bar{m}_{\text{water}} \) is the refractive index of water.

The refractive indices of BC, non-light-absorbing aerosols and water, which are used in this study, are \( 1.8+0.54i \) (Kuang et al., 2015), \( 1.53+10^{-7}i \) (Wex et al., 2002b) and \( 1.33+10^{-7}i \), respectively. Then, the corresponding \( g \) values under the given RH and PNSD can also be calculated. More details on using the Mie model to calculate the aerosol phase function for different RH conditions can be found in Zhao et al. (2017).

3.2 Calculating \( g \) by using the random forest machine learning model (FRML)

In this study, the random forest machine learning model from the Scikit-Learn machine learning library (Hu et al., 2017; Pedregosa, 2011) was used to calculate \( g \). The random forest model has two parameters: the number of input variables (\( n_{\text{sec}} \)) and the number of trees grown (\( n_{\text{tree}} \)). In this study, the \( n_{\text{sec}} \) and \( n_{\text{tree}} \) are determined by minimize the relative difference of the \( g_{\text{ML}} \) and \( g_{\text{Mie}} \). Details of choosing the values of \( n_{\text{sec}} \) and \( n_{\text{tree}} \) are shown in section 5 of the supplementary. The \( n_{\text{sec}} \) and \( n_{\text{tree}} \) are set as eight and ten, respectively.
The eight input parameters include the three dry scattering coefficients, three dry backscattering coefficients, RH and κ.

The measured datasets are divided into two parts: one for the training data of the random forest model, and the other for test data. All training datasets come from field measurements at Gucheng station, whereas the datasets from PKU are employed to test the accuracy of the model. With split datasets from different sites, the feasibility of the random forest model in the NCP can be guaranteed.

Before calculating $g_{Mie}$, we compare the measured $\sigma_{act}$ from the dry nephelometer and calculate $\sigma_{act}$ from the Mie scattering model. These data, where the relative difference between the measured and calculated $\sigma_{act}$ is within 30%, are used for the following analyses. With this, the inaccuracy from the measurement of the instruments can be avoided to some extent. More details regarding the used data are shown in section 3 of the supplementary material.

To avoid the uncertainties of the measurements when training the random forest machine learning model, both the required input parameters and the predictor $g_{Mie}$ values, come from the calculation of the Mie scattering model using the measurement of the aerosol PNSD and BC from the field campaign of Gucheng. For each measured PSND and BC, the corresponding $\sigma_{act}$ and $\beta_{act}$ under dry condition at the wavelength of 450nm, 525nm and 635nm are modeled based on the Mie theory. With the concurrently measured $\kappa$ values from the humidified nephelometer, the $g$ values under different RH can be determined too. Then the modeled $\sigma_{act}$, $\beta_{act}$ under dry condition, the $\kappa$ values and the RH are used as the input data for the model and the corresponding $g_{Mie}$ values are used as the predict data.

3.3 Aerosol DARF estimations

The earth-atmosphere systems can be significantly influenced by aerosols, which scatter and absorb the energy. In this study, the Santa Barbara DISORT (discrete ordinates radiative transfer) Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998) is employed to estimate the DARF. The characteristics of DARF with the variations in $g$ are studied.

The instantaneous DARF is calculated at the TOA for cloud-free conditions. DARF is defined as the difference between radiative flux at the TOA under present aerosol conditions and aerosol-free conditions:

$$DARF = (f_a \downarrow - f_a \uparrow) - (f_m \downarrow - f_m \uparrow)$$

where $(f_a \downarrow - f_a \uparrow)$ is the downward radiative irradiance flux with given aerosol distributions and $(f_m \downarrow - f_m \uparrow)$ is the radiative irradiance flux under aerosol free conditions. The DARF at 50km is
calculated because almost all of the aerosols are located at the range of 0 and 50 km in the parameterization scheme of aerosol vertical distribution (Liu et al., 2009). The wavelengths in the range from 0.25 to 4 μm are calculated for irradiance in this study.

Input data for the SBDART are listed below. Vertical profiles of the aerosol optical properties, which include the aerosol extinction coefficient (σₐₑ), aerosol single scattering albedo (SSA) and g with a height resolution of 50 m, come from results of the Mie scattering, and the parameterized aerosol vertical distributions. Methods for parameterization and calculation of the aerosol optical profiles can be found in section 4 of supplementary material or relate to Kuang et al. (2016) and Zhao et al. (2017). Atmospheric meteorological parameter profiles come from the results of the intensive radiosonde observations at the Meteorological Bureau of Beijing (39°48′ N, 116°28′ E) at the local time of 13:30 from July to September in 2008. Kuang et al. (2016) studied these measured profiles and found that the vertical distributions of these parameters, which include profiles for water vapor, pressure and temperature, can be used as a good representation of the meteorological parameter profiles in the NCP during the summer. The corresponding measured mean results during field measurement are used in this study and the details of these profiles are shown in section 4 of the supplementary material. Surface albedo values are obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) V005 Climate Modeling Grid (CMG) Albedo Product (MCD43C3). The mean results of the surface albedo of Beijing from Jul to September in 2008 are used. The remaining input data for the SBDART are set to their default values (Ricchiazzi et al., 1998).

4 Results and Discussion

4.1 Characteristics of \( g_{\text{Mie}} \)

4.1.1 Characteristics of \( g_{\text{Mie}} \) at different sites

Fig. 1 gives the statistical results for the calculated g properties at Gucheng, PKU and UCAS. The RH at the three sites shows almost the same diurnal variation pattern in Fig. 1 (a) (b) and (c). The RH reaches a peak in the morning at approximately 6:00 am, and then reaches its lowest value at approximately 16:00 in the afternoon. However, the mean values of RH are 77.7%±20.9% at Gucheng, 47.8%±20.8% at PKU and 33.49±15.22% at UCAS. The \( g_{\text{Mie}} \) values under dry conditions that are calculated by the measured PNSD have almost no diurnal patterns. The \( g_{\text{Mie}} \) values at PKU (0.614±0.025) are slightly lower than those at Gucheng (0.601±0.021) and UCAS (0.595±0.023) as shown in Fig. 1 (d), (e) and (f). The difference in \( g_{\text{Mie}} \) values results from different aerosol properties.
at these sites. From fig. S6, the peak diameter of the mean and median PNSD at Gucheng locates around 150 nm. However, the peak diameter of the mean and median PNSD at PKU locates at around 100 nm. The peak values of the mean and median diameter of the aerosol PNSD at UCAS locates at around 60 nm. At the same time, there are large partitions of small particles that are lower than 60 nm at PKU and UCAS. However, these particles, which are lower than 100 nm, contribute little to the total aerosol scattering. The aerosol PNSD at PKU is more dispersed than that of the Gucheng and UCAS, which corresponds to a larger variation in the g values. From fig. S6 (b), (i) and (j), the size distribution of the aerosol scatter coefficient at around 500 nm contributes less to the scatter coefficient at PKU than at that of the Gucheng and UCAS. Thus these particles with the diameter larger than 500 nm contribute more to the aerosol scattering coefficient. As g_{Mie} increase with the aerosol diameter, the aerosol g_{Mie} under dry conditions at PKU tends to be larger than that at Gucheng and UCAS.

However, ambient g_{Mie} values have different patterns at different sites, as shown in Fig. 1 (j), (i) and (l). The g_{Mie} values have an RH-related diurnal pattern at Gucheng, with a mean value of 0.66±0.073, but show no diurnal variation at PKU and UCAS, where the mean values of g_{Mie} are 0.63±0.049 and 0.618±0.033, respectively. The variations of ambient g_{Mie} values are mainly resulted from the variation of the aerosol hygroscopic growth under the ambient condition, which is highly related to the ambient RH. The g_{Mie} value is significantly influenced by RH when the RH is higher than 80%, which will be detailed in section 4.1.2. Ambient g_{Mie} values at Gucheng, PKU and UCAS can vary from 0.57 to 0.8, 0.55 to 0.76 and 0.56 to 0.72 respectively, comparable to those of Andrews et al. (2006), which range from 0.59 to 0.72.

4.1.2 Influence of RH on g

To assess the influence of RH on g, the g_{Mie} values are calculated under different RH conditions for each aerosol PNSD. The statistical results of g_{Mie} versus RH are shown in Fig. 2. The g_{Mie} value has a mean of 0.61 at dry conditions and can vary widely (0.54 to 0.67), which corresponds to approximately 10% of the variation. However, the mean g_{Mie} value can vary from 0.65 to 0.8 when the RH reaches 90%. The g_{Mie} enhancement factor, which is defined as the ratio of g_{Mie} at a given RH and g_{Mie} under dry conditions, can reach a mean value of 1.2 at an RH of 90%, which means that the g_{Mie} value under wet conditions is approximately 20% higher than that under the dry conditions.
This finding is consistent with that of Hartley and Hobbs (2001), who found that $g$ is highly related to the RH. However, the aerosol complex refractive index has little influence on $g$ and the uncertainties for $g$ are less than 0.004 based on the Monte Carlo simulation of the $g$ at different complex refractive index. More details of discussing the influence of aerosol complex refractive index on $g$ can relate to section 6 of the supplementary materials.

4.2 Calculating $g_{ML}$ by using the machine learning model

4.2.1 Feasibility of using the random forest model

We establish two independent random forest machine learning models to predict $g_{ML}$ values under dry conditions and under ambient RH conditions separately.

When running the random forest machine learning model for $g$ values under dry conditions, $\sigma_{sc}$ and $b_{sc}$ at three different wavelengths are used as the input for independent variables. The other two input parameters, RH and $\kappa$, are set to zero. The predictor $g$ values come from the results of the Mie scattering model. Fig. 3(a) shows the calculated $g_{ML}$ values and predicted $g_{ML}$ values by the random forest machine learning model under dry conditions at the site of PKU. The results show that the $g_{ML}$ values and $g_{ML}$ values show good consistency with an $R^2$ value of 0.92. There are 95% of the cases that the relative difference between $g_{ML}$ and $g_{ML}$ are within the relative differences of 2.56%.

Fig. 3(b) shows the comparison of the predicted $g_{ML}$ values under different RH conditions and $g_{ML}$ values calculated by the Mie scattering model. The correlation coefficient between $g_{ML}$ and $g_{ML}$ reaches 0.93 and 95% of the relative differences within 4.02%. The random forest model can be a good method to predict $g$ values under different RH conditions with high accuracy and the uncertainties of predicting $g$ values using the random forest machine learning model is estimated to be 4.02%.

The filled colors of the dots in Fig. 3 represent the concurrently measured $\sigma_{sc}$. It is shown that with an increase in $\sigma_{sc}$, $g$ values tend to be larger, which is in accordance with the particle scattering properties. When a particle has larger diameters, the $\sigma_{sc}$ of the particle is higher, and there tends to be a larger partition of forward scattering light.

Wiscombe and Grams (1976) studied the relationship between $b$ and $g$ and gave the expression between them as follows:

$$g = -7.143889 \cdot b^3 + 7.464439 \cdot b^2 - 3.96356 \cdot b + 0.9893 \quad (8).$$
This equation is widely used to calculate $g$ from $b$ (Andrews et al., 2006; Horvath et al., 2016; Kassianov et al., 2007). We use the field measurement results to test its reliability. The comparison results between calculated $g$ values from the Mie scattering model and parameterized $g$ values from equation $\beta$ are shown in Fig. S3. From Fig. S3, we can see that the parameterized $g$ values are prevalently larger than the calculated $g$ values by approximately 10%. When the $\sigma_{\text{abs}}$ is smaller, the deviations become larger. Some other empirical relationships between $b$ and $g$ (Moosmüller and Ogren, 2017) are also tested. These parameterization schemes have almost the same result as Wiscombe and Grams (1976). This result means that the previously established parameterization scheme is not applicable in the NCP.

4.2.2 Sensitivity of the random forest model

Sensitivity studies are carried out to assess the influence of each input variable on $g_{\text{MIE}}$. Based on the works of Müller et al. (2011), the uncertainties in total scattering are 4% (450nm), 2% (525nm), and 5% (635nm) for experiments with ambient air and laboratory generated white particles. For backscattering, the differences are higher and amount 7% (450nm), 3% (525nm) and 11% (635nm). The uncertainties of the measured RH by the RH sensors is 1.7% for RH ranges from 0 to 90% (Kuang et al., 2017) and the uncertainties of the derived $\kappa$ values is 6% (Kuang et al., 2017). The Monte Carlo simulations are conducted to study the sensitivities of the $g_{\text{MIE}}$ to the input parameters in three steps. First, the mean results of the measured dry $\sigma_{\text{abs}}$, dry $\beta_{\text{MIE}}$, RH and $\kappa$ values are used to predict the $g$ value. Second, the dry $\sigma_{\text{abs}}$ at 450nm are randomly changed with a mean value of 0 and standard deviation of 4% and the other input are kept unchanged as the input. The corresponding standard deviation of the predicted $g$ value is used as the sensitivities of the predicted $g$ values to the $\sigma_{\text{abs}}$ at 450nm. At last, the sensitivities are carried out accordingly for each of the input parameter. With this, the uncertainties of the $g_{\text{MIE}}$ values to the input parameters are estimated. The total uncertainties of predicting $g$ RH are derived when all of the input parameters are randomly changed with their corresponding uncertainties. For each test, the Monte Carlo simulations are carried out for 20000 times.

Table 2 gives the two time of the standard deviation of the $g_{\text{MIE}}$ values corresponding to the uncertainties of the input parameters. Form Table 2, it is shown that the uncertainties of measured $\sigma_{\text{abs}}$ has little influence of the $g_{\text{MIE}}$ with 0.487%, 0.492% and 0.486% for wavelength of 450nm, 525nm and 635nm respectively. However, the measurement of the three $\beta_{\text{MIE}}$ have larger uncertainties and lead to greater influence on predicting $g_{\text{MIE}}$ with 0.651%, 0.486% and 0.710%. The uncertainty of the RH has...
little influence on predicting $g_{ML}$ with 0.487%. However, the uncertainty of derived $\kappa$ values (6%) influence the $g$ values most with 1.92%. The total uncertainties of predicting $g$ due to the uncertainties of the measurement is 1.95%. All in all, the total uncertainties of predicting the $g_{ML}$ is estimated to be 4.47% considering the 4.02% uncertainties of the random forest machine learning model from section 4.2.

### 4.2.3 Validation of the random forest machine learning model

Datasets of the UCAS campaign are also used to validate the random forest machine learning model. On one hand, the $g_{ML}$ values are calculated by using the random forest machine learning model with the measurements of the humidified nephelometer. On the other hand, ambient $g$ values are calculated by using the measured phase function from the CCD-LADS $g_{CCD}$ according to the definition shown in formula 2. Then the $g$ values calculated with the two methods are compared.

Comparison results of these two kinds of $g$ values are shown in fig. 4. Form fig. 4, the values of $g_{ML}$ and $g_{CCD}$ show good consistence. There are 95% of the conditions that the relative differences between the $g_{ML}$ and $g_{CCD}$ are in the range of 6.5% which is a little higher than the relative difference of the $g$ values (4.02%) between machine learning method and the Mie scattering method. During the period, the $\sigma_{\text{re}}$ range from 30 to 260 $\text{Mm}^{-1}$ which lead to cleaner conditions in UCAS than in Gucheng and PKU. Correspondingly, most of the $g_{ML}$ values are small and locate at the range of 0.54 to 0.62 which are obviously lower than those in other campaigns. At the same time, the surrounding condition at UCAS during the winter is relative dry, which results to small $g$ values. These conditions may partially explain the relatively higher difference between the $g_{ML}$ and $g_{CCD}$. With this validation, we conclude that the random forest machine learning model can give a reasonable $g$ value based on the measurements of the humidified nephelometer system.

### 4.3 Estimating the impacts of $g$ on DARF

#### 4.3.1 Uncertainties of replacing the calculated phase function with the $PF_{HG}$

When the $PF_{HG}$ is used to parameterize the calculated phase function by using the Mie theory ($PF_{ML}$), there are some deviations and the influence of these deviations should be estimated. The relative difference between the DARF from the $PF_{ML}$ and from the $PF_{HG}$ is used to estimate uncertainties when using the $PF_{HG}$. First, the $PF_{HG}$ profiles are used as inputs to estimate DARFs. The $PF_{ML}$ is then replaced with the $g$-related $PF_{HG}$ which is parameterized by $g_{ML}$ from the $PF_{ML}$ and the...
DARFs are calculated again. These relative differences between the DARFs from the above two steps are recorded and compared. The relative differences at different zenith angle conditions are calculated to comprehensively estimate the influence of the HG phase function.

Fig. 5 shows the estimated DARFs at different zenith angles. In Fig. 5(a), DARF at the TOA can vary from -2.55 to -4.8 w/m². When the $\text{PF}_{\text{Mie}}$ is replaced by the $\text{PF}_{\text{HG}}$, the calculated DARF ranges from -2.6 to -5.1 w/m². The relative difference of the DARFs between the two methods ranges from 1.3% to 7.1%, as shown in Fig. 5(b). It is concluded that using the g-related $\text{PF}_{\text{HG}}$ to replace the $\text{PF}_{\text{Mie}}$ to estimate aerosol radiative effects is applicable, with a deviation of less than 7%, in the NCP.

4.3.2 Impacts of g variations on DARF estimation

Variations in g can lead to significant variations in the estimated DARF (Kuang et al., 2016; Andrews et al., 2006; Mccomiskey et al., 2008). In this study, the uncertainties of the g values from the input parameter is estimated to be 1.95% when predicting g and the total variation in running the random forest machine learning model is estimated to be 4.47%. At the same time, the g can varies about 10% for different aerosol PNSD and can be enhanced by 20% with the increment of RH from 30% to 90%. It is very important to know the extent of the variation in DARF corresponding to the uncertainties from g.

The variation in DARF from the uncertainties of g is calculated by increasing or decreasing g by 1.95%, 4.47%, and 10% to the original g values, and then comparing the corresponding DARFs with the original DARFs. To study the influence of RH on g and DARF, the DARF with the g values calculated from the dry parameterized aerosol population profile, is estimated.

Fig. 6 shows the estimated DARFs with different variation in g and the corresponding variation in the estimated DARF. The results show that when g varies by 1.95%, the DARF can vary 4%, However, variations of 4.47% and 10% in g values can lead to variations in the estimated DARF with 9.4% and 21%, respectively. The estimated DARF using the parameterized aerosol profile, which considers the aerosol hygroscopic growth, is smaller than the DARF using the g profiles from the dry aerosol population. The g values under dry condition are smaller than that of the wet ambient. Thus, there is larger partition of energy that is scattered forward which leads to less outgoing backscattering energy and a larger value of the estimated DARF. When the DARF are estimated ignoring the impacts of aerosol hygroscopic growth on g, the relative difference can be as high as 20% for all of the zenith angles. It is necessary to considering the aerosol hygroscopic growth when calculating the g values.
5 Conclusions

The characteristics of g in the NCP are studied based on the Mie scattering theory and field measurements from sites of Gucheng and PKU. The results show that g\textsubscript{M\textit{ie}} values are 0.604±0.025 at Gucheng and 0.615±0.021 at PKU. The ambient g\textsubscript{M\textit{ie}} values at Gucheng show obvious diurnal variations due to variations in RH. When the ambient RH reaches 90%, g\textsubscript{M\textit{ie}} can be enhanced by 20%. and the g values under different aerosol population can vary 10%. Comparison of the calculated g\textsubscript{M\textit{ie}} values from the Mie scattering model and the parameterized g values from the Wiscombe and Grams (1976) method shows that the parameterized g is overestimated by approximately 10% and that the deviations are even greater when the measured σ\textsubscript{a} is below 200 Mm\textsuperscript{-1}.

The random forest machine learning model and datasets from the humidified nephelometer are employed to calculate g\textsubscript{M\textit{ie}} values. The input data of the random forest model contain measured σ\textsubscript{a} and β\textsubscript{m} at three wavelengths, RH and the hygroscopic parameter κ. Except for RH, all input data came from measurements from the humidified nephelometer system (Kuang et al., 2017). The random forest model significantly improve the accuracy of predicting g\textsubscript{M\textit{ie}}. The uncertainties of the predicted g\textsubscript{M\textit{ie}} values are constrained to be within 2.56% under dry conditions and 4.02% under ambient conditions, and the uncertainties from the measurement of the humidified nephelometer can lead to a variation of 1.95% in g, which is mainly resulted from the inaccuracy of the derived κ. The total uncertainty of calculating g using the random forest machine learning model is 4.47%. This is the first time that datasets from the humidified nephelometer system and machine learning are combined to study g. At the same time, this method can accounting for the influence of aerosol by hygroscopic growth on g.

The new method for calculating g is validated by comparing the g\textsubscript{M\textit{ie}} values from the random forest machine learning model and the g\textsubscript{CCD} values from the measured phase function by using the CCD-LADS. The g values with this two methods show good consistence with 92% of the data within the relative difference of 6.2%.

SBDART model is used to study the impacts of g on DARF. We first studied the relative differences between the estimated DARFs by using the PF\textsubscript{HG} and the calculated phase function by using the Mie theory, the measured mean aerosol PNSD and BC mass concentration at the site of PKU. The results show that the relative differences in DARF can be contained within 7.1% when replacing
The phase function with $g$-related $\Phi_{HG}$. The HG phase function can be a feasible parameterization scheme to study DARF in the NCP.

The sensitivity study shows that the maximum uncertainties of DARF are 4\%, 9.4\% and 21\%, which correspond to the uncertainties of the $g$ from the instrument measurement, the machine learning model and the variation of aerosol PNSD. However, when the DARF are estimated ignoring the effects of aerosol hygroscopic growth on $g$, the relative differences of the DARF is as large as 20\% for all of the zenith angles. It is necessary to parameter the $g$ with accounting for the effect of aerosol hygroscopic growth.

This work can further our understanding of the role of $g$ in the radiative effects of aerosols and can help reduce uncertainties in estimating DARF.

Acknowledgements

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Table 1. Field information, dataset information and instruments that are used in this study.

<table>
<thead>
<tr>
<th>Location</th>
<th>Time period</th>
<th>Datasets</th>
<th>Instruments</th>
<th>Phase function</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gucheng, Hebei</td>
<td>15 Oct to 25 Nov, 2016</td>
<td>SMP, APS</td>
<td>Aurora, Humidified Nephelometer</td>
<td>None</td>
</tr>
<tr>
<td>PKU, Beijing</td>
<td>21 Mar to 10 Apr, 2017</td>
<td>SMPS, APS</td>
<td>Aurora, Humidified Nephelometer</td>
<td>None</td>
</tr>
<tr>
<td>UCAS, Beijing</td>
<td>3 Jan to 27 Jan, 2016</td>
<td>SMPS, APS</td>
<td>Aurora, Humidified CCD-LADS</td>
<td></td>
</tr>
</tbody>
</table>
Table 2. The sensitivities of \( g \) to the input parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( \sigma_{\text{esc},450} )</th>
<th>( \sigma_{\text{esc},525} )</th>
<th>( \sigma_{\text{esc},635} )</th>
<th>( \beta_{\text{esc},450} )</th>
<th>( \beta_{\text{esc},525} )</th>
<th>( \beta_{\text{esc},635} )</th>
<th>RH</th>
<th>( \kappa )</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parameter(%)(^1)</td>
<td>4</td>
<td>2</td>
<td>5</td>
<td>7</td>
<td>3</td>
<td>11</td>
<td>6</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>( g(%) )(^2)</td>
<td>0.487</td>
<td>0.492</td>
<td>0.486</td>
<td>0.651</td>
<td>0.487</td>
<td>0.710</td>
<td>0.486</td>
<td>1.920</td>
<td>1.950</td>
</tr>
</tbody>
</table>

*1. The uncertainties of the measured parameters.

*2. The uncertainties of \( g \) values due to the uncertainties of the measurement parameters.
Figure 1. (a)(b)(c) Average diurnal pattern of RH, (d)(e)(f) g values calculated from dry aerosols, and (h)(i)(g) g values from ambient aerosols. The panels (a), (d) and (h) are the results from Gucheng. Panels (b), (e) and (i) are the results from PKU. Panels (c),(f) and (g) are the results of UCAS. The box and whisker plots represent the 5th, 25th, 75th and 95th percentiles.
Figure 2. Probability distributions of $g$ under different RH conditions. The ticks on the left show $g$ values at different RH values, and the ticks on the right show the $g$ enhancement factor, which is defined as the ratio of $g$ at a given RH to the $g$ value at dry conditions (RH=30%). The solid line (cyan) shows the mean result of $g$ values and the enhancement factor at different RH values.
Figure 3. Comparison of calculated $g$ values ($g_{\text{Mie}}$) from the Mie model and predicted $g$ values ($g_{\text{ML}}$) from the random forest model under (a) dry conditions and (b) ambient conditions at the site of PKU. Colored dots represent the concurrently measured $\sigma_{\text{sca}}$ corresponding to the time of $g$. 
Figure 4. Comparison of the calculated g values (g_{CCD}) from the CCD-LADS measured phase function and the calculated g values (g_{ML}) by using the random forest machine learning model.
Figure 5. (a) Estimated DARFs at different zenith angles when using the g-related HG phase function (dotted line) and the phase function calculated by using the Mie scattering theory (solid line). (b) The relative difference between the DARFs in (a).
Figure 6. The variation in DARF when g varies by a range of 1.95% (the filled dark color), 10% (grey color), and 20% (light grey color). Different line styles represent the corresponding mean relative differences in DARF compared to the original value.