Response to anonymous referee#2

Main comments:

Comment: In the introduction, the authors motivate their study by saying that satellites only provide one or two measurements per day, and AERONET inversions are limited to a narrow range of solar zenith angles (page 341, line 8 to 24). The authors should be fair, and their abstract and conclusion (after line 27 on page 355) should clearly state that according to their findings, observing an incomplete diurnal cycle has only second-order consequences on direct radiative effect estimates.

Response: We agree with you, and some sentences are added in the conclusions and discussions.

Comment: The authors use a fixed refractive index for the core and shell of their aerosol particles (page 346, lines 10 and 19). (1) To what chemical compositions do those refractive indices correspond? (2) Are refractive indices constant with wavelength? If so, wouldn't that be a larger source of error than diurnal variations in optical properties?

Response to (1): Thanks for your comment. The core of aerosol particles correspond to light absorbing carbonaceous aerosol (LAC) , its refractive index is assumed to be $1.80 - 0.54i$, and the shell of aerosol particles correspond to less absorbing aerosol components (include inorganic salts, acids, and most of the organic compounds), its refractive index is assumed to be $1.55 - 10^{-7}i$. And the assumptions on the refractive indices used in this research are clarified in the following.

In the following parts, the n_{non} and i_{non} refer to the real and imaginary part of the refractive index of less absorbing components, and the n_{BC} and i_{BC} refer to the real and imaginary part of the refractive index of LAC. For LAC, no accurate refractive index is valid in open literatures, and a variety of values for its refractive index have been used in different climate and aerosol optical models(Bond et al., 2013). And the valid values of refractive index for black carbon in some open literatures are listed in Table 1. The value of refractive index for LAC in this research is in accordance with that used in (Ma et al., 2012), due to the mixing state retrieved from (Ma et al., 2012) is used, and this value is the average value for wavelengths of 450, 550 and 700nm from (D'Almeida et al., 1991).

Table 1. Refractive indices of Black Carbon in open literatures at wavelength of 550nm

Source	Black Carbon
Dalzell and Sarofim (1969)	$1.56 - 0.56i$
Ouimette and Flagan (1982)	$1.56 - 0.47i$
Hasan and Dzubay (1983)	$1.97 - 0.65i$
Sloane (1984)	$1.90 - 0.55i$
Covert et al. (1990)	$1.95 - 0.66i$
Hess et al. (1998)	$1.74 - 0.44i$
Seinfeld and Pandis (2006)	$1.96 - 0.66i$
Bond and Bergstrom (2006)	$1.95 - 0.79i$

Table 2. The refractive indices of different less absorbing species at 550nm in the open literatures.

Source	Ammonium Sulfate	Ammonium Nitrate	Nitrate	Non-sea-salt Sulfate	Sea salt	Organic Mater	Residue
(D'Almeida et al., 1991)	$1.43-10^{-8}i$						
(Morgan et al., 2010)	$1.53-0i$	$1.60-0i$				$1.63-0.021i$	
(Sloane, 1984)	$1.53-0i$					$1.55-0i$	$1.53-0.005i$
(Cheng et al., 2008b)			$1.54-10^{-7}i$	$1.54-10^{-7}i$	$1.54-10^{-7}i$	$1.55-0.001i$	$1.58-0.005i$

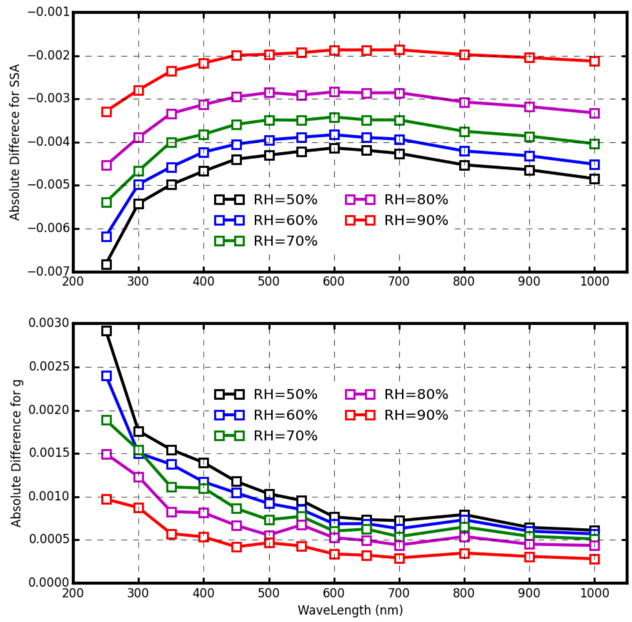

Figure 1. The absolute difference for SSA and g, when i_{non} is changed from 10^{-7} to 0.001.

For less absorbing components, the refractive indices of different less absorbing aerosol species in the open literatures are listed in Table 2. The imaginary part of their refractive indices are all very small and close to zero, in this research, i_{non} is assumed to be 10^{-7}. The real part of their refractive indices also don't vary much, the range is relatively small. And the filter-based chemical composition results (Liu et al., 2014) during the observation period demonstrate that sulfate, nitrate and organic matter dominate the composition of aerosol particles reside on accumulation mode. This means that, although the chemical composition of less absorbing components varies, the n_{non} will locate within a small range, and the value is assumed to be 1.55 in this research. With this value of refractive index of less absorbing components, good agreement is achieved between calculated and measured scattering coefficients(Ma et al., 2011). It is noteworthy that the magnitude of the imaginary parts of the refractive indices of different less absorbing components shown in Table 2 range from 0 to 10^{-3}. To insure the rationality of the use of i_{non}, a simple test is done. In this test, the average PNSD of aerosol at dry state and average mixing state during the observation period are used. For i_{non}, we change the i_{non} from 10^{-7} to 0.001 and other values of refractive indices stay the same with those used in this research. The results are shown in Figure 1, and only the results of wavelengths range from 250nm to 1000nm are shown due to the energy of solar irradiance mainly locate between this spectral range. It can be seen from this figure that even the i_{non} changed significantly, it only has negligible impacts on the calculation of SSA and g, especially for high RH conditions which are prevalent during the Haze in China (HaChi) campaign.

Response to (2): About the wavelength dependence of refractive indices of core and shell, in this research, the refractive indices are constant with wavelength. The spectral refractive indices of some aerosol species used in the widely used aerosol optical model: Optical Properties of Aerosols and Clouds (OPAC) (Hess et al., 1998), are shown in Figure 2. The real parts of refractive indices of less absorbing components and the real and imaginary part of refractive index of black carbon vary little for wavelengths range from 250nm to 1μm where most of the energy of solar irradiance locate.

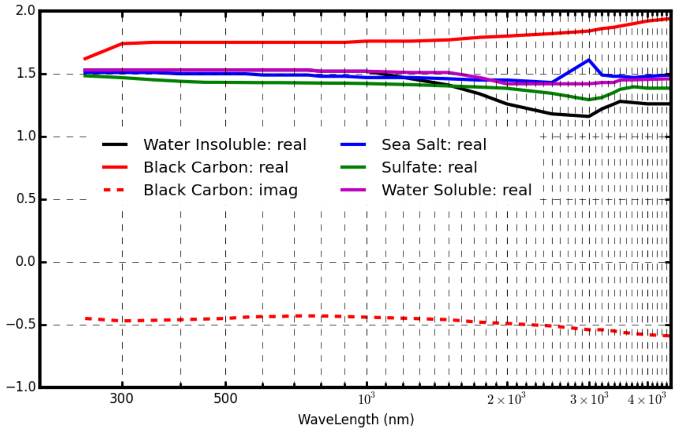

Figure 2. The real or imaginary parts of the refractive indices of aerosol chemical positions used in OPAC.

Moreover, a Monte Carlo simulation was applied to investigate how much the uncertainties of refractive indices of core and shell will impact on the calculation of SSA and g at different wavelengths. The uncertainties of the input parameters for the simulation given in terms of one standard deviation are listed in Table 2, and those uncertainties are chosen according to (Cheng et al., 2008a;Ma et al., 2012). The possible values of each parameter are assumed to be normally distributed with standard deviations listed in Table2. And the configuration of other parameters is

Table 3. Uncertainties of the input parameters for the Monte Carlo simulation, giving in terms of one standard deviation.

Parameter	Standard deviation (σ, %)
$n_{non} = 1.55$	0.5
$i_{non} = 10^{-7}$	0
$n_{BC} = 1.80$	4
$i_{BC} = 0.54$	6.6

consistent with those used in the simple test introduced previously. And the simulated uncertainties for SSA and g induced by the uncertainties of refractive indices of less absorbing components and LAC at different RH levels and wavelengths are shown in Figure 3. The results demonstrate that the uncertainties for SSA and g are small and less than 0.01, especially for high RH conditions. However, averagely speaking, the differences between the maxima and minima during daytime for SSA and g can reach up to 0.06 and 0.1, respectively.

By all accounts, we think that the wavelength dependence of refractive indices of less absorbing components and LAC will not be a larger source of error than the diurnal variations in aerosol optical properties.

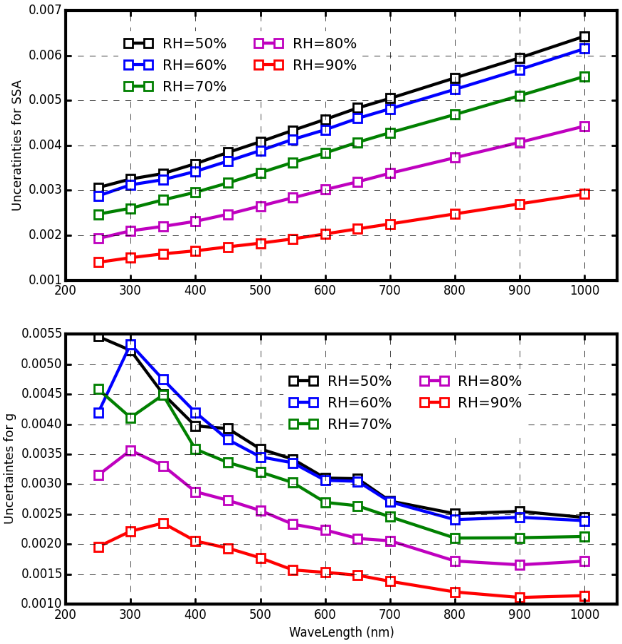

Figure 3. Uncertainties for SSA and g at different RH levels and wavelengths.

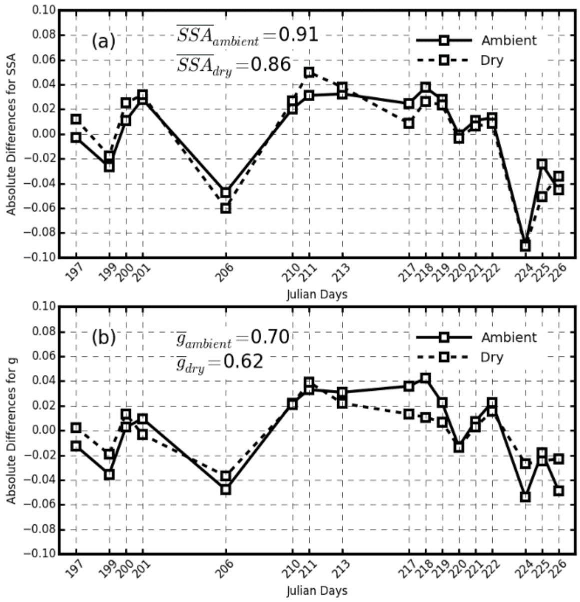

Figure 4. The absolute differences between daytime average of SSA (or g) and corresponding 17 day average of SSA (or g).

Comment: Throughout the paper, day-to-day variability of aerosol optical properties appears to be a major factor, but it is never quantified. According to Figure 1 and 2, day-to-day variability is much larger than diurnal variability. It would be useful to compare the two in terms of direct radiative effect. In addition, when discussing variability in direct effect (from page 353, line 22), variability for the case 1 (compared to its 17-day average) should be shown, and the high variability of case 5 and 8 deserves to be discussed (especially since the authors mention it on page 354, line 16).

Response: Thanks for the comment. We agree with you that aerosol optical properties have large day-to-day variability as shown in Figure 4. The day-to-day variability of SSA is larger than its diurnal changes during daytime. The diurnal variation of g during daytime is comparable to its day-to-day variability. However, in this paper, we mainly focus on the influence of the diurnal variations of SSA and g on the estimation of direct aerosol radiative effect (DARE). It is easier to capture the day-to-day variability than diurnal variability of aerosol optical properties. Such as measurements from satellites and AERONET observations, they might be capable of capturing the significant day-to-day variability of aerosol optical properties, however, it is difficult for them to capture the diurnal variations of aerosol optical properties, especially for SSA and g.

Thanks for your suggestion. The day-to-day variability of Case 1 is added in Figure 5, and the high variability of Case 5 to 8 is also discussed in the revised manuscript.

Comment: The authors claim that their findings can be applied beyond the North China Plain (page 354, lines 16-19), but this is not obvious and needs a more specific discussion. Which regions share the same diurnal pattern of relative humidity? In which seasons? Are aerosols in those regions similarly hygroscopic? I believe that the results are not as universal as claimed by the authors.
Response: Thanks for your comment. We agree with you that the results are not universal at different regions and seasons due to complication of aerosol properties and meteorology conditions. We rephrased the sentence in the text as follows: "The diurnal pattern of RH shown in Figure 3 is prevalent at many regions around the world (Ephrath et al., 1996;Gebhart et al., 2001;Fan et al., 2010;Sun et al., 2013), the scheme of Case 11 maybe also suitable for these regions when the RH is frequently higher than 60%, especially for regions where aerosol particles are similarly or more hygroscopic compared to the hygroscopicty of aerosols introduced in this research".

2. **Other Comments**
Comment: Page 340, line 6 and page 355, line 2: Saying that diurnal variations are "evident" is not useful. It would be better to say what they are.
Response: Thanks for your suggestion. We have revised it accordingly.

Comment: Throughout the paper, I recommend the use of direct radiative effect rather than forcing. The latter term is nowadays commonly used for the radiative effects of anthropogenic aerosols only (see IPCC 2013 chapter 7, or Myhre et al., 2013).
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 341, lines 4 and 20: constant in time? They also vary with wavelength.
Response: Thanks for the comment. Constant in time, vary with wavelength is considered.

Comment: Page 341, lines 5-7: The authors seem to give the conclusions of the study a little early.
Response: Thanks for the comment. The statement is rephrased as follows: "However, variations of the aerosol optical properties, including AOD, SSA and g, are important information for the estimates of daily average DARE, and the monthly and annually averaged DARE as well".

Comment: Page 341, near line 17: The study by Kaufmann et al. (2000) is relevant here.

Response: Thanks for the suggestion. The research of this study will be added in the revised manuscript.

Comment: page 342, line 3: "rather small" Be more quantitative: how small?
Response: Thanks for the comment. The quantitative information is added in the revised manuscript.

Comment: Page 343, line 21-22: The current sentence makes it sound like the temporal resolution depends on relative humidity. Please rephrase.
Response: Thanks for the comment. We have rephrased it as follows: "The particle number size distribution (PNSD) at dry state ranging from 3nm to 10 μm was observed jointly by an Aerodynamic Particle Sizer (APS, TSI Inc., Model 3321) and a Twin Differential Mobility Particle Sizer (TDMPS, Leibniz-Institute for Tropospheric Research (IfT), Germany;Birmili et al. (1999)) with a temporal resolution of 10 min, and the relative humidity (RH) of sampling air is controlled lower than 30 %".

Comment: Page 344, line 13: Rigorously speaking, g is an approximation of the phase function. Using that approximation introduces errors, see Boucher (1998).
Response: Thanks for the comment. We agree with you about this, and this is clarified in the revised manuscript.

Comment: Page 344, line 13: Say here that the AERONET site is not located in the same place as the HaChi measurements.
Response: Thanks for the suggestion. We have revised it accordingly.

Comment: Page 345, equation 3: This assumes that the $r_{ext-LAC}$ does not depend on D_p. Is that assumption reasonable?
Response: Thanks for the comment. Physically speaking, $r_{ext-LAC}$ will vary with D_p, however, in this study, $r_{ext-LAC}$ is an optically equivalent parameter retrieved from the closure between the hemispheric backscattering fraction (HBF) measured by nephelometer and the calculated HBF by using Mie theory(Ma et al., 2012). Our purpose with $r_{ext-LAC}$ in this study is using it for the calculation of aerosol optical properties, and an optically equivalent parameter is enough. Hence, we think it is reasonable.

Comment: Page 345, equation 4 and page 347, equation 11: I recommend to place brackets to clearly indicate what terms are under the summation signs.
Response: Thanks for the comment. We have revised it accordingly.

Comment: Page 345, line 11: A reference is required for the assumed density.
Response: Thanks for the comment. The reference is added.

Comment: Page 345, line 18: Again, a supporting reference is needed for that assumption. Is it a strong assumption?
Response: Thanks for your comment. This is not a strong assumption, pure black carbon is

hydrophobic(Bond et al., 2013), and the reference is added.

Comment: Page 345, equation 6: Is it really RH on the left-hand side of the equation?
Response: Yes

Comment: Page 346, line 1: Please give the value of $\sigma_{s/a}$.
Response: Thanks for the comment. The value of $\sigma_{s/a}$ is given in the revised manuscript.

Comment: Page 346, line 4: Please give a short summary of size-resolved κ method, so the paper stand on its own.
Response: Thanks for your suggestion. The short summary of size-resolved κ method is given in the revised manuscript.

Comment: Page 346, line 5: Note that using Mie theory assumes that particles are spherical.
Response: Thanks for the comment.

Comment: Page 348, line 4: Are calculations for the short spectrum only? Are diurnal variations in solar zenith angle fully accounted for?
Response: Thanks for the question. Only the short spectrum which corresponding to solar irradiance is considered, and the solar zenith angle is fully accounted for, and is declared in the part of introducing the calculation of direct aerosol radiative effect.

Comment: Page 348, line 16: Please give a typical value for surface albedo at the measurement site.
Response: Thanks for the comment. A typical value for surface albedo is given in the revised manuscript.

Comment: Page 348, line 19: Say here that those hours are approximately those of sunrise and sunset (according to page 350, line 6-8).
Response: Thanks for your suggestion, we have revised it accordingly.

Comment: Page 348, line 22: Most studies report the 24-hour average, but here it seems to be the daytime average. Any good reason why?
Response: Thanks for the comment. In this study, the reported value is also 24-hour average. It will be clarified in the revised manuscript.

Comment: Page 348, line 23 and Table 1: "ambient" is not a great choice of word, because that case in fact refers to accounting for the full temporal variation of optical properties. I suggest " full temporal" for the name of case 1.
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 348, line 26 and Page 349, line 1: Can you give approximate hours in the day for those two values of solar zenith angle?
Response: Thanks for your comment. The approximate hours are given in the revised manuscript.

Comment: Technically, case 2 should be the reference for case 3 to 11, but thankfully, results for case2 are similar to those for case 1
Response: Thanks for your comment.

Comment: Page 349, line 10 : "variation of aerosol optical properties" -> " of selected aerosol optical properties"
Response: Thanks for the suggestion. We have revised it accordingly.

Comment: Page 350, line 1: Note that the two peaks in AOD are not shown on Figure1.
Response: Thanks for the comment. We rephrased the sentence to " The value of AOD between 7 and 8 o'clock in the morning, and that at 16 o'clock in the afternoon are relatively higher. The relative departures of AOD from daily mean can be up to 20% on average."

Comment: Page 350, line 9-11, page 351, lines 8-10, page 352 line 24 to page 353 line 6: There is no need to motivate your study again, the introduction has done that. Delete those sentences.
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 351, line 3-4: It is true that single-scattering albedo of the dry aerosol varies in time. Is that due to a change in composition?
Response: Thanks for the comment. Yes, it is mainly due to the change of mass fraction of black carbon, and the mixing state of black carbon is of second-order importance.

Comment: Page 351, lines 5-7: This correlation is in fact built-in the Mie calculation as done by the authors.
Response: Thanks for the comment. We rephrased this sentence.

Comment: Page 351, line 20: I am not sure it is so easy to understand, because it implies that the diurnal changes in size distribution are small. It is not obvious it should be the case.

Response: Thanks for the comment. Averagely speaking, g for aerosol at dry state shows little

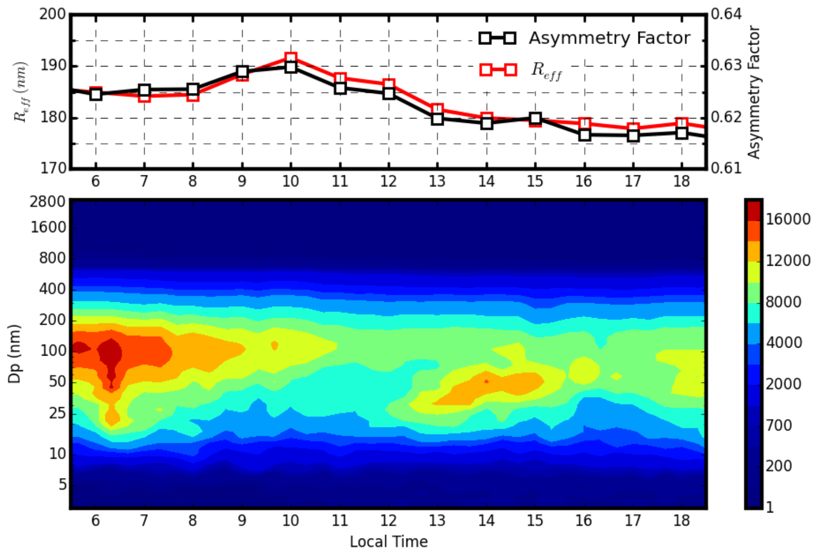

Figure 4. The diurnal variations of PNSD, the effective radius and g of aerosol particles at dry state.

diurnal variation during daytime, and shown in Figure 2 of the manuscript. However, this does not mean that diurnal changes in size distribution are small. The average diurnal changes of aerosol size distribution during the 17 selected days during daytime are shown in Figure 5, and also the diurnal changes of effective radius (R_{eff}) and g of aerosol particles. This figure demonstrate that the size distribution has evident diurnal changes, and the diurnal changes of g are highly correlated with those of R_{eff}. However, their diurnal changes are small, for g, the difference between maxima and minima is 0.013, for R_{eff}, the difference between maxima and minima is 13.8nm. This result demonstrate that rather than PNSD, the R_{eff} is directly connected with g.

Comment: Page 351, line 25: Please give the values of direct radiative effect in case 1, for the sake of completeness.

Response: Thanks for the comment. The value of direct aerosol radiative effect of the base case is presented in the revised manuscript.

Comment: Figures 4 and 5 can be merged into one.

Response: Thanks for your comment. We have revised it accordingly.

Comment: Page 352, line 1: " the results of case 2" -> " the small differences in case2"

Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 352, line 5: Why is case 4 not discussed?

Response: Thanks for the question, the discussion is added in the revised manuscript.

Comment: Page 352, lines 12 and 17: "hit onto the ground" does not read well, Rephrase to " Reaches the surface"

Response: Thanks for the suggestion, we have revised it accordingly.

Comment: "Figure 2: Please re-label the panels so that they are listed in alphabetical order in the caption.
Response: Thanks for the comment. We have revised it accordingly.

Technical Comments:
Comment: Page 341, line 12: "limited for" -> " limited to"
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 342, line 15: "are therefore important" ->" is therefore important"
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 347, line 7: " according to the definition" -> " its definition"
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 353, line 20, also captions of Figure 4, 5, and 6: typo: compare -> compared.
Response: Thanks for the suggestion, we have revised it accordingly.

Comment: Page 353, line 27: exactly -> exact
Response: Thanks for your suggestion. We have revised accordingly.

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