Response to Reviewers

Manuscript Number : acp-2015-729

Manuscript Title : Optical properties of atmospheric fine particles near Beijing during the HOPE-J^{3}A Campaign

We thank both reviewers for their thoughtful and thorough reviews of our manuscript. We are pleased that the reviewers found merit in the measurements and potentially valuable to the scientific community. There were a number of issues raised by the reviewers; we have tried to answer every point comprehensively and to address deficiencies in the analysis and interpretation of our data. Where warranted, we re-analyzed some of our data in response to the reviewer comments. Both reviewers thought the manuscript too long. Accordingly, we consolidated some sections and edited the entire manuscript to improve its clarity and brevity. The Results and Conclusions sections are now appreciably shorter. Point-by-point responses to the reviewers' comments are attached below.

Response to Reviewer #1 comments

Xu et al. present results from a study carried out around Beijing, China. The report on measurements of optical properties (extinction, scattering and absorption) and composition of PM1, contributing to the rapidly growing literature on the characteristics of the severe air pollution in this region. I find that the measurements seem to be of good quality and that the results will be of interest to the community of scholars working in this area. However, I have concerns regarding some of the analyses presented, which are discussed further below. I also think that this work is much longer than it needs to be and suggest that it could easily be cut by 30% with no loss in content and an improvement in readability simply through a reduction in redundant discussion points. I find that substantial revisions will be necessary before this work will be publishable in ACP, although believe that with effort it should ultimately be.

Specific concerns:

[1] P33680/L4: I find the meaning of "chemical extinctions" to be ambiguous here as it is not a commonly used term.

We changed "chemical extinctions" to "chemical apportionment of light extinction" in the revised manuscript.

[2] Equation S2: Although two references are given where this equation was used, has it been validated? It assumes additivity of MEE values on a diameter-by-diameter basis. A more common "mixing rule" is that associated with refractive indices. Is the MEE linearly related to the refractive index, thus justifying this equation, for example? Some additional discussion should be provided.

Under the assumption that particles were completely separated (externally mixed) and can be adequately estimated as separate terms of each single chemical component, j, the extinction coefficient of component j can be calculated from (Petersson, et al., Aerosol Sci. 35, 995-1011, 2004; Hand and Malm, J. Geophys. Res., 112, D16203, 2007):

$$\alpha_{ext,j} = \int_0^\infty \frac{\pi}{4} D_p^2 Q_{ext}(m_j, D_p, \lambda) N(D_p) dD_p$$

where $N(D_p)$ is the number of particles with mean diameter D_p per unit volume in the size bin dD_p, and $m = n + ik$ is the complex refractive index. Q_{ext} is the extinction efficiency and can be calculated with Mie theory for spherical particles.

The mass extinction efficiency(MEE) is a function of the Mie extinction efficiency, particle diameter and species density (ρ_j) (Hand and Malm, J. Geophys. Res., 112, D16203, 2007):

$$MEE_j = \frac{3}{2} \frac{Q_{ext}(m_j, D_p, \lambda)}{\rho_j D_p} \quad \text{(equation S1 in previous supplement)}$$

If the mass size distribution of each species j does not vary with total mass concentration, the light extinction coefficient of species j can be calculated with :

$$\alpha_{ext,j} = MEE_j M_j$$

where M_j is the mass concentration per unit volume.

If the mass size distribution of each species varies with the total mass concentration, equation S2 can be used for the MEE calculation.

$$MEE_j = \frac{\sum_{bin=1}^{D_{bin}} MEE(j, D_{bin}) M_{j,bin}}{\sum_{bin=1}^{D_{bin}} M_{j,bin}} \quad \text{(equation S2 in previous supplement)}$$

The total extinction is a linear combination of all the species :

$$\alpha_{ext} = \sum_j MEE_j M_j$$

Since the size-segregated mass concentration of each chemical composition was not available in this work, Equation S2 was not used. We used equation S1 (with the literature reported complex refractive index and the measured mean size distribution, Table S1) for the calculation of the MEE values at $\lambda = 470$ nm and $\lambda = 550$ nm.

In the common "mixing rule" method (volume-weighted refractive indices), the aerosols were treated as internally mixed. In the IMPROVE method, the particles are assumed to be completely separated and treated as externally mixed. The "mixing rule" was not used in this work.

[3] P33681: Was the SMPS really “calibrated” or was it simply “validated.” Calibration assumes some adjustment in the operating conditions or processing method based on the obtained results.

We replaced "calibrated" with "validated" in the revised manuscript.

[4] P33681: The authors provide no information as to the sensitivity, detection limits or uncertainties in the IBBCEAS measurements of light scattering and extinction. This information is critical to the assessment of this work and the reader should not have to look up the Zhao et al. paper. Also, the instrument performance should be reported based on the time between filter periods, not the averaging time, as this will determine the actual measurement uncertainty.

We added this paragraph in the "Experimental" section:

> The detection limits for the scattering and extinction channels with 9 s integration time were 0.54 Mm^{-1} and 0.15 Mm^{-1}, respectively. The total uncertainty in the extinction measurement was estimated to be less than 4% and arose from uncertainties in the mirror reflectivity (R) (~1%), the ratio of cavity length to the cell length containing the air sample when the cavity mirrors were purged (R_L) (~3%), and particle losses in the system (~2%). The total uncertainty in the scattering measurement was about 3%, with dominant contributions from uncertainties in the experimentally determined scattering calibration coefficient (K') (2%), and the uncertainty associated with particle losses in the cavity (2%).
>
> Based on a Mie scattering calculation, the truncated fraction of total

scattering was about 0.22% for a 1 μm diameter spherical particle with a complex refractive index (CRI) of $m = 1.6 + i0$ at $\lambda = 470$ nm. This truncation effect was therefore negligible compared to the measurement uncertainty and no correction for the truncation underestimate was applied to our data.

Potential uncertainties associated with changes in the instrument environment were considered but found to be unimportant. The instrument was located in a temperature-controlled room, the temperature inside the albedometer enclosure was maintained at 28.3 ± 0.8°C, and the sample flow was controlled with a mass flow meter. Example data of the transmitted intensity measured with the CCD spectrometer and the scattering signal measured with the PMT are shown in Fig. S1. The cavity was flushed with particle-free air every hour to acquire the $I_0(\lambda)$ spectrum. No obvious drift in the LED light intensity was observed even after 6 hours of measurement, indicating the high stability of the instrument under these operating conditions.

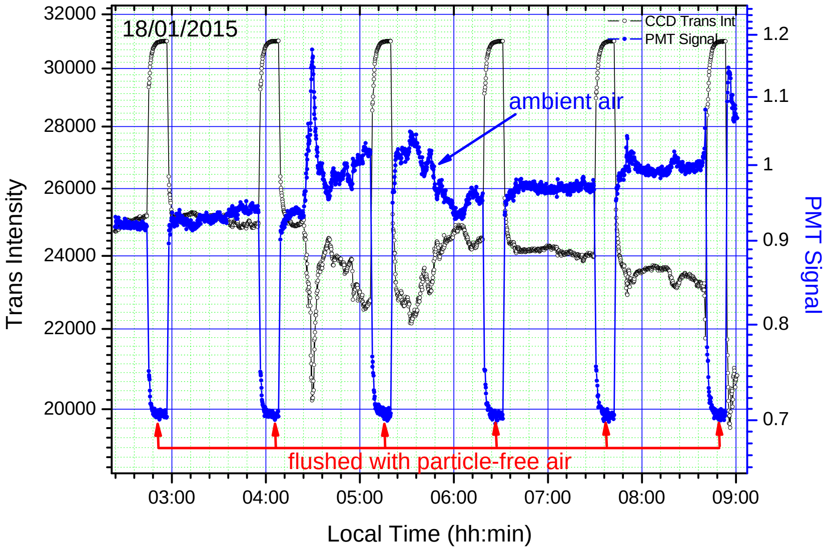


Figure S1. Example data of the transmitted intensity measured with the CCD spectrometer and the scattering signal measured with the PMT of the cavity-enhanced albedometer during the experimental period.

[5] P33684 and general: Everywhere that the authors use the term "CRI" for "complex refractive index" I suggest they should instead substitute "effective CRI" since they are using an effective parameter, not a try physical parameter. This is necessary since they are using what amounts to a volume mixing assumption even though it is known that black carbon, in particular, does not conform to volume mixing relationships and can have a non-spherical particle morphology.

Where the authors state "The CRI is one of the intensive optical properties of *atmospheric aerosols, and determined by the aerosols' size, shape, mixing state and chemical composition" they should clarify that this is an effective property that averages over variations in the properties listed. The CRI is a fundamental property of a material, not of particles. They should also state that they are implicitly assuming that BC can be treated in a volume mixing approach and spherical particle assumption, which is not fundamentally correct but commonly done.*

DONE. We have used "effective CRI" instead of "CRI".

We replaced the statement "*The CRI is one of the intensive optical properties of atmospheric aerosols, and determined by the aerosols' size, shape, mixing state and chemical composition*" with "The effective CRI is an effective property that averages over the aerosols' size, shape, mixing state and chemical composition.".

We also added a statement about the retrieval of the effective CRI.

> The measured extinction and scattering coefficients ($\alpha_{\mathrm{ep},470}$ and $\alpha_{\mathrm{sp},470}$) and the particle number size distribution, $N(D_\mathrm{p})$, were used to determine the effective CRI on the assumption that particles were spherical and that the black carbon aerosol (BC) can be treated in a volume mixing approach

[6] P33686 and IMPROVE: The authors should note that the IMPROVE algorithm was developed based on analysis of data collected in mostly remote environments (specifically ,US national parks) and thus may not be appropriate for application to urban measurements due to potential differences in typical size distributions within a mode(which affects the MEE and f(RH) terms) and the split between "small" and "large" modes. Also, the authors never define what the f(RH) terms mean (presumably the awkwardly stated "the hygroscopic increase of inorganic component.")

The IMPROVE algorithm, although it is a simplified predictor of extinction, is nevertheless a useful tool to estimate the contribution of different particle components to haze levels and the relative magnitude of these contributions (Pitchford, et al., J. Air & Waste Manage. Assoc., 57, 1326-1336, 2007).

Despite being developed from data collected in remote environments, the IMPROVE method is still a good choice for the estimation of the chemical apportionment of light extinction. It has been extensively used to characterise atmospheric aerosols in China, notably in the megacities of Beijing (Li et al., 2013), Guangzhou (Jun et al., 2009; Tao et al., 2012;Zhang et al., 2013), Shanghai, Xi'an (Cao et al., 2012), Tianjin (Han et al., 2012), Nanjing (Shen et al., 2014), and Suzhou (Tian et al., 2015).

In many cases, the revised IMPROVE algorithm gives consistent estimates of the observed aerosol characteristics. However, observations from both Chinese megacities and US monitoring sites show that the revised IMPROVE algorithm underestimates (-11 - -26%) the $PM_{2.5}$ extinction during high aerosol loading and overestimates (+25 - +54%) the values under low aerosol loading (Cheng et al., 2015).

In this work, with improved mass extinction efficiencies, the agreement among the measured and calculated $PM_{1.0}$ extinction was good when the measured extinction coefficient was lower than 300 Mm^{-1} (with a correlation slope of 1.04 ± 0.04). When the observed extinction coefficients were larger than 300 Mm^{-1}, the reconstructed values using the modified IMPROVE algorithm were 16% lower than the observed values. The IMPROVE algorithm could be appropriate for application to urban measurements in this study.

Potential differences in aerosol loading in Chinese cities and US sites may bias the results of the IMPROVE algorithm. Further field measurements are therefore needed to reduce the bias, especially for locations with high aerosol mass loadings. In this work, we also found that major absorbing aerosol components also needed to be considered.

We added the definition of f(RH) in the revised manuscript:

> where f(RH) is the water growth factor of inorganic components, f_S(RH) and f_L(RH) are the water growth factors for the small and large particle size modes for sulfate and nitrate, respectively, and f_{SS}(RH) is the hygroscopic growth factor for sea salt.

References:

Cao, J. J., Wang, Q. Y., Chow, J. C., Watson, J. G., Tie, X. X., Shen, Z. X., Wang, P., andAn, Z. S.: Impacts of aerosol compositions on visibility impairment in Xi'an, China, Atmos.Environ., 59, 559–566, 2012.

Han, S., Bian, H., Zhang, Y., Wu, J., Wang, Y., Tie, X., Li, Y., Li, X., Yao, Q.: Effect of aerosols on visibility and radiation in spring 2009 inTianjin, China. Aerosol Air Qual. Res., 12, 211−217, 2012.

Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Gu, J., and Fan, S.: Aerosol chemistry and the effect of aerosol water content on visibilityimpairment and radiative forcing in Guangzhou during the 2006 Pearl River Delta campaign. J. Environ. Manage.,90, 3231−3244,2009.

Li, X.,K. He, C. Li,F. Yang, Q. Zhao, Y. Ma,Y. Cheng,W. Ouyang, andG. Chen.:$PM_{2.5}$mass, chemical composition, and light extinction before and during the 2008 Beijing Olympics,J. Geophys. Res. Atmos.,118,12158–12167, 2013.

Shen, G., Xue, M., Yuan, S., Zhang, J., Zhao, Q., Li, B., Wu, H., and Ding, A.: Chemical compositions and reconstructed light extinctioncoefficients of particulate matter in a mega-city in the western YangtzeRiver Delta, China. Atmos. Environ., 83, 14−20, 2014.

Tao, J., J.-J. Cao, R.-J. Zhang, L. H. Zhu, T. Zhang, S. Shi, and C.-Y. Chan.: Reconstructed light extinction coefficients using chemical compositions of $PM_{2.5}$ in winter in urban Guangzhou, China. Adv. Atmos. Sci., 29(2), 359–368, 2012.

Tian, M., Wang, H. B., Chen, Y., Yang, F. M., Zhang, X. H., Zou, Q., Zhang, R. Q., Ma, Y. L., and He, K. B.: Characteristics of aerosol pollution during heavy haze events in Suzhou, China, Atmos. Chem. Phys. Discuss., 15, 33407-33443, 2015.

Zhang, G., Bi, X., Chan, L. Y., Wang, X., Sheng, G., and Fu, J.: Sizesegregatedchemical characteristics of aerosol during haze in an urbanarea of the Pearl River Delta region, China. Urban Climate, 4, 74−84, 2013.

[7] P33687: The authors state "The dry mass extinction efficiencies for ammonium sulfate, ammonium nitrate and OM at λ = 470nm were calculated by using the Mie theory and were compared with that values at λ= 550nm." These are not directly comparable due to the wavelength differences. I think that this is accounted for by the "scale factors" given on L9, but it is not clear as written what is being done or where these scale factors come from or why they differ so strongly between the different components. Maybe this is what is meant by "find out more in the Supplemental Section"?

Continuation of the reply to comment [2]. Under the assumptions of the IMPROVE algorithm (in which particles are treated as separate entities), the difference in the wavelength will only affect the dry mass extinction efficiency (MEE) terms. We modified the MEE terms (equation 3 in the discussion paper) of each individual particle components with a scaling factor so as to be directly comparable to measurements at λ = 470 nm.

The scaling factor ($Scaling\ factor_j = MEE_{j,\lambda=470nm} / MEE_{j,\lambda=550nm}$) was the ratio of the MEEs of each species at λ = 470 nm and λ = 550 nm, calculated from the literature reported complex refractive index and the measured mean number size distribution with Mie theory.

We used equation S1 (in previous supplement) for the MEE calculation at different wavelengths. The wavelength dependent MEE of each species under different polluted days is shown in Figure S1 (in previous supplement).

$$MEE_j = \frac{3}{2}\frac{Q_{ext}(m_j, D_p, \lambda)}{\rho_j D_p}$$

where the values of complex refractive index (m_j) and species density (ρ_j) were obtained from literature reported values (as shown in Table S1). The size distribution was obtained from the measured mean size distribution under different polluted days (as shown in Figure 5 in the discussion paper).

[8] Equation 5: It is not clear how the sea salt is being accounted for here. It is also not clear how the authors are treating ammonium.

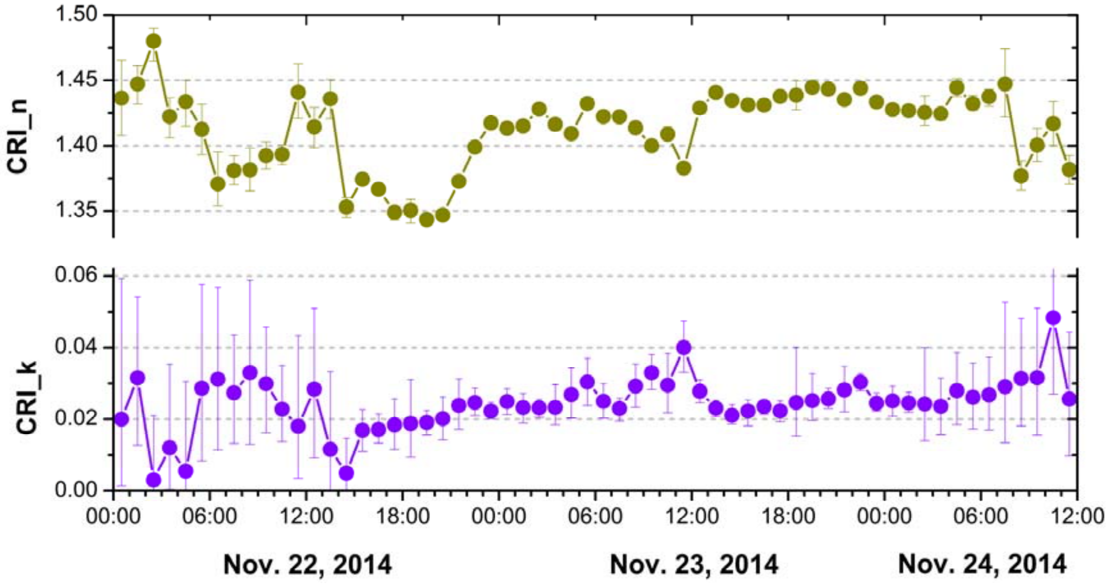
The sea salt mass concentration was estimated by the Cl^- mass concentration multiplied by a factor of 1.8. This text was added to the manuscript. The ammonium cation was not used directly in the IMPROVE algorithm. $NH_4^+$was assumed to be fully neutralized by SO_4^{2-} and NO_3^- and treated as ammonium sulfate ($(NH_4)_2SO_4$) and ammonium nitrate(NH_4NO_3), respectively.

[9] Section 3.3: The authors calculate the MAE from the total absorption divided by the total PM1 mass. It is much more common to see this parameter defined relative to the amount of BC (or EC), rather than the total PM1 as it is the BC that contributes the absorption. I suggest that the authors use the terminology MAE_PM1 (and where the_PM1 indicates subscript PM1) throughout to make clear that these are not BC-specific MAE values and avoid confusion.

DONE.

[10] P33689 and Fig. 3: It is evident in looking at the figure that the measurements at 3min are insufficient for accurate retrieval of the effective CRI when the signal levels are "low" (e.g. period 1, 2). The authors should strongly consider a longer averaging time(perhaps 1 h).

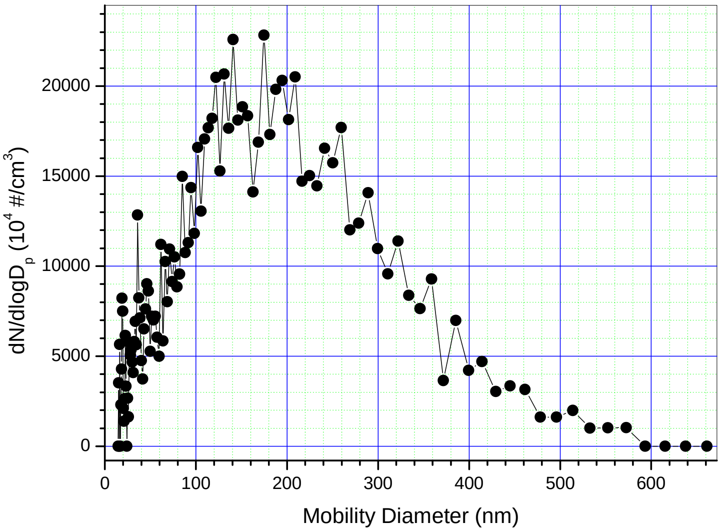
DONE. We have redrawn this Figure with 1 hour averaging time.



[11] Fig. 3: It appears that the size cuts off at 661.2 nm, presumably reflecting the limits of the SMPS and the PM1 size cut. Does the calculated scattering fall to zero at this size, indicating that the authors have captured all of the actual scattering in their calculations? If not, is there any particular dependence on time or conditions that could influence the retrieval of the effective CRI values? Could this explain some of the differences between "periods?"

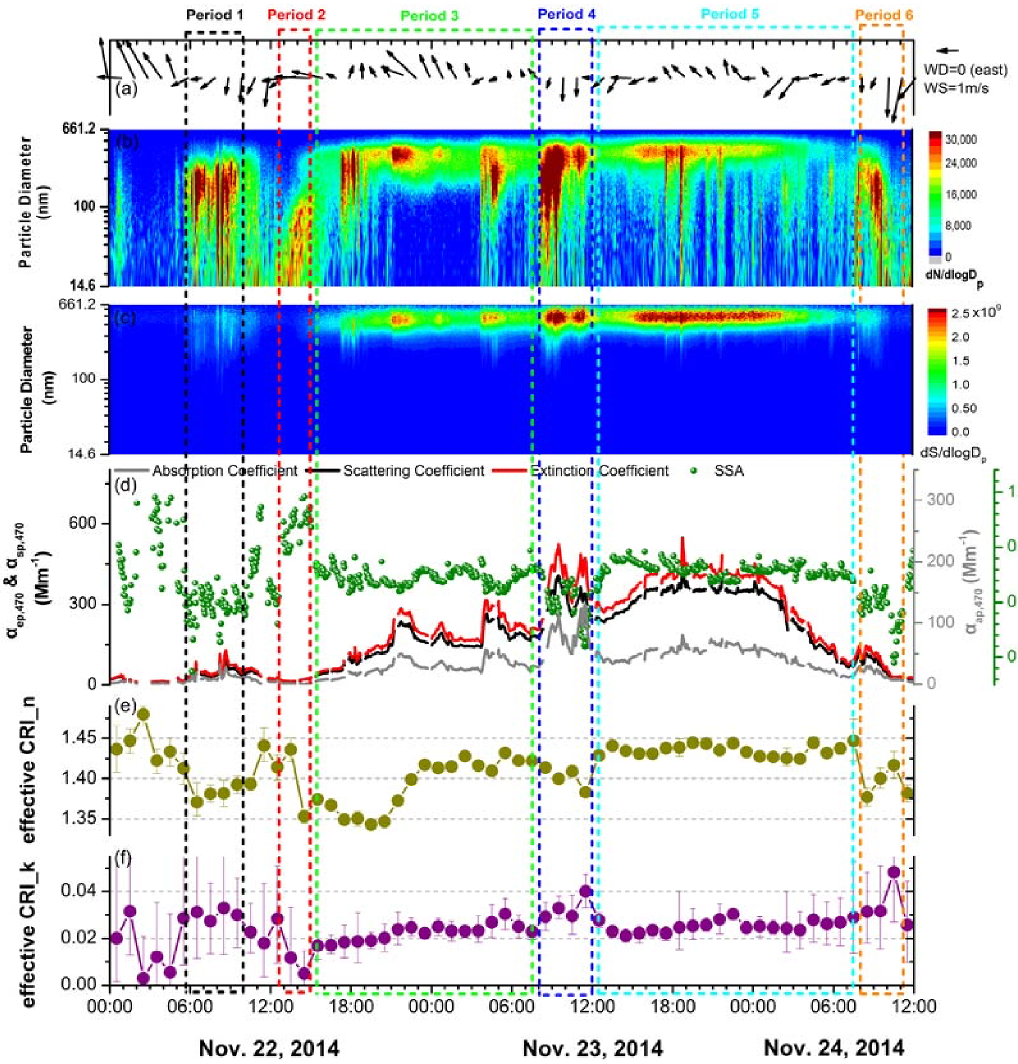
The SMPS measures the electrical mobility diameter (d_m) while the PM1.0 cutter is the aerodynamic diameter (d_{va}). In the simplest case of spherical particles, $d_m = \frac{d_{va}}{\rho_p}$, where ρ_p is the average particle density. For atmospheric particles, ρ_p is typically about 1.55. The upper size limit of the SMPS at 661.2 nm therefore corresponds to an aerodynamic diameter of ~ 1025 nm.

A typical size distribution is shown below. No particles with mobility diameters larger than 590 nm were seen. So, the calculated result captured all of the actual scattering.



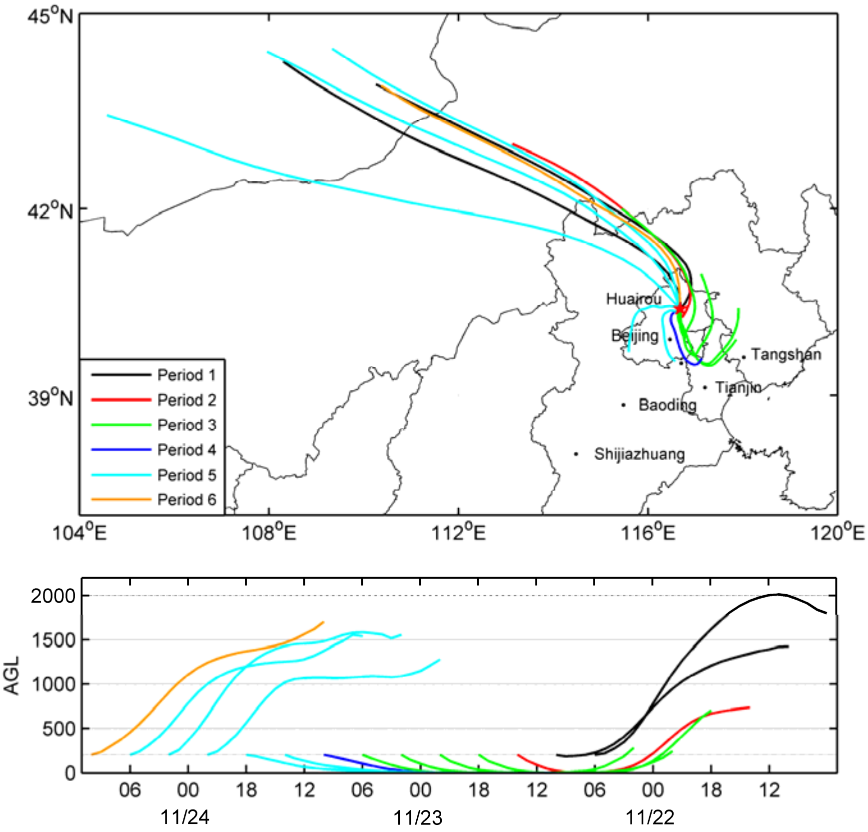
[12] Fig 3/4: I would find it more useful if Fig. 3 and Fig. 4 upper panel were combined. This would help the reader to see the relationships between the met conditions and the optical property measurements more clearly.

DONE. We have combined Fig. 4 upper panel and Fig. 3.



[13] Fig. 4: Why have the authors considered 10 clusters of trajectories but only 6 periods? I would think these should be equal. It would be much, much more helpful if the clusters were aligned with the periods (i.e. cluster 1 = period 1). Also, it is not clear how the "clusters" were determined. Or are these just individual trajectories?

DONE. We have redrawn the figure. The trajectories were calculated every 4 hours (at 2:00, 6:00, 10:00, 14:00, 18:00 and 22:00 local time) during the selected air pollution episode. All of them were individual trajectories.



[14] P33691, L4: It is not clear how the authors arrive at this conclusion.

We have rewritten this statement:

> Takegawa et al. (2009) and Guo et al. (2014) showed that newly formed aitken mode particles were mainly composed of sulfate and organic matter, indicating that most components were absolute scattering species. In this case, the values of SSA and *k* should be close to 1 and 0, respectively. The retrieved values of SSA (0.94±0.03) and *k* (0.008±0.005) matched these expectations and indicate that the newly formed particles were predominantly scattering species.

[15] P33692, L8: The authors state "However, the real part of the CRI (1.40 +/- 0.03) was larger than that of the earlier traffic-dominated pollution period." This conclusion is not justified, as the value reported for the "traffic-dominated" period was 1.38 +/- 0.06. These are identical within any reasonable uncertainties (and certainly within the stated uncertainties) and no difference can be inferred. Similarly, on P33692, L20 the authors conclude that 1.44 +/- 0.03 is "larger than" 1.40 +/- 0.03. This difference is closer to being real, but the authors here need to add "but within uncertainties."

DONE. We have clarified these statements based on the ability to distinguish between values with their stated uncertainties

[16] P33693, L13: It would seem that a new sub-section heading is appropriate here.

According to the suggestion of Reviewer 2, we moved this part (general discussion) to Section 4.1.

[17] P33693, L18: I find this to be confusing as the authors seem to be mixing changes in fractional contributions with changes in absolute concentrations (although it is not entirely clear). I actually find that the distinction between fractional and absolute concentrations is often unclear in this manuscript. And these numbers should be reconciled with those shown in Fig. 5b.

We have modified this sentence and other similar parts in the revised manuscript to reconcile the fractional and absolute concentration:

> The mean value of the total mass concentration on clear days was 9.49 μg m^{-3}, and the mean fractional contributions of OM, nitrate, and sulfate were 56% (5.32 μg m^{-3}), 12% (1.12 μg m^{-3}), and 14% (1.34 μg m^{-3}), respectively. On polluted days, the mean value of the total mass concentration was 41.64 μg m^{-3} with fractional contributions of 44% OM (18.47 μg m^{-3}), 22% nitrate (9.30 μg m^{-3}), and 11% sulfate (4.54 μg m^{-3}). In summary, in going from clear to polluted days, the fractional contribution of OM to the total mass decreased by 12%, whereas the fraction of nitrate almost doubled and that of sulfate decreased slightly.

[18] P33693, L20: The authors state that previous results indicate that inorganic species become more important during haze. They should relate their measurements here directly and explicitly to this, rather than leaving it to the reader to infer a connection. Their measurements can speak to this.

DONE. We revised this sentence:

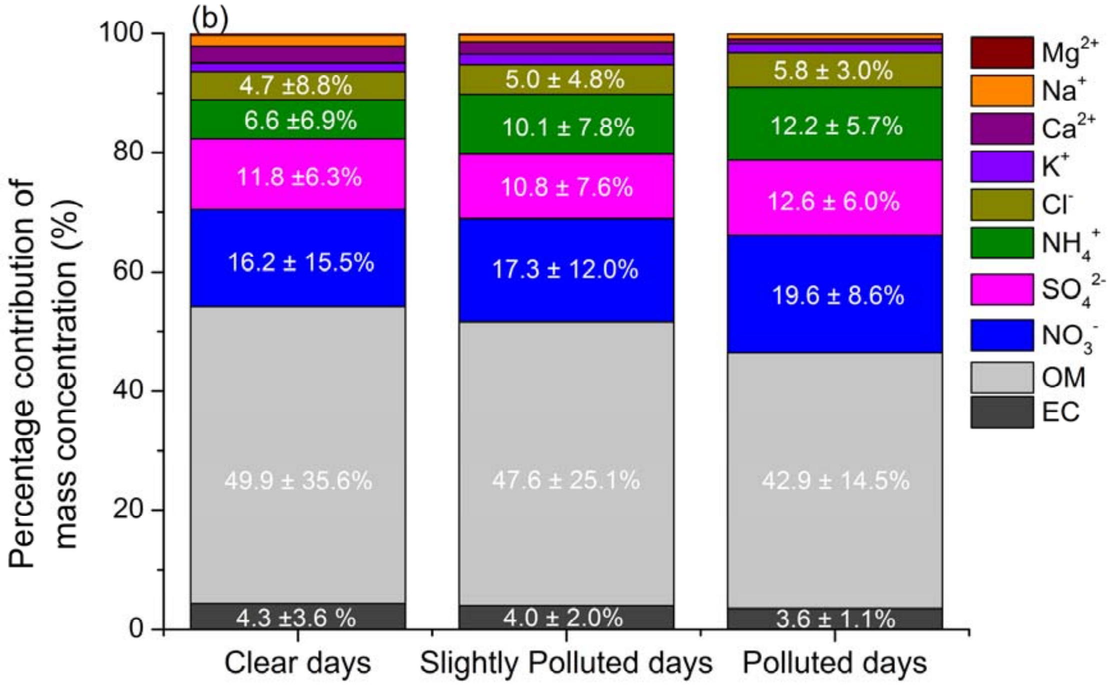
> Organic matter was the largest contributor to $PM_{1.0}$. The significant increase in the concentration and fractional contribution to $PM_{1.0}$ extinction coefficient of inorganic species, particularly nitrates (discussed in Section 4.3), indicate that inorganic species become more important during haze, an observation which is consistent with previously measurements during an extreme haze episode in Beijing in January 2013 (Wang et al., 2015).

[19] P33693, L24: The word "dramatic" should be removed.

DONE.

[20] Fig. 5b: Should state what this is a percentage of. Presumably mass concentration, but it is not stated in the caption.

DONE. We modified the y-axis label to "Percentage contribution of mass concentration".



The Figure caption was also modified accordingly.

> "(b) percentage contribution of the mass concentrations of OM, EC and eight water-soluble ion components on clear, slightly polluted and polluted days."

[21] I find "SNA" to be an awkward acronym and suggest it is removed, especially as it is only used a few times.

DONE.

[22] P33694: The authors are again a bit loose in terms of specificity when discussing absolute versus relative concentrations. I suggest that they revise to generally be clearer in this regard throughout the manuscript. They should state "relative" or "fractional" anytime that they are talking about relative or fractional

concentrations or abundances, and reserve the term "concentration" for when they are talking about absolute values. I find much of the discussion on the bottom of this page (which is discussing results from another study, not this one) to be unclear due to confusion between absolute and relative abundance.

DONE. We revised the manuscript to clarify these properties.

[23] P33695 and Table 3: The authors compare their observations to a seemingly randomly selected set of other measurements from around the world. The authors should provide some actual discussion here, and not just repeat the information in the table in the text. Is there a specific reason for comparing these sites? What is the point here?

The Huairou observation site is a new suburban site at which aerosol optical properties have not previously been reported. To put our observations in context, we selected several urban, suburban, and rural sites from around the world to compare the effect of the pollution level on light scattering and absorption.

We modified the associated paragraph in the revised manuscript:

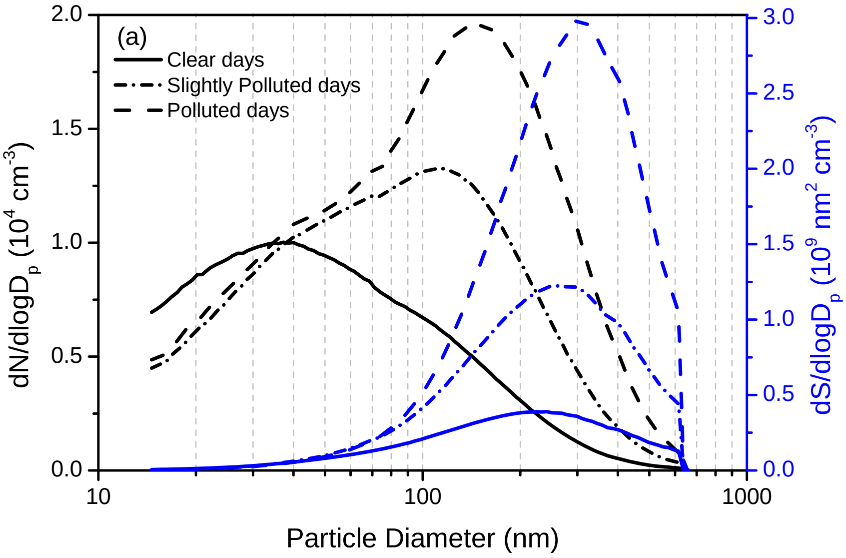
> The Huairou site is a new suburban site at which aerosol optical properties have not previously been reported. To put our observations at Huairou in context, the scattering and absorption coefficients and SSA observed in this campaign are compared in Table 3 to those at other locations (urban, suburban, and rural sites). As would be expected given the high concentrations of particulate matter in much of China, the mean $\alpha_{sp,470}$ value at Huairou was considerably higher than values observed in America and Europe, including the Los Angeles basin measurements in Pasadena (Thompson et al., 2012) and the urban site of Granada (Titos et al., 2012). Within China, the Huairou values for scattering and absorption were higher than in Shanghai (Li et al., 2013) and similar to the urban site of Guangzhou in China (Garland et al., 2008). Compared with other non-urban polluted sites in China, both $\alpha_{sp,470}$ and $\alpha_{ap,470}$ at Huairou were lower than Xinken (Cheng et al., 2008), Yufa (Garland et al., 2009) and much lower than Xianghe (Li et al.,2007). Moreover, the $\alpha_{sp,470}$ values were comparable to those observed at Shangdianzi, an atmospheric background site located ~150 km northeast of the urban center of Beijing (Yan et al., 2008). The average value of $\alpha_{ap,470}$ at Huairou was lower than those seen at other urban and suburban locations in China, with the exceptions of Guangzhou (reflecting the lower SSA values observed in Huairou) and the rural site of Shangdianzi.

[24] P33695, L21: The authors state "Higher SSA values suggest a more mixed origin for particles, including industrial emissions, domestic coal combustion, and agricultural emissions." I do not believe that this is correct. One could have a binary source just as easily as one could have a single source that could lead to a given SSA value. Or three sources. It is not clear why higher SSA values equal "more mixed" sources. Perhaps they just need to be more precise as to what they mean by this statement. And the authors appear to ignore the role of secondary formation in this discussion. This statement should be justified further if it is to be kept. Also, in the sentences that follow the authors are a bit loose in providing references, giving them for some of the studies but not others.

We agreed with the reviewer's comment. Since this statement was not a conclusion of our study, we have removed it from the revised manuscript.

[25] Fig. 5a: I suggest that the authors also show the scattering weighted size distributions.

DONE. According to the Mie theory, the slope of the plot of the scattering coefficient versus surface area concentration corresponds to an average geometric scattering efficiency (Q_{scat}) of atmosphere aerosol. The mean surface size distributions for the three different pollution categories were added in Fig. 5 a.



[26] P33697: The authors should report standard deviations associated with the PM1 PM fractional contributions.

DONE. We have added the standard deviations associated with the PM1 fractional contributions:

> On average, OM, nitrate, sulfate, ammonium, chloride and EC comprised 46.3% (13.3 ± 11.1 μg m^{-3}), 18.0% (5.2 ± 5.5 μg m^{-3}), 11.6% (3.3 ± 3.5 μg m^{-3}), 10.3% (3.0 ± 3.6 μg m^{-3}), 5.3% (1.5 ± 1.9 μg m^{-3}) and 3.9% (1.1 ± 0.9 μg m^{-3}) of observed $PM_{1.0}$, respectively.

[27] Section 4.2: I find that there is a great deal of redundancy in this section, with discussion of the particle components provided no less than 3 separate times in varying levels of detail. I strongly encourage that this section could be greatly streamlined. Also, there seems to be a lot of summarizing of other results with limited direct linking to the current study. As already noted, such connections should be made more explicit.

DONE. We rewrote Section 4.2 to make it more streamlined.

[28] Section 4.2.3: The authors conclude that the extensive property distributions were not normal distributions. There is no reason to think that they should be since there is a lower limit (zero). Since values cannot be <0, it is not possible for them to be normally distributed. This statement should be removed or revised.

DONE. We removed this conclusion.

[29] Section 4.2.4: The authors should be more explicit as to how the lower boundary layer heights would lead to a shift in the timing of the morning rush hour peaks. In the amplitude, sure. But it is not clear from what is stated how this would influence timing. Also, the conclusions regarding the "striking" consistency of the increase in the absorption coefficients between the different periods seems to go against the conclusion regarding different boundary layer heights. If emissions do not depend on meteorology, then the increase in concentration should be larger when the boundary layer is lower. These disparate thoughts should be reconciled.

As discussed in the paper, the time delay was mainly due to intense emission of light absorption particles during the morning rush hour and secondary aerosol formation. The boundary layer height mainly affects the diurnal variation pattern.

The sentence "*a strikingly consistent increase given that there is an order of magnitude difference in* $\alpha_{ap,470}$ *for the different pollution days before this period.*" probably gave a misunderstanding to the reader, which was not related to the boundary layer heights. What we would like to express is: (1) The increment of $\alpha_{ap,470}$ was about 10 to 20 Mm^{-1} from 06:00 to the maximum at 08:00 to 09:00; (2) For different pollution levels (even the magnitude of $\alpha_{ap,470}$ ranged from 10 Mm^{-1} in clear days to about 100 Mm^{-1}, one order changed), the increment of $\alpha_{ap,470}$ was still about 10 to 20 Mm^{-1} from 06:00 to the maximum at 08:00 to 09:00. It was notable that the increase in absorption coefficient was quite consistent across different pollutant days. There is a relatively stable contribution of traffic emissions regardless of the type of pollution day because the level of pollution does not (much) affect vehicular use.

[30] P33700, L10: The use of a negative sign for an underestimate would be helpful.

DONE.

[31] Fig. 8: The authors should indicate whether they have performed a 1-sided or 2-sidedfit.

The regression was a standard 2-sided, linear least squares fit to the data.

[32] P33700, L20: It is not clear how the 16% value was calculated here. This should be stated.

It was calculated from the average of the ratios of the measured extinction (y-axis) to the reconstructed extinction (x-axis) for all points > 300 Mm^{-1}. Text to this effect was added to the manuscript.

[33] Figure 8 and P33700: I find the "reconstructed mass" to be very confusing. How was this determined? This is not clear. It appears that it might just be the "summation of the concentrations all 5 groups." (P33703). But how is this different than just the PM1 mass? In other words, how does "reconstructed" mass differ from "measured" mass concentration? I feel like the authors are just saying here that A = A. I strongly suggest that this section needs to be clearer, both the methodology and the point.

We have added following discussion in the revised manuscript:

> According to the IMPROVE formula, the $PM_{1.0}$ mass concentration was reconstructed as the sum of its major chemical components (Pitchford et al.,

2007):

$$\begin{aligned}\text{Reconstructed\_PM}_{1.0} &= [(NH_4)_2SO_4] + [NH_4NO_3] + [SS] + [OM] + [EC] \\ &= 1.375\times[SO_4^{2-}] + 1.29\times[NO_3^-] + 1.8\times[Cl^-] + 1.6\times[OC] + [EC]\end{aligned}$$

The reconstructed $PM_{1.0}$ mass concentration (Fig. 8b) using the modified IMPROVE algorithm was well correlated with the measured $PM_{1.0}$ mass concentration (the summation of the concentrations of eight water-soluble ion compositions and carbon concentration (including elemental carbon, [EC], and organic mass, 1.6×[OC])) (R^2 = 0.99, slope = 1.00, intercept = 0.28) (Fig. 8d), indicating that the modified IMPROVE algorithm can be used to estimate the chemical apportionment for extinction in this campaign.

[34] Section 4.3: Personally, I find little value in this section over what has already been presented. I say this because the "chemical apportionment" is just a linear transformation of the chemical measurements that were already discussed. This section is therefore highly redundant with section 4.2. I strongly suggest that the authors merge section4.2 and 4.3. Or, if they prefer to keep these separate, to greatly reduce section 4.3 to focus on information that isn't effectively already stated in section 4.2 in the discussion of the concentration variations.

DONE. We have merged section 4.2 and 4.3 in the revised manuscript.

[35] Fig. 10: I find it odd that the slope for absorption and scattering is greater for the entire campaign than it is for any of the subset periods. How is this the case and what does this mean? Is this just an artifact of some of the periods having large non-zero intercepts?

On the basis of this comment, we reviewed the analysis of the data in Figure 8 and re-analysed the data using the reviewer's later suggestion (comment [38]).

The aerosol mass scattering efficiency (MSE, common units : m^2/g) is defined as the ratio of the total scattering coefficient (α_{scat}, common units : Mm^{-1}) to the volume mass concentration (M, common units : $\mu g/m^3$), which is similar for mass absorption efficiency (MAE) (Hand, and Malm, J. Geophys. Res., 112, D16203, 2007):

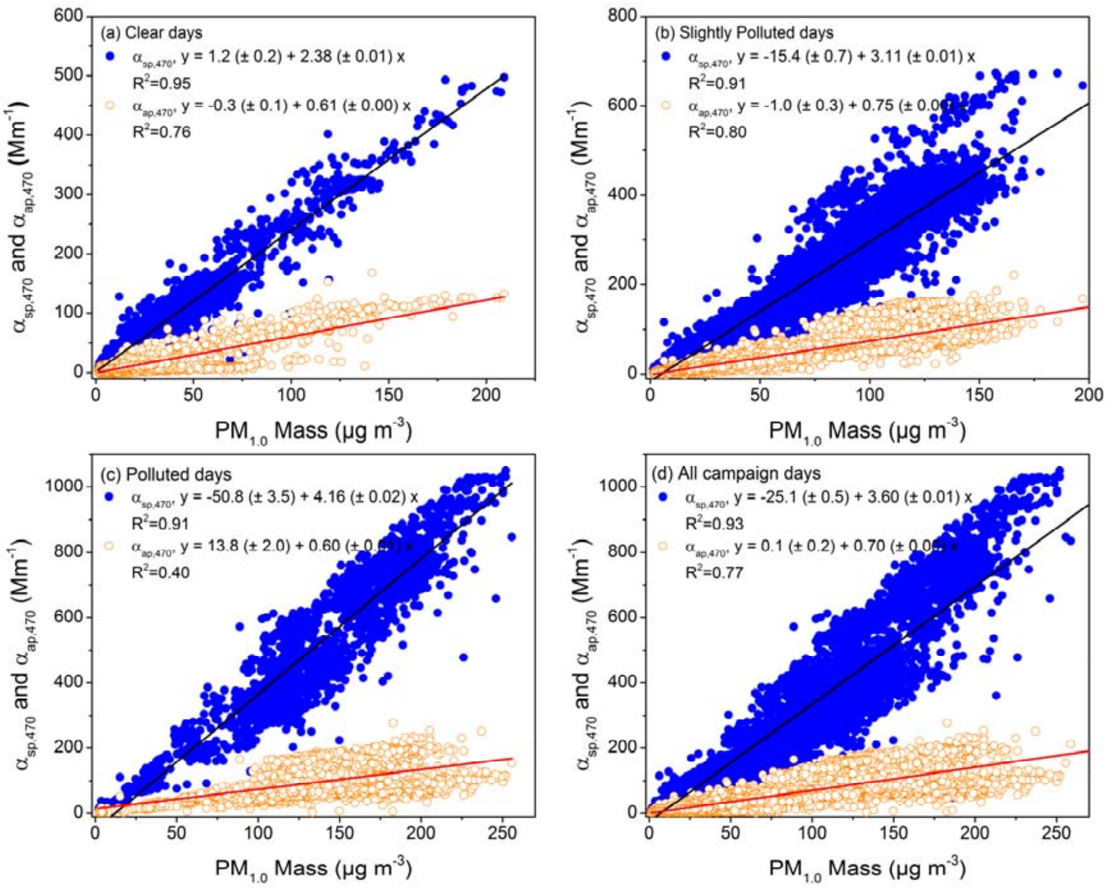
$$MSE = \frac{\alpha_{scat}}{M}$$

The simplest method for computing efficiencies is by direct measurement of aerosol scattering/extinction coefficient and the mass concentration. The average mass scattering efficiency can be estimated by dividing the averaging scattering coefficient by the average mass concentration, or the slope from a linear regression of α_{scat} and M.

In this work, the mass scattering/absorption efficiency was estimated from the slope of a linear regression of $\alpha_{\mathrm{scat,abs}}$ and M (each data point with 3 min time resolution). The $\alpha_{\mathrm{scat,abs}}$ was directly measured with the cavity-enhanced albedometer with high time resolution, and M was calculated from the average density and the volume concentrations measured with the SMPS ($M = \overline{\rho} \int V(D_p) dD_p$). The average density was calculated with $\overline{\rho}^{-1} = \sum_j \rho_j^{-1} X_j$, where ρ_j is the density of each chemical components (as listed in Table S1), X_j is the mass concentration ratio of each species (reconstructed with the IMPROVE formula, see reply to comment [33]) to the total $\mathrm{PM}_{1.0}$ concentration measured from filter sample.

In the discussion paper, we used the average $\overline{\rho}$ over the whole campaign to calculate the mass concentration, essentially treating $\overline{\rho}$ as constant under different pollution conditions. However, this approximation is not sufficiently accurate when there are large daily changes in the physical and chemical properties of atmospheric aerosols. The average $\overline{\rho}$ caused the larger slope for absorption and scattering for the entire campaign compared to the different pollutant conditions.

We revised our calculations by using $\overline{\rho}$ from each sample filter (at 12 hours time resolution). The revised results (see below) show that the absorption and scattering efficiencies for the entire campaign were between that of the subset periods. The figure was accordingly revised and the calculation method clarified in the manuscript.



[36] P33702, L15: The term "well-correlated "does not seem appropriate for the absorption measurements on polluted days (R^2 = 0.43).

We have removed the term "well-" in the revised manuscript.

[37] P33702: The authors should note the substantial non-zero intercepts. Or modify their fitting to fix the intercept to zero. And they should state whether they have performed1-sided or 2-sided fits. 2-sided fits are actually appropriate in this case, and if not performed the authors should justify this choice. They should also note that the uncertainties reported are fit errors, which are completely unrealistic and unrepresentative of the actual uncertainty on this value.

Excepting observations from polluted days, we contend that the y-intercepts are actually rather small. The larger deviation of the y-intercept on polluted days likely stems from the smaller proportion of points near the origin on these days (as would be expected for high particle loadings in the atmosphere): more points near the origin would constrain the fit of the y-intercept to smaller absolute values.

The reviewer is correct that the nature of the uncertainty is purely related to the fit statistics and are misleadingly small. We accordingly removed the uncertainties from the reported MSA and MSE values and explained this decision in the text:

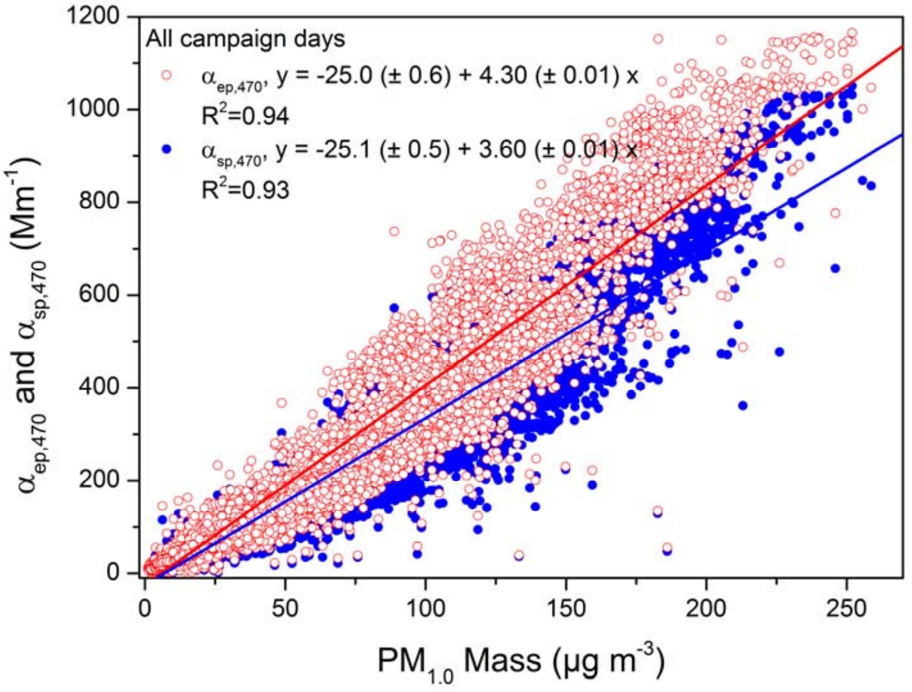
> Statistical uncertainties from the slope of the regression are misleadingly small and have been omitted in the reported MSE and MAE values.

[38] P33702: The authors report at the bottom of the page a MEE that is smaller than the MSE (4.35 vs. 4.66). This is impossible and obviously the result of fit error and limitations. That the MEE < MSE should give the authors pause. They must revisit this issue and explain it sufficiently, as it is a physical impossibility. The entire section should be revised accordingly, as it is clear that the derived values are not robust.

As described in the reply of comment [35], we have improved the data analysis method and revised this section accordingly:

> The derived mass extinction efficiency is 4.30 $m^2 g^{-1}$, which was comparable to the reported values during Aerosols 1999 (4.1–5.4 $m^2 g^{-1}$) (Quinn et al., 2001) and INDOEX 1999 (4.0–5.6 $m^2 g^{-1}$) (Quinn et al., 2002).

The fitted result was shown in the following Figure.



[39] P33703: I find all the discussion regarding the MSE values and how they depend on concentration to be a bit lacking. Specifically, it is well known that the primary factor that influence the MSE is particle size. The authors mention particle size variations, but almost as an afterthought. Particle size variations are the driver of the variations. It just so happens that particle size in this region correlates with mass concentration .I strongly suggest that the authors revise this to focus more on the fundamental relationship(size) and less on the secondary relationship (mass concentration).

We have added following discussion in the revised manuscript:

> The variability in MSE is typically more dependent on mass size distribution than on density or refractive index (Hand and Malm, 2007). As shown in Fig. 5(a), the particle diameter increased significantly with high aerosol mass concentrations. The MSEs in this work increased consistently with pollution level, primarily because larger particles scatter light more efficiently.

[40] P33703: The authors should report explicitly the EC/PM1 fraction, not just allude to it.

We have added the EC/PM1.0 fraction in the revised manuscript:

> At the same time, the EC fraction decreased with increasing pollution level (clear days: 0.48 ± 0.39 μg m^{-3} (4.3 ± 3.6 %), slightly polluted days: 1.18 ± 0.59 μg m^{-3} (4.0 ± 2.0 %) and polluted days: 2.72 ± 0.87 μg m^{-3} (3.6 ± 1.1%).

[41] P33703, L21: I find the conclusion regarding "brown carbon" formation to be highly speculative and not sufficiently justified/demonstrated. It is also severely lacking in discussion. For example, why would there be more "brown carbon" during the slightly polluted period than the polluted period? What would cause this? Is it reasonable? Discussion is necessary if this is to be retained. Also, some uncertainty assessment is required. How precise are the EC/PM1 estimates? What are the MAE values with respect to EC only? Are they reasonable? Do they differ between periods in the same way?

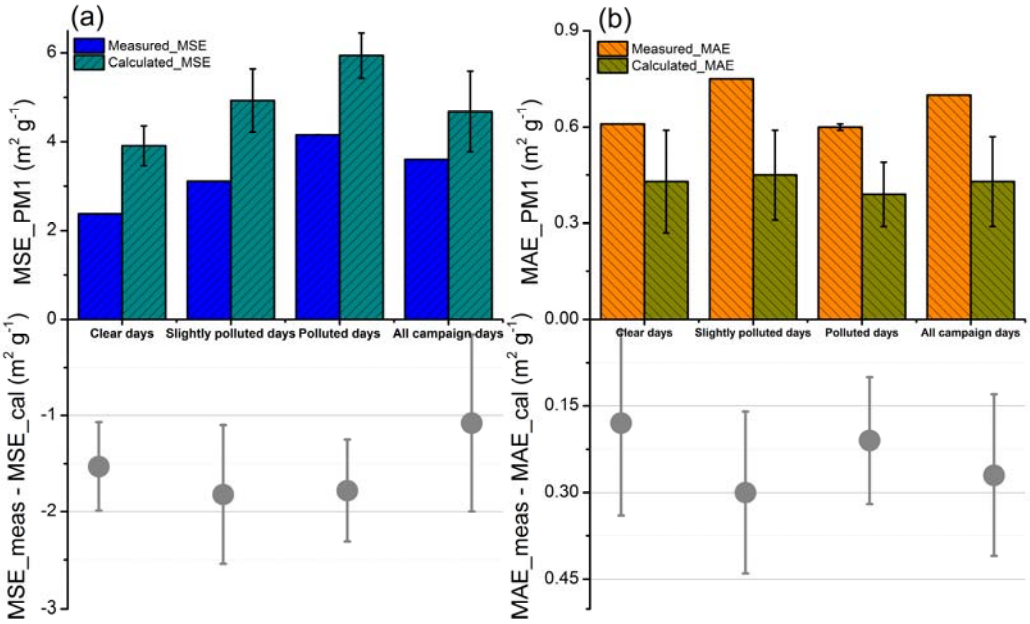
As described in the reply of comment [35], we improved the data analysis method and modified Fig 11.We revised the manuscript accordingly:

> The observed MAEs of $PM_{1.0}$ were 0.61, 0.75, and 0.60 m^2 g^{-1} for clear, slightly polluted and polluted days, respectively. At the same time, the EC fraction decreased with increasing pollution level (clear days: 0.48 ± 0.39 μg m^{-3} (4.3 ±

3.6 %), slightly polluted days: 1.18 ± 0.59 μg m^{-3} (4.0 ± 2.0 %) and polluted days: 2.72 ± 0.87 μg m^{-3} (3.6 ± 1.1%). As EC is the only absorbing species in the modified IMPROVE algorithm, the extinction coefficients reconstructed using equation (5) can be divided into two parts: absorption caused by EC and scattering caused by the other four groups. To compare with the measurement results, the reconstructed $PM_{1.0}$ mass concentration was used for the calculation of the reconstructed MSEs and MAEs of $PM_{1.0}$ particles (Fig. 11). The calculated MSEs were 1.64, 1.58 and 1.43 times larger than the observed MSEs for clear, slightly polluted and polluted days, respectively. In contrast, the calculated MAE values were 1.41, 1.67 and 1.54 times smaller than the experimental results. MSEs increased with increasing pollution level in both experimental and calculated results (the differences in measured and calculated MSE values were -1.53, -1.82, and -1.78 m^2 g^{-1}, respectively), whereas for MAEs, the experimental and calculated values increased in slightly polluted conditions, but decreased under polluted conditions (the differences in the measured and calculated values were 0.18, 0.30, and 0.21 m^2 g^{-1}, respectively). Even though further improvement of the calculation method is necessary, the large difference of MAE on slightly polluted days suggests aerosol absorption is incompletely represented. This discrepancy may indicate the presence of other light absorbing components such as brown carbon, BrC. Wang et al. (2013) have recently shown that BrC was the second-largest absorbing aerosol constituent in Beijing (with a contribution of about 5 - 25%) and exhibits a clear seasonal variation (dominates in late fall and winter, and extremely low in summer).

Reference:

Wang, L., Li, Z., Tian, Q., Ma, Y., Zhang, F., Zhang, Y., Li, D., Li, K., and Li L.: Estimate of aerosol absorbing components of black carbon, brown carbon, and dust from ground-based remote sensing data of sun-sky radiometers, J. Geophys. Res. Atmos., 118, 6534–6543, doi:10.1002/jgrd.50356, 2013.



[42] P33703: It is not clear how the comparison between the observed and reconstructed MEE and MSE values in this section relates to the reconstruction in the previous section .I feel as if these should be directly relatable.

The reported MSE and MAE was specially refer to atmosphere PM 1.0 particles in this work.

According to the IMPROVE formula, the $PM_{1.0}$ mass concentration was reconstructed as the sum of its major chemical components (Pitchford et al., 2007):

$$\begin{aligned}\text{Reconstructed\_PM}_{1.0} &= [(NH_4)_2SO_4] + [NH_4NO_3] + [SS] + [OM] + [EC] \\ &= 1.375\times[SO_4^{2-}] + 1.29\times[NO_3^-] + 1.8\times[Cl^-] + 1.6\times[OC] + [EC]\end{aligned}$$

As EC is the only absorber and treated as pure absorbing in the modified IMPROVE algorithm, the reconstructed extinction coefficients by using equation (5) can be easily divided into 2 parts : absorbing caused by EC and scattering caused by the other 4 groups. The reconstruction of scattering and absorption coefficients with IMPROVE were calculated with following formula.

$$\begin{aligned}\alpha_{scat,470nm,PM_{1.0}} = {} & 2.88\times[\text{Small Sulfate}] + 6.29\times[\text{Large Sulfate}] \\ & +3.14\times[\text{Small Nitrate}] + 6.68\times[\text{Large Nitrate}] \\ & +3.64\times[\text{Small Organic Mass}] + 7.93\times[\text{Large Organic Mass}] \\ & +2.23\times[\text{Sea Salt}]\end{aligned}$$

$$\alpha_{abs,470nm,PM_{1.0}} = 10.8\times[\text{Elemental Carbon}]$$

The MSE and MAE of PM1.0 were calculated with :

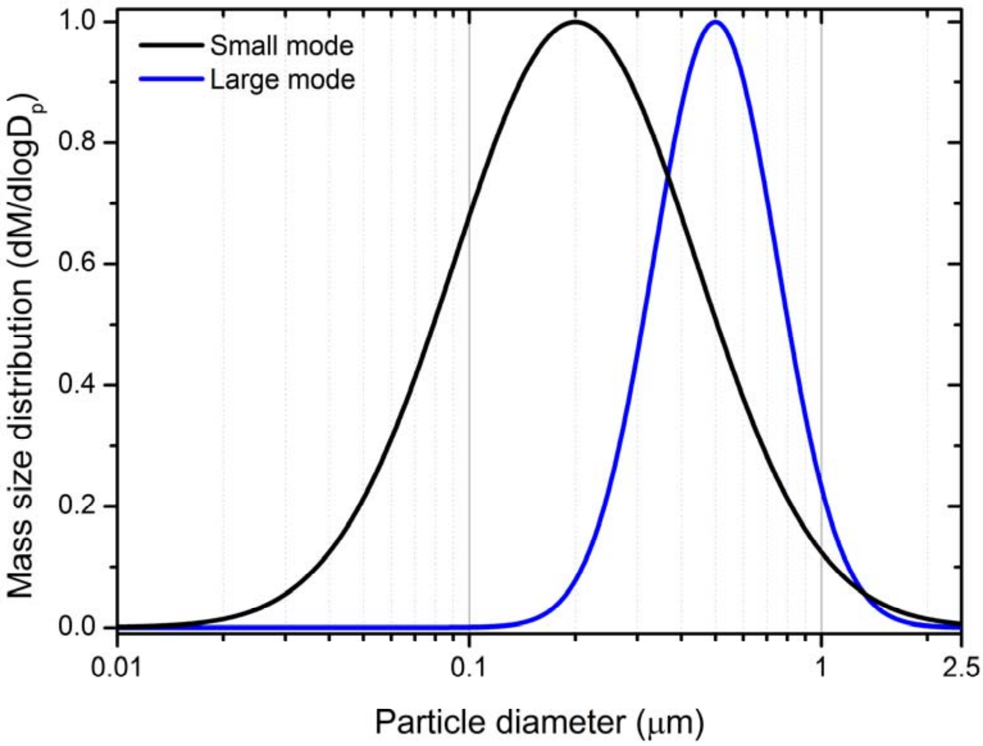
$$MSE\_PM1 = \frac{\alpha_{scat,470nm,PM1.0}}{\mathrm{Re}constructed\_PM1.0}$$

$$MAE\_PM1 = \frac{\alpha_{abs,470nm,PM1.0}}{\mathrm{Re}constructed\_PM1.0}$$

[43] P33703, L25 and Fig. 11: As with the linear fitting above, there is something in congruous about the MSE's for each of the individual cases being smaller than the reconstructed method while the campaign average matches well and is larger than any of the cases. How is this possible? I would think that the campaign average should be in between the different cases. Also, unless I am missing something it is entirely unclear how the reconstructed MSE's were calculated. Equations are only given for the MEE(in the supplemental). Same is true for the MAE values. I also find it difficult to understand the large calculated (i.e. reconstructed) values shown in Fig. 11. The individual component MEE values given in Eqn. 2 only range from 2.88 to 3.64 for in organics and organics (not counting the "large" component, which the authors stated were ignored).The fractional contribution to EC is too small for it to substantially increase the MSE. And the composition does not change substantially between periods in terms of the inorganic/organic difference and thus one would not expect the reconstructed MSE to change very much either. I find this bit to be lacking in key methodological details.(Note: I also don't understand why Eqn. 5 has "large" components when the authors seem to indicate that these are ignored. If they are ignored, they should just be removed from the equation. If they are not ignored, then much more detail is needed in section 3.2.)

We reanalyzed the data and redrew Fig. 11 in the manuscript (as shown in Reply to comment [41]). The revised MSEs and MAEs for PM 1.0 were 2.38 and 0.61, 3.11 and 0.75, 4.16 and 0.60, and 3.6 and 0.7 m^2 g^{-1} for clear, slightly polluted, polluted, and all campaign days, respectively. The values of MSE and MAE of the campaign average now fall between the different cases.

Following figure gives a representation size distribution of the small and large particle size modes in the revised IMPROVE algorithm (Pitchford, et al., 2007). Because of the size distribution of the small and large particle was mixed, we kept the "large particle size mode" term in our this work.



[44] P33706: I find the last two sentences of the conclusions to be particularly awkward in terms of writing in a way that makes them difficult to understand. I suggest these need revising, as do all of the conclusions in relation to the issues raised above.

DONE. We have rewritten these two sentences:

> Organic matter was consistently the dominant constituent by mass of the observed aerosols, and light extinction apportionment indicated that was it accordingly made the largest contribution to the extinction of $PM_{1.0}$. Under polluted conditions, the proportion of inorganic components, especially nitrate, was higher than under clean conditions and the contribution of inorganic components to visibility degradation was significant.

Technical corrections:

[45] Fig. 2: The axes labels should be changed to avoid the use of abbreviations (e.g. coeff= coefficient and concen = concentration)

DONE.

[46] There are numerous typos throughout the manuscript, too many to take the time

to document here. The authors should have this read over by a native English speaker.

DONE. The entire document has been carefully and thoroughly edited for brevity, clarity, and to remove any typing errors.

Response to Reviewers

Manuscript Number : acp-2015-729

Manuscript Title : Optical properties of atmospheric fine particles near Beijing during the HOPE-J^3 A Campaign

We thank both reviewers for their thoughtful and thorough reviews of our manuscript. We are pleased that the reviewers found merit in the measurements and potentially valuable to the scientific community. There were a number of issues raised by the reviewers; we have tried to answer every point comprehensively and to address deficiencies in the analysis and interpretation of our data. Where warranted, we re-analyzed some of our data in response to the reviewer comments. Both reviewers thought the manuscript too long. Accordingly, we consolidated some sections and edited the entire manuscript to improve its clarity and brevity. The Results and Conclusions sections are now appreciably shorter. Point-by-point responses to the reviewers' comments are attached below.

Response to Reviewer #2 comments

The authors describe field measurements during November 2014 - January 2015 in the North China Plain. They measure aerosol extinction, scattering, and absorption, together with size distribution and composition. They examine the aerosol optical properties in detail over two days with different pollution levels. The cavity-enhanced albedometer is a unique and interesting instrument. However, some important questions need to be addressed before publication.

Major Comments

1. The repeatability of the cavity-enhanced albedometer is mentioned, but its accuracy is not discussed. The following questions should be addressed:

- How has the cavity-enhanced albedometer been validated? How well did the measurement agree with Mie calculations for standard aerosol particles (PSL, ammonium sulfate)?

The performance of the cavity-enhanced albedometer was evaluated in measurements of laboratory generated, monodispersed polystyrene latex spheres (PSL, non absorbing) and the Nigrosine (absorbing) aerosols. The retrieved effective complex refractive index (CRI) from scattering channel and extinction channel independently agreed with the literature reported values. For further evaluation and validation of the instrument for field measurement, scattering measurement was compared with the TSI 3563 nephelometer. Please find more information in our previously published work (Zhao et al., 2014a, b).

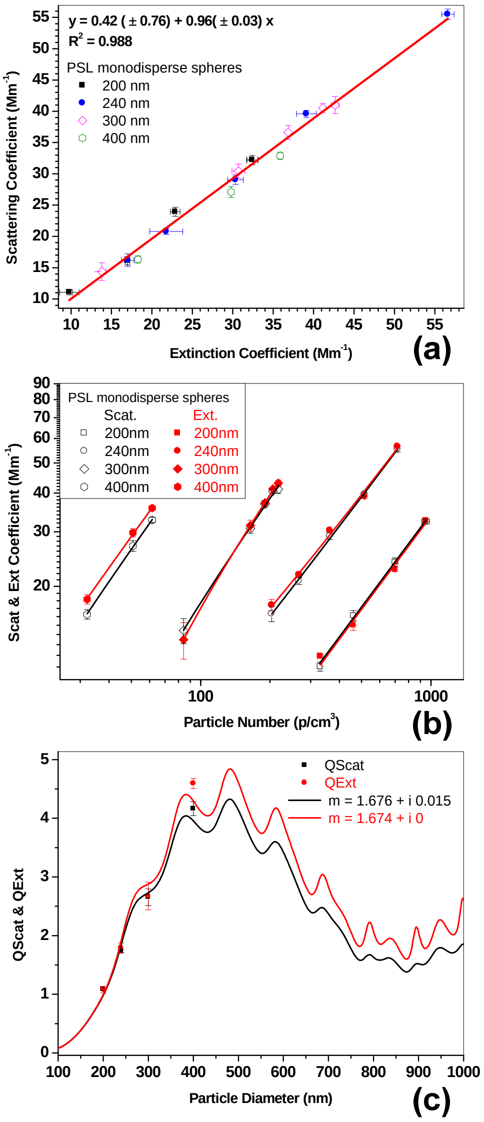


Fig. 8. in Reference (Zhao et al., 2014a) (a) Regression plot of the measured extinction and scattering coefficients, **(b)** scattering and extinction coefficients as a function of particle concentration, and **(c)** the scattering (Q_{Scat}) and extinction (Q_{Ext}) efficiencies as a function of particle diameter for monodisperse PSL spheres with four different particle diameters (200, 240, 300 and 400 nm) at λ= 470 nm.

We have added following statement in the revised manuscript.

> The performance of the cavity-enhanced albedometer was previously evaluated using laboratory-generated, monodisperse standard aerosol particles, and the scattering measurements were also found to be in close agreement with TSI 3563 nephelometer measurements (Zhao et al., 2014 a, b)

Reference:

Zhao, W., Xu, X., Dong, M., Chen, W., Gu, X., Hu, C., Huang, Y., Gao, X., Huang, W., andZhang, W.: Development of a cavity-enhanced aerosol albedometer, Atmos. Meas. Tech., 7,2551–2566, 2014a.

Zhao, W., Xu, X., Dong, M., Chen, W., Gao, X., Huang, W., and Zhang, W.: Development of a cavity-enhanced albedometer for simultaneous measurement of aerosol extinction and scattering coefficients, in imaging and applied optics 2014, OSA Technical Digest (online) (Optical Society of America), paper JTu4A.43, 2014b.

- How does the truncated fraction of total scattering vary with particle diameter, and what error does that introduce?

Size-dependent truncated fraction of total scattering for various truncation angles is shown in the following figure. Truncation effects are an important limitation of scattering measurements with nephelometers because light scattered at angles smaller and larger than the truncation angles is not detected. For instance, the TSI 3563 integrating nephelometer measurements are limited to scattering angles from 7 and 170º. The truncation errors lead to the underestimation of scattering coefficients, particularly for particles with large size.

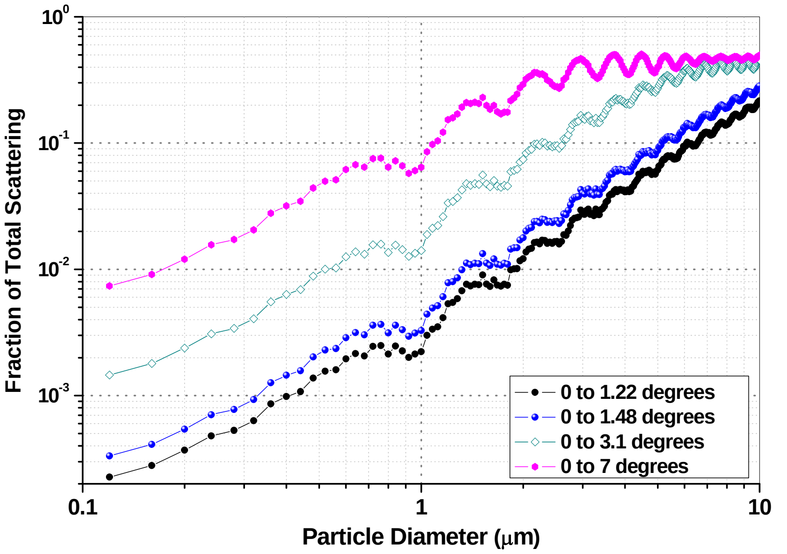


Fig. 3 in Reference (Zhao et al., 2014a). Size dependence of the truncated fraction of total scattering under different truncation angles: (1) 0–1.22°, calculated with $d_e = (d - d_0)/2$; (2) 0–1.48°: with $d_e = (L - d_0)/2$; (3) 0–3.1°: without truncation reduced tubes; and (4) 0–7°: for the used TSI Nephelometer. The simulations were made based on Mie scattering theory applied to monodisperse particles with a refractive index of $m = 1.6 + i0$ at $\lambda = 470$ nm.

Based on Mie scattering calculations, the truncated fraction of total scattering is about 0.22% with this truncation angle for a 1 μm diameter spherical particle with a complex refractive index (CRI) of $m = 1.6+i0$ at $\lambda = 470$nm. This small truncation effect is negligible in the context of the results we report here, as we discuss below.

- *What was the precision of the mirror reflectivity and scaling factor measurements?*

The absolute accuracy of the mirror reflectivity (R) and scaling factor (K') were limited by the uncertainty of Rayleigh scattering cross-section for He, N_2, and CO_2. The experimental uncertainty of Rayleigh scattering of N_2 is about 1%. The uncertainty of He is similar, but this value makes a minor contribution to the total uncertainties of R and K' (Washenfelder, et al., Atmos. Meas. Tech., 6, 861 - 877, 2013). The experimental uncertainty of CO_2 is about 4%.

In this work, the mirror reflectivity was calibrated using He and N_2. Ten different pairs of He and N_2 extinction measurements were used for the mirror reflectivity determination, of which 10 values of the mirror reflectivity were averaged. The mean value was used as the mirror reflectivity and the standard deviation in (1-R) values was about 1%.

A linear fit of the theoretical Rayleigh scattering coefficient of He, N_2 and CO_2 to the measured I_{scat}/I_{trans} (experimentally measured scattering intensity and transmitted intensity of the cavity) ratio was used for the calibration of K'. The error of the regression plot of K' was about 2%.

- Define the "scaling factor (K')".

We have added the following definition in the revised manuscript.

> The mirror reflectivity $R(\lambda)$ and the scaling factor (K', the calibration coefficient that related instrument response to scattering magnitude) for the scattering channel of the albedometer were determined by He, N_2 and CO_2 every week.

- Add an error budget for the cavity-enhanced albedometer and give the total uncertainty. The error budget would include uncertainty in temperature, pressure, Rayleigh scattering of calibration gases, truncation angle, aerosol sampling losses, and possibly other factors.

This is a similar issue raised by reviewer 1 (comment [4]). We added this paragraph in the "Experimental" section:

> The detection limits for the scattering and extinction channels with 9s integration time were 0.54 Mm^{-1} and 0.15 Mm^{-1}, respectively. The total uncertainty in the extinction measurement was estimated to be less than 4% and arose from uncertainties in the mirror reflectivity (R) (~1%), the ratio of cavity length to the cell length containing the air sample when the cavity mirrors were purged (R_L) (~3%), and particle losses in the system (~2%). The total uncertainty in the scattering measurement was about 3%, with dominant contributions from uncertainties in the experimentally determined scattering calibration coefficient (K') (2%), and the uncertainty associated with particle losses in the cavity (2%).
>
> Based on a Mie scattering calculation, the truncated fraction of total scattering was about 0.22% for a 1 μm diameter spherical particle with a complex refractive index (CRI) of $m = 1.6+i0$ at λ = 470nm. This truncation effect was therefore negligible compared to the measurement uncertainty and no correction for the truncation underestimate was applied to our data.
>
> Potential uncertainties associated with changes in the instrument environment were considered but found to be unimportant. The instrument was located in a temperature-controlled room, the temperature inside the albedometer enclosure

was maintained at 28.3 ± 0.8°C, and the sample flow was controlled with a mass flow meter. Example data of the transmitted intensity measured with the CCD spectrometer and the scattering signal measured with the PMT are shown in Fig. S1. The cavity was flushed with particle-free air every hour to acquire the $I_0(\lambda)$ spectrum. No obvious drift in the LED light intensity was observed even after 6 hours of measurement, indicating the high stability of the instrument under these operating conditions.

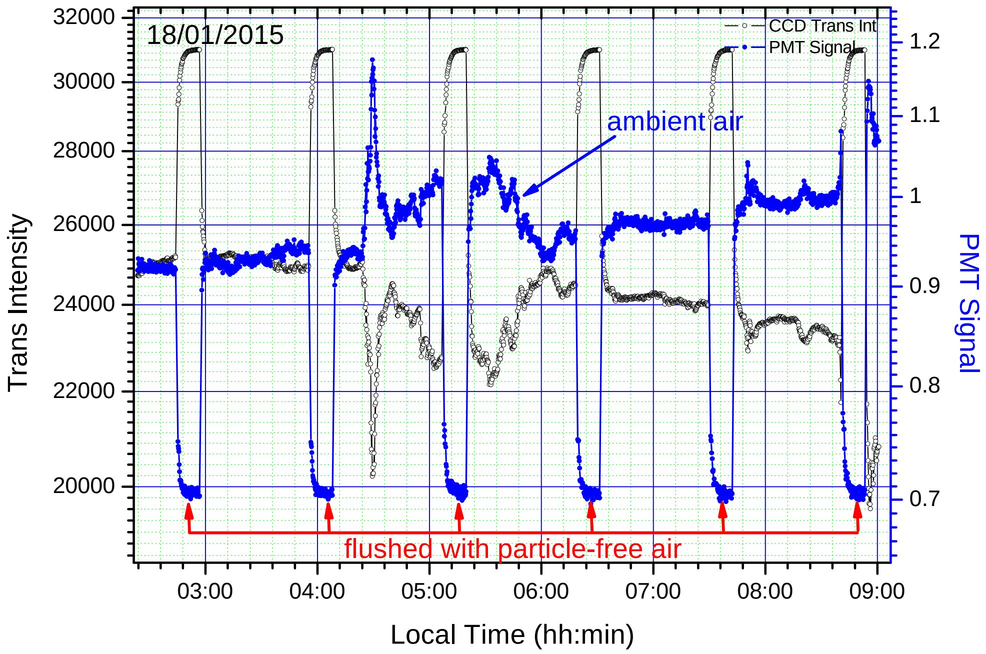


Figure S1 Example data of the transmitted intensity measured with the CCD spectrometer and the scattering signal measured with the PMT of the cavity-enhanced abldoemeter during the experimental period.

2. The introduction is well-organized and well-written in five paragraphs. In contrast, Section 4 (Results and discussion) and Section 5 (Conclusions) are long and contain too much detail. The paper would be strengthened if the authors edited sections 4 and 5 to shorten their discussion to the most important points and eliminate repetition.

Sections 4.2 and 4.3 have been merged in the revised manuscript and Sections 4 and 5 were shortened in the revised manuscript.

3. The authors use the IMPROVE algorithm (Pitchford et al 2007) for comparison to their measurements. The IMPROVE algorithm predicts optical extinction at 550 nm, and is not directly comparable to measurements at 470 nm. I do not understand how the authors correct for this. They state (pg. 33686, lines 14-18): "One point should be kept in mind that the above discussion of the IMPROVE algorithm was suitable for the reconstruction of atmospheric aerosol extinction at lambda = 550nm. In this study, the optical properties of PM1.0 were measured at lambda = 470nm. The IMPROVE algorithm need further improve to well represent the chemical apportionment of light extinction for PM1.0 particles."

Under the assumptions of the IMPROVE algorithm (in which particles are treated as separate entities), the difference in the wavelength will only affect the dry mass extinction efficiency (MEE) terms. We have modified the MEE terms (equation 3 in the discussion paper) of each individual particle components by multiplying a scaling factor to be directly comparable to measurements at λ = 470 nm. The scaling factor ($Scaling\ factor_j = MEE_{j,\lambda=470nm} \big/ MEE_{j,\lambda=550nm}$) was the ratio of the MEEs of each species at λ = 470 nm and λ = 550 nm calculated with the literature reported complex refractive index and the measured mean number size distribution with Mie theory.

4. The IMPROVE algorithm is a simplified prediction of extinction that is intended to be used when no size distribution measurement is available. The authors have the necessary information to directly calculate aerosol extinction, scattering, and absorption using Mie theory. The Mie calculation assumes that the aerosol are spherical and well-mixed, but it would be more accurate than the IMPROVE algorithm. The approach would be to use the the AIM or ISORROPIA model to determine salt concentrations from the measured ion concentration. The salt concentrations, their density and refractive index values from literature (see values in Hand and Kreidenweis, 2002, AST), and the measured size distribution would then be used with Mie theory to directly calculate aerosol absorption, scattering, and extinction.

For the optical closure between the measured and calculated total aerosol extinction coefficients, the reviewer is correct that the Mie theory calculation with the salt concentrations, their density and refractive index, and the measured size distribution provides a more accurate method (Washenfelder, et al., Geophys. Res. Lett., 42, 653-664, 2015). The reason why we chose the IMPROVE method for our data analysis is that we did not have accurate information on the fractional contribution of the chemical compositions to the aerosol extinction owing to the lack of quantitative information about the size distribution of each chemical component (Cheng, et al., Atmos. Environ. 42, 6351-6372, 2008). The IMPROVE algorithm, although it is a simplified predictor of extinction, is nevertheless a useful tool to estimate the contribution of each particle component to haze levels and the relative magnitude of haze contribution by the various particle components (Pitchford, et al., J. Air & Waste Manage. Assoc., 57, 1326-1336, 2007).

5. The authors have data on aerosol optical properties and size distribution with high time resolution. In contrast, the filter composition was measured every 12 h and the classifications of "clear", slightly polluted", and "polluted" are based on a 24 h government definition. Interpreting the high time resolution data within simplistic 24 h classifications and only looking at a few days means that it is difficult to draw conclusions. Are any high time resolution gas-phase measurements available? This would open two analysis possibilities: 1) Individual air quality index could be calculated at higher time resolution (using SO_2*,* NO_2*,* O_3*, CO measurements); or 2) The aerosol optical properties could be correlated with tracer species to identify the likely sources.*

We thank the reviewer for this suggestion. We added the $PM_{2.5}$, SO_2, NO_2, O_3, and CO data with a time resolution of 1 hour to the selected high time-resolution data (Figure S3), which provide supporting evidence for the air pollution sources analysis in Section 4.1.

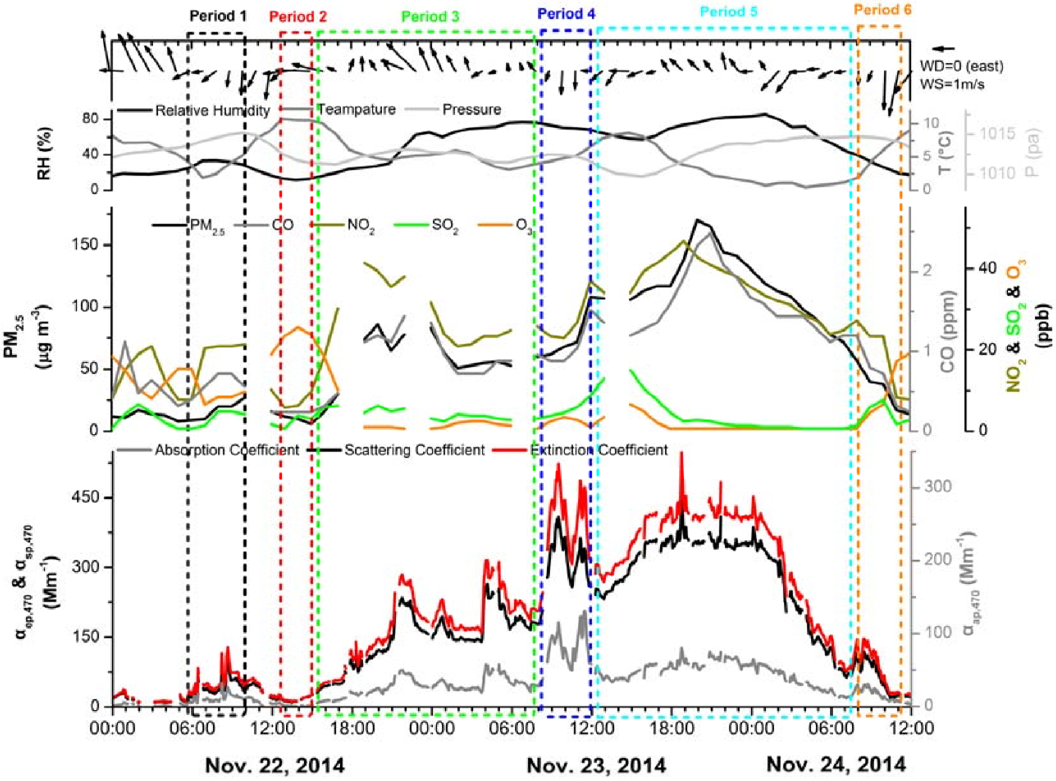


Figure S3. Temporal wind direction and speed, temperature, relative humidity and pressure of atmosphere air, $PM_{2.5}$, CO, NO_2, SO_2 and O_3 concentrations, and the measured aerosol extinction, scattering, and absorption coefficients during the air pollution episode.

We did not use other individual air quality index for the data analysis because the classification of the pollution level based on the mass concentration of $PM_{2.5}$ is more often used to explore the differences of the content of chemical component, aerosol optical properties and meteorological conditions (Che, et al., Atmos. Chem. Phys., 14, 2125–2138, 2014; Zheng, et al., Atmos. Chem. Phys., 15, 2969–2983,2015; Tan, et al., Atmos. Environ., 131, 196-208, 2016).

Other Comments

How often was the particle size distribution measured?

We have added the time in the revised manuscript.

> The particle size distribution between 14 and 662 nm was measured every 3 min with a scanning mobility particle sizer.

Pg. 33679, line 4: Change "higher than 500 μg m^{-3}*" to "higher than 500 μg* m^{-3} *for Beijing"*

DONE.

Pg. 33680, line 18: Add reference for MODIS data.

DONE. This reference has been added in the revised manuscript.

Chu, D. A., Kaufman, Y. J., Zibordi, G., Chern, J. D., Mao, J. T., Li, C. C., and Holben, B. N.: Global monitoring of air pollution over land from the Earth Observing System-Terra Moderate Resolution Imaging Spectroradiometer (MODIS), J. Geophys. Res.,108(D21), 4661, 2003.

*Pg. 33680, lines 23-24: "The inlet consisted of a PM1.0 ambient size cut (SF-PM1.0, 1.0*m^3 h^{-1}*, Seven Leekel Ingenieurburo GmbH), allowing only particles with an aerodynamic diameter smaller than 1 μm to enter the sampling line." Is this correct or does the inlet have a 50% cut-point at 1.0 μm?*

We have modified this statement in the revised manuscript.

> The inlet consisted of a $PM_{1.0}$ ambient size cut (SF-$PM_{1.0}$, 1.0 m^3h^{-1}, Sven Leckel Ingenieurburo GmbH) with a 50% cut-point at 1.0µm.

Pg. 33683, line 17: Add a definition of the Ambient Air Quality Index 633-2012. My understanding is that it is the highest of six pollutant values, but only PM2.5 is mentioned here.

Air Quality Index (AQI) is a scale designed to help understand the impact of air quality on health. The AQI level is based on the level of six atmospheric pollutants, namely SO_2, NO_2, PM_{10}, CO, O_3 and $PM_{2.5}$ measured at the monitoring stations. The individual AQI is assigned to the level of each pollutant and the final AQI is the highest of the six values. As particle pollution was usually the primary pollutant of atmospheric environment in China, we selected $PM_{2.5}$ concentration as the representer of IAQI.

We have modified this section as follows:

> In this work, the $PM_{2.5}$ pollution level was divided into three categories according to the technical regulation on Ambient Air Quality Index (National

> Environmental Protection Standard of the People's Republic of China, HJ 633–2012) (http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/jcg_fbz/201203/W020120410332725219541.pdf; A. Zhang et al., 2013; G. J. Zheng et al., 2015): clear day ($PM_{2.5}$ concentration ≤ 35μg m^{-3}, when the corresponding individual air quality index (IAQI) of 24 hours averaged $PM_{2.5}$ concentration ranged from 0 to 50), slightly polluted day (35μg m^{-3}<$PM_{2.5}$ concentration ≤115 μg m^{-3}, when IAQI of $PM_{2.5}$ ranged from 50 to 150),and polluted day (115 μg m^{-3} <$PM_{2.5}$ concentration ≤ 350 μg m^{-3}, when IAQI of $PM_{2.5}$ ranged from 150 to 400).

Pg. 33684, lines 13-14: "The CRI is one of the intensive optical properties of atmospheric aerosols, and determined by the aerosols' size, shape, mixing state and chemical composition." CRI depends only on chemical composition of a material. It does not change with size or shape or mixing state.

We have modified this statement.

> The effective CRI is an effective property that averages over the aerosols' size, shape, mixing state and chemical composition.

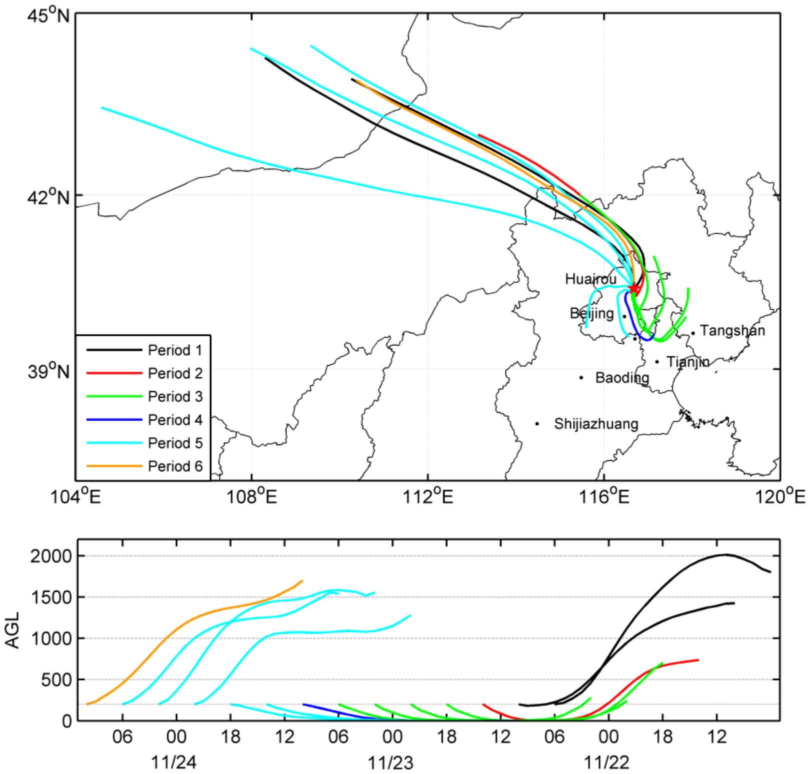
Pg. 33685, lines 10-12: "Projections of the contour lines (with a contour value of 2.298)on the n and k plane gave the standard errors Δn and Δk, respectively (Dinar et al., 2008;Zhao et al., 2014)."This statement is unclear.

We have modified this statement.

> **A contour plot of χ^2 versus *n* and *k* was used to estimate the standard errors of *n* and *k*.** The values of *n* and *k* that satisfy $\chi^2 < \chi_0^2 + 2.298$, which fell within the 1σ error bound of the best measurement (with 68% confidence level of χ^2 distribution), were considered acceptable. Projections of the contour lines (with a contour value of 2.298) on the *n* and *k* plane gave the standard errors Δn and Δk, respectively (Dinar et al., 2008;Zhao et al., 2014).

Pg. 33689, lines 14-19: It is very difficult to match the back trajectories to the periods of interest. Change Fig. 4 to show back trajectories for the highlighted periods.

DONE. We have recalculated the trajectories with 4 hours resolution during the selected air pollution episode.



Pg. 33693, lines 13-27: This general discussion is common to all of the six periods. It should be moved to Section 4.1.

DONE.

Change "Seven Leekel" to "Sven Leckel" throughout.

DONE.