**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

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*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).

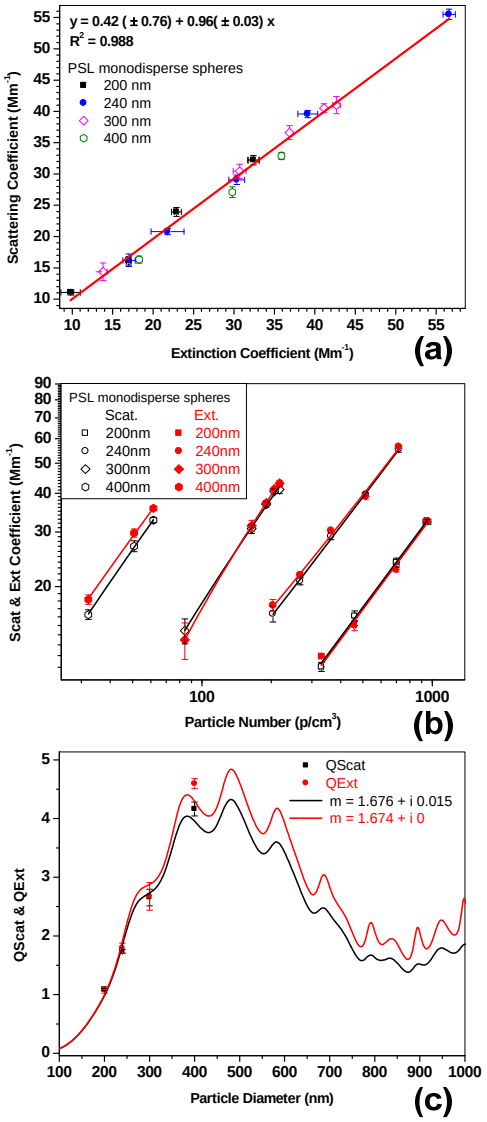



**Fig. 8. in Reference (Zhao et al., 2014a) (a)** Regression plot of the measured extinction and scattering coefficients, **(b)** scattering and extinction coefficients as a function of particle concentration, and **(c)** the scattering ($Q_{Scat}$) and extinction ($Q_{Ext}$) efficiencies as a function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

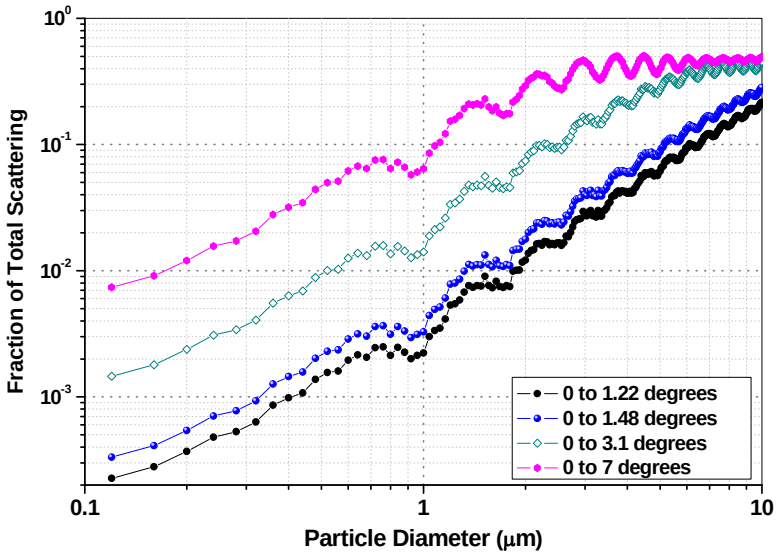



**Fig. 3 in Reference (Zhao et al., 2014a).** Size dependence of the truncated fraction of total scattering under different truncation angles: (1) 0–1.22°, calculated with $d_e = (d - d_0)/2$; (2) 0–1.48°: with $d_e = (L - d_0)/2$; (3) 0–3.1°: without truncation reduced tubes; and (4) 0–7°: for the used 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 $\lambda = 470$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.

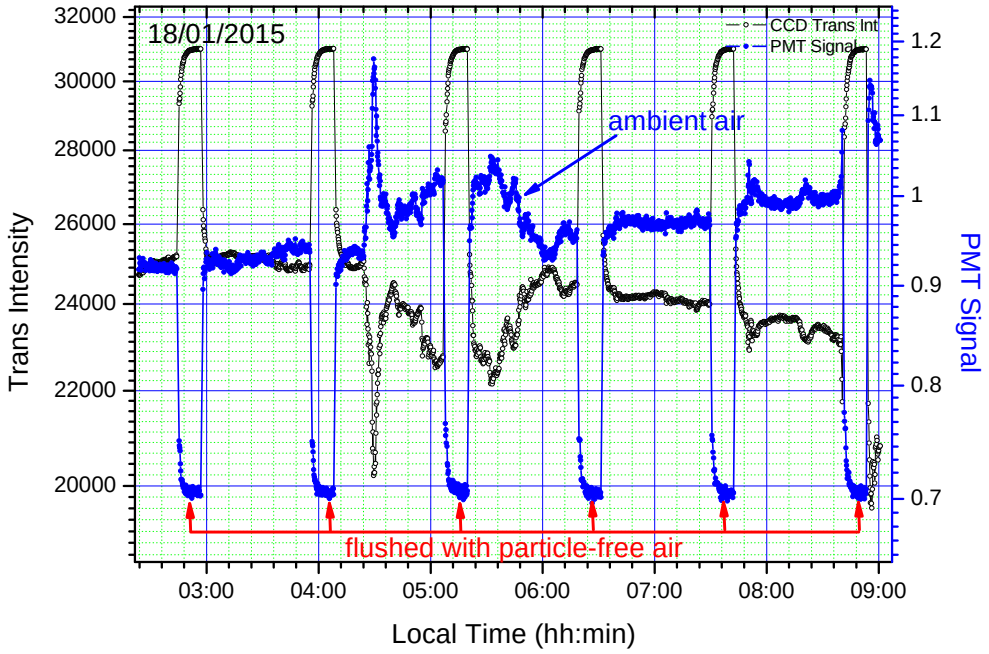



Figure S1 Example data of the transmitted intensity measured with the CCD spectrometer and the scattering signal measured with the PMT of the cavity-enhanced abldoemeter during the experimental period.

*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 ( $Scaling\ factor_j = MEE_{j,\lambda=470nm} / MEE_{j,\lambda=550nm}$ ) 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.

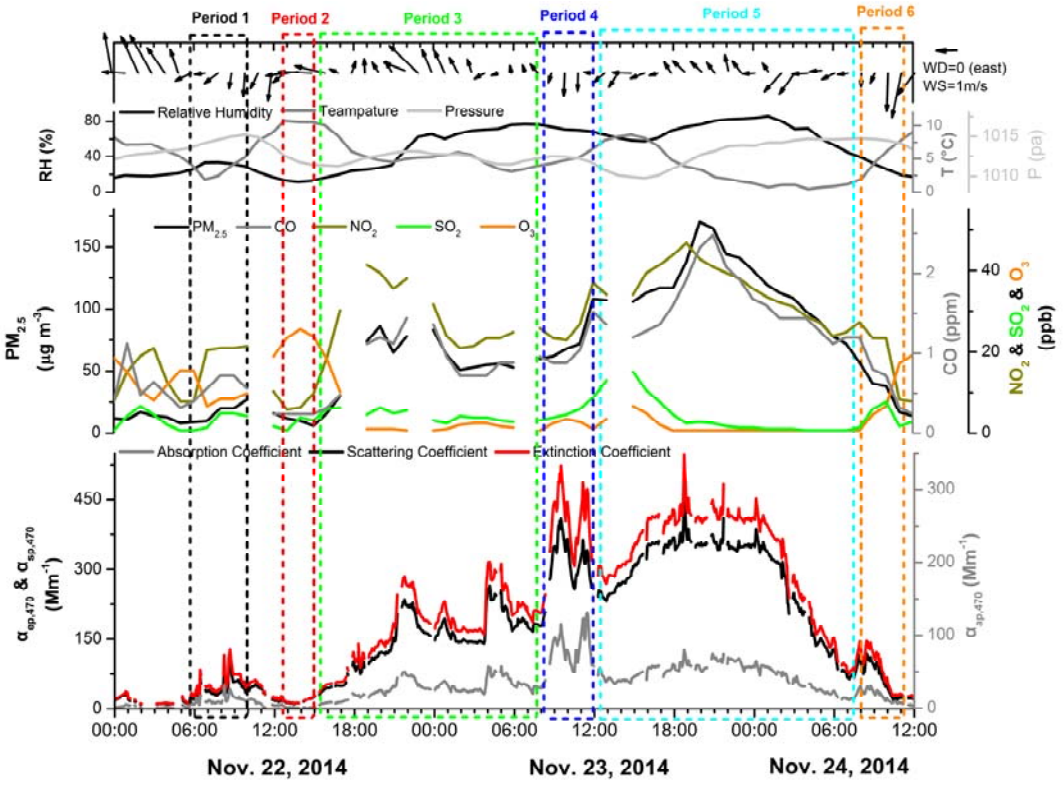



Figure S3. Temporal wind direction and speed, temperature, relative humidity and pressure of atmosphere air, $PM_{2.5}$, CO, $NO_2$, $SO_2$ and $O_3$ concentrations, and the measured aerosol extinction, scattering, and absorption coefficients during the air pollution episode.

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 $\leq$ 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.

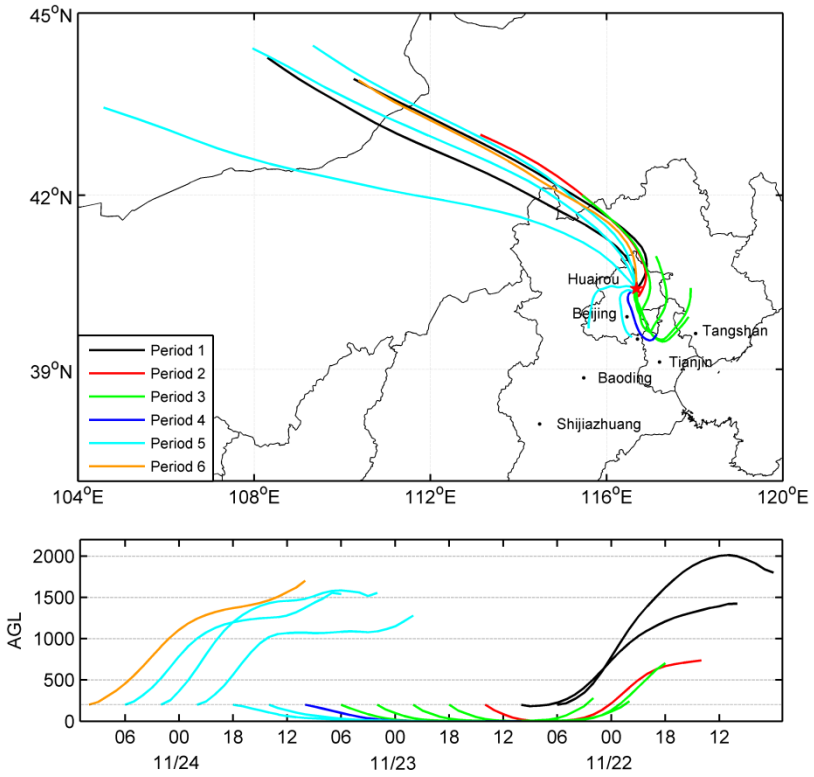
*Pg. 33685, lines 10-12: "Projections of the contour lines (with a contour value of 2.298)on the n and k plane gave the standard errors Δn and Δk, respectively (Dinar et al., 2008;Zhao et al., 2014)."This statement is unclear.*

We have modified this statement.

**A contour plot of $\chi^2$ 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 $\chi^2$ 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 Δ*n* and Δ*k*, 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.