

## Anonymous Referee #1:

We sincerely appreciate for your time and attention on our paper. The comments and suggestions you gave are very helpful for us to improve our paper. We now present point-by-point replies (in black) to all your comments (in green) in this response and the corresponding changes in the revised manuscript have been highlighted in blue.

### ● General Comments:

- 1. The aerosol inlet system on the aircraft should be discussed in the manuscript. It strong related to how to evaluate the accuracy of airborne aerosol measurement.*

**Reply:** Thanks for your comment. We have added some detailed description about aerosol inlet system in Section 2.2 as “The conical double diffuser aerosol inlet, designed for a Twin Otter, was installed on the Y-12. This inlet described by Hegg et al. (2005) has been used extensively on the UMD Cessna 402 (Brent et al., 2014) manufactured by the Droplet Measurements Technologies (MP-1806-A and MP-1807-A, Boulder, CO). The passing efficiency is expected to be near 100% for particle diameters up to 2.5  $\mu\text{m}$  and near 50% for particles between 3-4  $\mu\text{m}$  (Huebert et al., 2004; McNaughton et al., 2007).”

- 2. SSA is a very sensitive and unstable parameter. Authors should provide the detail descriptions on how to calculate the columnar SSA from airborne measurement data and their uncertainties? When different aerosol types or multi aerosol layer in vertical, how authors apply to the calculation?*

**Reply:** Thanks for your comment. We have added some detailed description about the calculation of columnar SSA in Section 3.1 as “The method we calculated the columnar SSA from airborne measurement data and estimation of its uncertainties were based on previous studies (Leahy et al., 2007; Schafer et al., 2014). The SSA ( $\omega_{\text{sample}}$ ) was calculated from  $\sigma_{\text{sca}}$  measurements at 450, 550, and 700 nm measured by a Nephelometer and  $\sigma_{\text{abs}}$  measurements at 565nm by a PSAP. The small mismatch in the wavelength around 550nm was corrected by linearly extrapolating the  $\sigma_{\text{abs}}$  values. We assumed that *in-situ* SSA measured between minimum and maximum flight altitudes represents the entire column. In order to compare a column SSA ( $\omega_{\text{column}}$ ) value with AERONET measurements, the sampled SSA values were averaged for duration of the profile sampling after weighting the values according to aerosol loading. In this study, aerosol loading could represent by  $\sigma_{\text{sca}}$  values and the profiles were limited to those samples collected from below 400 m AGL and continued to greater than 2000 m that may adequately represented the column value. For every profile, SSA data was weighted by the normalized magnitude of  $\sigma_{\text{sca}}$  using the following equation as given below:

$$\omega_{column} = \frac{\sum_{i=0}^N \left[ \frac{\sigma_{sca}}{\sigma_{sca(profile\ mean)}} * \omega_{sample} \right]}{N} \quad (1)$$

N equals the number of  $\omega_{sample}$  in the profile.”

Considering the vertical distribution of  $\sigma_{sca}$  and  $\sigma_{abs}$ , the SSA measured at higher altitudes (lower aerosol loading) is substantially less than in the lower troposphere or the aerosol enrichment layer. Thus, the weighted mean method was better than a simple average of  $\omega_{sample}$ , which would over-represent the absorption features of aerosol that has negligible effect on radiation at the surface where the CIMEL radiometer is located.

**3. Expanding the discussion on the Table 3. Why only 28 May, 2016 case shown in the table? Maybe the scatter plot of AERONET SSA vs Aircraft in situ for each flight can provide more strong conclusion.**

**Reply:** The discussion of Table 3 has been expended in the revised manuscript as the following: “These SSA values obtained in the NCP are lower than those observed in Africa and in northeast United States. The reason for the difference is probably due to different type of aerosol components in these different locations: primarily mineral dust aerosols in Africa and photochemically produced secondary aerosols in Northeast US, and the mixed of these two types of aerosols in the NCP.”

Due to weather conditions, the restriction of airspace, and the working status of CIMEL radiometer, it was very difficult in acquiring coincident surface and aircraft measurements, even with multiple flights on 10 different days. By given the thresholds used for temporal and spatial matching during the experiment period, only the case in 28 May matched the time and space qualification.

**4. How to calculate AOD for different layers (Fig 5) should describe in the manuscript.**

**Reply:** We have included the following description in Section 3.2 in the revised manuscript:

“During the experimental period, the majority of aerosol layers were well characterized by the sampled vertical profiles and most aerosols reside below the maximum flight levels. Mie theory was applied to calculate the extinction profiles and the AOD, and to estimate the impact of different aerosol vertical distributions on these optical properties. The AOD can be calculated by integrating the extinction coefficient over height as:

$$AOD_{(z1 \sim z2)} = \int_{Z_1}^{Z_2} \sigma_{ext}(z) dz \quad , \quad (7)$$

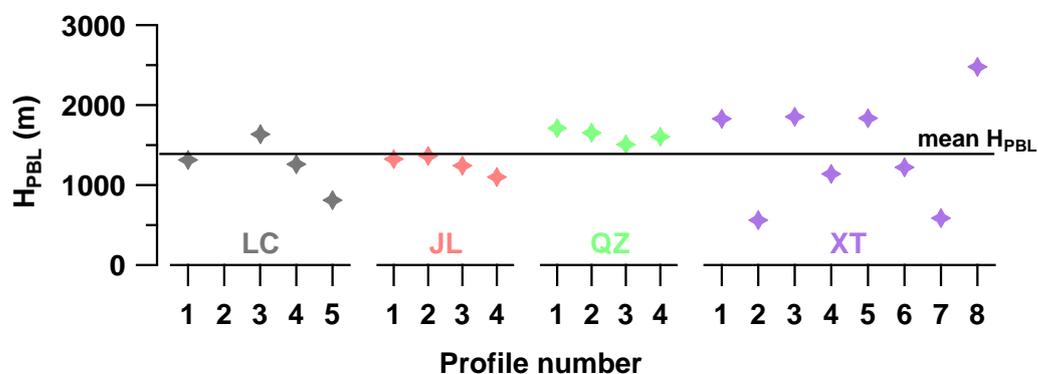
Where the  $\sigma_{sca}(z)$  is the extinction coefficient at a height of z and  $Z_2$  is above most of the aerosol.”

**5. In figure 5(b), it is not clear how authors define the PBL height? Although in the manuscript, the authors say they determined by aerosol scatter profile (Page 9, line 28), does that quantitatively define PBL height? In figure 5(b), how does**

*authors apply the normalization? Does use 1400m PBL height or individual PBL height from each profiles?*

**Reply:** Thanks for your comment. We have added some detailed description about PBL height in Section 4.2 as “The PBL height is determined by the shapes of  $\sigma_{\text{sca}}$  vertical profiles. When the pollution in the lower layer of the troposphere, the magnitude of  $\sigma_{\text{sca}}$  increased slightly with height until a layer where  $\sigma_{\text{sca}}$  sharply decreased. In this study, the mean decreasing rate is about  $0.81 \text{ Mm}^{-1} \text{ m}^{-1}$ , We defined the bottom of this layer as PBL height ( $H_{\text{PBL}}$ ).”

We did the normalization by the shapes of  $\sigma_{\text{sca}}$  vertical profiles. Considering the  $\sigma_{\text{sca}}$  gradient, we use individual PBL height from each profile. The  $H_{\text{PBL}}$  calculated (Section 4.2) in the manuscript is shown in Fig. 1 below, and the mean value of  $H_{\text{PBL}}$  is  $\sim 1400\text{m}$ .



**Fig.1** The calculated PBL height of each profile based on airborne observations. The mean value of  $H_{\text{PBL}}$  is  $\sim 1400$  m during the experimental period.

*6. It is not clear why the authors use the scale height (eq 6 and 7) here? Is that only to calculate the mean profile (or Fit mean in Fig 6)?*

**Reply:** The scale height ( $H_p$ ) represents the height when the aerosol is reduced to  $1/e$  of its surface value. We use  $H_p$  as one of parameters to describe a parameterized model of  $\sigma_{\text{sca}}$  distribution.  $H_p$  is determined from airborne observations. It is only calculated in the mean profile of  $\sigma_{\text{sca}}$  in Fig 6.

*7. RH and scattering coefficient have strong correlation. Does mean that the PBL aerosol is well mixed and the different in aerosol scatter is due to aerosol hygroscopicity?*

**Reply:** There is a high correlation between RH and  $\sigma_{\text{sca}}$  under relative dry conditions (both in clean PBL and lower polluted PBL,  $r^2=0.62$  and  $0.95$ , respectively), but not under humid conditions.

We have modified Fig. 6b & 7b in the revised manuscript by separating dry and humid conditions and improved the equations correspondingly. In the clean PBL,  $H_{\text{RS}}$  was proposed by representing a relative stable layer near surface where the vertical variation of  $\sigma_{\text{sca}}$  was not significant. The mean  $\sigma_{\text{sca}}$  under dry condition are slightly

lower than under humid condition in this layer, the difference of  $\sigma_{\text{sca}}$  maybe due to aerosol hygroscopicity. The same can be inferred that the magnitude of  $\sigma_{\text{sca}}$  increased slightly with height in the polluted lower PBL (Fig.2b below), suggesting that aerosol pollutants were well mixed and the difference of  $\sigma_{\text{sca}}$  may be due to aerosol hygroscopicity.

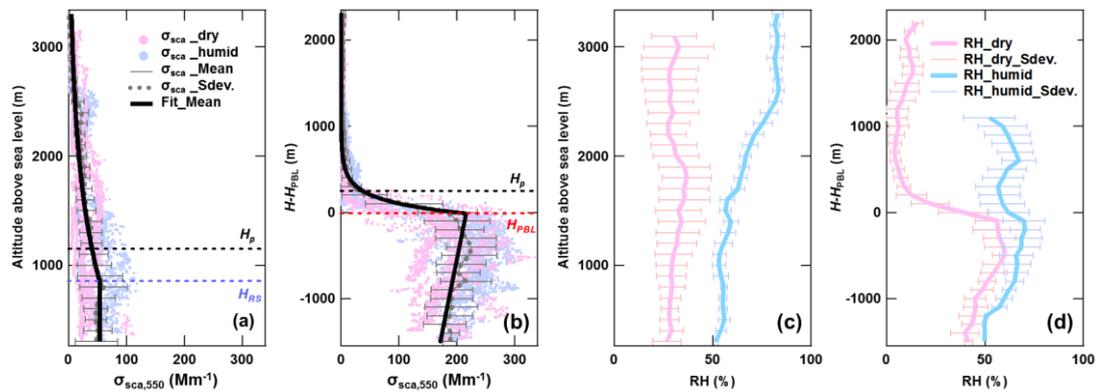


Fig. 2. Mean vertical distributions of  $\sigma_{\text{sca}}$  at 550 nm (in Mm<sup>-1</sup>) and relative humidity (%) during the flight campaign for those cases of (a, c) clean PBL and (b, d) pollution in the lower layer of the PBL where PBL heights have been normalized to the same altitude. Grey dashed lines represent mean  $\sigma_{\text{sca}}$  vertical profiles, the light pink and blue dots represent 1s Nephelometer-measured  $\sigma_{\text{sca}}$ , under dry of humid condition respectively. Thick lines show the calculated fitting curves of the  $\sigma_{\text{sca}}$  profiles (see Eq. 6 and 7). Magenta and blue lines represent RH data collected under dry or humid conditions (c, d). The horizontal error bars represent the standard deviations at every 100 m level.

- **Specific Comments:**

1. *Figure 2a: why the large discrepancy between airborne and sounding data? In the airborne T profile, one can not easily determine the inversion layer.*

**Reply:** The difference between airborne and sounding data could be due to the mismatch of temporal and spatial measurements. Due to the considerations of flight safety and the restricted control of airspace, the research aircraft and the sounding balloons were rarely collocated. In Fig. 2 of the manuscript, there is about 1-1.5 hours interval between aircraft and sounding measured T profiles.

The T profile in Fig. 2 of the revised manuscript is the average of 3 spirals shown in the following figure. Two inversion layers are identified by inspection at ~1000 m and ~2500 m, which matched well with sounding profile. In the T profile in the manuscript, it is hard to determine the inversion layer probably due to the averaged calculation and smoothed curve.

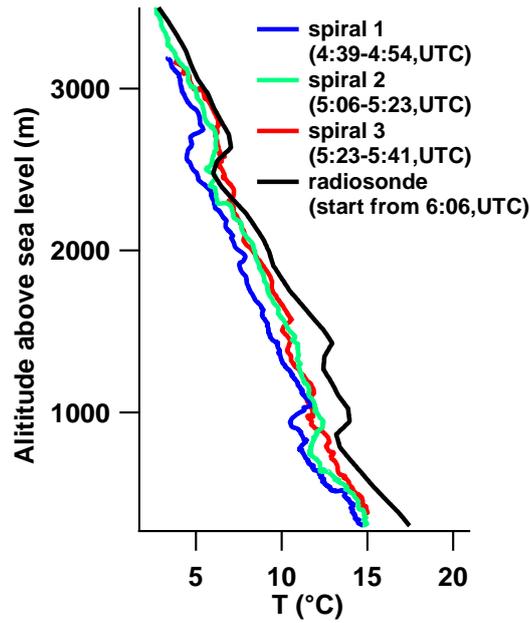


Fig. 3 Vertical profiles of temperature (T) from radiosonde and aircraft measurements made on 8 May 2016 over the Xingtai supersite. Each colored line represents the measurement in one spiral.

### 2. Equation (1): what is the $r$ value?

**Reply:** The  $\gamma$  is an experimentally determined variable of the hygroscopicity, with water uptake increasing with increasing  $\gamma$ . Due to the generally hygroscopic nature of aerosol, there is a change in the scattering coefficient measurement. We carried out a calibration to provide a stable scattering data under humid condition. The  $\gamma$  was determined to be 0.33. The scattering values are adjusted using a correction factor  $f(\text{RH})$ . Details of Equation (1) can be referred the study of Shinozuka et al. (2007) and Beyersdorf et al. (2016).

### 3. Equation (2): what is the $C$ value? $B$ is beta right?

**Reply:**  $C$  is the angular truncation factor. When using the nephelometer for extinction budget studies, correction factors should be applied to account for the effects of angular nonidealities (primarily, the truncation of near-forward scattering). Actual range of total scattering angles captured by nephelometer less than ideal range of  $0^\circ$  to  $180^\circ$ .  $C$  was empirically derived by angstrom exponent as Table 1. Details of Equation (2) can be referred the study of Anderson and Ogren (1998).

Table 1 Parameter adjustments with empirically derived angular truncation correction factors

$\lambda$	Angular Truncation Factor	Angstrom Exponent	Detection Limit
450 nm	$C^{450} = 1.165 - 0.046 \times A_{500}$	$A_{500} = -\log(b_{\text{sca}}^{450}/b_{\text{sca}}^{550})/\log(450/550)$	$4.4\text{E-}07 \text{ m}^{-1}$
550 nm	$C^{550} = 1.152 - 0.044 \times A_{575}$	$A_{575} = -\log(b_{\text{sca}}^{450}/b_{\text{sca}}^{550})/\log(450/550)$	$1.7\text{E-}07 \text{ m}^{-1}$
700 nm	$C^{700} = 1.120 - 0.055 \times A_{625}$	$A_{625} = -\log(b_{\text{sca}}^{550}/b_{\text{sca}}^{700})/\log(550/700)$	$2.6\text{E-}07 \text{ m}^{-1}$

$B_{\text{sca}}$  is measured scattering coefficient, and  $B_{\text{sca\_adj}}$  is the adjusted scattering coefficient (corresponding to  $\sigma_{\text{sca}}$ ) in the following analysis.

**4. Does adjusted Beta sca (Beta sca\_adj) use in the follow analysis? If yes, author should maintain the symbol consistency.**

**Reply:**  $B_{sca\_adj}$  is the adjusted scattering coefficient (corresponding to  $\sigma_{sca}$ ) in the following analysis. Thanks for your comment. We have revised this to maintain the symbol consistency in the manuscript.

**5. Page 5 line 4: for loading? (not clear)**

**Reply:** “for loading” has been deleted.

**6. The equation should better insert in a paragraph, not like equations(4) and (5).**

**Reply:** We have revised this as suggested:

“The measured SSA values were scaled proportionally to the aerosol loading at the altitude of the observation as in the following equation:

$$\omega_{column} = \frac{\sum_{i=0}^N \left[ \frac{\sigma_{sca}}{\sigma_{sca(profile\ mean)}} * \omega_{sample} \right]}{N}, \quad (4)$$

where N equals the number of  $\omega_{sample}$  in the profile.” and “Based on measurements of aerosol scattering and absorption coefficients, aerosol extinction coefficient ( $\sigma_{ext}$ , defined as the sum of  $\sigma_{sca}$  and  $\sigma_{abs}$ ) and SSA ( $\omega$ , defined as the ratio of  $\sigma_{sca}$  to  $\sigma_{ext}$ ) were calculated at 550 nm following Eq. 5,

$$\omega_{\lambda} = \frac{\sigma_{sca, \lambda}}{\sigma_{ext, \lambda}} = \frac{\sigma_{sca, \lambda}}{\sigma_{sca, \lambda} + \sigma_{abs, \lambda}}. \quad (5)$$

**7. Equation (6): what is Hp value used?**

**Reply:** We use scale height ( $H_p$ ) as one of parameters to describe a parameterized model of  $\sigma_{sca}$  distribution.  $H_p$  is determined from airborne observations. We have added the following description: “...and  $H_p$  is the aerosol scale height ( $H_p$  represents the height when  $\sigma_{sca}$  is reduced to 1/e of its surface value);  $H_{RS}$  represent a relative stable layer near surface where the vertical variation of  $\sigma_{sca}$  was not significant. In the cases of clean PBL,  $\sigma_{sca,0}=124 \text{ Mm}^{-1}$ ,  $H_p = 1146 \text{ m}$  and  $H_{RS} = 837 \text{ m}$ .”

**8. Equation (7): What is k value?**

**Reply:** In Section 4.2 of the manuscript, Equation (7) is now Equation (9) and we have defined k as the increasing rate of  $\sigma_{sca}$  in the PBL with a mean value of  $0.03 \text{ Mm}^{-1} \text{ m}^{-1}$  in this study.

**9. Page 10 (lines 5-6): The correlation.....is suitable, please remove since not necessary (not relevant) to discuss here.**

**Reply:** Thanks for your comment. We have revised this as “Under dry condition, there was a pronounced correlation ( $r^2=0.95$ ) between  $RH_{dry}$  and  $\sigma_{sca}$  profiles. But under humid condition, the correlation coefficient was 0.12, which suggest a poor correlation between  $RH_{humid}$  and  $\sigma_{sca}$  profiles.”.

**10. Page 10, line 21: Why affected by the long-range transport?**

**Reply:** We think that the distribution of  $\sigma_{\text{sca}}$  showed in Section 4.3 might but not necessarily be affected by long-range transport and local emissions. The enrichment of aerosol might have two causes. Aerosols could be lifted into the free troposphere (e.g., dust aerosol) and transported over a long distance, altering the aerosol vertical distributions over remote areas (Han et al., 2008). Topographically-generated local circulations can also carry high concentrations of surface air pollutants and change the PBL structure (Chen et al., 2009). Thus, we correct the expression of the manuscript as following:

“The vertically inhomogeneous distribution of  $\sigma_{\text{sca}}$  suggests that aerosol particles in the PBL might be significantly affected by the long-range transport of air pollutants or local emissions around the study area.”

**11. Page 10, line 23: Figure 9c, 9d.**

Reply: Thanks for your comment. This has been corrected in the revised manuscript.

## References:

- Anderson, T. L., and Ogren, J. A.: Determining Aerosol Radiative Properties Using the TSI 3563 Integrating Nephelometer, *Aerosol Science and Technology*, 29, 57-69, 1998.
- Beyersdorf, A. J., Ziemba, L. D., Chen, G., Corr, C. A., Crawford, J. H., Diskin, G. S., Moore, R. H., Thornhill, K. L., Winstead, E. L., and Anderson, B. E.: The impacts of aerosol loading, composition, and water uptake on aerosol extinction variability in the Baltimore-Washington, D.C. region, *Atmospheric Chemistry & Physics*, 16, 1003-1015, 2016.
- Leahy, L. V., Anderson, T. L., Eck, T. F., and Bergstrom, R. W.: A synthesis of single scattering albedo of biomass burning aerosol over southern Africa during SAFARI 2000, *Geophysical Research Letters*, 34, 261-263, 2007.
- Schafer, J. S., Eck, T. F., Holben, B. N., Thornhill, K. L., Anderson, B. E., Sinyuk, A., Giles, D. M., Winstead, E. L., Ziemba, L. D., and Beyersdorf, A. J.: Intercomparison of aerosol single - scattering albedo derived from AERONET surface radiometers and LARGE in situ aircraft profiles during the 2011 DRAGON - MD and DISCOVER - AQ experiments, *Journal of Geophysical Research Atmospheres*, 119, 7439 - 7452, 2014.
- Shinozuka, Y., Clarke, A. D., Howell, S. G., Kapustin, V. N., Mcnaughton, C. S., Zhou, J., and Anderson, B. E.: Aircraft profiles of aerosol microphysics and optical properties over North America: Aerosol optical depth and its association with PM<sub>2.5</sub> and water uptake, *Journal of Geophysical Research Atmospheres*, 112, 1037-1044, 2007.