RESPONSES TO REVIEWER#2 COMMENTS

General Response:

We appreciate the reviewer's comments on the manuscript entitled "Aerosol hygroscopic growth, contributing factors and impact on haze events in a severely polluted region in northern China". All comments are highly valuable and helpful for us to improve our manuscript. We have studied them carefully and have addressed them in the revised manuscript which includes additional investigations. Below are point-by-point responses to the reviewer's comments.

1. Add more descriptions or details on the instruments, e.g, what are measured directly and what are derived, uncertainties in their measured/derived quantities.

Response: The Raman lidar system directly measures atmospheric Mie scattering signals at 355, 532, and 1064 nm, and vibrational Raman scattering signals from H_2O and N_2 molecules at 386 and 407 nm. Aerosol optical properties (aerosol extinction and backscattering coefficient, Ångström exponent, and the depolarization ratio) and atmospheric water vapor mixing ratio profiles can then be derived from this information. The errors of all parameters used in this study are now given in the revised manuscript.

The co-located Doppler lidar (model TWP3-M) emits electromagnetic beams in different directions to the atmosphere above it, then directly receives backscattering signals after those beams interact with atmospheric turbulence. Based on the Doppler effect, this system can derive time series of horizontal wind velocity and direction at a time resolution of 5 min and a range resolution of 60 m below 1 km and 120 m above 1 km. The root-mean-square errors of the Doppler lidar-retrieved wind speed and direction are typically ≤ 1.5 m s^{-1} and $\leq 10^{o}$, respectively.

The GrayWolf six-channel handheld particle/mass meter (model PC-3016A) can directly monitor the total mass concentrations of $PM_{2.5}$ and PM_1 in the actual atmosphere. The minimum detection particle size is 0.3 µm, the counting efficiency for 0.3-µm particles is 50%, and 100% for particle sizes greater than 0.45 µm. Mass spectra are the raw data collected by the ACSM, and standard analysis software offered by Aerodyne Inc. is provided to derive mass concentrations of each chemical component. The uncertainties of ACSM-derived quantities are insignificant (Ng et al., 2011).

In the H-TDMA system, the directly measured distribution function as a function of growth factor can be calculated using number concentrations from condensation particle counter measurements. The TDMA fit algorithm (Stolzenburg et al., 2008) is then used to retrieve the particles' actual growth factor probability density function at RH = 85%. The TDMA fit algorithm assumes that groups in the PDF following one or more lognormal distribution functions (Gaussian shape), thus allowing for the possibility that particles of a given type are not all identical. Uncertainties of these calculated parameters are also insignificant. More details are given by Tan et al. (2013).

More details about the instruments can be found in the revised manuscript.

A brief review of ACSM:

“The non-refractory PM1 (NR-PM_1) chemical components including organics, sulfate, nitrate, ammonium, and chloride were measured in situ by an aerodyne quadrupole aerosol chemical speciation monitor (ACSM) at a time resolution of five minutes. Detailed information about the operation of the ACSM and its application in this campaign can be found elsewhere (Zhang et al., 2018). Briefly, aerosols with vacuum aerodynamic diameters of ~40–1000 nm are sampled into the ACSM through a 100-mm critical orifice mounted at the inlet of an aerodynamic lens. The particles are then directed onto a resistively heated surface (~600°C) where NR-PM_1 components are flash vaporized and ionized by a 70-eV electron impact. The ions are then analyzed by a commercial quadrupole mass spectrometer. Mass spectra are the raw data collected by the ACSM, and standard analysis software offered by Aerodyne Inc. is provided to derive mass concentrations of each chemical component. In this study, the ACSM was calibrated with pure ammonium nitrate following the procedure detailed by Ng et al. (2011) to determine its ionization efficiency. The aerosol aerodynamic particle size was determined by an aerodynamic lens. The uncertainties of ACSM-derived quantities are insignificant (Ng et al. 2011).”

A brief review of H-TDMA:

“The aerosol GF probability distribution function (GF-PDF) at RH = 85% was measured by an in situ H-TDMA. The H-TDMA system mainly consists of a Nafion dryer, a bipolar neutralizer, two DMAs, a CPC, and a Nafion humidifier. The first DMA is used to select monodispersed aerosols with a set mobility size (40, 80, 110, 150, and 200 nm in this study) after the sample is dried and neutralized by the Nafion dryer and the bipolar neutralizer. The selected particles are then humidified when passing through a Nafion humidifier with controlled RH (85%). The second DMA and the CPC are responsible for measuring the number size distribution of the humidified particles. Finally, the TDMA-fit algorithm is used to retrieve GF-PDF (Stolzenburg and McMurry, 2008). Uncertainties of these retrieved parameters are insignificant. More detailed descriptions about the H-TDMA system are given by Tan et al. (2013) and Y.-Y. Wang et al. (2017, 2018).”

2. The four variables representing aerosol hygroscopicity from different aspects, namely the aerosol backscattering enhancement factor [*f*(RH)], the aerosol particle growth factor, the aerosol acidity and the hygroscopicity parameter. To what extent are these variables correlated? What are the correlations among *f*(RH) , and Äÿ and acidity? Under what circumstances? The answers would make the manuscript potentially more useful, e.g. for aerosol modeling. If possible, expand studied RH range, as I understand there is available data (below the selected loft layers and with lower RH) from the measurements.

Response: The aerosol size growth factor (GF) measures the change in particle diameter due to water uptake. Different from GF, *f*(RH) represents the aerosol backscattering coefficient hygroscopic enhancement factor of the aerosol population and is mainly determined by the particle number size distribution (PNSD), chemical composition, and refractive index (Chen et al., 2014). The GF can affect *f*(RH) by changing the aerosol particle size and refractive index. On the one hand, the scattering cross-section of the aerosols will be enhanced due to aerosol size hygroscopic growth. On the other hand, aerosol size hygroscopic growth will reduce the refractive index, so the aerosol scattering efficiency becomes smaller. The relationship between the hygroscopicity parameter (κ) and GF can be expressed as follows:

$$= \frac{(GF^3 \quad 1)(1 \quad a_w)}{a_w} \;, \qquad (1)$$

where a_w is the water activity. The Köhler equation, RH $= a_w \quad S_k$, describes the equilibrium RH for a solution droplet, where S_k is the Kelvin factor. When the particle size is larger than 100 nm, the Kelvin effect can be ignored, and a_w in Eq. (1) can be replaced by RH. Chen et al. (2014) introduced a retrieval method to calculate κ based on in situ measured *f*(RH) and PNSD obtained from the North China Plain. They showed that the *f*(RH) curves elevate as the mean κ value increases. The aerosol acidity is a parameter that can affect aerosol hygroscopic growth, toxicity, and heterogeneity (Iinuma et al., 2004; Yao et al., 2007; Sun et al., 2010). When atmospheric aerosols are acidic, they are more strongly hygroscopic than in their neutralized form (Zhang et al., 2015). To the best of our knowledge, research on the simultaneous correlations among these four variables has not been carried out until now. We will do more detailed correlation studies in the future.

In reference to previous research (e.g., Fernández et al., 2015; Granados-Muñoz et al., 2015; Lv et al., 2017; Bedoya-Velásquez et al., 2018), a simultaneous increase in aerosol backscattering coefficient and RH values is the precondition for determining the range of heights and RHs considered in this study. Data below the selected layers aloft do not fulfill this requirement, so we cannot expand the RH range in this study. We will consider your comment in future work.

3. Figure 3 is an important figure for this manuscript, however I find it is hard to read or draw conclusions with it. And the description of Fig 3. is lack of clarity. Why and how the two cases are selected are poorly demonstrated in the text. Consider adding time series of surface water vapor mixing ratio and RH, as PM and chemical composition data are both obtained at the surface, and the authors are trying to draw some relationships between surface RH and these aerosol data.

Response: We have updated Figure 3. The associated discussion of the figure was also revised (see below).

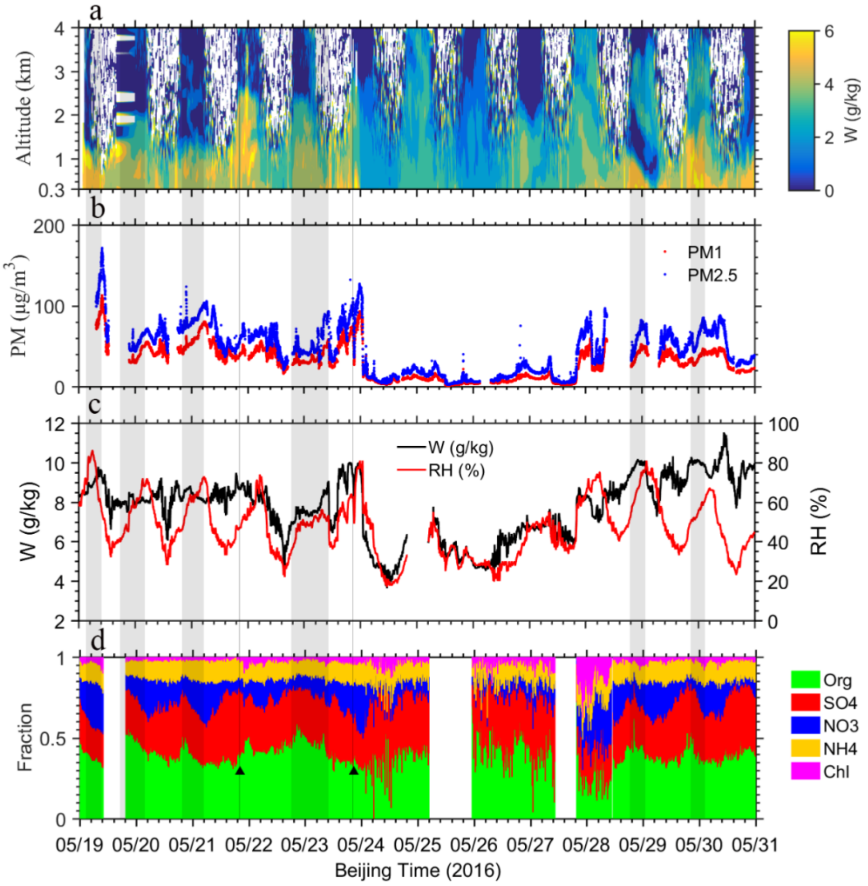

Fig. 3. Time series of (a) water vapor mixing ratio (W) profiles measured by the Raman lidar, (b) mass concentrations of PM_1 (red dots) and $PM_{2.5}$ (blue dots), and (c) surface W (black line) and relative humidity (RH, red line), and (d) chemical species mass fractions of PM_1 measured by the ACSM. Data are from 19–31 May 2016 at Xingtai. The shade grey areas are to enhance the readability of the article. The black triangles in (d) and grey lines in (a, b, c, d) represent the two cases chosen for further examination. Blank parts of the data are missing due to uncontrollable factors such as power supply.

More description of why and how the two cases are selected:

"As W in the lower atmospheric layer and the surface mass concentrations of PM_1 and $PM_{2.5}$ increases, the proportion of organic aerosols decreases (highlighted as shaded grey areas in Fig. 3), suggesting that the proportion of hygroscopic aerosols increased. This shows that strong aerosol hygroscopicity may aggravate air pollution conditions over Xingtai. ""Two instances when this relationship was not seen (highlighted as shaded grey areas in Fig. 3) are shown by the black triangles in Fig. 3d and marked with grey lines across Fig. 3. In the evening of 21 May 2016 (the leftmost triangle and grey line), W and the mass fractions of organics are comparable to those in the evening of

23 May (the rightmost triangle and grey line in Fig. 3). However, the mass concentrations of PM_1 and $PM_{2.5}$ at that time indicated by the leftmost grey line (in the evening of 21 May 2016) are significantly less than that in the evening of 23 May (indicated by the rightmost grey line). The cases occurring on 21 May 2016 (Case I) and 23 May 2016 (Case II) were selected for further study."

4. Page 9. Line 4-8, The authors give absolute errors of Raman-lidar-derived relative humidity and water vapor mixing ratio for a relatively dry case (20%<RH<35%) in Fig 1 and 2. However boundary layers are generally wetter, and the two case selected for the study both occurred under atmospheric environments with RH>80%. What is the error of Raman-lidar derived RH for wet environment? It would be more meaningful to add a relatively wet case for validation. Also what are other uncertainties from Raman Lidar, e.g, AE, depolarization ratio? Is there any difference in uncertainty lower and higher altitudes?

Response: We have added a figure like Fig. 1 but representing a wet environment (the new Fig. 2; see below). Error bars are shown in Fig. 5. It is difficult to determine the uncertainties of the atmospheric aerosol backscattering coefficient, the AE, and the depolarization ratio retrieved by the lidar. This is because they are related to the height, the performance of the lidar system itself, weather conditions, and the properties of aerosols at that time, etc. The height and system performance mainly affect the signal-to-noise ratio of the returned signal. So the uncertainties of these parameters might vary with height.

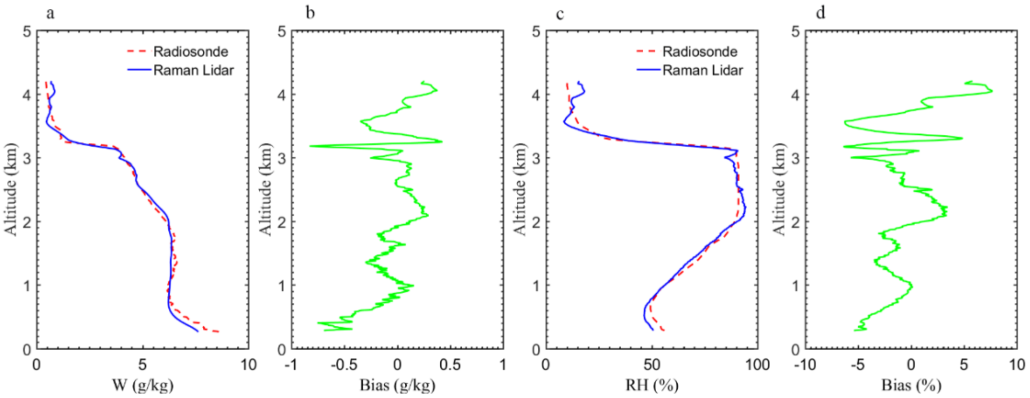

Fig. 2. (a, c) Water vapor mixing ratio (W) and relative humidity (RH) profiles at 2000 BJT 23 May 2016 retrieved by the Raman lidar (blue line) and the radiosonde (red dashed line), respectively, and (b, d) the absolute error in W and RH between the lidar and radiosonde retrievals (lidar minus radiosonde), respectively.

Other comments:

Page 7. Line 10-11, Please specify if the handheld particle/mass meter (PC-3016A) measures dry mass or total mass (including water uptake).

Response: The PC-3016A measures the total mass concentrations of PM_1 and $PM_{2.5}$, including water uptake, not just the dry mass.

Page 11. Line 12-13, "When AV=1.25, 50% of the total sulfate icons in the atmosphere consists of …and 50% consist of…" This sounds definite. Isn't this just a possible combination of different chemical components?

Response: These numbers were calculated using aerosol acidity and the pairing rule (Gysel et al., 2007). When AV=1.25, the mole number of the needed NH_4^+ to completely neutralize sulfate, nitrate, and chloride is 1.25 times than that of the measured NH_4^+. If the mole number of sulfate is 0.625 mol, the NH_4^+ paired with sulfate is 1 mol. The final calculation is: "50% of the total sulfate ions in the atmosphere consist of NH_4HSO_4, and the other 50% consist of $(NH_4)_2SO_4$".

Page 11 line 20, There are two "because" in this sentence, making it awkward.

Response: Fixed.

"The ACSM mainly measures the mass concentrations of SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, and organics. The chlorine ion was not considered because its concentration is extremely low."

Page 13, line 3-5 the whole sentence, starting with "To see if this is the true", reads awkward.

Response: It has been rephrased in the revised manuscript.

Page 13, line 5-7, "As W in the lower atmospheric layer and the mass concentrations of PM1 and PM2.5 increased, the proportion of organic aerosols decreased, suggesting that the proportion of hygroscopic aerosols increased." This relationship is not straightforward by looking at Fig 3. Please think of a way of pointing to the readers where to look, maybe by marking these cases. Also in the next paragraph, two cases are selected. Consider adding two vertical lines across Fig a-b-c) so that data can be better visualized.

Response: The figure and corresponding paragraphs have been revised. See the reply to the third comment for details.

Page 13, line 8-18, This paragraph is related to Figure 3 and is lack of clarity. "this relationship" in the first sentence needs to be explicitly defined. Line 9 "relatively higher", what does it compare to? The first sentence implies the two cases are similar because "this relationship was not seen" in the two instances. However reading along, there seems to be differences for the two cases. What are the similarities and what are the differences for the two cases are not clearly stated in this paragraph. Why are they selected as the studied cases?

Response: Related paragraphs have been revised (see the reply to the third comment).

Page 14, line 4. How and why are these altitude ranges are selected? I understand that the authors choose RH=80% as the reference RH. Why don't use the whole well mixed boundary layer, which can give a wider range of RH? I would be curious to see the hygroscopic growths under a wider range of RH. Would the regression relationships between RH and the various hygroscopic growth factors still be valid? If not, how much deviation there would be? Just thinking from aerosol modeling point of view, the result of this paper is potentially applicable in model parameterizations of aerosol hygroscopic growth if a wider RH range can be studied.

Response: As mentioned above, a simultaneous increase in aerosol backscattering coefficient and RH values is the precondition for determining the range of altitudes and RHs considered in this study (section 3.2 in original manuscript). In reference to previous studies (e.g., Fernández et al., 2015; Granados-Muñoz et al., 2015; Lv et al., 2017; Bedoya-Velásquez et al., 2018), the boundary layer height was not taken into account in the case selection. Your comments are sound, but the experimental data used in this study are limited and preclude doing what you suggest. We will consider your comment in future work.

12. Figure 7. If I understand correctly, the reverse of the slopes of fitted lines would be the Acid Value (*AV*). So maybe consider switch x and y axes for this figure. Then the slopes would be the *AV* with no need to calculate the reverses.

Response: We switched the x- and y-axes in this figure as suggested. However, we think that the original figure looks better.

Page 14, 19-20, "This suggests that aerosol particles were transported to Xingtai from the same source region". Are there local emissions? Can it be excluded?

Response: The description has been revised as follows:

"This suggests that the aerosols in each case were transported into their respective layers at low speeds from almost the same direction. In other words, there is no change in the aerosol type of both cases within the region of interest."

Page 18. Line 5 "a positive result". Please be explicit.

Response: This means that the results between the remotely sensed and in situ measurements are consistent.

Page 20, line 2. I think the authors meant relative humidity by "water vapor content". This may have appeared in other places in the draft. Please don't mix use.

Response: Revised.

Newly Added References:

Chen, J., Zhao, C. S., Ma, N. and Yan, P.: Aerosol hygroscopicity parameter derived from the light scattering enhancement factor measurements in the North China Plain, Atmos. Chem. Phys., 14(15), 8105–8118, doi:10.5194/acp-14-8105-2014, 2014.

Ng, N. L., Herndon, S. C., Trimborn, A., Canagaratna, M. R., Croteau, P. L., Onasch, T. B., Sueper, D., Worsnop, D. R., Zhang, Q., Sun, Y. L. and Jayne, J. T.: An Aerosol Chemical Speciation Monitor (ACSM) for routine monitoring of the composition and mass concentrations of ambient aerosol, Aerosol Sci. Technol., doi:10.1080/02786826.2011.560211, 2011.

Stolzenburg M.R. and McMurry P. H.: Equations governing single and tandem DMA configurations and a new lognormal approximation to the transfer function, Aerosol Sci Tech, 42, 421–432, 2008.

Tan H., Xu H., Wan Q., Li F., Deng X., Chan P. W., Xia D. and Yin Y.: Design and application of an unattended multifunctional H-TDMA system, J. Atmos. Ocean Tech., 30, 1136–1148, doi:10.1175/JTECH-D-12-00129.1, 2013.