RESPONSES TO REVIEWER #1 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.

Major Comments:

Title: Consider an Oxford comma.

Abstract: It appears that the message of the Abstract is that PM caused by anthropogenic activity is more hygroscopic than natural PM, but this does not come across clearly. The English and sentence structure of the entire Abstract require serious revision.

Response: We have rewritten the abstract so that the findings are more clearly stated. The English has also been improved.

Introduction: The introduction should provide a comprehensive overview of why the manuscript is relevant to the community. The central portion of the introduction should be moved to experimental techniques, and more literature survey or relevance should be included in the body of the introduction. For example, what other events lead to haze? Is there any proportion that suggests hygroscopic growth is a minor, moderate, or major contributor to haze – either in general or specifically in the Hebei province? What consequences does this intense haze have, either in Hebei, or elsewhere in the world?

Response: We have extensively revised the introduction by adding a comprehensive overview of why the study is relevant to the community. We have also done a more extensive literature survey about haze events and have added more descriptions about experimental techniques. The latter provides a brief overview of field instruments used to study aerosol hygroscopic growth. Aerosol hygroscopic growth has a major impact on haze events, but there are many other factors leading to haze such as emissions, weather conditions, planetary boundary layer (PBL)-aerosol interactions, and aerosol chemical and physical properties.

A comprehensive overview of why the study is relevant to the community:

"Aerosols, as solid or liquid particles suspended in the air, help regulate Earth's climate mainly by directly scattering or absorbing incoming radiation, or indirectly changing cloud optical and microphysical properties (IPCC, 2013). Many studies suggest that aerosols have a direct impact on human health (Araujo et al., 2008;

Anenberg et al., 2010; Liao et al., 2015; Li et al., 2017). For example, exposure to fine airborne particulates is linked to increased respiratory and cardiovascular diseases (Hu et al., 2015). Atmospheric aerosols can also reduce visibility. Poor visibility is not only detrimental to human health but also hazardous to all means of transportation (Zhang et al., 2010; Zhang et al., 2018)." Therefore, studies on aerosol formation and its influence are important for predicting climate change and improving the human habitat.

Other factors influencing haze:

"Poor visibility is caused by the presence of atmospheric aerosols whose loading depends on both emission and meteorology. The increase in anthropogenic emissions directly affects the formation of haze, such as biomass burning, and factory and vehicle emissions (Watson, 2002; Sun et al., 2006; Q. Liu et al., 2016; Qu et al., 2018). During some major events like the 2008 Summer Olympic Games, drastic measures were taken to reduce emissions which led to a significant improvement in air quality (Huang et al., 2014; Shi et al., 2016; Y.-Y. Wang et al., 2017). This attests to the major role of emissions in air quality. Surface solar radiation and weather such as wind conditions also affect aerosol pollution (Yang et al., 2015). It has been widely known that aerosols interact with the planetary boundary layer (PBL; Quan et al., 2013; Li et al., 2017; Qu et al., 2018; Su et al., 2018). More aerosols reduce surface solar radiation, resulting in a more stable PBL which enhances the accumulation of pollutants within the PBL. Numerous studies have highlighted that the diurnal evolution of the PBL is crucial to the formation of air pollution episodes (Tie et al., 2015; Amil et al., 2016; Kusumaningtyas and Aldrian, 2016; Li et al., 2017; Qu et al., 2018). Besides feedbacks, the stability of the PBL affects the dispersion of pollutants."

Roles of the hygroscopic effect:

"Aerosol hygroscopicity also significantly affects visibility due to the swelling of aerosols (Jeong et al., 2007; Wang et al., 2014). A number of studies have shown that aerosol hygroscopic growth can accelerate the formation and evolution of haze pollution in the North China Plain (NCP; e.g., Quan et al., 2011; Liu et al., 2013; Wang et al., 2014; Yang et al., 2015)."

More information about experimental techniques to measure the hygroscopicity:

"There are many ways to measure aerosol hygroscopicity. A widely used parameter, the aerosol particle size hygroscopic growth factor (GF), is defined as the ratio of the wet particle diameter ($D_{p,wet}$) at a high relative humidity (RH) to the corresponding dry diameter ($D_{p,dry}$). The GF at a certain particle size can be detected by a hygroscopicity tandem differential mobility analyzer (H-TDMA; e.g., Liu et al., 1978; Swietlicki et al., 2008; Y.-Y. Wang et al., 2017). In general, the H-TDMA system mainly consists of two differential mobility analyzer (DMA) systems and one condensation particle counter (CPC). The DMA is first used to select particles at a specific size, and the second DMA and the CPC are used to measure the size distribution of humidified particles. Another instrument known as the differential aerosol sizing and hygroscopicity spectrometer

probe (DASH-SP) can also measure the GF at different RHs (Sorooshian et al., 2008). The DASH-SP couples one DMA and an optical particle size spectrometer (OPSS). The dry size-dependent particles are selected by the DMA, then exposed to different RH environments and finally measured in the OPSS (Sorooshian et al., 2008; Rosati et al., 2015)."

Field campaign and instruments: I would merge this section with the Methodology section. The authors don't explain why the time resolution of the lidar is 15 min, and this may be of interest to the reader and would show that the authors have tailored their use for their study. Do other lidar users (whether they use it form the ground or satellite) use 15 min time resolution? Does it depend on how clean the air is? Is there a relationship between altitude and signal-to-noise, particularly at high altitude bins? Later we discover the AE and depolarization ratio, but they haven't been mentioned in this section. This needs to be explained in greater detail to be suitable for publication. Why are the three wavelengths chosen? What does "atmospheric Mie scattering signals" mean? That phrasing is far too vague. Are the authors hinting that UV telescopes detect Mie scattering, perhaps expected from small water droplets or aqueous aerosols? Based on wavelengths used and expected aerosol size ranges, is scattering supposedly in the Mie regime? These questions should be outlined clearly for the interested reader. Finally, whilst detailed information on the ACSM can be found elsewhere, a brief overview is necessary. Ionization scheme? Quad or TOF detection? Can an ACSM measure refractory chloride? Is this an issue? Etc. Furthermore, why is there no mention to the TDMA yet? P7 Lines 15-16 aren't enough to justify leaving out basic information of the TDMA, especially if critical data has been obtained from it.

Response: We have merged this section with the methodology section in the revised manuscript. In this field campaign, the time resolution was set to 15 min based on original factory settings. These settings have been used in previous studies. A different interval can be used but this would affect the lidar power. The signal-to-noise ratio depends on height. For a ground-based lidar, this ratio is low due to the attenuation of lidar signals. Three wavelengths were chosen to increase our detection ability, noting that there is little choice in the selection of wavelengths in atmospheric lidar applications. This fundamental knowledge was not included in the manuscript to keep the text concise.

We also have added the following new descriptions about the AE and the depolarization ratio.

"The system employs a pulsed neodymium-doped yttrium aluminum garnet laser as a light source and emits three laser beams simultaneously at 355, 532, and 1064 nm with a time resolution of 15 min and a range resolution of 7.5 m based on its original factory settings. … As for the optical receiving unit of this lidar system, optical fiber (OF), dichroic beam splitter (DBS), and ultra-narrowband filters following an ultraviolet telescope divides atmospheric Mie scattering signals and vibrational Raman scattering signals from H_2O and N_2 molecules (at 355, 386, and 407 nm, respectively).

Atmospheric Mie scattering signals at 532 and 1064 nm are divided by OF, DBS and ultra-narrowband filters after a visible infrared telescope. Based on the perpendicular and parallel components at 532 nm received by the lidar system, the aerosol depolarization ratio, a parameter that measures the shapes of aerosols, can be calculated. In general, the more irregular the particle shape, the larger the value of the depolarization ratio (Chen et al., 2002; Baars et al., 2016). The AE can also be calculated using lidar signals at 532 and 1064 nm, which is inversely related to the average size of the aerosols (Ångström, 1964; Tiwari et al., 2016)."

In addition, a brief overview of the ACSM and the H-TDMA was added in this section. 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 ACSM only measures bulk chemical species in PM_1. Also, all measured chemical compositions are non-refractory. According to other studies using a similar method (e.g., Gysel et al., 2007; Wu et al., 2016; Liu et al., 2016; Y.-Y. Wang et al., 2018), chlorine ions are usually ignored when estimating the hygroscopicity parameter using the ZSR mixing rule (Eq. 12 in the original manuscript) because its concentration is always low. Zhang et al. (2018) have found that the mass concentration of chloride is low in Xingtai based on the PM_1 species there.

A brief overview of the ACSM:

"The non-refractory PM1 (NR-PM1) 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^{o}C) where NR-PM1 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 overview of the 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)."

Methodology: As mentioned above, I'd merge this section with the previous one. Whilst the authors have derived equations rather clearly, and the flow of logic is very good in this section, one major comment for this section is to expand on the Fernald method, and to discuss why (7) and (8) are used. What are the advantages and disadvantages of one versus the other? Which one is more commonly used? Is one more accurate for specific conditions or wavelengths than the other? Also, in Section 3.3, no mention as to how NH4+ mass concentrations are obtained, nor any of the other supporting measurements. Why don't the authors rearrange information e.g. in Section 3.4. to here, but more importantly, why do the authors leave out so much information on the ACSM? Also, I got lost in clearly understanding what Case I and Case II are supposed to represent. The authors need to rearrange the writing so that one sentence can describe clearly the difference between the two. As I understand, Case I is a clean day and Case II is a hazy day, and that information is clear in the Abstract but not in the body of the document. The authors also need to draw clearer attention to the fact that Case I and II were chosen on days of similar water vapor content, which is hard to understand from the text.

Response: We have merged this section with the previous one in the revised manuscript. More information about the Fernald method and equations (7) and (8) were added. The dual-parameter fit equation is similar to the single-parameter equation, but with an additional parameter, i.e., a scale factor. In this study, two commonly used parameterized equations were used to verify the consistency of the results. Results from the model that best fit measurement data are shown in the figures. Only the 532-nm wavelength was considered. This is why the equations are not wavelength-dependent. The mass concentration of the measured NH_4^+ was measured by the ACSM. This has been clearly stated in the revised manuscript. We have added more information about the ACSM in the section about instruments. The selection criteria of the cases were rephrased in the revised manuscript, which includes a clear description of the main difference (different pollution conditions) and similarity (same ambient relative humidity) between the two cases.

Description of the Fernald method:

"Here, we use the Fernald method to retrieve AECs (Fernald et al., 1972; Fernald, 1984), which is an analytic solution to the following basic lidar equation for Mie scattering:

$$P_s(z) = ECZ^{-2}[\ _1(z) + \ _2(z)]T_1^2(z)T_2^2(z), \quad (3)$$

where $P_s(z)$ is the return signal, E is the energy emitted by the laser, C is the calibration constant of the lidar system, and $_1(z)$ and $_2(z)$ are the backscattering cross-sections of atmospheric aerosols and molecules at altitude z, respectively. $T_1(z)$ and $T_2(z)$ are the transmittances of aerosols and air molecules at height z."

More description about (7) and (8):

"Finally, a relationship between $f\ (RH)$ and RH was established. The most commonly used equations are the single-parameter fit equation (e.g., Hänel, 1980; Kotchenruther and Hobbs, 1998; Gassó et al., 2000) and the dual-parameter fit equation (e.g., Hänel, 1980; Carrico, 2003; Zieger et al., 2011). The single-parameter fit equation introduced by Hänel (1976) is

$$f\ (RH) = \frac{1\quad RH}{1\quad RH_{ref}}\quad , \tag{8}$$

where γ in an empirical parameter. Larger γ values in this formulation denote a stronger hygroscopic growth.

The dual-parameter fit equation is (Fernández et al., 2015)

$$f\ (RH) = a(1\quad RH)^{-b}. \tag{9}$$

The single- and dual-parameter fit equations are similar, but with an additional scale factor parameter, a, in the case of the dual-parameter fit equation. The parameter b is also an empirical parameter with larger values of b indicating particles with stronger hygroscopicities. In this study, both parameterized equations are used to verify the consistency of the results. The equation that fits the measurement data best is selected."

Selection of aerosol hygroscopic cases:

"How aerosol hygroscopic growth cases were chosen is described here. First, atmospheric mixing conditions were examined using radiosonde-based vertical potential temperature ( ) and W profiles. Cases with near-constant values of and W in the analyzed layer (variations less than 2°C and 2 g kg^{-1}, respectively) represent good atmospheric mixing conditions (Granados-Muñoz et al., 2015). Then aerosol backscattering coefficient profiles at 532 nm were calculated using the Fernald method (see details in section 2.2.1). A simultaneous increase in atmospheric RH and the aerosol backscattering coefficient is also needed, which might indicate aerosol hygroscopic growth (Bedoya-Velásquez et al., 2018). Based on the above criteria, individual cases with the same ambient humidity and different pollution conditions were selected for studying the influence of aerosol hygroscopicity on haze events."

Results and discussion: The major concern here is that no inferences are made, except for the wind directions and the possible airmass sources for the two cases. Describing the results is insufficient for this section.

Response: More inferences have been made in the revised manuscript. More discussion of the results has been added.

-First paragraph of page 14: the authors describe their results and conclude 'good mixing atmospheric conditions' for both cases. They fail to explain why that's important for the measurements though. Please explain why good mixing is necessary, or what does it tell us. Parcel is stable? Important for data retrieval? Is there any mention to boundary layer height? How does dilution affect aerosol load / visibility? Why are heights for Case I and II different?

Response: W and θ variations are less than 2 g kg^{-1} and 2°C, respectively, showing that good mixing atmospheric conditions were present in both cases (Granados-Muñoz et al., 2014). This information suggests atmospheric vertical homogeneity in the layers considered in the study. Moreover, we can infer that the increase in the aerosol backscattering coefficient is caused primarily by the increase in RH in the range of values of interest (Veselovskii et al., 2009; Fernández et al., 2015; Granados-Muñoz et al., 2015; Lv et al., 2017). This is why good mixing is necessary.

Why are the heights for Cases I and II different? A simultaneous increase in aerosol backscattering coefficient and RH values is the precondition for determining where the layer is located. 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.

-First paragraph on page 15: overstated. Also, if the authors conclude the haze is likely (from lidar data e.g., Fig. 4 e, f, k, and l) due to hygroscopic growth, I would bolster my minor comment for Fig. 4 that the data visualization is weak to support this claim / conclusion.

Response: As done in previous studies (e.g., Granados-Muñoz et al., 2015; Lv et al., 2017; Bedoya-Velásquez et al., 2018), what we try to show in this paragraph is that the changes in AE and depolarization ratio gives us more confidence to believe that the increase in aerosol backscattering coefficient with height in each case is primarily due to hygroscopic growth. More details are in the original manuscript on page 14 (lines 1–20).

-Figure 7: there is no decryption on how NH4+ has been predicated.

Response: The mass concentration of NH4+ is predicted using Eq. (10) in the revised manuscript.

"Figure 7 shows mass concentrations of measured ammonium (NH_4) as a function of predicted NH_4 assuming full neutralization of sulfate, nitrate, and chloride during the full day of 21 May 2016 (blue dots, Case I) and 23 May 2016 (green dots, Case II). The solid blue and green lines are the least-squares regression lines for Case I and Case II, respectively. The 1:1 line is shown in red."

-P16 Line 21 – P17 Lines 1-2: I question the validity of this assertion. If I understand correctly, the authors conclude the aerosols in Case II are more acidic than Case I based on the regression slopes in Fig. 7. This is overstated, particularly if it comes only from

one very short time window throughout the campaign. I would challenge it's 'consistent with the results presented'. What are the errors in the slope?

Response: We conclude that Case II aerosols are more acidic than Case I aerosols based on the regression slopes and the parameter AV for each case. The data used for the linear fitting of the whole day were obtained by the ACSM, and AV was calculated at the closest time of each case. The root-mean-squared errors of the liner regression best-fit lines are 0.63 and 0.48 on 21 May 2016 and 23 May 2016, respectively. The following has been added to the revised manuscript:

"The acidity of aerosols in Case II is greater than that in Case I, suggesting that aerosols in Case II were more hygroscopic than those in Case I. This is consistent with the results presented in section 3.2.1."

I think the results can support the conclusion, but data visualization needs to be clearer and include some error analysis of sorts, whether it be a confidence interval or standard deviation. Finally, equations for kappa evaluation need to be clearly stated with the proper values from ACSM, or clearly mentioned in the text.

Response: In the methodology section, we analyzed the absolute error between the water vapor mixing ratios retrieved by the Raman lidar and the radiosonde. The error bars of the relevant parameters are plotted in Fig. 5. We did not provide the uncertainty of the aerosol backscattering enhancement factor because it relies on the uncertainties of many factors including aerosol properties, ambient RH, hygroscopic growth itself, and so on. The H-TDMA and the ACSM cannot provide an error bar for only one point.

Concerning the κ evaluation, we first calculated the number of moles of SO_4^{2-}, NO_3^-, and NH_4^+ based on their mass concentrations obtained by the ACSM. The mass concentrations and mole numbers of the main inorganic salts in PM_1 [NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$] are then evaluated based on an ion-pairing scheme (Eq. 11 in the original manuscript) and their density values (see Table 1). Finally, the hygroscopicity parameter (κ) was calculated based on the ZSR mixing rule (Eq. 12 in the original manuscript) using moles of all species and their corresponding hygroscopicity parameters (Table 1). This has been clearly stated in the revised manuscript.

Conclusion: No major comments that aren't addressed in the previous section.

Minor Comments:

P2 Line 1: Remove "particles" and make "aerosol" plural.

Done.

P2 Lines 3-4: Rephrase "…the hygroscopic growth effect…" to "…hygroscopic growth…".

Done.

P2 Line 4: What do the authors mean by "contributing factors"?

Response: The "contributing factors" are the main factors affecting the hygroscopic

properties of aerosols.

P2 Lines 4-5: “rich measurements” is poor phrasing in English. The sentence in general should be refined.

Done.

“This study investigates the impact of the aerosol hygroscopic growth effect on haze events in Xingtai, a heavily polluted city in the central part of the North China Plain, using a large array of instruments measuring aerosol optical, physical, and chemical properties.”

P2 Line 5: Include “a city” after the comma.

Done.

P2 Line 6: This may refer to multiple instances throughout the document, but I encourage the authors to double check any improper usage of words. “most serious” Is a very poor word choice, please revise. A possible solution includes “…suffers from persistent haze…”, but words to that effect are encouraged.

Done.

“This study investigates the impact of the aerosol hygroscopic growth effect on haze events in Xingtai, a heavily polluted city in the central part of the North China Plain …”

P2 Line 7: To stay in line with the tense of the Abstract, perhaps change “are” to “were”. Also, was the lidar ground-based or satellite-based? If the lidar was ground-based, please rearrange the sentence or remove “ground-based instruments”.

Response: The lidar was ground-based.

“Key instruments used and measurements made include the Raman lidar for atmospheric water vapor content and aerosol optical profiles, the PC-3016A GrayWolf six-channel handheld particle/mass meter for atmospheric total particulate matter (PM) that have diameters less than 1 μm and 2.5 μm (PM_1 and $PM_{2.5}$, respectively), the aerosol chemical speciation monitor (ACSM) for chemical components in PM_1, and the hygroscopic tandem differential mobility analyzer (H-TDMA) for aerosol hygroscopicity.”

P2 Lines 8-9: Perhaps add ‘PM’ as the acronym of particulate matter? Also, what type of diameter, presumably aerodynamic diameter?

Done.

“… a GrayWolf six-channel handheld particle/mass meter for atmospheric total particulate matter (PM) that have aerodynamic diameters less than 1 μm and 2.5 μm (PM_1 and $PM_{2.5}$, respectively) …”

P2 Line 11: I am not sure what the authors mean by “the evolution”. Are they referring to aerosol growth?

Response: The term “the evolution” means the changes. It has been modified in the revised manuscript.

"The changes in PM_1 and $PM_{2.5}$ agreed well with that of the water vapor content …"

P2 Lines 13-16: I'd describe Case I before Case II and remove the unnecessary colon and replace it with 'of', for example.

Done.

"The lidar-estimated hygroscopic enhancement factor for the aerosol backscattering coefficient during a relatively clean period (Case I) was lower than that during a pollution event (Case II) with similar relative humidity (RH) of 80–91%."

P2 Line 16: Maybe use the plural, 'were'?

Done.

P2 Line 17: Why is 'cases' not capitalized?

Re: It has been modified.

P2 Line 18: What is an aerosol acidity value?

Response: The aerosol acidity (AV) in this study was defined as follows:

$$AV = (2 \cdot SO_4^{2-}/96 + NO_3^-/62 + Cl^-/35.5)/(NH_4^+/18), \quad (11)$$

where SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+ represent the mass concentrations (in μg m^{-3}) of the three species measured by the ACSM. The molecular weights of SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+ are 96, 62, 35.5, and 18. Aerosols are considered "bulk neutralized" if $AV = 1$ and "strongly acidic" if $AV > 1.25$. Details in the section 2.2.3 in the revised manuscript.

P2 Line 22: Why keep both 'aerosol' and 'particle'?

Response: The term has been modified.

P3 Line 2: I'd replace "Under the same water vapor conditions…" with "For similar ambient RH…"

Done.

"For similar ambient RH levels, the high content of nitrate facilitates the hygroscopic growth of aerosols, which may be a major factor contributing to heavy haze episodes in Xingtai."

P3 Lines 2-5: Please rephrase or merge the two sentences using simpler language.

Response: Rephrased. Details in the last response.

P4 Line 2: Again, no need for "aerosol particles", please rephrase to "Atmospheric aerosols"

Done.

P4 Lines 2-4: Reconsider the citation – as well as sentence structure – since what the authors state is well-accepted. If a citation is necessary, one of the classical textbooks

should do. Also, please avoid phrasing like "most important". The sentence could be rephrased, e.g.: "Atmospheric aerosols help regulate Earth's climate, mainly by scattering or absorbing incoming radiation" or words to that effect.

Done.

"Aerosols, as solid or liquid particles suspended in the air, help regulate Earth's climate mainly by directly scattering or absorbing incoming radiation, or indirectly changing cloud optical and microphysical properties (IPCC, 2013)."

P4 Lines 4-6: Again, citation may not be necessary, and both arguments can be collapsed in one sentence and citing a classical aerosol / optics textbook.

Done. Details in the last response.

P4 Lines 8-11: Please review sentence structure, because I'm assuming what the authors wrote is not what the authors mean. Hygroscopic growth is self-explanatory, the result however is that the scattering properties change (I would challenge the authors that it strictly increases scattering efficiency or scattering overall).

Response: The sentence has been removed.

P4 Line 13: Remove "crucial" … as for that matter, the second half of the sentence is a repetition.

Done.

"A number of studies have shown that aerosol hygroscopic growth can accelerate the formation and evolution of haze pollution in the North China Plain (NCP; e.g., Quan et al., 2011; Liu et al., 2013; Wang et al., 2014; Yang et al., 2015)."

P4 Lines 15-16: Please revisit the sentence structure, and rephrase e.g., "…water uptake. This growth can be detected by…"

Response: It has been rephrased.

"A widely used parameter, the aerosol particle size hygroscopic growth factor (GF), is defined as the ratio of the wet particle diameter ($D_{p,wet}$) at a high relative humidity (RH) to the corresponding dry diameter ($D_{p,dry}$). The GF at a certain particle size can be detected by a hygroscopicity tandem differential mobility analyzer (H-TDMA; e.g., Liu et al., 1978; Swietlicki et al., 2008; Y.-Y. Wang et al., 2017)."

P4 Line 15 – P5 Line 18: Please see my major comments. This should belong to the experimental section.

Response: This passage provides a brief overview of field instruments used for studying aerosol hygroscopic growth. We also mention the advantages of using a lidar instead of other instruments to study aerosol hygroscopicity. Among the instruments mentioned, only the H-TDMA and Raman lidar were used in this study. Details can be found in the replies to the major comments.

P5 Line 20: Please rephrase "…gain deeper insights…".

Response: This sentence have been removed.

P5 Line 22: Please rephrase “A specific goal…”.

Done.

“The goal of this study is to further investigate how aerosol hygroscopic growth affects haze events and what are the controlling factors by combining surface and vertical measurements of aerosol optical, physical, and chemical properties.”

P6 Lines 3-4: Rephrase “together with other suites of instruments measuring a variety of aerosol properties.” with “…coupled with supporting measurements.”

Response: This sentence has been modified.

“The goal of this study is to further investigate how aerosol hygroscopic growth affects haze events and what are the controlling factors by combining surface and vertical measurements of aerosol optical, physical, and chemical properties.”

P6 Lines 12-14: Restructure the sentence by beginning with “Raman lidar was Used…”.

Done.

“A Raman lidar was used to analyze the relationship between atmospheric water vapor content and PM_1 or $PM_{2.5}$ mass concentrations, …”

P6 Line 15: This is important and may apply to more instances throughout the document. What does “aerosol optical property” mean? It is far too vague when describing data retrieval. Do the authors mean scattering efficiency? Scattering cross-section? Scattering intensity? Extinction coefficients? Please avoid these generalizations; they are not suited for publication and imply poor working knowledge by the authors.

Response: The “aerosol optical property” means the aerosol extinction and backscattering coefficients, Ångström exponent (AE), and the depolarization ratio. This has been stated in the revised manuscript.

P6, second paragraph: This needs to be revisited. Please see my major comments regarding this section.

Re: It has been revised according to your comments.

P7 Line 1: Change “Collocated” to “Co-located”. Please check throughout.

Done.

P7 Line 4: Perhaps mention that the ascension velocity was ‘typically’ 5-6 m/s?

Done.

P7 Line 5: Again, please change “collocated” to “co-located”.

Done.

P7 Line 17: BJT has already been defined so I’d encourage the authors to be consistent with their acronym usage and replace “Beijing local time” with this acronym.

Done.

P8 Line 10: What does "…signal is affected by radiation…" mean?

Response: In the daytime, the strong background light will reduce the original signal-to-noise ratio of the Raman signal.

P9 Line 5: Please insert "reasonably" such that the phrase reads "…agree reasonably Well…"

Done.

"The W profiles agree reasonably well with an absolute error between them less than 0.5 g kg^{-1}."

P9 Line 8: If the authors state a percent error for relative humidity, why do they state an absolute error for W? Also, in ref. to equations (3) – (5), can the authors be clear why they choose to display two separate figures for W and RH? Why not combine in one figure, or why report two separate ones at all? Is it for lidar retrieval validation? Unless the percent error for RH is the actual units, not a relative (percentage-weighted) error?

Response: The percent error of RH is the actual unit, not a relative (percentage-weighted) error. Figures 1 and 2 have been merged.

P9 Line 11: Delete the first sentence, it's redundant in my opinion.

Done.

P9 Line 16: Could the authors add a little more information regarding the Fernald method for the readers?

Response: We have added some information about the Fernald method.

"Here, we use the Fernald method to retrieve AECs (Fernald et al., 1972; Fernald, 1984), which is an analytic solution to the following basic lidar equation for Mie scattering:

$$P_S(z) = ECZ^{-2}[\ _1(z) + \ _2(z)]T_1^2(z)T_2^2(z), \quad (3)$$

where $P_S(z)$ is the return signal, E is the energy emitted by the laser, C is the calibration constant of the lidar system, and $_1(z)$ and $_2(z)$ are the backscattering cross-sections of atmospheric aerosols and molecules at altitude z, respectively."

P10 Line 8: What is a "hygroscopic parameter"? Are they empirical fits? Do they have a physical meaning? The authors cite some literature, yet do not mention quite exactly what a and b are.

Response: The single- and dual-parameter fit equations are similar, but with an additional scale factor parameter, a, in the case of the dual-parameter fit equation. The parameter b is also an empirical parameter with larger values of b indicating particles with stronger hygroscopicities.

P10 Line 15: Again, vague wording like "…is a key parameter…" should be avoided. Furthermore, is the literature scarce on aerosol acidity and hygroscopic growth? I'd encourage the authors to find more relevant literature to cite.

Response: The sentence has been rephrased, and more relevant literature has been cited.

"Aerosol acidity is associated with aerosol hygroscopic growth (e.g. Sun et al., 2009; Fu et al., 2015; Zhang et al., 2015; Lv et al., 2017)."

P10 Line 16: "…aerosols in the atmosphere tend to be more hygroscopic than their neutralized form…" is awfully similar phrasing to the cited literature, Zhang et al., Environ. Sci. Technol., 2007. Apart from the awfully similar phrasing, isn't there a better reference? Might I suggest, but not limit to, Zhang et al., Atmos. Chem. Phys, 2015 (doi: 10.5194/acp-15-8439-2015)?

Response: This sentence has been rephrased as follows.

"When atmospheric aerosols are acidic, they have stronger hygroscopicities than when in their neutralized forms (Zhang et al., 2015)."

P10 Lines 17-18: Why does high hygroscopicity of aerosols enhance light scattering? Also, remove "particles" and make "aerosol" plural (check throughout). Finally, relate this to P4 Lines 8-11, are the authors being consistent?

Response: It has been rephrased as follows.

"The swelling of aerosols due to hygroscopic growth enhances their ability to scatter solar radiation."

P11 Line 21: Is chloride not considered because its concentration is extremely low or because the ACSM only measures non-refractory chloride? Is this even an issue for a city as inland as Xingtai?

Response: As mentioned above, the ACSM only measures non-refractory aerosol species including non-refractory chloride. According to other studies using a similar method (e.g., Gysel et al., 2007; Wu et al., 2016; Liu et al., 2016; Y.-Y. Wang et al., 2018), chlorine ions are usually ignored when estimating the hygroscopicity parameter using the ZSR mixing rule (Eq. 12 in the original manuscript) because its concentration is always low. Zhang et al. (2018) have found that the mass concentration of chloride is low in Xingtai based on the PM_1 species there.

P12 Line 15: Perhaps "e.g., Tobin et al., 2012"?

Done.

P12 Line 16: I would replace "temporal evolution" with "timeseries".

Done.

"Figures 3b and 3c show the simultaneous time series of the surface mass concentrations of PM_1 and $PM_{2.5}$, and W and RH, respectively."

P12 Lines 17-18: Is this a qualitative inference from the authors, or can they provide a correlation of sorts to support their claim?

Response: Yes, this is a qualitative inference.

P13 Line 3: Please rephrase "…and since…".

Done.

"And Zou et al. (2018) shows that aerosol hygrocopicity is related to aerosol chemical composition over the North China Plain."

P13 Lines 3-4: Citing one source hardly makes hygroscopicity 'highly' dependent on the composition of the aerosol. Please rephrase or support with data.

Done.

"And Zou et al. (2018) shows that aerosol hygrocopicity is related to aerosol chemical composition over the North China Plain."

P13 Lines 5-6: Unclear. Are the authors implying that hygroscopic growth diluted the organic fraction (on a mass basis) detected by the ACSM?

Response: No. Inorganics are the main aerosol components contributing to aerosol hygroscopicity (Liu et al., 2014). The decrease in the organics fraction suggests the increase in the inorganics fraction (hygroscopic aerosols).

P13 Line 17: "…cropped up." is not appropriate. Please change.

Done.

"… but they ignored some unexpected cases behind this positive correlation."

P13 Line 18: Remove "a", and I would challenge Case I and Case II help 'fully' understand the phenomenon.

Done.

"The two unexpected cases that occurred on 21 May 2016 (Case I) and 23 May 2016 (Case II) were selected for further study."

P14 Line 4: Why are these altitude ranges chosen? If the point of the article is to assess haze as a health or visibility issue, wouldn't it make sense to take data below the boundary layer? Or are these heights below the boundary layer?

Response: A simultaneous increase in aerosol backscattering coefficient and RH values is the precondition for determining where the layer is located. 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.

P13 Line 21 – P14 Line 1: Please rephrase, awkward sentencing.

Done.

"Two cases were selected: one on 21 May 2016 (Case I) and the other on 23 May 2016 (Case II) at the closest time of the radiosonde launch time at 1915 BJT."

P14 Lines 2-3: Can the authors explain either now or in the previous section why the AE and depolarization ratio are useful? What do they tell us?

Response: Based on your previous comments, detailed information about the AE and the depolarization ratio has been added to the instruments section. The last paragraph on page 14 of the original manuscript also has a description.

"Based on the perpendicular and parallel components at 532 nm received by the lidar system, the aerosol depolarization ratio, a parameter that measures the shapes of aerosols, can be calculated. In general, the more irregular the particle shape, the larger the value of the depolarization ratio (Chen et al., 2002; Baars et al., 2016). The AE can also be calculated using lidar signals at 532 and 1064 nm, which is inversely related to the average size of the aerosols (Ångström, 1964; Tiwari et al., 2016)."

P14 Line 15: Fix "collocated" to "co-located", as per previous comments, unless the doppler lidar is collocated somewhere? Also, no need to specify again the range of the 'region of interest'.

Done.

P14 Lines 13-20: Lots of repetition, unclear and inconsistent sentence structuring, and improper use of citation, and if the authors wish to keep the citation, a more appropriate description or analysis of said 'source region' is required.

Response: The revised paragraph is as follows.

"Figure 4 shows the time series of the horizontal wind velocity and direction retrieved from the co-located Doppler lidar system. From 1830–2030 BJT, Case I (Fig. 4c) and Case II (Fig. 4d) winds within their respective layers are mainly from the north and northwest, respectively, and have relatively low speeds (< 5 m s^{-1}, Fig. 4a and 4b). 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."

P14 Line 21: Perhaps the authors can use a symbol (abbreviation) for the scattering coefficients, rather than the words?

Done.

"The RH and $_{532}$simultaneously increase with altitude in the Case I (Fig. 5c and 5d) and Case II (Fig. 5i and 5j) layers of interest."

P15 Line 12: Remove "Specifically".

Done.

P15 Line 20: Remove "distinctly".

Done.

P16 Lines 6-7: Is this an accurate representation for aerosols in Xingtai, Hebei, or Northern China overall, or it's just a generalization? I'm not sure in the current state Lines 6-9 are necessary in this manuscript.

Response: It is a generalized description. We have removed these sentences in the revised manuscript.

P16 Lines 12-13: Remove sentence.

Done.

P17 Line 1: Again, please correct “aerosol particles”.

Done.

P17 Lines 19-21: Again, this is overstated. How different are kappa value of 0.557 vs. 0.610? I would encourage the authors to use phrasing like ‘suggests’ or ‘point towards’, rather than definitive conclusions, which I don’t think can be made from the presented data.

Done.

“This suggests that the aerosol hygroscopicity for Case II was higher than that for Case I.”

P17 Line 22: Remove “ion”, fix “aerosol particles”, and remove “significant”.

Done.

“It also suggests that under the same ambient RH conditions, the nitrate content in aerosols can cause differences in the hygroscopicity of aerosols.”

P18 Line 3: Replace “Concerning the aerosol scattering enhancement factor, during the last decade, many…” with “In the last decade, many…”

Done.

“In the last decade, many studies have compared remotely sensed and in situ aerosol scattering enhancement factor measurements using a humidified tandem nephelometer and have shown positive results (Zieger et al., 2011, 2012; Sheridan et al., 2012; Tesche et al., 2014; Lv et al., 2017).”

P18 Line 4: Is the nephelometer an example? Or have all studied used the nephelometer?

Response: To our knowledge, almost all studies have used the nephelometer (e.g., Zieger et al., 2011, 2012; Sheridan et al., 2012; Tesche et al., 2014; Lv et al., 2017).

P18 Line 6: Replace “for use in” with “for”.

Done.

The H-TDMA is also a reliable instrument for measuring the aerosol hygroscopicity due to water uptake (Liu et al., 1978).

P18 Line 7: Please fix “aerosol particles”.

Done.

The aerosols diameter GFs observed by the ground-based H-TDMA at the closest time of each case are examined next.

P18 Line 12: Please fix “aerosol particles”.

Done.

P18 Lines 13-14: Is the ‘kappa model’ supposed to be capitalized?

Response: We used the symbol instead.

Tables and Figures:

Figure 1: The radiosonde line does not look dashed to me, neither does it look dashed in the legend. Please amend how the authors see fit.

Done.

Figure 2: Please keep consistency with data display. Traces should appear like they do in Fig. 1. I don’t know if displaying the difference is useful, unless at those heights where the difference is marked, it implies poor lidar performance? If so, please reflect in the main text, because it is arguable how well they agree (as per main text, P9 Lines 4-5)

Re: Figures 1 and 2 were merged. The biases in Fig. 1 and Fig. 2 (original manuscript) are both absolute errors.

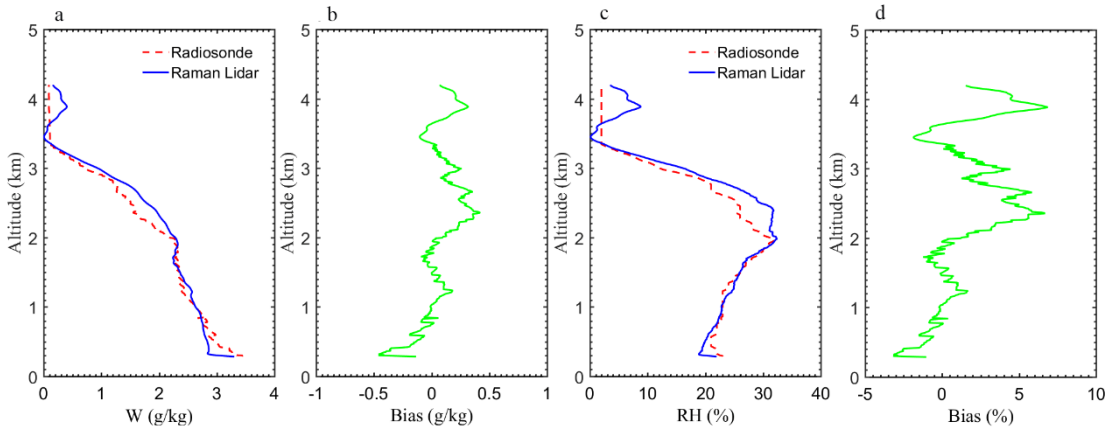

Fig. 1. (a, c) Water vapor mixing ratios (*W*) and RH profiles at 0515 BJT 24 May 2016 retrieved by the Raman lidar (blue line) and the radiosonde (red dashed line), respectively, and (b, d) the absolute error in *W* and RH between the lidar and radiosonde retrievals (lidar minus radiosonde), respectively.

Figure 3: In the caption, please explain the missing data.

Done.

“Blank parts of the data are missing data due to uncontrollable factors such as power supply failures.”

Figure 4: To be consistent with the text, change “Angstrom” to “Ångström”, and I don’t know if the heights for Cases I and II should be reported to 1 decimal place, unless that is instrument precision (appears so e.g., from P14 Line 4)? Finally, I don’t know how impactful this figure is visually if the x-axes are different for Case I and II. I would suggest either keeping x-axes consistent or overlapping the traces for the two cases in one plot. Differences aren’t obvious in the current display.

Response: We have changed "Angstrom" to "Ångström". The accuracy of the height for the instrument is one decimal place. The more important role of Fig. 5 is to determine if the cases meet our prior selection criteria (section 3.2 in original manuscript), as was done in other studies (e.g., Fernández et al., 2015; Granados-Muñoz et al., 2015; Lv et al., 2017; Bedoya-Velásquez et al., 2018). Further research was conducted when there was sufficient reason to believe that the aerosol backscattering coefficient increase with height was primarily due to the increase in RH. Whether the x-axes are consistent does not affect our judgment. In addition, we have updated Fig. 5.

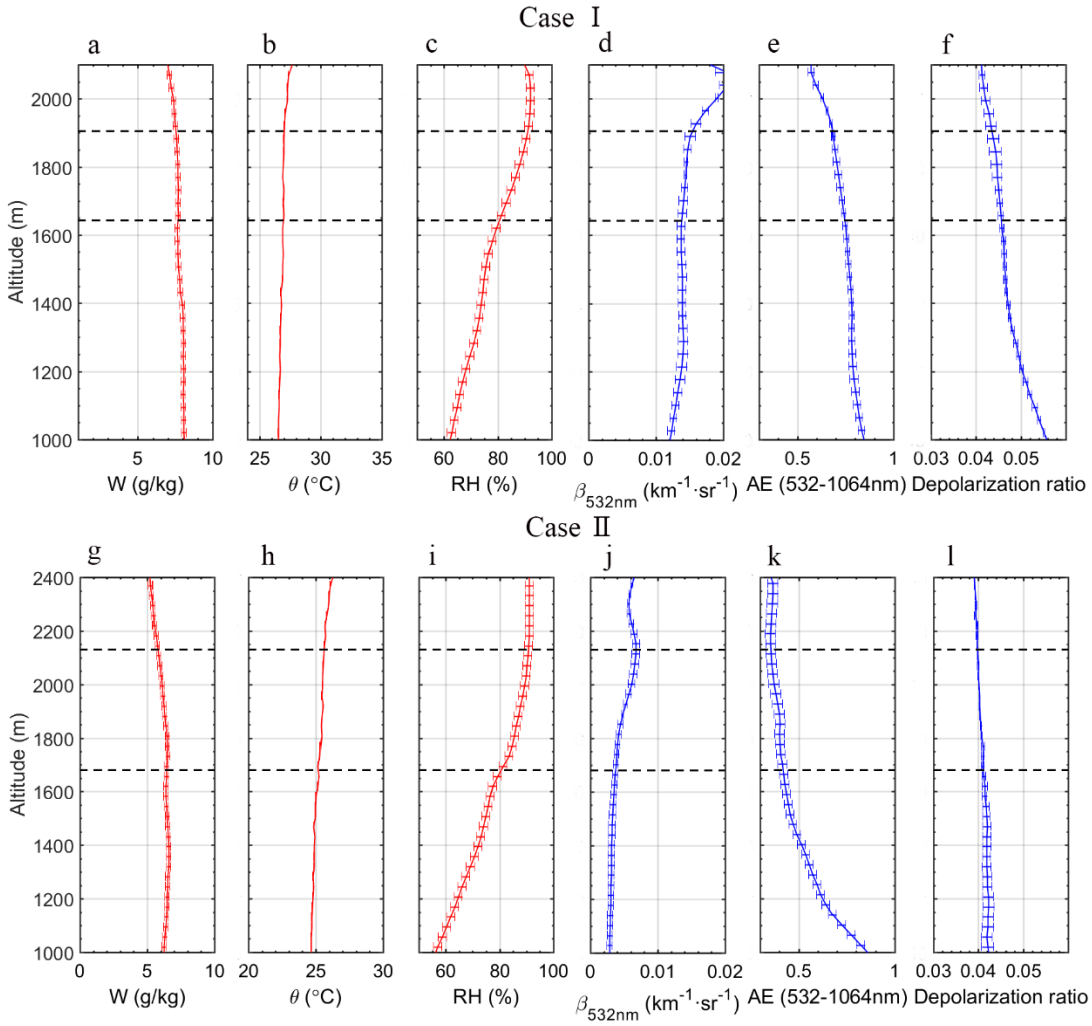

Fig. 5. The vertical profiles of (a, g) water vapor mixing ratio (W), (b, h) potential temperature (θ), (c, i) relative humidity (RH) calculated from radiosonde data, (d, j) backscattering coefficient at 532 nm (β_{532}), (e, k) the Ångström exponent [AE (532-1064nm)], (f, l) depolarization ratio retrieved from Raman lidar data for Case I (top panels) and Case II (bottom panels). Horizontal dashed lines show the upper and lower boundaries of the layer under analysis (1642.5–1905.0 m for Case I and 1680.0–2130.0 m for Case II). Horizontal error bars denote the uncertainty of each property.

Figure 5: No major comments.

Figure 6: No major comments.

Figure 7: No major comments.

Figure 8: No major comments.

Figure 9: Please fix "Aerosol particle", but more importantly, in the caption, explain this is not data, but a model based on Eq. 3 (as per the text, unless I'm mistaken).

Done.

"Fig. 9. Aerosol size hygroscopic growth factor (GF) as a function of relative humidity (RH) for (a) Case I and (b) Case II. The different colors represent different particle sizes (Dp). These are the results of a model based on Eq. 3 from Gysel et al. (2009)."

Tables 1-3: No comments.

Table 5: The results weakly support the conclusions of the document. I would encourage the authors to be more transparent with their data, perhaps in a Supporting Information section. Any simple errors to report, e.g. 95% confidence intervals? How was the raw data from the H-TDMA obtained? A timeseries to serve as example perhaps?

Re: We decided to remove Table 5.

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Aerosol hygroscopic growth, contributing factors, and impact on haze events in a severely polluted region in northern China

Jun Chen[1], Zhanqing Li[1,2,*], Min Lv[3], Yuying Wang[1], Wei Wang[1], Yingjie Zhang[4], Haofei Wang[5,6], Xing Yan[1], Yele Sun[4], Maureen Cribb[2]

[1]State Key Laboratory of Earth Surface Processes and Resource Ecology, College of Global Change and Earth System Science, Beijing Normal University, Beijing 100875, China
[2]Department of Atmospheric and Oceanic Sciences and ESSIC, University of Maryland, College Park, Maryland, USA
[3]School of Geographic Science, Nantong University, Nantong 226000, China
[4]State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[5]College of Resource Environment and Tourism, Capital Normal University, Beijing, 100048, China
[6]State Environment Protection Key Laboratory of Satellite Remote Sensing, Institute of Remote Sensing and Digital Earth, Chinese Academy of Sciences, Beijing, 100101, China

Correspondence to: Zhanqing Li (zli@atmos.umd.edu) and Yuying Wang (wang.yuying@mail.bnu.edu.cn)

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Abstract:

~~The hygroscopic growth of aerosol particles is a key factor of air pollution because it can significantly reduce visibility. In order to better understand~~This study investigates the impact of the aerosol hygroscopic growth effect on haze events ~~and contributing factors, we made use of rich measurements during an intensive field campaign conducted~~ in Xingtai, ~~Hebei province of China that has suffered from the most serious pollution~~a heavily polluted city in the ~~Northern~~central part of the North China Plain~~. Key measurements are from~~ , using a large array of instruments measuring aerosol optical, physical, and chemical properties. Key instruments used and measurements made include the Raman lidar ~~and ground-based instruments such as a~~ for atmospheric water vapor content and aerosol optical profiles, the PC-3016A GrayWolf ~~6~~six-channel handheld particle/mass meter for atmospheric total particulate matter (PM) that have diameters less than 1 μm and 2.5 μm (PM_1 and $PM_{2.5}$, respectively), the aerosol chemical speciation monitor (ACSM~~),~~) for chemical components in PM_1, and ~~a~~the hygroscopic tandem differential mobility analyzer (H-TDMA~~). The evolution of~~) for aerosol hygroscopicity. The changes in PM_1 and $PM_{2.5}$ agreed well with that of the water vapor content due to the aerosol hygroscopic growth effect. Two cases were selected to further analyze the effects of aerosol ~~particle~~ hygroscopic growth on haze events. The lidar-estimated ~~aerosol~~ hygroscopic enhancement factor ~~during a pollution event (Case II) was greater than that~~for the aerosol backscattering coefficient during a relatively clean period (Case I) was lower than that during a pollution event (Case II) with similar relative humidity (RH~~):~~) levels of 80–91%. ~~The hygroscopic growth was fitted by the~~

The Kasten model was used to fit the aerosol particle hygroscopic growth factor whose parameter *b* ~~differ~~differed considerably~~: 0.9346 vs.~~ between the two cases, i.e., 0.1000 ~~for cases~~ (Case I) versus 0.9346 (Case II ~~and I respectively.~~). The aerosol acidity value ~~of~~calculated from ACSM data for Case I (1.35) was less than that for Case II (1.50) ~~was greater than that of Case I (1.35)~~ due to different amounts of inorganics such as NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$, ~~consistent with the difference in the aerosol hygroscopicity parameter κ calculated from the chemical species of PM_1 obtained by the ACSM. Data from the~~ . Model results based on H-TDMA data showed that ~~all of the~~ aerosol ~~particle size~~ hygroscopic growth factors in each ~~particle~~ size category (40, 80, 110, 150, and 200 nm) at different RH levels (80–91%) ~~during~~for Case ~~II~~I were ~~higher~~lower than those ~~during~~for Case ~~I. Under~~II. For similar ambient RH levels, the ~~same water vapor conditions, aerosol~~ high content of nitrate facilitates the hygroscopic growth ~~was one~~ of ~~the~~aerosols, which may be a major ~~factors~~factor contributing to heavy haze ~~pollution. Concerning aerosol chemical composition, nitrate was the primary component contributing to aerosol hygroscopicity over~~episodes in Xingtai.

Key words: Raman lidar; aerosol hygroscopic growth; water content; haze; remote sensing

1. Introduction

~~Atmospheric aerosol~~ Aerosols, as solid or liquid particles ~~are one of~~suspended in the ~~most important components of the atmosphere that play a key role in~~ air, help regulate Earth's climate ~~system,~~ mainly ~~because aerosol particles can~~ by directly ~~and strongly scatter and absorb visible light (Mie, 1908). Moreover, atmospheric aerosol particles can act as cloud condensation nuclei and ice nuclei, a process that might change~~scattering or absorbing incoming radiation, or indirectly changing cloud optical and microphysical properties (~~Twomey, 1977). Atmospheric aerosol particles through hygroscopic growth can influence air quality and visibility (Haywood~~IPCC, 2013). Many studies suggest that aerosols have a direct impact on human health (Araujo et al., 2008; ~~Y. F., Wang~~Anenberg et al., ~~2012; Y. Y. Wang~~2010; Liao et al., 2015; Li et al., 2017). ~~Hygroscopic growth~~For example, exposure to fine airborne particulates is ~~a process whereby the aerosol scattering capacity increases significantly at high relative humidity (RH) levels, leading to enlarged aerosol particles (Covert et al., 1972; Hänel, 1976; Jeong et al., 2007). A recent study on the 2012–2013 winter haze events in Beijing (Tie et al., 2017) showed that atmospheric water vapor plays a critical role in~~ linked to increased respiratory and cardiovascular diseases (Hu et al., 2015). Atmospheric aerosols can also reduce visibility. Poor visibility is not only detrimental to human health but also hazardous to all means of transportation (Zhang et al., 2010; Zhang et al., 2018).

Poor visibility is caused by the presence of atmospheric aerosols whose loading depends on both emission and meteorology. The increase in anthropogenic emissions directly affects the formation of ~~severe haze events in China and~~ haze, such as biomass burning, and factory and vehicle emissions (Watson, 2002; Sun et al., 2006; Q. Liu et al., 2016; Qu et al., 2018).

During some major events like the 2008 Summer Olympic Games, drastic measures were taken to reduce emissions which led to a significant improvement in air quality (Huang et al., 2014; Shi et al., 2016; Y.-Y. Wang et al., 2017). This attests to the major role of emissions in air quality. Surface solar radiation and weather such as wind conditions also affect aerosol pollution (Yang et al., 2015). It has been widely known that aerosols interact with the planetary boundary layer (PBL; Quan et al., 2013; Li et al., 2017; Qu et al., 2018; Su et al., 2018). More aerosols reduce surface solar radiation, resulting in a more stable PBL which enhances the accumulation of pollutants within the PBL. Numerous studies have highlighted that the diurnal evolution of the PBL is crucial to the formation of air pollution episodes (Tie et al., 2015; Amil et al., 2016; Kusumaningtyas and Aldrian, 2016; Li et al., 2017; Qu et al., 2018). Besides feedbacks, the stability of the PBL affects the dispersion of pollutants.

Aerosol hygroscopicity also significantly affects visibility due to the swelling of aerosols (Jeong et al., 2007; Wang et al., 2014). A number of studies have shown that aerosol hygroscopic growth ~~is one of the crucial factors in enhancing heavy haze events.~~

~~The hygroscopic growth factor (GF) measures the change in particle diameter due to water uptake that~~ can accelerate the formation and evolution of haze pollution in the North China Plain (NCP; e.g., Quan et al., 2011; Liu et al., 2013; Wang et al., 2014; Yang et al., 2015). There are many ways to measure aerosol hygroscopicity. A widely used parameter, the aerosol particle size hygroscopic growth factor (GF), is defined as the ratio of the wet particle diameter ($D_{p,wet}$) at a high relative humidity (RH) to the corresponding dry diameter ($D_{p,dry}$). The GF at a certain particle size can be ~~measured~~detected by a ~~humidified~~hygroscopicity tandem differential mobility analyzer (H-TDMA~~) (e.g., Liu et al., 1978; Swietlicki et al., 2008; Y.-Y. Wang et al.,~~

~~2017). The aerosol hygroscopic optical~~ ; e.g., Liu et al., 1978; Swietlicki et al., 2008; Y.-Y. Wang et al., 2017). In general, the H-TDMA system mainly consists of two differential mobility analyzer (DMA) systems and one condensation particle counter (CPC). The DMA is first used to select particles at a specific size, and the second DMA and the CPC are used to measure the size distribution of humidified particles. Another instrument known as the differential aerosol sizing and hygroscopicity spectrometer probe (DASH-SP) can also measure the GF at different RHs (Sorooshian et al., 2008). The DASH-SP couples one DMA and an optical particle size spectrometer (OPSS). The dry size-dependent particles are selected by the DMA, then exposed to different RH environments and finally measured in the OPSS (Sorooshian et al., 2008; Rosati et al., 2015).

The aerosol optical hygroscopic enhancement factor [~~$f(RH)$~~][f(RH)] has ~~also~~ also been employed~~,~~ to investigate aerosol hygroscopicity, which is defined as the ratio ~~between~~of aerosol optical properties (~~e.g.,~~ aerosol extinction~~ coefficient, total,~~ scattering ~~and~~or backscattering coefficients) ~~under wet atmospheric conditions and the corresponding reference value under~~ between wet and dry conditions (Kotchenruther et al., ~~1999). It can be measured by a humidified~~ 1998). Two tandem ~~nephelometer~~ nephelometers are used to measure f(RH) (e.g., Covert et al., 1972; Feingold and Morley, 2003; Titos et al., 2018). One nephelometer measures the aerosol optical properties of dry ambient aerosols at RH < 40%, and another measures that of wet aerosols at different RHs adjusted by a humidifier placed between them (Koloutsou-Vakakis et al., 2001; Titos et al., 2018). MacKinnon (1969) ~~found~~was the first to find that the lidar backscattering signal is affected by ~~the~~ environmental ~~RH level. Subsequent~~RHs. Later studies have demonstrated the ~~use~~possibility of using the lidar ~~for~~

~~observing the~~to observe aerosol hygroscopic growth ~~phenomenon~~ (Tardif et al., ~~2003~~2002; Pahlow et al., 2006; Veselovskii et al., 2009; Di Girolamo et al., 2012; Fernández et al., 2015; Granados-Muñoz et al., 2015; Lv et al., 2017; Bedoya-Velásquez et al., ~~2018; Lv et al., 2017).~~

~~Raman lidar is particularly valuable for measuring atmospheric water vapor by using the vibrational Raman scattering signal from water vapor molecules (H_2O) and nitrogen molecules (N_2) (Whiteman, 2003; Barnes et al., 2008).~~2018). Compared with tandem nephelometers, lidar technology allows for measurements under unmodified ambient atmospheric conditions without drying ambient aerosols, Actual aerosol properties are not as affected when measured this way (Lv et al., 2017; Bedoya-Velásquez et al., 2018). The lidar also provides an opportunity to study the vertical characterization of aerosol hygroscopicity. Many ground-based Raman lidar systems have been operated ~~in~~around the world for measuring both atmospheric water vapor and aerosol profiles at higher spatial and temporal resolutions (Leblanc et al., 2012; Froidevaux et al., 2013; Wang et al., 2015; Bedoya-Velásquez et al., 2018). ~~The Lidar technology allows for measurements under unmodified ambient atmospheric conditions instead of drying and then humidifying air samples that may alter aerosol physical and chemical properties, a major limitation for in situ observation techniques such as the H-TDMA and nephelometer (Bedoya-Velásquez et al., 2018). Raman lidar systems also provide~~These measurements ~~at higher spatial and temporal resolutions which~~ are useful for examining the effects of aerosol hygroscopic growth on pollution events (e.g., Y.-F. Wang et al., 2012, 2017; Su et al., 2017). ~~Despite the numerous~~Many studies~~, these factors~~ on aerosol hygroscopic growth are ~~still poorly known, especially their influential factors in terms~~based on the surface measurements, but few studies have investigated the vertical characterization of aerosol

~~chemical composition.~~ hygroscopicity.

Xingtai ~~is~~as a city with a high density of heavy industries ~~that has been frequently~~was ranked as one of the most polluted cities in ~~China. We have thus attempted to gain deeper insights into the physical, chemical, optical and hygroscopic properties of aerosol particles~~central NCP. A joint field campaign was carried out in ~~order to understand~~this region in the summer of 2016. Some studies based on this campaign have been done for understanding the causes and evolution of pollution ~~in the region. A specific~~events in this region (Y.-Y. Wang et al., 2018; Zhang et al., 2018). These studies have shown that aerosols in Xintai are highly aged and internally mixed due to strong secondary formation. The goal of this study is to ~~study the~~ further investigate how aerosol hygroscopic growth ~~factor~~affects haze events and ~~its~~what are the controlling ~~factor for atmospheric particulate matter that have diameters less than 1 μm~~factors by combining surface and ~~2.5 μm (PM_1 and $PM_{2.5}$, respectively) based on Raman lidar~~vertical measurements ~~made at Xingtai in late May 2016 together with other suites of instruments measuring a variety~~ of aerosol optical, physical, and chemical properties. ~~Two representative cases are also selected to single out the influences of aerosol chemical compositions.~~

The following section describes the ~~field experiment,~~ instruments and ~~data used~~methodology. Section 3 presents the ~~methodology~~results and discussion. Section 4 ~~describes the results. A~~provides a brief summary of ~~this~~the study ~~is given in Section 5~~.

2. ~~Field campaign~~Instruments and ~~instruments~~methodology

~~In order~~**2.1 Instruments**

A Raman lidar was used to analyze the relationship between ~~the~~ atmospheric water vapor content and ~~the~~ PM_1 ~~and~~or $PM_{2.5}$ mass concentrations, and to explore the atmospheric aerosol ~~particle~~ hygroscopic growth effect on haze events~~, a Raman lidar was used~~. The lidar is an automated system that retrieves atmospheric water vapor mixing ratios (W) and aerosol optical property profiles ~~throughout the day. The lidar system used in this study~~(aerosol extinction and backscattering coefficients, Ångström exponent (AE), and the depolarization ratio) throughout the day. The system employs a pulsed neodymium-doped yttrium aluminum garnet laser as a light source and emits three laser beams simultaneously at 355, 532, and 1064 nm with a time resolution of 15 min and a range resolution of 7.5 m~~. The optical receiving unit includes an ultraviolet telescope and a visible infrared telescope. The ultraviolet telescope received~~ based on its original factory settings. The lidar sends 5,000 laser beams in the first four minutes and ten seconds of the 15-minute cycle, then the mean value of the received 5,000 signals are stored as the signal profile to enhance the signal-to-noise ratio. When a laser beam is sent into the atmosphere, the received backscattering signal generally includes Mie scattering by aerosols, Rayleigh scattering by atmospheric molecules, and Raman scattering caused by the rotation and vibration of the molecules. The size of many molecules and atoms in the atmosphere are typically much smaller than the wavelength of the laser, so Rayleigh scattering occurs when they interact (Strutt, 1871). Mie scattering describes the interaction between large particles (mainly atmospheric aerosols) and laser beams. As for the optical receiving unit of this lidar system, optical fiber (OF), dichroic beam splitter (DBS), and ultra-narrowband filters following

an ultraviolet telescope divides atmospheric Mie scattering signals and vibrational Raman scattering signals from H_2O and N_2 molecules (at 355, 386, and 407 nm~~). The atmospheric Mie scattering signal at 532 nm and 1064 nm is received by the visible infrared telescope.,~~ respectively). Atmospheric Mie scattering signals at 532 and 1064 nm are divided by OF, DBS and ultra-narrowband filters after a visible infrared telescope. Based on the perpendicular and parallel components at 532 nm received by the lidar system, the aerosol depolarization ratio, a parameter that measures the shapes of aerosols, can be calculated. In general, the more irregular the particle shape, the larger the value of the depolarization ratio (Chen et al., 2002; Baars et al., 2016). The AE can also be calculated using lidar signals at 532 and 1064 nm, which is inversely related to the average size of the aerosols (Ångström, 1964; Tiwari et al., 2016).

~~Collocated~~Co-located radiosondes were launched twice a day, i.e., at ~0715 and ~1915 Beijing Time (BJT), during the field campaign. The GTS1 detector collected profiles of atmospheric RH, temperature, and pressure at a resolution of 1~~%, .~~0~~.1°C~~ %, 0.1 °C, and 0.1 hPa, respectively. The radiosonde ascension velocity was typically ~5–6 m s^{-1}.

A ~~collocated~~co-located Doppler lidar system (TWP3-M) was also in operation at Xingtai. This system ~~provides~~emits electromagnetic beams in different directions to the upper air, then directly receives the backscattering signals after those beams interact with atmospheric turbulence. Based on the Doppler effect, this system can derive time series of horizontal wind velocity~~, horizontal wind~~ and direction~~, and vertical wind speed~~ 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 (RMSEs) of the Doppler lidar-retrieved wind speed and direction are typically ≤ 1.5 m s^{-1} and ≤ 10°, respectively. The maximum and minimum detection distances of this system are

3–5 km and 0.1 km, respectively.

~~AGrayWolf 6~~A GrayWolf six-channel handheld particle/mass meter (model PC-3016A) was used to directly monitor the total mass concentrations of $PM_{2.5}$ and PM_1 in the actual atmosphere (Yan et al., 2017). ~~An aerosol chemical speciation monitor (ACSM) measures aerosol chemical composition (~~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. The non-refractory PM_1 (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 ~~operations~~operation of the ACSM and ~~data analysis~~ its application in this campaign can be found elsewhere (~~e.g., Sun et al., 2016;~~ Zhang et al., ~~2016, 2017).~~ 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).

The aerosol GF probability distribution function (GF-PDF) at RH = ~85% was

~~retrieved~~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 ~~in this study has also been described in more details in~~ 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). All data are reported in ~~Beijing local time~~BJT in this study.

~~3.~~2.2 Methodology

~~3~~2.2.1 Water vapor retrieval

Using the ratio of the Raman signals of H_2O (P_H) and N_2 (P_N), ~~atmospheric water vapor content (W) can be~~W is calculated as follows (Melfi, 1972; Leblanc et al., 2012; Su et al., 2017):

$$W(z) = C_W \Delta q \frac{P_H(z)}{P_N(z)} \quad , \tag{1}$$

$$\Delta q = \frac{\exp[-\int_0^z (\alpha_N^m + \alpha_N^p) dz]}{\exp[-\int_0^z (\alpha_H^m + \alpha_H^p) dz]} \quad , \tag{2}$$

where C_W is the Raman lidar calibration constant which can be calculated using ~~corresponding~~co-located radiosonde data (Melfi, 1972; Sherlock et al., 1999). The parameters α_N^m and α_H^m are the molecular extinction coefficients at 386 ~~nm~~ and 407 nm, respectively.

These can also be calculated using temperature and pressure profiles from radiosonde measurements (Bucholtz, 1995). The parameters α_N^p and α_H^p are the aerosol extinction coefficients (AECs) at 386 ~~nm~~ and 407 nm, respectively. Here, we use the Fernald method to retrieve AECs (Fernald et al., 1972; Fernald, 1984~~).~~), which is an analytic solution to the following basic lidar equation for Mie scattering:

$$P_s(z) = ECZ^{-2}[\beta_1(z) + \beta_2(z)]T_1^2(z)T_2^2(z), \quad (3)$$

where $P_s(z)$ is the return signal, E is the energy emitted by the laser, C is the calibration constant of the lidar system, and $\beta_1(z)$ and $\beta_2(z)$ are the backscattering cross-sections of atmospheric aerosols and molecules at altitude z, respectively. $T_1(z)$ and $T_2(z)$ are the transmittances of aerosols and air molecules at height z. Note that during the daytime, the height of the retrieved W profile ~~will be~~is limited because the Raman signal is affected by radiation (Tobin et al., 2012).

We can also calculate the vertical distribution of RH based on the vertical profile of W retrieved from Raman lidar measurements and the temperature and pressure profiles provided by radiosonde data. The following equations are used to retrieve the RH profile:

$$RH(z) = \left[\frac{e(z)}{e_s(z)}\right] \times 100\%, \quad (~~3~~4)$$

$$e(z) = \frac{W(z)p(z)}{0.622 + W(z)}, \quad (~~4~~5)$$

$$e_s(z) = 6.1078 \exp\left[\frac{17.13[T(z) - 273.16]}{T(z) - 38}\right], \quad (~~5~~6)$$

where $e(z)$ and $e_s(z)$ are the vertical profiles of water vapor pressure (in hPa) and saturation vapor pressure (in hPa) at a certain temperature, respectively, $W(z)$ is the W profile

obtained from the Raman lidar, $p(z)$ is the pressure profile (in hPa), and $T(z)$ is the temperature profile (in ~~Kelvin~~K) provided by radiosonde data.

To assess the accuracy of the retrieval algorithm, Raman lidar- and radiosonde-derived W and RH profiles at 0515 BJT on 24 May 2016 and their differences are shown in Fig. 1. The W profiles agree reasonably well with an absolute error between them ~~of~~ less than 0.5 g kg^{-1}~~.~~ ~~Figure 2 shows results of the vertical profiles of RH retrieved by the Raman lidar and the Radiosonde at 0515 BJT 24 May 2016~~. Absolute errors between Raman lidar- and radiosonde-derived RH profiles are generally less than 5%. The same inversion results for a relatively wet case on 23 May 2016 are given in Fig. 2. In general, large absolute errors tend to occur at the inflection points. Figures 1 and 2 suggest that the retrieval algorithm can produce reasonable results.

~~3~~2.2 Selection of aerosol hygroscopic cases and their optical properties

How aerosol ~~particle~~ hygroscopic growth cases were chosen is described here. First, atmospheric mixing conditions were examined using radiosonde-based vertical potential temperature (θ) and W profiles. Cases with near-constant values of θ and W in the analyzed layer (variations less than 2°C and 2 g kg^{-1}, respectively) represent good atmospheric mixing conditions (Granados-Muñoz et al., 2015). Then aerosol backscattering coefficient profiles at 532 nm were calculated ~~based on the Fernald method (Fernald, 1984). Cases were chosen with a simultaneous increase in atmospheric RH and aerosol backscattering coefficient. These steps are needed to ensure that the variations in aerosol properties are due to water uptake and not to changes in the aerosol load (Bedoya-Velásquez et al., 2018). Aerosol hygroscopic properties of the selected cases were investigated in terms of the enhancement factor for the backscattering~~

~~coefficient which is defined as follows:~~using the Fernald method (see details in section 2.2.1).

A simultaneous increase in atmospheric RH and the aerosol backscattering coefficient is also needed, which might indicate aerosol hygroscopic growth (Bedoya-Velásquez et al., 2018). Based on the above criteria, individual cases with the same ambient humidity and different pollution conditions were selected for studying the influence of aerosol hygroscopicity on haze events. Aerosol hygroscopic properties of the selected cases were investigated in terms of the hygroscopic enhancement factor for the aerosol backscattering coefficient defined as follows:

$$f_\beta(RH,\lambda)=\frac{\beta(RH,\lambda)}{\beta(RH_{ref},\lambda)}, \quad (6$$

$$f_\beta(RH,\lambda)=\frac{\beta(RH,\lambda)}{\beta(RH_{ref},\lambda)}, \quad (7)$$

where $\beta(RH,\lambda)$ and $\beta(RH_{ref},\lambda)$ represent aerosol backscattering coefficients at a certain RH value and at a reference RH value, respectively, at wavelength λ. In this study, we selected $RH_{ref}=80\%$ which is the lowest RH in the layer ~~to be analyzed~~.

Finally, a ~~two-parameter fit equation was used to obtain the~~ relationship between ~~RH~~ $f_\beta(RH)$ and ~~$f_\beta(RH)$ (Kasten, 1969):~~RH was established. The most commonly used equations are the single-parameter fit equation (e.g., Hänel, 1980; Kotchenruther and Hobbs, 1998; Gassó et al., 2000) and the dual-parameter fit equation (e.g., Hänel, 1980; Carrico, 2003; Zieger et al., 2011). The single-parameter fit equation introduced by Hänel (1976) is

~~$f_\beta(RH)=a(1-RH)^{-b}$, (7)~~

~~where a and b are the hygroscopic parameters that define the enhancement. The larger the value of b is, the more hygroscopic are the particles (Fernández et al., 2015). The Hänel model (Hänel et al., 1976) was also used to calculate $f_\beta(RH)$:~~

$$f_\beta(RH)=\left(\frac{1-RH}{1-RH_{ref}}\right)^{-\gamma}. \quad f_\beta(RH)=\left(\frac{1-RH}{1-RH_{ref}}\right)^{-\gamma}, \quad (8)$$

where γ in an empirical parameter. Larger ~~γ~~ γ values in this formulation ~~denotes~~denote a stronger hygroscopic growth. ~~Whichever model had the best fit to the data was selected.~~

~~3~~The dual-parameter fit equation is (Fernández et al., 2015)

$$f_\beta(RH)=a(1-RH)^{-b}. \quad (9)$$

The single- and dual-parameter fit equations are similar, but with an additional scale factor parameter, a, in the case of the dual-parameter fit equation. The parameter b is also an empirical parameter with larger values of b indicating particles with stronger hygroscopicities. In this study, both parameterized equations are used to verify the consistency of the results. The equation that fits the measurement data best is selected.

2.2.3 Calculation of aerosol ~~particle~~ acidity

~~The~~Aerosol acidity ~~of aerosol particles is a key parameter affecting~~is associated with aerosol hygroscopic growth (e.g. Sun et al., 2009; Fu et al., 2015; Zhang et al., 2015; Lv et al., 2017). ~~Acidic~~When atmospheric aerosols ~~in the atmosphere tend to be more hygroscopic~~are acidic, they have stronger hygroscopicities than when in their neutralized ~~form~~forms (Zhang et al., ~~2007). High hygroscopicity~~2015). The swelling of ~~aerosol particles~~aerosols due to hygroscopic growth enhances their ability to scatter ~~light.~~solar radiation. We examined acidity by comparing the measured NH_4^+ mass concentration ~~and~~with the needed amount ~~needed~~ to fully neutralize sulfate, nitrate, and chloride ions ($NH_{4\ predicted}^+$) detected by the ACSM (Sun et

al., 2009; Zhang et al., 2015; Lv et al., 2017):

$$NH_{4\ predicted}^{+} = (2\times SO_4^{2-}/96 + NO_3^{-}/62 + Cl^{-}/35.5)\times 18\,, \quad (9\,10)$$

where SO_4^{2-}, ~~NO_3^{-}~~ NO_3^{-}, and Cl^{-} represent the mass concentrations (in μg m^{-3}) of the three species~~.~~ measured by the ACSM. The molecular weights of ~~–~~ SO_4^{2-}, NO_3^{-}, ~~Cl^{-}~~ Cl^{-}, and NH_4^{+} are 96, 62, 35.5, and 18, respectively. ~~Aerosol particles~~Aerosols are considered "more acidic" if the measured NH_4^{+} mass concentration is significantly lower than that of ~~the predicted NH_4^{+}. Aerosol particles~~ $NH_{4\ predicted}^{+}$. Aerosols are considered "bulk neutralized" if the two values are similar (Zhang et al., 2007; Sun et al., 2009; Zhang et al., 2015; Lv et al., 2017).

The acidity of ~~aerosol particles~~aerosols can be ~~measured~~quantified by ~~the~~a parameter called the acid value (*AV*) (Zhang et al., 2007~~) defined as follows:~~):

$$AV = (2\times SO_4^{2-}/96 + NO_3^{-}/62 + Cl^{-}/35.5)/(NH_4^{+}/18)\,. \quad (10\,11)$$

~~Aerosol particles~~The chemical formula and numbers after the equal sign have the same meanings as in Eq. (10). Aerosols are considered "bulk neutralized" if *AV* = 1 and "~~strong~~strongly acidic" if *AV* > 1.25. When *AV* = 1.25, 50% of the total sulfate ions in the atmosphere ~~consists~~consist of NH_4HSO_4, and the other 50% consist of $(NH_4)_2SO_4$.

~~3~~2.2.4 Aerosol chemical ion-pairing scheme

~~Zieger et al. (2014) showed that the~~The magnitude of ~~$f(RH)$~~ f(RH) is correlated with the inorganic mass fraction~~.~~ (Zieger et al., 2014). However, ~~the~~ GFs differ with different inorganic salts. To examine the mass fractions of neutral inorganic salts, ACSM measurements were used to calculate their mass concentrations and volume fractions (Gysel et al., 2007). This approach

is based on ~~an~~the ion-pairing scheme introduced by Reilly and Wood (1969). ~~Because the~~The ACSM mainly measures the mass concentrations of ~~SO_4^{2-}, NO_3^-~~ SO_4^{2-}, NO_3^-, NH_4^+, ~~Cl^-~~ Cl^-, and organics~~, the~~. The chlorine ion was not considered here because its concentration is ~~extremely~~ low. The aerosol chemical ion combination scheme is given by the following equations:

$$\begin{aligned} n_{NH_4NO_3} &= n_{NO_3^-} \\ n_{NH_4HSO_4} &= \min(2n_{SO_4^{2-}} - n_{NH_4^+} + n_{NO_3^-}, n_{NH_4^+} - n_{NO_3^-}) \\ n_{(NH_4)_2SO_4} &= \max(n_{NH_4^+} - n_{NO_3^-} - n_{SO_4^{2-}}, 0) \\ n_{H_2SO_4} &= \max(0, n_{SO_4^{2-}} - n_{NH_4^+} + n_{NO_3^-}) \\ n_{HNO_3} &= 0 \end{aligned} \quad , \qquad (\text{~~11~~}12)$$

where n donates the ~~number of the moles,~~mole numbers, and "min" and "max" are minimum and maximum values (Gysel et al., 2007). The volume fractions of inorganic salts can be calculated based on the ion combination scheme and the parameters in Table 1. Furthermore, for a multicomponent particle, the Zdanovskii-Stocks-Robinson mixing rule (Zdanovskii, 1948; Stokes and Robinson, 1966) can be applied to calculate the hygroscopicity parameter κ:

$$\kappa = \sum_i \varepsilon_i \kappa_i \quad . \qquad (\text{~~12~~}13)$$

where κ_i is the hygroscopicity parameter of each individual component. The parameter ε_i is the volume fraction of each component.

~~4.~~3. Results and discussion

~~43~~.1 Observations of W and mass concentrations of PM_1 and $PM_{2.5}$

Figure 3a shows the time series of the lidar-derived W at Xingtai from 19–31 May 2016.

The height of the retrieved W profile was limited because of ~~the~~ solar radiation during the daytime (e.g., Tobin et al., 2012). Overall, W ~~was~~is generally less than 6 g kg^{-1} between 0.3–4 km with a strong daily variability during the period analyzed~~ period. The~~. Figures 3b and 3c show the simultaneous ~~temporal evolution~~time series of the surface mass concentrations of PM_1 and $PM_{2.5}$~~ are shown in Fig. 3b. The variability~~, and W and RH, respectively. The variabilities in PM_1 and $PM_{2.5}$ mass concentrations ~~was~~are strongly coupled with that in W~~.~~ at the surface and in the lower atmospheric layer. Others have also found the same relationship between W in the lower atmospheric layer and the surface mass concentration of $PM_{2.5}$ (e.g., Y.-F. Wang et al., 2012, 2017; Su et al., 2017). Su et al. (2017) suggested that this ~~was~~is due to the aerosol hygroscopic growth effect. ~~To see if this is the true and since~~ The aerosol hygroscopicity is ~~highly dependent on the~~related to aerosol chemical composition over the North China Plain (Zou et al., 2018~~), the~~). Figure 3d shows simultaneous mass fractions of the chemical species ~~of~~comprising PM_1~~ are shown in Fig. 3c~~. As W in the lower atmospheric layer and the surface mass concentrations of PM_1 and $PM_{2.5}$ ~~increased~~increases, the proportion of organic aerosols ~~decreased,~~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. ~~3c.~~3d and marked with grey lines across Fig. 3. In the evening of 21 May 2016 (the leftmost triangle~~), the water vapor content was relatively higher.~~ 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 $PM_{2.5}$ ~~were~~at that time indicated by the leftmost grey line (in the evening of 21 May 2016) are significantly less than ~~those~~that in the ~~morning of 21 May and the~~ evening of 23 May (~~the rightmost triangle in Fig. 3c). The mass fractions of organics at the times~~ indicated by the ~~triangles were similar.~~rightmost grey line). Su et al. (2017) and Y.-F. Wang et al. (2012, 2017) have studied the relationship between atmospheric water vapor and haze events over Beijing and Xi'an, respectively, using Raman lidar measurements. Their analyses showed a positive correlation between W, and PM_{10} and $PM_{2.5}$ mass concentrations, but they ~~did not analyze in detail the reasons behind~~ ignored some unexpected cases ~~that cropped up. To fully understand~~ behind this ~~phenomenon, the~~positive correlation. The two unexpected cases that occurred on 21 May 2016 (Case I) and 23 May 2016 (Case II~~),~~) were selected for ~~a~~ further study.

43.2 Cases studies of aerosol hygroscopic growth

43.2.1 Lidar-estimated hygroscopic measurements

~~Cases~~Two cases were selected: one on 21 May 2016 (Case I) and the other on 23 May 2016 (Case II~~ measurements~~) at the closest ~~to~~time of the radiosonde launch time at 1915 BJT~~, were first selected.~~. Figure 4 shows the vertical distributions of W, θ, the aerosol backscattering coefficient at 532 nm (β_{532}), the backscatter-related Ångström exponent (AE) based on measurements at 532 and 1064 nm, and the particle linear depolarization ratio at 532 nm for Case I and Case II. The altitude ranges ~~for each case~~ are 1642.5–1905.5 m for Case I and 1680.0–2130.0 m for Case II. ~~The atmospheric mixing conditions in each layer were examined using~~ W and θ ~~which were~~ calculated from radiosonde-measured temperature and RH profiles~~.~~

~~The~~ were used to examine the atmospheric mixing conditions in the individual layers. Table 2 lists the gradients ~~(in km^{-1})~~ of the variables within each layer ~~are shown in Table 2~~. The gradient in W changes little within the layer of interest, decreasing monotonically with altitude at a rate of -0.34 g kg^{-1} km^{-1} and -1.42 g kg^{-1} km^{-1} for Case I and Case II, respectively. The gradient in θ shows a ~~monotonically~~monotonic increase within the layers of interest (0.27°C km^{-1} for Case I and 0.96°C km^{-1} for Case II). Overall, W and θ variations are less than 2 g kg^{-1} and 2°C, respectively, showing that good mixing atmospheric conditions were present in both cases (Granados-Muñoz et al., 2015). This confirms that aerosols within the analyzed layer of each case were well mixed.

Figure ~~5~~4 shows the time series of the horizontal wind velocity and direction retrieved from the ~~collocated~~co-located Doppler lidar system. ~~For Case I within its region of interest (1642.5–1905.0 m), the time series of horizontal wind velocity and direction (Fig. 5a and 5c) at five-minute intervals show that from~~From 1830–2030 BJT, ~~winds over the study area~~Case I (Fig. 4c) and Case II (Fig. 4d) winds within their respective layers are mainly ~~came~~ from the north and ~~had~~northwest, respectively, and have relatively low speeds (< 5 m s^{-1}~~). Figure 5b and 5d (Case II) show that winds mainly come from the northwest and also had relatively low speeds (< 5 m s^{-1}) within the analyzed layer (1680.0–2130.0 m) from 1830–2030 BJT.~~, Fig. 4a and 4b). This suggests that ~~aerosol particles~~the aerosols in each case were transported ~~to Xingtai from~~into their respective layers at low speeds from almost the same ~~source region (Bedoya-Velásquez et al., 2018).~~direction. In other words, there is no change in the aerosol type of both cases within the region of interest.

The ~~aerosol backscattering coefficients and~~ RH and β_{532} simultaneously increase with

altitude in the Case I (Fig. 5c and 5d) and Case II (Fig. 5i and 5j) layers of interest. The AE and depolarization ratio were retrieved in order to differentiate the fine/coarse mode predominance and shape of the ~~aerosol particles~~aerosols (Fig. ~~4e~~5e, f, k, and l). A decrease in AE and the depolarization ratio ~~means~~suggests that there is an increase in the predominance of coarse-mode particles and an increase in the sphericity of particles due to water uptake, respectively (Granados-Muñoz et al., 2015; Lv et al,. 2017; Bedoya-Velásquez et al., 2018).

Based on the ~~aerosol backscattering coefficient at 532 nm~~ β_{532} and RH profiles retrieved from Raman lidar measurements, the enhancement factor for the backscattering coefficient at 532 nm, $f_\beta(RH)$, is calculated for both cases using Eq. (~~6~~7). The reference RH value was set to 80% in this study, ~~which is~~ the lowest RH recorded in the layers of interest of both cases. This study applies the ~~two-parameter Kasten model [Eq. (7)] and the one~~single-parameter Hänel model [Eq. (8~~)]~~.)] and the dual-parameter Kasten model [Eq. (9)]. Table 3 lists the parameterized results of each model for each case, and ~~Figure~~Fig. 6 shows the best-fit lines. The ~~enhancement factor~~ $f_\beta(RH)$ for Case II is greater than that for Case I. ~~Specifically, the aerosol backscattering at 532 nm increased~~ β_{532} increases by a factor of 1.094 (Case I) and 1.794 (Case II) as RH ~~changed~~changes from 80% to 91%. The magnitudes of ~~f(85%)~~ $f_\beta(85\%)$ for Case I and Case II are 1.0283 and 1.0770, respectively. The *b* value from the Kasten parameterization is much larger in Case II (0.9346) than in Case I (0.1000), and the γ value from Hänel parameterization for Case II (0.6538) is also much ~~greater~~larger than that for Case I (0.09895). Chen et al. (2014) studied the aerosol hygroscopicity parameter derived from light-scattering enhancement factor [$f(RH)$] measurements made in the ~~North China Plain~~NCP and showed that $f(RH)$ for polluted cases is ~~distinctly~~ higher than that for clean

periods at a specific RH. This is consistent with the results of this study where the mass concentrations of PM_1 and $PM_{2.5}$ during Case II (69.36 μg m^{-3} for PM_1 and 94.88 μg m^{-3} for $PM_{2.5}$) were greater than those during Case I (34.08 μg m^{-3} for PM_1 and 45.00 μg m^{-3} for $PM_{2.5}$). An observational study of the influence of aerosol hygroscopic growth on the scattering coefficient at a rural area near Beijing also demonstrated that ~~during urban pollution periods,~~ aerosols ~~displayed~~had relatively strong water-absorbing properties during urban pollution periods (Pan et al., 2009).

43.2.2 The influences of chemical composition inferred from ACSM measurements

~~Inorganic salt aerosols are mostly hygroscopic by nature, and sulfates and nitrates frequently make up a large part of inorganic aerosols (Tang, 1980). Especially for fine aerosol particles (sizes between 0.1 and 1.0 μm) that scatter visible light more efficiently, the roles of inorganic salt aerosols are often important (Tang, 1996). Liu et al. (2014) have also~~Liu et al. (2014) have pointed out that inorganics are the primary aerosol component contributing to aerosol hygroscopicity especially in the size range of 150–1000 nm. The acidity of ~~aerosol particles~~aerosols is a key parameter affecting aerosol hygroscopic growth (Sun et al., 2009; Lv et al., 2017). ~~Generally speaking, neutral aerosols are less hygroscopic than their acidic forms (Zhang et al., 2007).~~ The dominant form of ~~the~~ inorganics can be examined by comparing measured NH_4^+ and predicted NH_4^+ (Lv et al., 2017; see section ~~3~~2.2.3 for details).

Figure 7 shows the ~~relationship~~relationships between ACSM-measured NH_4^+ and predicted NH_4^+ based on PM_1 chemical species information ~~obtained from~~collected during the ~~ACSM~~full day of each case. The slopes of the linear regression best-fit lines are 0.72 and

0.68 on 21 May 2016 (Case I) and 23 May 2016 (Case II), respectively. The RMSEs of the liner regression best-fit lines are 0.63 and 0.48 on 21 May 2016 and 23 May 2016, respectively. The parameter *AV* for Case I is 1.35 and for Case II is 1.50. These values suggest that there was insufficient NH_3 in the atmosphere to neutralize H_2SO_4, HNO_3, and HCl in each case and that the dominant form of inorganics was NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$. The acidity of ~~aerosol particles~~aerosols in Case II is greater than that in Case I, suggesting that aerosols in Case II were more hygroscopic than those in Case I. This is consistent with the results presented ~~here~~in section 3.2.1.

A hygroscopicity parameter~~, kappa (κ),~~ κ, was developed by Petters and Kreidenweis (2007~~)~~). κ can be calculated using the chemical composition information from Eq. (13) (Gysel et al., 2007; Y.-C. Liu et al., 2016; see section 2.3.4). To further confirm the effect of aerosol hygroscopic growth on haze events, κ is computed for each case based on the dominant form of the inorganics ~~determined previously~~mentioned above.

~~The~~Figure 8 shows the chemical species obtained from ground-based ACSM measurements of PM_1 around the times of the cases ~~are shown in Fig. 8~~. In Case I (Fig. 8a), PM_1 was mainly made up of organic particles (39%) and sulfate (39%), followed by nitrate (8%), ammonium (13%), and chloride (1%). In Case II (Fig. 8b), PM_1 was made up of 37% organics, 25% sulfate, 22% nitrate, 12% ammonium, and 1% chloride. Based on the aerosol chemical ion-pairing scheme introduced in Section ~~3~~2.2.4 and the aerosol properties shown in Table 1, chloride and organics were neglected because of their relatively small ~~content~~contents and comparatively low ~~hygroscopicity~~hygroscopicities (Gysel et al., 2007; Petters and Kreidenweis, 2013). ~~The~~Table 4 lists the mass concentrations and volume fractions of NH_4NO_3,

NH_4HSO_4, and $(NH_4)_2SO_4$ for each case ~~are given in Table 4.~~as well as κ computed using Eq. (13). The mass concentration of H_2SO_4 is equal to zero. Liu et al. (2014) have shown that κ for NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$ is equal to 0.68, 0.56, and 0.60, respectively. ~~Values of κ computed using Eq. (12) are given in Table 4.~~ The parameter κ for Case I (0.557) ~~was~~is less than that for Case II (0.610). This ~~is consistent with our previous results, namely,~~suggests that the ~~enhancement factor of the backscattering coefficient at 532 nm [$f_\beta(RH)$]~~aerosol hygroscopicity for Case II was higher than that for Case I. ~~This~~It also suggests that under the same ~~water vapor~~ambient RH conditions, the nitrate ~~ion~~ content in ~~aerosol particles~~aerosols can cause ~~significant~~ differences in the hygroscopicity of aerosols.

~~4~~3.2.3 Comparison with H-TDMA measurements

~~Concerning the aerosol scattering enhancement factor, during~~In the last decade, many studies have compared remotely sensed and in situ aerosol scattering enhancement factor measurements ~~(~~using a humidified tandem nephelometer~~)~~ and have shown ~~a~~ positive ~~result~~results (Zieger et al., 2011, 2012; Sheridan et al., 2012; Tesche et al., 2014; Lv et al., 2017). The H-TDMA is also a reliable instrument for ~~use in~~ measuring the aerosol diameter ~~GF~~hygroscopicity due to water uptake (Liu et al., 1978). ~~The aerosol particle diameter~~Aerosol GFs observed by the ground-based H-TDMA at times nearest to the ~~closest time~~times of each case are examined next.

~~Table 5 lists~~Based on H-TDMA-derived aerosol ~~particle size hygroscopic~~ GFs at an RH level of ~~~~85% for different particle sizes~~. All aerosol particle size hygroscopic GFs for Case II are higher than those for Case I.~~ (40, 80, 110, 150, and 200 nm), GFs for different aerosol

particle sizes in both cases were extrapolated to different RH levels using Eq. (3) from Gysel et al. (\(2009) who used the kappaκ model introduced by Petters and Kreidenwies (2007). Figure 9 shows that Case II aerosol particle size hygroscopic GFs at each RH level (80–91%) are higher than those of Case I. Although the aerosol backscattering enhancement factor and aerosol particle $f_\beta(RH)$ and GF are completely different parameters for calculating the hygroscopicity of aerosol particlesaerosols and are difficult to compare quantitatively, the H-TDMA results offer a sense of confidence that aerosol hygroscopicity has an important influence on the formation of heavy haze.

In general, both the lidar-estimated aerosol backscattering hygroscopic enhancement factor and the ACSM and H-TDMA measurements support the proposed hypothesis that the main reasondifferent hygroscopic properties of aerosols are mainly responsible for the strong coupling between the variability in PM_1 and $PM_{2.5}$ mass concentrations is strongly coupled with thatand the variability in W which has to do with the different hygroscopic properties of aerosols.

5.4.Conclusions

During late May 2016, the *W*water vapor mixing ratio in the 0.3–4 km layer over Xingtai was generally less than 6 g kg^{-1} with a strong daily variability. Overall, the simultaneous temporal change ofchanges in the mass concentrations of PM_1 and $PM_{2.5}$ waswere strongly associated with that of the atmospheric water vapor content due to the hygroscopicity of the aerosol particlesaerosols. Two cases where this relationship was not seen were identified and further examined. Case I represents a relatively clean case, and Case II represents a polluted

case. The lidar-estimated aerosol backscattering coefficient hygroscopic enhancement factor [$f_\beta(RH)$] for Case II is greater than that for Case I. The γ ~~value~~and b values from the Hänel ~~parameterization~~and Kasten parameterizations, respectively, for Case II ~~(0.6538) was~~were larger than ~~that~~those for Case I ~~(0.09895).~~. A key parameter affecting the hygroscopicity of ~~aerosol particles~~aerosols, namely, the acid value~~,~~ (AV), was examined by comparing measured NH_4^+ and predicted ~~NH_4^+. The acid value~~ NH_4^+ based on data obtained by the ACSM. The AV for Case I (1.35) was less than that for Case II (1.50) and the main form of inorganics was NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$. The aerosol chemical composition determined by the ACSM showed that the ~~value of the~~ aerosol hygroscopicity parameter κ for Case II (0.610) was greater than that for Case I (0.577) due to the greater mass fraction of nitrate salt. Based on H-TDMA measurements, model results showed that the aerosol size hygroscopic growth factor (GF) in each particle size category (40, 80, 110, 150, and 200 nm) for Case II was greater than that for Case I.

The ~~aerosol backscattering enhancement factor [$f_\beta(RH)$], the aerosol particle growth factor, the aerosol acidity~~ $f_\beta(RH)$, GF, AV, and ~~the hygroscopicity parameter~~κ are completely different quantities for calculating the hygroscopicity of ~~aerosol particles~~aerosols and are difficult to compare quantitatively. The lidar-estimated ~~aerosol hygroscopic enhancement factor~~ $f_\beta(RH)$ and ACSM and H-TDMA measurements show that the hygroscopic growth of ~~aerosol particles~~aerosols has a strong influence on the process of air pollution. Under the same atmospheric ~~water vapor content~~relative humidity conditions, the stronger the hygroscopicity of aerosols, the more likely they cause severe air pollution. The mass fraction of the nitrate ion in ~~aerosol particles~~aerosols was one of the main factors that determined the hygroscopic ability

of aerosols in the study area (Xingtai). These findings not only reveal ~~a major cause of air pollution~~why haze events in Xintai can be severe, but they also provide ~~a~~ scientific ~~basis for~~evidence that may be used to persuade the local government to ~~put more effort into preventing~~prevent and ~~controlling~~control environmental contamination in this ~~commonly known as the most~~heavily polluted ~~place in~~part of China.

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Table 1. Aerosol properties of selected compounds used for the calculation of the hygroscopicity parameter– κ, i.e., the density (ρ_i) and (κ_i) of each compound.

species	NH_4NO_3	NH_4HSO_4	$(NH_4)_2SO_4$	H_2SO_4
density[a]	1.725	1.78	1.76	1.83
κ [b]	0.68	0.56	0.52	0.91

(a) Tang and Munkelwitz (1994); Carrico et al. (2010);

(b) Fountoukis and Nenes (2007); Carrico et al. (2010); Liu et al. (2014).

Table 2. Range of values and gradient values over the analyzed layer for the water vapor mixing ratio (W), the potential temperature (θ), the backscattering coefficient at 532 nm (β_{532}), the Ångström exponent [AE (532–1064 nm)], and the depolarization ratio at 532 nm for Cases I and II.

	Case I			Case Ⅱ		
	Range		Gradient (km^{-1})	Range		Gradient (km^{-1})
Altitude (m)	1642.5	1905.0	—	1680.0	2130.0	—
W (g kg^{-1})	7.65	7.56	-0.34	6.42	5.78	-1.42
~~θ (℃)~~ θ (℃)	26.93	27.00	0.27	25.18	25.61	0.96
RH (%)	80	91	—	80	91	—
β_{532nm} (km^{-1} sr^{-1})	0.01379	0.01535	—	0.003711	0.006762	—
AE (532–1064 nm)	0.74	0.68	-0.23	0.42	0.35	-0.16
Depolarization ratio	0.046	0.044	-0.0076	0.041	0.039	-0.0044

Table 3. The fitting parameters and R^2 of the ~~fit~~fits for the Kasten and Hänel models.

	Case I			Case II		
	a	b	R^2	a	b	R^2
Kasten model	0.8508	0.1000	0.97	0.1916	0.9346	0.95
	γ		R^2	γ		R^2
Hänel model	0.09895 ± 0.0047		0.97	0.6538 ± 0.0662		0.84

Table 4. Calculated mass concentrations and volume fractions of NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$, and the hygroscopicity parameter (κ) for Case I and Case II.

	Case I			Case II		
	NH_4NO_3	NH_4HSO_4	$(NH_4)_2SO_4$	NH_4NO_3	NH_4HSO_4	$(NH_4)_2SO_4$
mass conc. ($\mu g\ m^{-3}$)	3.60	8.31	8.30	12.2979	10.3795	3.0616
volume fraction	0.18	0.41	0.41	0.48	0.40	0.12
κ		0.557			0.610	

Table 5. Different aerosol particle size hygroscopic growth factors at a relative humidity level of ~85% for Case I and Case II.

—	GF(85%, D_p)				
	40 nm	80 nm	110 nm	150 nm	200 nm
Case I	1.32	1.36	1.39	1.40	1.41
Case II	1.33	1.39	1.40	1.41	1.42