# **Response to comments by referee 3 Specific comments:**

The spatial and seasonal characteristics of mixing layer height (MLH) over northern China plain (NCP) were revealed by the authors using variety of measurements, primarily focusing on northern NCP and southern Hebei. The authors attempt to explain the different feature of MLH development between the two interested regions by examining observed wind shear, buoyancy and turbulent stability. In addition, this study pointed out that the MLH plays a key role in forming the heavy near-ground particular mater (PM) pollution besides emissions. The paper is well organized and the reasoning for MLH spatial variations and its association with air pollution is comprehensively discussed. I recommend to publish this paper in ACP journal after addressing following minor issues.

# **Comment 1:**

For Fig. 6: What are the reasons for the difference in buoyancy term profiles between the site XT and BJ during winter, as shown in Fig. 6g? In addition, the absolute values of buoyancy term seem larger than that of shear term; does this mean the buoyancy term rather than the shear term is the dominant component in determining turbulent energy?

# **Response 1:**

Thank you for your suggestion. As we described in section 2.4, the Gradient Richard

number (*Ri*) is the ratio of buoyancy term 
$$(\frac{g}{\bar{e}}\frac{\Delta\bar{e}}{\Delta z})$$
 and shear term  $(\left(\frac{\Delta\bar{u}}{\Delta z}\right)^2 + \left(\frac{\Delta\bar{v}}{\Delta z}\right)^2)$ .

For the static instability, the buoyancy term is usually negative, and the buoyancy force will suppress the turbulent development; for the neutral stratification, the buoyancy term is usually zero; and for the static stability stratification, the buoyancy is usually positive, which will promote the turbulent development. While the shear term usually has positive value and attribute to the mechanical turbulence. In our study, the averaged buoyancy term is positive and larger than the shear term, leading to the Ri larger than 1, this indicated that the mechanical production rate can not balance the turbulent kinetic energy's consumption by buoyancy. Therefore, from a statistical point of view, the atmospheric turbulence is stable on the BJ, XT and LT stations. Turbulent energy is affected by various factors, except for the buoyancy term and the mechanical product term, there are also the turbulent transport contribution and the dissipation terms. Through analysis of the Ri value is just for a simplify and effective evaluation. Although the averaged result exhibited a more significant effect of buoyancy term than the shear term, there are many different occasions that the Ri is less than 1, and the shear term may play a dominant role.

As we mentioned in line 407-410, the higher buoyancy term in XT may be resulted from the warm advection from the Loess Plateau. Since the warm advection usually develops from southwest to the northeast and results in strong thermal inversion above the NCP plain, the warm advection will has stronger impact at the XT station than the BJ station. Thus, this will lead to a higher buoyancy term contribution in XT.

# Comment 2:

Line 381-382: It appears that the profiles were averaged over only two time points (i.e. 8:00 am and 08:00 pm), right? Can the average of only two time points represent the entire day MLH features which are most significant during noon time?

# **Response 2:**

Thank you for your suggestion. Yes, the profiles were averaged over only two time points (i.e. 8:00 am and 08:00 pm). Analysis of these two time points could explain the lower MLH in XT at 8:00 am and 08:00 pm, but could not exactly illustrate the entire day MLH features. Although we can not better explain the day MLH features with these limited data, our study still provide a fundamental knowledge about the reasons for MLH contrast between northern NCP and southern Hebei. We can also make a prediction about these parameters features during noon time: it is known that the MLH development is mainly affected by the solar radiation during daytime and such radiation is almost consistent on the NCP plain, since the XT has weaker turbulent develop condition (i.e., weaker mechanical force and stronger buoyancy inhibition) than the BJ station, the MLH development at the XT station might be weaker than the BJ station. Such limitation in our study is illustrated in the final paragraph of the conclusion section.

# Comment 3:

Line 444-447: In addition to the emission discrepancy and different meteorological factors (like MLH), the aerosol-radiation interactions are a potential candidate to explain different PM pollution between the two interested regions. Therefore, it is better to say 60% is the upper bound of contribution due to meteorological factors.

# **Response 3:**

Thank you for your suggestion. Consider your comment, it is certainly reasonable to say 60 % is the upper bound of contribution due to meteorological factors. Therefore, the relevant contents were modified in section 4.3 in our revised manuscript.

# Comment 4:

Line 474: a recent ref should also be cited here:

Wang G, et al. (2016) Persistent sulfate formation from London Fog to Chinese haze. Proceedings of the National Academy of Sciences 113(48):13630-13635.

# **Response 4:**

Thank you for your suggestion. The paper that you mentioned has been cited in our revised manuscript.

# Comment 5:

Line 409: "enhance" should be "enhanced" or "enhancement of".

# **Response 5:**

Thank you for your suggestion. We have already modified the relevant content in the revised manuscript.

## 1 Mixing layer height on the North China Plain and meteorological

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## evidence of serious air pollution in southern Hebei

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## 22 Abstract

To investigate the spatiotemporal variability of the mixing layer height (MLH) on the 23 North China Plain (NCP), multi-site and long-term observations of the MLH with 24 ceilometers at three inland stations (Beijing (BJ), Shijiazhuang (SJZ) and Tianjin (TJ)) 25 and one coastal site (Qinhuangdao) were conducted from 16 October 2013 to 15 July 26 27 2015. The MLH of the inland stations in the NCP were highest in summer and lowest in winter, while the MLH on the coastal area of Bohai was lowest in summer and 28 29 highest in spring. As a typical site in southern Hebei, the annual mean of the MLH at SJZ was 464±183 m, which was 15.0 % and 21.9 % lower than that at the BJ 30 31  $(594\pm183 \text{ m})$  and TJ  $(546\pm197 \text{ m})$  stations, respectively. Investigation of the shear term and buoyancy term in the NCP revealed that these two parameters in southern 32 Hebei were 2.8 times lower and 1.5 times higher than that in northern NCP within 33 0-1200 m in winter, respectively, leading to a 1.9-fold higher frequency of the 34 Gradient Richardson number >1 in southern Hebei compared to the northern NCP. 35 36 Furthermore, combined with aerosol optical depth and  $PM_{2.5}$  observations, we found 37 that the pollutant column concentration contrast (1.2 times) between these two areas was far less than the near-ground PM2.5 concentration contrast (1.5 times). Through 38 39 analysis of the ventilation coefficient in the NCP, the near-ground heavy pollution in southern Hebei mainly resulted from the lower MLH and wind speed. Therefore, due 40 41 to the importance of unfavorable weather conditions, heavily polluting enterprises should be relocated and strong emission reduction measures are required to improve 42

43 the air quality in southern Hebei.

## 44 **1. Introduction**

45 The convective boundary layer is the region where turbulence is fully developed. The height of the interface where turbulence is discontinuous is usually referred to as 46 the mixing layer height (MLH) (Stull, 1988). The mixing layer is regarded as the link 47 between the near-surface and free atmosphere, and the MLH is one of the major 48 factors affecting the atmospheric dissipation ability, which determines both the 49 volume into which ground-emitted pollutants can disperse, as well as the convective 50 time scales within the mixing layer (Seidel et al., 2010). In addition, continuous MLH 51 observations will be of great importance for the improvement of boundary layer 52 parameterization schemes and for the promotion of meteorological model accuracy. 53

Conventionally, the MLH is usually estimated from radiosonde profiles (Seidel et 54 55 al., 2010). Although meteorological radiosonde observations can provide high-quality 56 data, they are not suitable for continuous fine-resolution MLH retrievals due to their high cost and limited observation intervals (Seibert et al., 2000). As the most 57 advanced method of MLH detection, remote sensing techniques based on the profile 58 measurements from ground-based instruments such as sodar, radar, or lidar that have 59 60 the unique vertically resolved observational capability are becoming increasingly popular (Beyrich, 1997; Chen et al., 2001; He et al., 2005). Because sound waves can 61 be easily attenuated in the atmosphere, the vertical range of sodar is generally limited 62 to within 1000 m. However, the optical remote sensing techniques can provide higher 63 height ranges (at least several kilometers). The single-lens ceilometers developed by 64 Vaisala have been widely used in a variety of MLH studies (Alexander et al., 2017; 65 Emeis et al., 2004, 2009, 2011; Eresmaa et al., 2006; Münkel et al., 2004, 2007; 66 Muñoz and Undurraga, 2010; Schween et al., 2014; Sokół et al., 2014; Tang et al., 67 2015, 2016; Wagner et al., 2006, 2015). Compared with other remote sensing 68 instruments, this type of lidar has special features favorable for long-term and 69 multi-station observations (Emeis et al., 2009; Wiegner et al., 2014; Tang et al., 2016), 70 71 including the low-power system, the eye-safe operation within a near infrared laser 72 band, and the low cost and ease of maintenance during any weather conditions 73 (excluding rainy, strong windy or sandstorm weather conditions) with only regular window cleaning required (Emeis et al., 2004; Tang et al., 2016). 74

75 The North China Plain (NCP) region is the political, economic and cultural center of China. With the rapid economic development, energy use has increased 76 substantially, resulting in frequent air pollution episodes (Guo et al., 2011; Li et al., 77 78 2013; Liu et al., 2016; Tang et al., 2017b; Wang et al., 2014; Wang et al., 2013a; Xu et al., 2016; Zhang et al., 2014). The haze pollution has had an adverse impact on human 79 health (Tang et al., 2017a) and has aroused a great deal of concern (Tang et al., 2009; 80 Ji et al., 2012; Zhang et al., 2015a). To achieve the integrated development of the 81 Jing-Jin-Ji region, readjustment of the regional industrial structure and layout is 82 83 imperative. To this end, the industrial capacity of heavily polluting enterprises in the areas with unfavorable weather conditions should be reduced, and these heavily 84 85 polluting enterprises should be removed to improve the air quality. For the remaining enterprises, the industrial air pollutant emissions structure should be changed, and 86

strong emission reduction measures must be implemented. Although the government 87 has carried out some strategies for joint prevention and control, with the less 88 89 well-understood distributions of regional weather condition on the NCP, how and where to adjust the industrial structures on the NCP are questions in pressing need of 90 answers. As one of the key factors influencing the regional heavy haze pollution 91 (Tang et al., 2012, 2016, 2017b; Quan et al., 2013; Hu et al., 2014; Zhu et al., 2016; 92 93 Zhang et al., 2016a), the MLH to some extent represents the atmospheric environmental capacity, and the regional distribution and variation of MLH on the 94 NCP can offer a scientific basis for regional industrial distribution readjustment, 95 96 which will be of great importance for regional haze management.

Nevertheless, due to the scarcity of MLH observations on the NCP, reliable and 97 98 explicit characteristics of MLH on the NCP remain unknown. Tang et al. (2016) utilized the long-term observation data of MLH from ceilometers to analyze the 99 characteristics of MLH variations in Beijing (BJ) and verified the reliability of 100 ceilometers. The results demonstrated that MLH in BJ was high in spring and summer 101 and low in autumn and winter with two transition months in February and September. 102 A multi-station analysis of MLH on the NCP region was conducted in February 2014, 103 104 and the characteristics of high MLH at coastal stations and low MLH at southwest piedmont stations were reported (Li et al., 2015). Miao et al. (2015) modeled the 105 seasonal variations of MLH on the NCP and discovered that the MLH was high in 106 spring due to the strong mechanical forcing and low in winter as a result of the strong 107 thermodynamic stability in the near-surface layer. The mountain-plain breeze and the 108 109 sea breeze circulations played an important role in the mixing layer process when the background synoptic patterns were weak in summer and autumn (Tang et al., 2016; 110 111 Wei et al., 2017).

To overcome previous studies' deficiencies, our study first conducted a 22-month 112 113 (from 16 October 2013 to 15 July 2015) observation of MLH with ceilometers on the NCP. The observation stations included three inland stations (BJ, Shijiazhuang (SJZ) 114 and Tianjin (TJ)) and one coastal site (Qinhuangdao (QHD)). First, we will describe 115 the spatial and temporal distribution of MLH on the NCP. Subsequently, reasons for 116 spatial difference of MLH on the NCP will be explained in the discussion section. 117 Finally, the meteorological evidence of serious air pollution in southern Hebei will be 118 119 studied.

## 120 2 Data and methods

## 121 2.1 Sites

To study the MLH characteristics on the NCP, observations with ceilometers were 122 conducted at the BJ, SJZ, TJ and QHD stations from 16 October 2013 to 15 July 2015 123 124 (Fig. 1 and Table S1). The SJZ, TJ and QHD sites were set around Beijing in the 125 southwest, southeast and east directions, respectively. The BJ station was at the base of the Taihang and Yanshan Mountains on the northern NCP. The MLH observation 126 127 site was built in the courtyard of the Institute of Atmospheric Physics, Chinese Academy of Sciences (116.32° E, 39.90° N). SJZ was near the Taihang Mountain in 128 southern Hebei; the location was in the Hebei University of Economics (114.26 $^{\circ}$  E, 129 38.03° N). The TJ site was set in the courtyard of the Tianjin Meteorological Bureau, 130

which was located south of the urban area, with a geographic location of 117.20° E,
39.13° N. The QHD station was an eastern coastal site of Bohai Bay, which was set
up in the Environmental Management College of China (119.57° E, 39.95° N), and
the surrounding areas are mostly residential buildings with no high structures. Since
the TJ site was approximately 50 km away from the coast and the QHD station was
only 2 km, the TJ station, by contrast, was supposed to be an inland station.



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Fig. 1 Locations of the ceilometers observation sites (BJ, SJZ, TJ and QHD) are
marked with red and bold abbreviations; other PM<sub>2.5</sub> observation sites (ZJK, CD, LF,
TS, CZ, BD, HS, XT, HD, DZ, LC, JN, BZ, DY, ZB and WF) and the sounding
observation sites (BJ, LT and XT) are marked on the map with black abbreviations.
The size and color of the circular mark are representative of the annual mean
near-ground PM<sub>2.5</sub> concentration; the larger and darker the circle is, the greater the
concentration is.

## 145 2.2 Measurement of MLH

The instrument used to measure the MLH at the four stations was an enhanced 146 single-lens ceilometer (Vaisala, Finland), which utilized the strobe laser lidar (laser 147 148 detection and range measurement) technique (910 nm) to measure the attenuated backscattering coefficient profiles. As large differences existed in the aerosol 149 concentrations between the mixing layer and the free atmosphere, the MLH can be 150 determined from the vertical attenuated backscattering coefficient ( $\beta$ ) gradient, 151 whereby a strong, sudden change in the negative gradient  $(-d\beta/dx)$  can indicate the 152 153 MLH. In the present study, the Vaisala software product BL-VIEW was utilized to calculate the MLH by determining the location of the maximum  $|-d\beta/dx|$  in the 154 attenuated backscattering coefficient. To strengthen the echo signals and reduce the 155 156 detection noise, spatial and temporal averaging should be conducted before the gradient method is used to calculate the MLH. The BL-VIEW software was utilized 157 with temporal smoothing of 1200 s and vertical distance smoothing of 240 m. The 158 instrument installed at the BJ station was a CL31 ceilometer and the CL51 159

ceilometers were used at the SJZ, TJ and QHD stations. Some of the properties of 160 these two instruments are listed in Table 1, and basic technical descriptions can be 161

162 found in Münkel et al. (2007) and Tang et al. (2015).

To ensure the consistency of the MLH measurements with the two different 163 ceilometer versions, before we set up the ceilometer observation network in the NCP, 164

we made a comparison of the MLHs observed by CL31 and CL51 at BJ from October 165

1 to October 8, 2013 (Fig. S1). The MLH observed by CL31 was highly relevant to 166

those observed by the CL51 ceilometers, with correlation coefficients (R) of 0.86-0.92. 167

Therefore, the impact of version discrepancy on the MLH measurement can be 168 neglected. 169

170 Since the ceilometers can reflect rainy conditions and the precipitation will influence the MLH retrieval, the precipitation data were excluded. In addition, a 171 previous study has compared MLH measurements retrieved from ceilometers and 172 sounding data (Tang et al., 2016). The results revealed that the ceilometers 173 underestimate the MLH under neutral conditions caused by strong winds and 174 overestimate the MLH when sand storms occur. Therefore, data points for these three 175 special weather conditions were eliminated manually. The criterion to exclude these 176 data points is as follows: (a) precipitation, i.e., a cloud base lower than 4000 m and 177 the attenuated backscattering coefficient of at least  $2 \times 10^{-6}$  m<sup>-1</sup>sr<sup>-1</sup> within 0 m and the 178 cloud base, (b) sandstorm, i.e., the ratio of PM2.5 to PM10 suddenly decreased to 30 % 179 or lower and the  $PM_{10}$  concentration was higher than 500 µg m<sup>-3</sup>, and (c) strong winds, 180 i.e., a sudden change in temperature and wind speed (WS) when cold fronts passed by 181 (Muñoz and Undurraga, 2010; Tang et al., 2016; Van der Kamp and McKendry, 182

2010). 183

Table 1 Instrument properties of	CL31 and CL51	
Parameter	CL31	CL51
Detection range (km)	7. <u>5</u> 7	1 <u>5</u> 3.0
Wavelength (nm)	910	910
Report period (s)	2-120	6-120
Measurement interval		
(s)Report accuracy	<u>2</u> 5	<u>610</u>
<del>(m)</del>		
Measurement		
resolution (m)Peak-	<u>10</u> 310	<u>10</u> 310
power (W)		

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#### 2.3 Other data 185

The hourly data of near-ground relative humidity (RH) and temperature (T) in the 186 NCP region were obtained from the China Meteorological Administration 187 (http://www.weather.com.cn/weather/101010100.shtml/). To study the reason for the 188 MLH difference between the northern NCP and southern Hebei, meteorological 189 sounding data were included in this paper. The data were provided by the upgraded 190 radiosonde network of China, where the GTS1 digital electronic radiosonde was 191 required to be operationally launched twice per day at 08:00 LT and 20:00 LT by the 192 193 China Meteorological Administration (Guo et al., 2016). Considering the deficiency

of sounding data at the SJZ and QHD stations, data from the Xingtai (XT) and
Laoting (LT) stations were used instead after a consistency test with the reanalysis
data (Fig. S2). The reanalysis data at these four sites were downloaded from the
website of European Centre for Medium-Range Weather Forecasts
(http://apps.ecmwf.int/datasets/data/interim-full-mnth/levtype=pl/).

The near-ground  $PM_{2.5}$  and  $PM_{10}$  concentrations at the 20 observation sites from December 2013 to November 2014 were provided by the Ministry of Environmental Protection with a time resolution of 1 h (<u>http://www.zhb.gov.cn/</u>). Details for the near-ground  $PM_{2.5}$  and  $PM_{10}$  observation sites are shown in Table S1 and Fig. 1.

The aerosol optical depth (AOD) data within the NCP region were retrieved with the dark target algorithm from the Moderate Resolution Imaging Spectra-radiometer aerosol products on board the National Aeronautics and Space Administration Earth Observing System Terra satellite from December 2013 to November 2014 (Zhang et al., 2016b) (https://ladsweb.nascom.nasa.gov/search/index.html/), then the AOD data was interpolated into 0.1°×0.1° to produce the regional distribution in the NCP.

## 209 2.4 Atmospheric stability criterion

The Gradient Richardson number (Ri) is usually used to estimate the atmospheric turbulent stability within the mixing layer and is defined as follows (Eq. 1):

212 
$$Ri = \frac{\frac{g\Delta\theta}{\bar{\theta}\Delta z}}{\left(\frac{\Delta\bar{u}}{\Delta z}\right)^2 + \left(\frac{\Delta\bar{v}}{\Delta z}\right)^2}$$
(1)

213 Where  $\Delta z$  is the height increment over which a specific calculation of *Ri* is being 214 made; g is the acceleration of gravity;  $\overline{\theta}$  is the mean virtual potential temperature 215 within that height increment; and  $\Delta \overline{u}$  and  $\Delta \overline{v}$  are the mean wind speeds in zonal and 216 meridional directions within the height increment.

Using *Ri* to diagnose turbulence is a classical approach and has been covered in many textbooks on boundary-layer turbulence (Stull, 1988; Garratt, 1994). It can be

# 219 interpreted as the ratio of the buoyancy term $\left(\frac{g}{\overline{\theta}}\frac{\Delta\overline{\theta}}{\Delta z}\right)$ to the shear term $\left(\left(\frac{\Delta\overline{u}}{\Delta z}\right)^2 + \frac{1}{2}\right)$

220  $\left(\frac{\Delta \overline{v}}{\Delta z}\right)^2$  in the turbulent kinetic equation. When the *Ri*>1, the turbulence was 221 suppressed and the mixing layer development will be restrained (Stull, 1988). In our 222 study, the frequency of *Ri*>1 was used to represent the atmospheric stability in the

223 NCP. The larger the frequency is, the more stable turbulent stratification is.

## 224 **3. Results**

## 225 **3.1 Frequency distribution of MLH**

Since October 2013, continuous operation of the ceilometers observation network 226 227 in the NCP has provided 22 months of MLH data. For the purpose of analyzing the MLH temporal and spatial variation, the hourly averages of MLH for a whole year 228 (from December 2013 to November 2014) at the BJ, SJZ, TJ and QHD stations were 229 230 chosen in the following sections. Hourly means of MLH under rainy, sandstorm and windy conditions were removed, resulting in data availability of 81, 89, 83 and 77 % 231 at the BJ, SJZ, TJ and QHD stations, respectively. In this study, March, April and May 232 233 are defined as spring; June, July and August are defined as summer; September, October and November are defined as autumn; and December, January and Februaryare defined as winter.

236 To study the regional distribution characteristic of MLH on the NCP, we analyzed the frequency of the daily maximum MLH distribution in Fig. 2. The daily maximum 237 MLH at the BJ, SJZ and TJ stations could reach 2400 m. The large daily maximum 238 values mostly existed in spring and summer, while the low values always appeared in 239 autumn and winter and were as low as 200 m. The daily maximum MLH values at the 240 BJ, SJZ and TJ stations were mainly distributed between 600 and 1800 m, 400 and 241 242 1600 m and 800 and 1800 m, accounting for 74.2, 72.0 and 67.0 % of the total samples, respectively. Notably, the daily maximum MLH in SJZ was lower than at the 243 MLHs at the BJ and TJ stations in spring, autumn and winter. Values below 600 m at 244 245 the SJZ station occurred primarily in autumn and winter. The most frequent daily maximum MLH existed in the range of 1000-1200 m, which was 200-600 m lower 246 247 than that at the TJ station. This demonstrated a weaker atmospheric diffusion capability at the SJZ station in spring, autumn and winter than the northern NCP 248 249 stations.

The frequency distribution of the daily maximum MLH at the coastal site showed different features. The daily maximum MLH in QHD was mainly distributed between 800 and 1800 m with a relatively small seasonal fluctuation (Fig. 2d). Values lower than 600 m were mainly distributed in summer, which was probably influenced by the frequent occurrence of a thermal internal boundary layer (TIBL) in summer. Reasons for this are illustrated in section 4.1.



256

Fig. 2 Frequency distribution of the daily maximum MLH at the (a) BJ, (b) SJZ, (c)

TJ and (d) QHD stations from December 2013 to November 2014.

<sup>259 3.2</sup> Spatiotemporal variation of MLH

## 260 3.2.1 Seasonal variation

Monthly variations of MLH at the BJ, SJZ, TJ and QHD stations are shown in Fig. 3. The monthly means of the regional MLH ranged between 300 and 750 m. The maximum and minimum MLH existed in June 2014 at the BJ station and in January 2014 at the SJZ station, with values of 741 and 308 m, respectively. Most of the monthly averages were between 400 and 700 m, which accounted for 81.3 % of the total samples.

The MLH at the BJ, SJZ and TJ stations showed obvious seasonal variations with 267 high values in spring and summer and low values in autumn and winter. Seasonal 268 269 means of MLH at the three stations followed the same order: summer>spring>autumn>winter, with maximum values of 722±169, 623±161 and 270  $655\pm165$  m in summer, respectively, and minimum values of  $493\pm131$ ,  $347\pm153$  and 271 436±178 m in winter, respectively (Table S2). Obvious annual changes of the MLH 272 with large values in spring and summer and low values in autumn and winter at the BJ, 273 SJZ and TJ stations implied that MLH is influenced by seasonal changes of solar 274 radiation (Stull, 1988). 275

Nevertheless, the seasonal variation of MLH at the coastal site of Bohai was 276 277 different from that at the inland stations. The MLH in QHD exhibited a decreasing trend from spring to summer and an increasing trend from autumn to winter, with the 278 maximum seasonal mean of 498±217 m in spring and the minimum seasonal mean of 279 280 447±153 m in summer. Moreover, the MLH in spring and summer at QHD was much lower than those at other stations. Similar to our analysis of frequency distributions of 281 282 daily maximum MLH in section 3.1, the lower MLH at QHD in spring and summer 283 mainly resulted from the frequent occurrence of the TIBL. A detailed explanation of the TIBL impact was included in section 4.1. The effect of TIBL on the coastal 284 boundary layer was consistent with previous studies (Zhang et al., 2013; Tu et al., 285 286 2012), which demonstrated that ceilometers can properly retrieve the coastal MLH as 287 well.



288

Fig. 3 Monthly variations of MLH at the BJ, SJZ, TJ and QHD stations fromDecember 2013 to November 2014.

## 291 **3.2.2 Diurnal variations**

292 Seasonal variations of diurnal MLH change patterns were investigated to reveal the 293 24 h evolution characteristics of the MLH on the NCP. As shown in Fig. 4, diurnal

variations of MLH in different seasons all had single peak patterns. With sunrise and

increased solar radiation, MLH at the four stations started to develop and peaked in 295 the early afternoon. After sunset, turbulence in the MLH decayed quickly, and the 296 297 mixing layer underwent a transition to the nocturnal stable layer (less than 400 m). The annual averaged diurnal ranges of MLH at the BJ, SJZ, TJ and QHD stations 298 were 782, 699, 914 and 790 m, respectively. The annual averaged diurnal range of 299 MLH in SJZ was approximately 100-200 m smaller than those at the other stations, 300 301 which was associated with its shallow daytime MLHs in spring, autumn and winter (Figs. 4a, 4c and 4d). This also indicated the worse pollutant diffusion ability in SJZ. 302

303 Growth rates averaged over the four stations during each season were plotted with gray columns in Fig. 4. It was obvious that the growth rates of the MLH varied by 304 season. The MLH developed the earliest in summer (at approximately 7:00 LT) and 305 reached the highest growth rates (164.5 m h<sup>-1</sup>) at approximately 11:00 LT, and the 306 307 time when MLH started to develop was found to be 1 hour later (at approximately 308 8:00 LT) in spring and autumn than in summer. Furthermore, the MLH developed the latest (at approximately 9:00 LT) and slowest in winter, with the maximum growth 309 rate (101.8 m h<sup>-1</sup>) occurring at approximately 11:00 LT. 310



Fig. 4 Diurnal variations of MLH at the BJ, SJZ, TJ and QHD stations in (a) spring, (b) summer, (c) autumn and (d) winter seasons are indicated by lines and scatters. The growth rates averaged over the four sites are drawn with gray columns for each season to represent the MLH growth velocity, and only positive values are shown in the figure.

311

Annual averages of MLH at the BJ, SJZ, TJ and QHD stations were also calculated, and the values were 594±183, 464±183, 546±197 and 465±175 m, respectively. The MLH at SJZ was approximately 21.9, 15.0 and 0.2 % lower than at the BJ, TJ and QHD stations, respectively. Therefore, according to the analysis above in sections 3.1 and 3.2, an obvious phenomenon can be observed in the MLH distribution on the NCP: the MLH in southern Hebei was lower than in the northern NCP in spring, 323 autumn and winter but was almost equal to the northern areas in summer.

## 324 **4. Discussion**

Through preliminary study of the spatiotemporal variation of MLH on the NCP region, we found something interesting: (a) the MLH at the coastal site was lower than the inland sites in summer; (b) the MLH in southern Hebei was lower than the northern NCP in spring, autumn and winter, but was almost consistent between these two areas in summer. Reasons for these two phenomena will be illustrated in the following sections (4.1 and 4.2). Finally, we will investigate the meteorological evidence for serious haze pollution in southern Hebei in section 4.3.

## 332 4.1 The TIBL impact in coastal site

From the studies in sections 3.1 and 3.2, we found that the maximum MLH at the 333 334 QHD station was larger and arrived earlier than the BJ, SJZ and TJ stations in summer (Fig. 4b). However, this characteristic was not evident in other seasons (Figs. 4a, 4c 335 and 4d). The sea-land breeze was a local circulation that occurs when there is no 336 large-scale synoptic system passes. In our study, we first excluded days with 337 large-scale synoptic systems. Then, according to the coastline orientation, if the 338 southeast wind at the TJ station and south and southwest winds at the QHD station 339 340 occurred at approximately 11:00 LT, and the northwest wind started to blow at approximately 20:00 LT, then this type of circulation was supposed to be a sea-land 341 circulation. The prevailing southeast wind at the TJ station and the south and 342 southwest wind at the QHD station were regarded as sea breezes (Fig. 5). 343

The sea breeze usually brings a cold and stable air mass from the sea to the coastal 344 345 region. When the top of the local mixing layer was higher than the top of the air mass, 346 a TIBL will develop within the mixing layer under the influence of the abrupt change of aerodynamic roughness and temperature between the land and sea surfaces. Then, 347 the local mixing layer will be replaced by the TIBL. In the presence of warm air on 348 land, the cold sea air advects downwind and is warmed, leading to a weak temperature 349 difference between the air and the ground. In consequence, the TIBL warms less 350 351 rapidly due to the decreased heat flux at the ground, and the rise rate is reduced. In addition, since the TIBL deepens with distance downwind and usually can not extend 352 353 all the way to the top of the intruding marine air, the remaining cool marine air above the TIBL will hinder vertical development of the TIBL (Stull, 1988; Sicard et al., 354 355 2006; Puygrenier et al., 2005; Tomasi et al., 2011). With distance inland, the top of the intruding marine air will enhance and exceed the local MLH; if so, the TIBL will 356 not form, and the TIBL impact will be impaired with distance inland (Stull, 1988). 357 Accompanied by the weak synoptic system and the frequent occurrence of sea breezes 358 in summer, the TIBL formed easily and the MLH peak time and value at the QHD 359 360 station were earlier and lower than other stations (Figs. 3 and 4). For the TJ station, 361 with a distance of approximately 50 km out to sea, the TIBL will not extend so far. Therefore, although the TJ station can be affected by the sea breeze, the local MLH 362 363 cannot be influenced by the TIBL.



364

365 Fig. 5 Monthly diurnal wind vectors at the BJ, SJZ, TJ and QHD stations from

366 December 2013 to November 2014.

## 367 4.2 Reasons for low MLH in southern Hebei

368 Turbulent stability was mainly responsible for the MLH development, and the generation of turbulent energy was highly correlated with the heat flux (mainly 369 sensible heat fluxes) produced by radiation and the momentum flux caused by wind 370 shear (Stull, 1988). As presented in section 2.4, the Ri could describe the turbulent 371 372 stability not only from the perspective of thermal forces but also from the perspective of mechanical forces; it was calculated in this section with meteorological sounding 373 374 profiles to study the reason for MLH differences between southern Hebei and the northern NCP, and the frequency values of Ri>1 were given in this study. With larger 375 376 frequency comes more stable stratification. Considering the geographic locations (Fig. 1), the lack of sounding data at the SJZ station was replaced by sounding data from 377 378 the XT station; meanwhile, sounding data from the LT station was used instead of the data from QHD. Each of the four parameter profiles (WS, shear term, buoyancy term, 379 and the frequency of Ri>1) at the BJ, XT and LT stations are depicted in Fig. 6. The 380 381 profiles were averaged over 8:00 LT and 20:00 LT and vertically smoothed using a 100-m running average to reduce unexpected fluctuations for viewing purposes only. 382





Fig. 6 Vertical profiles of (a, e) horizontal WS, (b, f) shear term, (c, g) buoyancy term and (d, h) frequency of Ri>1 at the BJ, XT and LT stations in summer (upper panel) and winter (lower panel).

387 Using the winter and summer as examples, when we analyzed the seasonal means 388 of shear term and the buoyancy term between the XT and the BJ stations, some 389 distinct features were observed. As shown in Figs. 6f and 6g, the shear term and the buoyancy term in XT was 2.8 times lower and 1.5 times higher than that in BJ within 390 0-1200 m in winter, respectively. The largest discrepancies of the wind shear term and 391 buoyancy term between southern Hebei and the northern NCP could reach  $2.84 \times 10^{-5}$ 392  $s^{-2}$  at the altitude of 800 m and  $3.93 \times 10^{-4} s^{-2}$  at 200 m, respectively. As a result, the 393 frequency of Ri>1 in XT was approximately 1.9 times larger than that in BJ within 394 0-1200 m, leading to a much more stable stratification in southern Hebei (Fig. 6h). 395 396 The shear term, buoyancy term and the frequency of Ri>1 in spring and autumn displayed similar characteristics to those in winter, and the averaged frequency of 397 398 Ri>1 in southern Hebei was approximately 1.5 and 1.3 times larger than those in northern NCP in spring and autumn, respectively (Fig. S3). While in summer, the 399 shear term, buoyancy term and the frequency of Ri>1 were almost the same between 400 southern Hebei and the northern NCP (Figs. 6b, 6c and 6d). 401

402 As a result, the lower MLH in southern Hebei was due to a more stable atmospheric 403 turbulent structure than the northern NCP in spring, autumn and winter. This probably 404 resulted from the frequent effect of cold air on the northern NCP, and such cold air was usually too weak to reach southern Hebei (Su et al., 2004). Then the cold front 405 406 resulting from the cold air system will enhance the wind shear over the northern NCP. In addition, a previous study has revealed that the warm advection from the Loess 407 408 Plateau usually developed from southwest to northeast, and the higher buoyancy term (Fig. 6g) and lower MLH in southern Hebei will be partially related to the enhanced 409

thermal inversion at the altitude of 1500 m (Hu et al., 2014; Zhu et al., 2016). In summer, due to the northward lift and westward intrusion of the subtropical high on the NCP, the diminishing existence of the weak cold air on the northern NCP accompanied with the regional scale strong solar radiation and strong turbulent activities will lead to a small turbulent stability contrast between southern Hebei and the northern NCP.

416 In addition, other researchers proposed that absorbing aerosols above the MLH can be another factor affecting the MLH (Peng et al., 2016; Wang et al., 2013; Li et al., 417 418 2016). Absorbing aerosols gives rise to an increasing temperature aloft but a 419 decreasing temperature at the surface, which will enhance the strength of capping 420 inversion and inhibit the convective ability. In contrast, absorbing aerosols within the 421 mixing layer could reduce the capping inversion intensity despite the reduction in the 422 surface buoyancy flux and raise the MLH (Yu et al., 2002). Considering the higher 423 concentrations of surface PM<sub>2.5</sub> in southern Hebei, absorbing aerosols could have 424 some impacts on MLH development. However, the comprehensive influences from the feedback of absorbing aerosols above and below the MLH are hard to explain 425 426 without sufficient knowledge of vertical variations in absorbing aerosols at the four 427 stations. Additionally, the mixed state and morphology of absorbing aerosols dominant the absorption effects (Jacobson, 2001; Bond et al., 2013). Therefore, 428 without sufficient observation data, it is difficult to discuss the possible influences of 429 air pollution feedback on MLH development in this study. Elaborate experiments of 430 vertical profiles and the morphology of absorbing aerosols are needed in future 431 432 studies.

## 433 **4.3 Meteorological evidence of serious air pollution in southern Hebei**

When we analyzed the near-ground PM2.5 and PM10 concentration distributions on 434 the NCP from December 2013 to November 2014, a unique phenomenon was found 435 and shown in Fig. 1 and Fig. S4. The annual means of near-ground PM2.5 436 concentration in southern Hebei (SJZ, XT and HD) was 133.3 µg m<sup>-3</sup> (225.3 µg m<sup>-3</sup> 437 for the PM<sub>10</sub> concentrations), while in the northern areas (BJ and TJ), it was 86.5 µg 438  $m^{-3}$  (126.0 µg  $m^{-3}$  for the PM<sub>10</sub> concentrations), and the difference in the near-ground 439 PM<sub>2.5</sub> concentration between the two areas can be as high as 1.5-fold (1.8-fold for the 440 PM<sub>10</sub> concentrations). Since AOD represents the aerosol column concentration, it is a 441 442 much better indicator for the emissions difference than the PM<sub>2.5</sub>. Additionally, the averaged annual AOD in southern Hebei was only 1.2 times of that in the northern 443 NCP (Fig. 7). If the difference in AOD represents the emission discrepancy, the 444 445 remaining differences of PM<sub>2.5</sub> may be induced by the meteorology. In other words, meteorological conditions may play an important role in heavier haze formation in 446 447 southern Hebei and the meteorological condition contrast between these two areas 448 may contributed at most approximately 60% (considering the aerosol-radiation interactions) to the PM<sub>2.5</sub> concentration discrepancy. 449





Fig. 7 Distribution of the annual mean values of AOD from December 2013 to
November 2014 in the NCP. The PM<sub>2.5</sub> concentrations of the 13 observation sites
were also marked beside each station. The major sites in the northern NCP (BJ and TJ)
and southern Hebei (SJZ, XT and HD) are enclosed by white rectangles.

Previous studies revealed that the most significant meteorological factors for 455 456 regional heavy haze formation in the NCP were RH and MLH (Tang et al., 2016; Zhu et al., 2016). In addition, the T influences the particles' physicochemical reaction rate 457 458 and the ventilation coefficients (V<sub>c</sub>) can be used as an index to evaluate the total 459 diffusion ability of the atmosphere; thus, the RH, T and V<sub>c</sub> were compared and analyzed among the four stations (BJ, SJZ, TJ and QHD) in the next section. The 460 regional particle growth and the atmospheric dissipation ability will be discussed 461 462 separately, each from a meteorological point of view.

## 463 **4.3.1 Meteorological factors for particle growth**

Distributions of annual means of T and RH are shown in Fig. 8, and the 464 distributions of seasonal means of T and RH were added in Figs. S5 and S6. The T 465 value in southern Hebei was similar to that on the northern NCP but was higher than 466 that at the coastal site (Figs. 8a and S5). This indicated an almost consistent 467 468 temperature condition for an atmospheric physicochemical reaction (Garratt et al., 1994; Zhang et al., 2010). However, differences existed in RH between southern 469 Hebei and the northern NCP. The RH in the SJZ station was always higher than that in 470 the BJ and TJ stations but was slightly lower than that at the coastal sites (Figs. 8b and 471 S6). The annual averages of RH at the BJ, SJZ, TJ and QHD sites were 51.2, 65.7, 472 473 57.0 and 68.6 %, respectively, and the RH at SJZ was 22.1 and 13.2 % higher than 474 that at the BJ and TJ sites, respectively (Table S3). Since RH is a key factor for haze development, higher RH is beneficial to fine particle growth through hygroscopic 475 476 growth processes and heterogeneous reactions (Zhao et al., 2013; Fu et al., 2014; Liu et al., 2011; Hu et al., 2006; Wang et al., 2016; Zhang et al., 2015; Seinfeld et al., 477 1998). Thus, a higher RH provided a favorable meteorological condition for haze 478 479 development, which could be partially responsible for heavier pollution in southern



Fig. 8 Distribution of annual means of (a) T and (b) RH in the NCP region fromDecember 2013 to November 2014.

## 484 4.3.2 Meteorological factors for particle dissipation

As MLH and WS can represent the atmospheric dissipation capability in the 485 vertical and horizontal directions, respectively, in addition to the MLH, we analyzed 486 the WS variations on the NCP. Similar to our analysis in section 4.2, as SJZ and QHD 487 488 had no sounding data and due to the close geographic proximity among SJZ and XT as well as LT and QHD, sounding data from the XT and LT stations were used instead 489 of the data at SJZ and QHD, respectively. The WS profiles were averaged every 100 490 491 m at each station and are depicted in Figs. 6 and S3. Except for summer, the WS in southern Hebei was far less than that on the northern NCP in spring, autumn and 492 493 winter (Figs. 6e, S3a and S3e) but was nearly consistent in summer (Fig. 6a). This 494 finding indicated a weaker horizontal diffusion capability in southern Hebei than that 495 on the northern NCP.

The  $V_c$  is an important factor in pollutant dissipation and air quality studies; it accounts for the vertical dispersion and advection of pollutants. With a larger  $V_c$ , strong dissipation ability follows. The  $V_c$  is defined as the product of MLH and wind transport ( $U_T$ ) as shown in Eq. (2).

500

502

 $V_c = MLH \times U_T$  (2)

501 When we utilized the wind profiles in Figs. 6 and S3 with equal spacing in the vertical

direction, U<sub>T</sub> could be regarded as the mean wind transport, i.e., U<sub>T</sub>= $\frac{1}{n}\sum_{i=1}^{n}U_i$  where

Ui is the WS observed at each level and n is the number of levels within the mixing 503 layer (Nair et al., 2007). Since the WS was a climatic parameter, the WS profiles at 504 08:00 LT and 20:00 LT were used to approximate V<sub>c</sub> approximately. Considering the 505 monthly averaged MLH at the BJ, SJZ and QHD stations, the monthly Vc were 506 507 depicted in Fig. 9. V<sub>c</sub> at southern Hebei was always lower than that in the northern NCP during the whole study period. The seasonal means of Vc at the BJ, SJZ and 508 QHD stations in spring, summer, autumn and winter were 4112.0, 2733.3 and 4008.5; 509 3227.5, 2908.8 and 2593.7; 2481.4, 1421.9 and 2581.7; and 2397.2, 1117.7 and 510 2900.0 m<sup>2</sup> s<sup>-1</sup>, respectively. It was clear that the SJZ station usually had the lowest  $V_c$ , 511 and the annual averaged Vc at SJZ was almost 1.5 and 1.5 times smaller than the BJ 512 and QHD stations, respectively (Table S3). As a result, the particle dissipation 513

514 capability in southern Hebei was much weaker than that in the northern NCP and 515 coastal areas.

516





Fig. 9 Seasonal variations of  $V_c$  at the BJ, SJZ and QHD stations from December 2013 to November 2014. The  $V_c$  is defined as the product of MLH and wind transport (Nair et al., 2007) (Eq. (2)). With a larger  $V_c$ , strong dissipation ability follows.

Therefore, with lower MLH, lower WS and higher RH occur in southern Hebei compared to the northern NCP, the near-ground  $PM_{2.5}$  showed a large contrast between these two areas. However, the AOD had little difference between southern Hebei and the northern NCP. Apart from the emission contrast, the meteorological condition contrast between these two areas heavily contributed to the heavy haze in southern Hebei and the industrial structure is in need of readjustment for the NCP (Fig. 10).



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Fig.10 The schematic diagram of the meteorological causes for heavy haze insouthern Hebei.

## 531 5. Conclusions

To gain new insight into the spatiotemporal variation of the regional MLH, the present study conducted a simultaneous observation with ceilometers at three inland stations (BJ, SJZ, and TJ) and one coastal site (QHD) to obtain high spatial and temporal resolution MLH data. The experiment period lasted for 22-months from October 16, 2013, to July 15, 2015, and a whole year of data (from December 2013 to November 2014) were utilized for further study. Conclusions were drawn as follows.

The ceilometers can not only retrieve the inland MLH but also retrieve the coastal MLH properly. The MLHs in the inland areas of the NCP were high in spring and summer and low in autumn and winter. While under the impact of TIBL, the coastal
MLH had an opposite variation trend of inland sites and the lowest MLH in QHD
occurred in summer. The TIBL impaired the local MLH development at the coastal
site and caused the mixing layer to peak early in summer; this effect weakened with
distance inland.

The MLH in southern Hebei was lower than that on the northern NCP, especially in spring, autumn and winter. This mainly resulted from the more stable turbulent structure (weak shear term, higher buoyancy term and larger frequency of Ri>1) than the northern NCP, and the stable stratification in southern Hebei was partially related to the Siberian High and warm advection from the Loess Plateau. In summer, the atmospheric stability was almost consistent between southern Hebei and the northern NCP, and the MLHs between these two areas were nearly identical.

552 The lower MLH and WS in southern Hebei restricted the atmospheric environmental capability and the pollutant dissipation ability, respectively. 553 Accompanied by higher RH values (stronger pollutant growth ability), the adverse 554 weather conditions will cause severe haze to occur easily in southern Hebei, and the 555 industrial layout in the NCP is in need of restructuring. Heavily polluting enterprises 556 557 should be relocated to locations with better weather conditions (e.g., some northern areas and coastal areas), and strong emission reduction measures should be 558 implemented in the remaining industrial enterprises to improve air quality. 559

Overall, the present study is the first to conduct a long-term observation of the 560 MLH with high spatial and temporal resolution on a regional scale. The observation 561 562 results will be of great importance for model parameterization scheme promotion and provide basic information for the distribution of weather conditions in the NCP region. 563 The deficiency of this study is that we did not account for the transport effect on 564 PM<sub>2.5</sub> concentrations. Because pollutants are usually transported from south to north 565 in the NCP region during haze episodes (Zhu et al., 2016; Tang et al., 2015), pollutant 566 transport has a greater impact on the northern areas and has less influence on the 567 568 results of this analysis. The absence of sounding data at noon is another shortcoming, and the daytime observations will be implemented in future experiments. 569 570 Nevertheless, with the data only at 8:00 LT and 20:00 LT, we still provide a fundamental knowledge about the reasons for MLH contrast between northern NCP 571 572 and southern Hebei, and our study can provide reasonable and scientific suggestions for industrial layout and air pollution emission reduction measures for the NCP 573 region., which This will be of great importance for achieving the integrated 574 575 development goals.

576

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