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Atmospheric Chemistry and Physics Discussions

Rapid formation of intense haze episode in Beijing

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Abstract

Although much efforts have been put on studying air pollution, our knowledge on the mechanisms of frequently occurred intense haze episodes in China is still limited. In this study, using three years of measurements of air pollutants at three different height levels on a 325-meter Beijing meteorology tower, we found that a positive particulate matter-boundary layer feedback mechanism existed at three vertical observation heights during intense haze polluted periods within the mixing layer. This feedback was characterized by a higher loading of $PM_{2.5}$ with a shallower mixing layer. Measurements showed that the feedback was related to the decrease of solar radiation, turbulent kinetic energy and thereby suppression of the mixing layer. The feedback mechanism can explain the rapid formation of intense haze episodes to some extent, and we suggest that the feedback mechanism should be considered in air quality models for better predictions.

1. Introduction

With the rapid economic growth and urbanization, an increasing frequency of haze episodes along with the air pollution has become of great concern in China during the last decade (Cao et al., 2016; Huang et al., 2014; Kulmala, 2015; Wang et al., 2014; Wang et al., 2015). For example, during December 2016 a series of intense haze episodes took place in Eastern China, characterized by surface $PM_{2.5}$ concentrations exceeding 500 ug m^{-3} in several measurement sites in Beijing and its surrounding sites (http://www.mep.gov.cn/gkml/hbb/qt/201701/t20170102_393745.htm). Severe air pollution has serious effects on human health. A recent study reported that the particulate matter has significantly decreased the life span of residents as many as 5.5 years in Northern China (Chen et al, 2013). In a global scale, the air pollution was estimated to cause over 3 million premature deaths every year (Lelieveld et al., 2015).

Increased emissions from fossil fuel combustion due to vehicle traffic, industrial activities and power generation, along with exceptionally strong secondary aerosol formation, were thought to be responsible for these haze episodes (Cheng et al., 2016; Huang et al., 2014; Pan et al., 2016; Petäjä et al., 2016; G Wang et al., 2016a; Zhang

et al., 2015; Zhao et al., 2013). Meanwhile, the formation of intense haze episodes was considered to be affected by meteorological conditions (Wang et al., 2014; Quan et al., 2013; Wang et al., 2016b; Zheng et al., 2016). For example, the mixing layer height is a key parameter that constrains the dilution of surface air pollution, and the development of mixing layer is highly related to the amount of solar radiation absorbed by the air and reaching the surface (Ding et al., 2016; Stull, 1988; Sun et al., 2013; Tang et al., 2016; Wilcox et al., 2016).

In this study, using unique measurements on the Beijing 325-meter-high meteorology tower, we present direct evidence on the feedback that relates the decreasing mixed layer height with increasing particulate matter concentrations, and this feedback is critical to the formation of intense haze episodes in Beijing.

2. Methods

2.1 Calculation of mixing layer height with ceilometer

The mixing layer height was measured with the enhanced single-lens ceilometers (CL 31, Vaisala, Finland), which utilized the strobe laser lidar technique (910 nm) to measure the attenuated backscattering coefficient profiles. Detection range of the CL31 is 7.6 km with the report period of 2-120 s. Detail information can be found in previous studies (Tang et al., 2016). Since the distribution of particle concentrations is uniform in the mixing layer and has significant differences between the mixing layer and free atmosphere, the height at where a sudden change exists in the attenuated backscattering coefficient profile indicates the top of the mixing layer height (Münkel et al., 2007). The Vaisala software product BL-VIEW was used to determine the mixing layer height by finding the position with the maximum negative gradient ($-d\beta/dx$) in the attenuated backscattering coefficient profiles as the top of the mixing layer (Tang et al., 2016;Münkel et al., 2007).

2.2 Measurements of energy flux at 325m Beijing meteorology tower

The turbulent fluxes of sensible heat (Q_H), latent heat (Q_E) and The turbulence kinetic energy (TKE) were measured at 140m level using eddy covariance technique. The raw data (10 Hz) of wind components (u, v and w) and sonic temperature (Ts) recorded with three-dimensional sonic anemometers (Model CSAT3, Campbell Scientific Inc., Logan, Utah, USA) and of water vapor

concentrations (q) with open-path infrared gas analyzers (Model LI-7500, LiCor Inc., Lincoln, Nebraska, USA). The fluxes of heat (Q) were calculated as the covariance between the instantaneous deviation or fluctuations of vertical velocity (w_i') and their respective scalar (s_i') averaged over a time interval of 30 min:

$$Q = \overline{w's'} = \frac{1}{N}\sum_{i=1}^{N} w's'$$

Where the over-bar denotes a time average, N is the number of samples during the averaging time and the fluctuations are the differences between the instantaneous readings and their respective means. The TKE were calculated as follows (Stull, 1988):

$$\frac{TKE}{m} = \frac{1}{2}\left(\overline{u'} + \overline{v'} + \overline{w'}\right) = \overline{e}$$

where m is the mass (kg), e is the TKE per unit mass ($m^2\ s^{-1}$). A more detailed description of the calculation and post processing of flux is provided in Song et al. (2013).

2.3 Measurements of $PM_{2.5}$ concentration and gases in the 325m Beijing meteorology tower.

The mass concentration of $PM_{2.5}$ at 8m, 120m and 280m observation platform were measured with three TEOM RP1400 simultaneously. (Thermo Scientific, http://www.thermoscientific.com). The resolution and precision of the instrument for one-hour was of 0.1 μg m^{-3} and ±1.5 μg m^{-3}. The filters were exchanged when the loading rates were approximately to 40% and the flow ratio were monitored and calibrated monthly. The volume mixing ratios of ozone and NOx were measured with 49i and 42i (Thermal Environment Instruments (TEI) Inc.), respectively. Detailed introduction can be found in Wang et al. (2014).

2.4 Other supporting measurements

Total solar radiation was measured with a direct radiometer (TBQ-2, Junzhou, China). Direct radiation was measured with a direct radiometer (TBS-2, Junzhou, China). UV radiation in the range of 400nm-220nm was measured using CUV3 radiometer (USA). The estimated experiment error for the three instruments are 3%, 1% and 2%, respectively (Hu et al., 2012). The original data were obtained at one-minute intervals and the hourly average values were used in this study. The chemical composition of organic, sulphate, nitrate, ammonium and chloride in non-refractory submicron aerosol were measured with an Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-Tof-AMS, Aerodyne Research Inc., Billerica, MA, USA) (DeCarlo et al., 2006). Detailed information about instrument, calibration and data process have been introduced by Zhang et al. (2014).

3 Results and Discussion

A typical intense haze episode occurred during the heating season in urban Beijing during 17 to 22 November 2010. This episode was associated with synoptic stagnation in the North China Plain (Figure S1) and was characterized by low wind speeds and irregular wind direction (Figure 1). Several meteorological variables had distinct temporal patterns during different stages of pollution, including reduced solar radiation and surface temperatures and increased relative humidity during the most intense presence of haze (Fig. 1) The temporal patterns of $PM_{2.5}$ concentrations were very similar at the two lower measurements heights (8 m and 120 m, Fig. 1d), even though the concentration was clearly the highest close to the surface. The $PM_{2.5}$ concentration measured at 280 m behaved in a different way, especially during the most intense period of the haze when the mixed layer height was very low (Fig. 1e). The decoupling of the 280-m platform from the other two lower ones at low mixed layer heights is apparent in our 3-year measurement data set, especially when comparing O_3 and NO_x concentrations between the three measurement platforms (Figs. S2 and S4). During the haze period, the maximum $PM_{2.5}$ concentrations at 8, 120 and 280 m were 505, 267 and 339 g m^{-3}, respectively. The higher maximum concentration at 280 m compared with 120 m can be ascribed to the transport of pollutants from surrounding regions of Hebei and Tianjin Provinces typical for polluted periods (Sun et al., 2013). The mixing layer height varied from 130 m to 1640 m during the haze episode, ranging between about 200 and 500 m during the most intense period of the haze period on 18 November 2010 (Fig. 1e).

The vertical distribution of attenuated backscatter density obtained from ceilometer measurements indicate vertical mixing conditions accompanied with an inversion layer and high relative humidity in the surface as shown in Figure 2. The strong inversion and high relative humidity occurred on morning of 18 November 2010, with a lapse rate of 2K / 100 m, relative humidity of 78% and north-direction wind speed of around 2 m / s detected by the vertical sounding. The turbulent kinetic energy at 140 m was reduced to around 0.1~0.7 m^2/s^2 due to decreased solar radiation, as shown in Figure1(a). In this manner, the development of a mixing layer was significantly suppressed during the intense haze episode.

In order to demonstrate how the mixing layer height modifies $PM_{2.5}$ concentrations, we used three years of simultaneous winter-time air pollutant measurements in the Beijing tower at 8 m, 120 m and 280 m platforms. We divided

the observed $PM_{2.5}$ concentrations into highly-polluted and less-polluted conditions using a threshold value of 75 ug m^{-3} for $PM_{2.5}$ to distinguish between these conditions. This is consistent with Chinese Environment Protection Bureau definition of a haze pollution events. With this threshold value, we found that 31% and 69% of total measurement time corresponded to highly-polluted and less-polluted conditions, respectively. We plotted the $PM_{2.5}$ data as a function of the mixing layer height at the three observation heights during both highly-polluted and less-polluted conditions and fitted an exponential curve to these data (Figure 3). The $PM_{2.5}$ concentration has a clear anti-correlation with the mixing layer height during the intense haze episodes. At all measurement heights, the $PM_{2.5}$ concentration increased as the mixing layer height decreased, and this pattern was very strong under polluted conditions (Figure 3). It is worth noting that the increase was mainly from the $PM_{1\text{-}2.5}$ fraction that increased from 42% to 65% as mixing layer height decreased from more than 1400 m to lower than 300 m (Figure S3b). A major portion of particulate mass between 1 and 2.5 μm originates from secondary aerosol formation processes in urban air (Wang et al., 2014; Zhang et al., 2015). The reduction in solar radiation due to these fine particle matters reaching the surface reduces the turbulent kinetic energy and the development of mixing layer, as shown in Figure 4.

We assign part of the observed increase in $PM_{2.5}$ and simultaneous decrease in the mixing layer height to a positive feedback from particulate matter-mixing layer interaction (Petäjä et al. 2016, Ding et al. 2016), which occurred at the same time as primary emissions and secondary formation were confined into a smaller volume of air. The feedback occurred at all the three observation platforms and was most intensive at 8 m. In an urban environment, NO_x originates mainly from local anthropogenic emissions, whereas the sources of particulate matter include bot primary emissions and secondary formation (Ehn et al., 2014; Jimenez et al., 2009; Zhang et al., 2015; Zhao et al., 2013). As shown in Figure S4, the median NO_x concentration at 8 m was 250% higher under highly-polluted conditions compared with less-polluted conditions as the mixing layer height decreased to 100-200 m, while the corresponding number for the $PM_{2.5}$ concentration was 360%.

The increase of the $PM_{2.5}$ concentration from less-polluted to highly-polluted conditions is mainly due to concentrated particulate matter caused by a decreased mixing layer height, which is accompanied by primary particle emissions, secondary

aerosol formation and feedback from particulate matter-mixing layer height interactions. Compared with the increased amounts of NOx, we can roughly estimate that in maximum 110% of the increased $PM_{2.5}$ originates from secondary aerosol formation processes in this study. Of the remaining 250% of the $PM_{2.5}$ increase, potentially a large fraction originates from particulate matter-mixing layer height interactions, but we cannot quantify this fraction at the moment.

4 Conclusions

The development of mixing layer height in an urban city is affected by the intensity of incoming solar radiation and by anthropogenic heating in the city. Our measurement at the 325-meter meteorology tower showed that the solar and ultraviolet radiation reaching the surface decrease considerably at increased pollution levels., which leads to further increases in concentrations of $PM_{2.5}$ and its precursor gases from both direct emissions and secondary formation. This feedback mechanism may be an important reason for rapid increase of particulate matter from moderate-polluted conditions to periods of intense pollution in an urban atmosphere. The particulate matter-mixing layer height feedback is probably a critical factor for the formation of intense haze periods in Beijing and other polluted cities.

Acknowledgements

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Competing financial interests

The authors declare no competing financial interests.

Author contributions

M.K, Y.S.W, T.P and Y.H.W, have the original idea. Y.S.W, G.T, T.S, Z.L, B.H, L.W, X.Z, D.J, W.G and Y.S conducted the longtime measurements and provided the data. Y.H.W, G.T, S.T, P.Z, M.E, C.Y, V.K, T.P and M.K interpreted the data and plotted the figures. Y.H.W wrote the manuscript, with contribution from all co-authors.

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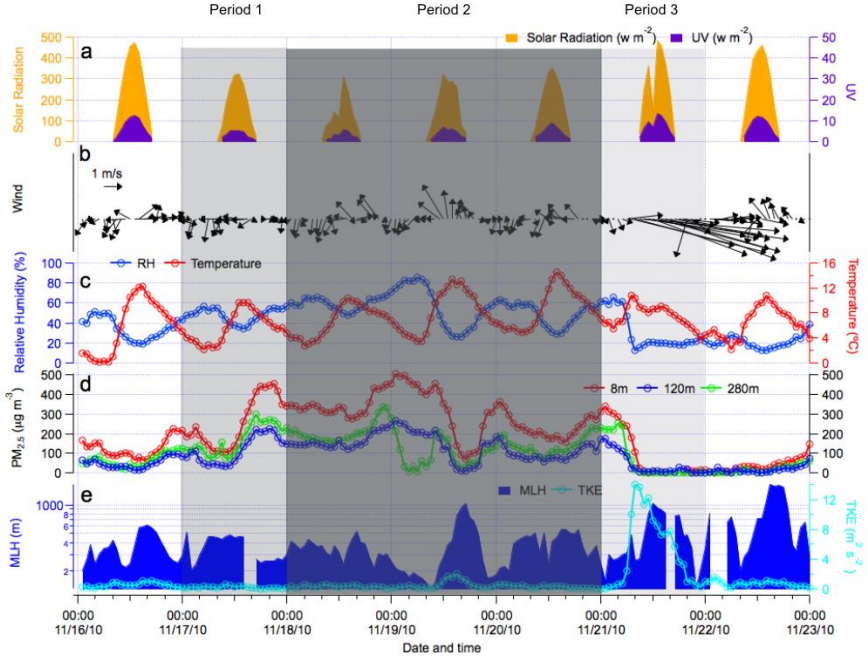
376 Figure captions



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378

379 **Figure 1**. Measurements of (a) solar radiation and ultraviolet radiation at 8 m, (b)
380 wind speed and direction at 8 m, (c) relative humidity and air temperature at 8 m, (d)
381 mass concentration of $PM_{2.5}$ at 8 m, 120 m and 280 m, (e) mixing layer height at 8 m
382 and turbulence kinetic energy at 140 m in the Beijing 325-meter meteorology tower
383 during an intensive air pollution episode in November of 2010. The evolution of the
384 air pollution episode can be divided into the period 1 （clean period to air pollution
385 accumulation period, period 2 (pollution period) and period 3 (pollution to clean
386 period).

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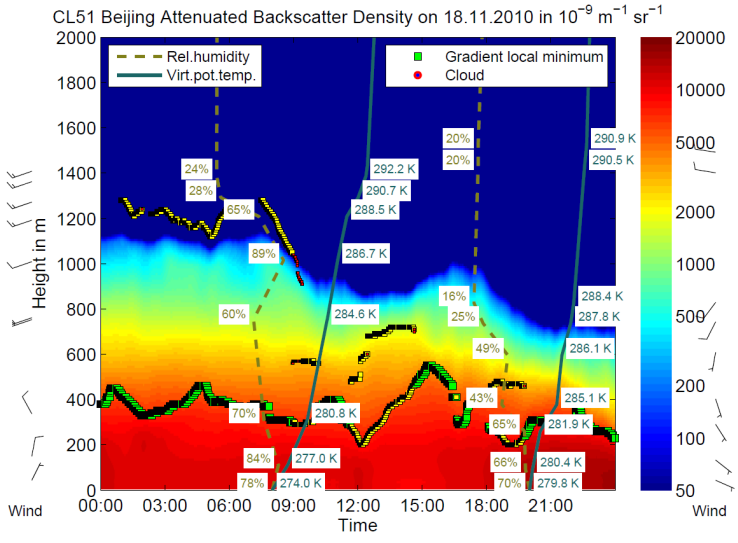

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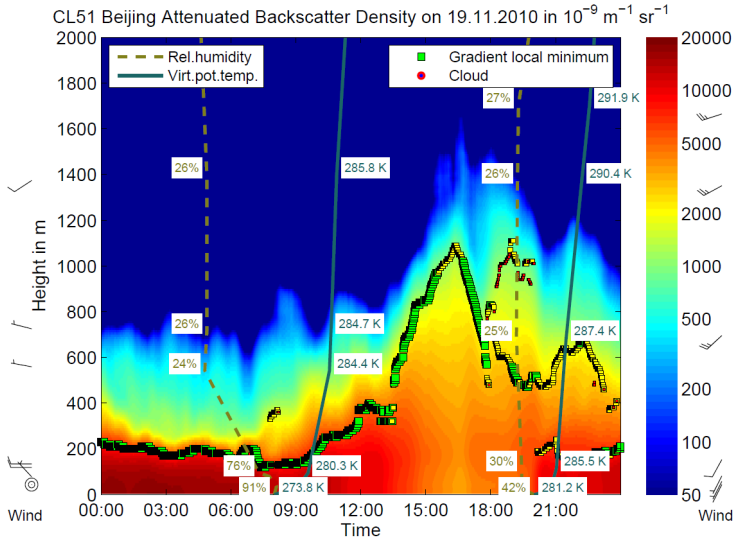

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391 **Figure 2**. Observed attenuated backscatter density, calculated mixing layer height
392 using ceilometer and vertical wind speed, wind direction, relative humidity, virtual
393 potential temperature using sounding data during November 18 (top) and 19 (bottom).
394 The black flag in the left and right side of the figures stand for vertical wind speed
395 and wind direction obtained from sounding measurements at 08:00 and 20:00 of
396 Beijing time, respectively. The circle in the left side of figure stands for calm wind.
397 The dotted yellow lines and solid green lines stand for vertical distribution of virtual

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potential temperature and relative humidity from sounding at 08:00 and 20:00, respectively. The mixing layer height was determined from the local minimum of the backscatter density gradient, and the colour in the figure stands for backscatter density from ceilometer. From both figures, we can clearly see that mixing layer has important role in regulating distribution of air pollutants.

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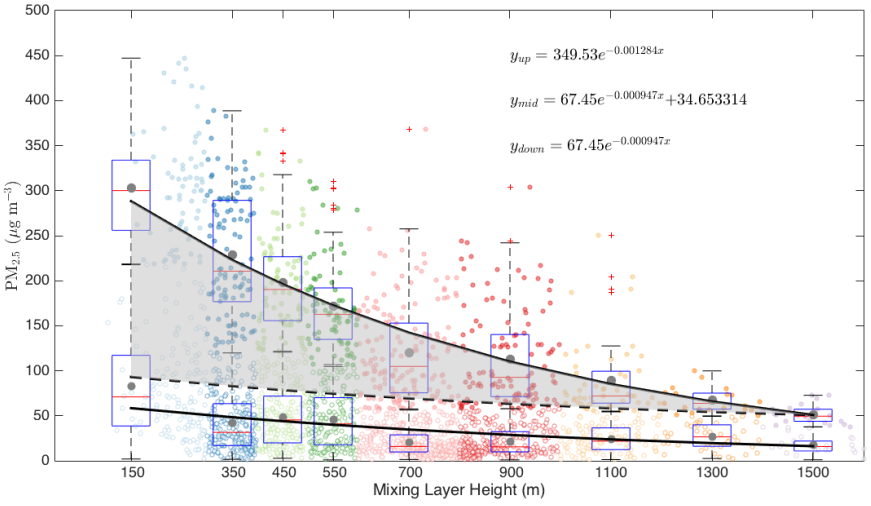

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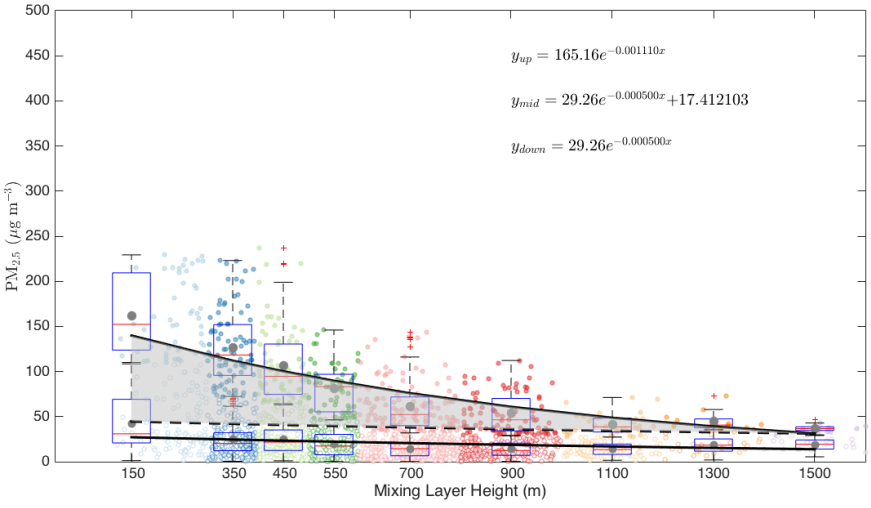
406 (a)



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408 (b)

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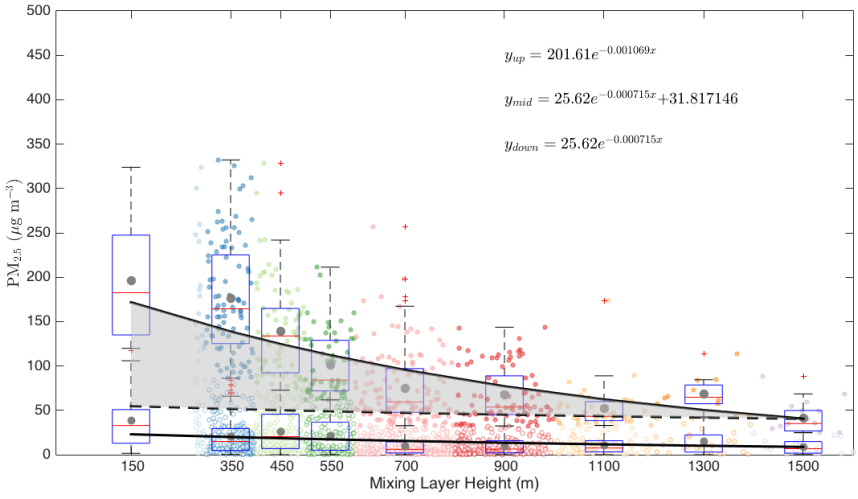

(c)

Figure 3. The variability of the $PM_{2.5}$ mass concentration as a function of the mixing layer height at 8 m (a), 120 m (b) and 280 m (c). The data related to the upper fitting line represents $PM_{2.5}$ concentrations larger than 75 ug m^{-3}, while the date related to the lower fitting line represents $PM_{2.5}$ concentrations less than 75 ug m^{-3}. The dark grey points represent mean values; the red line represents median values. The shadowed area corresponds to an increased amount of $PM_{2.5}$ with decreased mixing layer height assuming that $PM_{2.5}$ has the same variation pattern under highly- polluted conditions as in less polluted time.

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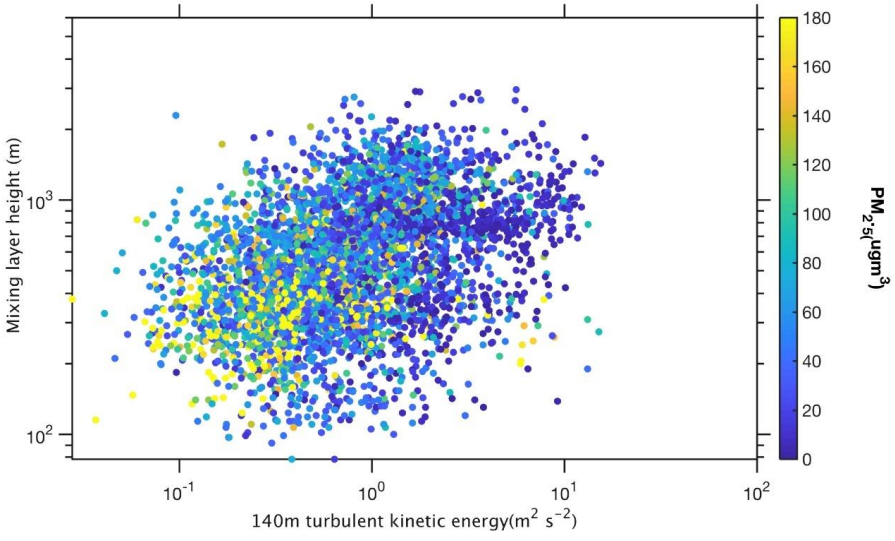

Figure 4. Turbulent kinetic energy at 140 m as a function of mixing layer height and $PM_{2.5}$ concentrations at 140 m.