

## A point to point response to the reviewers' comments

We thank the two reviewers for their comments, and we do think their comments and suggestions improved our manuscript a lot. Here are points to points responses (in blue colored), accordingly, we also revised manuscript (in red colored).

### Reviewer #1

This paper presents valuable observational evidence for interactions between boundary layer mixing properties and haze aerosol loading. It describes an important set of long-term measurements from a polluted urban location in Beijing, combining atmospheric composition measurements at a number of different altitudes with ceilometer measurements of atmospheric mixing height. This provides a unique opportunity to provide conclusive evidence for a feedback between aerosol and mixing height, but unfortunately this has not been fully achieved, and the claims that are made are not fully supported.

The study shows that PM is higher when the mixing height is lower, but does not present deeper analysis. The inverse relationship between aerosol and mixing height may represent a dynamical feedback, or it may just reflect the one-way control of PM levels by mixing height. It is ascribed to the former here (line 178), but the evidence provided to back this up is weak, and no argument is presented on why the simpler mechanism (reduced vertical mixing alone) is insufficient to explain it. This needs to be addressed before the paper is suitable for publication.

The ratio between NO<sub>x</sub> and PM shown in Fig S5 could potentially provide valuable new insight into the mechanisms involved, but this is not explored in the paper. Similarly, increases in secondary aerosol production shown in Fig S6 are interesting but not properly explored. Neither Fig S5 nor Fig S6 are even referred to in the paper.

The paper calls for the feedback between aerosol and boundary layer turbulence to be included in air quality models. In practice online models already include this process, and what is missing is strong observational evidence to back up existing theory and validate our current understanding. The study described in this paper has the potential to provide this important evidence, and I strongly encourage the authors to complete this analysis and revise their paper, after which it could make a valuable and important contribution to the scientific literature.

**Response: we thank the reviewer for careful reading and the comments. We think these comments are really important to improving the manuscript. We calcified the reviewer's comments to two comments, as following:**1. The study shows that PM is higher when the mixing height is lower, but does not present deeper analysis. The inverse relationship between aerosol and mixing height

may represent a dynamical feedback, or it may just reflect the one-way control of PM levels by mixing height. It is ascribed to the former here (line 178), but the evidence provided to back this up is weak, and no argument is presented on why the simpler mechanism (reduced vertical mixing alone) is insufficient to explain it. This needs to be addressed before the paper is suitable for publication. **We agree that inverse relationship**

between aerosol and MLH represent a dynamical feedback. The increase of PM will decrease MLH by reduce solar radiation and thereby TKE, and decreased MLH will also lead to more secondary formation due to concentrated precursors. Also, we carefully ascribed partly the increase in PM<sub>2.5</sub> and simultaneous decrease in the mixing layer height to a positive feedback from particulate matter-mixing layer interaction, since other factors may work during this process. For example, a recent study by Liu et al., (2018) suggested that relative humidity played an important role in feedback between secondary particle matter and boundary layer height. 2. The ratio between NOx and PM shown in Fig S5 could potentially provide valuable new insight into the mechanisms involved, but this is not explored in the paper. Similarly, increases in secondary aerosol production shown in Fig S6 are interesting but not properly explored. Neither Fig S5 nor Fig S6 are even referred to in the paper.

The sources of NOx and PM are mainly from surface emission and formation, therefore, the concentrations are both higher at lower observation platform than concentrations at higher observations platform. We added statements in our revised manuscript about figure s6, as following: As shown in figure S7, the chemical concentration of NR-PM<sub>1</sub> increased significantly from 12.1  $\mu\text{g m}^{-3}$  to 56.4  $\mu\text{g m}^{-3}$  with the variation of MLH decreased from more than 1400 m to less than 200 m

#### General Comments

The conclusions of the paper remain weak and speculative, principally because clear evidence has not been provided in the paper to support the suggestions made.

The figures in the supplementary material are not labelled or described well, and their relevance to the body of the paper is unclear. Please justify their inclusion carefully.

Figures S5-S7 are not referred to in the text, and should be introduced or removed.

Further specific comments are provided below.

The written English in the paper is reasonable but does not fully reach the standards needed for a scientific publication, and some polishing of this will be required in the final version.

Response: Thanks for the comment. We have revised the manuscript on grammars and writing.

#### Specific Comments

Page 3: particle concentrations are often not uniform in the mixed layer, particularly near sources or where there are residual layers present. How does the approach used resolve multiple layering? The maximum gradient approach used has known deficiencies; how sensitive is the mixing height retrieval to the approach taken?

Response: Thanks for the comment. The maximum gradient approach was used in our study to retrieve mixing layer height. Utilizing with approach with ceilometer measurements, three layers can be retrieved within 4000 m height from surface. Usually, the first layer was considered as mixing layer height. In our previous study, we found that mixing layer height retrieved by this method was consistent with sounding data results during polluted days, while the method can not be used during dust conditions and neutral boundary layer (Tang et al., 2016). As the reviewer has pointed out clearly that the distribution of particle matter is not uniform in the mixed layer, particularly during a stable boundary layer occurred. We can not get exact distributions of PM in the vertical scale, so the assumption of relative uniform of PM in the mixed layer could be carefully made. We also recognize that detailed technic work is challenge and needed in future.

Line 80: Please describe where these measurements were made.

Response: Thanks for the comment. The measurements were made in the yard of institute of atmospheric physics (IAP), where the 325 meter tower is located. We also added descriptions on our revised manuscript.

Line 132: Fig 1 does not show reduced temperatures: the temperature appears higher than before or after

Response: Thanks for the correction. We corrected this statement in our revised version.

Line 154-156: The study of TKE is essential for supporting the links between mixing height and PM levels, but it is not mentioned again except in reference to Fig 4 in the following paragraph.

Response: Thanks for the comment. We added more discussion about variation of TKE in Figure 1. The TKE was quite low during this intensive haze episode from 18 November to 21 November, with an average value around  $0.3 \text{ m}^2\text{s}^{-2}$ . However, the TKE increased significant on morning of 21 November as surface wind increased from 1.2 m/s to around 6 m/s, which was possible dur to cold front as shown in Figure S1.

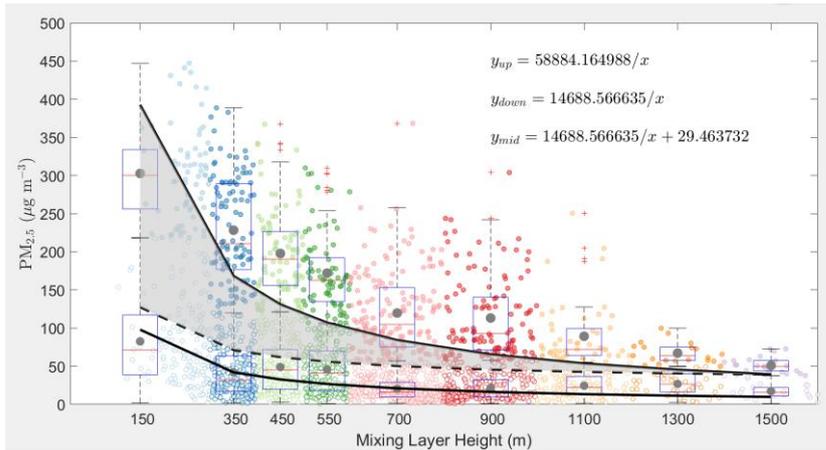
Line 167: What is the justification for fitting an exponential curve to the data in Figure 3? Fitting a reciprocal curve would make more sense (doubling mixing height halves concentrations). Please explain the rationale here and the evidence supporting it.

Response: Thanks for the comment. In general, the reciprocal fitting can not represent a positive feedback. So in this case, the exponential fitting curve is more appropriate. This can also be supported by the root-mean-square error (RMSE) of these two fitting methods. The RMSE of the exponential fitting is much smaller than the reciprocal fitting in any case (Table R1).

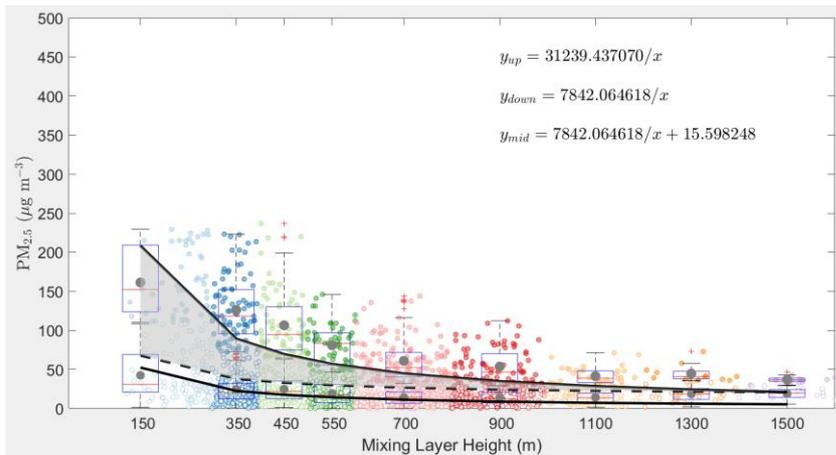
Moreover, we have tested the reciprocal fitting function for the data as shown in Figs. R1, R2, and R3. It overestimated the PM 2.5 concentration when the mixing layer height is very low compared to the exponential fitting function (Fig. 3). This also indicates that a much higher PM 2.5 concentration is needed in order to obtain a very low mixing layer height without a positive feedback.

Table R1: The root-mean-square error (RMSE) in the unit of  $\mu\text{g m}^{-3}$  between fitted functions and measured data.

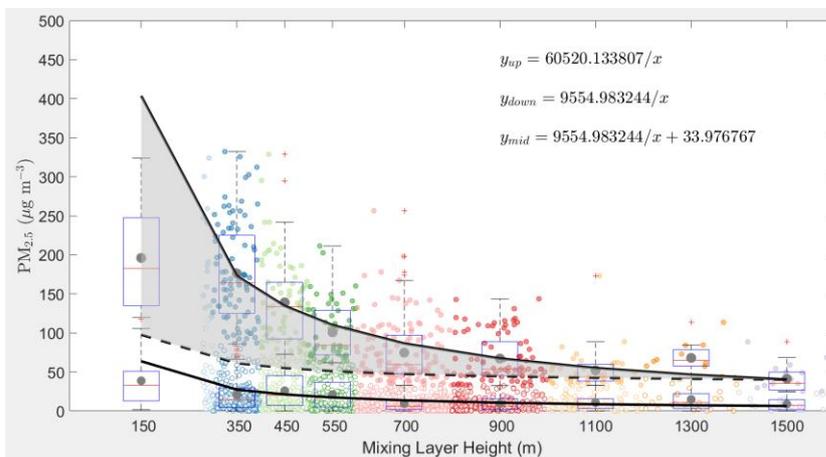
Fitting function	Pollution intensity	At 8 m	At 120 m	At 280 m
reciprocal	High	53.5589	27.8437	69.7236
reciprocal	Low	12.0393	8.2159	9.3756
exponential	High	9.2853	11.4142	19.0527
exponential	Low	10.4951	6.2643	6.6181



8m



120 m



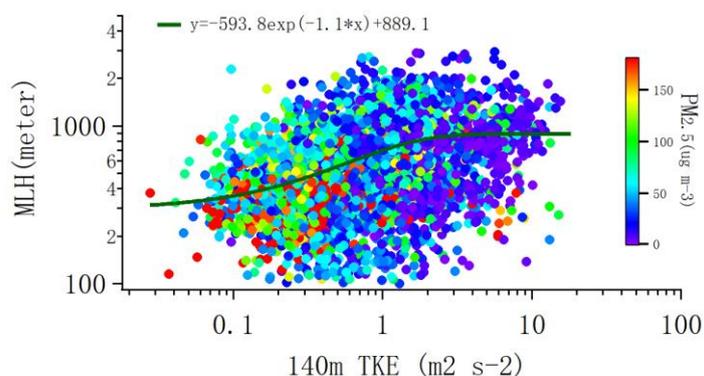
280m

Figure r1 . The variability of the PM<sub>2.5</sub> mass concentration as a reciprocal function of the mixing layer height at 8 m (R1), 120 m (R2) and 280 m (R3).

Line 171-7: These arguments are the most important made in the paper, and need clear observational support. What relationship is shown in Figure 4? What is the signal underlying the scatter? Extracting a quantitative relationship from the data shown here and using it to support the arguments made would strengthen the paper greatly.

Response: Thanks for the comment. We fitted an exponential function between Turbulent kinetic energy at 140 m and mixing layer height. We also added some more statement in our

revised manuscript. An exponential function between Turbulent kinetic energy at 140 m and mixing layer height was fitted, which could provide us some simple quantification. As presented in the function, the MLH will be doubled from around 400 m to 800 m if TKE increased from around  $0.1 \text{ m}^2 \text{ s}^{-2}$  to  $1 \text{ m}^2 \text{ s}^{-2}$ , and these are typical MLHs during polluted conditions in Beijing.



Line 202: Anthropogenic heating isn't mentioned in the paper prior to this.

Response: Thanks for the comment. We removed it in our revised version.

Line 316: The references from this point onward are out of alphabetic sequence in the reference list.

Response: Thanks for the comment. We made it in the corrected order.

Line 391: What is the distinction between the yellow and green mixing height lines in Figure 2? This needs to be explained more clearly in the caption.

Response: Thanks for the comment. The yellow square and green square stand for first layer and second layer, respectively, and usually the first layer was used as mixing layer height.

Fig S1: Please simplify this figure (perhaps by reducing to 4 panels) as there is too much extraneous detail here. Annotate to highlight key features that you want the reader to be aware of. Please label High and Low pressure systems more clearly, and label each panel with the date and time.

Response: Thanks for the comment. We reduced the figures to 4 panels and added date and time in each panels, explanations in figure captions part about the high and Low pressure systems.

Fig S2: Make figure clearer, label panels and state the time period used. There is no analysis of this figure in the text; please state how it contributes to the arguments in the paper (or otherwise remove it).

Response: Thanks for the comment. Actually, the air pollutants were well mixed during daytime, primary pollutants showed higher concentration near surface, while the secondary pollutant, ozone showed higher concentrations in higher altitude. We have analysis at Line 141. 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  $\text{O}_3$  and  $\text{NO}_x$  concentrations between the three measurement platforms (Figs. S2 and S5).

Fig S3: The panels in this figure are small and difficult to read, and there is no apparent connection between them. If they are needed, present as two separate figures. In the first, explain what is meant by direct radiation (what wavelength range? excluding all

diffuse components?) and use a compatible color scale between the two panels so that the results can be compared easily (dark blue is 8m haze in one panel and 280m clean in the other). X-axis labels are missing in the PM1 figure.

Response: Thanks for the comment. The direct radiation is the proportion of the almost rectilinear solar radiation, which reaches the earth's surface from an angle with a distance of 0.25 to the centre of the sun and reaches a normal area, which is oriented perpendicularly to the direction of radiation. We have revised the figures according to your suggestion and presented them as two separate figures. The PM1 figure shared the same X-axis labels as radiation figure, as explained in figure caption, the X-axis was mixing layer height.

Fig S4: Please state the time period and explain x-axis (no axis label is provided)

Response: Thanks for the comment. The data was statistics of two years mixing layer height with variation of PM2.5, NOX and ozone. The X-label was added in the revised version.

Fig S5: Define Fb here. This figure does not appear relevant to the paper, and should be removed.

Response: Thanks for the comment. We have removed the figure.

Fig S5(2): Please correct the numbering of this and subsequent figures

Response: Thanks for the correction. We have numbered the figure correctly.

Fig S6: The increase in secondary aerosol is interesting here, but it would be more informative to show how the contribution of secondary aerosol to total aerosol changes here. However, the figure is not discussed in the paper, and should either be discussed or removed.

Response: Thanks for the comment. Inorganic aerosol like sulfate, nitrate and ammonium are can be considered as secondary aerosol. However, identifying primary and secondary of organic aerosol from a 2 years data needs more time with source apportionment method, and the current work cannot resolve it. We added more discussions in the revised version, as following: As shown in figure S7, the chemical concentration of NR-PM<sub>1</sub> increased significantly from 12.1 ug m<sup>-3</sup> to 56.4 ug m<sup>-3</sup> with the variation of MLH decreased from more than 1400 m to less than 200 m.

Fig S7: What period is this over? Again, this figure should be discussed or dropped.

Response: Thanks for the comment. The measurement was conducted from July 2009 to August of 2012, the figure has been removed.

Typos and minor issues

Please use European date order conventions (day, month, year) throughout. The x-axis labels in Fig 1 use US conventions, while Fig S5 in the supplement uses Chinese conventions.

Response: Thanks for the comment. We revised the Figure 1 and added time and year in X-label.

Title: episode -> episodes

line 107: in -> on (or at)

line 111: remove to

line 118: 220-400?

line 142: microgram unit missing

line 178: assign -> ascribe

line 184: bot ??

line 396-7: stand for -> represents

line 415: date -> data

[Response: Thanks for the correction.](#)

**Reviewer 2#**

This paper characterizes the interactions between atmospheric mixing layer dynamics and fine particulate matter pollution using long-term measurements of vertical distributions of PM<sub>2.5</sub> and NO<sub>x</sub>, atmospheric mixing layer height, vertical meteorological parameters, energy flux, etc. in an urban site in Beijing. Based on the relationship between PM<sub>2.5</sub> concentration, mixing layer height, solar radiation, and turbulent kinetic energy, the authors claimed that they found a feedback mechanism between mixing layer height and fine particulate matter pollution that could explain the rapid formation of severe haze pollution episodes in Beijing.

This work addresses an important topic that are of interest to many of the readers in atmospheric science community. However, many data presented in the paper are not thoroughly analyzed and discussed, and the evidence claimed by the authors are not strong enough to support their conclusion about the aerosol pollution-mixing layer development feedback mechanism. There are many important issues that need to be addressed before the publication of the paper in ACP can be considered.

[Response: Thanks for the comment.](#)

Major comments:

The authors claimed that the fine aerosols can reduce the solar radiation reaching the surface, resulting in a decrease in the turbulent kinetic energy (TKE) and a suppression of atmospheric mixing layer development, which further increase aerosol concentrations from direct emission and secondary formation (i.e., the feedback mechanism).

However, they did not provide clear evidence that fine aerosols play a non-negligible role in regulating TKE and mixing layer heights. As shown in Fig 1, the TKE decreases dramatically from 8:00 to 20:00 on 21 November 2010, while aerosol loadings are pretty low during this period. This suggests that the variation of TKE is largely driven by non-haze related factors. Therefore, to claim the feedback mechanism, it is important to quantify to what extent fine aerosols can reduce or regulate the TKE and the development of the mixing layer in severe haze episodes.

[Response: Thanks for the comment. The increasement of aerosol loading in the atmosphere can suppress vertical development of turbulent activity. However, other factors also will influence the variation of TKE, as the reviewer pointed out, the TKE decreases dramatically from 8:00 to 20:00 on 21 November 2010, while aerosol loadings are pretty low during this period. The dramatically variation of TKE was due to wind shear, as shown in Figure 1\(a\), the wind speed increased dramatically from 1m/s to 6m/s on the surface, so the wind shear could be occurred. As presented in figure s1, a cold front passed Beijing region on 21 November.](#)

Other specific comments:

In the Introduction, the review of literatures is too brief. A summary of the current knowledge and remain issues regarding the interactions between boundary layer dynamics and aerosol pollution should be included, and the novelty of the present study should be clearly pointed out.

[Response: Thanks for the comment. We added a summary of current knowledge and remain issues in our revised manuscript. Also, we pointed out the novelty of the present study. By using](#)

filed measurements combined with model simulation, a positive Feedback between aerosol pollution, relative humidity and boundary layer was important in aerosol production, accumulation and severe haze formation in Beijing (Liu et al., 2018). Wang et al., 2018 found that PBL schemes in their atmospheric chemistry models are not sufficient to describe the explosive growth of PM<sub>2.5</sub> concentration in Beijing-Tianjin-Hebei region due to absence of an online calculation of aerosol-radiation feedback, and/or a deficient description of extremely weak turbulent diffusion. In this study, using unique measurements on the Beijing 325-meter-high meteorology tower, we show clear relationship between mixing layer height and turbulent kinetic energy at 140m observation platform. We also present direct evidence on the feedback that relates the decreasing mixed layer height with increasing particulate matter concentrations.

P4, Sect. 2.3 and 2.4. Please specify the altitude at which the measurements of O<sub>3</sub>, NO<sub>x</sub>, radiation, and aerosol chemical composition were performed.

Response: Thanks for the comment. We added statement that all these measurements were conducted in the IAP station.

Line 111. Remove “to”, and “ratio” should be “rate”.

Response: Thanks for the correction, we corrected.

Line 121-123. The HR-ToF-AMS was used to measure aerosol chemical composition. However, the data were not discussed in the paper, though a figure (Fig. S6) was included in the supplementary material.

Response: Thanks for the comment. We added statements in our revised manuscript as following: As shown in figure S7, the chemical concentration of NR-PM<sub>1</sub> increased significantly from 12.1 ug m<sup>-3</sup> to 56.4 ug m<sup>-3</sup> with the variation of MLH decreased from more than 1400 m to less than 200 m.

Line 138-141 and Figs. S2 and S4. The decoupling of the 280-m platform from the other two lower ones for O<sub>3</sub> was shown to be much smaller than that for PM<sub>2.5</sub> and NO<sub>x</sub>. What is the reason for this difference?

Response: Thanks for the comment. The possible reason was that the concentrations of PM<sub>2.5</sub> and NO<sub>x</sub> were higher near surface than that at higher altitude, while O<sub>3</sub> concentration was higher at higher altitude (within 1000m), also O<sub>3</sub> is a secondary product on the surface.

Line 171-173 and Fig. S3b. Was the PM<sub>1</sub> measured by HR-ToF-AMS? If so, the authors should point out that measured PM<sub>1</sub> mass concentrations do not include the refractory components such as soot and dust, whereas the PM<sub>2.5</sub> concentrations include these components. In addition, compared to PM<sub>1-2.5</sub>, the origin of PM<sub>1</sub> is generally more secondary. Therefore, the increase of PM<sub>1</sub> concentration but decrease of its mass fraction in PM<sub>2.5</sub> (as the decrease of mixing layer height) may offer insights into the contributions of primary emission and secondary formation to the haze pollution.

This merits further discussions in the paper.

Response: Thanks for the comment. The mass concentration of PM<sub>1</sub> was measured by Thermo TEOM 1400, which is the same instrument as PM<sub>2.5</sub> measurement. In general, We agree your statement that PM<sub>1</sub> is more secondary than PM<sub>1-2.5</sub>. However, during intensive haze period, the formation of PM<sub>1-2.5</sub> is more significant than PM<sub>1</sub>, which is possible due to aqueous phase reactions and gas-particle partitioning of gases. For larger sized particles, these two process are more significant (Wang et al., 2015).

Line 184. “bot” should be “both”.

Response: Thanks for the correction.

Some references in the reference list do not follow an alphabetical order.

[Response: Thanks for the correction. We made the correct order in our revised version.](#)

#### Reference

Tang, G., Zhang, J., Zhu, X., Song, T., Munkel, C., Hu, B., Schäfer, K., Liu, Z., Wang, L., Xin, J., Suppan, P., and Wang, Y.: Mixing layer height and its implications for air pollution over Beijing, China, *Atmos. Chem. Phys.*, 16, 2459-2475, 10.5194/acp-16-2459-2016, 2016.

Liu, Q., Jia, X., Quan, J., Li, J., Li, X., Wu, Y., Chen, D., Wang, Z. and Liu, Y.: New positive feedback mechanism between boundary layer meteorology and secondary aerosol formation during severe haze events , doi:10.1038/s41598-018-24366-3, 2018.

Wang, Y. H., Z. R. Liu, J. K. Zhang, B. Hu, D. S. Ji, Y. C. Yu, and Y. S. Wang, Aerosol physicochemical properties and implications for visibility during an intense haze episode during winter in Beijing, *Atmos. Chem. Phys.*, 15(6), 3205-3215, doi:10.5194/acp-15-3205-2015, 2015.