Response to Reviewer #1:

This paper investigated the relationship between PBLH and surface PM concentrations over China. The interaction between PBLH and surface pollutants under different topographic and meteorological conditions has been carefully considered. However, I have some concerns about the conclusion of the paper. The authors have investigated many parameters that may influence the relationship between PBLH and surface PM concentrations. But all the derived correlations are relatively low. It seems risky to get the strong conclusion based on those correlations.

Response: We are very grateful to the reviewer for his/her valuable comments on our work, which are quite constructive and helpful. We carefully considered all of these comments, and modified some strong conclusion regarding the PBLH-PM relationships, as well as the analysis. Our detailed responses to the reviewer's questions and comments are listed below.

General comments:

1. Page 10. I recommend to put the introduction of MERRA in Section 2, which will make the flow more clear.

Response: Per your kind suggest, we moved the introduction of MERRA data to Section 2 (revised Section 2.2.3).

2. Section 3.1. The authors discussed the differences between CALIPSO and MERRA in detail. Will the differences influence the conclusion about the relationship between PBLH and surface pollutants? If so, how much will it be?

Response: Thanks for this valuable comment. In fact, the reanalysis data take account of large-scale dynamical forcing, and have the ability to produce a general PBLH climatology (Guo et al., 2016) which is used to compare with that derived from CALIPSO in this study. However, the reanalysis data do not consider the impact of aerosols; only limited upper atmospheric measurements are assimilated, and the effects of aerosol-PBL interaction are poorly represented (Ding et al., 2013; Simmons, 2006; Huang et al., 2018). Thus, the reanalysis data offer limited ability to investigate detailed PBLH-PM relationships. Therefore, the observation-based retrievals (CALIPSO PBLH or MPL PBLH) are used to produce the PBLH-PM relationships over China. A detailed discussion has been incorporated into the revised Section 3.1.

3. Section 3.2. The correlation coefficient is very low here (Figure 3). I guess it is too risky to make the statement that PBLH has negative correlation with PM2.5 without conditions, which appeared in both abstract and conclusion.

Response: We greatly appreciate this constructive comment. Indeed, the PBLH is not always negatively correlated with $PM_{2.5}$. The weak correlation coefficients cause some

difficulties in deriving a clear relationship between PBLH and $PM_{2.5}$. In addition to PBLH, $PM_{2.5}$ is also controlled by many other factors (e.g. emissions, wind, synoptic patterns, stability, etc.), and thus, the variation of $PM_{2.5}$ is not necessarily related to PBLH, especially when other factors play dominant roles (e.g. strong wind). In such situations, there are rather weak or uncorrelated relationships between PBLH and $PM_{2.5}$. Strong aerosol-PBL interactions only occur under certain conditions. In our analysis, heavy aerosol loading, plains areas, and weak wind speed would be favorable conditions for relatively strong negative correlations between PBLH and $PM_{2.5}$. This discussion has been incorporated into the revised Section 4.

In addition, we revised the overly strong statements to avoid misleading the reader, and show three examples as follows:

In the abstract, "A generally negative correlation is observed between PM and the PBLH…" has been revised to "Albeit the PBLH-PM correlations are roughly negative for most cases, their magnitude, significance, and even sign vary considerably with location, season, and meteorological conditions."

In conclusion, "We observe widespread negative correlations…" has been revised to "Albeit the PBLH-$PM_{2.5}$ correlations are generally negative for the majority of conditions, their magnitude, significance, and even sign vary greatly by region and timing."

In conclusion, "Strong correlations between PBLHs and aerosols occur in low-altitude regions." has been revised to "The PBLH-$PM_{2.5}$ correlations are found to be more significant in low-altitude regions."

Moreover, we previously used the Pearson correlation coefficient, which is representative in a linear relationship. However, the PBLH-$PM_{2.5}$ relationships are nonlinear under most conditions, and this fact would contribute to the low Pearson correlation coefficients. To partly address this problem, we introduce an inverse function ($f(x) = A/x + B$) to fit the PBLH-$PM_{2.5}$ relationships more closely with set the weighting function as the normalized density. In Figure R1 (the revised Figure 5), we jointly use the regular linear regression and the fitted inverse function to characterize the PBLH-$PM_{2.5}$ relationships. Over North China Plain, the nonlinear inverse function shows high consistency with the average values for each bin, and well represents the behavior of the most dense area in the scatter plot with an improved correlation (correlation coefficient -0.49). Similar improvements in the fitting method are also found in other regions, but are still not significant for Pearl River Delta and Northeast China (relatively clean regions).

The fitting methods are described in the revised Section 2.3. We updated the fitting method description in the revised manuscript, which shows better performance in characterizing the PBLH-$PM_{2.5}$ relationships.

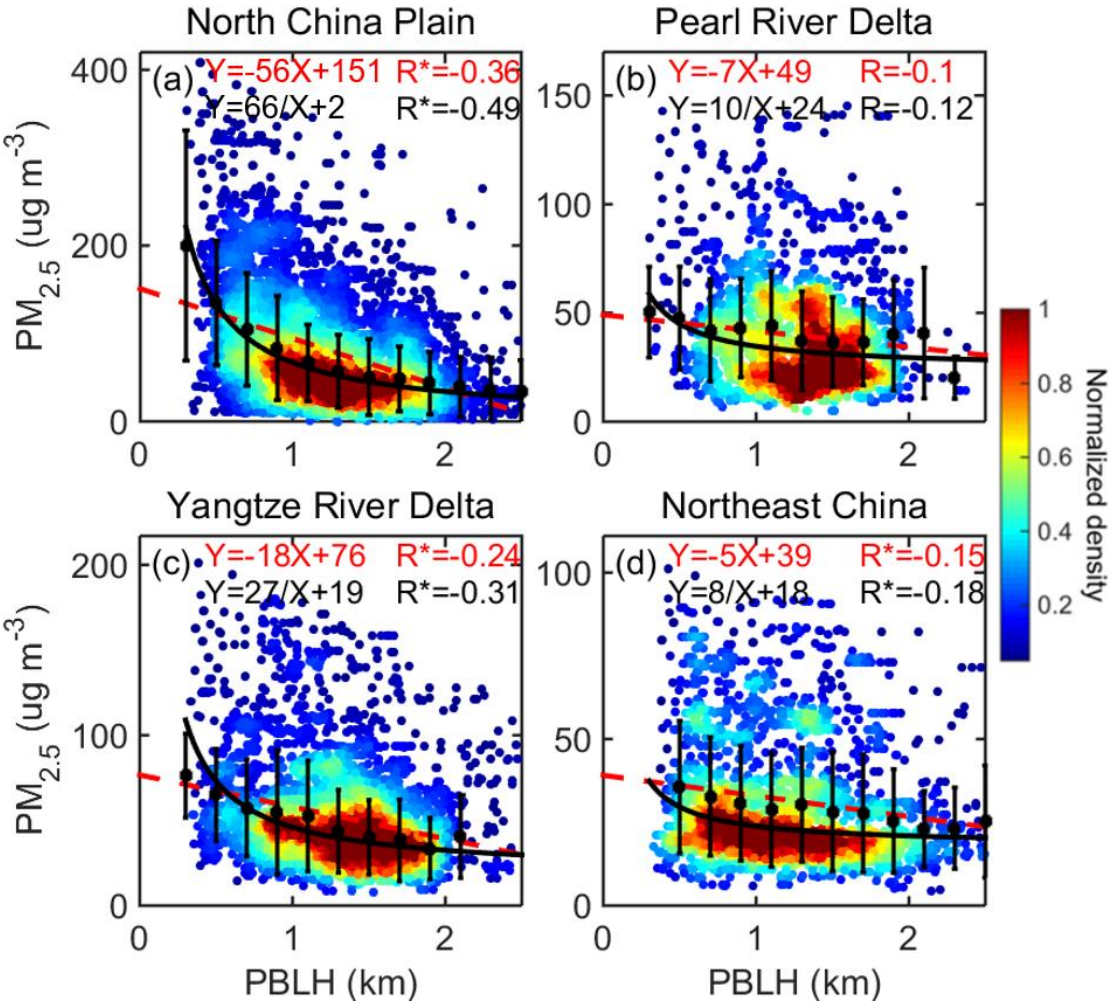

Figure R1. The relationship between CALIPSO-derived PBLH and early-afternoon $PM_{2.5}$ over (a) NCP, (b) PRD, (c) YRD, and (d) NEC. The black dots and whiskers represent the average values and standard deviation for each bin. The red dash lines indicate the regular linear regressions, and the black lines represent the inverse fit ($f(x) = A/x + B$). The detailed fitting functions are given at the top of each panels, along with the Pearson correlation coefficient (red) and the correlation coefficient for the inverse fit (black). Here and in the following analysis, R with asterisks indicates the correlation is statistically significant at the 99% confidence level. The color-shaded dots indicate the normalized sample density.

4. The authors use many figures in the supplement to support the discussion in the text. Meanwhile, the text is not self-explanatory without the graphs. I suggest the authors to reconsider the arrangement of the whole graphs including what has been included in the manuscript. You may want to delete the description that is not very relative or add some figures that are really necessary.

Response: Thanks for pointing this out. We reorganized the supporting information (SI). Figure S2 was revised to present the $PM_{2.5}$ climatology, and was moved to the main text (the revised Figure 4). Figure S4 was revised to present the relationship between MPL PBLH and $PM_{2.5}$ (or normalized $PM_{2.5}$) at Beijing, and was moved to the main text (the revised Figure 7). Previous Figure S3 and Figure S9 were deleted from the SI. As such, we

believe that our main points are delivered and reflected in the revised main text. The SI presents some additional analyses to complement the points in the main text with more evidences.

5. Section 3.6. I understand the authors would like to perform some preliminary analysis here. But exploring the feedback of absorbing aerosols by only analyzing the correlation between PBLH and PM2.5 looks not convincing for me. You may want to perform some further analysis to make the conclusion more solid or discard this part.

Response: We appreciate your suggestion. Indeed, the analysis in Section 3.6 is insufficient, and we deleted this section as suggested. In the discussion, we mention the feedback of absorbing aerosols would be a potential factor affecting the PBLH-PM relationships, which merits further analysis that will be presented in a future paper, given the long length of the current paper.

6. I suggest the authors to add the applications of the findings in the conclusion. How will the findings influence the model development or policy design in the future?

Response: Per your comment, we added following statement to the Section 4:

"Such information can help improve our understanding of the complex interactions between air pollution, boundary layer, and horizontal transport, and thus, can benefit policy making aimed at mitigating air pollution at both local and regional scales. Our study also contributes to the quantitative understanding of aerosol-PBL interaction and further improvement of surface pollutant monitoring and forecasting capabilities."

Specific comments:

1. *Page 3, line 48, the term of "anthropogenic gases" sounds strange. Anthropogenic emissions?*

Response: We revised the "anthropogenic gases" to "gaseous pollutants".

2. *Page 3, line 49, "they are much more visible". Please clarify what are compared with.*

Response: Per your comment, we revised the statement as "PM pollutants are of greater concern to the public partly because they are much more visible than gaseous pollution…"

3. *Page 6, line 114. The grammar seems not proper.*

Response: Per your kind suggestion, we revised the statement as "These empirical relationships between PBLH and surface pollutants are expected to improve our understanding and forecast capability for air pollution..."

4. *Page 6, line 124. The source of the meteorological data is missing.*

Response: Per your comment, we added the source.

5. *Page 6, line 129. The reason for the usage of "noontime" day is missing.*

Response: Thanks for pointing this out. We changed "noontime" to "early-afternoon" and added some clarifying text:

"To match the CALIPSO retrievals with equator crossings at approximately 1330 local time, we use the surface meteorological and environmental data in early-afternoon, averaged from 1300 to 1500 China standard time (CST). During this period, the PBL is well developed with relatively strong vertical mixing, which is a favorable condition for investigating aerosol-PBL interaction."

6. *Page 11, line 234. The English looks not proper in "The PM2.5 seasonal pattern is generally opposite that of PBLH".*

Response: We revised the statement as "The $PM_{2.5}$ seasonal pattern is generally coupled to that of PBLH…"

7. *Page 11, line 238. The grammar seems not proper.*

Response: We revised the statement as "Both the PBLH and $PM_{2.5}$ also show strong seasonality over NCP. PRD is a relatively clean region, and $PM_{2.5}$ maintains low values (<50 μg m^{-3}) through all seasons"

References:

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Ding, A. J., et al., Intense atmospheric pollution modifies weather: a case of mixed biomass burning with fossil fuel combustion pollution in eastern China, Atmos. Chem. Phys., 13(20), 10545-10554, 2013.

Simmons, A., ERA-Interim: New ECMWF reanalysis products from 1989 onwards, ECMWF newsletter, 110, 25-36., 2006.