Anonymous Referee #1 Received and published: 21 October 2020

General comments: The interaction between aerosols and the boundary layer is a hot topic in the study of the formation mechanism of air pollution in polluted areas. The aim of this paper is to evaluate the fundamental interaction between PM and ABL structure and to further quantitatively estimate the effect of aerosol radiative forcing (ARF) on ABL structure. The paper addressed relevant scientific questions and presented novel concepts, ideas and tools. The scientific methods and assumptions were almost valid and clearly outlined so that substantial conclusions were reached. The description of experiments and calculations were almost complete and precise to allow their reproduction by fellow scientists except some points. I think the manuscript could be considered to be accepted after major revision.

Response: Thank the reviewer for the encouragements and constructive suggestions. According to the reviewer's suggestions, we have done our best to revise our manuscript. The modifications have been highlighted in red in the following marked-up manuscript version.

Major comments:

1. Seven cases spanning two months were selected in the paper to discuss the threshold value of aerosol radiative forcing's effect on the contaminated areas' boundary layer structure. Are these cases representative, and would the thresholds change in other cases?

Response: Thank the reviewer for the comments. Firstly, this campaign was launched in Beijing city to obtain the vertical profile observations of meteorological elements in the boundary layer. This experiment lasted from November 2018 to January 2019, and we obtained two-month data sets that can reflect the atmospheric boundary layer structure and atmospheric pollution in winter in Beijing. Second, we need to restate that the threshold value of aerosol radiative forcing's effect on the boundary layer structure was obtained based on the whole two-month data rather than the several cases. Only in the qualitative analysis of the relationship between the aerosol radiation effect and the boundary layer we selected cases to analyze and explain. It means the Figs. 4-7 involved in the quantitative analysis of aerosol radiative forcing influences on the boundary layer structure were processed and obtained based on the whole two-month datasets. We think the threshold value results could be representative and reflect specific effects of aerosol radiation forcing on boundary layer structure in winter in Beijing.

2. With only a finite number of points in Fig. 4, does the current fitting relationship pass the significance test?

Response: Thank the reviewer for the comments and suggestions. We need to explain that the current fitting relationships in Fig. 4 have passed the significant test. More details were shown below:

We used SPSS V19.0 software to calculate the relationship coefficients between $PM_{2.5}$

and TOA, ATM, SFC, and |SFC-ATM|, respectively, shown in Table 1. The significance levels between $PM_{2.5}$ and TOA, SFC, and |SFC-ATM| are respectively less than 0.01, indicating that they have passed the 99% significance test and have a significant correlation, respectively. The significance level between $PM_{2.5}$ and ATM is 0.021, grater than 0.01 and less than 0.05, indicating that they have passed the 95% significance test and have a reasonable correlation.

Table 1. Relationship test

		N	Relationship coefficient (R^2)	Significance level
a	$PM_{2.5}$ & TOA	13	0.75	0.000
b	$PM_{2.5}$ & ATM	13	0.40	0.021
c	$PM_{2.5}$ & SFC	13	0.83	0.000
d	$PM_{2.5}$ & |SFC-ATM|	13	0.81	0.000

3. In Fig. 1b, the results for aerosol radiative forcing have values only for individual moments of the day, and a detailed explanation of how they relate to hourly variations in atmospheric conditions and PM concentrations is needed.

Response: Thank the reviewer for the constructive suggestions. According to the reviewer's suggestions, we have added a detailed explanation of how the aerosol radiative forcing relates to hourly variations in atmospheric conditions and PM concentrations in the revised manuscript.

4. What is the physical mechanism by which |SFC-ATM| affects the threshold of atmospheric stability?

Response: Thank the reviewer for the comments. |SFC-ATM|, defined as the absolute value of the difference between SFC and ATM, represents aerosols' combined action on the solar radiation reaching the aerosol layer and the ground. Larger values of |SFC-ATM| indicate either stronger aerosol scattering (higher SFC) or absorption effects (higher ATM), or indicate both stronger aerosol scattering and absorption effects. No matter which one causes the increased |SFC-ATM|, they all imply a more significant temperature difference between the surface and the above atmosphere layer. That means a higher |SFC-ATM| would lead to a more stable atmospheric stratification, which would suppress the turbulence development.

5. When calculating TKE, why a one-hour wind standard deviation was chosen rather than a half-hour or two-hour standard deviation? In lines 141-152, the temporal and spatial scales of TKE need to be clarified.

Response: Thank the reviewer for the comments and suggestions. Considering the time series of the boundary layer meteorological elements profile were displayed on the hourly scale, we choose to calculate one-hour TKE for better analyze the relationship among them. Regarding the temporal and spatial scales of TKE have been added in the calculation part of Section 2.

6. Fig. 1 is of low quality and should be improved. In Fig. 1-(a)-III, why does the PM not increase with decreasing ABLH?

Response: Thank the reviewer for the comments and suggestions, and we have improved the quality of Fig.1. In Fig.1-(a)-III, $PM_{2.5}$ concentrations were generally below ~40 μg m^{-3}, when there was a decreasing ABLH, the $PM_{2.5}$ concentrations have slightly increased. The $PM_{2.5}$ concentrations did not increase as significantly as those in heavy pollution phases of cases I and II. Due to the drop of vertical diffusion height, the PM was accumulated at the ground level, increasing the surface PM concentrations to some degree. However, during the clean period III, Beijing was controlled by clean and dry winds, the air humidity was quite low. With less PM loading and low humidity near the surface, the heterogeneous reaction was not intense. The weak secondary aerosol formation would not lead to an outbreak of PM concentrations near the surface. That is the reason that the PM did not increase much with decreasing ABLH.

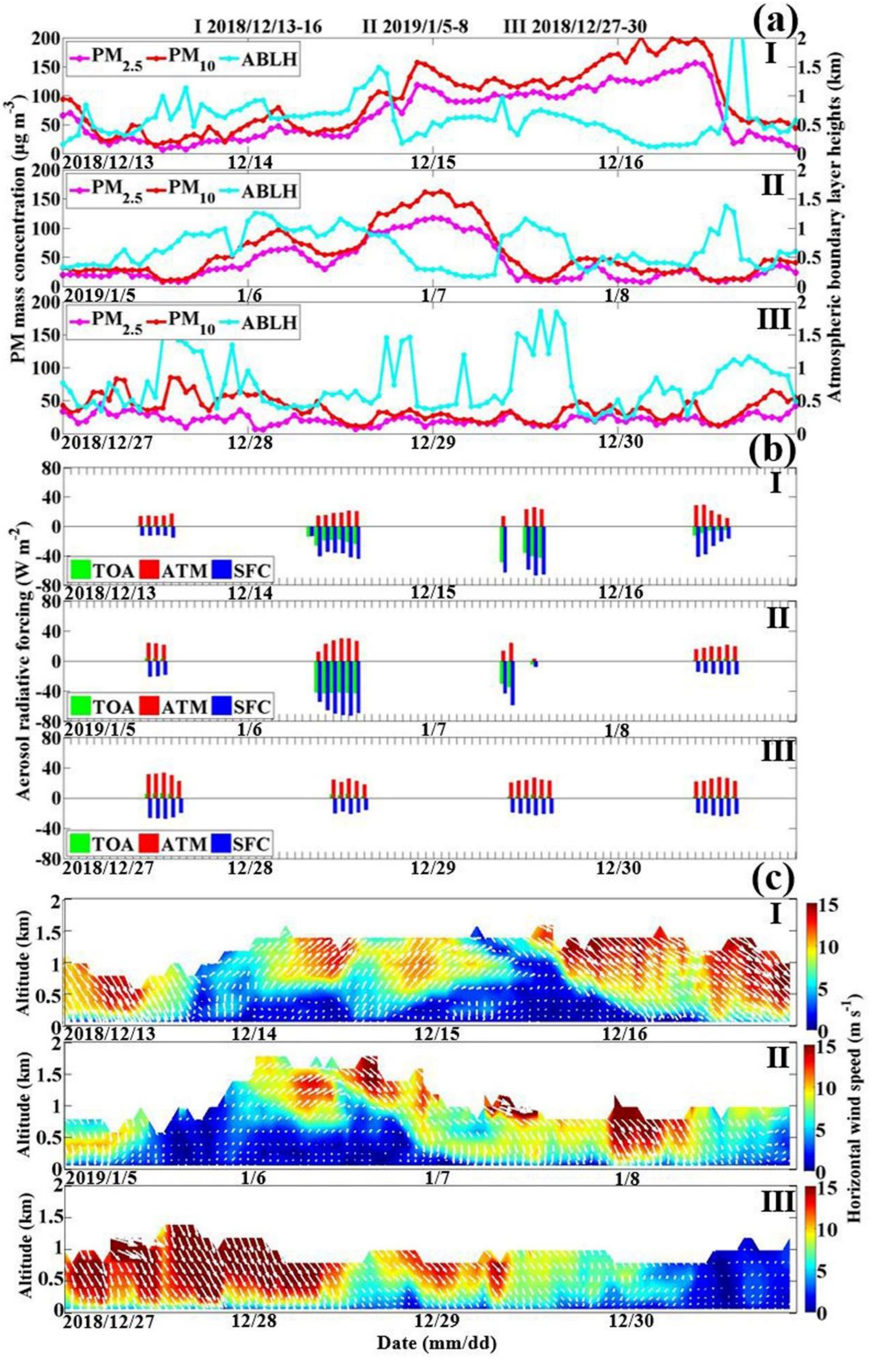

Figure 1. Temporal evolution of (a) the PM mass concentration and atmospheric boundary layer height ($PM_{2.5}$: solid pink lines; PM_{10}: solid red lines; ABLH: solid blue

lines), (b) aerosol radiative forcing at the top (TOA; green bars), surface (SFC; blue bars) and interior of the atmospheric column (ATM; red bars), and (c) horizontal wind vector profiles (shaded colors: wind speeds; white arrows: wind vectors) during the typical haze pollution episodes of I (2018/12/13-16) and II (2019/1/5-8) as well as the typical clean period of III (2018/12/27-30).

7. In Figure 1-(b) I and II, the TOA varied significantly. What is the reason?

Response: Thank the reviewer for the comments and suggestions. As shown in Fig. 4, TOA forcing was proportional to the $PM_{2.5}$ concentration. With the increase in $PM_{2.5}$ concentration, elevated aerosol loading near the surface would scatter more solar radiation back into outer space and cause less solar radiation reaching the ground, corresponding to a cooling of the surface and making negative SFC. TOA means the aerosol radiative forcing at the top of the atmosphere column and is the sum of ATM and SFC. Considering that anthropogenic aerosols are mostly scattering aerosols, the SFC forcing is generally stronger than ATM, corresponding to a cooling of the earth-atmosphere system. The TOA forcing was thus usually negative and had a similar trend with SFC. Thus, in Figure 1-(b) I and II, with PM concentrations increasing, the TOA varied significantly.

8. There are very interesting results for PM and temperature in Figure 2. What are the diurnal characteristics of the potential temperature? Does potential temperature affect the diurnal concentration of $PM_{2.5}$?

Response: Thank the reviewer for the comments and suggestions. Figure 2 shows temporal variations in the vertical profiles of (a) the virtual potential temperature gradient ($\partial\theta v/\partial z$), (b) pseudoequivalent potential temperature gradient ($\partial\theta se/\partial z$) and (c) temperature inversion phenomenon (shaded colors: inversion intensity) during the typical haze pollution episodes of I (2018/12/13-16) and II (2019/1/5-8) as well as the typical clean period of III (2018/12/27-30). Figures 2(c) have shown the relationship between PM and temperature structure. For example, when the temperature vertical gradient is positive means a temperature inversion occurs. This abnormal temperature structure would lead to a stable stratification with a positive potential temperature gradient. Figure 2(a)-(b) exactly present the potential temperature conditions corresponding to the temperature structure in Fig. 2(c). The temporal variations in the vertical profiles of (a) the virtual potential temperature gradient ($\partial\theta v/\partial z$) and (b) pseudoequivalent potential temperature gradient ($\partial\theta se/\partial z$) can represent a diurnal variation in potential temperature (stratification stability) which influence the diurnal change in $PM_{2.5}$. The specific analysis was provided in section 3.2.

9. In Figure 4, other dots represent mean data. How is it calculated?

Response: Thank the reviewer for the comments. In Fig. 4, the other dots represent mean data calculated by averaging the daily data at a fixed step length. The daily data means daily mean values of TOA, ATM, SFC, and corresponding daily averaged $PM_{2.5}$ mass concentration from 27 November 2018 to 25 January 2019 in Beijing. The mean

$PM_{2.5}$ concentrations were obtained by averaging daily $PM_{2.5}$ concentrations at intervals of 10 μg m^{-3}. The mean TOA, ATM, and SFC were obtained after the corresponding daily TOA, ATM, and SFC average, respectively. For example, all daily $PM_{2.5}$ concentrations greater than 40 μg m^{-3} and less than 50 μg m^{-3} were averaged as a mean $PM_{2.5}$ concentration, and TOA values (ATM; SFC) corresponding to this daily $PM_{2.5}$ concentration range were also averaged as a mean TOA (ATM; SFC). We have added a more detailed calculation description in the Fig. 4 caption.

10. The empirical relationships of TKE and |SFC-ATM| are very interesting in Figure 6 (left upper panel). It established the thermodynamic relationship between ARF and TKE by using the measured data. Why does the fitting relationship fit so well below 300 meters?

Response: Thank the reviewer for the comments. As we can see in Fig. 6, the exponential relationship between TKE and |SFC-ATM| was notable in the lower layers (below ~300 m) and gradually deteriorated with the increasing altitude. We all know that aerosols are mainly concentrated in the lower atmosphere, contributing the most to the SFC and ATM forcing. The stratification stability induced by the aerosol radiative effect would mainly occur in lower layers. The much better exponential relationship between TKE and |SFC-ATM| in the lower layers exactly further confirmed that the considerable change in atmospheric stratification caused by aerosols indeed existed and was mainly shown in the lower layers. With the increase of altitude, aerosol loading is in decline; thus, aerosol radiative effect on the atmospheric stability drops. Furthermore, at a relatively high altitude, the aerosol is few, and the radiation effect has almost no influence on the stability of the atmosphere layer.

11. The ARF threshold is about 55 W m^{-2}. What about the concentration of $PM_{2.5}$? Is it possible to derive a threshold concentration for $PM_{2.5}$ based on current observational relationships. The $PM_{2.5}$ threshold would be a very meaningful target for air pollution control.

Response: Thank the reviewer for the comments and constructive suggestions. As we can see from Fig. 4(d), the exponential relationship between $PM_{2.5}$ and |SFC-ATM| was founded. According to the linear fitting equation of $y=0.49x+31.21$ (x: $PM_{2.5}$; y: |SFC-ATM|), it is possible to derive a threshold concentration for $PM_{2.5}$ based on the current |SFC-ATM| threshold of about 55 W m^{-2}.

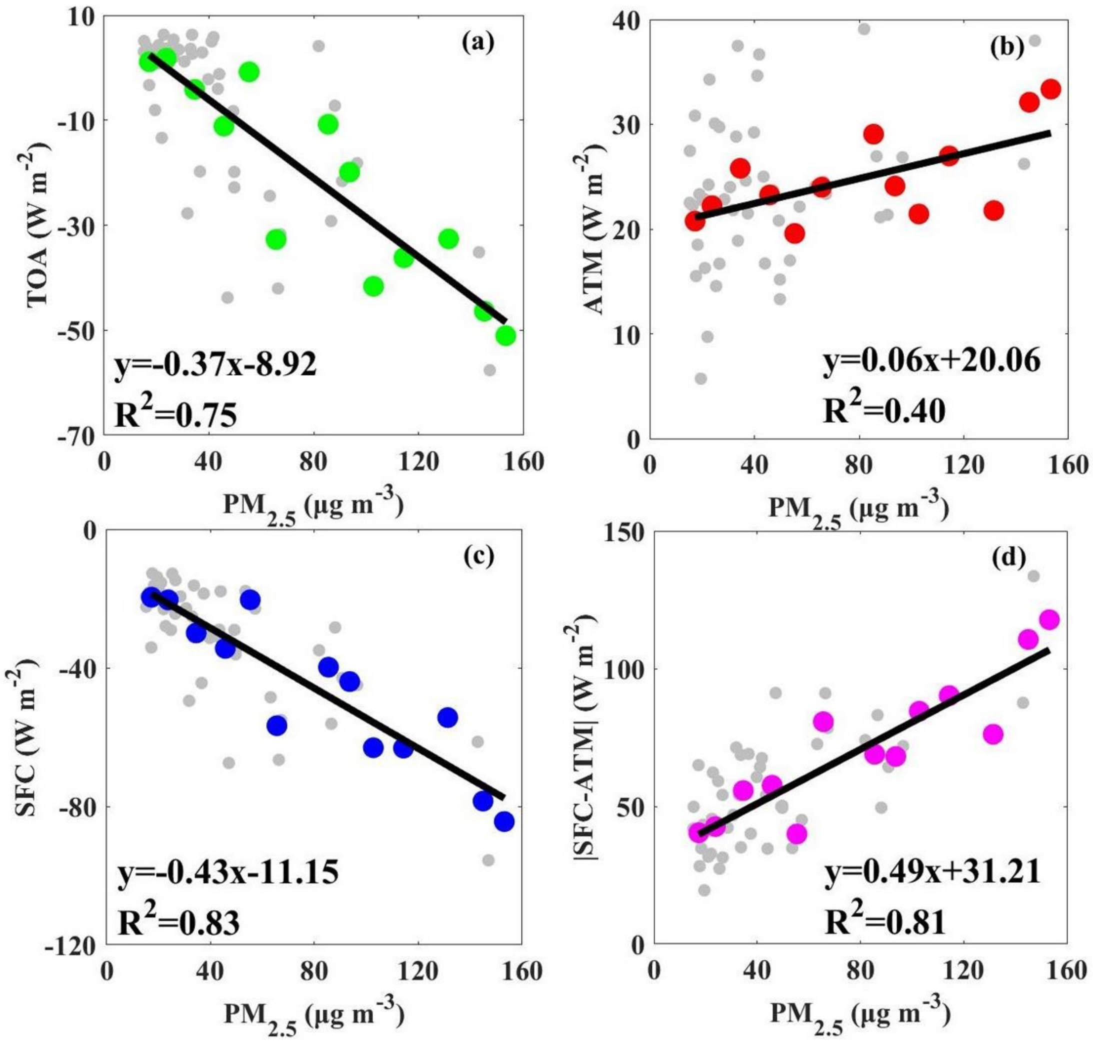

Figure 4. Scatter plots of the $PM_{2.5}$ mass concentration (x) versus aerosol radiative forcing at the surface (SFC; y; a), interior of the atmospheric column (ATM; y; b) and top of the atmospheric column (TOA; y; c) as well as the absolute difference of SFC and ATM (|SFC-ATM|; y; d), respectively (gray dots: daily data; other dots: mean data). (The daily data means daily mean values of TOA, ATM, SFC, and corresponding daily averaged $PM_{2.5}$ mass concentration from 27 November 2018 to 25 January 2019 in Beijing. The mean $PM_{2.5}$ concentrations were obtained by averaging daily $PM_{2.5}$ concentrations at intervals of 10 μg m^{-3}. The mean TOA, ATM, and SFC were obtained after the corresponding daily TOA, ATM, and SFC average, respectively. For example, all daily $PM_{2.5}$ concentrations greater than 40 μg m^{-3} and less than 50 μg m^{-3} were averaged as a mean $PM_{2.5}$ concentration, and TOA values (ATM; SFC) corresponding to this daily $PM_{2.5}$ concentration range were also averaged as a mean TOA (ATM; SFC)).

12. The review of aerosol radiative forcing in the introduction needs to be strengthened.

Response: Thank the reviewer for the comments and constructive suggestions. As the reviewer's suggested, the review of aerosol radiative forcing in the introduction has been strengthened.

13. Conclusion needs to be subdivided and further simplified.

Response: Thank the reviewer for the comments and constructive suggestions. As you suggested, the Conclusion has been subdivided and further simplified.

14. In Figure 8, TKE >2 $m^2 s^{-2}$, |SFC-ATM| ~55 W m^{-2}. Are these thresholds generalizable?

Response: Thank the reviewer for the comments. Firstly, this campaign was launched in Beijing city to obtain the vertical profile observations of meteorological elements in the boundary layer. This experiment lasted from November 2018 to January 2019, and we obtained two-month data sets that can reflect the atmospheric boundary layer structure and atmospheric pollution in winter in Beijing. Second, the threshold value of aerosol radiative forcing's effect on the boundary layer structure was obtained based on the whole two-month data. It means Fig. 8 involved in the quantitative analysis of aerosol radiative forcing influences on the boundary layer structure were processed and obtained based on the whole two-month datasets. We think the threshold value results could be representative and reflect specific effects of aerosol radiation forcing on boundary layer structure in winter in Beijing.

Minor comments:

English writing should be polished. Some sentences were hard to read.

1. e.g. line 18-20 "Multi-episode contrastive analysis stated the key to determining whether haze outbreak or dissipation was the ABL structure (i.e., stability and turbulence kinetic energy (TKE)) satisfied relevant conditions." Should be "Multi-period comparative analysis indicated that the key to determining whether the haze outbreak or dissipation occurs is whether the ABL structure (i.e., stability and turbulent kinetic energy (TKE)) satisfies the relevant conditions."

Response: Thank the reviewer very much for this gramma suggestion. We have corrected it.

2. Line 22-23. "SFC and ATM is respectively the ARF at the surface and interior of the atmospheric column" should be "SFC and ATM are the ARFs at the surface and interior of the atmospheric column, respectively."

Response: Thank the reviewer very much for this gramma suggestion. We have corrected it.

3. Line 37-38. (Li et al., 2020; Xu et al., 2019), should be cited at the end of this sentence.

Response: Thank the reviewer for this suggestion. We have corrected it.

4. Line 316 two "dropped to".

Response: Thank the reviewer for pointing out this mistake. We have corrected it.