Anonymous Referee #2 Received and published: 14 November 2020

1. The authors attempted to propose a parameter, $|SFC-ATM|$ for quantification of the impact of aerosol radiative forcing (ARF) on the atmospheric boundary layer (ABL) structure. Why did the author use the ARF of the interior of the atmosphere column (ATM) rather than the ARF in the ABL since most of aerosols or particulate matters are trapped in the atmospheric boundary layer?

Response: Thanks for the reviewer's comment. First of all, when quantifying the impact of aerosols on climate change, it is more to judge its impact on the earth-atmosphere system as a whole, so the top of the atmosphere's choice will be more reasonable. Secondly, in our previous work, we used the path radiation in MODIS data as a key parameter for calculating the atmospheric SSA, which represents the radiation value at the top of the atmosphere (TOA). In order to facilitate the comparison and verification in the later calculation process, we chose the same height to calculate the relevant radiation results, which can perform unified calculation and analysis both on the top of the atmosphere. We believe this can get more representative results in aerosol radiation research (Gong et al., 2014; Lee et al., 2007; Xin et al., 2016). Finally, as the reviewer said, aerosols are concentrated in the boundary layer and few in the stratosphere. It is because most aerosols exist in the boundary layer that we have verified in the previous sensitivity test, and the calculations at the top of the boundary layer and the top of the atmosphere are as follows:

$$\Delta F^{AEROSOL} = (\Delta F^{TOA}_{aero} - \Delta F^{TOA}_{non-aero}) - (\Delta F^{SFC}_{aero} - \Delta F^{SFC}_{non-aero}) \quad (1)$$

$$\Delta F = F^{\text{downward}} - F^{\text{upward}} \quad (2)$$

Where ΔF denotes the net downward flux (downward minus upward radiation); the subscripts "TOA" and "SFC" denote the top of the atmosphere/boundary layer and the surface; and “aero” and “non-aero” denote dusty and clean skies (Chou et al., 2002). Since there are few aerosols at high altitudes, the ΔF aero- ΔF non-aero itself is derived from the boundary layer difference. The ΔF aero- ΔF non-aero at high altitudes is negligible. So the radiative forcing generated by aerosols will not be significantly different because of the ABL or the top of the atmosphere. For these three reasons, we finally chose the top of the atmosphere for analysis.

Chou, M., Chan, P., and Wang, M.: Aerosol radiative forcing derived from SeaWiFS-retrieved aerosol optical properties, J. Atmos. Sci., 59, 748–757, http://dx.doi.org/10.1175/1520-0469(2002), 2002.

Gong, C., Xin, J., Wang, S., Wang, Y., Wang, P., Wang, L., and Li, P.: The aerosol direct radiative forcing over the Beijing metropolitan area from 2004 to 2011, J. Aerosol Sci., 69, 62-70, https://doi.org/10.1016/j.jaerosci.2013.12.007, 2014.

Xin, J., Gong, C., Wang, S., and Wang, Y.: Aerosol direct radiative forcing in desert and semi-desert regions of northwestern China, Atmos. Res., 171, 56-65, https://doi.org/10.1016/j.atmosres.2015.12.004, 2016.

Lee K., Li Z., Wong M., Xin J., Wang Y., Hao W., and Zhao F.: Aerosol single scattering albedo estimated across China from a combination of ground and satellite measurements, J. Geophys. Res.: Atmos., 112(D22), https://doi.org/10.1029/2007JD009077, 2007.

2. Impact of ARF on reduction of surface-reaching shortwave radiation and heating/cooling of the atmosphere is dependent on not only aerosol loadings in the atmosphere (e.g., AOD) but also aerosol optical or radiative properties such as single-scattering albedo (SSA). What value(s) of SSA was (were) used in the numerical simulations with the SBDART radiation transfer model and how the threshold value changes single-scattering albedo (SSA)? It will be helpful if the author may provide more details about the configurations and inputs utilized in the simulations.

Response: Thanks for the reviewer's comment. We have added relative details in the manuscript after the first referee round. "The algorithm of SBDART (Santa Barbara DISORT Atmospheric Radiative Transfer) (Levy et al., 2007) is the core model to calculate the radiative forcing parameters. A standard mid-latitude atmosphere is used in SBDART in Beijing. AOD and Angstrom Exponent (AE) at 550 nm were obtained from sun-photometer. Multiple sets of Single Scattering Albedo (SSA) and backscattering coefficient were calculated based on MIE theory, and surface albedo & path radiation were read from MODIS (MOD04), which is used to calculate radiative forcing at the top of atmosphere (TOA). The TOA results were combined with MODIS observations, the result which has the lowest deviation is defined as the actual parameters of aerosols, and this set of parameters would be used to calculate the radiative forcing at the surface, top, and interior of the atmospheric column (Gong et al., 2014). Hourly radiative forcing parameters, including the ARF at the top (TOA), surface (SFC), and interior of the atmospheric column (ATM) at an observation site in Beijing can be calculated based on this algorithm. More detailed descriptions are provided in our previous work (Gong et al., 2014; Xin et al., 2016)." was added in Section 2.

3. Is it necessary to use both virtual potential temperature gradient and pseudoequivalent potential temperature gradient to define the atmospheric stability since both have very similar time-height cross section distribution patterns? Please provide a description on how to use these two gradients to define the atmospheric stability and what are the advantages of using these two gradients rather than potential temperature gradient in determining the atmospheric stability?

Response: Thank the reviewer very much for this comment. Using both virtual potential temperature gradient and pseudoequivalent potential temperature gradient to define the atmospheric stability is more accurate and closer to the real atmosphere condition. Because the real atmosphere consists of saturated and unsaturated air masses. The negative virtual potential temperature gradient means absolute unstable stratifications for both saturated and unsaturated air masses, rare except in the lower layers where it is possible. When the virtual potential temperature gradient is positive while the pseudoequivalent potential temperature gradient is negative means a stratification of conditional instability. The atmosphere stratification is unstable for a saturated air mass and stable for an unsaturated air mass. The stratification of conditional instability will become unstable once the saturated air mass reaches the condensation height due to strong local convection or substantial uplift of dynamic factors. The positive pseudoequivalent potential temperature gradient means absolute stable stratifications for both saturated and unsaturated air masses. However, the potential temperature gradient in determining the atmospheric stability only refers to unsaturated air masses. These are the reason that we choose to use both virtual potential temperature gradient and pseudoequivalent potential temperature gradient to define the atmospheric stability.

4. Figs. 2-3: It is suggested to replot these figures by including specific months and dates in x-axis for a better view. In addition, right y-axis should be $PM_{2.5}$ rather than PM for both figures. Please correct them.

Response: Thank the reviewer very much for this comment and suggestion. As you suggested, we have replotted Fig. 1-3 to add specific months and dates in the x-axis, shown below. However, the right y-axis should be PM mass concentration for both time series of $PM_{2.5}$ and PM_{10} concentration has been plotted.

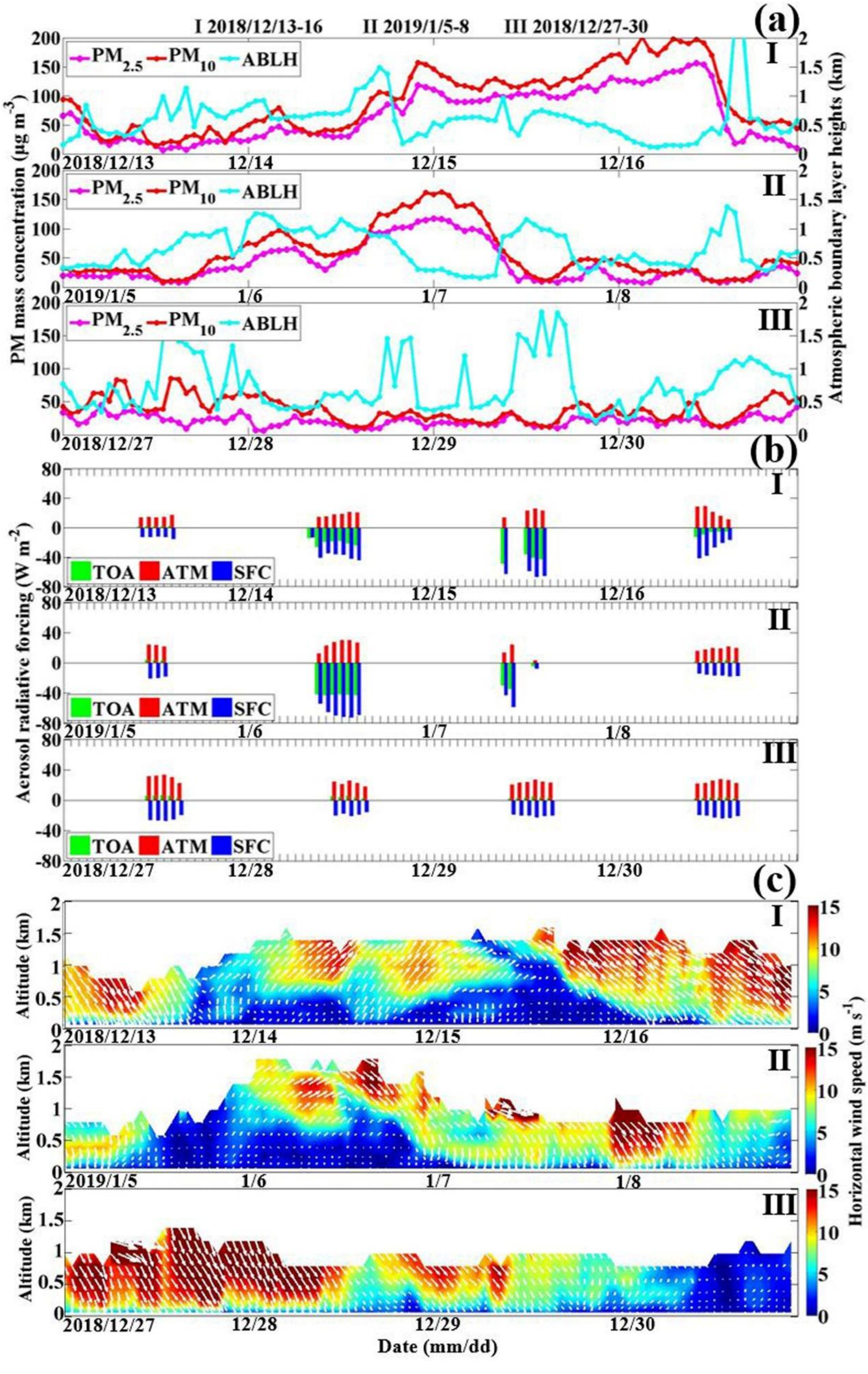

Figure 1. Temporal evolution of (a) the PM mass concentration and atmospheric boundary layer height ($PM_{2.5}$: solid pink lines; PM_{10}: solid red lines; ABLH: solid blue

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).

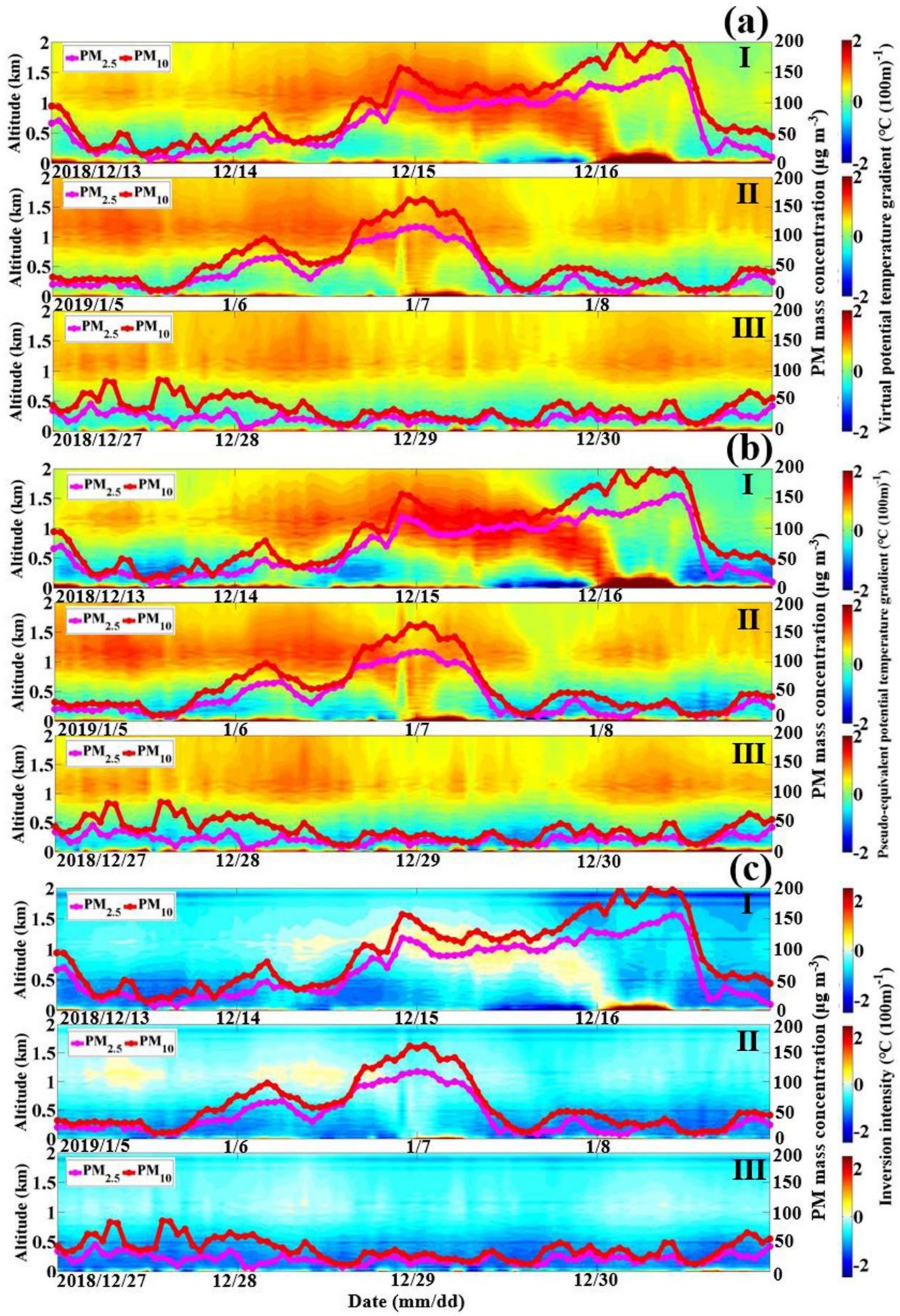

Figure 2. Temporal variation in the vertical profiles of (a) the virtual potential temperature gradient ($\partial\theta v/\partial z$), (b) pseudoequivalent potential temperature gradient ($\partial\theta se/\partial z$) and (c) temperature inversion phenomenon (shaded colors: inversion intensity) during 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).

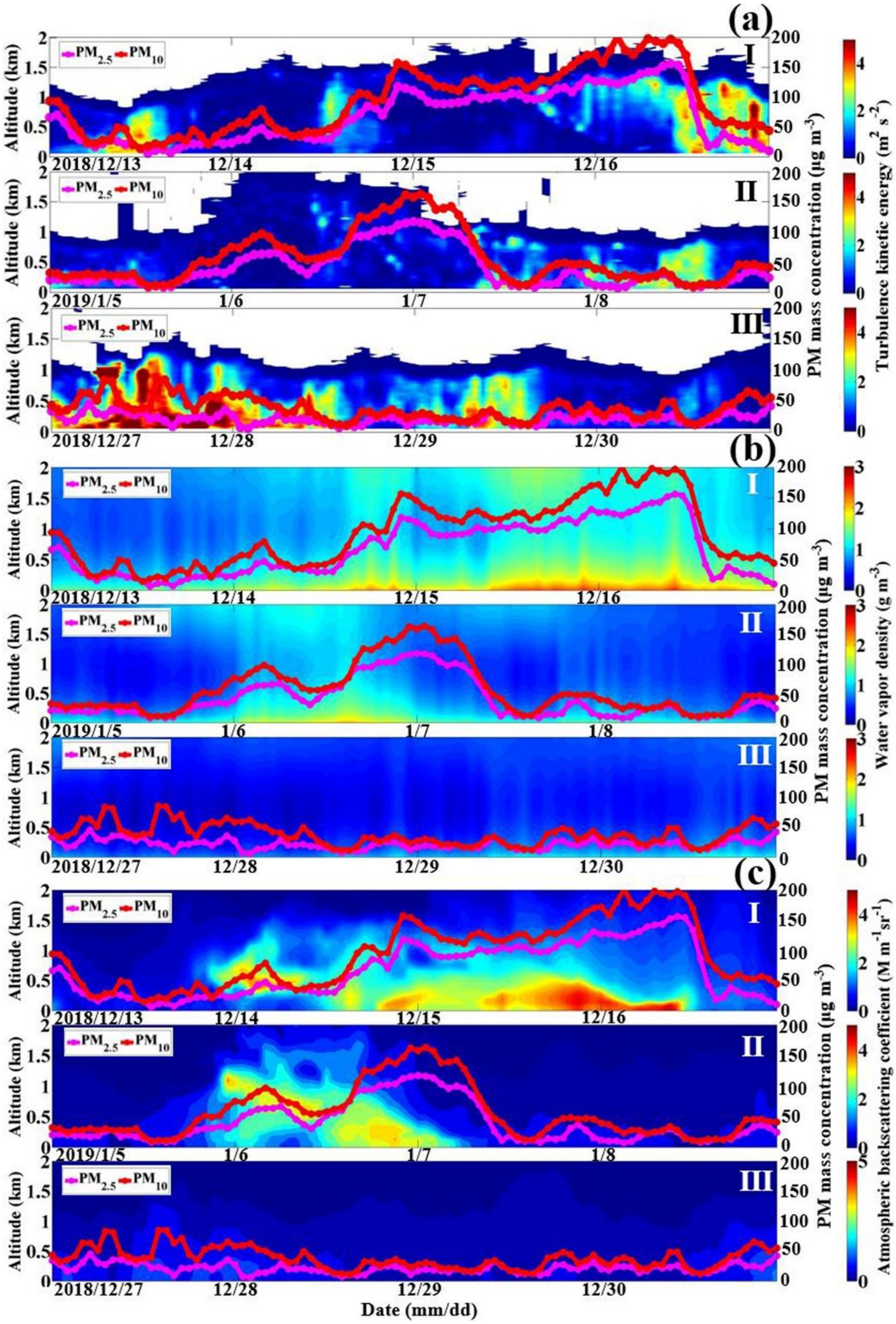

Figure 3. Temporal variation in the vertical profiles of (a) the turbulent activity (shaded colors: TKE), (b) atmospheric humidity (shaded colors: vapor density) and (c) vertical distribution of suspended particles (shaded colors: BSC) during the typical haze

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).

5. Fig.3a: Usually, higher $PM_{2.5}$ concentrations, lower surface-reaching shortwave radiation, and weaker turbulent activity (i.e., lower TKE). However, such a relationship is not clear in the ABL on day 1 for Episode II and day 4 for Episode III.

Response: Thank the reviewer very much for this comment. Usually, in the daytime of the clean day, with the mixing layer developing the turbulent activity would be strong. In the ABL on day 1 for Episode II and day 4 for Episode III, the $PM_{2.5}$ concentrations were really low while the turbulent activity (i.e., lower TKE) was not too strong. Both mechanical and thermal actions determine turbulence activity. The wind fields during day 1 for Episode II and day 4 for Episode III were relatively weak, while the other clean periods were always corresponding to strong winds. With little mechanical action on turbulence generation, the TKE during these periods were not as strong as other clean periods.

6. L250-251, For the statement of "the atmospheric stratification during Episodes I and II was altered", please provide specific calculation to illustrate how the stratification was altered". Similar statements were also found in several places in the manuscript.

Response: Thank the reviewer very much for this comment. Regarding the statement "the atmospheric stratification during Episodes I and II was altered" in line 250-251 was concluded based on the previous analysis. The specific description is shown below:

"During the remainder of the 2nd day, the PM mass concentration continued to increase with south winds blowing and reached its highest level at midnight with a $PM_{2.5}/PM_{10}$ mass concentration of ~110/150 μg m^{-3} during both episodes I and II. The highest BSC values mainly occurred from the ground to a height of 1 km at this time, implying that a portion of the suspended particles was pushed down to the near-surface. *Noteworthily, regardless of the wind field, the atmospheric stratification states during this rising phase changed more notably. Before southerly wind transport occurred, the evolution of the stability indicator (∂θv/∂z; ∂θse/∂z) profiles during episodes I and II was analogous to that during episode III (Figs. 2(a)- (b)). The stratification states at the different heights (0-1 km) were either unstable or neutral, with negative or zero ∂θv/∂z values, respectively, whereby no clear nor strong temperature inversion phenomenon occurred in the lower atmosphere layer (Fig. 2(c)). The corresponding ABLHs were the same (Fig. 1(a)). However, the atmospheric stratification from ~0.5-1 km during the episode I and from 0-1 km during episode II became quite stable during the PM increase period, with positive values of ∂θse/∂z and almost no turbulent activity (TKE: ~0 m^2 s^{-2}) (Fig. 3(a)). In contrast to an increased ABLH during clean period III, the ABLHs during episodes I-II sharply decreased.* Considering that aerosol scattering and absorbing radiation could modify the temperature stratification (Li et al., 2010; Zhong et al., 2018), the aerosol radiation effect is too weak at a low PM level to change the latter, which defines the atmospheric stability. With the elevated PM level due to southerly transport, ARF also increased, with SFC (ATM) reaching ~-40 (~20) W m^{-2}

and ~-75 (~30) W m^{-2} during episodes I and II, respectively. Less radiation reaching the ground and more heating the atmosphere above the ground, and in comparison to clean episode III, the atmospheric stratification during episodes I and II was altered".

As described above, with the PM rising and the ARF increasing in episodes I and II, the corresponding atmospheric stratifications were altered compared to that in clean episode III and the previous no PM rising period.

7. Fig.4: It is difficult to understand that aerosol radiative forcing at top of the atmospheric column (TOA) has so close relationship with surface $PM_{2.5}$ concentrations. Please provide an explanation. Again, it is better to calculate the ARF for the integrated ABL rather than the interior of the atmospheric column.

Response: Thank the reviewer very much for this comment. As shown in Fig. 4(a), 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. The ARF calculation for the interior of the atmospheric column rather than the integrated ABL has been explained in Question 1.

8. Why did the authors use the absolute value of difference between SFC and ATM? Why not use ATM–SFC since ATM is positive and SFC is negative? In fact, the ATM-SFC represent a combined impact of aerosol radiative effect on surface-reaching shortwave radiation and the atmospheric layer. It is not surprised to see ATM-SFC increases with increasing $PM_{2.5}$ concentrations (see Fig. 4d). Here the authors still use scatter plots to quantify the relationship between aerosol radiative effect and surface $PM_{2.5}$ in terms of model results. Are there any observational data available to verify the results?

Response: Thank the reviewer very much for this comment. First of all, we all know that the difference between SFC and ATM means a combined impact of the aerosol radiative effect on surface-reaching shortwave radiation and the atmospheric layer. The reason we use the absolute value of SFC-ATM is that ATM is positive and SFC is negative; thus the SFC- ATM is always negative. The absolute value of SFC-ATM represents the same meaning as ATM-SFC. Secondly, we plotted this scatter plot (Fig. 4d) to show the relationship between the combined impact of the aerosol radiative effect on surface-reaching shortwave radiation and the atmospheric layer and $PM_{2.5}$ concentrations. It shows |SFC-ATM| increases with increasing $PM_{2.5}$ concentrations. We need to explain that the aerosol radiative forcing (ie., SFC and ATM) can be obtained only by models. Regarding the observational data verify, Zhong et al. (2018) once verified the relationship between the global radiant exposure measured at the surface and $PM_{2.5}$ concentrations, shown as below. To further investigate the impact of elevated $PM_{2.5}$ on the loss in surface solar radiation, they calculated daytime mean $PM_{2.5}$ mass concentration, direct, diffuse, and global radiant exposure in December 2016 to 10th January 2017 in Beijing. We can see that the radiation reaching the ground decreased with the $PM_{2.5}$ concentration increasing, consistent with the relationship between SFC and $PM_{2.5}$ concentration in Fig. 4(c). However, the radiation in the atmosphere is hard to be measured yet. Thus, the aerosol radiative effect on the earth-atmosphere system is mainly based on the aerosol radiative forcing calculated by models.

J. Zhong et al. *Atmospheric Research 209 (2018) 59–64*

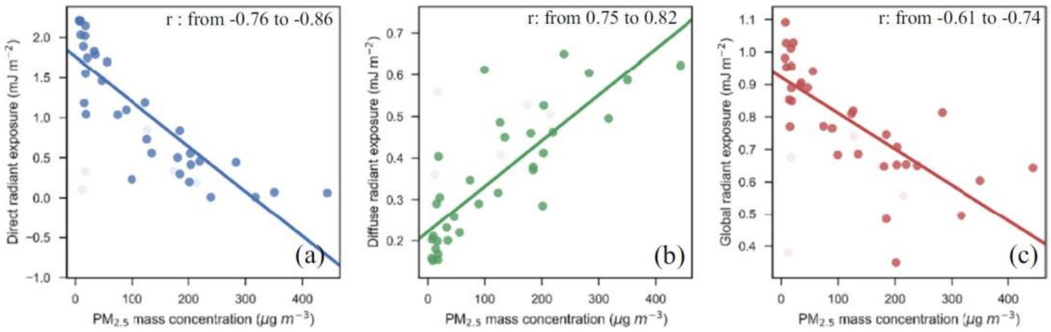

Fig. 2. The correlation of daytime mean $PM_{2.5}$ mass concentration and daytime mean radiant exposure from 1st December 2016 to 10th January 2017. (a) $PM_{2.5}$ and direct radiant exposure; (b) $PM_{2.5}$ and diffuse radiant exposure; (c) $PM_{2.5}$ and global radiant exposure; (semitransparent points represents the days with high-layer moisture, and r shows the variations without semitransparent points).

Zhong J., Zhang X., Wang Y., Liu C., and Dong Y.: Heavy aerosol pollution episodes in winter Beijing enhanced by radiative cooling effects of aerosols, Atmos. Res., 59-64, 10.1016/j.atmosres.2018.03.011, 2018.

9. Fig.6: Please add a), b), c), and d) each panel, respectively, and specify clearly in the figure caption.

Response: Thank the reviewer very much for this suggestion. We have added a), b), c), and d) each panel, respectively, and specify clearly in the figure caption, shown below.

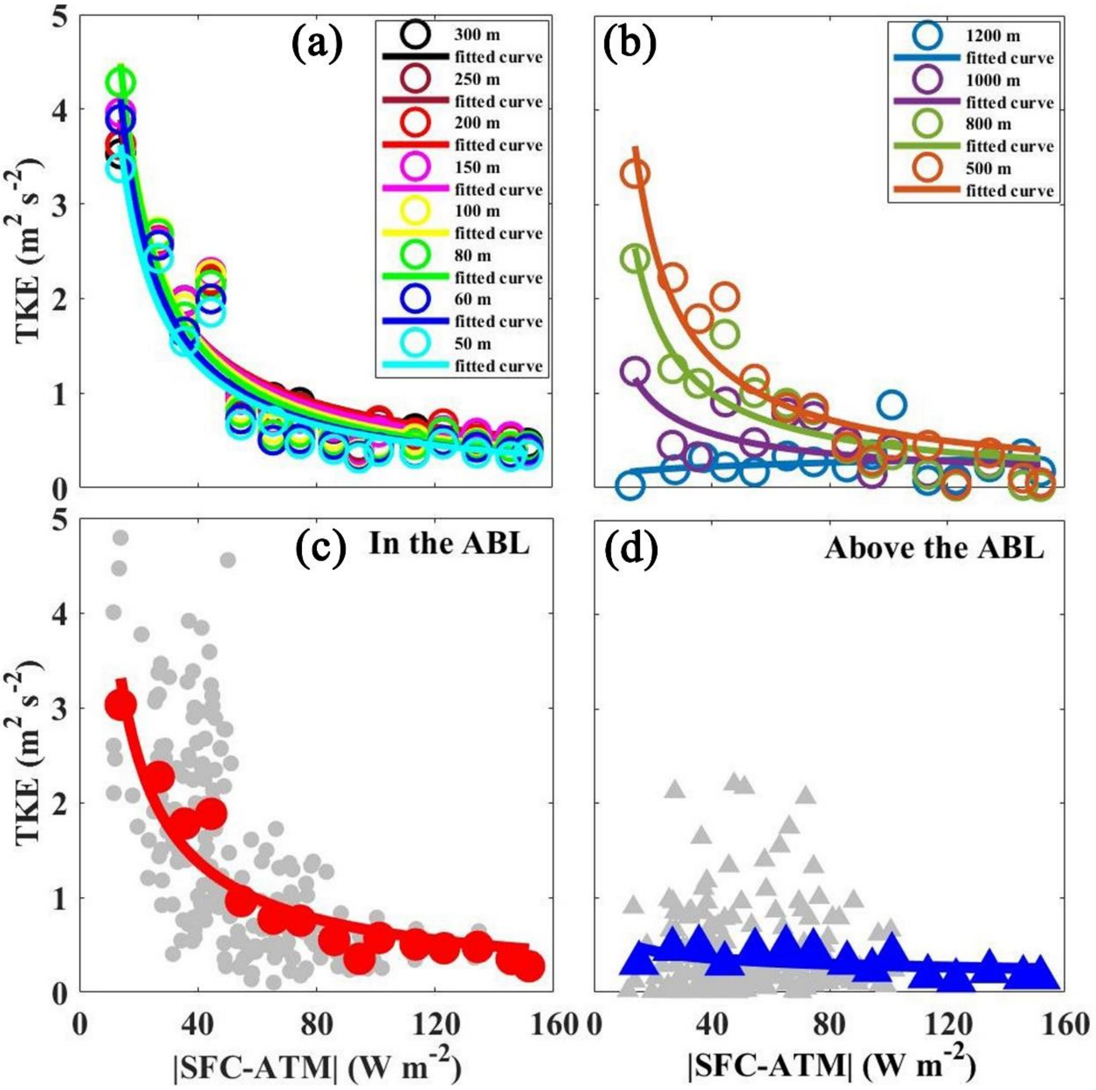

Figure 6. Scatter plots of the mean absolute difference of the aerosol radiative forcing at the surface and interior of the atmospheric column (|SFC-ATM|; x) versus the mean turbulence kinetic energy (TKE; y) at the different altitudes (a; b). Scatter plots of |SFC-ATM| (x) versus TKE (y) in the ABL (c) and above the ABL (d) (gray dots: hourly data; other dots: mean data). The hourly data were collected over a two-month period in Beijing from 27 November 2018 to 25 January 2019. (The hourly data means hourly mean values of |SFC-ATM| and corresponding hourly TKE. The mean |SFC-ATM| was obtained by averaging hourly |SFC-ATM| at intervals of 10 W m^{-2}, then the mean TKE was obtained after the average of the corresponding hourly TKE.).

10. L87-91: This is definitely not true if the authors claimed that "this paper is the first time to analyze the interaction between ….". Many studies have devoted to understanding and quantifying the interactions between aerosol radiative effect and the atmospheric boundary layer thermodynamic and dynamic structures up to now. Some examples include Zhao et al., 2019, Zhang et al., 2020, Miao et al., 2020, Liu et al. 2020, etc.

Response: Thank the reviewer very much for this suggestion. This kind of mistake has been pointed out, and we have corrected it in the first referee round. We thank the reviewer again for pointing out this problem and have modified it.

11. Line 510: Again, this study is definitely not the first one. Please delete any statement like this.

Response: We thank the reviewer again for pointing out this problem, and we have modified it.

12. L15: I am very concerned with the statement with "…because most studies have been superficial". Please delete or modify it.

Response: Thank the reviewer very much for this suggestion. This kind of mistake has been pointed out, and we have corrected it in the first referee round. We thank the reviewer again for pointing out this problem and have modified it.