February 9, 2021

Dear Editor:

We are submitting our revised manuscript, entitled "**The impact threshold of the aerosol radiative forcing on the boundary layer structure in the pollution region**" to *Atmospheric Chemistry and Physics*.

We thank the reviewer #2 for the detailed and helpful comments to improve the manuscript. Responses to the individual comments are provided below. Reviewer comments are in **bold**. Author responses are in blue plain text. Modifications to the manuscript (Tracked changes) are highlighted in red. Line numbers in the responses correspond to those in the final submitted version.

The submitted manuscript has been revised based on reviewer #2's comments.

Sincerely,

Jinyuan Xin,
Professor
Institute of Atmospheric Physics, Chinese Academy of Sciences
Beijing, China

Response to Reviewer #2's comments:

I appreciate the efforts that the authors made to address all the review comments on the original submission. However, I do find limited improvement in the revised manuscript. I still have several concerns with the conclusion and the writing in the revised version. Please see below for the detail.

We thank the reviewer for the encouragements and constructive suggestions. The response to each comment is listed below.

1. As one of the major results, the authors claimed that they were able to identify a threshold value of ARF (i.e., 55 W m^{-2}) in determining the stability of the atmospheric boundary layer (ABL) (see lines 27-29). However, such a threshold value is determined from a scatter plot (i.e., Fig.6) with limited data points that were generated by an aerosol radiation transfer model simulation (i.e., SBDART). The simulation results are dependent on not only aerosol loadings (e.g., AOD), but also aerosol optical properties (e.g., SSA) and other metrological inputs. I believe that different model configurations including types of aerosol optical property and meteorological conditions or inputs (e.g., clouds) may have different statistical relationship as presented in Fig.6. I am not sure that this threshold value (55 W m^{-2}) has a general meaning in Beijing and other regions. For instance, the relations between the ABL structure and the ARF parameter |SFC-ATM| (Fig.6) could be very different when a dust event with high concentration occurred in this region. This concern is not well addressed in the revised version.

Thank the reviewer for the comments and constructive suggestions.

Firstly, this campaign was launched in Beijing city to obtain the vertical profile observations of meteorological elements, time series of PM concentration and corresponding aerosol optical properties in the boundary layer. This experiment lasted from November 2018 to January 2019, and the threshold value of aerosol radiative forcing's effect on the boundary layer structure was obtained based on the whole two-month data.

Second, the ARF was obtained from SBDART taking AOD etc. as input parameters. AOD means aerosol optical depth, an important parameter characterizing the aerosol optical properties and radiation properties. Cloud screening is important for the photometers to observe AOD, thus the measurements was taken under daytime cloudless conditions to remove the impact of clouds. Because the AOD measurements need to exclude the cloud effects, we calculated the aerosol radiative forcing under clear sky conditions. The atmospheric parameter profiles of the observation period were taken as metrological inputs of the SBDART and were considered to reflect local weather conditions. Therefore, ARF is the radiation forcing generated by pure aerosols, which was calculated under certain meteorological conditions during the clear sky observation period. The corresponding measured TKE is screened under the same weather conditions.

Additionally, AOD was obtained in Beijing and represented the local aerosol properties where anthropogenic aerosols dominate with few dust aerosols. Whereas no matter whether Beijing is dominated by anthropogenic aerosol or dust aerosol during the observation period, the measured AOD represents the overall local aerosol optical depth during the observation period, and the ARF obtained through SBDART with AOD as the input parameter also represents the aerosol radiative forcing in Beijing during the observation period. Hypothetically, there is mainly dust aerosols, the ATM value would be larger. Conversely, SFC is more significant. However, |SFC-ATM| represents the overall aerosol direct radiation effect, including both scattering effect and absorption effect. Therefore, different aerosol composition of Beijing during the study period would make different AOD etc. and thus different |SFC-ATM| values. The |SFC-ATM| obtained in this study was representative in the winter (Dec. 2018-Jan. 2019) in Beijing. The threshold value obtained from the relationship between ARF and measured TKE is of certain reference significance for the study of aerosol-ABL interaction in winter under urban conditions in Beijing.

However, based on the reviewer's comments, it needs to be emphasized that the threshold will certainly vary from region to region; For example, in the dust-prone area in Northwest China, the aerosol mainly behaves absorption effect, so the ATM value is large, and the |SFC-ATM| value in the observation period must be generally different from that in Beijing where scattering aerosols dominate. The specific threshold value would vary from that of Beijing.

2. If you read the manuscript closely, many sentences are still not written carefully from scientific perspective. The statement of "once |SFC-ATM| exceeded ~55 W m^{-2}, the ABL structure would quickly stabilize" (Lines 28-29) is an example. Are you sure that the ABL became stable in this case? Based on my experience with large eddy simulations and field experiments, the ABL was still in a weakly unstable to neutral with the heavy PM pollution conditions. There are very chances that the ABL can reach the stable status during the heavy pollution events even in the nighttime. The 2nd example is, "… poor air quality due to rapid economic growth". This is not accurate. Actually, it was mainly due to rapid increase in anthropogenic emissions or a large amount of fossil fuel consumption. For instance, the rapid growth in economy did not cause any big trouble for air quality in US over the past several decades. The 3ds one is "Heavy air pollution episodes have always occurred with persistent inversions". There are too many sentences like these, which require very careful revision. I know that ACP will provide a language edit service, but I assume they are only for language polishing and I am not sure whether they are able to correct any inaccurate descriptions behind those sentences.

Thank the reviewer very much for these constructive suggestions.

Concerning the 1st comment "Once |SFC-ATM| exceeded ~55 W m^{-2}, the ABL structure would quickly stabilize (Lines 28-29)", we would like to clarify it in two aspects:

(1) This result came from the statistical analysis of the datasets in the winter in Beijing. As shown in Figure R1, we can find an exponential relationship between ARF and TKE. With the increase of |SFC-ATM|, TKE decreases exponentially. And TKE decreased with increasing |SFC-ATM| and hardly changed when |SFC-ATM| exceeded the critical point. Considering exponential curve characteristics, we found that once the aerosol radiative effect defined by |SFC-ATM| exceeded 50-60 W m^{-2} (average of ~55 W m^{-2}), the TKE sharply decreased from ~ 2 m^2 s^{-2} to lower than ~ 1 m^2 s^{-2}, and then changed little with further increasing |SFC-ATM|.

(2) As we previously analyzed in section 3.2, haze pollution did break out with a stable atmosphere at night: For example, in episode II (Jan 7), the level of particulate matter increased due to southerly transport in the daytime; The enhanced direct aerosol radiation effect strengthens the ground cooling, which further promotes the occurrence of stable boundary layer at night and thus the outbreak of pollution. However, even in the daytime, when severe haze pollution occurred, its significant aerosol direct radiation effect changes the vertical temperature structure largely, which will also promote the stability of the boundary layer, as shown in episode I (Dec. 15 and Dec. 16) (Figure R2, marked as dashed back lines and black arrows). The high level of particulate matter existing under the stable boundary layer in the previous night arouse very strong aerosol direct radiation forcing in the daytime, facilitating the maintenance of a stable boundary layer in the daytime. In Beijing, strong emissions, southerly transport and the high PM level existing under the previous night's stable boundary layer would make the PM concentration extremely high in the daytime and thus the aerosol direct radiation forcing, which will promote the occurrence of the strong stable boundary layer in the daytime. In turn, the diffusion of particulate matter is further inhibited, further aggravating the pollution. This is why the severe haze pollution process in Beijing often lasts for two or three days. It is the interaction between aerosol and boundary layer that makes the pollution continuously intensified and difficult to dissipate, which is the central idea that we have been emphasizing in this study. Many previous studies also measured and reported it (Zhong et al., 2018, 2019; Zhao et al., 2019): As shown in Figure R3 (marked as black arrows) and Figure R4 (marked as dashed black lines), the high PM concentration lasts for several days in Beijing, which is often accompanied by continuous temperature inversion structure (stable boundary layer). Even in the daytime, the boundary layer would be stable under heavy haze pollution condition.

About the 2nd one, we reconsidered this sentence "Most areas in China, such as the North China Plain, have suffered from poor air quality due to rapid economic growth". As a developing country, China's rapid economic development is currently largely due to the rapid industry development. It is a rapid increase in anthropogenic emissions or a large amount of fossil fuel consumption that contributes to poor air quality in most China, such as North China Plain. As the reviewer suggested, we admitted that it is not accurate enough, and have modified it in the revised version. We checked this whole manuscript to avoid the similar mistakes.

Regarding the 3rd one "Heavy air pollution episodes have always occurred with persistent inversions", we reconsidered it and understood why the reviewer pointed out it. This kind description was too subjective and we strongly agree with the reviewer's suggestion and have corrected all the kind of description.

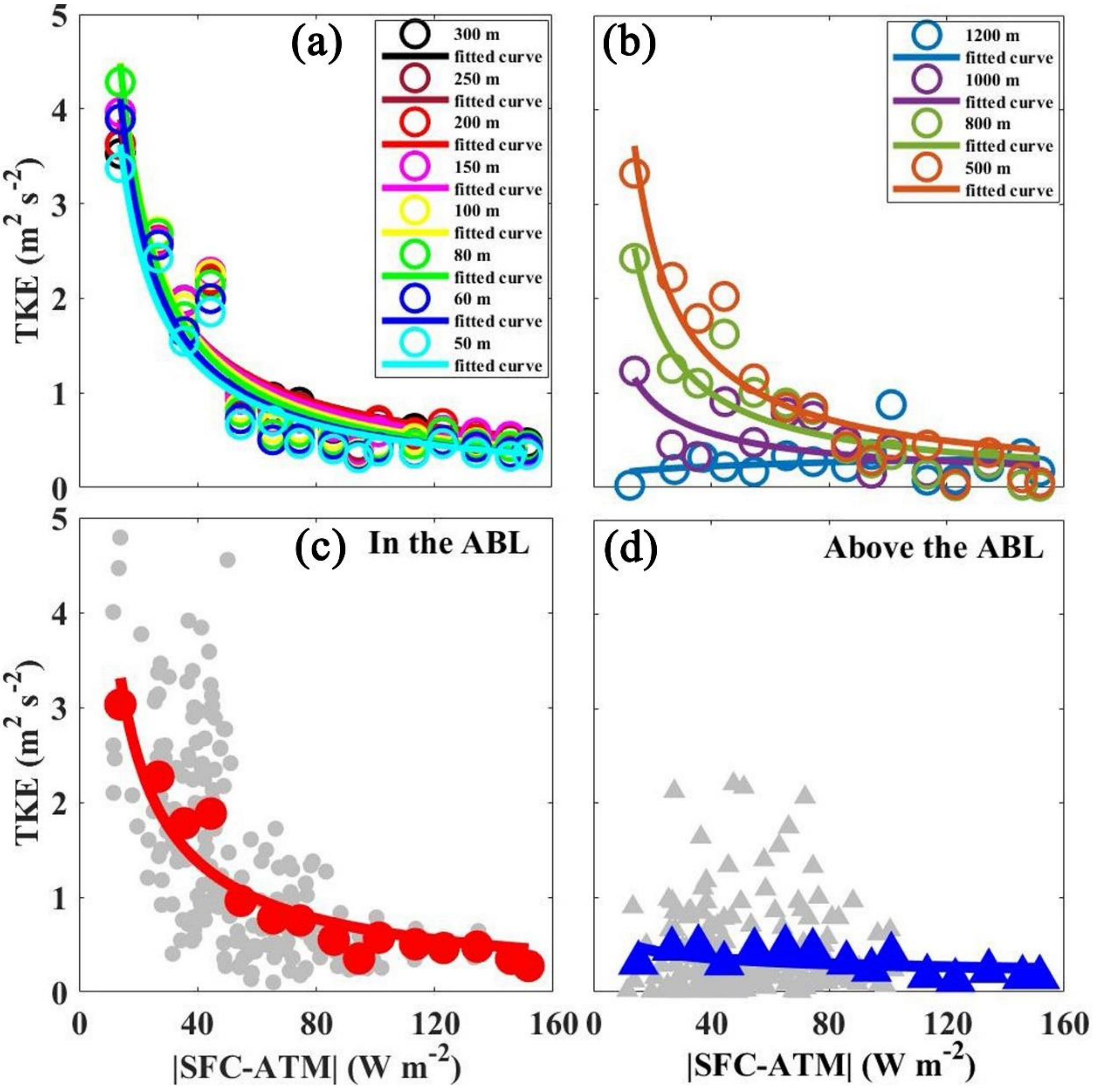

Figure R1 (Figure 6 in the manuscript). Scatter plots of the mean absolute difference of the aerosol radiative forcing at the surface and interior of the atmospheric column (|SFC-ATM|; *x*) versus the mean turbulence kinetic energy (TKE; *y*) at the different altitudes (a; b). Scatter plots of |SFC-ATM| (*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.).

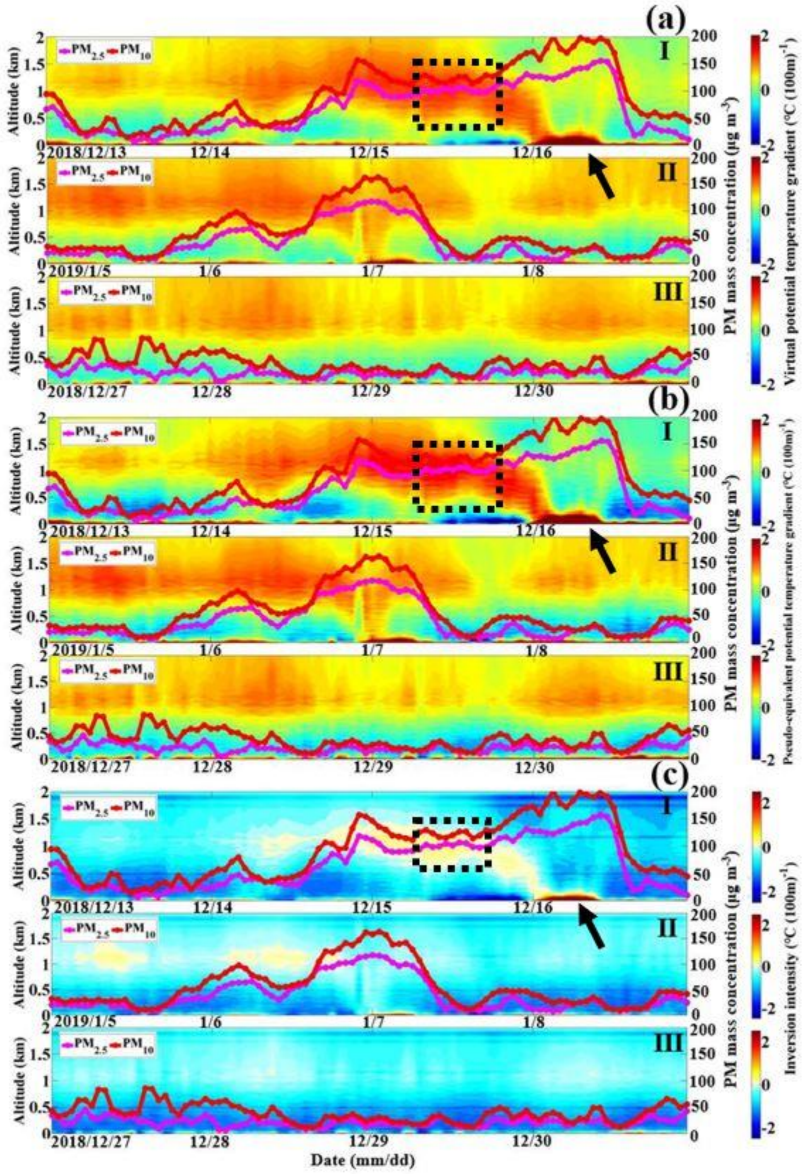

Figure R2 (Figure 2 in the manuscript). Temporal variation in the vertical profiles of (a) the virtual potential temperature gradient ($\partial\theta_v/\partial z$), (b) pseudo-equivalent potential temperature gradient ($\partial\theta_{se}/\partial z$) and (c) temperature inversion phenomenon (shaded 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).

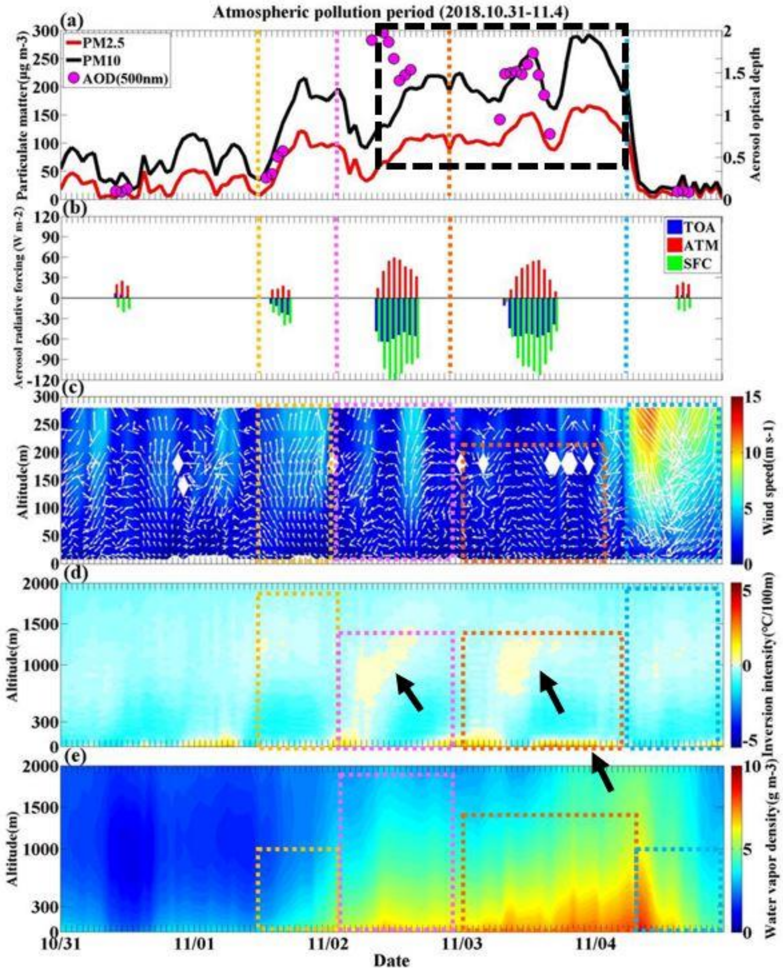

Figure R3 (Figure 5 in the study of Zhao et al. (2019)). Temporal evolution of (a) PM mass concentration (solid lines) with AOD (pink solid circles), (b) aerosol radiative forcing at the top (TOA; blue bars), surface (SFC; green bars) and interior of the atmospheric column (ATM; red bars), (c) horizontal wind vector profiles (shaded colors: wind speeds; white arrows: wind directions), (d) temperature inversion intensity profiles and (e)water vapor density profiles during a typical atmospheric pollution episode in Beijing from 31 October to 4 November 2018.

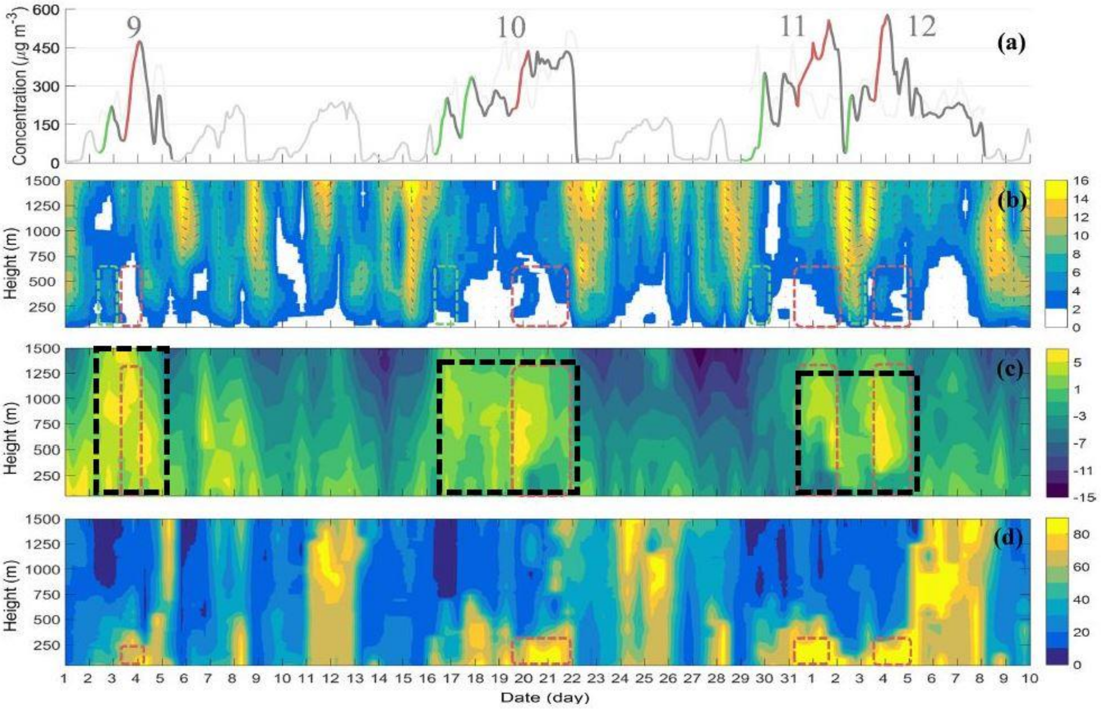

Figure R4 (Figure 4 in the study of Zhong et al. (2018)). Temporal variations in urban mean $PM_{2.5}$ and vertical distributions of meteorological factors in December 2016. (a) $PM_{2.5}$ mass concentration (dark gray or gray: Beijing; light gray: Baoding); (b) winds (vectors; red vectors: southwesterly winds) and wind velocity (shadings; units: m s-1); (c) temperature (shadings; units:℃); (d) RH (shadings; units: %). Green boxes: rising processes; red boxes: cumulative explosive processes.

3. The structures of the manuscript, especially several long paragraphs need to be re-organized. The first example is the one from Lines 64 to 102. The authors mixed the reference review and the objectives of this study together in the same paragraph. A similar issue can be found on Page 7 (L178-201), P12-14(L224-280), P14-16 (L281-341), P20-22(L416-458), etc. It is very tedious when you read these paragraphs. I would like to suggest rework them and make those paragraphs readable and understandable.

Thanks very much for this suggestion. We are very grateful for the reviewer's suggestion of such modification. As suggested, we have reworked those paragraphs and recognized them more readable and understandable in the revised version.

4. L114-118: Please add a new table to present all the input parameters (e.g., SSA, AOD) that were used in the SBDART model calculations.

Thanks very much for this suggestion. We added a "Table S2" in the Supplement to show all the input parameters that are used in the SBDART model calculations.

5. L72-74: To my understanding, surface heat flux rather than TKE is the key

driving the development of the ABL. Instead, TKE is an useful parameter describing the turbulence intensity.

We thank the reviewer for pointing out this problem. We agree with the reviewer that surface heat flux is the key driving the development of the ABL, as we wrote in lines 52-53: “Moreover, the change in solar radiation reaching the ground drives the diurnal ABL evolution…”. However, we didn’t describe this sentence accurately and we very appreciated the reviewer’s suggestion. This sentence was corrected in the revised version.

6. L444-446: Again, as pointed out in the 2nd comment above, it is impossible that the ABL can reach extremely stable state under the ABL with heavy PM pollution. Please check vertical profiles of air temperature or potential temperature.

Regarding more on this comment, we have clarified it in the 2nd comment above. Thank the reviewer again.

Besides, we double checked the vertical profiles of potential temperature obtained by microwave radiometer (MWR0109) and found that the dataset is reliable:

Figure R5 shows the accuracy evaluation of MWR measurements of atmospheric temperature and humidity in terms of vertical variation trends, correlation coefficients, biases and RMSEs against radiosonde data. For the temperature in the boundary layer (below 2000 m), as shown in Fig R5(a-d), the consistency between the MWRs and radiosonde is well. The negative biases and RMSEs vary in the ranges ~-2-0 °C and ~1-2°C, respectively. The bias and RMSE values are the lowest at ~1000-2000 m, reaching ~0-0.5 °C and ~1-1.3°C, respectively. The vertical profiles of air temperature or potential temperature of microwave radiometer are thought to be reliable in the boundary layer and more information on the accuracy of microwave radiometer data has been discussed in our previous literature (Zhao et al., 2019).

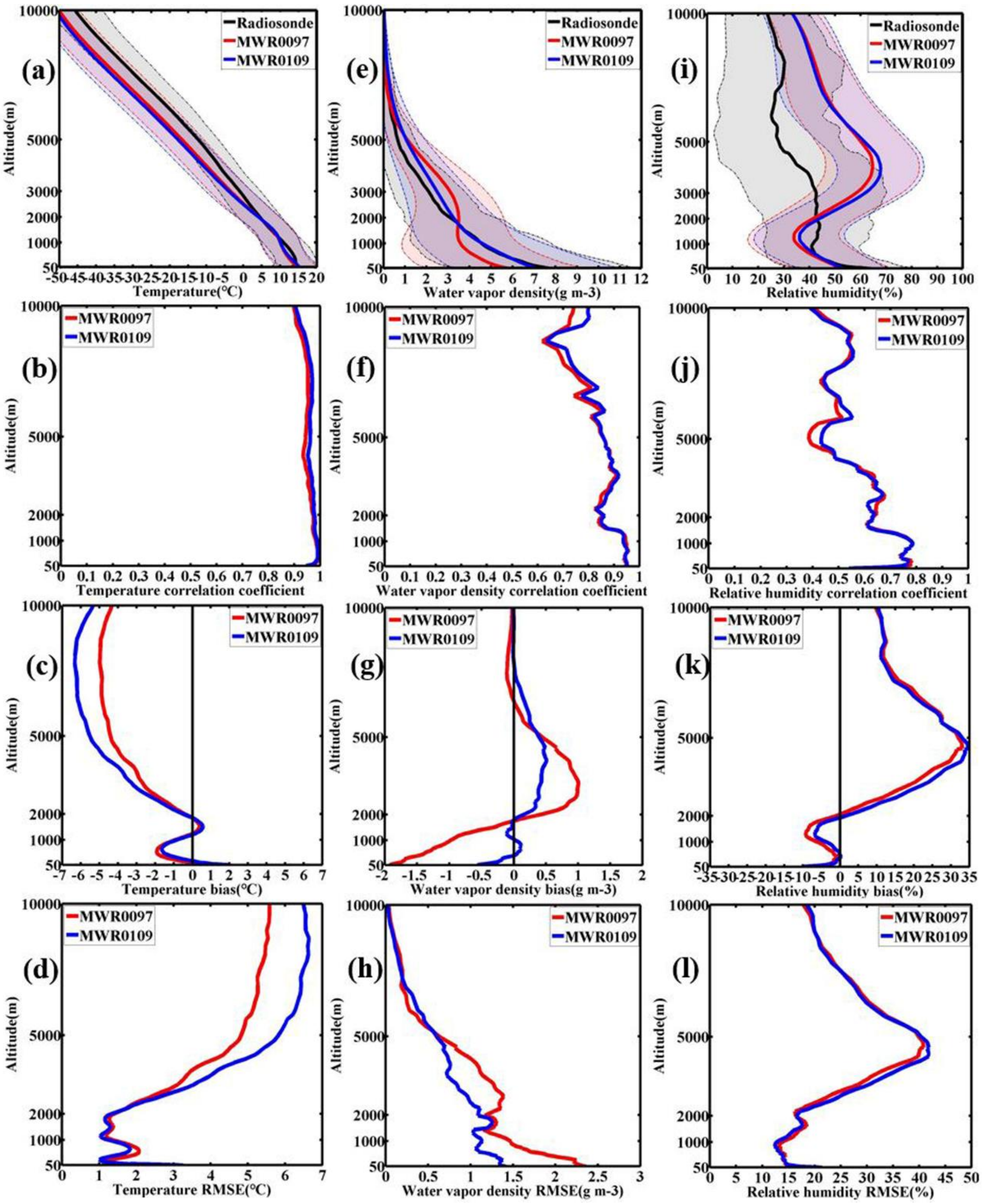

Figure R5 (Figure 2 in the study of Zhao et al. (2019)). The comparison diagrams for (a) temperature, (e) water vapor density and (i) relative humidity profile trends obtained by the microwave radiometers (MWRs) (red lines: MWR0097; blue lines: MWR0109) and radiosonde (black lines) (shaded area: standard deviations), as well as the correlation coefficients, biases, and RMSEs of MWR-retrieved (b-d) temperature, (f-h) water vapor density and (j-l) relative humidity profiles against radiosonde data under all skies selected from 25 August to 12 November 2018 (RMSE: root-mean-square error).

7. In Figure 1-(c)-I, it is surprised to see that wind speeds were higher than 10 to 15 m s^{-1} above 500 m from noon on Dec 15 to noon on Dec. 16 while surface $PM_{2.5}$

concentrations continued to increase before reaching the maximum value. Please check the observational winds used in the plotting.

Thank the reviewer for this comment.

Firstly, as the reviewer suggested, we double checked the observational winds used in this Fig. 1-(c)-I in the manuscript, and the data is reliable. Then, regarding the reviewer's doubt, we need explain it combined with the thermodynamic structure of boundary layer and further clarified it in the revised version. For better explanation, we integrated related profiles of the wind field, stability index, temperature inversion and turbulent activity of episode I as Figure R6 below. Figure R6(a)-(f) is corresponding to Fig. 1(c)-I, Fig. 2(a-c)-I, Fig. 3(a)-I, and Fig. 3(c)-I in the manuscript, respectively.

As marked as black lines in Fig. R6(b-d), there was continue temperature inversion structure in ~0.5-1.0 km and the atmospheric stratification was quite stable from noon on Dec. 15 to nighttime of Dec. 15-16. Since the temperature inversion layer acted as a lid at altitudes of ~0.5-1.0 km, downward momentum transport would be blocked, further explaining the lower atmosphere layer's calm/light winds (Fig. R6(a)). With surface cooling at night, the temperature inversion gradually turned to ground-touching temperature inversion at 0-0.2 km altitude at midnight of De. 15-16. This abnormal temperature structure lasted till noon on Dec. 16, mainly due to the strong aerosol radiative effect of pre-existing high PM level. As expected, we can see the strong wind above ~1.0 km at night of Dec. 15 gradually extended downward and eventually occurred above the ground-touching inversion in the forenoon of Dec. 16 (Fig. R6(a)). It further indicated the downward momentum transport was blocked by the temperature inversion structure. Therefore, with calm/light winds and weak turbulent activity below the temperature inversion lid on Dec. 15, the PM concentrated exactly below the inversion lid (below ~ 0.5 km) and maintained high concentrations, as the BSC distribution shows in Fig. R6(f). With strong ground-touching inversion (0-0.2 km altitude) forming in the forenoon of Dec. 16, the accumulated particles near the surface were further inhibited right in the stable atmosphere layer (below ~ 0.2 km), as reflected by the BSC distribution (Figure R6(f)). The pollutant layer was compressed downward accompanied by intense heterogeneous hydrolysis reactions at the moist particle surface, contributed to the further increase of near-surface $PM_{2.5}$ concentrations.

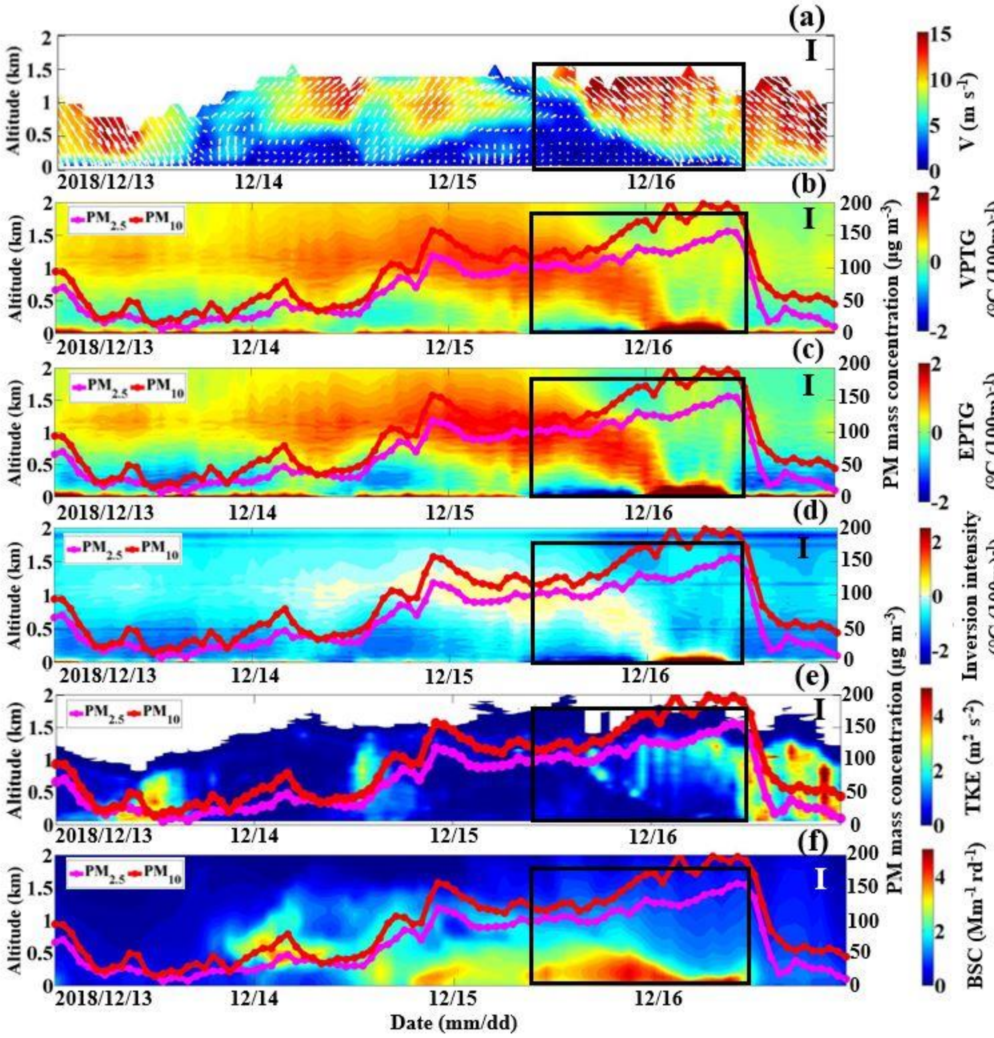

Figure R6. Temporal variation in the vertical profiles of (a) the horizontal wind vectors (shaded colors: wind speed (V)), (b) the virtual potential temperature gradient (shaded colors: $\partial\theta_v/\partial z$; VPTG), (c) pseudo-equivalent potential temperature gradient (shaded colors: $\partial\theta_{se}/\partial z$; EPTG), (d) temperature inversion phenomenon (shaded colors: inversion intensity), (e) turbulent kinetic energy (shaded colors: TKE) and (f) atmospheric back scattering coefficient (shaded colors: BSC) during the typical haze pollution episodes of I (2018/12/13-16).

8. Figure 1-(b), why does the calculated ARF show the values at several hours only rather than all the hours each day (indicated by the color bars). Please clarify.

Thank the reviewer for this comment.

ARF is calculated by SBDART with the input parameters of aerosol optical depth (AOD) and Ångström Exponent (AE) and so on. As we know, the AOD and AE are observed by automatic (CIMEL: Gregory, 2011) or manual (Microtops II: Morys et al., 2001; Ichoku et al., 2002) sun-photometers:

(1) This kind of instrument detects the direct solar radiation and retrieves AOD based on the Beer-Lambert law. Therefore, the measurements are usually carried out from 10:00 and 16:00 local time (LT) when the solar radiation is relatively strong.

(2) Cloud screening is also important for the photometers to observe AOD, thus the measurements should be taken under daytime cloudless conditions to remove the impact of clouds.

Besides, as we know, the definition of ARF is:

Due to the aerosol direct radiation effect, the absorption and scattering of aerosols change the radiation budget of the earth-atmosphere system along the solar radiation path, thus generating forcing, namely ARF. It also implies that aerosol radiative forcing exists during the daytime when there is solar radiation.

All above explains why ARF only shows the values at several hours rather than all the hours each day.

9. Figure 4: The captions of Figure 4. (a) and (c) were mixed up.

We thank the reviewer again for pointing out this mistake and have modified it in the revised manuscript.

References:

Gregory, L.: Cimel Sun-photometer (CSPHOT) Handbook, 2011.

Ichoku, C., Levy, R., Kaufman Y. J., Remer L. A., Li R., Martins V. J., Holben B. N., Abuhassan N., Slutsker I., and Eck T. F.: Analysis of the performance characteristics of the five-channel Microtops II Sun photometer for measuring aerosol optical thickness and perceptible water vapor, J. Geophys. Res., 107, doi:10.1029/2001JD001302, 2002.

Morys, M., Mims III F. M., Hagerup S., Anderson S. E., Baker A., Kia J., and Walkup T.: Design, calibration, and performance of MICROTOPS II handheld ozone monitor and sun photometer, J. Geophys. Res., 106, 14573–14582, doi:10.1029/2001JD900103, 2001.

Zhong, J., Zhang, X., Dong, Y., Wang, Y., Liu, C., Wang, J., Zhang, Y., and Che, H.: Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of PM2.5 during winter heavy pollution episodes in Beijing from 2013 to 2016, Atmos. Chem. Phys., 18, 247–258, doi.org/10.5194/acp-18-247-2018, 2018.

Zhong, J., Zhang, X., Wang, Y., Wang, J., Shen, X., Zhang, H., Wang, T., Xie, Z., Liu, C., Zhang, H., Zhao, T., Sun, J., Fan, S., Gao, Z., Li, Y., and Wang, L.: The two-way feedback mechanism between unfavorable meteorological conditions and cumulative aerosol pollution in various haze regions of China, Atmos. Chem. Phys., 19, 3287–3306, doi.org/10.5194/acp-19-3287-2019, 2019.

Zhao, D., Xin J., Gong C., Quan J., Liu G., Zhao W., Wang Y., Liu Z., and Song T.: The formation mechanism of air pollution episodes in Beijing city: Insights into the measured feedback between aerosol radiative forcing and the atmospheric boundary layer stability, Sci. Total Environ., 692, 371–381, doi.org/10.1016/j.scitotenv.2019.07.255, 2019