Responses to comments of "Interaction between aerosol and thermodynamic stability within the PBL during the wintertime over the North China Plain: Aircraft observation and WRF-Chem simulation [Preprint acp-2021-769]" to *Atmospheric Chemistry and Physics*.

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We would like to thank the editor Dr. Li, Z. and the reviewers for giving constructive criticisms and comments, which are very helpful in improving the quality of the manuscript. We have made the point-by-point response to the comments below and revised the manuscript accordingly. We hope that the revised version can meet the favorable approval and journal requirements. The referee's comments are reproduced (*black, italic*) along with our replies (blue) and changes made to the text (red) in the revised manuscript. All the authors have read the revised manuscript and agreed with the submission in its revised form. Please check them.

Responses to Reviewers

Anonymous Referee #2

General comments:

This work investigates the roles of the synoptic pattern, PBLH, aerosol type and vertical distribution in aerosol-PBL interactions by using aircraft measurements, model simulation. Several parallel numerical experiments are conducted to investigate the radiative effects of scattering and absorbing aerosols under different aerosol vertical distributions. Moreover, the long-term variation in PBL stability from 1980 to 2020 over the NCP region is examined. However, the current method and model settings in this work cannot well support the conclusion proposed, and need to be reconsidered. In addition, I personally think that hardly the case study for 2 days with a flawed method can be beneficial in determining which pollutants to target and achieving precise controls of air pollution. Here list some major concerns that need to be addressed.

Response:

Dear Reviewer,

We would like to thank you for your time in reviewing this manuscript. Many thanks for your meticulous judgments and suggestions, which are very helpful in improving our manuscript. We have made the point-by-point response to the comments below and revised the manuscript according to your substantive comments, which helps improve the quality of this paper. The revision mainly includes a more extensive description of the model setup as well as a long-term modeling for a more robust conclusion.

Major comment:

Comment NO.1:

The present study focuses on the case on 3-4 Jan 2020. However, the WRF-Chem model simulation was started on 2 Jan with only 16 hours as model spin-up. As well acknowledged, the atmospheric lifetime of aerosol is more than one week. That is to say, such a short spin-up time cannot reflect the aerosol background, chemical environment (OH radical, VOC levels and etc) and regional transport at all. Thus, it is not possible that the secondary scattering aerosol like sulfate and nitrate aerosol was well reproduced. I suggest that the authors either prolong the model time or use the other model output as the chemical initial condition.

Response: Thank you for your valuable comments. We agree with your concerns about the long period (more than one week) required for secondary aerosol production. As a matter of fact, the chemical outputs from the previous modeling periods between 1200UTC 25 December 2019 and 0000UTC 2 January 2020 (7.5 days) were used as the initial chemical conditions for the modeling. We are sorry for the ambiguous statement regarding the 16-h spin-up time, which was used to achieve a quasi-steady state of the model's meteorological process. The previous modeling results of 7.5 days, which was regarded as the spin-up time for the chemistry, were discarded in the analysis. In the revision, we have added a detailed description of the spin-up time and the initial chemical condition.

Changes in Manuscript:

[Page 6 Lines 149-155 (in the "Track Changes" version)]

"The simulations were conducted from 1200UTC on December 25, 2019 to 0000UTC on January 31, 2020, with the first 7.5 days as spin-up time for chemistry. The model was run with an 84 hours model cycle, with the first 12 hours discarded as spin-up time and the last 72-hour results used for the final analysis. The chemical outputs from previous runs were used as the initial conditions for the subsequent overlapping 84-hour simulation. The simulations of the case study were carried out from 0000 UTC on January 2 to 1800 UTC on January 4, 2020 with the first 16 hours as the model spin-up time, and the chemical outputs from the previous run were used as the initial conditions."

Comment NO.2:

Another issue concerning the model simulation is that the model adopted a 3-km grid resolution but used an emission inventory with ~30km grid, which is not very matched with each other in spatial. Please clarify. Besides, since that NCP has experienced significant emission reduction in past years, please specify the base year of the emission inventory that was used in this work.

Response: Thank you for your critical comments. In this study, the three nesting domains with horizontal resolutions of 27, 9 and 3 km were performed. Nesting is a useful technique that can be used in WRF-Chem where a single, or several higher resolution model domains (nests) are located within a coarser, parent domain. This technique makes it possible to downscale from data with large grid space to the high-resolution scales, using the parent domain as a provider of lateral boundary conditions for the nest. The resolution of the outermost domain is 27 km, which is of the same order of magnitude as the emission inventory with a ~30km grid. Moreover, previous

studies also used a similar method in the WRF-Chem modeling with the MEIC emission inventory, which is partially listed in Table Comment 2.1.

Table Comment 2.1. List of the domain information of the references.

Reference	Emission inventory	Domain
Sha, T., et al, STOTEN, 2021	MEIC, 2016	D01: 27 km; D02: 9 km
Shu, Z., et al, ACP, 2021	MEIC, 2012	D01: 48 km; D02: 12 km; D03: 3 km
Liu, C., et al, AE, 2021	MEIC, 2010	D01: 36 km; D02: 12 km; D03: 4 km
Qu, Y., et al, AE, 2020	MEIC, 2010	D01: 81 km; D02: 27 km; D03: 9 km; D04: 3 km

In addition, as you suggested, we have included the base year (2017) of the emission inventory that was used in this study in the revised version. The MEIC in 2017 is the newest and the most accurate version that can well represent the emissions in recent years, though the NCP has experienced emission reduction in the past several years.

Reference:

Liu, C., et al. "Evaluation of WRF-Chem simulations on vertical profiles of $PM_{2.5}$ with UAV observations during a haze pollution event." Atmospheric Environment (2021).

Qu, Y., et al. "Vertical structure and interaction of ozone and fine particulate matter in spring at Nanjing, China: The role of aerosol's radiation feedback" Atmospheric Environment (2020).

Sha, T., et al. "Improvement of inorganic aerosol component in $PM_{2.5}$ by constraining aqueous-phase formation of sulfate in cloud with satellite retrievals: WRF-Chem simulations." Science of The Total Environment (2021).

Shu, Z., et al. "Elevated 3D structures of $PM_{2.5}$ and impact of complex terrain-forcing circulations on heavy haze pollution over Sichuan Basin, China." Atmospheric Chemistry and Physics 21.11(2021):9253-9268.

Changes in Manuscript:

[Page 6 Lines 157-159 (in the "Track Changes" version)]

"Anthropogenic emissions were adopted from the Multi-resolution Emission Inventory for China (MEIC) in 2017 developed by Tsinghua University (http://meicmodel.org)"

Comment NO.3:

According to the model settings, the Morrison double-moment microphysics was utilized, which means that the aerosol-cloud interaction has been included in all the simulations. Thus, the differences in these simulations reflect not only the changes in ARE but also perturbations in CCN due to different emission scenarios, for example, EXP_WFexBC, EXP_WF20BC and EXP_WF20Aer. I recommend the authors reconsider and reinterpret the model result and check if it can support the conclusion.

Response: Thank you very much for your careful comments. As you mentioned, the aerosol indirect effect is a significant issue that we have previously considered in the model setup. To eliminate the influence of the aerosol indirect effect, the modeling has already set "progn = 0" and "cldchem onoff = 0". These settings only considered aerosol direct effect and turned off the indirect effect. As a result, the changes in CCN do not contribute to the variations in cloud number concentrations and have little bearing on ARE. We have added the description of the modeling setting in the revision to avoid potential misunderstandings of the readers.

Changes in Manuscript:

[Page 7 Lines 181-182 (in the "Track Changes" version)]

"To eliminate the influence of cloud condensation nuclei under various emission scenarios, the aerosol indirect effect was turned off in the modeling."

Comment NO.4:

Since the respective contributions of the absorptive and scattering aerosol are compared, the validations of the aerosol components are suggested to be conducted, especially BC, rather than just evaluating $PM_{2.5}$. It seems that the aircraft measurement and model simulation are totally isolated in terms of chemical profiles. How did the model represent the vertical profile of aerosols? It would be interesting and imperative to compare the model and the measurements.

Response: Thank you for reminding us to focus on the aircraft measured BC vertical profile when compared with the modeling result. This suggestion is of great value for the combination of aircraft measurement and WRF-Chem simulation. We have revised it as you suggested.

Changes in Manuscript:

[Page 8 Lines 203-208 (in the "Track Changes" version)]

"In addition, validations of aerosol concentrations between the modeling and in-situ observations are shown in Fig. 3. Both simulation and observation display a high level of air pollution on 3 January, and good air quality on 4 January 4. The vertical profiles of BC concentration suggest a good simulation performance, which can characterize the vertical variations and daily differences (Fig. 3a). The modeled surface $PM_{2.5}$ mass concentrations in Baoding City compare well with the ground-based measurements, especially during the daytime (Fig. 3b)."

[Page 11 Lines 226-229 (in the "Track Changes" version)]

"The statistical validations of BC concentration vertical profiles show an *R* of 0.67 and a total MB of -0.18 μg m^{-3}. The statistical validations of $PM_{2.5}$ mass concentration indicate an *R* of 0.79, a total MB of -4.91 μg m^{-3}, and an NMB of 4.69% during the daytime (Table 3). Therefore, in this study, we consider that the WRF-Chem simulation is in line with the observation and can capture the weather characteristics as well as the general distributions and variations in air pollutants."

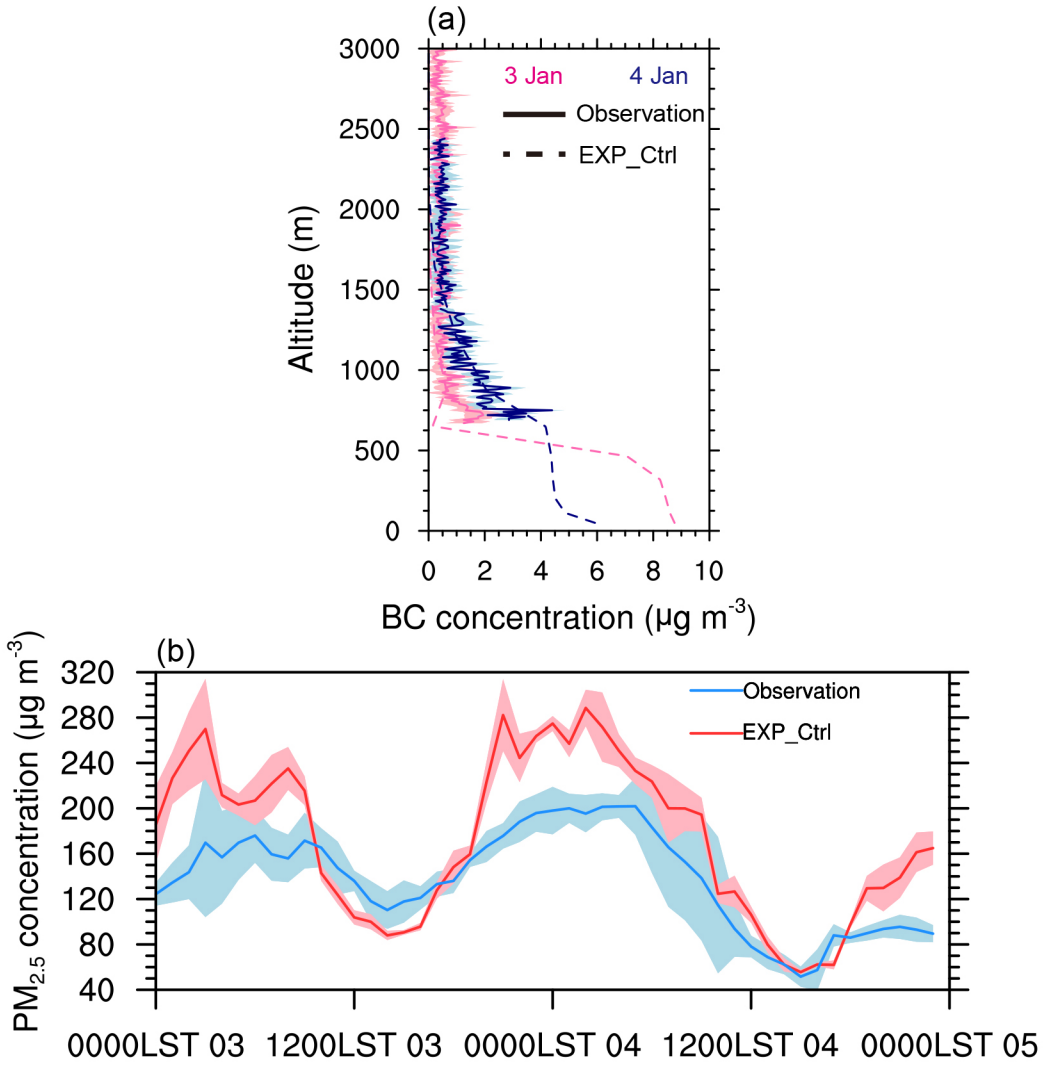

Figure 3: Validation of aerosol concentration between the modeling (EXP_Ctrl) and in-situ observations. (a) aircraft measured BC concentration vertical distributions; (b) ground-based observed $PM_{2.5}$ concentration. The shaded areas indicate the error bars (standard deviation).

Comment NO.5:

As pointed out, the combination of aircraft and model simulation was not so much. I do not think that the simulation needs to confine to these two days. Hardly the case study for just two days can represent the general conditions in this small region.

Response: Thank you for your valuable comments and insightful suggestions. As you suggested, in the revision, we have added the statistical analysis of a long-term simulation in Baoding city for nearly one month from January 3 to 30, 2020. The long-term simulation results give a more robust and representative conclusion, which can compensate for the two-day case investigation. Please see the revision below for the details.

Changes in Manuscript:

[Pages 24-29 Lines 394-469 (in the "Track Changes" version)]

3.3.3 Statistical properties of the PBL and AREs under different synoptic conditions

It is noticeable that different aerosol vertical distributions between the two days contribute to distinct AREs due to the synoptic condition and PBL thermal stability differences from the measurements and simulations. In particular, the absorptive BC aerosols have both stove and dome effects, which affect the PBL thermal structure. Here, we further analyze the modeling results for nearly one month from January 3 to 30, 2020 in Baoding city to give a more significant and representative conclusion.

Fig. 11 shows the correlations between the daily average 10 m meridional wind speed, lapse rate within 1.5 km, and PBLH. The negative correlation between the 10 m meridional wind speed and the lapse rate within 1.5 km (Fig. 11a) suggests that the increased south wind stabilizes the PBL, whereas the strong north wind destabilizes the PBL. The variation in lapse rate has a direct impact on the development of the PBL, as evidenced by the PBLH modification shown in Fig. 11b. Fig. 12 compares the distinct

vertical distributions of aerosols caused by north and south winds. Samples with a daily average wind speed within $\pm$ 0.05 m s^{-1} are discarded to avoid the north-south reverse of wind direction in a day. Eventually, 16 days with the prevailing north wind and 8 days with the prevailing south wind are used. The result indicates that the synoptic condition influences the PBL thermal structure, thereby affecting the vertical dispersion of aerosols. The warm and polluted air is carried to the NCP by the south winds, which stabilize the PBL, exacerbating the surface air pollution. The cold and clean air is carried to the NCP by the north winds, forming an unstable stratification and transporting pollutants to the upper layer.

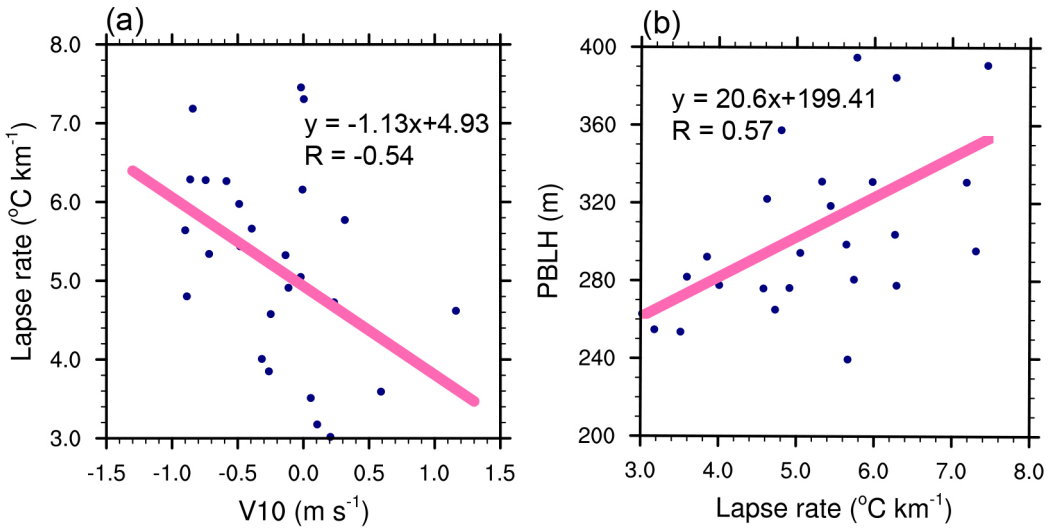

Figure 11: Scatter plots of the correlations between (a) 10 m meridional wind speed (positive: south wind; negative: north wind) and lapse rate within 1.5 km and (b) lapse rate within 1.5 km and PBLH. The data are daily averages from January 3 to 30, 2020.

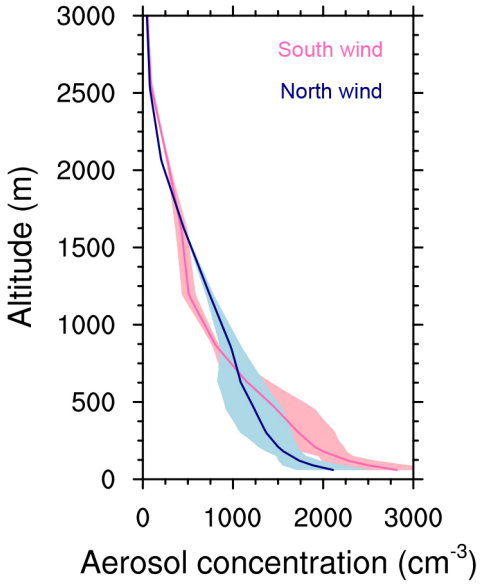

Figure 12: Vertical distributions of the aerosol number concentrations (particle diameter: 0.15-2.5 μm) under the prevailing south wind and north wind, respectively. The shaded areas indicate the error bars (standard deviation).

When evaluating the AREs of light-absorbing and light-scattering aerosols, the temperature profile variations show various patterns due to differences in aerosol concentration and vertical distribution caused by synoptic conditions, particularly the wind direction. Fig. 13 demonstrates that light-absorbing aerosols heat the atmosphere while light-scattering aerosols contribute to a cooling effect. Aerosols are constrained to the low layer under south wind conditions, and the BC aerosols result in a warming effect below 1 km (stove effect), while the scattering aerosols cool the layer below 0.6 km (umbrella effect). In contrast, the PBLs exhibit strong turbulence mixing when influenced by the north winds, and the aerosols are carried to the aloft layer. The aloft scattering particles prevent incident solar radiation from reaching the low layer, resulting in cooling effects below 1 km (umbrella effect), whereas the aloft absorbing aerosols heat the upper layer between 0.5 and 1.5 km (dome effect). The remarkable aerosol effects under south winds attribute to the accumulation of aerosols under adverse weather conditions. The contrasting aerosol vertical distributions caused by the varying synoptic conditions lead to different AREs, which is consistent with the results obtained on January 3 and 4, 2020.

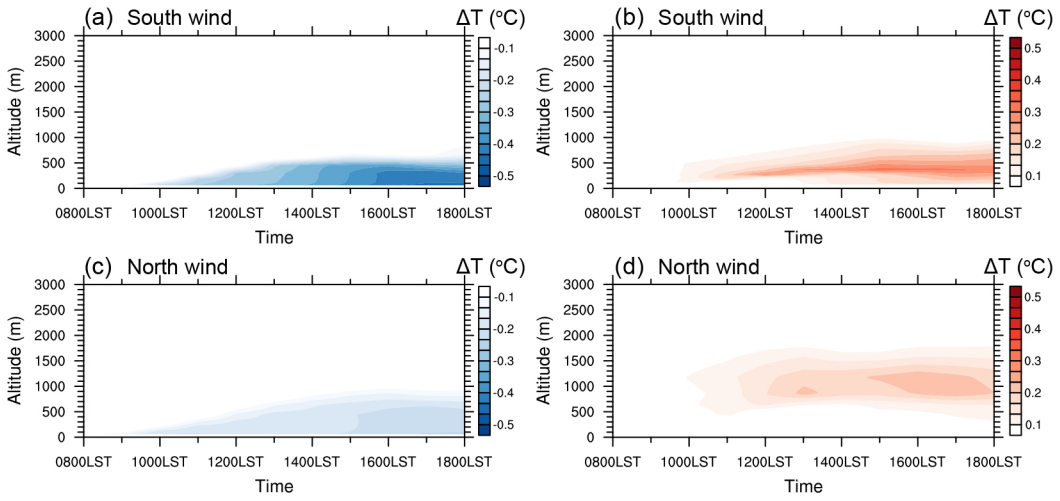

Figure 13: Temporal evolution of the temperature profile variation influenced by aerosol radiative effect (ARE). (a) ARE by other aerosols (EXP_WFexBC – EXP_WoF) and (b) ARE by BC (EXP_Ctrl – EXP_WFexBC) under the prevailing south wind; (c) ARE by other aerosols and (d) ARE by BC under the prevailing North wind.

Furthermore, based on the nearly one-month simulations, we quantify the variations in lapse rate within 1.5 km and PBLH under different synoptic conditions, respectively, caused by absorptive BC aerosols and other light-scattering aerosols. The results in Fig. 14 reveal that the BC stove effect induces a 0.04 ℃ km^{-1} increase in lapse rate within 1.5 km and a 3 m increase in PBLH under the stable stratifications with the prevailing south winds. However, the BC dome effect causes a 0.085 ℃ km^{-1} decrease in lapse rate within 1.5 km and a 3 m decrease in PBLH under the unstable stratifications with the prevailing north winds. The umbrella effect of scattering aerosols in both stable and unstable conditions reduces the lapse rate by about 0.15 ℃ km^{-1} and reduces the PBLH by about 3.5–4 m. The vertical distribution of absorbing aerosols has a significant impact on their aerosol-PBL feedback. The absorbing aerosols concentrated in the low layer have a strong radiative heating effect on the atmosphere develop the PBL in the case of stable weather patterns under the influence of the south wind. The absorbing aerosols in the upper layer heat the atmosphere and inhibit the development of the PBL in the case of unstable weather patterns under the influence of the north wind. The inhibition effect of scattering aerosols on the PBL, on the other hand, is independent of the aerosol height distributions and is solely dependent on the aerosol concentrations.

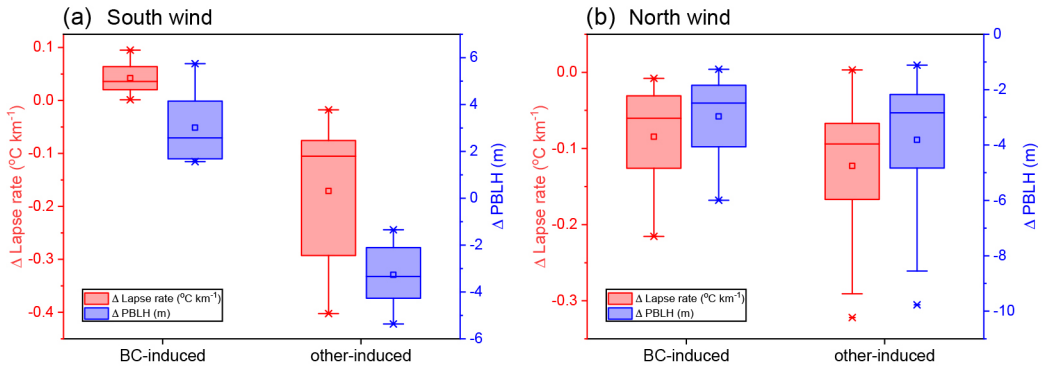

Figure 14: Box plots of the variations in lapse rate within 1.5 km and PBLH influenced by the BC and other aerosols under (a) the prevailing south wind and (b) the prevailing north wind, respectively. The squares represent the mean values, the horizontal lines inside the boxes are the medians, and the bottom and top sides of the boxes represent the first and third quartiles. The whiskers are the minimum (maximum) values within 1.5 interquartile ranges of the lower (upper) quartile. The asterisks indicate the minimum (maximum) values.

Comment NO.6:

Figure 12 is not a good way to show the impact of PBL and pollution. PBLH cannot well reflect the structure itself. And for CO, it is a relatively long-lived species in the atmosphere with a background concentration of around 100 ppb. The short-term perturbations of aerosol on PBL just for two days cannot substantially influence the concentration since the background concentration in the lower troposphere is way larger than the perturbation caused by ARE.

Response: Thank you for your constructive criticisms. The original reason we chose CO to study the effects of PBL on aerosol is that CO is a rather stable component in the atmosphere which is little impacted by other particles. We agree with your assessment that the CO concentration cannot be substantially influenced by the perturbation caused by ARE, as a result, the correlations shown in Fig. 12 are not immediately apparent. The primary aims of Figs. 11-12 were to provide a spatial statistical conclusion of the aerosol-PBL feedback, but the revision gives a long-term simulation finding that is

more representative than the prior analysis. In any case, we have replaced the prior Figs. 11-12 with the updated results presented in the response of Comment NO.5.

Minor issues:

Comment NO.7:

Table2: NMB makes no sense when evaluating air temperature.

Response: Thank you. The temperature has an MB of -0.87℃, which is within acceptable limits when compared with the previous study (Ding, A., et al., GRL, 2016, temperature MB: -1.25~1.02℃). Due to the low temperature value, the NMB is relatively higher. The correction coefficient between the ground-based (aircraft) observation and EXP_Ctrl is 0.87 (0.98) that indicating the model well describes the temperature variation.

Reference:

Ding, A., et al. "Black carbon enhances haze pollution in megacities in China." Geophysical Research Letters (2016).

Comment NO.8:

Correct the unit of mass concentration to μg in main figures.

Response: Thank you. We have corrected them in Fig. 3 and Fig. 7.

Comment NO.9:

In Figure 13, Label the correlation coefficient and specify the location of LR in the caption. Is b-c the correlations between ESWM and LR, SH and LR or their anomalies? The data needs to be double-checked.

Response: Thank you. The correlation coefficient is labeled in b-c. LR is calculated in domain d03 depicted in Fig. 1b between 1000 hPa and 850 hPa, which has been

specified in the revision. In addition, we have stated that b-c is the correlations between their anomalies in the revision.

Changes in Manuscript:

[Page 30 Lines 491-495 (in the "Track Changes" version)]

"Figure 15: (a) Time series of the standardized anomaly of the wintertime boundary layer lapse rate (LR) in domain d03 between 1000 hPa and 850 hPa, the index of Siberian High (SH), and the index of East Asian Winter Mooson (EAWM) from 1980 to 2020. The scatter plots of the correlations between (b) the standardized anomaly of the ESWM and LR, as well as between (c) the standardized anomaly of the SH and LR. Standardized anomaly is calculated by dividing anomalies by the climatological standard deviation."

Again, we would like to thank you for taking your time to review this manuscript and providing insightful comments and advice. we believe that this work has been much improved with your constructive and informative remarks.

Dr. Yong Han

On behalf of all the authors