Dear Editors and Referees:

Thank you very much for your review and comments concerning our manuscript entitled "Elevated 3D structures of PM_{2.5} and impact of complex terrain-forcing circulations on heavy haze pollution over Sichuan Basin, China" [MS No.: acp-2020-1161]. Those comments are all valuable and helpful for revising and improving the manuscript. We have studied comments carefully and have accordingly made the revisions. Revised parts are highlighted with Track Changes in the revised manuscript. In the following we quoted each review question in the square brackets and added our response after each paragraph.

Response to Referee #1

[The paper analyzes an episode with high concentrations of PM2.5 in the Sichuan Basin (China), combining observations and numerical simulations. The paper is potentially interesting, in particular for the peculiar interaction between meso and local circulations and pollutant emissions, which leads to the formation of an elevated pollutant layer. However, the discussion of the results should be improved before the paper can be accepted for publication.]

Response 1: Many thanks for your encouraging comments. We have revised the manuscript accordingly. All the revisions have been highlighted with Track Changes in the revised manuscript. The point-by-point responses to the reviewer's comments are as follows.

General comments

[1. A general meteorological overview of the event, including a synoptic characterization, is missing in the paper.]

Response 2: Following the referee's comment, We have plotted the 700hPa geopotential heights and wind vectors in three stages (newly added Figure 3) over the SCB and the surrounding regions with the meteorology analysis data of ERA-Interim.

We also characterized the overview of the synoptic conditions in the revised Sect. 2.4 as follows:

"The meteorological overview of the haze event was characterized by the 700 hPa fields of geopotential heights and wind vectors (Fig. 3). A trough in the mid-latitude westerlies moved eastward from the eastern edge of the TP to the western SCB margin during P1, the trough of low pressure evolved over the SCB region during P2, and the westerly trough shifted out the SCB region with the low-pressure system disappearing in the P3 (Fig. 3). The changes in atmospheric circulations in the three stages reflected the meteorological modulation of heavy haze development over the SCB in associated with the effect of TP topography on the westerlies."



Figure 3. The 700 hPa geopotential height fields and wind vectors averaged during (a) P1, (b) P2 and (c) P3 stages with the trough line (brown line) and low-pressure center (L). The SCB was outlined with the red solid lines.

From the formation to the maintenance and the dissipation periods of haze pollution, the near-suface prevailing northeasterly winds strengthened gradually over the SCB. During the formation and maintenance stage, the surface winds were weak (1.4–1.7 m s⁻¹) over the SCB, which was insufficient to dispel the air pollutants, but to continuously accumulate PM_{2.5} locally from light to heavy pollution conditions (Fig. 7a, Fig. 7b). By the dissipation period, the northeasterly winds intensified and removed PM_{2.5} from the SCB (Fig. 7c).

[2. The Authors adopt a grid ratio of 1:4, while an odd grid ratio is recommended because for even values interpolation errors arise due to the nature of Arakawa C-grid staggering. The authors should at least discuss this choice.]

Response 3: Thanks for the referee's suggestion. In the revised manuscript, we have accordingly added the following discussions in the revised Sect. 2.3:

"Considering the complex terrain underlying of the SCB's deep basin and surrounding plateaus and mountains in Southwest China, we adopted a grid ratio of 1:4 for simulation experiments with a precisely defined horizontal resolution. It should be noted that the even grid ratio may cause interpolation errors at the nested-domain boundary conditions owing to the nature of Arakawa C-grid staggering.

[3. The Authors say that the "vertical turbulent diffusion coefficient of the boundary layer was reduced". This aspect should be better discussed, since it might significantly affect the results.]

Response 4: Following the referee's comments, we have added the below discussions into the revised Sect. 2.3.

"High PM_{2.5} levels in the atmosphere could significantly reduce the near-ground solar radiation for stable atmospheric stratification, which decreases the vertical turbulent diffusion in the boundary layer (Wang et al., 2019). This is an important mechanism in the formation of severe haze pollution with the explosive growth of PM_{2.5} (Zhong et al., 2018). The overestimated vertical diffusion capacity under poor air quality conditions (Ren et al., 2019) causes deviations in air pollutant concentrations simulated in air quality models (Wang et al., 2018). In this study, the vertical turbulent diffusion coefficient of the atmospheric boundary layer was cut halfway for better simulation of the 3D structures of PM_{2.5}, during the heavy air pollution event over the SCB region."

References

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[4. No information about the vertical discretization is given. An adequate vertical resolution is fundamental to evaluate the thermal stratification over complex terrain.]

Response 5: Thanks for the referee's comments. The information about the vertical discretization is added in the revised manuscript (lines 114-116) as follows:

"An adequate vertical resolution is fundamental for evaluating thermal stratification over a complex terrain. Therefore, 35 vertical layers were set with fine resolutions of 30–120 m in the boundary layer."

[5. The Authors propose a series of statistical indexes for evaluating model results, both for meteorological variables and PM2.5. From these statistical indexes it is difficult to judge the performance of the model, regarding in particular the time evolution of observed and simulated variables. I strongly suggest to show some representative time series to better evaluate the model performance at some representative location.]

Response 6: Following the referee's suggestion, the hourly variations of $PM_{2.5}$ concentrations, 2 m air temperature, surface relative humidity and near-surface wind speed in Chengdu (site 1), Suining (site 10) and Zigong (site 13) were shown in Figures S1 and S2 in the supplement of manuscript. The comparisons between

observation and simulation also were evaluated with the reasonable WRF-Chem modeling performance.



Figure S1. Hourly variations of observed (black curves) and simulated (red curves) $PM_{2.5}$ concentrations respectively in (a) Chengdu (site 1), (b) Suining (site 10) and (c) Zigong (site 13) during the haze pollution episode.



Figure S2. Hourly variations of observed (black curves) and simulated (red curves) 2 m air temperature, surface relative humidity and wind speed results respectively in (a) Chengdu (site 1), (b) Suining (site 10) and (c) Zigong (site 13) during the haze pollution episode in 2017.

[6. Figure 4 presents a comparison between the vertical profiles of potential temperature, wind speed and relative humidity from observations and model results. Also in this case it is difficult to evaluate model results, since only mean profiles and the variation range over the entire period are presented. I suggest to show also some representative profiles at some specific hours. In particular, it would be interesting to evaluate how the WRF model is able to capture the vertical temperature profile, since atmospheric stability is crucial for pollutant dispersion. In many points in the paper a temperature inversion is cited, but the simulation of this temperature inversion is never discussed. For example, at lines 243-250, "thermo-dynamical structures" and "stable stratification" are cited, but, without a representative figure, it is difficult to follow the discussion of the results.]

Response 7: Following the referee's suggestions, the vertical air temperature profiles were evaluated with the comparisons of vertical air temperature profiles between observation and simulation during different haze periods at (a) 11:00 p.m. on 2 January, (b) 05:00 a.m. on 3 January, (c) 02:00 p.m. on 3 January, (d) 02:00 p.m. on 4 January, (e) 08:00 p.m. on 4 January, (f) 05:00 p.m. on 5 January, (g) 08:00 a.m. on 6 January, (h) 11:00 a.m. on 6 January and (i) 11:00 a.m. on 7 January 2017. (Figure S3 in the supplement of manuscript) with the following description added in the revised Sect. 3.1:

"Compared with the observed air temperature, the WRF-Chem simulations were evaluated to reasonably capture the vertical temperature profiles for understanding atmospheric stability in the vertical thermodynamic structures of the boundary layer over the SCB (Fig. S3)."

We have clarified the description of "thermo-dynamical structures" and "stable stratification" into the revised Sect. 3.3.

"The potential temperature vertical gradients (Fig. 5), which are used for assessing atmospheric stability, were estimated respectively with 4.0 K/km, 7.8 K/km

and 5.2 K/km in the boundary layer during the three periods of haze pollution with near-surface strong temperature inversion (Fig. S3), presenting the thermodynamic structure with stable stratification in the atmospheric boundary layer, weakening the air pollutant dispersion."



Figure S3. Comparisons of vertical profiles of air temperature between observation (black curves) and simulation (red curves) at (a) 11:00 p.m. on 2 January, (b) 05:00 a.m. on 3 January, (c) 02:00

p.m. on 3 January, (d) 02:00 p.m. on 4 January, (e) 08:00 p.m. on 4 January, (f) 05:00 p.m. on 5 January, (g) 08:00 a.m. on 6 January, (h) 11:00 a.m. on 6 January and (i) 11:00 a.m. on 7 January 2017.

[7. Although the paper is rather well written, a review by a native English speaker would be beneficial.]

Response 8: Thanks for your positive comments. A native English speaker in ELSEVIER Language Editing Services has reviewed the paper to improve the language (please see the following certificate).



Specific comments:

[Page 2, line 79: "Section 2 introduced...". Here and in other parts of the paper I would use the present tense (when referring to tables, figures...).]

Response 9: Thanks for the careful review, we have changed with the present tense (when referring to tables, figures...).

[Figures 7 and 8: the location of the cross sections should be indicated in Fig. 1.]

Response 10: We have indicated the location of the cross sections in the modified Figure 1.



Figure 1. (Left panel) three nesting domains D1, D2 and D3 of WRF-Chem simulation with the terrain heights (m in a.s.l.) and (right panel) the location of 18 urban observation sites (black dots, Table 1) including site 1 (Chengdu) with the intensive sounding observations and site 15 (Ya'an) with the ground-based MPL detection in the SCB with the surrounding Tibetan Plateau (TP), Yunnan-Guizhou Plateau (YGP), Mountains Daba (Mt. Daba) and Wu (Mt. Wu) in Southwest China. The red dash lines indicate the location of the cross sections respectively along 30.67° N and 104.02° E.

Response to Referee #2

[This paper analyzed the three-dimensional distribution of PM2.5 concentrations in Sichuan Basin during a heavy haze pollution episode in January 2017. The topic is quite interesting; However, many discussions are only general descriptions of phenomena and processes, lacking in-depth analysis and discussion. This makes the article as a whole difficult to follow.]

Response 1: Many thanks for your encouraging comments. We have revised the manuscript accordingly with in-depth analysis and discussion. All the revisions have been highlighted with Track Changes in the revised manuscript. The point-by-point responses to the reviewer's comments are as follows.

[1. Line 115: The Multi-resolution Emission Inventory for China (MEIC) has been

updated to 2017 (http://www.meicmodel.org), while the authors used the data of 2012 for the simulation period in January, 2017. The author should explain the mismatch. Further, the first domain (D1) of the study area includes China and its neighboring countries/regions. In this section, the author only stated that the anthropogenic emission sources used in their study is MEIC data, but as far as I know, MEIC data only includes the anthropogenic emission sources in China, while the emissions from natural sources and neighboring countries/regions are not included. How did the author consider this in their simulation process? If the emission data of neighboring countries/regions are not included, there will be great uncertainty about the results of the section 3.5 (Contribution of local emission and outflow transport), because the surrounding emissions are ignored.]

Response 2: Thanks for the comments. We have accordingly added the following explanation and discussions in the revised conclusions (Sect. 4.):

"MEIC 2017 was not available for the WRF-Chem model. The SCB is located in Southwest China with larger uncertainties in the anthropogenic emission inventory compared to Eastern China. An accurate emission inventory could improve air pollution simulations and air quality change assessments in future studies."

Furthermore, we have clarified the emissions from natural sources and neighboring countries/regions in the revised manuscript (Sect. 3.5) as follows:

"The Model of Emissions of Gases and Aerosols from Nature (v2.1) was applied to the natural emission sources in the simulation with dust emission parameterization. The SCB region in the northeastern part of Southwest China, characterized by a deep-bowl structure, is isolated by plateaus (TP in the west and YGP in the south) and mountains with a clean atmospheric environment. Haze pollution events with extremely high PM_{2.5} concentrations over the SCB are ascribed to the accumulation of local anthropogenic pollutants and air pollutant transport over the basin (Wang et al., 2018; Qiao et al., 2019; Zhao et al., 2019). High local anthropogenic pollutant emissions in the SCB dominate regional air pollution over the SCB (Liao et al., 2017). The transport of air pollutants from neighboring countries in South Asia is mostly concentrated in the neighboring regions of the southern TP and southern YGP (Wang et al., 2018; Zhao et al., 2019; Yin et al., 2020). Therefore, the anthropogenic emission data of South Asian neighboring countries of China are not included in the WRF-Chem simulation on haze pollution over the SCB during 2–8 January 2017, considering the less effects of northward cross-border transport of air pollutants from South Asian neighboring countries on air pollution in SCB with prevailing northeasterly wind during Asian winter monsoon season with a negligible contribution to the wintertime heavy haze pollution over the SCB region."

References

Liao, T., Wang, S., Ai, J., Gui, K., Duan, B., Zhao, Q., Zhang, X., Jiang, W., and Sun, Y.: Heavy pollution episodes, transport pathways and potential sources of PM_{2.5} during the winter of 2013 in Chengdu (China), Science of the Total Environment, 584, 1056-1065, https://doi.org/10.1016/j.scitotenv.2017.01.160, 2017.

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Yin, D., Zhao, S., Qu, J., Yu, Y., Kang, S., Ren, X., Zhang, J., Zou, Y., Dong, L., Li, J., He, J., Li, P., and Qin, D.: The vertical profiles of carbonaceous aerosols and key influencing factors during wintertime over western Sichuan Basin, China, Atmospheric Environment, 223, 1352-2310, https://doi.org/10.1016/j.atmosenv.2020.117269, 2020.

[2. Line 157: 3.1 Model evaluation As we know, China has adopted active pollution source control policies in the last 5 years, and the intensity, the temporal and spatial distribution of emission sources will vary greatly from year to year. The author selected the 2012 MEIC inventory as its emission data. Thus the model evaluation result may not be convincing.]

Response 3: We agree with the referee that China has adopted active pollution source control policies in the last 5 years, and the intensity, the temporal and spatial distribution of emission sources will vary greatly from year to year. We have added the explanation and discussions about the 2012 MEIC emission data in the revised conclusions as follows:

"MEIC 2017 was not available for the WRF-Chem model. The SCB is located in Southwest China, with larger uncertainties in the anthropogenic emission inventory compared to Eastern China, An accurate emission inventory could improve air pollution simulations and air quality change assessments in future studies."

[3. Line 216: To examine the vertical structures of PM2.5 concentrations over SCB, we selected the urban site 1 (104.02° E; 30.67° N) in Chengdu (cf. Fig. 1) as a reference point to investigate the distributions of PM2.5 and the atmospheric circulations respectively in the vertical-meridional and vertical-zonal cross-sections. Why do you select the urban site 1 (104.02° E; 30.67° N) in Chengdu for the vertical discussion. Do you have any special purpose? Chengdu is located in the far west side of the SCB, and other sites in the central area of SCB maybe are better choices, as the wind vectors shows in Figure 6.]

Response 4: We selected the urban site 1 (104.02° E; 30.67° N) in Chengdu for the vertical discussion with the following purposes:

1) The terrain effect of TP, the "world roof" on the mid-latitude westerlies could modulate haze pollution in the downstream region over China (Xu et al., 2016). The SCB is immediately to the east of TP with a large elevation drop exceeding 3000 m over a short horizontal distance. The unique terrain effect generates the asymmetries of meteorological and air pollutant distribution over the SCB (Zhang et al., 2019). Chengdu on the far west side of the SCB was selected to better understand the elevated 3D structures of PM_{2.5} with the impact of TP terrain-forcing circulations on the haze pollution episode over the SCB.

2) Chengdu (site 1), is a metropolis in the SCB with high anthropogenic emissions and the most polluted environment in Southwest China (Ning et al., 2018). It is important to investigate how the urban surface high $PM_{2.5}$ levels evolved vertically in the atmosphere with the combination between the high urban emissions and TP's terrain-forcing lifting over the SCB.

Furthermore, we have plotted the cross-sections of $PM_{2.5}$ and wind vectors along the near-surface prevailing northeastern wind across the central SCB (blue line in

Figure S4 of manuscript supplement). The vertical changes of $PM_{2.5}$ with the terrain-forcing local circulations by YGP-terrain effects were remarkably presented in the different stages of the heavy haze pollution event.



Figure S4. Northeast-southwest cross-sections along the near-surface prevailing wind (blue line) over the SCB (left panel), $PM_{2.5}$ concentrations (color contours: $\mu g m^{-3}$) and wind vectors (right panel) in (a) the relative clean environment at 12:00 a.m. on 2 January, (b) heavy air pollution formation stage at 12:00 a.m. on 3 January, and (c) maintenance stage at 8:00 a.m.on 6 January 6 and (d) dissipation stage at 8:00 a.m. on 7 January, 2017.

References

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Ning, G., Wang, S., Ma, M., Ni, C., Shang, Z., Wang, J. and Li, J.: Characteristics of air pollution in different zones of Sichuan Basin, China, Sci. Total. Environ., 612, 975–984, 440 https://doi.org/10.1016/j.scitotenv.2017.08.205, 2018.

[4. Line 275: Figure 10 showed the PM2.5 concentrations emitted from the regional air pollutant sources over the SCB region and the relative contribution rates to air pollution changes.

The expression here shows the author's conceptual misunderstanding of the source of PM2.5. How can the "PM2.5 concentrations" be "emitted"?]

Response 5: We modified the expression as "Figure 11 shows the $PM_{2.5}$ concentrations originating from local emissions of primary $PM_{2.5}$, gaseous precursors of $PM_{2.5}$ over SCB and the relative contribution rates to air pollution changes."

[5. Line 278: The SCB's regional air pollutant emissions provided surface PM2.5 concentrations from 40.6 to 136.2 µg m-3, contributing 75.4–94.6 % of surface PM2.5 concentrations for the heavy pollution episode over SCB, indicating its dominant role over this isolated deep basin in Southwest China. What does "indicating its dominant role over this isolated deep basin in Southwest China" mean? It is hard to follow.]

Response 6: In the revised manuscript, we have clarified this sentence as "The SCB's regional air pollutant emissions provided surface $PM_{2.5}$ from 40.6 to 136.2 µg m-3, contributing 75.4–94.6% of total concentrations for the heavy pollution episode over SCB. This indicates the dominant role of local air pollutant emissions on air quality changes over this isolated deep basin in Southwest China."

[6. Line 279: The regionally emitting PM2.5 concentrations averaged over SCB were 88.64, 91.04 and 65.96 μ g m-3 for the formation, maintenance and dissipation periods, respectively. Same as above. How can the "PM2.5 concentrations" be "emitted"?]

Response 7: The expression has been corrected as "The surface $PM_{2.5}$ concentrations sourced from the regional air pollutant emissions over the SCB were averaged, with 88.64, 91.04 and 65.96 µg m⁻³ for P1, P2, and P3, respectively."

[7. Line 284: We think the exchanges of PM2.5 between the polluted air over SCB and the cleaner environment air over the surrounding plateaus and mountains in Southwest China play a role in this process. (Figs. 7 and 8). How do you think the PM2.5 can be "exchanged" between the polluted air over SCB and the cleaner environment air?]

Response 8: Thanks for the referee's careful review. The sentence has been modified as "This could be attributed to the exchanges between the $PM_{2.5}$ -rich airmass over SCB and $PM_{2.5}$ -poor airmass in the surrounding plateaus and mountains over

Southwest China (Fig. 8 and 9)."

[8. Line 560: Table 5. Please give the definite range of the eastern TP edge (ETP), northern YGP edge (YGP) and DBM region.]

Response 9: The definite ranges of the three regions were defined with the altitudes of 750–3500 m over 30.5–33.0° N, 102.7-105.3 °E (the eastern TP edge), 750–3000 m over 27.8–29 °N, 103.5–108.5 °E (northern YGP edge) and above 750 m over 31.5–33.0 °N, 106.0–109.4 °E (DBM region), as shown in Fig. S5.

The above description has been added in the Sect. 2.3.



Figure S5. The roughly periphery of eastern TP edge (ETP), northern YGP edge (YGP) and DBM region (black lines) in the study with the terrain heights.

[9. Line 600: Figure 5(a). Why only 8 hours data are presented here? There is an abnormal value around half past 10 a.m., please give the reason.]

Response 10: The MPL is located at site 15 of the western margin of SCB (Fig. 1). The layer of high $PM_{2.5}$ concentrations with the vertical hollow was observed between 1–2 km during the 8-hr haze maintenance stage P2 at site 15.

The abnormal values as might be caused by the background noise of MPL,

giving rise to an aberrant point of observation with extremely high extinction coefficients at this point. The abnormal values around half past 10 a.m. have been removed in the modified Figure 6.



Figure 6. Vertical and time cross-sections of PM_{2.5} mass concentrations (µg m⁻³) from (a)

MPL-4B retrievals products and (b) simulation results at site 15 (Fig. 1; Table 1) in the western SCB edge during 7:00 a.m.–2:00 p.m. on 5 January 2017.

[10. Line 610 -615: Figure 8. Does this cross section along 104.02° E? Please specify. Same as previous mentioned, why do you select this site 1 (104.02° E; 30.67° N)? Do you think it may be a better choice if you put the cross section along the wind vectors from northeast to southwest?]

Response 11: Yes, Figure 9 (in the revised manuscript) actually exhibited the cross sections along the Chengdu (104.02 °E). The vertical cross section along the northeast-southwest wind vectors were provided in Figure S4 (please see our response 4). The separate height-longitude and height-latitude cross sections could better represent the vertical circulation changes and $PM_{2.5}$ distribution over the SCB.

[11. Line 625: Figure 10. How the values of surface PM2.5 concentrations are calculated? The regional average of the SCB or the average of several monitoring sites in SCB?]

Response 12: Both surface $PM_{2.5}$ concentrations and the contribution proportions in Figure 11 (in the revised manuscript) were calculated with the regional averages over the SCB rather than the averages of several monitoring sites in cities. The caption of Figure 11 has been revised as follows:

"Figure 11. Hourly variations of surface $PM_{2.5}$ concentrations originating from the SCB's anthropogenic emissions (blue filled areas) and the contribution proportions to the basin surface $PM_{2.5}$ levels (red curve) during 1–8 January 2017 based on the regional averages over the SCB."