Response to Reviewers

Thank you for the reviewers' comments on our manuscript entitled "Impacts of urbanization on air quality and related health risks in a city with complex terrain" (egusphere-2022-486). These constructive comments are all valuable for revising and improving our manuscript. We study these comments carefully and have made correction as requested. Here are point-by-point responses (in blue color), and the changes are reflected in the revised manuscript (in red color). The line numbers in the authors' responses are obtained from the revised manuscript, in which all the revisions have been accepted.

Anonymous Referee #1:

The paper presents WRF-Chem simulations to analyze changes in $PM_{2.5}$ and O_3 concentrations due to urbanization effects in the Chinese city of Chengdu. Two months are simulated, specifically January for $PM_{2.5}$ and July for O_3, and sensitivity experiments are performed for each of the two simulations to evaluate the impact of land-use changes associated with urban expansion and the impact of anthropogenic emissions. The authors also estimate the changes in premature mortality associated with the changes in air pollution concentrations. The study is well designed to differentiate between the meteorological changes associated with urbanization caused by land-use changes and the changes in emissions and is, in my opinion, a nice example of how a modeling study can be used to disentangle different factors. The paper is also overall very well written.

I have, however, some concerns related to the lack of information about the boundary layer depth and vertical structure despite its supposedly large impact on the results (general comment 1) and the seemingly strong overestimation of $PM_{2.5}$ in the model (general comment 2).

Response: We appreciate the time and efforts you have dedicated to providing valuable comments on our manuscript. The manuscript has been modified and much improved based on those constructive comments. As for the two general comments mentioned by the reviewer, in the revised manuscript, we have added more information on the planetary boundary layer (general comment 1), and we have run new simulations on $PM_{2.5}$ (general comment 2). A point-by-point response can be found below.

General comments

1. The text refers repeatedly to the boundary layer height to explain, e.g., the temporal variations in $PM_{2.5}$ (e.g., line 287, line 308), which makes of course perfect sense. These statements are, however, rather phrased as assumptions or hypotheses and the structure and development of the boundary layer is never discussed or even evaluated to make sure that the model actually captures this crucial factor correctly. In contrast to the authors' statement (line 266), I would say that the model does not reproduce the diurnal cycle of $PM_{2.5}$ well (Fig. 5). The correlation coefficient is not that high and the model also generally overestimates the observed values during most of the time. This could potentially be connected to a poor representation of the boundary layer.

Response: We thank the reviewer for pointing out the inadequacies of our simulations. We have tried our best to revise our manuscript based on the following comments.

a. I would recommend to start by comparing vertical profiles in the model with the radiosounding data to determine how well the model actually captures the vertical structure of the boundary layer. This is also commonly done before comparing the model output to surface observations to evaluate the larger scale first. This may even be helpful in better understanding the model performance for surface wind to see whether the large-scale flow is actually represented correctly.

Response: Thanks for the constructive comment. We take your suggestion and compare vertical profiles in the model with the sounding observations in Section 3.3.2 Evaluation of model performance. Please see lines 300-304 in the revised manuscript. As shown in Figure R1, the WRF-Chem model can roughly capture the vertical structure of the tropospheric atmosphere and the large-scale flow, indicating that the simulated vertical structure of the boundary layer is reasonable.

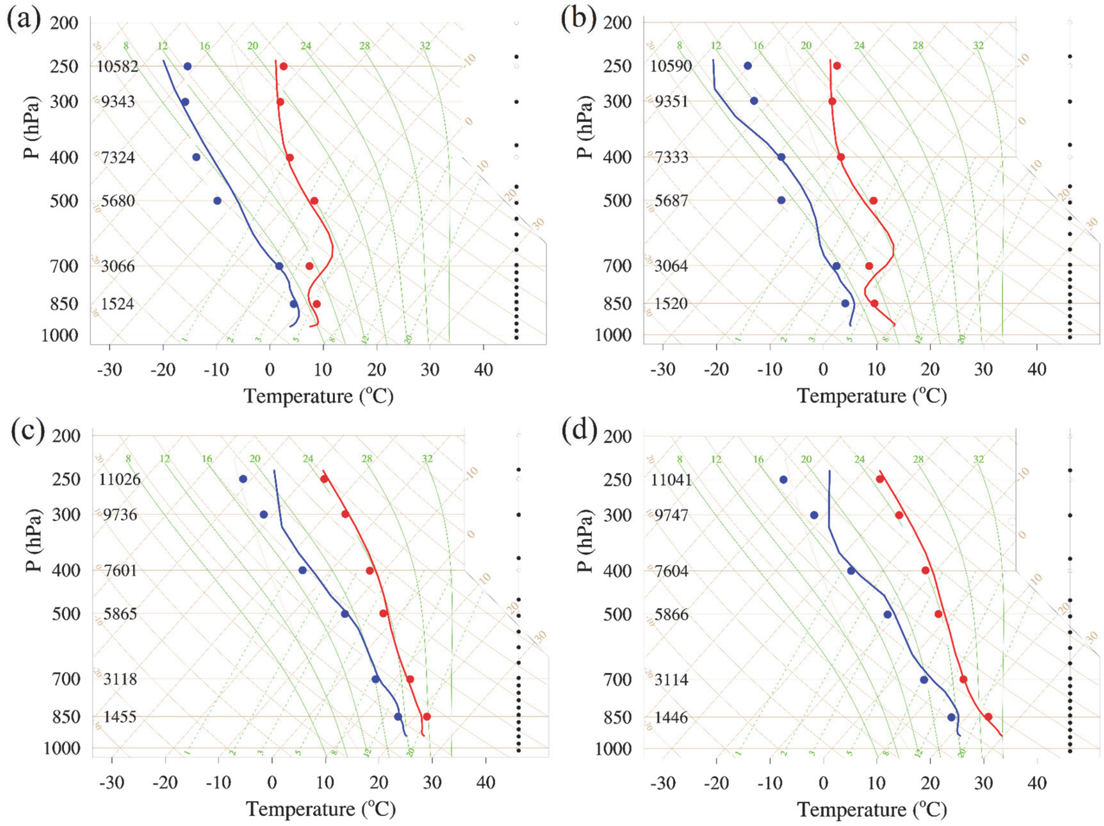

Figure R1. The skew-T diagram at (a) 00:00 UTC and (b) 12:00 UTC in January 2017. (c) and (d) are the same as (a) and (b), but for July 2017. The red and blue lines are the simulated air temperature and dew point temperature, while the red and blue points are the sounding temperature and dew point temperature. These results are monthly averages.

b. Can it be shown that the temporal development of the boundary-layer depth and vertical structure can explain the modeled development in $PM_{2.5}$ and O_3 as discussed in the paper? Similarly, the text also hypothesizes about the impact of the urban heat island (line 339: "Urban land use can enhance surface heating leading to an increase in air temperature … resulting in and increase in the boundary layer height"). Can this actually be shown in the simulations?

Response: Thanks for the constructive comment. In the revised Section 3.3.3, the temporal-vertical cross sections of boundary layer height, $PM_{2.5}$ (Figure 7a) and O_3 (Figure 8a) are given to explain their diurnal evolution. O_3 concentrations and boundary layer height are high during the day and low at night, while $PM_{2.5}$ concentrations are high at night and low during the day. Based on new Figure 7 and 8, we deeply reorganize the language in Section 3.3.3 to revisit the spatiotemporal variations in $PM_{2.5}$ and O_3. Please see lines 328-343 for $PM_{2.5}$ and lines 352-371 for O_3.

Urban land use can lead to an increase in air temperature due to increased sensible heat, and an increase in boundary layer height due to higher air temperature, which will decrease surface $PM_{2.5}$ concentrations but increase surface O_3 concentrations. This can be confirmed by our simulation results (Figure R2 and R3), and has also been reported by other scholars (e.g. Jiang et al., 2008; Li et al., 2019). However, the increases in air temperature and boundary layer height were not shown in original Figure 8 and 9 as we only gave the differences in $PM_{2.5}$ and O_3 concentrations in these two figures. In the revised manuscript, we add supporting information containing the relevant content. Please see lines 18-27 in the supporting information.

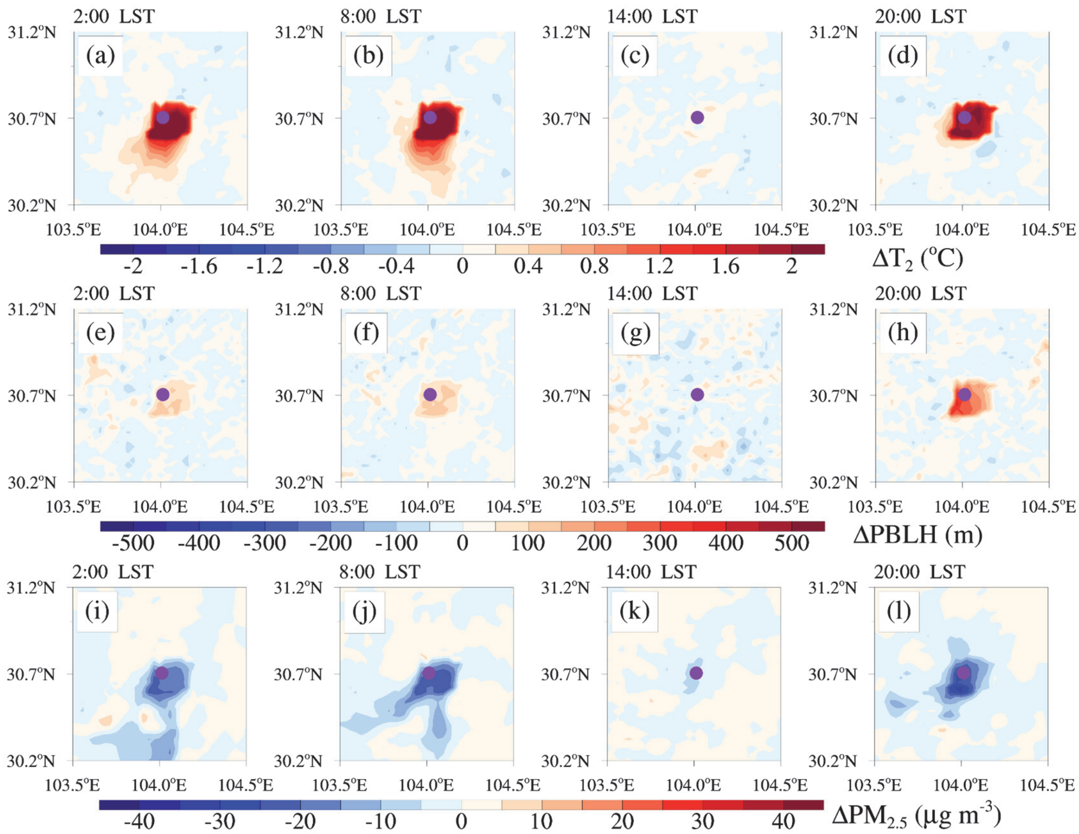

Figure R2. Horizontal distributions of differences in (a-d) 2 m air temperature, (e-h) boundary layer height and (i-j) $PM_{2.5}$ concentrations between Jan_Base and Jan_noCD (Jan_Base − Jan_noCD). The purple dots represent the locations of Chengdu.

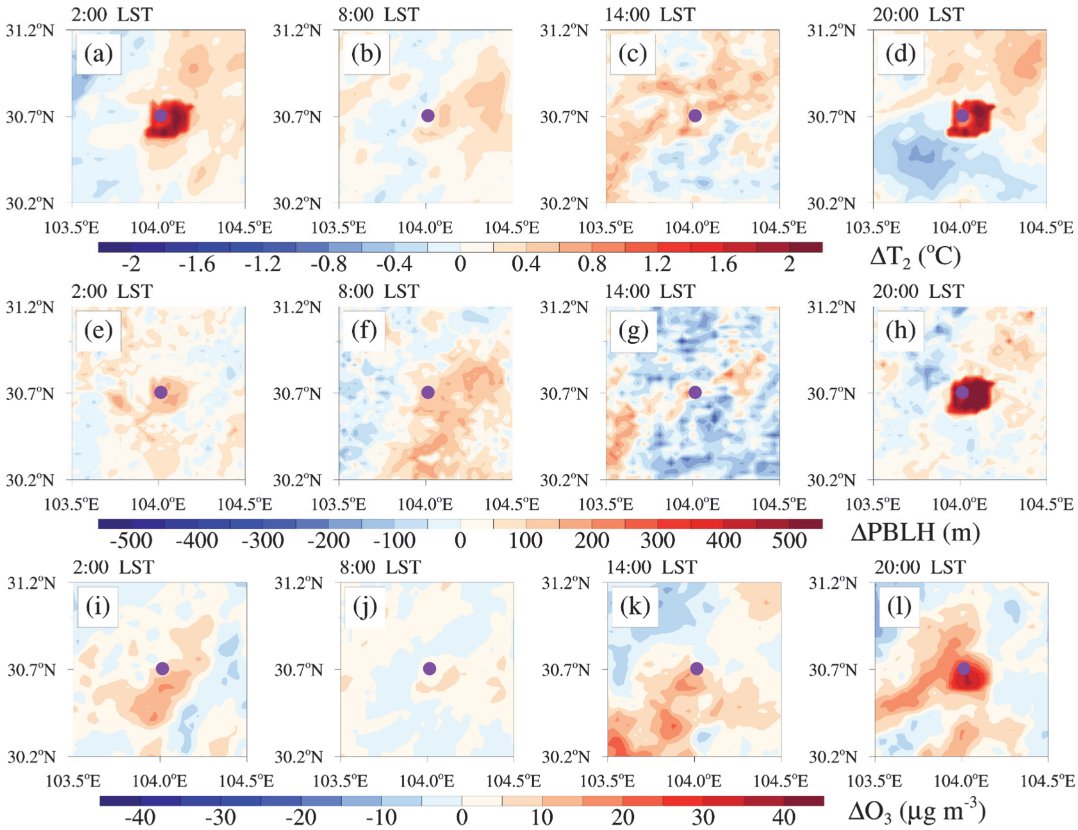

Figure R3. Horizontal distributions of differences in (a-d) 2 m air temperature, (e-h) boundary layer height and (i-j) O_3 concentrations between Jan_Base and Jan_noCD (Jan_Base − Jan_noCD). The purple dots represent the locations of Chengdu.

References

Jiang, X., Wiedinmyer, C., Chen, F., Yang, Z.-L., and Lo, J. C.-F.: Predicted impacts of climate and land use change on surface ozone in the Houston, Texas, area, Journal of Geophysical Research, 113, 10.1029/2008jd009820, 2008.

Li, Y., Zhang, J., Sailor, D. J., and Ban-Weiss, G. A.: Effects of urbanization on regional meteorology and air quality in Southern California, Atmospheric Chemistry and Physics, 19, 4439-4457, 10.5194/acp-19-4439-2019, 2019.

c. Have the authors performed any sensitivity tests with different PBL parameterizations? They can have a large impact on the boundary-layer structure and depth, which could potentially have a large impact on the results.

Response: Thanks for the constructive comment. Yes, we investigated and tested different PBL parameterizations before we performed numerical simulations. Table R1 shows the statistical metrics between simulations and observations for MYJ and YSU schemes, the two most common PBL parameterizations in WRF. Overall, the MYJ scheme performed better than the YSU scheme, especially in the overestimation of $PM_{2.5}$ concentration. In some of the literature we surveyed (Hu and Wang, 2021; Shu et al., 2021), the MYJ scheme was recommended for complex terrain conditions. Thanks to reviewer #2 for **Comment 9** and the references, we add a comparison of our model performance with previous studies to demonstrate the robustness of model results. Please see lines 309-311 and 314-318 for details. By the way, the references provided by reviewer #2 also use the MYJ scheme.

Table R1. Statistical metrics between observations and simulations.

		MYJ scheme				YSU scheme			
Variables	$\overline{O}$	$\overline{S}$	MB	RMSE	COR	$\overline{S}$	MB	RMSE	COR
$PM_{2.5}$ (μg m^{-3})	128.5	152.5	23.4	77.9	0.44	209.9	81.4	155.2	0.34
T_2 (°C)	9.0	9.2	0.2	2.0	0.76	8.7	–0.3	2.2	0.76
TD_2 (°C)	5.2	2.4	–2.8	4.1	0.44	2.0	–3.2	4.4	0.50
WS_{10} (m s^{-1})	1.3	2.7	1.3	2.0	0.36	2..4	1.1	1.8	0.32

Note. $\overline{O}$, $\overline{S}$, MB, RMSE and COR are the average of observations, the average of simulations, the mean bias, the root mean square error and the correlation coefficient, respectively. T_2, TD_2 and WS_{10} are 2-m air temperature, 2-m dew point temperature and 10-m wind speed, respectively.

References

Hu, Y. and Wang, S.: Formation mechanism of a severe air pollution event: A case study in the Sichuan Basin, Southwest China, Atmospheric Environment, 246, 10.1016/j.atmosenv.2020.118135, 2021.

Shu, Z., Liu, Y., Zhao, T., Xia, J., Wang, C., Cao, L., Wang, H., Zhang, L., Zheng, Y., Shen, L., Luo, L., and Li, Y.: Elevated 3D structures of $PM_{2.5}$ and impact of complex terrain-forcing circulations on heavy haze pollution over Sichuan Basin, China, Atmos. Chem. Phys., 21,

9253-9268, 10.5194/acp-21-9253-2021, 2021.

2. As mentioned above, the model seems to overestimate $PM_{2.5}$ relatively strongly, with a mean bias of 23.4 μg m^{-3}, which the authors consider to be small (line 264). This is almost the same magnitude as the monthly mean change related to anthropogenic emissions (26.6 μg m^{-3}), which the authors call significant (line 373), and twice the monthly mean change related to urban expansion. During individual days, the overestimation reaches even values of up to 200 μg m^{-3} (estimated from Fig. 5). I am somewhat concerned about the value of the results from the sensitivity tests considering the equally large model error and maybe that some processes affecting $PM_{2.5}$ concentrations are not simulated correctly.

Response: Thanks for the constructive comment. The quality of atmospheric chemical models depends on many factors, of which accurate meteorological, chemical and emission processes are critical. These three requirements are seldom uniformly met, so it is often unclear whether inadequacies in air quality models are due to meteorological, chemical or emissions (or all three) weaknesses. A simple common judgment is that when the normalized mean bias (NMB) is within ±15%, the simulation of pollutants is acceptable. Thanks for pointing out that our model overestimated $PM_{2.5}$ concentrations and thanks to reviewer #2 for the reminder to check emissions data. Considering that emissions are in most cases the major factor limiting the accuracy of air quality forecasts. We check the anthropogenic emissions in the Multi-resolution Emission Inventory for China (MEIC) and empirically cut the anthropogenic emissions by about 20%. The new NMB value of $PM_{2.5}$ is 9.9%, and the mean bias value of $PM_{2.5}$ drops from 23.4 μg m^{-3} to 12.7 μg m^{-3}. In the revised manuscript, all results on $PM_{2.5}$ are from new simulations.

As for the overestimation or underestimation of pollution in some periods, we believe that this may be related to meteorological weakness, especially for wind flows. In practice, it is often difficult to ensure that the meteorological conditions are correct for each time period in a long-term simulation. Since the effects of urbanization are long-term, and the health risks of pollutants are statistically significant, we focus on the entire simulation period to avoid some special situations, such as cold fronts and setting off fireworks. Furthermore, the differences in air pollutant concentrations between baseline and sensitivity simulations are used when assessing the impacts of urbanization on health risks of air pollutants. Overestimation or underestimation in some periods may not significantly change the differences in health risks over the entire time period. For example, both the original and new simulations show that urban land use could decrease premature mortalities due to $PM_{2.5}$ by about 6.9%.

3. The paper reports very precise numbers for the changes caused by urbanization, e.g., the number of premature deaths resulting from $PM_{2.5}$ (9386, 2609, 1321, and 1485 from ANAC, CVD, RD, and COPD) and the corresponding decreases of 424, 111, 55, and 56 without anthropogenic emissions. Considering that the equations given in Section 2.3 can likely provide only estimates and that I assume there must be large spatial variations in population density and air pollution throughout the city as well, which will affect the total premature deaths, I find the precision of these numbers somewhat misleading. Similarly, in the introduction a paper is cited by Liao et al. (2015) listing temperature and PM_{10} concentrations caused by urbanization with a resolution of 0.1ºC and 0.1 µg m^{-3} while providing a range of more than 1 ºC and more than 40 µg m^{-3} and no information about the magnitude of the urban expansion.

Response: Thanks for the constructive comment. We agree that it is inappropriate to report precise numbers of premature deaths due to $PM_{2.5}$ and O_3. The formulas for calculating the health risks of air pollutants are derived from epidemiology and the results are statistically significant. Premature deaths (ΔM) depend on the exposed population (Pop), the baseline mortality rate (y_0), the concentration-response function (β), the pollutant concentration (C) and the threshold concentration (C_0). Among these factors, the greatest uncertainty comes from β, which reflects the relationships between exposure and attributable deaths. Therefore, β at 95% confidence intervals (95% CI) is used in most studies. In the revised manuscript, premature deaths are presented based on β at 95% CI. The spatial distribution of population and pollutant concentrations undoubtedly affects the number of premature deaths. Some scholars use gridded population data to obtain a fine distribution of premature deaths (Li et al., 2020; Liu et al., 2018). But this is mostly the case for the population distribution in a particular year. The output of the numerical model is itself grided, but pollution monitoring stations rarely fall on a grid. Therefore, in most cases, the model results have errors due to interpolation (Please see **Comment 19a** for details.), and even increasing the resolution of models does not seem to solve this problem (Liu et al., 2020). For the above reasons, we only give the total number of premature deaths in Chengdu instead of the precise distribution of premature deaths. And the uncertainties in the assessment of premature deaths are given in the revised manuscript. Please see lines 204-209 for details. Furthermore, we use percentage change to characterize the health risks associated with urbanization. Please see lines 456-477 for details.

$$\Delta M = Pop \cdot y_0\left(1 - e^{-\beta(C-C_0)}\right)$$

As for significant digits in the citation, Liao et al. (2015) studied the impacts of urban expansion in the entire Yangtze River Delta with over 20 mega/large cities. They further analyzed the differences between daytime and nighttime, resulting in a large range of changes. Our results are averaged over the study period, and all numbers in this paper are accurate to one decimal place. Thanks for the reminder and reviewer #2's **Comment 1**. In the revised manuscript, we reorganize the language of the citation and review more studies on the impacts of urbanization in other regions. Please see lines 62-73 for details.

References

Li, Y., Zhao, X., Liao, Q., Tao, Y., and Bai, Y.: Specific differences and responses to reductions for premature mortality attributable to ambient $PM_{2.5}$ in China, Sci Total Environ, 742, 140643, https://doi.org/10.1016/j.scitotenv.2020.140643, 2020.

Liu, H., Liu, S., Xue, B., Lv, Z., Meng, Z., Yang, X., Xue, T., Yu, Q., and He, K.: Ground-level ozone pollution and its health impacts in China, Atmospheric Environment, 173, 223-230, https://doi.org/10.1016/j.atmosenv.2017.11.014, 2018.

Liu, T., Wang, C., Wang, Y., Huang, L., Li, J., Xie, F., Zhang, J., and Hu, J.: Impacts of model resolution on predictions of air quality and associated health exposure in Nanjing, China, Chemosphere, 249, 126515, https://doi.org/10.1016/j.chemosphere.2020.126515, 2020.

4. I found it rather interesting that the model simulations for both $PM_{2.5}$ and O_3 show that very high concentrations can also be observed for large distances downstream of Chengdu. Assuming that there are also large populations in the vicinity of Chengdu, the overall impact would thus be even larger. I understand that an analysis of the effects around the city would be clearly beyond the scope

of the paper, but I simply found this another interesting aspect.

Response: Thanks for the constructive comment. The phenomenon of downstream spread of air pollutants, also known as regional transport, has also been reported in many studies (Gong et al., 2020; Qiao et al., 2019). This is an important phenomenon and future research can be carried out in quantifying the contribution of Chengdu to air quality in adjacent areas, or vice versa.

Chengdu is situated on the Chengdu Plain that is widely known as “the Abundant Land” owing to its fertile soil, favorable climate and novel Dujiangyan Irrigation System. We agree that there are a large number of people in the vicinity of Chengdu, but the population density is very uneven since most of the area around Chengdu is mountainous. On the other hand, there are still many challenges in high-resolution simulation of mountainous areas. Therefore, it is difficult to obtain the precise distribution of health risks in mountainous areas, but there is no doubt that this is a topic worthy of continued effort.

References

Gong, C., Liao, H., Zhang, L., Yue, X., Dang, R., and Yang, Y.: Persistent ozone pollution episodes in North China exacerbated by regional transport, Environ Pollut, 265, 115056, 10.1016/j.envpol.2020.115056, 2020.

Qiao, X., Guo, H., Tang, Y., Wang, P., Deng, W., Zhao, X., Hu, J., Ying, Q., and Zhang, H.: Local and regional contributions to fine particulate matter in the 18 cities of Sichuan Basin, southwestern China, Atmos. Chem. Phys., 19, 5791-5803, 10.5194/acp-19-5791-2019, 2019.

Specific comments

1. Line 54: To which socioeconomic developments is the text referring?

Response: Thanks for the constructive comment. We reorganize the section of the introduction, and there is no such phrase in the revised manuscript.

2. Line 61: “the thermal contrast of the topography” is not entirely clear? Thermal circulations are caused by temperature gradients in the atmosphere.

Response: Thanks for the constructive comment. In the revised manuscript, in order to put our research into context of the present study, we review the state of the art of mountain-air pollution research. The mountain-plain wind is caused by horizontal temperature differences between air over mountain massifs and the air over the surrounding plains. Please see lines 90-93 for details.

3. Line 66: I would suggest to be more precise here and specify in which ways the diffusion conditions are more complicated.

Response: Thanks for the constructive comment. Mountain wind systems, like mountain-plain wind, can often recirculate urban air pollutants and thus worsen air quality in mountainous cities. We emphasize this in the revised manuscript. Please see lines 93-96 for details.

4. Line 89: "Chengdu has the most complex terrain in the world". This is a strong statement. How is that defined and determined?

Response: Thanks for the constructive comment. We agree that this conclusion is arbitrary. In the revised manuscript, we have deleted this sentence.

5. Fig. 1: The figure shows SO_2 emissions, but their role is never actually mentioned in the paper. There are also high-emission areas in the southeast corner of the figure. Are these other large cities?

Response: Thanks for the constructive comment. We give the distribution of SO_2 emissions to illustrate that anthropogenic emissions in Chengdu are much larger than those in its surrounding areas. The high-emission areas in the southeast corner of the figure are from another megacity, Chongqing. While the straight-line distance between Chengdu and Chongqing is over 250 km. Thanks for the reminder and reviewer #2's **Comment 3**. We replace the original Figure 1b with the distribution of observed sites in Chengdu. Please see new Figure 1 for details.

6. Air quality and meteorological data: How many stations are there in Chengdu, i.e., the ones that are averaged for the analysis (line 129)? And how strongly do the locations differ?

Response: Thanks for the constructive comment. There are one sounding, one meteorological station and eight air quality stations in Chengdu. Please see Figure 1c for details. Since the air quality stations are mainly concentrated in the city center, we use the average of air pollutant concentrations at all stations to represent air pollutant concentrations in Chengdu. In the revised Section 2.1 Air quality and meteorological data, we briefly describe these stations and the data processing. Please see lines 135-139 for details.

7. Line 137: At which height is the lowest model level? Is the boundary layer also properly resolved during the night?

Response: Thanks for the constructive comment. There are 32 vertical layers below the model top of 100 hPa and 12 vertical layers below 2 km to resolve the boundary layer processes. The size of the lowest vertical grid is ~25 m. We add this information in the revised manuscript. Please see lines 152-154 for details. Due to the lack of observations of the boundary layer, we are not entirely sure that the simulated boundary layer height is proper. However, in terms of the vertical distribution of soundings (Figure 4c, 4d, 5c and 5d) and pollutants (Figure 7a and 8a), the evolution of the simulated boundary layer is reasonable.

8. Line 175: The text says that β has units of ($\mu g\ m^{-3})^{-1}$, which makes sense to have a non-dimensional exponent in eq. 5, but Table 3 mentions units of %.

Response: Thanks for the constructive comment. β is the concentration-response function (CRF), which relates a unit change in air pollutant concentrations to a change in health endpoint incidence. It is identified by short- or long-term epidemiology studies. It is a ratio, and usually the estimated slope of the log-linear relation between concentration and mortality. In the revised manuscript, we assign a more precise meaning to β. Please see lines 191-194 for details.

9. Line 180 and Table 3: The references in the text and in the figure caption are not the same.

Response: Thanks for the constructive comment. β for $PM_{2.5}$ and MDA8 O_3 were from Chen et al. (2017) and Yin et al. (2017), respectively. β and y_0 included in Table 3 were summarized by Wang et al. (2021). In the revised manuscript, we unify the references in these two places. Please see lines 198-199 for details.

10. Lines 192 and 439: How are "pollution episodes" defined? Are these continuous periods, during

which the air quality standard is exceeded, i.e., they can have different lengths? Does "pollution" always mean exceeding the standards (e.g., line 232) in this paper?

Response: Thanks for the constructive comment. In China, the national ambient air quality standard for $PM_{2.5}$ is that daily average $PM_{2.5}$ concentrations cannot exceed 75 μg m^{-3}; the national ambient air quality standard for O_3 is that daily maximum 8-h average (MDA8) O_3 concentrations cannot exceed 160 μg m^{-3}. They have different lengths. Thus, Figure 2a shows the distribution of daily average $PM_{2.5}$ concentrations while Figure 2b shows the distribution of MDA8 O_3 concentrations. In the revised manuscript, we add a note on the definition of "pollution episodes". Please see lines 215-217 for details.

11. Line 213: What does "administrative division adjustment" mean?

Response: Thanks for the constructive comment. In May 2016, Jianyang urban district was placed under the jurisdiction of Chengdu, and thereby the population of Chengdu in 2016 increased by more than 1 million compared with 2015. Chengdu's population has grown rapidly both because of the influx of people and the consolidation of surrounding areas, which will further affect the premature deaths attributable to air pollutants. However, since our study is for Chengdu as a whole, this information seems to be redundant and has been removed from the manuscript.

12. Line 222: I am not sure how meaningful it is to calculate and discuss an increase or decrease between the first and last year if there is no real trend and the inter-annual variability is larger than the overall trend.

Response: Thanks for the constructive comment. O_3 concentrations monitored by the National Environmental Monitoring Center of China were reported in the unit of μg m^{-3} under standard atmospheric conditions (273.15 K, 1 atm) before September 2018 and changed to 298.15 K conditions afterward. After revising this, we find that although the population of Chengdu was increasing from 2015 to 2021, the premature deaths due to $PM_{2.5}$ decreased while the premature deaths due to O_3 increased slightly. Please see lines 244-246 for $PM_{2.5}$ and lines 250-253 for O_3.

13. Table 4: Are these numbers calculated for individual days and then summed over the whole year?

Section 2.3 does not say for which time period the given equations are applicable. It is mentioned that equation 5 is applied to MDA8 O_3, but it does not say either over which time period the $PM_{2.5}$ values have to be averaged.

Response: Thanks for the constructive comment. Yes, the numbers of premature death are first calculated for individual days and then are summed over the whole year/month. The MDA8 O_3 concentrations and daily average $PM_{2.5}$ concentrations are used to calculate premature mortalities attributable to O_3 and $PM_{2.5}$, respectively. Because the standard and β for $PM_{2.5}$ are based on the daily average concentrations of $PM_{2.5}$ while those for O_3 are based on MDA8 O_3 concentrations. In the revised Section 2.3, we add the details of the calculations. Please see lines 202-204 for details.

14. Line 235: "cold westerly winds from the north" seems to be a contradiction. Are the winds from the west or from the north?

Response: Thanks for the constructive comment. We are sorry for this confusing sentence. The winds are from the west. In the revised manuscript, we have clarified this point. Please see lines 265-266 for details.

15. Line 239: Why do the humidity differences cause the inversion? Actually, it seems that the temperature decreases with height, i.e., there is no inversion, but only a stable layer.

Response: Thanks for the constructive comment. When Chengdu is controlled by the southerly warm air flow (purple arrow in Figure R4b), the inversion tends to appear around 700 hPa (purple rectangles in Figure R4c and d) under the effect of warm advection. Since the southerly airflow originating from the Bay of Bengal is rich in moisture, the humidity below the inversion is much higher than that above the inversion (The smaller the difference between the temperature and the dew point temperature, the higher the relative humidity.). Unlike January, there is no inversion in July (Figure 5c and 5d in the revised manuscript). In the revised manuscript, we analyze the meteorological conditions separately for January and July so that readers can distinguish the difference between the two backgrounds. Please see lines 261-290 for details.

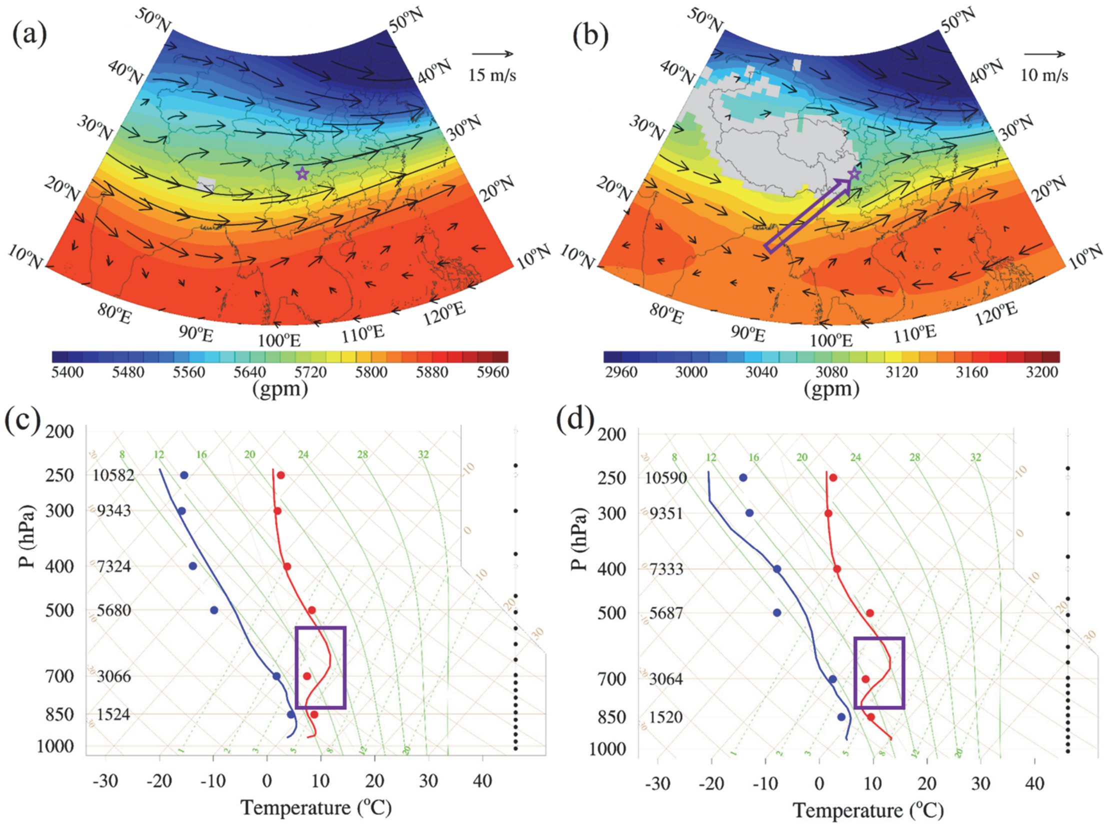

Figure R4. The weather charts at (a) 500 hPa and (b) 700 hPa for January 2017 based on the NCEP FNL reanalysis data. The purple pentacles show the location of Chengdu. The skew-T diagram at (c) 00:00 UTC and (d) 12:00 UTC in January 2017. The red and blue lines are the simulated air temperature and dew point temperature in Jan_Base simulation, while the red and blue points are the sounding temperature and dew point temperature. These results are monthly averages.

16. Fig. 3: The 850-hPa level should be lower than the Tibetan Plateau. So what do the plotted fields over the Tibetan Plateau actually show? The areas above 850 hPa should best be masked in the plots. Also, are these monthly averages?

Response: Thanks for the constructive comment. For various purposes, such as driving numerical models, reanalysis data are often interpolated to different pressure layers. We agree that the 850-hPa layer is lower than the Tibetan Plateau. The areas above the selected pressure layer are masked in the revised Figure 4 and 5. Please see line 273 for Figure 4 and line 292 for Figure 5. The weather charts and the temperature profiles for January and July are monthly averages. We have added a note about this in the captions to Figure 4 and 5.

17. Fig. 4: (i) What do the black dots to the right of the subfigures (where one usually finds the wind arrows) mean? (ii) Are the profiles from the radiosoundings or also from the NCEP analysis as Fig. 3? Are they monthly means? (iii) Why are both 00 and 12 UTC shown if the differences are never discussed?

Response: Thanks for the constructive comment. (i) The black dots to the right of the skew-T plots usually represent the locations of the wind barbs in the base map of the skew-T plot (https://www.ncl.ucar.edu/Applications/skewt.shtml#ex2). However, the wind barbs are missing in this study because the data on the winds are incomplete and we are mainly concerned with the temperature profiles. (ii) The profiles are from the radiosoundings instead of the NCEP FNL analysis data. And they are monthly averages as Figure 4a and 4b. Thanks for the reminder. We have added a note about this in the captions of Figure 4 and 5. Please see lines 278-279 for Figure 4 and lines 296-297 for Figure 5. (iii) The radiosounding data are available at 00:00 and 12:00 UTC, corresponding to 08:00 and 20:00 LST (LST is UTC+8h) in Chengdu, which can show the diurnal difference in the vertical distribution of the atmosphere. The good agreement between the vertical profiles in the model and the sounding data indicates that our model can capture the diurnal variation of the boundary layer.

18. Line 269: A comparison of RH may not actually say much about the model performance in simulating humidity correctly because it combines the effects of humidity and temperature. It might be better to use, e.g., mixing ratio or specific humidity if the observations are available to convert the units.

Response: Thanks for the constructive comment. We often use relative humidity in weather reports out of habit. But we agree that relative humidity is not an accurate indicator of atmospheric water vapor because it largely depends on temperature. In the revised manuscript, we use dew point instead of relative humidity (the dew point is included in the surface meteorological data). Please see lines 312-314 for details. The dew point is the temperature to which air must be cooled to become saturated with water vapor. It provides a measure of the actual amount of water vapor in the air–a higher dew point means there is more moisture in the air.

19. Fig. 5

a. Are the measurements averages over all stations in the city as suggested in the previous section? If so is the model output equally averaged over all urban grid cells or over all grid cells closest to the stations?

Response: Thanks for the constructive comment. There are one sounding, one meteorological station and eight air quality stations in Chengdu (Figure 1c). Therefore, for the sounding and meteorological variables, the simulated values are the average of all grid cells closest to the station. We use the same method to first calculate the simulated $PM_{2.5}/O_3$ concentrations of the eight stations, and then average the simulated values as $PM_{2.5}/O_3$ concentrations for the entire city. We do not distinguish between urban and rural stations since these stations are basically located in the city center. We acknowledge that this approximation will affect the results of premature deaths because pollutant concentrations and population densities vary in different parts of the same city. We give more clarification in the revised Section 2 Data and methods, including the handling of data and the uncertainty about premature deaths. Please see lines 135-139 and lines 202-209 for details.

b. The figure and figure caption do not say which of the shown datasets (black dots or colored lines) is the model and which the observations. Based on a statement in the text that the model underestimates RH, I assume that the observations are the dots.

Response: Thanks for the constructive comment. Yes, the black dots are the observations and the colored lines are the model results. In the revised Figure 6, we add this in the figure caption. Please see lines 323-325 for details.

c. Wind speed, RH, and temperature seem to have a resolution of only 1 m s^{-1}, 5 or 10%, and 1ºC, respectively, which makes it really difficult to compare the diurnal cycles because, e.g., wind speed remains basically constant for hours. This will also affect the quantitative comparison (correlation coefficient). Is this the actual resolution of the data?

Response: Thanks for the constructive comment. We recheck the observation data as well as the code for processing these data. Temperature, dew point temperature, wind speed and $PM_{2.5}/O_3$ concentration in the observation data are measured hourly with resolutions of 1 ℃, 1 ℃, 1 m s^{-1} and 1 μg m^{-3}. Therefore, meteorological variables may have the same value for several hours, especially for wind speed since the wind speed is small. This may reduce the correlation coefficient between observations and simulations. Nevertheless, the correlation coefficients of all variables can still exceed the 99% significance level, indicating that our simulations are generally reasonable.

d. Why is wind direction not shown?

Response: Thanks for the constructive comment. In the revised Figure 6, we replace the wind speed in the original Figure 5 with the wind barbs to illustrate the wind direction. Northerly winds prevail in Chengdu in both January and July. Please see line 323 for details.

20. Line 292: "mountain-valley breezes" – Where can they be seen in the figure? Chengdu seems to be located in a northeasterly flow directed towards the mountains all day and night. How does that agree with diurnal circulations?

Response: Thanks for the constructive comment. After repeated discussions with the co-authors, we agree that this study is a classic example of regional mountain-plain circulation rather than local mountain-valley circulation. During daytime, the plain-to-mountain wind (plain wind) characterized by easterly and upslope flows (Figure R5c and d) draws $PM_{2.5}$-rich air mass from the polluted area to the eastern slope of the Tibetan Plateau. During nighttime, the circulation reverses, the mountain-to-plain wind (mountain wind) enhances $PM_{2.5}$ concentration over Chengdu (Figure R5a and b). Moreover, under the influence of the prevailing northeasterly wind (Figure 6), high-concentration $PM_{2.5}$ can be transported to downstream areas. These rules also apply to O_3 (Figure 8), which is the climatology of mountain areas. In the revised manuscript, we replace the mountain-valley breeze with the mountain-plain wind, and we explore the impact of mountain-plain wind on $PM_{2.5}$ and O_3 pollution in Chengdu. Please see lines 335-343 for $PM_{2.5}$ and lines 360-367 for O_3.

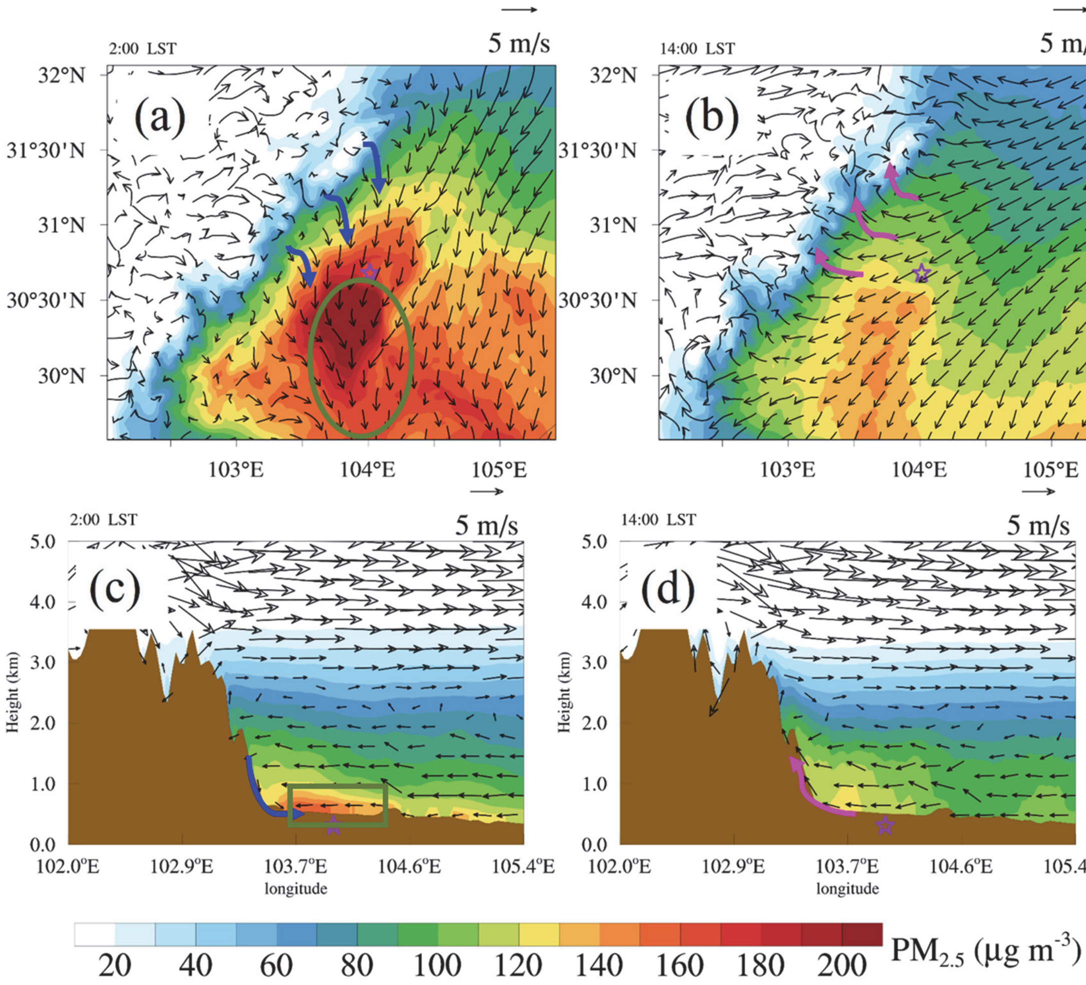

Figure R5. Horizontal distributions of $PM_{2.5}$ with wind vectors at the lowest model level at (a) 2:00 and (b) 14:00 LST. East-west vertical cross sections of $PM_{2.5}$ with wind vectors at (c) 2:00 and (d) 14:00 LST. Purple pentacles show the location of Chengdu. Brown-shaded areas represent the terrain. These results are the monthly average based on Jan_Base simulation. The blue and magenta arrows highlight the mountain wind and plain wind, respectively.

21. Line 294: It is very difficult to see in the horizontal cross sections whether there are actually converging flows, because the figures show the flow at the first model level (I assume so because the figure caption does not say), which means very large elevation differences. The shown wind field is about 2-3 km higher in the leftmost part of the figure than in the right part. Based on the figures, one could also argue that the high concentrations are simply located at the foot of the mountains, i.e., they are trapped by the terrain.

Response: Thanks for the constructive comment. Yes, horizontal distributions of $PM_{2.5}$, O_3 and wind are the results at the lowest model level. We have added this to the figure caption in the revised manuscript. Please see lines 347-348 for $PM_{2.5}$ and lines 375-376 for O_3. The position of the convergent flows is shown by the green oval in Figure R5a, where the terrain is relatively flat (the green rectangle in Figure R5c). Driven by northeasterly wind and mountain wind, surface $PM_{2.5}$ converges to the west boundary of the Chengdu Plain and then will be transported to the eastern Tibetan Plateau edge and the northern Yunan-Guizhou Plateau edge areas, where it will be trapped by the terrain.

22. Line 298: Is this really a secondary circulation that is forced by the terrain? Could it not be the upper-level westerly flow that is shown in Fig. 3? This would also agree with the absence of this "secondary circulation" in July, when a weak-gradient situation prevails at upper levels.

Response: Thanks for the constructive comment. We are sorry for not giving a clear definition of the secondary circulation. The secondary circulation here refers to the mountain-plain wind (blue and magenta arrows in Figure R5c and d) rather than upper-level westerly flow. The daytime heating and nighttime cooling contrast between the mountain atmosphere over the outer slopes of the mountain massif and the free atmosphere over the surrounding plain produces the horizontal pressure differences that drive this wind system. In July, under the influence of the high-pressure systems, the horizontal pressure gradient is small so the mountain-plain wind is not as pronounced as in January. Nevertheless, it is still distinguishable (Figure 8). In the revised manuscript, we use the mountain and plain wind directly instead of vague words, i.e., the secondary circulation. Please see lines 335-343 for $PM_{2.5}$ and lines 360-367 for O_3.

Technical comments, typos, etc.

1. Several of the figure axis labels are very small and thus almost impossible to read, e.g., axes labels in Fig. 2 (in particular the days of the week, but also the months), Fig. 4., Figs. 6-11.

Response: Thanks for the constructive comment. In the revised manuscript, we have enhanced the quality of these figures with higher resolution and larger font size. Furthermore, we have redrawn other figures in this paper to make them clear.

2. For past events, the past tense should be used: e.g., "increased by 24.2%" (line 214). Similarly, lines 217, 232 and 438. There may be others.

Response: Thanks for the constructive comment. We carefully check the tense in this paper, using the past tense for past events and the present tense for present events.

3. Lines 25 and 30: It would be good to add that the mentioned changes in $PM_{2.5}$ concentrations are based on monthly means.

Response: Thanks for the constructive comment. In the revised manuscript, we add that changes in $PM_{2.5}$ and O_3 are based on monthly means. Please see lines 28-31 and lines 33-35 for details.

4. Line 26: MDA8 is not defined

Response: Thanks for the constructive comment. We add the definition of MDA8 to the revised abstract. Please see line 29 for details.

5. Line 55: "increases in anthropogenic emissions"

Response: Thanks for the constructive comment. We have unified the singular and plural forms here as well as in the rest of this paper.

6. Line 60: The reference list contains only Qian et al. (2022).

Response: Thanks for the constructive comment. We correct this typo to Qian et al., 2022 in the revised manuscript. Please see line 75 for details.

7. Line 67: "notable pollution episodes"

Response: Thanks for the constructive comment. We reorganize the section of the introduction, and there is no such phrase in the revised manuscript.

8. Line 67: Chow et al. (2013) is not included in the reference list.

Response: Thanks for the constructive comment. We recheck and update all references in this paper, including the addition of the book titled Mountain weather research and forecasting edited by Chow et al. (2013). Please see line 90 and lines 749-751 for details.

9. Line 71: I don't understand the meaning of the sentence "Although the principles …"

Response: Thanks for the constructive comment. We reorganize the section of the introduction, and there is no such sentence in the revised manuscript.

10. Line 91: It would be really helpful to label these terrain features in Fig. 1a?

Response: Thanks for the constructive comment. We take your suggestion and mark the names of these mountains in the new Figure 1a. Please see lines 122-125 for details.

11. Fig. 1: It would be helpful to indicate the location of Chengdu in subfigure (b). I assume it corresponds to the red location in the center with high emission values?

Response: Thanks for the constructive comment. We remove the original Figure 1b since the role of SO_2 emission is never actually mentioned in the paper. Instead, we give the boundary of Chengdu and the distribution of monitoring sites in Chengdu. We mark the location of Chengdu in Figure 1a and 1b. Please see lines 122-125 for details.

12. Line 115: "Data and methods" might be a more common section title.

Response: Thanks for the constructive comment. The title of Section 2 has been changed from "Materials and methods" to "Data and methods". Please see line 127 for details.

13. Line 200: "The high temperature and strong sunlight contribute to the elevated …"

Response: Thanks for the constructive comment. In the new manuscript, the sentence "Elevated O_3 concentrations in warm months are contributed to the high temperature and strong sunlight." is revised to "The high temperature and strong sunlight contribute to the elevated O_3 concentrations in warm months". Please see lines 229-230 for details.

14. Line 189: "two crucial air pollutants that account for air pollution". This is a somewhat awkward sentence. "that account for air pollution" can easily be removed.

Response: Thanks for the constructive comment. In the new manuscript, the awkward sentence "$PM_{2.5}$ and O_3 are two crucial air pollutants that account for air pollution." has been removed.

15. Line 237: "to the west of Chengdu"

Response: Thanks for this comment and **Specific comments 16**. Since the 850-hPa level is much lower than the Tibetan Plateau, the Southwest Vortex that appears to the west of Chengdu is masked out on the weather charts. Thus, the corresponding sentence does not exist in the revised manuscript.

16. Line 240: "cold air"

Response: Thanks for the constructive comment. The phrase "cold air" is revised to "blocking of air", and the entire sentence is "The blocking of air and the temperature inversion were two important reasons for frequent $PM_{2.5}$ pollution episodes during this period.". Please see lines 269-271 for details.

17. Line 314: "and mixing" (remove "well")

Response: Thanks for the constructive comment. The word "well" has been removed in the revised manuscript. Please see lines 359-360 for details.

18. Line 360: "can increase" suggests that this is a maximum value. Or is the text also referring to a monthly average as for $PM_{2.5}$? It would actually help to always add "monthly mean" when talking about monthly mean values (e.g., line 404) to avoid misunderstandings.

Response: Thanks for the constructive comment. In the revised manuscript, we take your suggestion and always add "monthly mean" when we talk about monthly mean values, including in the abstract, text and figure captions.

19. Fig. 10: Is there a reason why the color scale is not adjusted to show the whole range? Based on the color bar it looks like the highest values are around 10 $\mu g\ m^{-3}$, whereas the text actually mentions a monthly average of 26.6 $\mu g\ m^{-3}$.

Response: Thanks for the constructive comment. Figures 9-12 use the same color bar to visualize the different effects of urban land use and anthropogenic emission on $PM_{2.5}$ and O_3. In the revised manuscript, we adjust the color scale in Figures 9-12 so that they can show the whole range of pollutant concentrations. Please see lines 395, 416, 433 and 451 for Figure 9, 10, 11 and 12, respectively.

20. Line 407: "assess" instead of "access"

Response: Thanks for the constructive comment. In the revised manuscript, we have corrected this typo. Please see line 461 for details.

21. Line 545: This reference is not cited in the manuscript.

Response: Thanks for this comment and **Technical comment 8**. This reference is incorrectly cited in the text as Chow et al. (2013) because the book was edited by Prof. Chow. We have corrected this typo in both the text and references. Please see line 90 and lines 749-751 for details.

Anonymous Referee #2:

General comments

Accelerated urbanization across the globe has resulted in considerable changes in land surface parameters and subsequently affect meteorological conditions and air pollutant levels. In this study, Zhan et al. reveal the changes in criteria air pollutants between 2015 and 2021 and probe the environmental consequence of rapid urbanization over a typical megacity situated in southwestern China, Chengdu. They also quantify the premature mortalities attributed to exposure to ozone and $PM_{2.5}$. This paper is well-written and presents results that would be interesting to the air quality modeling community from a practical perspective. I have several concerns that the authors should consider when revising the manuscript, as listed below. I recommend publication after the following comments are adequately addressed.

Response: We would like to thank the referee for the valuable and affirmative comments on our manuscript. We have carefully revised our manuscript based on the following comments.

Specific comments

1. The literature review could be better. The authors only provide an example of urbanization impacts focused on the YRD by Liao et al. (2015), while extensive studies have been focusing on identifying the effects of urbanization on the regional meteorological phenomena and air quality (including the Beijing-Tianjin-Hebei area and the Pearl River Delta). Furthermore, recent studies have widely acknowledged the critical role of urbanization in altering air quality in Chengdu [Wang et al., 2021, 2022a]. Thus, these studies should be discussed and properly cited.

Response: Thanks for the constructive comment. We take your suggestion and deeply reorganize the introduction, in which we add a review of research on the effects of urbanization on regional meteorology and air quality in other regions, such as the Beijing-Tianjin-Hebei region, the Pearl River Delta region and the Sichuan Basin. Please see lines 62-73 in the revised manuscript for details. Thanks for the recommended literatures. We have learned a lot from these literatures, all the recommended literatures are cited in the revised manuscript.

2. Line 34-36: This sentence is a bit vague and I genuinely don't understand this sentence - please rephrase.

Response: Thanks for the constructive comment. The original sentence "This reminds us that the development of cities is also important for the urban air quality apart from the emissions reduction" is revised as "This reminds us that, in addition to regulating anthropogenic emissions, urban planning is also important for the urban air quality, especially for secondary pollutions like O_3." in the new manuscript. Please see lines 40-41 for details.

3. Figure 1: It seems that the shapefile used by the authors (NCL default shapefile) is wrong. Please check. Also, please clarify the source of SO_2 emission for subplot (b). Moreover, it would be valuable to provide the boundary of Chengdu city in Figure 1 for making it clear.

Response: Thanks for the constructive comment. We have revised the NCL default shapefile based on the map database provided by Dr. Yongjie Huang (https://github.com/huangynj/NCL-Chinamap). Thanks for the reminder and reviewer #1's **specific comment 5**. We remove the original Figure 1b since the role of SO_2 emission is never actually mentioned in the paper. Instead, we give the boundary of Chengdu and the distribution of monitoring sites in Chengdu. Please see the new Figure 1 in line 123.

4. Line 190: The criterion for $PM_{2.5}$ and MDA8 ozone is a bit vague. I believe that it is annual $PM_{2.5}$ concentrations less than 75μg/m^3 and MDA8 ozone less than 160μg/m^3. Please clarify the time period for these metrics.

Response: Thanks for the constructive comment. In China, the national ambient air quality standard for $PM_{2.5}$ is that daily average $PM_{2.5}$ concentrations cannot exceed 75 μg m^{-3}; the national ambient air quality standard for O_3 is that MDA8 O_3 concentrations cannot exceed 160 μg m^{-3}. The definition of $PM_{2.5}$/O_3 pollution is based on Chinese national standards. We have clarified these metrics in the revised manuscript. Please see lines 215-217 for details.

5. Line 193-196: The authors apply the annual mean MDA8 ozone concentrations for illustrating the variations of ozone across Chengdu over time. However, annual mean MDA8 ozone is not a meaningful metric as wintertime low MDA8 ozone would pull low ozone levels. In general, it is recommended for using the warm season (April-September) MDA8 ozone average (see Wang et al., (2022b)) or the 90th percentile of MDA8 ozone (which is based on Chinese NAAQS GB3095-2012).

Response: Thanks for the constructive comment. We agree that the annual mean MDA8 O_3 concentration is not a meaningful metric for the interannual variation of O_3. Since the annual metric for O_3 in China is the 90th percentile of MDA8 O_3 (GB3095-2012; HJ663-2013), we adopt the 90th percentile of MDA8 O_3 concentrations in the revised manuscript. Please see lines 221-225 for details.

6. It would be better to use "heat maps of (a) daily average $PM_{2.5}$ and (b) MDA8 O_3 concentrations" rather than "distribution" for the caption of Figure 2.

Response: Thanks for the constructive comment. The new caption of Figure 2 has been revised to "Figure 2. Heat map of (a) daily $PM_{2.5}$ and (b) MDA8 O_3 concentrations in Chengdu from 2015 to 2021." Please see lines 233-234 for details.

7. Line 219-220: "the premature mortalities due to O_3 fluctuate." This is an incomplete sentence.

Please check.

Response: Thanks for the constructive comment. In the revised manuscript, this incomplete sentence is removed. Instead, we directly give the number of premature deaths attributable to O_3. Please see lines 247-250 for details.

8. Line 221: “annual average” might be “7-year average”. Please check.

Response: Thanks for the constructive comment. Yes, it is “7-year annual average” instead of “annual average”. In the revised manuscript, we have corrected this typo. Please see lines 22, 240-243, 250 and 492-493 for details.

9. Line 275-278: Is the WRF-Chem model performance comparable with prior studies over Chengdu (or Sichuan Basin)? It would be valuable to briefly compare the model performance with previous studies (Yang et al., 2021; Wu et al., 2022) for demonstrating the robustness of model results.

Response: Thanks for the constructive comment. In the previous version, we do not compare the WRF-Chem model performance with prior studies over Chengdu. Thanks for the recommended literatures. We add a brief comparison of our model results with those from prior studies in the revised manuscript, which further suggests that our simulations are reasonable. Please see lines 305-311 and lines 314-318 for details.

10. Line 369-370: This sentence is a bit vague and I genuinely don’t understand this sentence - please rephrase.

Response: Thanks for the constructive comment. We apologize for this confusing sentence here. In the revised manuscript, a new sentence “Rising anthropogenic emissions of air pollutants and their precursors can significantly increase ambient air pollution.” is adopted. Please see lines 423-425 for details.

11. Line 385-395: The authors attribute the ozone changes in Chengdu to the Ozone-NOx-VOCs regime but do not provide any details about the formation regime. A comprehensive discussion on

the underlying mechanism of the VOCs-limited ozone regime in urban Chengdu is needed (Wang et al., 2022a).

Response: Thanks for the constructive comment. Yes, we attribute only a slight increase in O_3 concentrations from anthropogenic emissions to the non-linear sensitivity of O_3 and its precursor (VOCs and NO_x). Thanks to your literature and references therein, in the revised manuscript, we add a discussion on O_3 formation regime in Chengdu. From 2013 to 2020, metropolitan Chengdu remains VOCs-limited regime, and the effect of reducing NO_x emissions may be partially offset by changes in VOCs. Please see lines 440-446 for details.

References

[1] Wang, H., et al. (2021). Impact of different urban canopy models on air quality simulation in Chengdu, southwestern China. Atmospheric Environment, 267, 118775. https://doi.org/10.1016/j.atmosenv.2021.118775

[2] Wang, H., et al. (2022a). Impact of Urbanization on Meteorology and Air Quality in Chengdu, a Basin City of Southwestern China. Frontiers in Ecology and Evolution, 10, 845801. https://doi.org/10.3389/fevo.2022.845801

[3] Wang, Y., et al. (2022b). Long-term trends of ozone and precursors from 2013 to 2020 in a megacity (Chengdu), China: Evidence of changing emissions and chemistry. Atmospheric Research, 106309. https://doi.org/10.1016/j.atmosres.2022.106309

[4] Wu, K., et al. (2022). Drivers of 2013–2020 ozone trends in the Sichuan Basin, China: Impacts of meteorology and precursor emission changes. Environmental Pollution, 300, 118914. https://doi.org/10.1016/j.envpol.2022.118914

Thanks for the recommended literatures. We have learned a lot from these literatures, all the recommended literatures are cited in the revised manuscript. Please see lines 719-721, 722-724, 729-732 and 735-738 of the references.