June 20, 2020 Dear Editor,

Thanks for your efforts on this manuscript. We have received comments from the reviewers of our manuscript, and we would like to thank the reviewers for their careful reading and their insightful comments. To address the reviewers' comments, we have revised the manuscript, and the revised text is highlighted in red.

Best Regards, Xuexi Tie

Reply to Anonymous Referee #1

Thanks for the reviewer's helpful comments. We have given our point-to-point response to your comments and suggestions in the revised manuscript. To carefully address the comments of the reviewer, we add more content and figures. We would like to think that the revised manuscript is greatly improved after addressing the reviewer's comments.

General comments:

In this manuscript, the authors focus on the effect of warming Tibetan Plateau on air quality in the Sichuan Basin, China. Specifically, they address the 2°C warming causes an increase in the PBL height and a decrease in the relative humidity in the basin. The elevated PBL height strengthens vertical diffusion of PM2.5, while the decreased RH significantly reduces secondary aerosol formation. The authors highlight that the recent warming plateau has improved air quality in the basin. The results of this work are based on the WRF-Chem simulations and extensive observation. The analysis is mostly sound, the manuscript is well written, but some details need clarify. I recommend a minor revision with my comments listed below.

Specific comments:

Comment 1. In line151, please further explain what does "'top-down' method" means here and how to use the 'top-down' method to constrain the emission inventory via comparing the simulations with the measurements?

Response: The 'top-down' method is to compare the simulated value with the observed value time and again until the simulated values, including the averaged level and the trend, are close to the observed ones. Generally, we use mean bias (MB), root mean square error (RMSE), and index of agreement (IOA) to evaluate the model performance. The higher the IOA, the closer the simulated value is to the observed value. In this study, the statistical indices of agreement (IOAs) of pollutants (O₃, CO and PM_{2.5}) are greater than 0.7.

The 'bottom-up' emission inventory used in this study is constructed by national and provincial emission factors and activity data based on a statistical approach, so it is difficult to obtain accurate activity data. Also, the emission factors representative at a local level is difficult to

measured. Therefore, the spatial pattern of the inventory at a local level needs to be improved. In addition, the 'bottom-up' emission inventory is not updated every year. In practice, the 'bottom-up' emission inventory is used to drive the model, and the 'top-down' method is used to constrain the emission. Top-down constraints on emissions is helpful to improve the accuracy of the 'bottom-up' emission inventory. The detailed introduction of these two approaches are referred to Zhang et al. (2009) and Fu et al. (2012).

In the revised version, we have added a brief introduction to the 'top-down' method, and the text is "*The emission inventory is constructed by a 'bottom-up' approach based on national and provincial activity data and emission factors. To improve the emission inventory accuracy, we use a 'top-down' method here to constrain the emission inventory. We compare the simulated value with the measured value time and again until the simulations are close to the measurements.*" In lines 151 - 155.

- Fu, T. M., Cao, J. J., Zhang, X. Y., Lee, S. C., Zhang, Q., Han, Y. M., et al. (2012). Carbonaceous aerosols in China: top-down constraints on primary sources and estimation of secondary contribution. *Atmospheric Chemistry and Physics*, 12(5), 2725–2746. http://doi.org/10.5194/acp-12-2725-2012
- Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., et al. (2009). Asian emissions in 2006 for the NASA INTEX-B mission. *Atmospheric Chemistry and Physics*, 9(14), 5131–5153. http://doi.org/10.5194/acp-9-5131-2009

Comment 2. In line 164-165, in the configuration of the sensitivity simulation, how to set the temperature increment to 2K? Is it just increase the temperature in all levels and all grids of the model above Tibetan Plateau (TP)? Does the 2K increment set at the beginning of model simulation or need nudging in every step of the simulation? Are the temperature increment same in verticals or just at the surface?

Response: We have given a detailed description for the 2K sensitivity simulation in the revised version. According to the ERA-interim reanalysis data, the warming is only happening in the troposphere (600 hPa - 250 hPa). As a result, in the sensitivity simulation, we set the 2K increment in the troposphere (600 hPa - 250 hPa) over the Tibetan Plateau. In order to ensure a persistent influence of the 2K increment, we add the 2K increment at the initial and boundary conditions of the model, and also drive the initial condition with a 2K increment every day.

These texts are added in the revised manuscript. "According to the meteorological records at weather stations, surface air temperature risen by an average of 2°C from 2013 to 2017 over the Tibetan Plateau (Table S1). ERA-interim reanalysis data also show that the troposphere (600hPa - 250hPa) over the plateau is warming during the 2013-2017 period, and the temperature increment shows a parabolic pattern with the altitude, by an average increase of \sim 2°C (Figure S1). Thus, we design a sensitivity simulation, with a temperature increase of 2°C in the troposphere over the plateau. In the model, we set to the 2°C warming in the initial and boundary fields. In order to ensure a persistent influence of the 2°C warming, we drive the initial field with a 2°C increment every day. Then, by comparing the difference between the sensitivity simulation and the baseline simulation, we determine the impact of the 2°C warming over the Tibetan Plateau on air quality in the Sichuan Basin." In Lines 167 - 178.

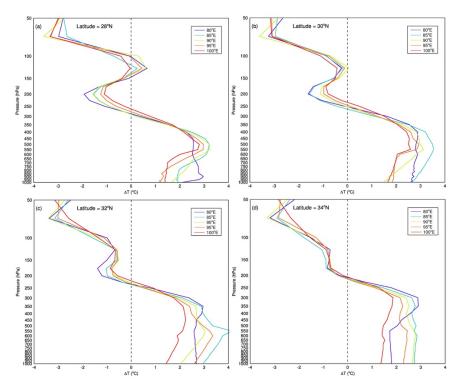


Figure S1 Vertical profile of temperature change along the longitude ($80^{\circ}E - 100^{\circ}E$) covering the plateau at 28°N, 30°N, 32°N and 34°N, ΔT is calculated by the annual temperature increase rate from 2013 to 2017 multiplying by the number of years (N = 5). Noted that the temperature in the troposphere over the Tibetan Plateau (600 hPa - 250 hPa) is inhomogeneously warming by 0 - 4 °C from 2013 to 2017, and we take an average warming increase of 2 °C.

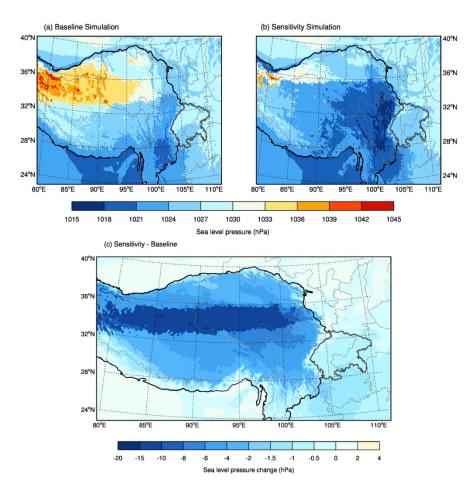
Comment 3. In line 231-232, is it correct here "the overestimated PM2.5 concentration is mainly caused by the overestimated wind speed"? Or underestimated wind speed?

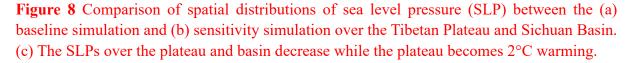
Response: Yes. We have explained that the overestimated $PM_{2.5}$ concentration here is mainly related to the wind departure in detail, including of an overestimated wind speed and a departure of wind direction. Figure S4 shows that the simulated temperature and humidity are well consistent with the observed, but the simulated winds are not consistent with the observed. Observational wind speed concentrates in the range of $1 - 2 \text{ m s}^{-1}$ (the average wind speed is 1.3 m s⁻¹), obviously lower than the simulated wind speed (mostly higher than 2 m s⁻¹, the average wind speed is 2.0 m s⁻¹,). The observed prevailing wind is northerly wind while the simulated is mainly easterly wind. Figure S6a shows that PM_{2.5} concentration is lower in the north to Sichuan Basin while higher in the east to the basin. Therefore, the simulated high PM_{2.5} concentration is mainly caused by a wind departure, which results in a false transport from the east to the basin. To clarify the explanation, we have revised the text as follows "During the period of Jan 17^{th} to Jan 20^{th} , the observed wind speed concentrates in the range of 1 - 2 m s⁻ ¹, with an average of 1.3 m s⁻¹, while the simulated wind speed is obviously higher, with an average of 2.0 m s⁻¹ (Figure S3). The observed prevailing wind is northerly wind while the simulated prevails easterly wind. Figure S6a shows that $PM_{2.5}$ concentration is lower in the north to the Sichuan Basin while higher to in the east to the basin. Therefore, the overestimated $PM_{2.5}$ concentration is mainly caused by the departure of winds, which results in a false transport from the east to the basin." In lines 241 - 249.

Comment 4. Could you further explain the thermodynamic reasons of the winds and PBLH changes due to 2K warming over TP in figure 7 and the description in line 263- 269 "easterly winds over the basin enhance while westerly wind over the plateau weaken...northerly winds over the basin slightly enhance,"?

Response: Yes, we have added the analysis of pressure gradient to explain the changes in winds (Figure 8, a new figure). The further explanation is as follows: "Wind patterns show that easterly winds over the basin enhance while westerly wind over the plateau weaken (Figure S6 and Figure 7). We further compare the difference in the surface pressure between the baseline and sensitivity simulations, and find out that surface pressure over the plateau and the basin all decreases when the plateau warms by $2^{\circ}C$ (Figure 8a and 8b). Over the plateau, the pressure drop has a decrease characteristic from west to east (Figure 8c), which results in a decreased pressure gradient and a weakened westerly wind. While in the basin, the pressure drop is less than the plateau. This leads to an increased pressure gradient from the basin to the plateau, inducing an intensified easterly wind. The enhanced easterly wind causes an increased transport of PM_{2.5} from the basin to the plateau. On the other hand, the weakened

westerly wind and the enhanced easterly wind are convergent at the border between the plateau and the basin (Figure 7), jointly leading to an increase in $PM_{2.5}$ concentration at the eastern edge of the plateau. Additionally, northerly winds over the basin slightly enhance, conducive to diluting the air and reducing $PM_{2.5}$ concentration. Both easterly winds transport and northerly winds dilution are favorable for a reduction of $PM_{2.5}$ concentration in the basin." In lines 278 - 291.





Comment 5. In line 293-295, similarly, could you further explain the mechanism of "a maximal temperature reduction located at 1.5 km to 3 km above the ground (Figure 9a)"?

Response: We have added the explanation: "This is probably due to a sharp topography decrease (from ~ 5 km in the plateau to < 1 km in the basin) that leads to a warm plume via subsidence. In the basin, there is a decrease in the temperature from the surface to ~ 4 km, with a maximal temperature reduction $(1 - 2^{\circ}C)$ located at 1.5 km to 3 km above the ground (Figure 10a). We speculate that changes in the surface pressure can account for the maximal

temperature reduction here. After the 2°C warming, surface pressure decreases in the basin (Figure 8), which produces more convergent airflow (as shown in Figure 7). The strengthened convergent airflow induces an intensified ascending motion, conducive to a reduction of temperature in the basin. As a result, the zone where the maximal temperature drop appears, overlaps with the zone with the maximal ascending motion. Furthermore, the intensified updraft increases the vertical temperature gradient and the instability in the lower troposphere of the basin, thereby causing a higher PBL height than that in the non-warming case (Figure 10b). On the contrary, the change in vertical temperature profile leads to a decreased vertical temperature gradient and increased thermal stability in the lower troposphere of the plateau, in which the PBL height decreases.

On the other hand, the convergent airflows by a weakened westerly wind over the plateau and a strengthened easterly wind in the basin (shown in Figure 8) triggers an ascending motion on the east side of the plateau, which is also beneficial to the development of the PBL height in the basin. Consequently, the elevated PBL facilitates vertical diffusion, leading to a reduction in $PM_{2.5}$ concentration over the basin. "In Lines 313 - 332.

Comment 6. Related to comments 4 and 5, the paragraph from line 302-311 did not make very clear discussion on the changes of wind and temperature gradient. I suggest the com- parison of the changes of pressure-difference between TP and basin, and see the circulation changes could easily explain the issues in comments 4 and 5.

Response: Thanks for your suggestions, in the revised manuscript, we have re-written the paragraph, and the text is referring to the response to Comment 5.

Comment 7. I don't think the ascending motion in this study is similar to the plateau "heat pump" effect raised by Lau (2016)

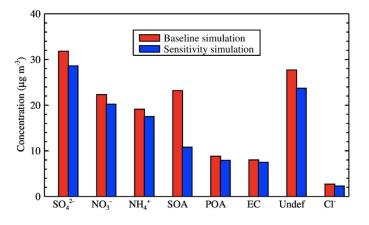
Response: Yes, they are not the same. Here, we consider of the reviewer's comment, and have deleted this statement that the ascending motion in this study is similar to that in the EHP mechanism.

Elevated Heat Pump (EHP) hypothesis proposed by Lau and Kim (2006) illustrate that absorbing aerosols (dust and black carbon) heat up the air over the south slope of the Tibetan Plateau, inducing an ascending motion in lower troposphere and a positive temperature anomaly in the mid-to-upper troposphere over the TP. According to the mass continuity principle, the air divergent in upper level and the air convergent in lower level, which further strengthens the upward motions. Under the circumstance, low-level convergence draws more warm and moist air from South Asia to increase monsoon rainfall. This thermodynamic mechanism shows that the heated plateau acts as an elevated heat pump.

In the present study, the temperature over the Tibetan Plateau rises by 2°C, which triggers an upward airflow on the eastern edge of the plateau. We would like to think that the role of the 2°C warming over the TP is similar to the positive temperature anomaly induced by absorbing aerosols in the EHP mechanism. Consequently, the 2°C warming leads to a convergent airflow and an ascending motion on the east edge of the plateau. The difference is that the EHP mechanism happens in the north-south direction, and our study explains the similar thermodynamic processes in the east-west direction.

Technical corrections:

Comment 8. I am misleading by the figure 6 in the first look and regards they are pie charts in percentage of species. Plot them as columns could be better.



Response: We have modified Figure 6 by a column chart, seen Figure 6 in the revised version.

Figure 6 Comparison of chemical composition of $PM_{2.5}$ concentration between the baseline simulation (red bar) and sensitivity simulation (blue bar) over the Sichuan Basin.

Comment 9. Setting figure 11 as figure 10c is reasonable.

Response: We have combined Figure 10 and Figure 11 together, and labeled Figure 11 in the revised version.

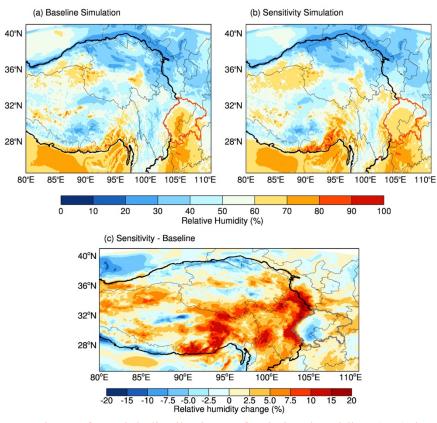


Figure 11 Comparison of spatial distributions of relative humidity (RH) between the (a) baseline simulation and (b) sensitivity simulation over the Tibetan Plateau and Sichuan Basin. (c) Spatial changes in RH after the plateau becomes 2°C warming, and the positive shows the RH increases while the negative shows the RH decreases.