Dear editor and reviewer,

Thank you very much for the comments and suggestions, which contribute to improve the quality of our paper. We have replied all comments and suggestions in our point-by-point response attached below. In order to highlight the changes what we have done, the color of the revised text will become blue.

Response to Anonymous Referee #1

RC1: In the paper entitled "Chemical composition, source and formation mechanism of urban PM2.5 in Southwest China", the authors used different techniques to investigate the properties of fine aerosols in Chengdu at the beginning of 2023. The investigated time period was divided into two pollution events and the reasons causing these pollution events were clarified. Although this paper showed us many pieces of information about the pollution, my feeling is that this paper is more like a report than a scientific paper. The methods used in this study are normal. The results are plenty but I did not find many new scientific findings from this paper. However, considering the hard work made in this study, I would like to give the authors a chance to revise their paper by highlighting the new findings in the revised manuscript. Thus, my suggestion is a major revision for this paper. The detailed comments are listed below.

Response. We are very grateful to the reviewer for giving us the chance to make revisions. Based on the reviewer's comments, we have made significant revisions to the structure and content of the manuscript. These revisions can be summarized as follows: (1) We have simplified some content that lacks innovation (section 3.1.1) or placed it in supplementary materials (Table 1 and the introduction to PMF factors) to ensure that the main text presents more important scientific information. (2) We conducted a more in-depth analysis of the obtained results: (a) when analyzing the regional transmission of pollutants, we not only analyzed the air mass transmission during different pollution periods, but also added the analysis results of concentration-weighted trajectory (Line 406-426), which can directly reflect the concentration contribution of regional transmission to the observation station; (b) we analyzed the sources and mixing structures of individual particles in the two haze periods, and

provided more scientific information and evidence for analyzing their formation mechanisms (section 3.3.4); (c) in order to provide readers with a clearer understanding of the evolution characteristics of pollution during the observation period, we have drawn a conceptual model of pollution evolution (Fig. 12), which displays the evolution characteristics of various meteorological and pollution indicators, particle mixing states and sources, and pollution formation mechanisms in the study period; (d) we discussed the significance of this study, particularly the importance of the TEM-EDS results in studying particle health and climate environmental effects (section 4.2). (3) We optimized the figures of the manuscript, such as presenting the content of Table 2 in the original manuscript in the form of figures (Figs. S5 and 7) and adding a conceptual model figure of pollution evolution (Fig. 12). These revisions ensures that the study results are more clearly expressed. We believe that through this major revision, the manuscript can present more and in-depth scientific information, allowing readers to better understand the sources and formation mechanisms of haze pollution in Chengdu. Thanks again for giving us the opportunity to make revisions.

1. Blank lines should be added between paragraphs.

Response. The formatting of the paragraph has been reset, i.e., blank a line before and after a paragraph.

2. Line 146, as the author mentioned, aerosols in China show new features recently. However, I did not find many new features presented in this paper. The one I found is that a less importance of sulfate and a stronger importance of nitrate in Chengdu. But what is the situation in other cities of China? I guess the situations are perhaps the same due to the policy released by the national government. Thus, I suggest the authors summarize the major new findings they found in this study and highlight them explicitly in the revised manuscript.

Response. Thanks for this important comment. We fully agree with the reviewer's comment that the original manuscript seriously lacks the innovative summary of this study, and the analysis of the data is not in-depth enough. Some of the results we reported, such as stronger nitrate contribution and weaker sulfate contribution, not only appeared in Chengdu but also in other cities in China. Therefore, during this revision process, we made significant revisions to the structure, content and presentation of the

manuscript. The revised manuscript provides a more in-depth analysis of the observed results, particularly by integrating the results of bulk-chemical and single-particle analysis, PMF results and WRF-Chem model to comprehensively analyze the formation mechanisms of the two pollution events. At the same time, we not only conducted a more in-depth analysis of the TEM-EDS results (section 3.3.4), emphasizing the importance of these results for the study of secondary effects of atmospheric particulate matter, but also pointed out that the integration of multiple methods is necessary for future air pollution research (section 4.2). To our knowledge, this study is the first to integrate these methods into the study of atmospheric pollution in Southwest China.

3. Line 231, please explain OM and EC here so that readers who are not familiar to these abbreviations can understand them.

Response. Thanks for this important comment. We have already explained OM and EC when they first appeared (Line 68 and Line 131).

4. Line 242, similar “to” the ….

Response. Corrected (Line 188).

5. Line 257, local sources include anthropogenic and natural emissions. The authors made a sensitivity test by switching off only anthropogenic emissions, so the results can only depict the influence of local anthropogenic sources

Response. Thanks for this valuable comment. We have added necessary explanations in the main text to ensure that readers can more accurately understand our research results (Line 205-206), i.e., “It is worth noting that this study mainly focuses on the contribution of anthropogenic sources, without considering the contribution of natural emissions, such as biogenic sources.”

6. Lines 295-345, as the information given here is not very related with the topic of the paper, I suggest the authors largely shorten it.

Response. We fully agree with the reviewer's comment that this section is only a comparison of the

obtained results, reflecting very little scientific information and not very relevant to the research topic. Therefore, we have largely shortened this section (reduce from 971 to 391 words) and the Table 1 has been placed in the supplementary materials.

7. Line 378, lower than that of Feb. 3 or Feb. 4? It was written that a haze alarm was released on Feb. 4.

Response. We would like to explain the haze alarm policy in Chengdu, which means that based on the current level of pollutant emissions and meteorological conditions in the coming days, the environmental protection department can predict the air quality in the coming days. If it is predicted that severe pollution may occur in the next few days, the environmental protection department will release a haze alarm. At present, there are three levels of haze alarm in Chengdu:

"Yellow" haze alarm: It is predicted that the daily average of air quality index (AQI) > 200 (or $PM_{2.5}$ mass concentration >115 μg m^{-3}) will last for 2 days (48 hours) or more, and did not meet higher level warning conditions.

"Orange" haze alarm: It is predicted that the daily average of AQI > 200 for 3 days (72 hours) or more, or $PM_{2.5}$ concentration >115 μg m^{-3} for 3 days (72 hours) or more, and $PM_{2.5}$ concentration >150 μg m^{-3} for 1 day (24 hours) or more, and did not meet higher level warning conditions.

"Red" haze alarm: It is predicted that the daily average of AQI > 200 for 4 days (96 hours) or more, and that the daily average of AQI > 300 for 2 days (48 hours) or more; or it is predicted that the daily average of AQI will reach 500.

In order to reduce the harm of haze to the public, government departments will take corresponding pollution reduction measures after releasing a haze alarm. For example, in this study, after releasing the "orange" haze alarm, a large number of pollution reduction measures were taken in Chengdu and surrounding areas, such as the time limit for motor vehicles was tightened, all open-air operations and all kinds of construction were prohibited and industrial enterprises were instructed to cease production or had limitations imposed on their production depending on the type of goods they were producing. Under the influence of these emission reduction measures, the concentration level of pollutants during

the alarm period usually does not significantly increase, and may even decrease. Therefore, it is reasonable that the $PM_{2.5}$ concentration during the orange haze alarm was lower than that during the peak pollution period before its release.

We can simply understand that after releasing a haze alarm, if no pollution control measures are taken, there will definitely be more serious pollution in the next few days. The fact is, in order to prevent the occurrence of such heavy pollution, government departments will take a large number of pollution reduction measures after the haze alarm is released. Then, many key sources of air pollution (such as motor vehicles, industry, combustion processes and construction sites) will be prohibited from emitting or reducing emissions. Finally, these pollution reduction measures not only prevent serious pollution events from occurring, but may also lead to a decrease in pollutant concentration levels.

8. Lines 378-382, the reasons can be various such as the change of meteorological conditions, so please give your evidence here.

Response. We fully agree with the reviewer's opinion that there are many factors that may cause a decrease in pollutant concentration. Here, we infer that the reduction in pollutant concentration is closely related to the implementation of emission reduction policies. This can be supported by the changes in the composition of $PM_{2.5}$ sources before and after the release of the "orange" haze alarm.

Compared to before the "orange" haze alarm, many strict pollutant reduction measures were implemented in Chengdu and surrounding areas during the alarm period, such as the time limit for motor vehicles was tightened, all open-air operations and all kinds of construction were prohibited and industrial enterprises were instructed to cease production or had limitations imposed on their production depending on the type of goods they were producing. Correspondingly, the contributions of sources related to these activities, such as vehicular emission, coal combustion, industrial processes and dust, have decreased by 7.9%, 16.5%, 6.3% and 10.9%, respectively (Fig. R1). In fact, the haze alarm period in this study corresponds to more unfavorable meteorological conditions. According to our observations, from NP-2 to Haze-2 ("orange" haze alarm period), the relative humidity increases from 50±6% to 72±8%, while the wind speed decreases from 0.7±0.4 m s^{-1} to 0.6±0.4 m s^{-1}. This is conducive to the generation and accumulation of pollutants. Therefore, based on the observed

meteorological conditions and changes in source composition, we can infer that the decrease in pollutant concentration here is closely related to various emission reduction measures during the "orange" haze alarm. Of course, if the reviewer still believes that our inference lacks sufficient evidence, we are also happy to delete it.

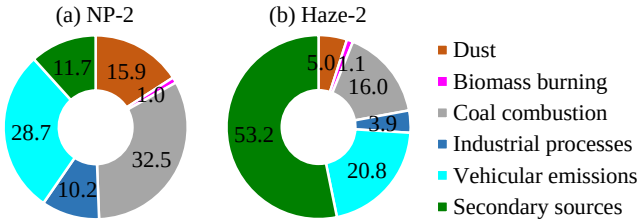

Figure S1: Composition of $PM_{2.5}$ sources during NP-2 and Haze-2 periods (%).

9. Table 2, is that better to show a figure instead of the table here as the numbers may not be that important.

Response. According to the reviewer's suggestion, the table has been presented in the form of figures (Fig. 7 and S5).

10. Line 495, should we look at Fig. 1(a) or Fig. 1(b)?

Response. Thanks for this important comment. In fact, we also found that it is unreasonable to go back to the beginning of the main text to search for the corresponding figures while reading the latter half of it. This may have caused trouble for readers.

At the same time, we found that our original content in this section was only a description of the geographical regions (cities) that the air masses passed during different periods and lack relevance to the topic of the study. Therefore, we have rewritten this section. The rewritten content not only compares the composition of air masses during different periods, but also relates air masses to pollution levels. Accordingly, we defined the six types of air masses as "clean" or "polluted" air masses. Then we found that the contribution of "clean" air masses during the NP1 period was higher than that of the two haze periods, and the air masses during the haze periods all passed through key potential source areas of pollutants. This proves the significant impact of regional transmission on pollution in Chengdu

at different time periods (Line 417-426).

11. Line 698, how did you calculate these contributions? Please give the definitions of the contributions and the methods obtaining them.

Response. We are very sorry for this unclear discussion. we have added the calculation of local contribution and regional transmission in section 2.4.2. The calculation formulas for the relative contribution of local sources and regional transmission is as follows:

$$\text{Regional transmission } (PM_{2.5}) = \frac{\text{Sensitivity scenario } (PM_{2.5})}{\text{Baseline scenario } (PM_{2.5})} \times 100\%$$

$$\text{Local sources } (PM_{2.5}) = 1 - \text{Regional transmission } (PM_{2.5})$$

12. Line 698, again, the local sources here include only anthropogenic sources.

Response. As our response to comment 5, we have added necessary explanations in the main text to ensure that readers can more accurately understand our research results (Line 205-206), i.e., "It is worth noting that this study mainly focuses on the contribution of anthropogenic sources, without considering the contribution of natural emissions, such as biogenic sources."

13. From Fig. S2 in the supplementary information, the model results of $PM_{2.5}$ and observations are not in a good consistency. Therefore, I doubt about the validity of the results shown in Section 3.6. In addition, my feeling is that the WRF-Chem simulation is not very associated with the topic of the paper, is that possible to remove this part?

Response. Thanks for this important comment. (1) At present, we have optimized the emission inventory and parameterization scheme, and re-run the simulation. The correlation between the simulated and observed values of the new simulation has been improved to 0.73, the NMB has been reduced to -24.2%, and the NME has been reduced to 27.1% (Fig. R2), which is a further improvement of the new simulation results compared with the previous results. Moreover, the evaluated parameters of the model simulation in this paper are within the acceptable range as reported by Huang et al (2021). Therefore, the simulation results in this study can be used for the analysis of the causes of $PM_{2.5}$

pollution in Chengdu. (2) When analyzing the formation mechanism of pollution, we found that there were differences in the sources and mixing states of particulate matter between the two haze periods. At the same time, there were significant differences in the composition of air masses and potential source areas between the two haze periods. These pieces of evidence suggest that regional transmission may have different impacts on the two haze events. However, we lack quantitative results. Therefore, we quantitatively studied the contribution of regional transmission at different time periods using the WRF-Chem model, which is crucial for clarifying the formation mechanism of pollution.

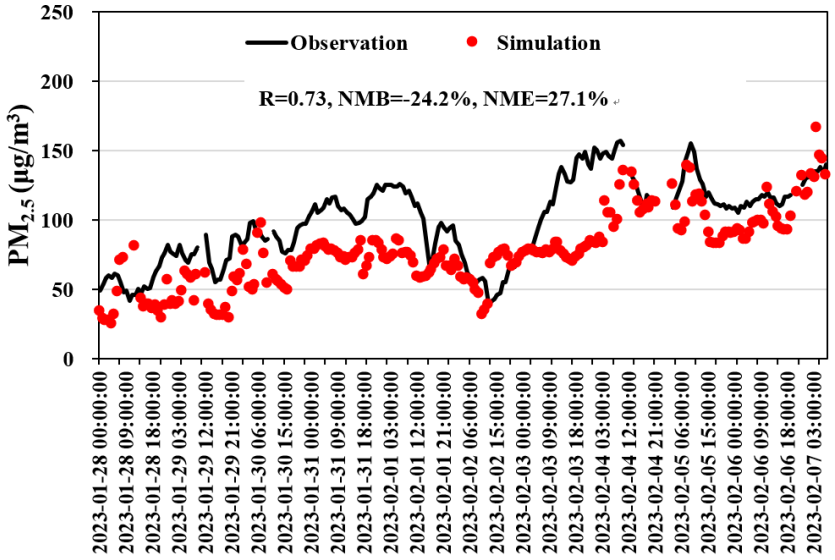

Figure R2: Temporal variation in the simulated and observed surface $PM_{2.5}$ concentration at the Chengdu station.

References

Huang, L., Zhu, Y., Zhai, H., Xue, S., Zhu, T., Shao, Y., et all., 2021. Recommendations on benchmarks for numerical air quality model applications in China - Part 1: $PM_{2.5}$ and chemical species. Atmos. Chem. Phys. 21, 2725-2743, https://doi.org/10.5194/acp-21-2725-2021, 2021.

14. Line 755, it is always better to share the data to the public instead of giving this sentence.

Response. Thanks for this important comment. According to the reviewer's suggestion, we have shared the data (both main text and supplementary materials) with the public.

15 In addition, the language of this manuscript has been edited by a professional organization, and the language editing certificate is as follows:

Certificate

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Contact author: Junke Zhang	**Manuscript:** Chemical composition, sources and formation mechanism of urban $PM_{2.5}$ in Southwest China: A case study in January 2023

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