

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 #2

RC2. In this study, the authors investigated the characteristics of aerosol chemical compositions by bulk-chemical and single-particle observations, analyzed the sources and formation mechanism of PM_{2.5} pollution at the beginning of 2023 in Chengdu. The results of this observation experiment, including chemical components, meteorological conditions and source analysis, are comprehensive introduced in this paper. However, with the lack of the innovation in methods or ideas, this work did not provide significant new insights and scientific highlights. I think this paper is more a presentation of observational data rather than a scientific analysis as research article. In addition, the observation period is only about half a month, which does not represent the pollution level in southwest China as described in title. I also agree with previous reviewers of this article about the lack of analysis and discussion of new scientific findings. As a result, I would encourage the authors to carry out in-depth analysis and innovative results with major revision. But much further work is still required at this stage.

Response. We are very grateful to the reviewer for giving us the chance to make revisions. 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 WCWT (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 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 and adding a conceptual model figure of pollution evolution. (4) Although our study period is short, it cannot be ignored that this period includes two typical haze processes. Clarifying their sources and formation mechanisms is also of great value for future air pollution control. Therefore, referring to similar studies in the past (Zhang et al., 2019; Zhu et al., 2016; Liu et al., 2013), we have defined our study as a "case study". Correspondingly, the title has been modified to: "**Chemical composition, sources and formation mechanism of urban PM_{2.5} in Southwest China: A case study in January 2023**". 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.

Reference:

Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., Gu, J., Chen, C., Yang, Y., Liu, X., Yang, T., Zhang, Y., Tian, H., and Hu, M.: Formation and evolution mechanism of regional haze: a case study in the megacity Beijing, China, *Atmos. Chem. Phys.*, 13, 4501-4514, <https://doi.org/10.5194/acp-13-4501-2013>, 2013.

Zhang, W., Zhang, Y. L., Cao, F., Xiang, Y., Zhang, Y., Bao, M., Liu, X., and Lin, Y. C.: High time-resolved measurement of stable carbon isotope composition in water-soluble organic aerosols: method optimization and a case study during winter haze in eastern China, *Atmos. Chem. Phys.*, 19, 11071-11087, <https://doi.org/10.5194/acp-19-11071-2019>, 2019.

Zhu, X. W., Tang, G. Q., Hu, B., Wang, L. L., Xin, J. Y., Zhang, J. K., Liu, Z. R., Münkkel, C., and S., W. Y.: Regional pollution and its formation mechanism over North China Plain: A case study with ceilometer observations and model simulations, *J. Geophys. Res-Atmos.*, 121, D14574,

<https://doi.org/10.1002/2016JD025730>, 2016.

1. L171-174: The diagram in the upper left corner of Figure 1(a) needs to remove the information of road network.

Response. Thanks for this important comment. In fact, we also feel that the expression of this diagram is not clear enough. Currently, we have provided a clearer map that presents the terrain characteristics of the study area and the detailed location of the observation station (Fig. 1), which is beneficial for readers to have a clearer understanding of the research area. Meanwhile, the analysis of air mass transmission is an auxiliary content, and relevant figure have been placed in the supplementary materials (Fig. S3).

2. L217: Please add references about density of 2 g cm^{-3} .

Response. The necessary references has been added (Line 164-165), such as Xu et al. (2021), Li et al. (2021), Li and Shao (2009) and Marple et al. (1993).

References:

Li, W. and Shao, L.: Transmission electron microscopy study of aerosol particles from the brown hazes in northern China, *J. Geophys. Res.*, 114, D09302, <https://doi.org/10.1029/2008jd011285>, 2009.

Li, W., Teng, X., Chen, X., Liu, L., Xu, L., Zhang, J., Wang, Y., Zhang, Y., and Shi, Z.: Organic coating reduces hygroscopic growth of phase-separated aerosol particles, *Environ. Sci. Technol.*, 55, 16339-16346, <https://doi.org/10.1021/acs.est.1c05901>, 2021.

Marple, V. A., Rubow, K. L., and Olson, B. A.: Inertial, gravitational, centrifugal, and thermal collection techniques, in aerosol measurement, *Aerosol Meas.*, 8, 206–233, <https://doi.org/10.1002/9781118001684.ch8>, 1993.

Xu, L., Liu, X., Gao, H., Yao, X., Zhang, D., Bi, L., Liu, L., Zhang, J., Zhang, Y., Wang, Y., Yuan, Q., and Li, W.: Long-range transport of anthropogenic air pollutants into the marine air: insight into fine particle transport and chloride depletion on sea salts, *Atmos. Chem. Phys.*, 21, 17715–17726,

<https://doi.org/10.5194/acp-21-17715-2021>, 2021.

3. L372: Add the definition of “orange” haze alarm.

Response. Due to the haze alarm in Chengdu includes three levels, namely “yellow”, “orange” and “red” alarms. And their definitions are relatively complex. Therefore, in order to provide readers with a comprehensive understanding of haze alarm information, we have introduced their definitions in the supplementary materials (Text S2).

4. L495-505: This description seems superfluous and does not fit the main idea of the article.

Response. Thanks for this important comment. We also found that our original content in this section was only a description of the geographical regions (cities) that the air masses passed and lack relevance to the topic of the study. Therefore, we have rewritten this section (Line 417-426). The rewritten content not only compares the composition of air masses during the three 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 different periods.

5. L556-558: The particulate removal process is complex and there is no direct indication that this is influenced by the hygroscopicity here.

Response. Thanks for this important comment. Our analysis approach in this section is as follows: (1) From Haze-1 to NP-2, the $PM_{2.5}$ mass concentration decreased by $85 \mu\text{g m}^{-3}$ within 31 hours. Correspondingly, this process is accompanied by short-term precipitation and strong winds, which are considered meteorological factors that cause a rapid decrease in pollutant concentration in the short term (Tsai et al., 2014; Zhang et al., 2015; Hu et al., 2021). (2) During the rapid decrease in $PM_{2.5}$ concentration, the contribution of secondary inorganic species (SNA measured by filter sampling method) or SIA-containing particles (OM-SIA and OM-SIA-soot particles measured by TEM-EDS method) with strong hygroscopicity decreased by 15.6% and 27.1%, respectively, while the

contribution of hydrophobic carbon species (OM and EC measured by filter sampling method) or particles (OM, soot and OM-soot measured by TEM-EDS method) increased by 2.8% and 23.7%, respectively. Therefore, we infer that the significant decrease in PM_{2.5} concentration and changes in its chemical composition are closely related to the precipitation and wind. Of course, if the reviewer still believes that our inference lacks sufficient evidence, we are also very happy to delete it.

Reference:

Hu, W., Zhao, T., Bai, Y., Kong, S., Xiong, J., Sun, X., Yang, Q., Gu, Y., and Lu, H.: Importance of regional PM_{2.5} transport and precipitation washout in heavy air pollution in the Twain-Hu Basin over Central China: Observational analysis and WRF-Chem simulation, *Sci. Total Environ.*, 758, 143710, <https://doi.org/10.1016/j.scitotenv.2020.143710>, 2021.

Tsai, Y. I., Kuo, S. C., Young, L. H., Hsieh, L. Y., and Chen, P. T.: Atmospheric dry plus wet deposition and wet-only deposition of dicarboxylic acids and inorganic compounds in a coastal suburban environment, *Atmos. Environ.*, 89, 696-706, <https://doi.org/10.1016/j.atmosenv.2014.03.013>, 2014.

Zhang, Z., Zhang, X., Gong, D., Quan, W., Zhao, X., Ma, Z., and Kim, S. J.: Evolution of surface O₃ and PM_{2.5} concentrations and their relationships with meteorological conditions over the last decade in Beijing, *Atmos. Environ.*, 108, 67-75, <https://doi.org/10.1016/j.atmosenv.2015.02.071>, 2015.

6. L605-620: Figure 7 and its associated descriptions are best moved to the supplement file.

Response. According to the reviewer's comment, we have moved Figure 7 and its associated descriptions to the supplementary materials (Text S3).

7. L698-704: How were the contributions of local sources and regional transmission calculated?

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:

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

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

8. L633: Please explain what is meant by “non-exhaust emissions”.

Response. Thanks for this important comment. A detailed introduction to “non-exhaust emissions” can be found in Charron et al. (2019). However, we found that this discussion is not closely related to the topic of this study, and therefore it has been deleted.

Reference:

Charron, A., Polo-Rehn, L., Besombes, J.-L., Golly, B., Buisson, C., Chanut, H., Marchand, N., Guillaud, G., and Jaffrezo, J.-L.: Identification and quantification of particulate tracers of exhaust and non-exhaust vehicle emissions, *Atmos. Chem. Phys.*, 19, 5187-5207, <https://doi.org/10.5194/acp-19-5187-2019>, 2019.

9. The grammar of the essay needs a thorough examination.(for example, L125: “investigate” ; L257: “sensitivity”; L315: “mitigate”...)

Response. Thanks for this important comment. 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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