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.

Dear Authors, I greatly appreciate the effort you made to improve the manuscript. Please consider the following additional comments:

Response to Comment 5, from referee #4: "In fact, the first comment on the number of motor vehicles in Chengdu appeared in the introduction section, and we provided necessary references, i.e. https://www.mps.gov.cn/n2254098/n4904352/c9244719/content.html (Line 94)".
The given reference is not in English. Please provide a reference that is understandable to all readers of ACP.
Response. Thanks for this important comment. We fully agree with that English reference is crucial for readers to understand scientific research results. However, the content here is about the ranking of Chengdu's motor vehicle ownership in China, and the latest data was released in October 2023, which is only three months away from now. Therefore, there is currently no reference reporting on the relevant data. Therefore, we have to cite relevant reports from government departments. However, according to this important comment, we have replaced other non English references. For example, "*such as the "Atmospheric Pollution Prevention and Control Action Plan (APPCAP)" during 2013–2017 (https://www.gov.cn/zwgk/2013-09/12/content_2486773.htm, last access: 10 January 2024) and the "Three-year Action Plan to Win the Blue Sky Defense War (BSDW)" during 2018–2020 (https://www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm, last access: 10 January 2024)*" has been revised as "*such as the "Atmospheric Pollution Prevention and Control Action Plan (APPCAP)" during 2013–2017 and the "Three-year Action Plan to Win the Blue Sky Defense War (BSDW)" during 2018–2020 (Geng et al., 2019; Huang et al., 2014; Huang et al., 2021b; Wang et al., 2022a)*" (Line 53-55).

Comments 6 and 10, from referee #4.
Please include the response to these comments in the text in order to clarify the issue for other readers.
Response. Thanks for this important comment. The responses have been appropriately added to the main text:
Line 278-280: "*Due to these emission reduction measures mainly targeting pollution sources that occur during the daytime. Therefore, although there was a significant increase in pollution at night, the pollution level in the daytime in the following days was lower than that on 3 February.*"
Line 455-458: "*It can be seen that although single-particle and bulk-chemical methods analyzed the chemical composition of particulate matter from two different perspectives, and there were differences in the contribution changes of similar species (such as SIA particles and SNA) with pollution evolution, but the two types of data exhibited similar trends. By combining these results, we can have a clearer understanding of the formation mechanism of pollution.*"

Comment 12, from referee #4: "In section 3.4 source apportionments, the sources profiles of each factor

should be given, and related references should be cited to support the source apportionment results."
I agree that the source profiles can be described in the supplemental material document. However, the references to support the association of a source to each profile are still missing.
Response. Thanks for this important comment. We have added all necessary references in the supplementary materials.

In addition, just after submitting my decision regarding the previous version of the manuscript, I received an additional report from an anonymous referee, which I am copying below. Although their comments are minor, I think that they can further improve the manuscript. Hence, I ask you to consider them too.

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Response to Anonymous referee

In this study, the authors synthesized multiple methods to study the characteristics, sources and formation mechanisms of atmospheric $PM_{2.5}$ pollution in Chengdu. They captured two typical pollution processes and conducted in-depth analysis of their formation mechanisms, and the results can be used as a reference for local pollution reduction. Compared to previous studies on this city, the research methods and results in this study have a certain degree of innovation. In addition, after carefully reading the first round of review comments and the authors' responses, I believe that the authors responded to the comments with high quality and made necessary modifications, which significantly improved the quality of the manuscript. However, there are still some issues that need to be further modified and improved before being considered for acceptance. The detailed comments are as follows.

1. The authors conducted a detailed analysis of the formation mechanism of two haze processes during the observation period. I would like to know if there is any difference in the formation mechanism of winter haze in Chengdu compared to previous studies? Therefore, it is very necessary to compare with previous study results and add relevant discussions.
Response. Thanks for this valuable comment. In fact, the referee #4 has raised similar comment, and we have added corresponding discussions. We compared our study results with the pollution formation mechanism reported in other regions of China (Northeast and North China Plain) and Chengdu in previous winters. Then emphasized the new results obtained in our study (Line 547-556): "*We found that the formation mechanism of haze in this study is different from previous winter study results in other regions in China, such as northern China. For example, Zhang et al. (2020b) found that residential coal burning and biomass burning were important factors causing winter haze in Northeast China. While, the contribution of industrial emissions to the formation of winter haze in the NCP region was much higher than that in Northeast China (Ma et al., 2016). Meanwhile, compared to previous winter studies in Chengdu (Liao et al., 2017; Li et al., 2017; Tao et al., 2013), the haze formation in this study presented some new characteristics. For example, (1) the key potential source areas during the haze period have shifted from the southeast in 2013 to the south; (2) mobile sources played a more important role, while the contributions of biomass burning and dust sources were significantly weaker; (3) the contribution of nitrate to the formation of heavy pollution was more prominent. This means that in order to develop efficient pollution reduction policies, it is very necessary to conduct targeted and timely research on the characteristics, sources and formation mechanisms of haze in the areas of concern.*"

Reference.

Li, L. L., Tan, Q. W., Zhang, Y., Feng, M., Qu, Y., An, J. L., and Liu, X. A.: Characteristics and source apportionment of $PM_{2.5}$ during persistent extreme haze events in Chengdu, southwest China, Environ. Pollut., 230, 718-729, https://doi.org/10.1016/j.envpol.2017.07.029, 2017.

Liao, T. T., Wang, S., Ai, J., Gui, K., Duan, B. L., Zhao, Q., Zhang, X., Jiang, W. T., and Sun, Y.: Heavy pollution episodes, transport pathways and potential sources of $PM_{2.5}$ during the winter of 2013 in Chengdu (China), Sci. Total Environ., 584-585, 1056-1065, https://doi.org/10.1016/j.scitotenv.2017.01.160, 2017.

Ma, L., Li, M., Zhang, H. F., Li, L., Huang, Z. X., Gao, W., Chen, D. H., Fu, Z., Nian, H. Q., Zou, L. L., Gao, J., Chai, F. H., and Zhou, Z.: Comparative analysis of chemical composition and sources of aerosol particles in urban Beijing during clear, hazy, and dusty days using single particle aerosol mass spectrometry, J. Clean. Prod., 112, 1319-1329, https://doi.org/10.1016/j.jclepro.2015.04.054, 2016.

Tao, J., Zhang, L. M., Engling, G., Zhang, R. J., Yang, Y. H., Cao, J. J., Zhu, C. S., Wang, Q. Y., and Luo, L.: Chemical composition of $PM_{2.5}$ in an urban environment in Chengdu, China: Importance of springtime dust storms and biomass burning, Atmos. Res., 122, 270-283, https://doi.org/10.1016/j.atmosres.2012.11.004, 2013.

Zhang, J., Liu, L., Xu, L., Lin, Q. H., Zhao, H. J., Wang, Z. B., Guo, S., Hu, M., Liu, D. T., Shi, Z. B., Huang, D., and Li, W. J.: Exploring wintertime regional haze in northeast China: role of coal and biomass burning, Atmos. Chem. Phys., 20, 5355-5372, https://doi.org/10.5194/acp-20-5355-2020, 2020b.

2. Line 107-108. The authors mentioned that "At the beginning of 2023, Chengdu experienced several severe haze events, during which the observed $PM_{2.5}$ mass concentration frequently exceeded the CNAAQS." Please provide more quantitative descriptions about the severity of this pollution, which will help readers understand the necessity of this study.

Response. According to the reviewer's suggestion, we have added a description of the pollution situation at the beginning of 2023 (Line 108-110): "*At the beginning of 2023, Chengdu experienced several severe haze events. The longest pollution process lasted for 12 days, and the highest daily average of $PM_{2.5}$ mass concentration reached 156 μg m^{-3}, which is 2.1 times the CNAAQS (daily average of 75 μg m^{-3}). Meanwhile, the proportion of mild and more severe pollution days in January and February reached 37.3%.*"

3. Line 172. The numbers of single particles analyzed during the four periods were 1325, 1159, 995 and 1870, respectively. I want to know if these analyzed particles are representative? Especially in the third period, the number of particles is less than 1000. This is crucial for the feasibility of the study results.

Response. Thanks for this important comment. We fully agree with the reviewer's comment, i.e., a larger sample size helps to obtain more accurate analysis results. However, during the TEM-EDS analysis process, each individual particle needs to be analyzed one by one, which is a very time-consuming, labor-intensive and costly process. Therefore, many previous studies were based on the analysis of hundreds or thousands of individual particle measurements. For example, Li et al., (2009) analyzed 810 particles in Beijing; Xu et al., (2020) analyzed 412, 486, and 887 aerosol particles at an inland urban site and a coastal urban site in China and a coastal site in southwestern Japan, respectively; Fu et al., (2012)

analyzed 834 particles in Shanghai; Li et al., (2021) analyzed 310, 280, 292 particles in Beijing, Hangzhou and Lesser Khingan Mountains, respectively. These results are believed to accurately reflect the characteristics of air pollution. Based on these previous studies, the numbers of particles analyzed for our four periods were 1325, 1159, 995 and 1870, respectively. We believe that our research results are reliable. In addition, accroding to the reviewer's comment, we will analyze as many particles as possible in future research.

Reference.

Fu, H., Zhang, M., Li, W., Chen, J., Wang, L., Quan, X. and Wang, W.: Morphology, composition and mixing state of individual carbonaceous aerosol in urban Shanghai, Atmos. Chem. Phys., 12, 693-707, https://doi.org/10.5194/acp-12-693-2012, 2012.

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

Li, W. J., Teng, X. M., Chen, X. Y., Liu, L., Xu, L., Zhang, J., Wang, Y. Y., Zhang, Y. and Shi, Z. B.: 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.

Xu, L., Fukushima, S., Sobanska, S., Murata, K., Naganuma, A., Liu, L., Wang, Y. Y., Niu, H. Y., Shi, Z. B., Kojima, T., Zhang, D. Z. and Li, W. J.: Tracing the evolution of morphology and mixing state of soot particles along with the movement of an Asian dust storm, Atmos. Chem. Phy., 20, 14321-14332, https://doi.org/10.5194/acp-20-14321-2020, 2020.

4. Line 252-253. "This inevitably leads to a the difference in the formation mechanism of air pollution in these two regions in China (Wang et al., 2021)" Why?

Response. Thanks for this important comment. This is mainly because temperature and relative humidity directly affect the homogeneous and heterogeneous generation processes of $PM_{2.5}$ chemical components. The necessary explanations have been added (Line 236-239): "*Due to the direct influence of T and RH on the homogeneous and heterogeneous generation processes of $PM_{2.5}$ chemical components, such as nitrates, sulfates and secondary organic compounds (Wang et al., 2021; An et al., 2019). Therefore, this difference inevitably leads to differences in the formation mechanisms of air pollution between the north and south of China.*"

Reference.

An, Z., Huang, R. J., Zhang, R., Tie, X., Li, G., Cao, J., Zhou, W., Shi, Z., Han, Y., Gu, Z., and Ji, Y.: Severe haze in northern China: a synergy of anthropogenic emissions and atmospheric processes, Proc. Natl. Acad. Sci. U.S.A., 116, 8657-8666, https://doi.org/10.1073/pnas.1900125116, 2019.

Wang, Y., Hu, M., Hu, W., Zheng, J., Niu, H., Fang, X., Xu, N., Wu, Z., Guo, S., Wu, Y., Chen, W., Lu, S., Shao, M., Xie, S., Luo, B., and Zhang, Y.: Secondary formation of aerosols under typical high-humidity conditions in wintertime Sichuan Basin, China: a contrast to the North China Plain, J. Geophys. Res-Atmos., 126, D03456, https://doi.org/10.1029/2021jd034560, 2021.

5. Line 396-399. The authors provided a detailed explanation for the synchronous increase of T and RH during the haze period in this study. They believe that the elevated RH caused an increase in T. I am curious why a similar phenomenon did not occur in Beijing, where RH increased but the T decreased.

Response. Thanks for this valuable suggestion. The relationship between meteorological factors and

pollution formation is complex, and there are obvious temporal and spatial differences in their relationship (Zhang et al., 2015; Zhang et al., 2019; Zhang et al., 2014; Yang et al., 2016). Meanwhile, the relationship between T and RH during the pollution process and their mutual influence mechanisms are beyond the scope of this study. Therefore, we have removed the discussion here.

Reference.

Yang, Y., Liao, H., and Lou, S. J.: Increase in winter haze over eastern China in recent decades: Roles of variations in meteorological parameters and anthropogenic emissions, J. Geophys. Res-Atmos., 121, 13,050-013,065, https://doi.org/10.1002/2016jd025136, 2016.

Zhang, R. H., Li, Q., and Zhang, R. N.: Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013, Sci. China Earth Sci., 57, 26-35, https://doi.org/10.1007/s11430-013-4774-3, 2014.

Zhang, X. Y., Xu, X. D., Ding, Y. H., Liu, Y. J., Zhang, H. D., Wang, Y. Q., and Zhong, J. T.: The impact of meteorological changes from 2013 to 2017 on $PM_{2.5}$ mass reduction in key regions in China, Sci. China Earth Sci., 62, 1885-1902, https://doi.org/10.1007/s11430-019-9343-3, 2019.

Zhang, Z. Y., Zhang, X. L., Gong, D. Y., Quan, W. J., Zhao, X. J., Ma, Z. Q., and Kim, S.-J.: Evolution of surface O_3 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. Fig. 9/Line 500-503. What are the potential effects of changes in the mixed structure of soot particles. I suggest the authors supplement necessary discussions.

Response. Thanks for this valuable suggestion. In fact, a preliminary discussion has been presented in section 4.2. According to the reviewer's suggestion, we have added a more in-depth discussion (Line 618-625): "*With the aggravation of pollution, soot particles mixed with other particles, and their particle sizes and morphologies undergone significant changes, which will further lead to changes in their hygroscopicity and optical properties, meaning that, ultimately, their climatic and environmental effects may differ significantly from when they exist alone (Adachi et al., 2010; Zhang et al., 2018). For example, Zhang et al. (2023a) found that the formation of organic coatings under the high RH could induce soot redistribution from the particle center to the edge in embedded soot-containing particles compared to partly coated soot-containing particles, and the soot redistribution reduces ~13% optical absorption enhancement of long-range transported soot particles, and the radiative absorption of long-range transported soot particles with a core-shell structure is overestimated by ~20% in the traditional Mie optical model*."

Reference.

Adachi, K., Chung, S. H., and Buseck, P. R.: Shapes of soot aerosol particles and implications for their effects on climate, J. Geophys. Res-Atmos., 115, D152061, https://doi.org/10.1029/2009jd012868, 2010.

Zhang, Y., Yuan, Q., Huang, D., Kong, S., Zhang, J., Wang, X., Lu, C., Shi, Z., Zhang, X., Sun, Y., Wang, Z., Shao, L., Zhu, J., and Li, W.: Direct observations of fine primary particles from residential coal burning: insights into their morphology, composition, and hygroscopicity, J. Geophys. Res-Atmos., 123, 12964-12979, https://doi.org/10.1029/2018jd028988, 2018.

Zhang, J., Li, W., Wang, Y., Teng, X., Zhang, Y., Xu, L., Yuan, Q., Wu, G., Niu, H., and Shao, L.: Structural collapse and coating composition changes of soot particles during long-range transport, J. Geophys. Res-Atmos., 128, e2023JD038871, https://doi. org/10.1029/2023JD038871, 2023a.

7. Line 524. Please provide a definition of "primary sources", which types of sources does it include? The current expression is unclear.

Response. Thanks for this important comment. According to the comment, we have added the necessary definitions (Line 495-497): "*Based on PMF, there were six factors identified in this study: dust, biomass burning, coal combustion, industrial processes, vehicular emissions and secondary sources (S3). Except for secondary sources, the other five factors can be referred to as primary sources.*"

8. Line 551-553. "During the NP-2 period, with the removal by precipitation and strong easterly winds, the contributions of secondary sources and vehicular emissions decreased by 14.3% and 9.6%, respectively". The significant reduction in secondary sources is easy to understand as they are hygroscopic. Why has the contribution of vehicular emissions also shown a significant decrease? The authors mentioned earlier that the main emission species of vehicular emissions are carbonaceous components, which are weakly hygroscopic or hydrophobic.

Response. This is mainly because Haze-1 was mainly caused by the accumulation of pollutants from vehicular emissions. As show in Fig. 8, compared to NP-1, the proportion of vehicular emissions increased by 19.5% during Haze-1 and reached 38.3%. Therefore, after precipitation and strong winds occurred, these accumulated vehicular emissions pollutants were removed, and their contribution correspondingly decreased. Meanwhile, the strong winds also brought pollutants emitted by other sources (such as coal combustion and dust sources), which further caused a decrease in the relative contribution of vehicular emissions. This explanation has been added to the main text (Line 529-535): "*During the NP-2 period, the precipitation and strong easterly winds not only reduced the contribution of secondary sources rich in hygroscopic species (such as SNA) by 14.3%, but also effectively cleared (decreased by 9.6%) a large amount of accumulated vehicular emissions pollutants during Haze-1. On the contrary, the contributions of coal combustion and dust sources increased by 17.9% and 6.8%, respectively. This is because the continuous easterly winds carried pollutants related to coal combustion in eastern Sichuan Province and Chongqing, and greatly increased the contribution of coal combustion. At the same time, this continuous easterly wind also drove the contribution of dust in NP-2 to reach its highest level across the four periods (15.9%).*"

9. In addition, some comments lack necessary references, such as Line 95-97. The format of references needs to be unified and standardized.

Response. We have checked the entire text and added necessary references (Line 96 and 102; S3 in Supplementary materials). Meanwhile, we have corrected the format of the references based on the "Copernicus_Word_template".