Dear editor and reviewers,

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 #3

Authors have carefully addressed the comments raised by two reviewers. I respect author's efforts. In its present form, the work is acceptable for final publication in ACP.

Response. We greatly appreciate the reviewer's review and recognition of this manuscript.

Response to Anonymous referee #4

The Sichuan Basin is one of the regions severely affected by haze in China. This study focused on Chengdu, the capital of Sichuan Province, and conducted in-depth analysis of the haze processes that occurred in early 2023 through field observations and model. By reviewing the review process of this manuscript, I found that the quality of the revised manuscript has significantly improved compared to the original version. Meanwhile, the results obtained in this study are interesting and can further enhance our understanding of the formation mechanism of air pollution in the city. However, there are still some scientific issues that need to be clarified or improved in the current version. Therefore, a revision is necessary before considering acceptance.

1. Line 50-55, some references about haze and non-haze study as well as control policy evaluation should be cited. Such as: Variations in $PM_{2.5}$, TSP, BC, and trace gases (NO_2, SO_2, and O_3) between haze and non-haze episodes in winter over Xi'an, China. Atmospheric Environment, 2015, 112, 64-71. Saccharides in summer and winter $PM_{2.5}$ over Xi'an, Northwestern China: Sources, and yearly variations of biomass burning contribution to $PM_{2.5}$. Atmospheric Research, 2018, 214, 410-417.

Inter-annual variability of wintertime $PM_{2.5}$ chemical composition in Xi'an, China: Evidences of changing source emissions. Sci. Total Environ. 545, 546–555.

Response. Thanks for providing these valuable references and they have already been cited in the manuscript (Line 38 and 51).

2. Line 60-62. I suggest the authors provide more comprehensive evidence and data on the improvement of air quality in China in recent years. It is insufficient to provide an example based solely on the study results from Beijing. In fact, there are already many comprehensive reports or literature on the implementation effectiveness of emission reduction policies.

Response. Thanks for this important comment. We have added a discussion on the improvement of air quality throughout China and the three major polluted areas in recent years (Line 58-62): "*For example, the annual mean $PM_{2.5}$ mass concentration in the China and its three key polluted areas, namely the Beijing-Tianjin-Hebei (BTH) and its surrounding areas, YRD and FWP, has decreased from 39, 60, 44 and 58 μg m^{-3} in 2018 to 29, 44, 31 and 46 μg m^{-3} in 2022, respectively. Meanwhile, the concentration of SO_2, one of the important gaseous precursors of $PM_{2.5}$, has decreased from 14, 20, 11 and 24 μg m^{-3} to 9, 10, 7 and 9 μg m^{-3}, respectively (https://www.mee.gov.cn/hjzl/sthjzk/zghjzkgb/, last access: 10 January 2024)*". These results can better support our viewpoint.

3. Line 100-102. "Despite numerous studies having used multiple methods to investigate the physical……" Please provide necessary references.

Response. The necessary references have been added, such as Chen et al., 2022, Huang et al., 2018, Huang et al., 2021a, Tao et al., 2014 and Wang et al., 2018a (Line 103-104).

References:

Chen, L., Zhang, J., Huang, X., Li, H., Dong, G., and Wei, S.: Characteristics and pollution formation mechanism of atmospheric fine particles in the megacity of Chengdu, China, Atmos. Res., 273, 106172, https://doi.org/10.1016/j.atmosres.2022.106172, 2022.

Huang, X., Zhang, J., Zhang, W., Tang, G., and Wang, Y.: Atmospheric ammonia and its effect on $PM_{2.5}$ pollution in urban Chengdu, Sichuan Basin, China, Environ. Pollut., 291, 118195, https://doi.org/10.1016/j.envpol.2021.118195, 2021a.

Huang, X., Zhang, J., Luo, B., Wang, L., Tang, G., Liu, Z., Song, H., Zhang, W., Yuan, L., and Wang, Y.:

Water-soluble ions in $PM_{2.5}$ during spring haze and dust periods in Chengdu, China: Variations, nitrate formation and potential source areas, Environ. Pollut., 243, 1740-1749, https://doi.org/10.1016/j.envpol.2018.09.126, 2018.

Tao, J., Gao, J., Zhang, L., Zhang, R., Che, H., Zhang, Z., Lin, Z., Jing, J., Cao, J., and Hsu, S. C.: $PM_{2.5}$ pollution in a megacity of southwest China: source apportionment and implication, Atmos. Chem. Phys., 14, 8679-8699, https://doi.org/10.5194/acp-14-8679-2014, 2014.

Wang, H., Tian, M., Chen, Y., Shi, G., Liu, Y., Yang, F., Zhang, L., Deng, L., Yu, J., Peng, C., and Cao, X.: Seasonal characteristics, formation mechanisms and source origins of $PM_{2.5}$ in two megacities in Sichuan Basin, China, Atmos. Chem. Phys., 18, 865-881, https://doi.org/10.5194/acp-18-865-2018, 2018a.

4. According to section 3.1, a haze event usually lasts for several days, while the collection time for each single particle sample was only 30 s to 3 min. I want to know how the authors ensure that the single particles they analyze are representative.

Response. We fully understand the concerns of the reviewer. However, TEM is a research method that analyzes the collected particles one by one. Therefore, it requires that the particles collected onto copper TEM grids should not overlap and each particle should be clearly visible. As is well known, the number concentration of particulate matter in the atmosphere is usually at a very high level, especially during pollution periods. This determines that the collection time for each sample cannot be too long. In fact, previous studies on air pollution using TEM method have adopted the same research approach as our study, and the obtained study results are considered reliable (Li and Shao, 2009; Li et al., 2014; Li et al., 2015; Xu et al., 2020; Xu et al., 2021; Zhang et al., 2021b).

References

Li, W. and Shao, L.: 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., Shao, L., Shi, Z., Chen, J., Yang, L., Yuan, Q., Yan, C., Zhang, X., Wang, Y., Sun, J., Zhang, Y., Shen, X., Wang, Z., and Wang, W.: Mixing state and hygroscopicity of dust and haze particles before leaving Asian continent, J. Geophys. Res-Atmos., 119, 1044-1059, https://doi.org/10.1002/2013jd021003, 2014.

Li, W. J., Chen, S. R., Xu, Y. S., Guo, X. C., Sun, Y. L., Yang, X. Y., Wang, Z. F., Zhao, X. D., Chen, J. M., and Wang, W. X.: Mixing state and sources of submicron regional background aerosols in the northern Qinghai-Tibet Plateau and the influence of biomass burning, Atmos. Chem. Phys., 15, 13365-13376, https://doi.org/10.5194/acp-15-13365-2015, 2015.

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

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.

Zhang, J., Yuan, Q., Liu, L., Wang, Y., Zhang, Y., Xu, L., Pang, Y., Zhu, Y., Niu, H., Shao, L., Yang, S., Liu, H., Pan, X., Shi, Z., Hu, M., Fu, P., and Li, W.: Trans-regional transport of haze particles from the North China Plain to Yangtze River Delta during Winter, J. Geophys. Res-Atmos., 126, D033778, https://doi.org/10.1029/2020jd033778, 2021b.

5. Line 263-264. “In particular, at the beginning of 2023, Chengdu has become the city with the highest number of motor vehicles in China” Please provide the data source.

Response. 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).

6. Line 294-295. “although there was a significant increase in pollution at night, the pollution level in the daytime was lower than that on 3 February.” Why? If the emission reduction policies during the haze alarm period have had a positive effect, what are the reasons for the high values at night? What are the main sources of pollution?

Response. Thanks for this important comment. (1) The emission reduction policies during the haze alarm period mainly target various anthropogenic sources, such as mobile sources, industrial processes and construction, which almost all appear during the daytime. This is why after the implementation of

emission reduction policies, the reduction of pollutants mainly occurred during the daytime. (2) The reasons for the high-level pollution at night are complex, such as a shallow boundary layer, secondary generation of pollutants or regional transmission. In fact, the phenomenon that the reviewer is concerned about occurred during the Haze-2 period, and according to our analysis, this pollution process is mainly caused by the regional transmission of secondary pollutants.

7. Line 369. "S-rich particles"? Is this a new type of particulate matter? If so, please provide necessary introduction.

Response. The "S-rich particles" should be "SIA particle" and it has been corrected (Line 347). We are very sorry for our carelessness.

8. Line 535-539. "This is mainly related to the various pollution reduction measures implemented in Chengdu, and even the broader SCB, in the past decade, such as……" I think some references are necessary here.

Response. The necessary reference has been added (Line 508-509), i.e., http://sthjt.sc.gov.cn/sthjt/c106120/2018/12/28/8f2a9dead56c4605ad2d26fe2e2fac43.shtml.

9. Line 577-579. "This means that if Chengdu's air quality is to achieve further improvement after reaching the current CNAAQS, it may need more attention paid to the contribution of local sources". I think this statement is unreasonable. In fact, the contribution of regional transmission reached 46%. Such a high contribution cannot be ignored in the future pollution control process. Therefore, I suggest that the authors revise it.

Response. Thanks for this important comment. We fully agree with the reviewer's viewpoint and the unreasonable statement has been removed. According to our analysis results, the future control of air pollution in Chengdu requires synchronous emission reduction of local sources and regional transmission.

10. Although the chemical composition results obtained based on bulk-chemical and single-particle analysis exhibited strong consistency, there is a significant difference in their magnitude of change. For example, from Haze-1 to NP-2, the contributions of SNA and SIA particles decreased by 15.6% and

27.1%, respectively; from NP-2 to Haze-2, the contributions of SNA and SIA particles increased by 24.2% and 42.4%, respectively. What factors are causing this difference?

Response. Thanks for this important comment. This is because SNA and SIA are two types of data, that is, SNA is the **mass** concentration of secondary inorganic components, while SIA is the **number** concentration of secondary inorganic particles. We must note that: (1) due to the differences in particle size distribution among different species, their contributions in number concentration and mass concentration are usually different. For example, in our study, the mass contribution of EC was only 5.8%–6.9%, while the corresponding species, namely soot-related particles, had a number contribution of 12.5%–23.9%; (2) the individual particles measured by TEM are usually a mixture of particles from multiple chemical components. These two factors ultimately lead to different degrees of variation in the pollution evolution for the two types of data. In fact, similar phenomena have also appeared in previous studies, such as (Zhang et al., 2020b; Zhang et al., 2021a). For example, in the study of Zhang et al. (2020b), they found that from the heavy Haze-I to Haze-II events, secondary inorganic ions (mass) significantly decreased from 62%-66% of the total $PM_{2.5}$ mass to 31%–35%, but OM (mass) markedly increased from 27%-30% to 53%-60%. Meanwhile, the S-OM particles fractions (number) significantly decreased from 60%-74% to 30%-32%, but K-OM fractions (number) largely increased from 4%-5% to 50 %-52%.

References:

Zhang, J., Liu, L., Xu, L., Lin, Q., Zhao, H., Wang, Z., Guo, S., Hu, M., Liu, D., Shi, Z., Huang, D., and Li, W.: 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.

Zhang, J., Huang, X., Yu, Y., Liu, Q., Zhang, J., Song, H., and Wang, Y.: Insights into the characteristics of aerosols using an integrated single particle–bulk chemical approach, Atmos. Res., 250, 105374, https://doi.org/10.1016/j.atmosres.2020.105374, 2021a.

11. Line 626-628. "……with the reduction of pollution and the enhancement of atmospheric oxidation in recent years (Zhao et al., 2020; Wang et al., 2023), the formation mechanism of haze is also undergoing dynamic changes……" What changes in pollution mechanisms have been found in this study compared to previous results? I think some necessary discussions need to be added to the main text.

Response. Thanks for this valuable comment. According to the reviewer's suggestion, 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 538-547): "*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.*"

12. In section 3.4 source apportionments, the sources profiles of each factors should be given, and related references should be cited to support the source apportionment results.

Response. Thanks for this important comment. In fact, an introduction to the sources profiles of each factor was included in the main text of the original manuscript. However, during the first round of review process, one referee suggested that these materials should be included in the supplementary materials. We also believe that this introduction is auxiliary and not the core content of our manuscript. Therefore, the relevant content has been included in the supplementary materials now (S3). We hope to gain the understanding of the reviewer.

13. Figures: (1) The marking of "fly ash/metal particles" in Figure 9 is not clear enough, please modify it. (2) Figure 11 is not clear enough, and its quality needs to be improved. (3) Figure 12. It is "relative humidity" rather than "humidity".

Response. We have completed the corresponding revisions. At the same time, we checked the quality

of all figures to ensure that they can be easily understood.