

Response to Reviewer Comments

Dear Reviewer,

We would like to thank you for your great effort and detailed work on this manuscript.

We have revised the manuscript and responded to each of the comments from the reviewers. In our response, your questions are shown in *italics*, and the responses are shown in standard text. For the ACP discussion, our research team also performed further analysis of the research results and made minor modifications to this manuscript.

We appreciate your help and time.

Sincerely yours,

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Title: Elucidating the pollution characteristics of nitrate, sulfate and ammonium in PM_{2.5} in Chengdu, southwest China, based on three-year measurements

Response to Anonymous Referee

This paper presented an overview of air pollution in Chengdu, southwest China based on study, a three-year observations of gas and particulate pollutants. Probably due to the special topography, the data from this site shows special characteristics of pollutants, different from most other polluted regions in China, e.g., North China plain and Pearl River Delta etc. Thermodynamic models and trajectory analysis have also been applied to analyze aerosol pH, partitioning of inorganic semi-volatile species and the contribution of potential source regions. Overall, I think that the datasets are very interesting and valuable, and authors have tried to give a comprehensive overview and analysis of the mechanisms beyond. However, there is still much room for improvement. I would suggest the authors to carefully consider my comments/suggestions in their revision before its final publication in ACP.

Response:

We appreciate your comments and have revised the manuscript accordingly. We firmly believe that your guidance is of great significance for improving our research.

Major concern:

1. Meteorological parameters and PM compositions show distinct diurnal variations compared to other regions such as NCP, PRD and YRD. Though chemistry is in this game, I guess that the special topography may play a dominant role in these features, which is missing from this submission. I'd suggest the authors to add these discussions.

Response:

We appreciate your comments and have revised the text. We also noticed that during the data analysis, the pollutant concentration in the daytime was higher, which was obviously different from that in NCP, PRD and YRD. We also collated the relevant research results in NCP, PRD and YRD and found that they were consistent with the

questions you raised. Therefore, according to your guesses and prompts, we conducted an in-depth analysis. The unique topographic structural features did have an important impact on the diurnal variation of meteorological factors in the Sichuan Basin, which may also be an important factor that causes the daily changes in air pollutants with unique characteristics. We also supplement and discuss the corresponding parts of the manuscript. Now, the text reads as follows:

In previous studies in Beijing-Tianjin-Hebei and the Pearl River Delta, the concentration of pollutants was affected by meteorological factors, and it was usually lower in the daytime than at night. In the Yangtze River Delta, the peak usually occurs in the morning, but in our study, the concentration was higher in the daytime than at night (Peng et al., 2011; Wang et al., 2018; Guo et al., 2017b). In addition to the diurnal variations in WS and atmospheric humidity, some studies have shown that due to the unique topographical structure of the Sichuan Basin, the atmospheric circulation between the Qinghai-Tibet Plateau, Yunnan-Guizhou Plateau and Sichuan Basin and the meteorological conditions of the Chengdu region are affected, such as the characteristics of air mass transport and typical “night rain” (more precipitation at night than in the day) under the influence of atmospheric circulation (Zhang et al., 2019b; Zhang et al., 2019a).

Reference:

- Guo, J., Xia, F., Zhang, Y., Liu, H., Li, J., Lou, M., He, J., Yan, Y., Wang, F., Min, M., and Zhai, P.: Impact of diurnal variability and meteorological factors on the PM_{2.5} - AOD relationship: Implications for PM_{2.5} remote sensing, *Environmental pollution*, 221, 94-104, 10.1016/j.envpol.2016.11.043, 2017b.
- Peng, G., Wang, X., Wu, Z., Wang, Z., Yang, L., Zhong, L., and Chen, D.: Characteristics of particulate matter pollution in the Pearl River Delta region, China: an observational-based analysis of two monitoring sites, *Journal of Environmental Monitoring*, 13, 1927-1934, 10.1039/c0em00776e, 2011.
- Wang, L., Li, W., Sun, Y., Tao, M., Xin, J., Song, T., Li, X., Zhang, N., Ying, K., and Wang, Y.: PM_{2.5} Characteristics and Regional Transport Contribution in Five Cities in Southern North China Plain, During 2013–2015, *Atmosphere*, 9, 157,

10.3390/atmos9040157, 2018.

Zhang, L., Guo, X., Zhao, T., Gong, S., Xu, X., Li, Y., Luo, L., Gui, K., Wang, H., Zheng, Y., and Yin, X.: A modelling study of the terrain effects on haze pollution in the Sichuan Basin, *Atmospheric Environment*, 196, 77-85, 10.1016/j.atmosenv.2018.10.007, 2019a.

Zhang, Y., Xue, M., Zhu, K., and Zhou, B.: What Is the Main Cause of Diurnal Variation and Nocturnal Peak of Summer Precipitation in Sichuan Basin, China? The Key Role of Boundary Layer Low-Level Jet Inertial Oscillations, *Journal of Geophysical Research: Atmospheres*, 124, 2643-2664, 10.1029/2018jd029834, 2019b.

2. *QA/QC. Quality assurance and control is essential for multi-year analysis. Maybe I overlooked it and I didn't find a description to assure the data quality. QA/QC would give the readers more confidence in your data and analysis, e.g., the extremely high NH₃ in the winter of 2017.*

Response:

We appreciate your comments and apologize for our unprofessional description. We agree with you very much because the instruments involved in this study are online monitoring equipment and have high time resolution (1 hour), so data quality assurance and control are the keys to determining the accuracy and scientific nature of this study. Therefore, we added this information to the supplementary materials. Now, it reads as follows:

Data quality control and assurance are important components of atmospheric comprehensive observation experiments. In addition to regular inspection and correction of the equipment through professional operation and maintenance to ensure the accuracy of experimental data, the quality control and processing of monitoring data, such as excluding outliers and data beyond the detection limit, are also an important. As shown in Figs. S1-4, the time sequence of monitoring data and the red part in the figure indicate that the data are missing and that the overall data integrity is good. The missing rates of PM₁₀, PM_{2.5} and PM₁ data in Fig. S1 are 5.0, 6.8 and 6.1%, respectively.

The missing rates of NO_3^- , SO_4^{2-} , NH_4^+ , OC and EC data in Fig. S2 are 18.3, 17.1, 20.9, 15.2 and 19.6%, respectively. In Fig. S3, the gaseous pollution of NO data is missing 18.2%, NH_3 is missing 11.3%, and other gases are missing 9%. The quality of meteorological data is good (Fig. S4), and the overall missing rate is 3.1% or less. On the whole, the observation data are good and do not affect the continuity of the data as a whole. The Cl^- , Na^+ , K^+ , Mg^{2+} and Ca^{2+} data are significantly missing, and this study only involved in analysis of the ISORROPIA-II thermodynamic equilibrium model. To ensure that each sample data point can be input into the model completely, 618 sample input models are selected according to the data quality control to eliminate the impact of missing ion data to ensure that the model analysis results are effective.

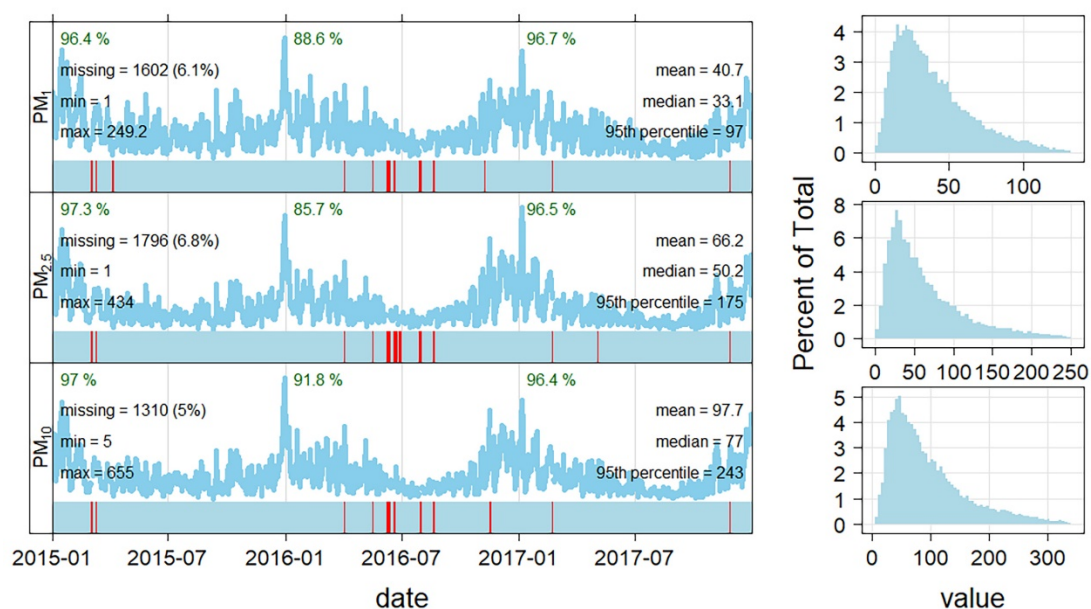


Fig. S1. PM₁₀, PM_{2.5} and PM₁ data quality assurance and control.

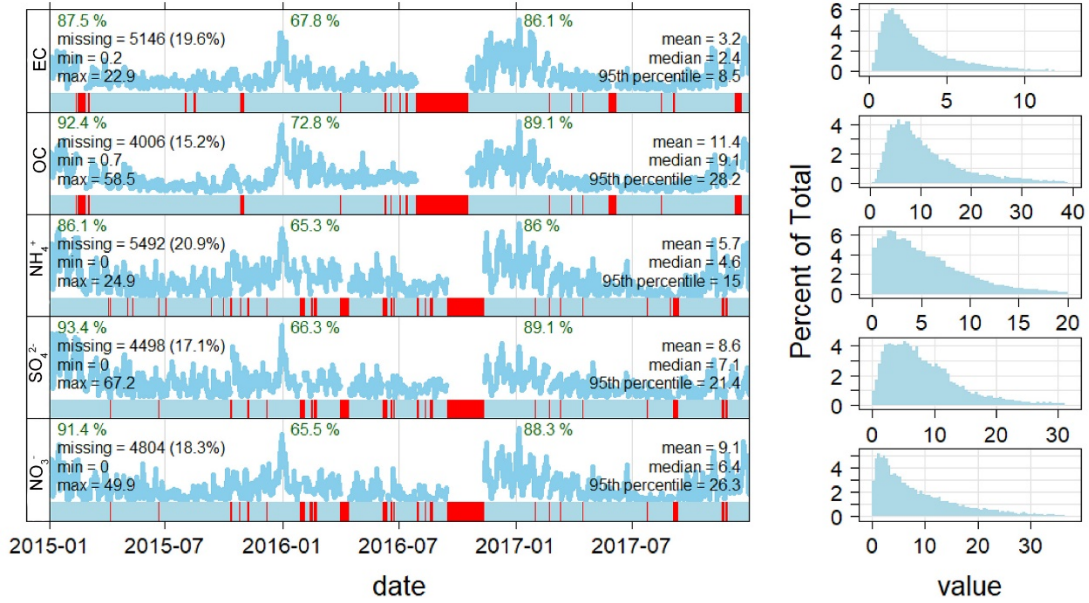


Fig. S2. NO₃⁻, SO₄²⁻, NH₄⁺, OC (organic carbon) and EC (element carbon) data quality assurance and control.

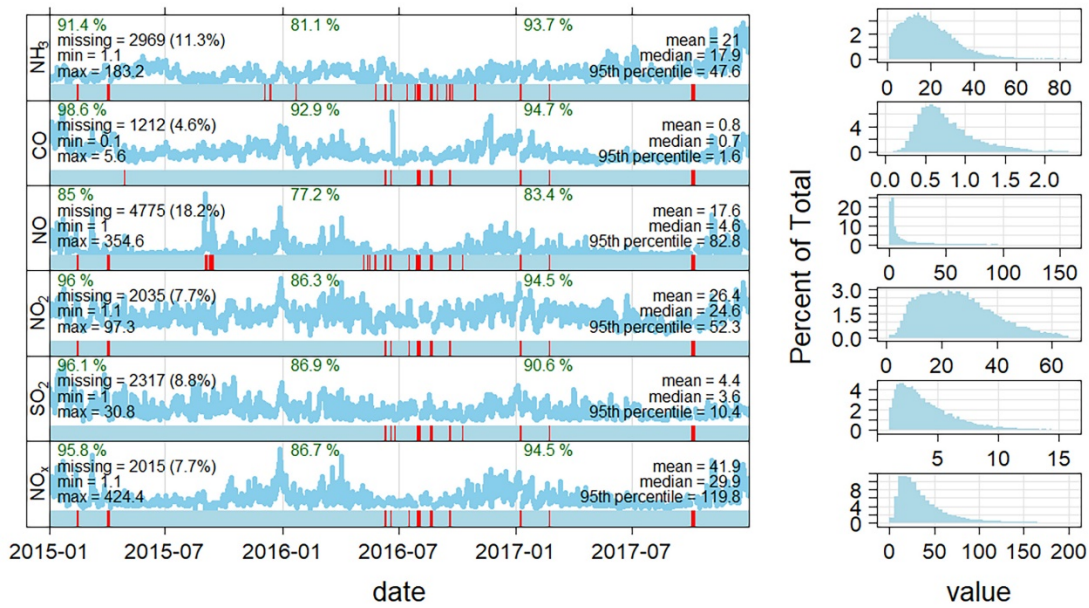


Fig. S3. NO_x, SO₂, NO₂, NO, CO and NH₃ data quality assurance and control.

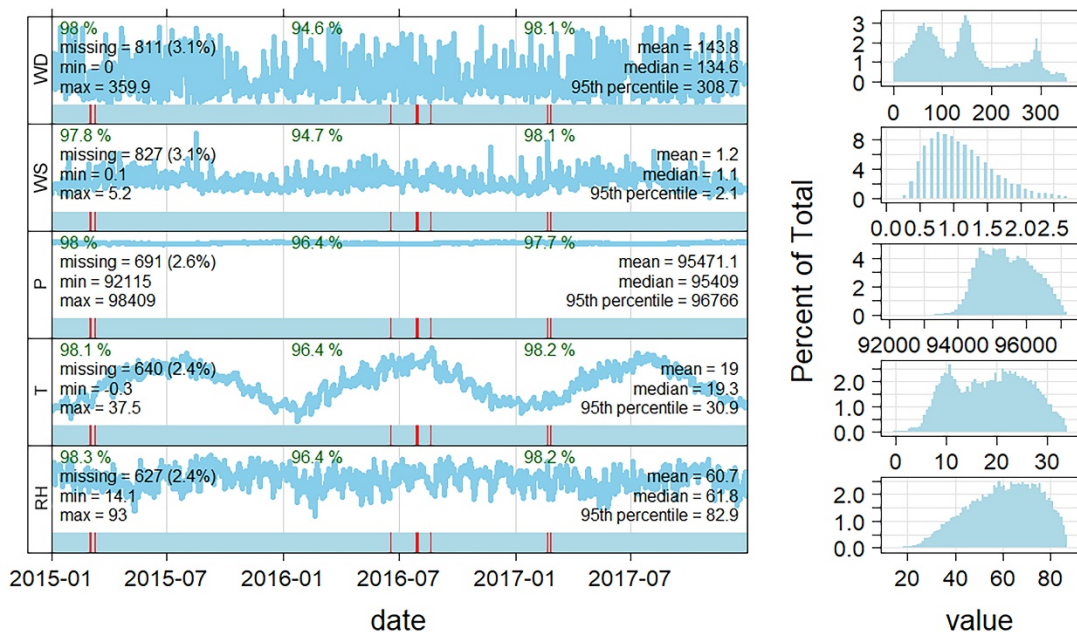


Fig. S4. Relative humidity (RH), temperature (T), atmospheric pressure (P), wind speed (WS) and wind direction (WD) data quality assurance and control.

3. To avoid jump between text and SI, the authors could consider moving some SI parts into the main text. For example, gas phase NH₃ that frequently used and discussed is missing from the main text.

Response:

We appreciate your comments and apologize for our unprofessional description. We have adjusted the positions of the figures according to the structure of the whole manuscript.

Minor comments:

Abstract: Line 21 “a long-term observational experiment was conducted from January 1, 2015 to December 31, 2017” Three years measurements are longer than a campaign-based experiment but I won’t call it “long-term”.

Response:

We appreciate your comments and apologize for our unprofessional description. We agree with you that the expression “long-term” observation instead of “three-year” observation is not accurate enough, so we use “three-year” observation.

Line 27 “Seasonal and diurnal variations have obvious characteristics, winter still has a high NSA concentration and emission intensity, and the concentration during the day was higher than that at night. “This is unusual; is it because of the valley topography

Response:

We appreciate your comments and apologize for our unprofessional description. As you commented in “Major concern: 1”, we have performed a comparative study of other regions of China, Beijing-Tianjin-Hebei, the Yangtze River Delta and the Pearl River Delta, and found that, as you believe, the diurnal variation of air pollutants in Sichuan Basin is indeed different from that in other regions. Based on related research results, the unique topographical structure of Sichuan Basin will indeed affect the meteorological conditions of Sichuan Basin by affecting the atmospheric circulation (interaction of the Qinghai Tibet Plateau, Yunnan Guizhou Plateau and Sichuan Basin). Therefore, the daily variation characteristics of pollutants may be significantly affected by meteorological conditions. In response to “Major concern: 1”, we have made corresponding modifications in the manuscript.

Line 34 “The ammonia-rich environment became increasingly obvious in the atmosphere of Chengdu” It is not clear what you want to say. Do you mean that you see an increase in NH3 concentration or partition rate?

Response:

We appreciate your comments and apologize for our unprofessional description. We have reconsidered our research purpose and found that the expression of the “ammonia-rich” environment is not accurate. Therefore, we revised the expression based on our current research results. In section 3.4.3 of the manuscript, the ratio of NH_4^+ to NO_3^- and SO_4^{2-} was analysed and found to increase from 2015 to 2017. The results may be attributed to the fact that the concentration of $\text{PM}_{2.5}$ gradually decreased under the implementation of relevant air pollution control measures, and its chemical composition also changed significantly; for example, the SO_4^{2-} concentration decreased more. Therefore, in the corresponding position of the manuscript, we corrected the

expression “ammonia-rich environment” and revised the relevant contents.

Page 3 line 69 “For example, photochemistry may affect the formation of NSA at high solar radiation, and the homogeneous reaction may dominate the formation of NSA in high relative humidity” I think you mean “heterogeneous” instead of homogeneous?

Response:

We appreciate your comments and apologize for our unprofessional description. Our purpose is to express the complex characteristics of NSA chemical conversions. According to previous studies, both homogeneous and heterogeneous reactions have chemical conversion processes of secondary inorganic aerosols, such as photochemical reactions, aerosol liquid-phase oxidation environments, and mineral dust catalysis. Therefore, we rewrote this sentence. Now, it reads as follows:

In addition, the chemical conversion of NO₂, SO₂ and NH₃ to form NSA is still very complex, and both homogeneous and heterogeneous reactions involve the chemical conversion of secondary inorganic aerosols, such as photochemical reactions, aqueous phase oxidation environments of aerosols and catalysis of mineral dust.

Page 3 line 82 “The characteristics of higher concentrations proportion of nitrate, sulfate and ammonium in PM_{2.5} were also found in other polluted areas in China, such as Beijing-Tianjin-Hebei, the Yangtze River Delta, the Pearl River Delta, the Fenwei Plain, “You may not use “other” since you also refer to Beijing Tianjin-Hebei.

Response:

We appreciate your comments and apologize for our unprofessional description. Now, the text reads as follows:

Higher concentrations of NSA in PM_{2.5} were also found in regions with more serious air pollution in China, such as Beijing-Tianjin-Hebei, the Yangtze River Delta, the Pearl River Delta, the Fenwei Plain, and the Chengdu-Chongqing region.

Page 5 line 123, Sect 2.1 According to the high NO concentrations, the site could be quite close to adjacent sources. This should be mentioned in the site description.

Response:

We appreciate your comments and apologize for our unprofessional description. As you believe, our observation station is located in the central area of the city, and the contribution of vehicle emissions may also be prominent, so we revised the description of the observation station. Now, it reads as follows:

Comprehensive observations were carried out at the Chengdu comprehensive observation station of atmospheric combined pollution (30.63°N, 104.08°E). The observation equipment was placed on the top of a building, approximately 25 m from the ground, and there was no obvious pollution source within approximately 200 m. The site is located in south section 1 of Yihuan Road, Wuhou District, Chengdu (Fig. 1), and traffic emission sources may be the main pollution emission source around the observation station. This is a typical residential, traffic and commercial mixed area that represents the characteristics of the urban atmospheric environment.

Page 7 line 154 "Temperature (T), relative humidity (RH) and the total concentrations (i.e., gas + aerosol) of Na⁺, SO₄²⁻, NH₃, NO₃⁻, Cl⁻, 155 Ca²⁺, K⁺ and Mg²⁺ were input into the ISORROPIA-II thermodynamic mode" Do you have HCl, and HNO₃ measured? I don't see it in your instrument list. You may need to do a back calculation to check the modelled value and see if you may retrieve these information iterative model calculations. You may need to calculate the uncertainties or bias due to these missing data in your model input.

Response:

We appreciate your comments and apologize for our unprofessional description. Your comment was very helpful for us in improving this research, and it also reminded us to pay attention to the key points when entering data in the ISORROPIA-II thermodynamic model. Therefore, we re-simulated and analysed the the model. In our observation experiment, HCl and HNO₃ were not measured, so the data of these two species could not be obtained. Therefore, in our data input, Cl⁻ and NO₃⁻ are entered, and NH₃ is the total ammonium (NH₃+NH₄⁺) of gas and aerosol inputs. We used this model to analyse the observation and simulation data of NSA in metastable and stable

state conditions in forward mode. We used NH₃ model simulation data and observation data to perform linear regression fitting to verify the model run effect, and the fitting slope of linear regression is 0.96 ($R^2=0.98$), which shows that the model effect is also good, and it can reflect the state of chemical components in aerosols. To reduce the uncertainties or bias caused by missing data, through strict data quality control during data input, we ensured the integrity and validity of each sample data, 618 samples were simulated, and the content of this model in the current manuscript was also re-described.

Page 7 line 159 “The simulated data and observed data were compared and analysed. Simultaneously, the aerosol water content (AWC) and pH of aerosols were calculated. The sensitivity of the interaction between aerosol chemical components (NSA) was analysed (Ding et al., 161 2019; Fountoukis et al., 2009). Could you show a comparison between the modelled and measured gas phase NH₃, HCl and HNO₃? This result can be used to check the reliability and performance of thermodynamic models.

Response:

We appreciate your comments and apologize for our unprofessional description. Through data quality assurance and control, combined with our research purpose, the usage of thermodynamic balance in this manuscript is modified. We agree that it is necessary to analyse the reliability and performance of the output results of the model. Therefore, we performed a comparative analysis of the output data and the observation data. Considering that the observation experiment did not measure HCl and HNO₃, we only analysed the observation and simulation data of NH₃ and compared the observation and simulation data of NSA. In addition, in section 2.4 of the manuscript, we also give an accurate description of the model and the methods used in this study. In response to the current comment, we made the following revision in section 2.4:

The simulated data and observed data were compared and analysed, and the observation data of NH₃ were consistent with the input data of the model. The linear regression fitting slope of NH₃ was 0.96 ($R^2=0.98$), which showed that the run result of the model had good reliability and performance.

Page 8 line 178 “the conditional probability function (CPF) was introduced the R Programming Language.” Complete the sentence.

Response:

We appreciate your comments and apologize for our unprofessional description. Now, the text reads as follows:

We used the conditional probability function (CPF) to analyse the characteristics of pollutants under the influence of wind direction (WD) and wind speed (WS). The analysis results using CPF were obtained using the R programming language, named openair.

Figure 2, why both fractional contribution of both organic and inorganic decrease at high PM_{2.5} concentrations? What’s the other compositions that are increasing?

Response:

We appreciate your comments and apologize for our unprofessional description. We were also initially puzzled by this problem, but by reading many research results, we also obtained a deep understanding. In our research, the proportion of organic (OC and EC) and inorganic (NSA) components in PM_{2.5} is analysed, which will certainly involve a large number of chemical components that have not been calculated and measured, which also reflects the complexity of the composition of PM_{2.5} chemical components. This variation in the chemical composition of PM_{2.5} has also been confirmed in studies in other regions of China. A long-term observation of OC and EC in PM_{2.5} from 2013 to 2018 in Beijing shows that with the accumulation of PM_{2.5} concentration, the concentrations of OC and EC increased, and the proportion of PM_{2.5} decreased (Ji et al., 2019). In a series of research reports on the evaluation of the Air Pollution Prevention and Control Action Plan (2013-2017), in the Chengdu-Chongqing region, the concentration of PM_{2.5} gradually decreased from 2013 to 2015, and the proportion of NSA in PM_{2.5} gradually increased, which also shows that when pollution is aggravated, the chemical composition of higher PM_{2.5} concentrations is more complex, and the unknown component will contribute to a certain quality (Wang et al., 2019). We have revised the corresponding part in the manuscript. Now, it reads as follows:

This phenomenon occurs because some chemical components are included in the statistical analysis. It also reflects that the chemical components of PM_{2.5} have more complex characteristics when pollution is aggravated. Some studies have analysed the changes in the chemical composition of particulate matter in regions with severe pollution in China in recent years, and the results show that the concentration of particulate matter has been significantly reduced, but other components (except NSA and carbonaceous aerosol) have higher contribution characteristics at higher particle concentrations (Geng et al., 2019; Wang et al., 2019). The variation trend of OC, EC and metal elements with increasing PM_{2.5} concentration is similar to that of NSA (Fig. 2c), and this variation trend of OC and EC is consistent with the results of long-term observation research carried out in Beijing (Ji et al., 2019).

Reference:

- Geng, G., Xiao, Q., Zheng, Y., Tong, D., Zhang, Y., Zhang, X., Zhang, Q., He, K., and Liu, Y.: Impact of China's Air Pollution Prevention and Control Action Plan on PM_{2.5} chemical composition over eastern China, *Science China Earth Sciences*, 62, 1872-1884, 10.1007/s11430-018-9353-x, 2019.
- Ji, D., Gao, W., Maenhaut, W., He, J., Wang, Z., Li, J., Du, W., Wang, L., Sun, Y., Xin, J., Hu, B., and Wang, Y.: Impact of air pollution control measures and regional transport on carbonaceous aerosols in fine particulate matter in urban Beijing, China: insights gained from long-term measurement, *Atmospheric Chemistry and Physics*, 19, 8569-8590, 10.5194/acp-19-8569-2019, 2019.
- Wang, Y., Li, W., Gao, W., Liu, Z., Tian, S., Shen, R., Ji, D., Wang, S., Wang, L., Tang, G., Song, T., Cheng, M., Wang, G., Gong, Z., Hao, J., and Zhang, Y.: Trends in particulate matter and its chemical compositions in China from 2013-2017, *Science China Earth Sciences*, 62, 1857-1871, 10.1007/s11430-018-9373-1, 2019.

Page 9 line 216 “The annual average mass concentration of NSA also changed significantly, and the difference was large. The Mann-Whitney U test showed that the variation in NO₃ - was nonsignificant ($p > 0.05$), and SO₄²⁻ and NH₄⁺ had obvious significance from 2015 to 2017 ($p < 0.05$), indicating that NO₃ - had not decreased

significantly, and there was an increase in 2017 compared to 2015.” Here, you could further discuss the reasons why the concentration SO₂ and sulfate decrease more than that of NO_x and nitrate.

Response:

We appreciate your comments and apologize for our unprofessional description. This similar problem has also been raised by *Anonymous Reveiwer #1*. We believe that the current greater emission reduction efforts are due to the implementation of the Air Pollution Prevention and Control Action Plan, and a series of pollution control measures have been implemented. Therefore, we have added in the manuscript the emission reduction and control measures taken by Sichuan Province in recent years to control air pollution. The concentrations of SO₂ and SO₄²⁻ decrease more than those of NO_x and NO₃⁻, indicating that it is necessary to strengthen the air treatment for NO_x emissions. We have made corresponding revisions in the manuscript.

Page 12, line 283 “This also shows that the implementation of air pollution reduction measures should increase the emission reduction intensity in terms of NO_x and NH₃ emissions, especially the implementation of autumn and winter air pollution prevention and control action.” You were talking about high NH₃ in 2017 and then taking to NO_x? I am missing a link here. Also I’d like to see an explanation about the high NH₃ concentration up to 60 ppb. This is very high for a monthly average. What’s the pH under this condition?

Response:

We appreciate your comments and apologize for our unprofessional description. We are here to make a brief summary of the monthly change trend of gaseous pollution; because we have revised a mistake in expression that caused ambiguity. We also noticed that in the second half of 2017, there was a higher NH₃ emission intensity, especially in winter. In combination with Section 3.5, we also found that there will be a higher concentration of pollutants at lower wind speeds (CPF analysis). There may be an emission source in the nearby area that played a more significant role in 2017. In addition, the PSCF analysis also has an obvious regional transport source in the

northeast direction. The contribution in this direction is significantly higher than that in 2015 and 2016. Therefore, we believe that the higher NH₃ emissions in 2017 are affected by local emissions and regional transport. Since chemical transport models were not available in this study to quantitatively analyse the contribution of local emissions and regional transport, it is necessary for us to conduct in-depth research in this area in the future. We recalculated and analysed the ISORROPIA-II thermodynamic equilibrium model through data quality assurance and control. Because the data were not suitable for annual change analysis, the pH value of aerosols at higher NH₃ concentrations in 2017 was not analysed.

Page 13 line 293 "Sulfate has a significant downward trend in all seasons from 2015 to 2017, especially in winter. This downward trend was due to implementation of the Air Pollution 294 Prevention and Control Action Plan" Such discussion should be put in Sect 3.1.

Response:

We appreciate your comments and apologize for our unprofessional description. We have adjusted the position of this discussion and revised the manuscript.

Page 15 line 325 "As shown in Fig. S4, from 9:00 to 11:00 a.m., the concentrations of SO₂, NO_x, NH₃, CO and other gases increased significantly, indicating that the primary emission of pollutants was relatively strong. At this time, higher RH (Fig S5) also provides favourable conditions for the formation of secondary aerosols and promotes the accumulation of NSA" But the RH in Fig. S5 is decreasing in contrast to an increase in aerosol concentrations?

Response:

We appreciate your comments and apologize for our unprofessional description. As you have noticed, RH is indeed decreasing from 9:00 to 11:00. In our analysis, despite the decrease, RH was still relatively high (approximately 65%). Therefore, we have revised and supplemented this description. Now, it reads as follows:

As shown in Fig. S7, from 9:00 to 11:00 a.m., the concentrations of SO₂, NO_x, NH₃

and CO increased significantly, indicating that the primary emission of pollutants was relatively strong. At this time, although RH is in a declining stage, it still has a relatively high atmospheric humidity (approximately 65%), and O₃ and NO₂/NO also occasionally show an increasing trend, indicating that the atmospheric oxidizability has also increased (Figs. S7 and S8). This situation also provides favourable conditions for the formation of secondary aerosols and promotes the accumulation of NSA.

Sect. 3.4.1 In general, it is true that the emissions of multi-pollutant may come from the same kinds of sources. But you cannot draw such a conclusion based on correlation studies. Because the variation of most pollutants, especially those of long-lifetime, is strongly influenced by the boundary layer developments, and may show a similar diurnal variation in spite of different origins (sources).

Response:

We appreciate your comments and apologize for our unprofessional description. We agree with you. During the ACP discussion stage of the manuscript, our research team also carefully reviewed this text and found that the current analysis is not appropriate and may affect the integrity of the manuscript. Therefore, we decided to delete this section.

Page 20 Line 430 “Figure 7 shows the variation characteristics of NSA chemical conversions and meteorological conditions with increasing RH. SOR and NOR increased with increasing RH, suggesting that SO₂ and NO₂ were more likely to produce sulfate and nitrate under higher RH conditions. In Fig. 7, how did you do the calculation, classifying the data according to RH or you keep all input the same but change RH only? In the former case, the apparent correlation with RH may not represent the real causation as chemical compositions and other parameters may change also change.

Response:

We appreciate your comments and apologize for our unprofessional description. We have revised this text. In response to your concerns, we classified and statistically

analysed the variation characteristics of NOR and SOR under different RH conditions according to RH observation data. Regarding the latter comment you raised, we also very much agree with you. The correlation is not enough to explain the relationship between chemical components and other parameters. Therefore, we combined the phase state of the chemical components analysed by the ISORROPIA-II thermodynamic equilibrium model to supplement the analysis.

Sect 3.5.2 I understand that the authors adopted this approach based on a published study. This approach, however, is subject to several problems, e.g., neglecting the dilution of pollution in the course of transport which may overestimate the contribution of distant sources, or the endpoint is not necessary at the ground level, or why 24 hour (aerosols have a longer lifetime) etc. If you still want to keep this part, please explicitly include these caveats in the text to avoid misinterpretation of this result.

Response:

We appreciate your comments and apologize for our unprofessional description. As you pointed out, aerosols do influence of emissions, diffusion, chemical conversions and deposition in the process of regional transport. In our study, PSCF is a kind of conditional probability function relationship. Using Meteorological data from the National Oceanic and Atmospheric Administration (NOAA) to analyse the potential sources of pollution is helpful for explaining the importance of regional joint prevention and control measures for air pollution control. We choose a 24-hour simulation time, mainly considering the following factors. The aerosol spatial distribution characteristics show that there is a high concentration of pollutants in the Sichuan Basin in Southwest China (Gui et al., 2019), and due to the unique topography of the Sichuan Basin, air pollution is obviously affected by the internal emission (Qiao et al., 2019). We agree that the endpoints of the backward trajectory are not on the ground, in fact, as you think. The results of PSCF reflect the potential source of pollution in a plane, rather than the three-dimensional spatial structure characteristics, and the endpoints of the backward trajectory are reflected in the design plane grid, which also better reflects the high-value regional distribution features of PSCF. Therefore, the PSCF reflects the two-

dimensional planar position distribution characteristics of potential sources, not the three-dimensional characteristics that reflect the transmission of pollution. In addition, the aerosol lifetimes of SO₂ (approximately 9.6 d) and NO_x (approximately 1 d) are also very different (Guo et al., 2014), and the research also shows that NH₃ is significantly contributed by local source emissions (Walker et al., 2004). Therefore, we comprehensively considered selecting a 24-hour backward trajectory to carry out PSCF simulation in the Chengdu region. The corresponding supplementary notes have been revised in the manuscript.

Reference:

- Gui, K., Che, H., Wang, Y., Wang, H., Zhang, L., Zhao, H., Zheng, Y., Sun, T., and Zhang, X.: Satellite-derived PM_{2.5} concentration trends over Eastern China from 1998 to 2016: Relationships to emissions and meteorological parameters, *Environmental pollution*, 247, 1125-1133, 10.1016/j.envpol.2019.01.056, 2019.
- Qiao, X., Guo, H., Tang, Y., Wang, P., Deng, W., Zhao, X., Hu, J., Ying, Q., and Zhang, H.: Local and regional contributions to fine particulate matter in the 18 cities of Sichuan Basin, southwestern China, *Atmospheric Chemistry and Physics*, 19, 5791-5803, 10.5194/acp-19-5791-2019, 2019.
- Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng, L., Molina, M. J., and Zhang, R.: Elucidating severe urban haze formation in China, *Proceedings of the National Academy of Sciences of the United States of America*, 111, 17373-17378, 10.1073/pnas.1419604111, 2014.
- Walker, J. T., Whitall, D. R., Robarge, W., and Paerl, H. W.: Ambient ammonia and ammonium aerosol across a region of variable ammonia emission density, *Atmospheric Environment*, 38, 1235-1246, 10.1016/j.atmosenv.2003.11.027, 2004.
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