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,

Xingang Liu and coauthors.
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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

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Response to Anonymous Referee#1

This study provided a good data basis for explaining the pollution characteristics of secondary inorganic aerosols in PM2.5 through a long-term atmospheric observation experiment. The formation mechanism and role of secondary inorganic aerosols during the formation of haze are still a research hot issue. The author not only explained the pollution characteristics of nitrate, sulfate and ammonium in Chengdu, but also analyzed its formation mechanism through observation data and ISORROPIA-II thermodynamic model simulation. Finally, the author also analyzed the distribution characteristics of pollution emissions and potential sources in Chengdu. At present, China is strengthening the control and treatment of air pollution, a long-term atmospheric observation experiment has a high research value for the analysis of the formation of air pollution and the implementation of haze abatement measures. However, there are some writing, grammatical and technical errors in the paper, and it is suggested that the author carefully revise and organize the presentation of the paper.

Response:

We appreciate your comments and have revised the full text; we also regret each error and have discussed and corrected them.

Line. 53-54, PM2.5 interpretation is inaccurate. "PM2.5(aerodynamic diameter less than 2.5 μm)"

Response:

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

particles with aerodynamic equivalent diameter ≤ 2.5 μm in ambient air ($PM_{2.5}$, also known as fine particles)

Line. 58-61, "NSA" is the abbreviation of nitrate, sulfate and ammonium in the paper? please rewrite this sentence.

Response:

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

Nitrate, sulfate, ammonium, organic matter and elemental carbon are the main components of $PM_{2.5}$, among which nitrate, sulfate, and ammonium (NSA) are the main secondary inorganic aerosols in $PM_{2.5}$.

Line. 77-78, suggest reinterpreting this sentence, how to understand the "regional transport"

Response:

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

In addition to the air pollution caused by the local emission of pollutants, the regional transportation of pollutants from its surrounding cities also has an important impact on the urban air quality.

Line. 82-83, if the author defines an abbreviation at the beginning of the paper, it is recommended to use the abbreviation below. Please use "NSA" abbreviations instead of nitrates, sulfates and ammonium.

Response:

We appreciate your comments and have revised the manuscript. Now, it 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.

Line. 87, please correct this writing, "(2013-207)"

Response:

We appreciate your comments and have revised the text. Now, it reads as follows:

In response to this situation, the Chinese government issued an Air Pollution Prevention and Control Action Plan (2013-2017) in 2013 to reduce pollutant emissions and improve air quality.

Line. 106-109, "high time resolution", what's the meaning of this?

Response:

We appreciate your comments and apologize for our unprofessional description. We mean that the time interval for gathering observation data is relatively small, at 1 hour. Compared with the daily average data of manual operation sampling, the time resolution is higher; we have revised and polished this sentence. Now, it reads as follows:

However, these analyses may be affected by the experimental equipment, observation stations and other conditions, and the time span of these atmospheric observations usually includes several pollution processes or lasts for weeks or months. Thus, it is difficult to analyse the long-term variations in characteristics of air pollution through comprehensive observation. In particular, there are few high-time-resolution (1 hour) observation experiments carried out with online automatic observation systems.

Line. 141, in Table 1, parameters not covered in this paper can be removed, such as PM1 and H2S

Response:

We appreciate your comments and have revised it. Now, it reads as follows:

Table 1. The experimental instruments used in this study

Instruments	Parameters	Manufacturer/Country
URG-9000	$NO_3^-/SO_4^{2-}/NH_4^+/Na^+/Mg^{2+}/Ca^{2+}/Cl^-/K^+$	Thermo Fisher Scientific/USA
SHARP 5030	$PM_{2.5}$	Thermo Fisher Scientific/USA
RT-4	OC/EC	Sunset Laboratory/USA
Xact-625	Metal elements	Cooper Environmental Services /USA
17i/450i/48i/49i	$NOx/NO_2/NO/NH_3/SO_2/CO/O_3$	Thermo Fisher Scientific/USA
WXT520	Meteorological parameters	VAISALA/Germany

OC: organic carbon; EC: element carbon

Line. 185-186, percentile (e.g. 0-25, 25-50, 50-75 and 75-100), please confirm it is consistent with the title of Fig. 13-15 (0-25%, 25-50%, 50-75%, and 75-100%.) in Supplementary materials.

Response:

We appreciate your comments and have revised the text in the Supplementary Materials. We have made the writing consistent throughout the manuscript and adopted percentiles (e.g., 0-25, 25-50, 50-75 and 75-100), and the "%" in the figure title was deleted.

Line. 185-186, it is suggested that the author briefly describe what measures should be taken.

Response:

We appreciate your comments. There is no description of "measures" in line 185, and we presume that the reviewer refers to the description in lines 228-230 about the measures taken in the Air Pollution Prevention and Control Action Plan. We also describe this part in detail. Now, it reads as follows:

From 2015 to 2017, the measures taken by Sichuan Province in the coordinated reduction of multiple pollutants have been continuously strengthened, and the scope of management and control has been continuously expanded, for example, in the improvement of desulfurization, denitrification and dust removal technologies in key industries, from accelerated improvement in 2015 to deeper improvement in 2017. The

process of eliminating small coal-fired boilers began in 2015 and was completed in 2017, when the ultra-low-emission coal-fired power plant transformation was promoted. In terms of vehicle emission control, we accelerated the elimination of "yellow label" vehicles (general term for gasoline vehicles with emission levels lower than the national I emission standard and diesel vehicles with emission levels lower than the national III emission standard when new vehicles are finalized) and “old vehicles” (the emission level does not meet the national stage IV emission standard) in 2015 and basically completed the elimination of yellow standard vehicles in 2017. The quality supervision of oil products has also been improved, and non-road mobile machinery pollution control requirements were proposed in the 2017 plan (ThePeople'sGovernmentofSichuanProvince, 2015, 2016, 2017).

References:

The People's Government of Sichuan Province. Detailed rules for the implementation of the action plan for the prevention and control of air pollution in Sichuan Province 2015 annual implementation plan. Website：http://www.sc.gov.cn/10462/10883/11066/2015/4/22/10333390.shtml，last access: June 17 2020.

The People's Government of Sichuan Province. Detailed rules for the implementation of the action plan for the prevention and control of air pollution in Sichuan Province 2016 annual implementation plan. Website：http://www.sc.gov.cn/zcwj/xxgk/NewT.aspx?i=20160401095908-612769-00-000，last access: June 17 2020.

The People's Government of Sichuan Province. Detailed rules for the implementation of the action plan for the prevention and control of air pollution in Sichuan Province 2017 annual implementation plan. Website：http://www.sc.gov.cn/zcwj/xxgk/NewT.aspx?i=20170527091543-450025-00-000，last access: June 17 2020.

Line. 263, “These variations have similar trends due to meteorological factors”, what do you mean?

Response:

We appreciate your comments and apologize for our unprofessional description. What we want to say is that the meteorological conditions also have obvious monthly variation characteristics, which may have some influence on the variation characteristics of NSA. Now, the text reads as follows:

The monthly variation characteristics of NSA from 2015 to 2017 are shown in Fig. 3. At the beginning and end of each year, the pollutant concentration is relatively high and relatively low in the middle of each year. The meteorological conditions also have obvious monthly variation characteristics (Fig. S5 a and b); from April to August, they have higher WS and lower RH, which is not only conducive to the dilution and diffusion of pollutants but also reduces the chemical conversions of pollutants by aqueous phase and influences the formation of secondary inorganic aerosols.

Line. 306, the legend in Fig. 3 is suggested to be modified, with one reserved, and also pay attention to modify other pictures, such as Fig.S2 and S8.

Response:

We appreciate your comments and apologize for our unprofessional description. We reviewed similar problems in the other figures and have corrected them.

Line. 309, replace "daily changes" with "diurnal variation".

Response:

We appreciate your comments and have revised the text.

Line. 400, please explain what "r" in Fig. 5 means?

Response:

We appreciate your comments and apologize for our unprofessional description. This text has been revised in the manuscript, and this error will not appear in the new manuscript.

Line. 452, in Fig.7h, "SO42- gas-particle phase partitioning"? inconsistent with the

NH4+ in the picture.

Response:

We appreciate your comments and apologize for our unprofessional description. The analysis content has been modified in this section, and the comments made by you in the manuscript have also been resolved.

Line. 470, please explain what "k" in Fig. 8 means?

Response:

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

Fig. 8. Molar ratio analysis of NSA (nitrate, sulfate and ammonium). (a) Interannual variation in the molar ratio of SO_4^{2-} and NH_4^+. (b) Interannual variation in the molar ratio of NO_3^- and NH_4^+. (c) Seasonal variation in the molar ratio of SO_4^{2-} and NH_4^+. (d) Seasonal variation in the molar ratio of NO_3^- and NH_4^+. k: Fitting slope of linear regression.

In Section 3.5.2, authors are advised to supplement PSCF analysis of NO2 and NO.

Response:

We appreciate your comments and have revised the text.

Line. 470, modify the Fig.9, remove the repeat "PSCF" in the picture

Response:

We appreciate your comments and apologize for our unprofessional description. This issue is also associated with the previous issue, and we have solved the problem.

Line. 521-525, the description is too simple, please rewrite the research results.

Response:

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

The results of using the ISORROPIA-II thermodynamic equilibrium model to simulate

NO_3^-, SO_4^{2-} and TNH_3 emission reduction control effects of 5%, 10%, 15% and 20%, respectively, are shown in Table S3, showing that controlling the concentration of NO_3^- and SO_4^{2-} is also helpful to reduce the concentration of NH_4^+ and indicating that controlling its precursor NOx and SO_2 is of great significance to reduce the secondary inorganic aerosol in $PM_{2.5}$ (the detailed results are described in the supplementary materials).

supplementary materials

Through observation data quality control, we screened 618 sample input ISORROPIA-II thermodynamic equilibrium models to ensure the integrity of the samples and the effectiveness of the data. The control variable method was used to explore the impact of a concentration reduction for other species. For example, to explore the impact of NO_3^- concentration reduction for SO_4^{2-} and NH_4^+, the NO_3^- data were calculated based on the 5, 10, 15 and 20% emission reduction ratio, and other parameters were input into the model using the observation data to explore the relative variable of SO_4^{2-} and NH_4^+ concentration. The simulation results are shown in Table S3. When only NO_3^- and SO_4^{2-} were reduced, NH_4^+ was significantly reduced, but the changes in SO_4^{2-} and NO_3^- were not obvious, and the relative variables of approximately 12% and 7% may be mainly affected by the change in phase state. When only TNH_3 was controlled, the relative variable of SO_4^{2-} was not obvious, and the concentrations of NO_3^- and NH_4^+ decreased, but the relative variable was not large. NSA has a good reduction effect under synergistic emission reduction control. The results show that reducing the amount of NO_3^- and SO_4^{2-} can not only reduce their concentrations but also help to reduce the concentration of NH_4^+. It also suggests that controlling the gaseous precursors NOx and SO_2 is of great significance to reduce the amount of secondary inorganic aerosol in $PM_{2.5}$. Studies in Mexico City have also shown that reducing total sulfates and total nitrates rather than total ammonium helps reduce $PM_{2.5}$ concentrations in an ammonium-rich environment (Fountoukis et al., 2009).

Table S3. Simulation of NO_3^-, SO_4^{2-} and TNH_3 emission reduction control effect (%) and its influence on pH based on the ISORROPIA-II thermodynamic model.

Reduction	Only NO_3^- Reduction				Only SO_4^{2-} Reduction			
	NO_3^-	SO_4^{2-*}	NH_4^+	pH*	NO_3^-	SO_4^{2-*}	NH_4^+	pH
5%	11.92	12.25914	8.33	4.0495	7.19	17.1088	9.77	4.08
10%	16.58	12.25911	11.13	4.0519	7.09	21.9593	13.65	4.13
15%	21.23	12.25909	13.91	4.0546	7.00	26.8093	17.50	4.19
20%	25.88	12.25906	16.69	4.0547	6.91	31.6596	21.58	4.25
	Only TNH_3 Reduction				Synergistic **			
	NO_3^-	SO_4^{2-*}	NH_4^+	pH	NO_3^-	SO_4^{2-*}	NH_4^+	pH
5%	7.51	12.25938	5.85	4.02	12.08	17.1090	12.86	4.07
10%	7.79	12.25965	6.20	3.99	16.85	21.9596	19.80	4.09
15%	8.10	12.25998	6.59	3.95	21.64	26.8097	26.17	4.11
20%	8.45	12.26040	7.03	3.91	26.37	31.6601	33.29	4.16

Notes: NO_3^-, SO_4^{2-} and TNH_3 are the concentration variables relative to the observation data;

pH is the average; TNH_3: $NH_3 + NH_4^+$;

*: In order to display the data difference, the number of digits after the decimal point was increased

**: NO_3^-, SO_4^{2-} and TNH_3 decreased in the same proportion

Reference:

Fountoukis, C., Nenes, A., Sullivan, A., Weber, R., Van Reken, T., Fischer, M., Matias, E., Moya, M., Farmer, D., and Cohen, R. C.: Thermodynamic characterization of Mexico City aerosol during MILAGRO 2006, Atmospheric Chemistry and Physics, 9, 2141-2156, 10.5194/acp-9-2141-2009, 2009.

Authors are requested to write rules uniformly. It is not recommended to use abbreviations in the title of the figures, such as NSA, AWC, SOR, NOR PSCF. In addition, there are "(a)", "(b)" and "(c)" in the picture, please explain what it means in the title.

Response:

We appreciate your comments and apologize for our unprofessional description. We have checked and corrected the abbreviations for consistency throughout the

manuscript.

In Fig. 5 and Fig. S4, please confirm the carbon monoxide (CO) unit, "ppb" and "ppm"?

Response:

We appreciate your comments and apologize for our unprofessional description. The unit of carbon monoxide (CO) is ppm, and we have revised it.

In Supplementary materials, Line. 90 and 96, pay attention to writing, it shouldn't be "2107"

Response:

We appreciate your comments and have revised it.

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,

Xingang Liu and coauthors.
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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

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Response to Anonymous Referee#2

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.

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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 rate of $PM_{2.5}$ data in Fig. S1 is 6.8%. 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.

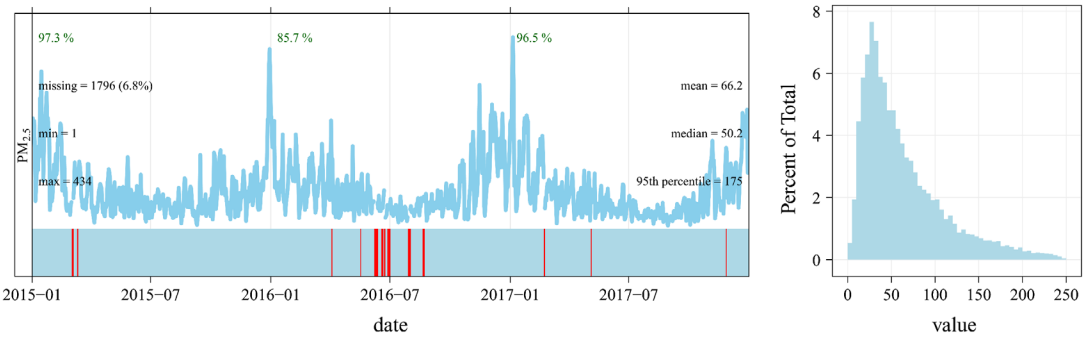

Fig. S1. $PM_{2.5}$ data quality assurance and control.

Fig. S2. NO_3^-, SO_4^{2-}, NH_4^+, OC (organic carbon) and EC (element carbon) data quality assurance and control.

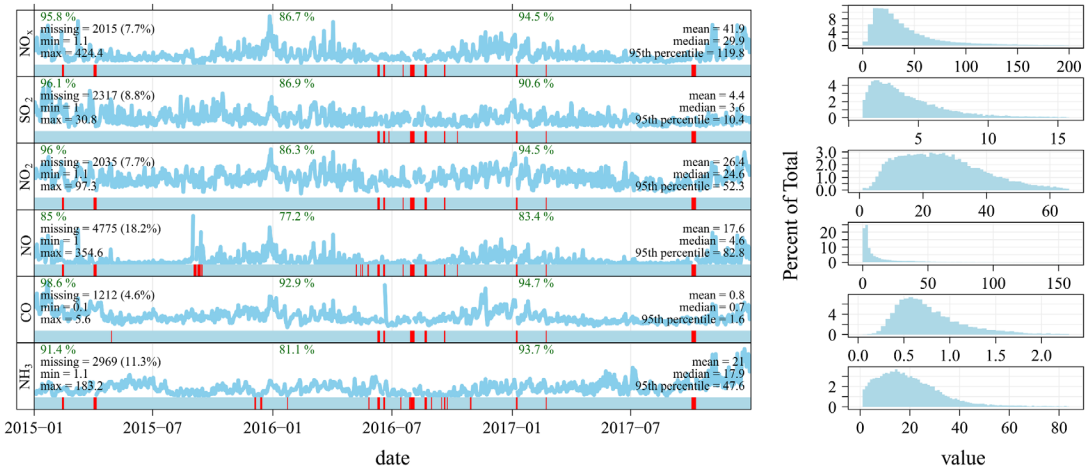

Fig. S3. NOx, SO_2, NO_2, NO, CO and NH_3 data quality assurance and control.

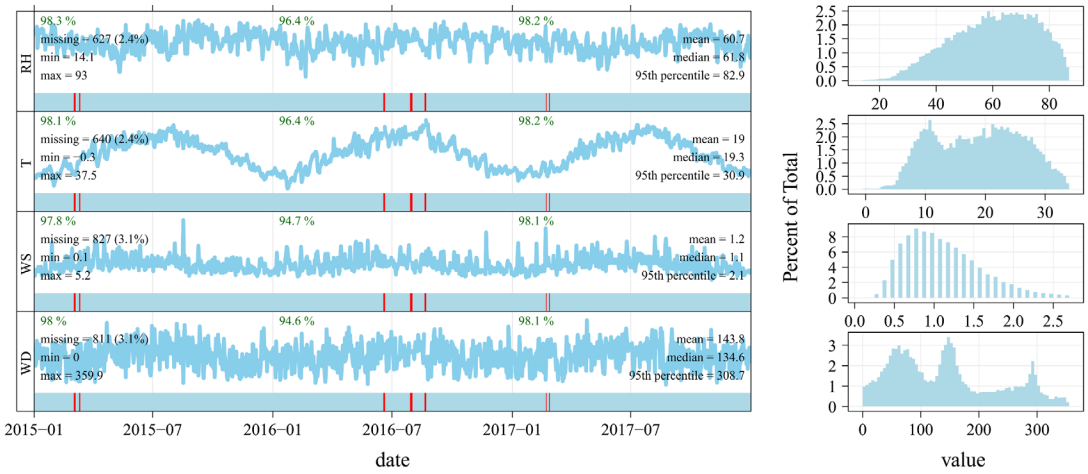

Fig. S4. Relative humidity (RH), temperature (T), wind speed (WS) and wind direction

(WD) data quality assurance and control.

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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 $PM_{2.5}$ 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 NO_2, SO_2 and NH_3 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 PM2.5 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 HNO_3 were not measured, so the data of these two species could not be obtained. Therefore, in our data input, Cl^- and NO_3^- are entered, and NH_3 is the total ammonium (NH_3+NH_4^+) 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 NH_3 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 HNO_3, we only analysed the observation and simulation data of NH_3 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 NH_3 were consistent with the input data of the model. The linear regression fitting slope of NH_3 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 PM2.5 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.

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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 SO4 2- 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 decease more than that of NOx 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 SO_2 and SO_4^{2-} decease more than those of NOx and NO_3^-, indicating that it is necessary to strengthen the air treatment for NOx

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 NOx 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 NOx? 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 NH_3 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 NH_3 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 NH_3 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, NOx, 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 SO_2, NOx, NH_3 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 O_3 and NO_2/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 SO_2 and NO_2 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 SO_2 (approximately 9.6 d) and NOx (approximately 1 d) are also very different (Guo et al., 2014), and the research also shows that NH_3 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:

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Elucidating the pollution characteristics of nitrate, sulfate and ammonium in $PM_{2.5}$ in Chengdu, southwest China, based on ~~long-term~~three-year ~~observations~~measurements

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Abstract

Nitrate, sulfate and ammonium (NSA) are the main secondary inorganic aerosols of $PM_{2.5}$ and play an important role in the process of air pollution. ~~However, few studies have analysed the variation characteristics of NSA in $PM_{2.5}$ and the effects of control measures through long-term observations.~~ In this study, a ~~l~~three-year~~long-term~~ observational experiment was conducted from January 1, 2015 to December 31, 2017 in Chengdu, southwest China. NSA pollution characteristics, chemical conversion generation, emission reduction control sensitivity analysis and pollutant regional transport characteristics were analysed. NSA is the most important chemical components of particles with aerodynamic equivalent diameter ≤ 2.5 μm in ambient air ($PM_{2.5}$), and the contribution of nitrate to the accumulation of $PM_{2.5}$ concentration is greater than that of sulfate and ammonium. NSA also have obvious characteristics of

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annual, monthly, seasonal, diurnal and weekly variations. Through observation data and model simulation, it was also found that the existence of an aerosol aqueous environment plays an important role in the formation and existence of NSA. Sensitivity analysis between NSAs found that controlling NO_3^- and SO_4^{2-} play an important role in reducing the contribution of NSA to $PM_{2.5}$, which also implies that the current control of NOx and SO_2 is important for improving air pollution.

~~The concentrations of sulfate and ammonium in $PM_{2.5}$ have been well reduced, but the effect of reducing nitrate was not obvious. 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. Although the workday concentration was slightly higher than the weekend concentration, the difference was nonsignificant.~~

~~The chemical conversion characteristics of NSA formation were comprehensively analysed,~~

~~and the aqueous phase oxidation process plays an important role in the conversion of NOx, SO_2 and NH_3 to NSA. The ammonia-rich environment became increasingly obvious in the atmosphere of Chengdu. Under these conditions, the sensitivity of NSA concentration variation was analyses using the ISORROPIA-II thermodynamic model, and the results show that by reducing NOx and SO_2 emissions, not only can reduce the nitrate and sulfate in $PM_{2.5}$, but also help reduce the formation of ammonium nitrate and ammonium sulfate to reduce ammonium. The results also show that while carrying out NSA emission reduction, it is also possible to generate potential risks of changes in aerosol pH.~~

Combined with meteorological conditions and ~~a~~potential source contribution function (PSCF) analysis, local emissions and regional emissions of pollutants are found to have important impacts on Chengdu's atmospheric environment. This research result not only provides an assessment of the current atmospheric emission reduction effect but also provides an important reference for ~~determining methods to further reduce the NSA~~

~~concentration in~~ atmospheric ~~$PM_{2.5}$~~pollution control.

Keywords: Secondary inorganic aerosols; Three-year~~Long-term~~ observations; Pollution characteristics; Chemical conversions; Source analysis; Chengdu

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1 Introduction

In recent years, with the rapid development of China's domestic economy and acceleration of the urbanization process, energy consumption and pollutant emissions have also increased, which increases the burden on the atmospheric environment, and severe air pollution has become ~~the~~ a focus of social concern (Liu et al., 2013a;An et al., 2019;Fu et al., 2014;Zhao et al., 2017). When air pollution forms, ~~$PM_{2.5}$ (aerodynamic diameter less than 2.5 μm)~~ mass concentrations of particles with aerodynamic equivalent diameter ≤ 2.5 μm in ambient air ($PM_{2.5}$, also known as fine particles) can reach a higher pollution level, which not only reduces atmospheric visibility but also carries a large number of toxic species into the human lungs, increasing the risks of cardiovascular and cerebrovascular diseases~~, as well as harming human health~~ (Chang et al., 2018;Tie et al., 2009;Kong et al., 2019;Zhao et al., 2018;Yang et al., 2015a). Nitrate, sulfate, ammonium, organic matter and elemental carbon are the main components of $PM_{2.5}$, among which nitrate, sulfate~~, nitrate~~ and ammonium (NSA) are the main secondary inorganic aerosols in $PM_{2.5}$ (Ji et al., 2019;Zheng et al., 2016). NSA mainly originates from the secondary aerosols produced by complex chemical reactions of NOx, SO_2 and NH_3 from coal combustion, vehicle exhaust emissions and agricultural sources (Liu et al., 2013b;Wang et al., 2016;Tian et al., 2017).

Because China's current main energy ~~consumption~~ resource is still fossil fuels, which are widely used in industry, for vehicles and residentially, the emission reduction space of NSA is still restricted by a large number of gaseous precursors of NSA (Zhao et al., 2018;Tong et al., 2019). In addition, the chemical conversion of NO_2, SO_2 and NH_3 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~~For example, photochemistry may affect the formation of NSA~~

~~at high solar radiation, and the homogeneous reaction may dominate the formation of SNA in high relative humidity~~ (Cheng et al., 2016;Sun et al., 2014;Wang et al., 2016;Ohta and Okita, 1990;He et al., 2014). The formation of sulfate can increase the acidity of aerosols (Sun et al., 2014). In contrast, the presence of NH_3 can play a role in neutralization and maintain the acid-base balance of aerosols (Wang et al., 2016). If improper control measures are taken in pollution reduction control, such as further ammonia emission reduction, the acidification of aerosols and environmental problems of acid rain may be aggravated ~~are the likely result~~ (Liu et al., 2019c). In addition to the air pollution caused by the local emission of pollutants, the regional transportation of pollutants from its surrounding cities also has an important impact on the urban air quality~~In addition to the local emission of pollutants, regional transport is also an important influencing factor~~. Determination of regional transport sources of pollutants, taking regional joint prevention and control measures, and jointly reducing the emissions of pollutants will enable better air control effects, particularly in the Beijing-Tianjin-Hebei region of northern China (Chen et al., 2019a).

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~~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, Chengdu-Chongqing region~~ (An et al., 2019;Li et al., 2017;Liu et al., 2019d). In response to this situation, the Chinese government issued an Air Pollution Prevention and Control Action Plan (2013-2017) in 2013 to reduce pollutant emissions and improve air quality (the Sate Council~~MinistryofEcologyandEnvironmentofthePeople'sRepublicofChina~~, 2013, last access: June 17, 2020). A large number of treatment measures have been taken in coal combustion, motor vehicle emissions and ~~phase out~~ outdated industrial capacities, and by 2017, China's ambient air quality control measures have achieved good results (Liu et al., 2019a;Chen et al., 2019b;Cheng et al., 2019;Li et al., 2019a). In Beijing, $PM_{2.5}$, NO_2 and SO_2 decreased by 35.2%, 17.9% and 69.8%, respectively, in 2017 compared

with 2013 (Beijing Municipal Ecology and Environment Bureau, 2018, last access: June 17, 2020~~BeijingMunicipalEcologyandEnvironmentBureau, 2017~~). In Chengdu, $PM_{2.5}$, NO_2 and SO_2 decreased by 42.3%, 15.9% and 64.5%, respectively, in 2017 compared with 2013 (Chengdu Municipal Ecology and Environment Bureau, 2018, last access: June 17, 2020~~ChengduMunicipalEcologyandEnvironmentBureau, 2017~~). To continue to promote air quality improvement, the Chinese government launched the " Three-Year Action Plan for Winning the Blue Sky Defense Battle" in 2018, which puts forward stricter requirements on how to further promote the implementation of emission reduction plans (the Sate Council, 2018, last access: June 17, 2020~~theSateCouncil, 2018~~). Through long-term~~long-term~~ observations, a comprehensive analysis of $PM_{2.5}$ chemical composition and source characteristics is carried out to verify the current implementation effects of emission reduction, and in-depth analyses of pollution reduction control characteristics ~~is~~ are of great significance for the next step in air pollution control. However, these analyses may be affected by the experimental equipment and observation station and other conditions, and the time span of these atmospheric observations usually includes several pollution processes or last for weeks or months. Thus, it is difficult to analyse the long-term variations characteristics of air pollution through comprehensive observation. In particular, there are few high time resolution (1 hour) observation experiments carried out by online automatic observation system~~However, observations with high time resolution are very rare, and the time period of these atmospheric observations usually includes several pollution processes or last for weeks or months; thus, it is difficult to analyse the long-term change characteristics of air pollution through comprehensive observational means~~ (Sun et al., 2013;Tie et al., 2017;Guo et al., 2014). Especially in the Sichuan Basin of Southwest China, there are few long-term observational experiments on NSA, which is the main chemical component of $PM_{2.5}$.

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The Sichuan Basin is among the most important areas of air pollution in China (Qiao et al., 2019;Gui et al., 2019;Zhong et al., 2019). Although there are many studies in this area, there are few long-term ~~view~~ studies of the hourly concentration data resolution of $PM_{2.5}$ chemical components. In this study, through ~~long-term~~three years of

observations (from ~~1~~ Januar1, y 2015 to ~~31~~ Decembe r 31, ~~r~~ 2017), we analysed the pollution ~~level and chemical conversion~~ characteristics of NSA, , as well as their formation mechanism and pollution control sensitivity. Finally, combined with local emissions and regional transport characteristics, we analysed the air pollution transport characteristics of Chengdu air pollution.~~in $PM_{2.5}$ in Chengdu and the concentration change sensitivities of nitrate, sulfate and ammonium. Finally, combined with local emissions and regional transmission characteristics, we analysed the regional transport characteristics of Chengdu air pollution.~~

2 Experiment and methods

2.1 Observation site

Comprehensive observations were carried out at the Chengdu comprehensive observation station of atmospheric combined pollution~~Super Station of Atmospheric Environmental Monitoring of Chengdu Academy of Environmental Protection Sciences~~ (30.~~65~~63°N, 104.~~05~~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 ~~the Wuhou District of Chengdu,~~ south section 1 of Yihuan Road, Wuhou District, Chengdu ~~between First Ring Road and Second Ring Road~~ (Fig. 1), and traffic emission source may be the main pollution emission source around the observation station. This is a typical residential, ~~transportation~~ traffic and commercial mixed area that represent the characteristics of the urban atmospheric environment. Chengdu is also a megacity in the Sichuan Basin of Southwest China, as well as an important part of the Chengdu-Chongqing region, which is among the regions with serious air pollution in China, and as shown in Fig. 1, Sichuan basin also has high aerosol optical thickness (AOD) ~~(Fig. 1)~~.

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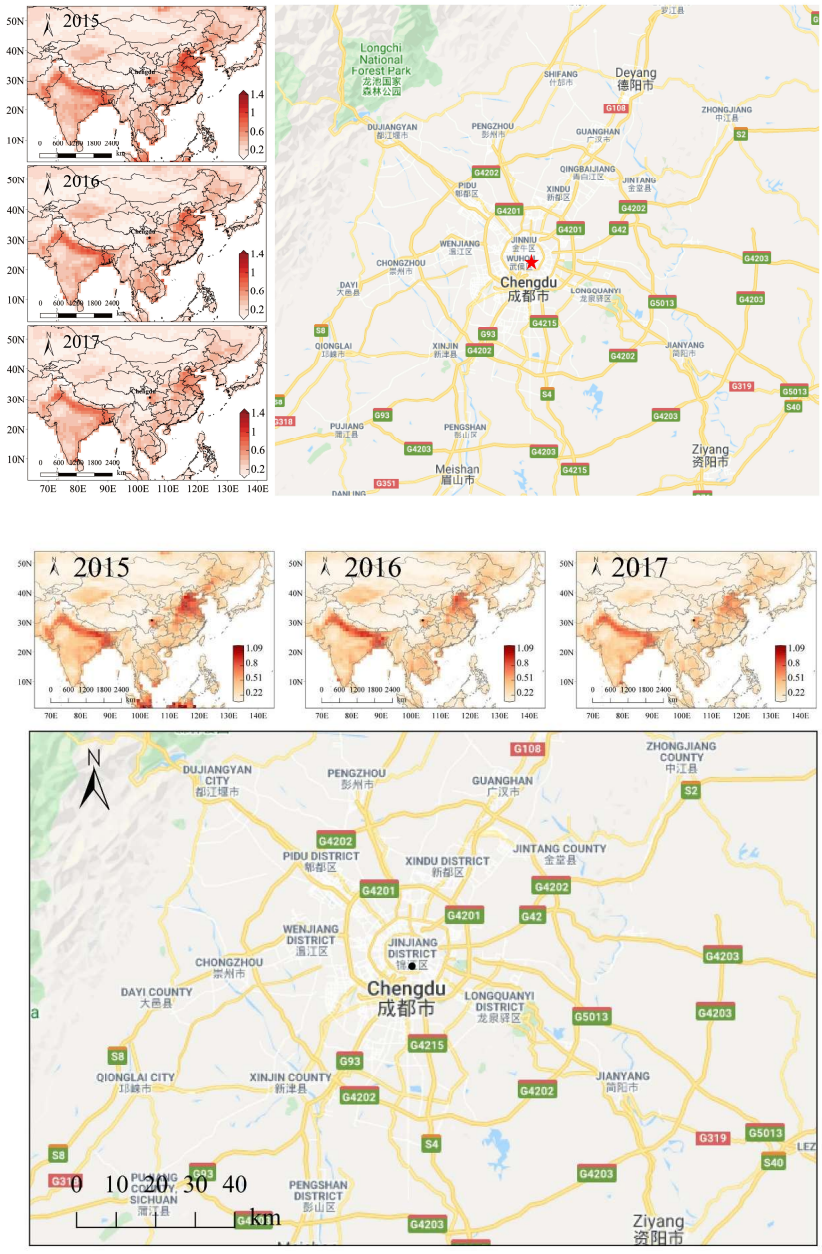

Fig. 1. Observation site in Chengdu. The image on the ~~left~~ top shows the aerosol optical depth (AOD, 550 nm) in from 2015 to 2017 ((National Aeronautics and Space

Administration, 2019, last access: June 17, 2020). The black dot in the image on the bottom shows the location of the observation site in Chengdu (Google Maps 2020, last access: June 17, 2020).~~National Aeronautics and Space Administration, 2019, Last access: 12 February 2020). The red star in the image on the right shows the location of the observation site in Chengdu (Google Maps 2020) (NationalAeronauticsandSpaceAdministration, 2019).~~

2.2 Instruments

During the research period, online experimental monitoring instruments were used to obtain the observation data with an hourly resolution (1 hour). The equipment list is shown in Table 1. Data quality control and assurance is an important part of the atmospheric comprehensive observation experiment, and this result is described in detail in the supplementary materials (Fig. S1-4).

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Table 1. The experimental instruments used in this study

Instrument Model	Parameters	Manufacturer/Country
URG-9000	NO_3^-/SO_4^{2-}/NH_4^+/Na^+/Mg^{2+}/Ca^{2+}/Cl^-/K^+	Thermo Fisher Scientific/USA
SHARP 5030	$PM_{2.5}$~~/PM_1~~	Thermo Fisher Scientific/USA
RT-4	OC/EC	Sunset Laboratory/USA
Xact-625	Metal elements~~Fe/Mn~~	Cooper Environmental Services ~~CES~~/USA
17i/450i/~~17i/42iY/~~48i/49i	~~SO_2/H_2S/~~NOx/NO_2/NO/NH_3/SO_2/CO/O_3	Thermo Fisher Scientific/USA
WXT520	Meteorological parameters	VAISALA/Germany

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OC: organic carbon; EC: element carbon

2.3 Chemical conversions and model methods

To clarify the conversion of gaseous pollutants to secondary aerosols, the ~~sulfur oxidation ratio (SOR~~nitrogen oxidation ratio (NOR)~~)~~ and sulfur oxidation ratio (SOR) ~~nitrogen oxidation ratio (NOR)~~ were used to reflect the conversions of NO_2~~SO_2~~ and SO_2 ~~NO_2~~ to ~~sulfate~~ NO_3^- and SO_4^{2-}~~ and nitrate~~, respectively (Sun et al., 2014;Yang et al., 2015a). ~~They~~ These ratios can be calculated using Eq. (1) and Eq. (2):

$NOR=nNO_3^-/(nNO_3^-+nNO_2)$ (~~2~~1)

$SOR=nSO_4^{2-}/(nSO_4^{2-}+nSO_2)$ (~~1~~2)

~~$NOR=nNO_3^-/(nNO_3^-+nNO_2)$ (2)~~

where n is the molar concentration.

The ISORROPIA-II thermodynamic model was used to analyse the variation characteristics ~~of the interaction~~ among aerosol chemical components (Fountoukis and Nenes, 2007;Guo et al., 2017a;Ding et al., 2019). Temperature (T), relative humidity (RH) and the ~~total~~ concentrations ~~(i.e., gas + aerosol)~~ of Na^+, SO_4^{2-}, NH_3, NO_3^-, Cl^-, Ca^{2+}, K^+ and Mg^{2+} were input into the ISORROPIA-II thermodynamic equilibrium model. In this study, we use "forward problems" mode to run the model, assuming that the aerosols were in a "metastable" state (salts do not precipitate under supersaturated conditions). At the time of data input, NH_3 data were the sum of NH_3 and NH_4^+. Previous studies had shown that the model has better performance when the RH is greater than 30%, and some studies believe that the model performance is greater than 40%, so this study maintains the RH at higher than 40% when data are input (Ding et al., 2019;Guo et al., 2016). The simulated data and observed data were compared and analysed, and the observation data of NH_3 was consistent with the input data of the model. The linear regression fitting slope of NH_3 was 0.96 (R^2=0.98), which shows that the run result of the model hasd good reliability and performance (Ding et al., 2019). Simultaneously, the aerosol water content (AWC) was calculated, and the sensitivity of the interaction between aerosol chemical components (NSA) and pH of aerosols ~~were~~ was analysed calculated~~. The sensitivity of the interaction between aerosol chemical components (NSA) was analysed~~ (Ding et al., 2019;Fountoukis et al., 2009). The pH ~~can be~~was calculated using Eq. (3):

$$pH = -\log_{10} H_{aq}^+ \cong -\log_{10} \frac{1000 H_{air}^+}{AWC} \quad (3)$$

where H_{aq}^+ (mol/L) is the concentration of hydronium ions in liquid water of atmospheric particulate matter, which can be calculated by the H_{air}^+ and AWC (μg/m^3) outputs from the ISORROPIA-II thermodynamic equilibrium model (Ding et al.,

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2019;Guo et al., 2017a).

~~Gas-particle phase partitioning can be used to describe the transformation characteristics between semivolatile inorganic salts (NO_3^- and NH_4^+) and corresponding gases (HNO_3 and NH_3) , which can be calculated by Eq. (4) and Eq. (5):~~

~~$$c(NO_3^-) = \frac{NO_3^-}{HNO_3+NO_3^-} \quad (4)$$~~

~~$$c(NH_4^+) = \frac{NH_4^+}{NH_3+NH_4^+} \quad (5)$$~~

~~where the units of NO_3^-, NH_4^+, NH_3 and HNO_3 were μg/m^3, and the HNO_3 data are from the ISORROPIA-II thermodynamic model output.~~

2.4 CPF and PSCF methods

We use conditional probability function (CPF) to analyse the action characteristics of pollutants under the influence of wind direction (WD) and wind speed (WS). The analysis result using CPF were obtained using the R programming language, named openair. ~~To analyse the relationship between pollutants and wind direction (WD) and wind speed (WS), the conditional probability function (CPF) was introduced the R Programming Language.~~ This function can be defined as CPF= $m_{\theta,j}/n_{\theta,j}$, where $m_{\theta,j}$ is the number of samples in the WD interval θ and WS interval j with mixing ratios greater than some~~, given a~~ 'high' pollution concentration (percentile of pollutants), and $n_{\theta,j}$ is the total number of samples in the same ~~wind direction-speed~~WD-WS interval (Uria-Tellaetxe and Carslaw, 2014). Usually, a higher given 'high' pollution concentration (percentile) is chosen, such as the 90th percentile, which will mask the lower percentile pollution concentration source contributions. In this work, to obtain a more complete contribution of pollution sources, a range of percentile values, ~~were selected for the CPF calculation, e.g.~~ 0-25, 25-50, 50-75 and 75-100 were selected for the CPF calculation.

The potential source contribution function (PSCF) is based on the analysis of pollution sources given~~, which is based on~~ the airmass backward trajectory and can be used to judge the long-distance regional transport of pollutants (Ji et al., 2019). In this study, ~~Meteoinfomap~~ MeteoinfoMap and TrajStat (Wang et al., 2009) were used, and the model simulation~~Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT)~~

data input model were provided by the National Oceanic and Atmospheric Administration (National Oceanic and Atmospheric Administration, 2019, last access: June 17, 2020~~National Oceanic and Atmospheric Administration, 2019, last access: 12 February 2020) (NationalOceanicandAtmosphericAdministration, 2019~~); these data were calculated to the 24-hr backward trajectories at the observation site at a height of 500 m every 1 hour from ~~1~~ January 1, 2015 to ~~31~~ December 31, 2017 (UTC+8). The calculated domain for PSCF was a range of 20-50° N, 75-115° E, and a grid cell with a resolution of 0.5°×0.5° was divided. The PSCF can be defined as Eq. (6):

$$PSCF_{ij} = \frac{M_{ij}}{N_{ij}} W_{ij} \quad (6)$$

$$W_{ij} = \begin{cases} 1.0 & (N_{ij} \geq 3N_{ave}) \\ 0.7 & (3N_{ave} > N_{ij} \geq 1.5N_{ave}) \\ 0.4 & (1.5N_{ave} > N_{ij} \geq N_{ave}) \\ 0.2 & (N_{ave} > N_{ij}) \end{cases} \quad (7)$$

where $PSCF_{ij}$ is the value for the ijth grid cell, M_{ij} is the total number of endpoints in the ijth grid cell, with pollution concentrations at the observation site (30.~~65~~63°N, 104.~~05~~08°E) that are greater than a given threshold value (the 75 percentiles ~~are~~ was selected for gaseous pollutants). N_{ij} is the number of backward trajectory endpoints that fall in the ijth grid cell (0.5°×0.5°) during the simulation period. 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. To reduce the uncertainty in N_{ij}, an empirical weight function W_{ij} was introduced in Eq. (7), where N_{ave} is the average of N_{ij} during the simulation period (Ji et al., 2019;Zhang et al., 2017;Wang et al., 2009).

3 Results and discussion

3.1 Pollution characteristics of the interannual and entire observation periods

The annual average mass concentration of NSA and its proportion in $PM_{2.5}$ are shown in Table 2. The annual averages of $PM_{2.5}$ were 67.78, 71.88 and 59.68 μg/m^3, corresponding to 2015, 2016 and 2017, respectively. However, the pollution of $PM_{2.5}$ in Chengdu was much higher than the annual secondary guideline value (35 μg/m^3, Ambient air quality standards/GB3095-2012) and the World Health Organization

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annual guideline value (10 μg/m^3). The same $PM_{2.5}$ pollution problem was also a serious problem in Beijing and Nanjing (Ji et al., 2019;Zheng et al., 2019). 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 NO_3^- was nonsignificant ($p > 0.05$), and SO_4^{2-} and NH_4^+ had obvious significance from 2015 to 2017 ($p < 0.05$), indicating that NO_3^- had not decreased significantly, and there was an increase in 2017 compared to 2015. SO_4^{2-} continues to decline, and NH_4^+ was also lower in 2017 ~~compared to~~than in 2016. Notably, SO_4^{2-} and NH_4^+ decreased significantly in 2017 compared with 2015, but the variation in NO_3^- was nonsignificant. Meanwhile, the annual averages of NO_3^-/SO_4^{2-} were 0.95, 1.02 and 1.45 for 2015, 2016 and 2017, respectively, indicating that the contribution of ~~vehicle~~ NOx emissions ~~as a mobile~~ source to $PM_{2.5}$ was increased compared with that of SO_2 emissions~~coal combustion as a stagnant~~ source (Li et al., 2017;Wang et al., 2015). As shown in Table S1, from 2013 to 2017, the emissions of NO_2 in Chengdu were obviously higher than those of SO_2, but $PM_{2.5}$, NO_2 and SO_2 all showed downward ~~trends, which benefited from the~~due to implementation of the Air Pollution Prevention and Control Action Plan launched by ~~the Chinese government, and~~ Sichuan Province~~Chengdu also launched a more detailed pollution control plan in 2014~~. From 2015 to 2017, the measures taken by Sichuan Province in the coordinated reduction of multiple pollutants have been continuously strengthened, and the scope of management and control has been continuously expanded, for example, in the improvement of desulfurization, denitrification and dust removal technologies in key industries, from accelerated improvement in 2015 to deeper improvement in 2017. The process of eliminating small coal-fired boilers began in 2015 and was completed in 2017, when the ultra-low-emission coal-fired power plant transformation was promoted. In terms of vehicle emission control, we accelerated the elimination of "yellow label" vehicles (general term for gasoline vehicles with emission levels lower than the national I emission standard and diesel vehicles with emission levels lower than the national III emission standard when new vehicles are finalized) and “old vehicles” (the emission level does not meet the national stage IV emission standard) in 2015 and basically completed the elimination of "yellow label" vehicles in

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2017. The quality supervision of oil products has also been improved, and non-road mobile machinery pollution control requirements were proposed in the 2017 plan (The People's Government of Sichuan Province, 2015, 2016, 2017, last access: June 17, 2020ThePeople'sGovernmentofSichuanProvince, 2015, 2016, 2017). Compared with 2015, NOx and SO_2 decreased by 5.98% and 32.35% respectively, in 2017, which shows that the treatment of NOx and SO_2 emissions has achieved remarkable results, of which SO_2 emission reduction effect is the best, followed by that of NOx. The effect of this emission reduction is due to air pollution prevention measures, especially the measures of "electricity instead of coal" and "natural gas instead of coal" (refers to increased use of electricity and natural gas in the residential sector to reduce coal combustion).

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Table 2. Comparison of annual mass averages (μg/m^3) and proportions (%) for SNA from 2015 to 2017.

	$PM_{2.5}$$NO_3^-$	SO_4^{2-}	NH_4^+	NO_3^- $PM_{2.5}$	NO_3^- /$PM_{2.5}$	SO_4^{2-} /$PM_{2.5}$	NH_4^+/$PM_{2.5}$
2015	67.789.13	10.37	6.14	67._ 9.1378	0.129	0.165	0.088
2016	71.889.27	8.53	6.16	71.889.27	0.123	0.133	0.089
2017	59.689.17	6.88	5.01	59.689.17	0.141	0.132	0.079

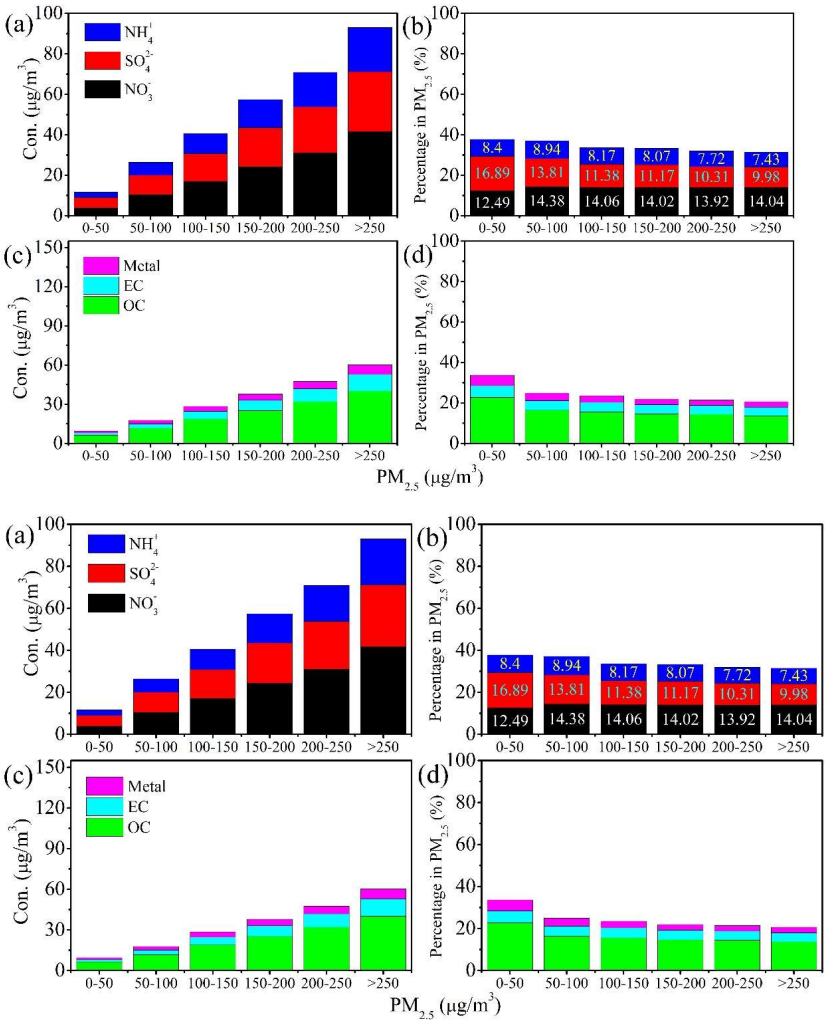

Fig. 2. Variation characteristics of the NSA (nitrate, sulfate and ammonium) and other chemical compositions with different concentrations of $PM_{2.5}$. (a) NSA mass concentration. (b) Percentage of NSA in $PM_{2.5}$. (c) ~~chemical~~ Chemical compositions of organic carbon (OC), element carbon (EC), and metal elements. (~~e~~d) Percentage of OC, EC and metal elements~~other chemical compositions~~ in $PM_{2.5}$.

The chemical composition of $PM_{2.5}$ from 2015 to 2017 varies with its concentration, as shown in Fig. 2. With the accumulation of $PM_{2.5}$ in the atmosphere, the concentration of NSA has also increased significantly, but their proportion of NSA in $PM_{2.5}$

decreased~~has a downward trend~~ (Fig. 2a and b). 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). When the $PM_{2.5}$ was less than 50 μg/m^3 and greater than 250 μg/m^3, the mass concentrations of NSA were 11.57 and 90.06 μg/m^3, respectively, and the proportions were 37.78 and 31.45% respectively. Comparing Fig. 2b and d, it was found that NSA was always the main contributor in the entire process of $PM_{2.5}$ accumulation, which was significantly higher than the proportions of OC and EC (Ji et al., 2019;Li et al., 2019b). In the accumulation process of $PM_{2.5}$ concentrations greater than 50 μg/m^3, NO_3^- ~~nitrate~~ accounts for a high proportion in SNA and ~~was~~ is stable at approximately 14%, and the proportion of SO_4^{2-} and NH_4^+ ~~sulfate and ammonium~~ continues to decrease (Li et al., 2019b;Wang et al., 2016). When the $PM_{2.5}$ concentration was less than 50 μg/m^3, the concentration of SO_4^{2-} was higher than that of NO_3^-, and the concentration of NH_4^+ was lower than the NH_4^+ concentration of $PM_{2.5}$ at 50 to 100 μg/m^3, possibly due to SO_4^{2-} ~~sulfate~~ concentration was higher than NO_3^- ~~nitrate~~, forming more chemically stable ~~ammonium sulfate~~$(NH_4)_2SO_4$ (Guo et al., 2017a). In addition, when $PM_{2.5}$ was less than 50 μg/m^3, low RH and strong solar radiation were also important ways to generate sulfate (Yao et al., 2018).

3.2 Monthly and seasonal variations

The monthly variation characteristics of NSA from 2015 to 2017 are shown in Fig. 3. At the beginning and the end of each year, the pollutant concentration is relatively high and relatively low in the middle of each year. The meteorological conditions also have

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obvious monthly variation characteristics (Fig. S5 a and b),~~The monthly variation characteristics of NSA from 2015 to 2017 are shown in Fig. 3. These variations have similar trends due to meteorological factors (Fig. S1);~~ from April to August, they have higher~~ temperature and~~ WS,~~ and~~ lower RH~~ and atmospheric pressure~~, which is not only conducive to the dilution and diffusion of pollutants but also reduces the chemical conversions of pollutants by aqueous phase and influencing the formation of secondary inorganic aerosols~~ and the concentrate ability of gaseous pollutants to concentrate particles~~ (Wang et al., 2016;Ji et al., 2019). Overall, the concentrations are higher in January and December and lower in July and August. The highest monthly average ~~of~~ NO_3^- reached 19.98 μg/m^3 in January 2017, and the highest monthly average of SO_4^{2-} and NH_4^+ were 22.08 μg/m^3 and 12.66 μg/m^3 in January 2015, respectively. The lowest concentrations of NSA appeared in August 2017, which were 1.96, 3.07 and 1.62 μg/m^3. The gaseous precursors of NSA also have obvious monthly variations, and the NOx and SO_2 trends were similar to those of NO_3^-~~ nitrate~~ and SO_4^{2-}~~ sulfate~~ (Fig. 3 and ~~S2~~4). NH_3 emissions were significantly different, with increases in warmer months (April-July) and colder months (September-December). On the one hand, NH_3 volatilization was promoted by relatively high ~~temperatures~~Ts (Fig. S5c); on the other hand, the use of agricultural fertilizers and livestock farming were also important sources of NH_3 in China. Second, from urban ~~areas~~region, fossil fuel combustion and motor vehicle emissions also contribute significantly (Liu et al., 2013b;Pan et al., 2016). Notably, NH_3 increased significantly from April to December 2017 compared with 2015 and 2016, especially during low-~~temperature~~ T months (Fig. ~~S2c~~4c). The result of an analysis of the monthly concentration variation of pollutants indicate that the implementation of pollution reduction and control measures should be strengthened at the beginning of each year (January to March) and the end of the year (October to December).~~This also shows that the implementation of air pollution reduction measures should increase the emission reduction intensity in terms of NOx and NH_3 emissions, especially the implementation of autumn and winter air pollution prevention and control action.~~

The seasonal variation in NSA was shown in Fig. ~~S3~~S6, and the concentration in winter was much higher than that in summer. NO_3^-~~ Nitrate~~ only declined in spring and summer

from 2015 to 2017, with an increase in autumn and winter (Fig. ~~S3A~~S6a). Seasonal variations in ~~ammonium~~ NH_4^+ were similar to those of NO_3^- ~~nitrates~~, with higher concentrations in winter and the lowest in summer (Fig. S6c). This may be because higher ~~temperatures~~ Ts and WSs not only can promote the decomposition of NH_4NO_3 ~~ammonium nitrate~~ in summer but also promote the dilution and diffusion of pollutant concentrations (Guo et al., 2017a;An et al., 2019). There is a significant downward trend in SO_4^{2-}, which continues to decrease in spring, summer and winter from 2015 to 2017 (Fig. S6b). In autumn, the concentration was the highest in 2016, and it was significantly lower in 2017 than in 2015 and 2016.~~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 Prevention and Control Action Plan, especially the measures of "electricity instead of coal" and "natural gas instead of coal"(refers to increased use of electricity and natural gas in the residential sector to reduce coal combustion).~~ The variation amplitude of NSA and gaseous pollutants in cold months was significantly higher than that in warm months (Figs. 3, ~~S2~~ 4 and ~~S3~~S6). This higher variation amplitude may be due to the differences in pollutant accumulation and scavenging processes. This finding also indicates that the instability of local pollutant emissions and regional transport during cold months was affected by meteorological conditions (Li et al., 2017;Ji et al., 2018). The large variation amplitude of pollutants in different months, similar to the changes in the Beijing-Tianjin-Hebei region of northern China and Chengdu, are due to the accumulation and removal of pollution by meteorological conditions and pollutants emissions (Ji et al., 2019;Qin et al., 2019;Zhang et al., 2019a).

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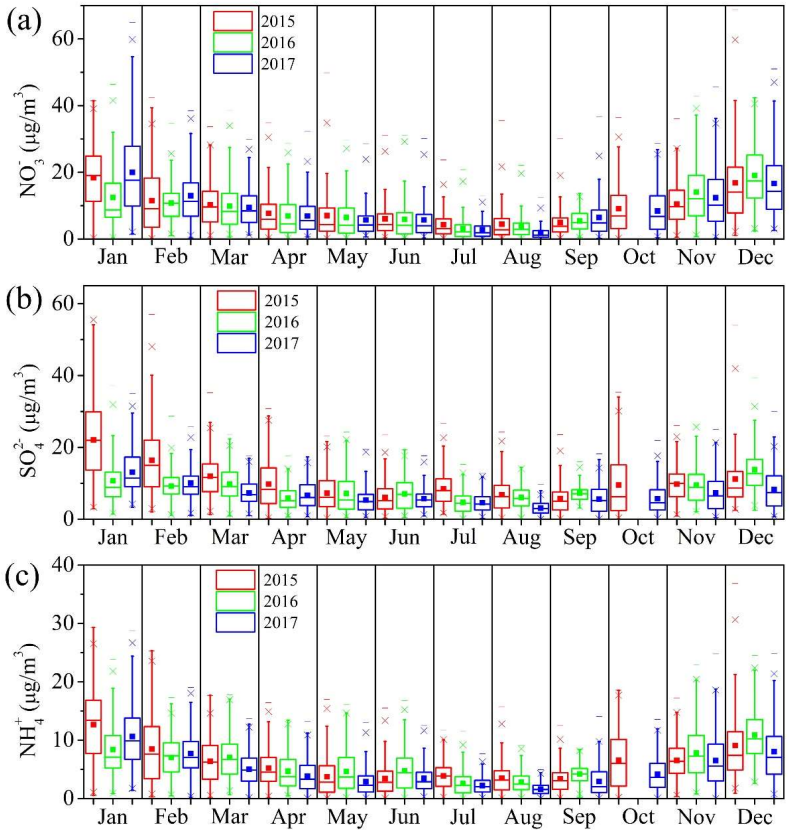
(a)
2015
2016
2017
NO_3^- (μg/m^3)
0
20
40
60
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec
(b)
2015
2016
2017
SO_4^{2-} (μg/m^3)
0
20
40
60
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec
(c)
2015
2016
2017
NH_4^+ (μg/m^3)
0
10
20
30
40
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec

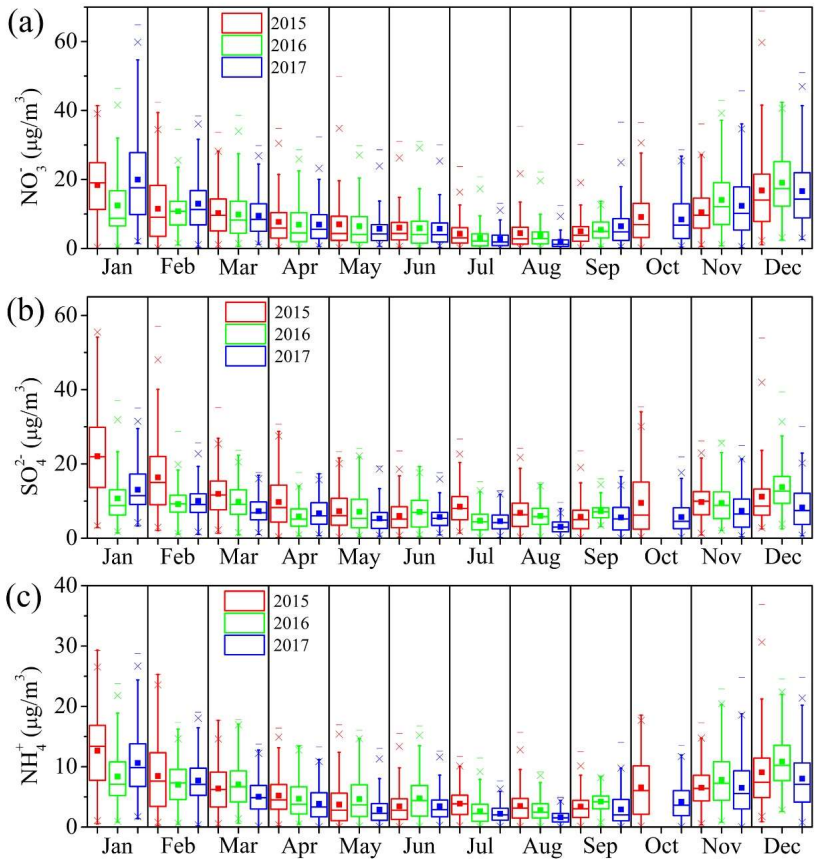

Fig. 3. Monthly variations in ~~NO_3^-, SO_4^{2-} and NH_4^+ mass~~NSA (nitrate, sulfate and ammonium) concentrations from 2015 to 2017. (a) NO_3^-. (b) SO_4^{2-}. (c) NH_4^+.

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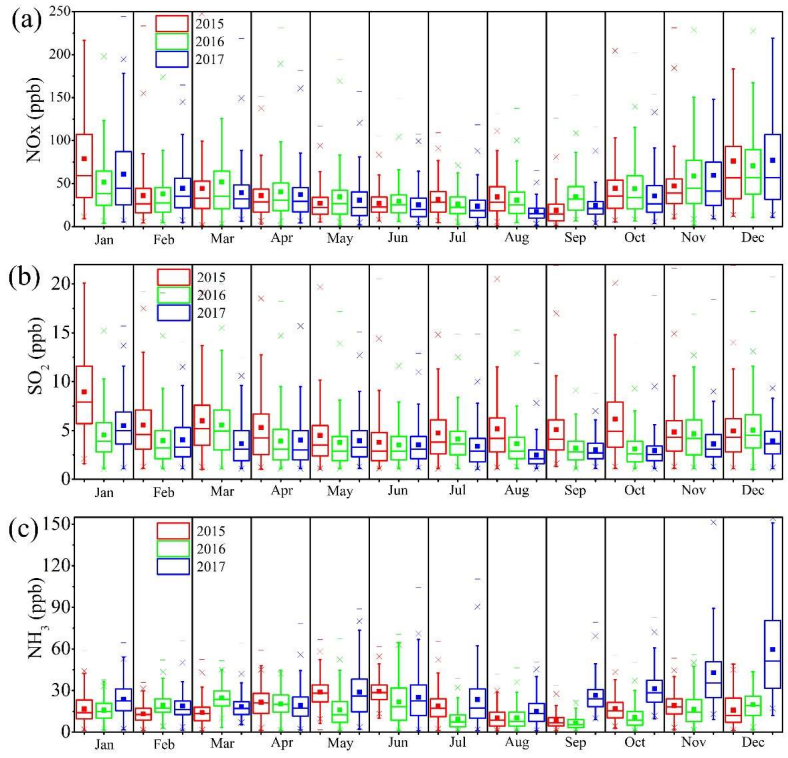

Fig. 4. Monthly variations in NOx, SO_2 and NH_3 concentrations from 2015 to 2017. (a) NOx. (b) SO_2. (c) NH_3.

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3.3 Diurnal and weekly variations

~~The diurnal variation in NSA is shown in Fig. 4; a similar trend was shown for daily changes of nitrate, sulfate and ammonium, which was higher in the daytime than in the evening. From 2015 to 2017, the diurnal variation trend of nitrate was similar, sulfate was obviously reduced, and the ammonium was only significantly reduced in 2017. The decrease in NH_4^+ may be closely related to the decrease in SO_4^{2-} (Fig. 4a). Some studies have shown that NH_4^+ in aerosols will first combine with SO_4^{2-} ions and HSO_4^- to form $(NH_4)_2SO_4$ or NH_4HSO_4 and then combine with NO_3^-. The significant drop in NH_4^+ in 2017 may be due to a decrease in SO_4^{2-}. Similarly, the NH_4^+ did not show a significant decrease in 2016, probably due to the increase in NO_3^- in 2016 (Table 2), combined~

~~with a portion of the NH_4^+. This finding also indicates that the concentration of NH_4^+ in particulate matter in Chengdu may be affected by the concentration of SO_4^{2-}.~~ From 2015 to 2017, the concentration of NSA was higher in the daytime than in the evening (Fig. ~~4a~~5a), and similar results were found in different seasons (Fig. ~~4b~~5b), which may be due to the combination of pollutant emissions and meteorological conditions. As shown in Fig. ~~S4~~S7, from 9:00 to 11:00 a.m., the concentrations of SO_2, NOx, NH_3~~,~~ and CO ~~and other gases~~ 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 O_3 and NO_2/NO also occasionally show an increasing trend, indicating that the atmospheric oxidizability has also increased~~higher RH~~ (Fig ~~S5~~S7 and 8)，this situation also provides favourable conditions for the formation of secondary aerosols and promotes the accumulation of NSA (Cheng et al., 2016;Wang et al., 2016;Sun et al., 2014). In addition, before 10 o'clock, relatively low WS will enable easy pollutant concentration accumulation. In contrast, the higher WS in the afternoon may be the main factor for the decrease in pollutant concentration (Fig. 4~~, S4~~5 and ~~S5~~S8). Photochemical reactions may also be one of the factors in the formation of NSA, and the concentration of O_3 peaks at approximately 15:00, which may be affected by the free radicals generated by photochemistry. At approximately 19:00, the ratio of NO_2/NO reached its highest value, and the concentration of NO_2 also increased significantly (Song et al., 2018;Zhu et al., 2019). At night, with the increase in RH (Fig. ~~S5~~S8), dissolved ozone, free radicals, hydrogen peroxide and NO_2 can catalyse SO_2 to form secondary aerosols through an aqueous phase reaction (Zhang et al., 2015;An et al., 2019). The seasonal diurnal variation in NSA was shown in Fig. ~~4b~~5b. The concentration of NSA in winter was obviously higher than that in summer, and the diurnal variation range was larger. The concentrations in spring and autumn was closer, but the diurnal variation in spring was larger than that in autumn. The larger diurnal variation range not only indicates serious pollution but also indicates the importance of other factors affecting air quality, such as meteorological conditions~~,~~ and secondary aerosol conversion conditions~~, and so on~~ (Ji et al., 2019;Yang et al., 2015b). The peak value of the NSA seasonal diurnal variation

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also varies in different seasons. The peak value appears at approximately 13:00 in winter, approximately 10:00 in spring and summer, and approximately 12:00 in autumn, possibly due to the influence of meteorological conditions. In previous studies in Beijing-Tianjin-Hebei and 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 variation of WS and atmospheric humidity, some studies have shown that due to the unique topographical structure of Sichuan Basin, the atmospheric circulation between the Qinghai-Tibet Plateau, Yunnan-Guizhou Plateau and Sichuan Basin and the meteorological conditions of 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., 2019c;Zhang et al., 2019b).

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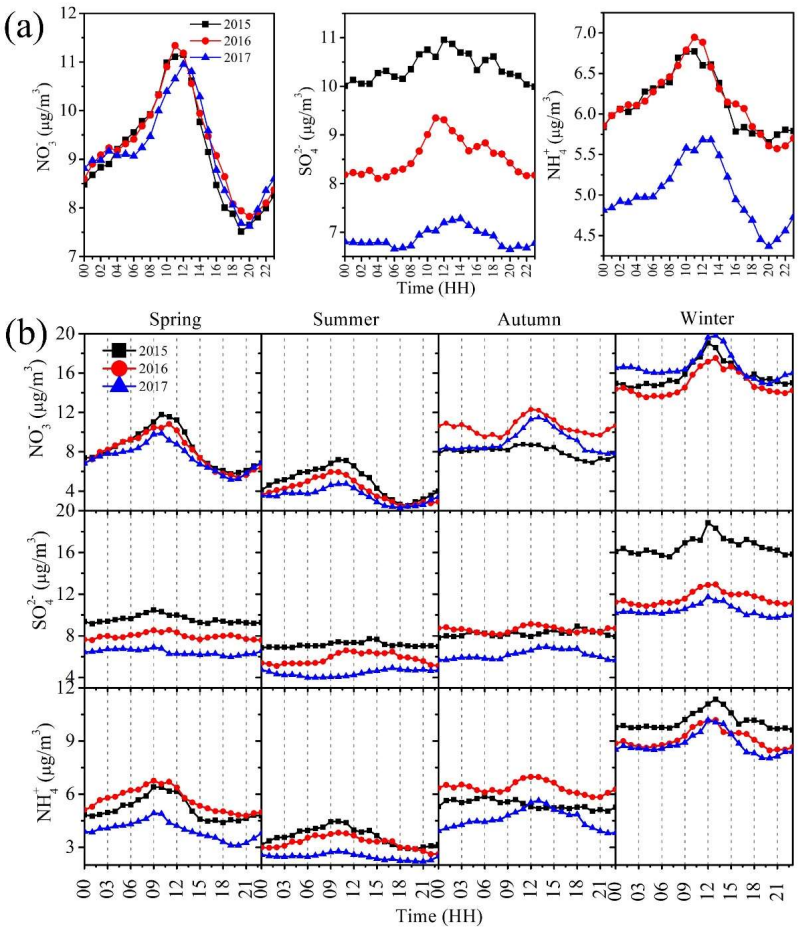
(a)
2015
2016
2017
NO_3^- (μg/m^3)
SO_4^{2-} (μg/m^3)
NH_4^+ (μg/m^3)
Time (HH)
(b)
Spring
Summer
Autumn
Winter
2015
2016
2017
NO_3^- (μg/m^3)
SO_4^{2-} (μg/m^3)
NH_4^+ (μg/m^3)
Time (HH)

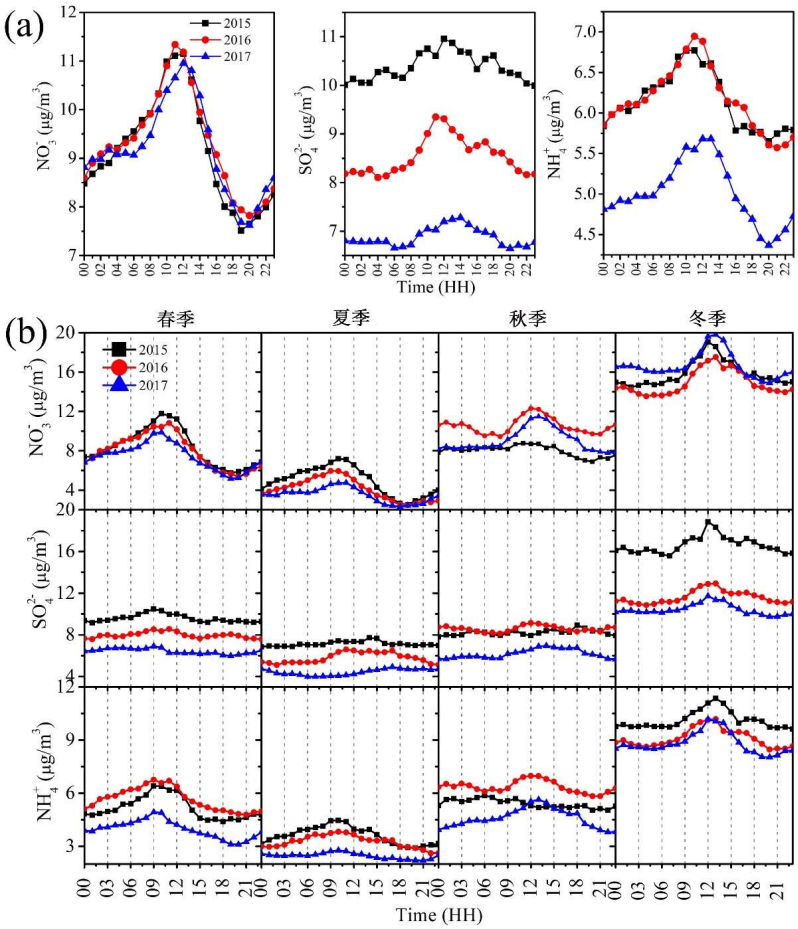

Fig. 45. Diurnal variations in SNA (nitrate, sulfate and ammonium from 2015 to 2017. (a) Annual average. (b) Seasonal average.

The weekly variation in NSA is shown in Figs. S6–89. During the overall observational period, workdays (Monday to Friday) showed higher variations than the weekends (Saturday and Sunday), with the highest variation being on Tuesday and the lowest being on Sunday. Despite the difference in mean values between Tuesday and Sunday, nonparametric tests show that the difference in mean values was nonsignificant (Mann-Whitney U test, $P > 0.05$). As shown in Figs. S6–79, the average trends of NO_3^- and

NH_4^+ were consistent from Monday to Sunday. The correlation coefficient was 0.94 ($P < 0.01$) from 2015 to 2017, which indicates that they have a common source and that the vehicle emissions also have an important contribution to NH_4^+ (Pan et al., 2016). The average values of NO_3^-, SO_4^{2-} and NH_4^+ from 2015 to 2017 were 9.21, 8.64 and 5.64 ug/m^3 on workdays and 8.56, 8.33 and 5.29 ug/m^3 on weekends, respectively. The average values of NOx, SO_2 and NH_3 were 42.43, 4.35 and 20.39 ppb on weekdays, 39.60, 4.34 and 19.67 ppb on weekends, respectively. Similarly, the mean difference between NSA and gaseous precursors (NOx, SO_2 and NH_3) was not significant by the Mann-Whitney U test on weekdays and weekends.~~Similarly, the Mann-Whitney U test showed no significant difference.~~ Population standard deviation comparisons of NO_3^-, SO_4^{2-} and NH_4^+ showed that workdays were higher than weekends, with 7.96, 6.04 and 4.35 on weekdays, 6.76, 5.69 and 3.88 on weekends, respectively, and it can also be seen from the box chart of NSA weekly variation that the concentration range on working days was slightly larger than that on weekends (Fig. ~~S8~~S10). The analysis of the diurnal variation of NSA gaseous precursors on weekdays and weekends shows that the variation trend is relatively consistent (Fig. S11), and the concentration of NOx on weekdays will be slightly higher at the peak of 9:00 to 10:00 than at weekends, which may be affected by the morning rush hour of vehicles. In this study, NSA and gaseous precursors are also slightly higher on weekdays than on weekends, which indicates that in Chengdu's air pollution prevention and control actions, the management of relevant industries and departments should be strengthened on weekdays.~~Compared with the diurnal variations on weekdays and weekends, the variations in nitrate and ammonium were more obvious than those of sulfate (Fig. S9). In Beijing, a vehicle restriction scheme based on motor vehicle license plates was implemented, that is, there are no restrictions on weekends, and the contribution of vehicle emissions pollution on weekends was lower than that on workdays . Similarly, Chengdu also implemented restriction measures according to the license plate of vehicles on weekdays, but the average concentration of pollutants on weekdays was slightly higher than that on weekends (Mann-Whitney U test, P > 0.05). This finding shows that while implementing a policy of motor vehicle restriction, improving the emission standards~~

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~~of motor vehicles and the quality of gasoline and diesel oil was an important measure.~~

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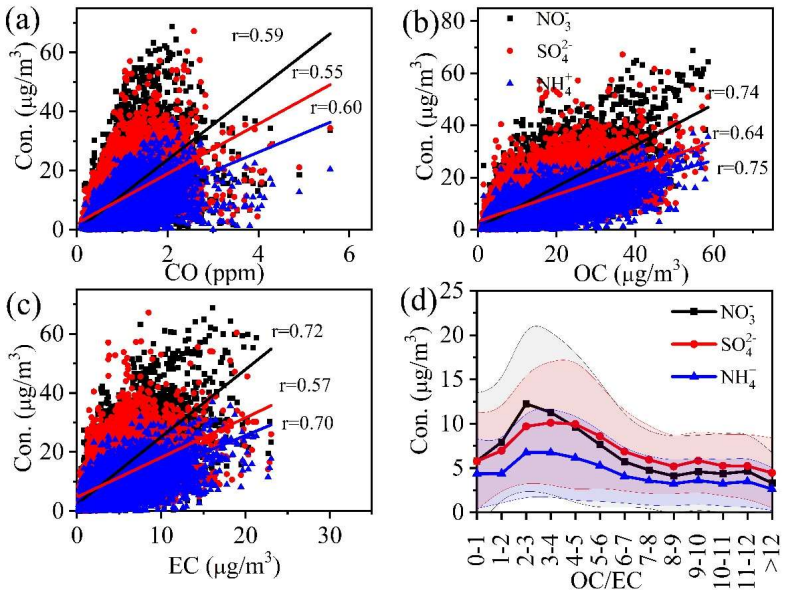
3.4 Chemical characteristics of SNA

~~3.4.1 Relationship between SNA and carbonaceous components~~

~~Precursor gases of NSA, such as SO_2, NOx and NH_3, are usually derived from coal combustion, vehicle exhaust and agricultural sources, and they are accompanied by emissions of carbonaceous aerosols . Fig. 5a, b and c show the relationship between NSA concentration and CO, OC and EC, showing a good Pearson's correlation ($p<0.01$), which indicates that the emissions of carbon aerosols were accompanied by the emissions of NSA precursor gases; these gases form NSA through complex chemical reactions, such as photochemical, aqueous chemical conversions and heterogeneous reactions . CO and EC usually originate from combustion sources, while OC originates from primary emissions and secondary conversion . The OC/EC value can be used to determine the sources of carbon aerosols, such as vehicle exhaust, coal combustion and biomass burning . As shown in Fig. 5d, when the concentration of nitrate and ammonium reached a peak, the OC/EC value was between 2-3, which was lower than the OC/EC value when the sulfate was at the peak (3-4). Previous studies have also shown that the OC/EC value of vehicle emissions was lower than that of coal combustion . Nitrate and ammonium also have similar trends, and their Pearson's correlation was 0.92 ($p<0.01$), which was higher than that of ammonium and sulfate (0.88). The correlation coefficients of NH_3 with NOx and SO_2 were 0.42 and 0.23, respectively, suggesting that vehicle emissions may also be a major source of ammonia .~~



~~Fig. 5. Relationships between SNA and CO and OC and EC. (a) SNA and CO. (b) SNA and OC. (c) SNA and EC. (d) SNA and OC/EC.~~

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3.4.~~2~~ 1 Chemical conversion characteristics of SNA

~~Figure~~ Fig. 6 shows the abilities of ~~SO_2~~ NO_2 ~~-~~and SO_2~~NO_2~~ to chemically convert to NO_3^- and SO_4^{2-} ~~sulfates and~~ nitrates ~~and the variation trend of ozone concentration and metal elements~~ at different $PM_{2.5}$ concentrations. With the increase in $PM_{2.5}$ concentration, ~~SOR~~ NOR and ~~NOR~~ SOR gradually increased, indicating that the formation ability of ~~sulfate~~ NO_3^- and SO_4^{2-} ~~and nitrate~~ increased during the formation of air pollution. In this study, when the $PM_{2.5}$ concentration is ≤ 50 μg/m^3, the average of NOR and SOR are 0.07 and 0.27, respectively, and when the $PM_{2.5}$ concentration is greater than 250 μg/m^3, the average of NOR and SOR increase to 0.22 and 0.41, , respectively, indicating that the chemical conversion and formation ability of secondary inorganic aerosol was obviously enhanced when the air pollution was aggravated. ~~As the concentration of $PM_{2.5}$ increases, the extinction properties of aerosols increase, the photochemical reaction conditions weaken, and the O_3 concentration shows a~~

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~~decreasing trend, as shown in Fig. 6a. With the accumulation of $PM_{2.5}$ concentration, metal elements (Fe and Mn) also showed an increasing trend and were similar to SOR and NOR. Previous studies have shown that mineral dust elements such as Fe and Mn can play a catalytic role in the formation of atmospheric sulfate . The Pearson's correlation statistics of SOR and NOR with Fe and Mn under different $PM_{2.5}$ concentration conditions are shown in Table S2; it is only under high $PM_{2.5}$ concentration conditions (>200 μg/m^3) that SOR and NOR have a positive correlation. This result is similar to those of previous studies in Beijing and Xi'an, where Fe and Mn play a limited catalytic role in sulfate formation .~~ ~~Some~~ Previous studies suggest that when ~~SOR~~ NOR and SOR ~~is~~ are greater than 0.~~1~~1, and 0.2, respectively, it has a intense conversions and formmation secondary inorganic aerosols~~there may be a photochemical reaction pathway leading to the conversion of SO_2 to sulfate~~ (Yang et al., 2015a). ~~Fig. 6a shows that in addition to the photochemistry contributing to SO_2 oxidation, there may be a more important pathway leading to the conversion of SO_2 to sulfate.~~

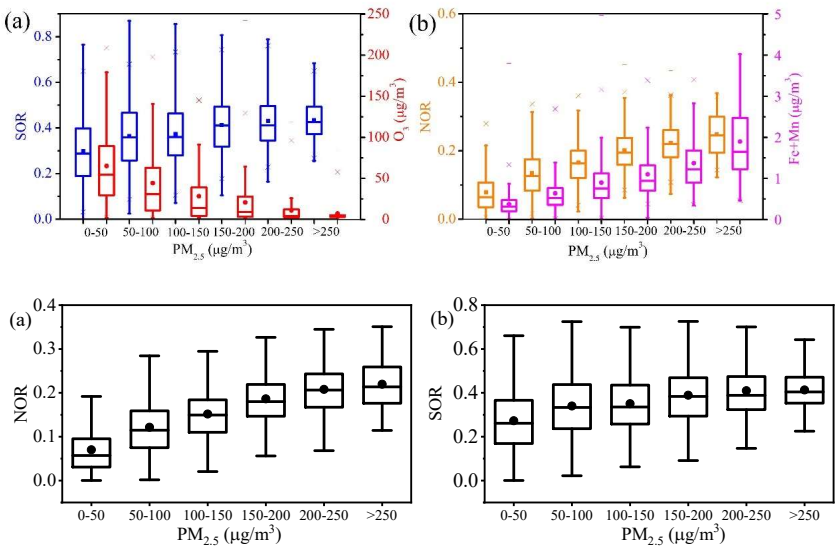

Fig. 6. Analysis of atmospheric chemical conversion ability at different $PM_{2.5}$ concentrations. (a) ~~S~~ NOR (nitrogen oxidation ratio). (b) SOR (sulfur oxidation

ratio)~~OR and O_3. (b) NOR and concentration of metal elements (Fe and Mn).~~

~~Figure~~ Fig. 7 shows the variation characteristics of NSA chemical conversion~~s and meteorological conditions~~ with increasing RH. ~~SOR~~ NOR and ~~NOR~~ SOR increased with increasing RH, suggesting that ~~SO_2~~ NO_2 and ~~NO_2~~ SO_2 were more likely to produce NO_3^- and SO_4^{2-} ~~sulfate and nitrate~~ under higher RH conditions. Previous studies have shown that in the presence of NH_3, NO_2 can promote the chemical conversion of SO_2 to SO_4^{2-} ~~sulfate~~ in the aqueous phase (Wang et al., 2016). In an aerosol aqueous phase environment ~~In aerosol water~~, alkaline aerosol (NH_3) components can promote the dissolution of SO_2 and formation of SO_4^{2-} ~~sulfate~~ under the oxidation of NO_2 (Cheng et al., 2016). Especially when the atmosphere was polluted, the formation of SO_4^{2-} ~~sulfate~~ by SO_2 through the aqueous phase environment can contribute most of the SO_4^{2-} ~~sulfate~~ (Sun et al., 2013). When the RH is greater than 80%, the NOR appears to decline, possibly because HNO_3 is semivolatile, and the T increases at this time (Fig. 7 c), which is not conducive to the condensation of gaseous HNO_3 to the particulate matter, which affects the amount of NO_3^- in $PM_{2.5}$ (Guo et al., 2017a). According to the ISORROPIA-II thermodynamic equilibrium model simulation, AWC ~~and pH~~ also increase with RH (Fig. 7~~c and~~ d), and ~~. T~~the increase of AWC can provide a liquid environment for aerosol, which is conducive to the dissolution and conversion of gaseous precursors of NO_2, SO_2 and NH_3, and promote the formation of more NSA. The Pearson's correlation coefficients of RH and NOR and SOR were 0.12 and 0.16 ($p<0.01$), and the AWC and NOR and SOR were 0.73 and 0.37 ($p<0.01$), respectively, showing a significant positive correlation, indicating the increase of AWC is beneficial to the conversion of NO_2 and SO_2 to NO_3^- and SO_4^{2-}.~~increase in AWC can dilute the concentrations of sulfate and hydrogen ions and promote an equilibrium shift in the SO_2 to sulfate during the aqueous phase~~.~~ The increase in RH and gradual decrease in T can also affect the gas-particle phase partitioning of HNO_3-NO_3^- (Fig. 7g) and NH_3-NH_4^+ (Fig. 7h), prompting more nitrate and ammonium to condense in aerosol liquid water. By comparing the NOR with the meteorological conditions and gas-particle distribution when RH is greater than 80% (Fig. 7b, e, f and g), the increase in T and the decrease in atmospheric pressure were not conducive to the conversion and presence of nitrate in the aqueous phase.~~ As shown

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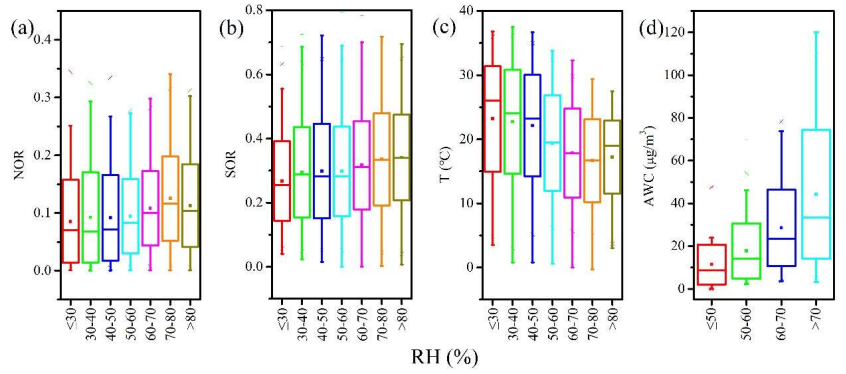
in Fig. S12, the simulated values of NSA (meta-stable state, liquid phase components) are compared with the observed data. The linear regression fitting slope is about 1 ($p<0.01$), indicating that the effect of liquid phase environment in $PM_{2.5}$ is obvious; in addition, stable state simulation is also performed, and the linear fitting slopes of the NSA liquid phase state data output from the model and the observations data are 0.73, 0.63 and 0.74, and the Pearson's correlations are 0.82, 0.71 and 0.80 ($p<0.01$), indicating that they are more often combined with AWC in the aerosol aqueous phase environment at stable ~~Therefore~~state. Previous studies have also confirmed that the aqueous phase environment of aerosols plays an important role in the formation of secondary inorganic aerosols (Wang et al., 2016;Cheng et al., 2016). ~~, Figs. 6 and 7 also illustrate that the aqueous phase oxidation environment may contribute to the generation of a larger portion of NSA.~~



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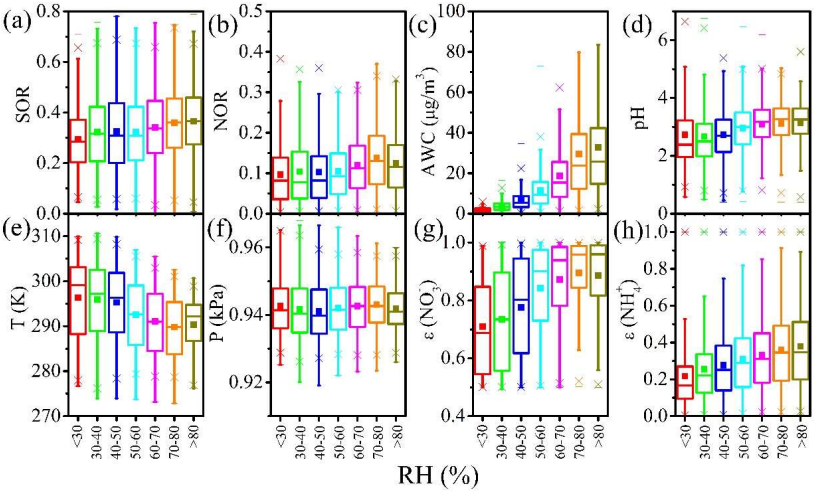
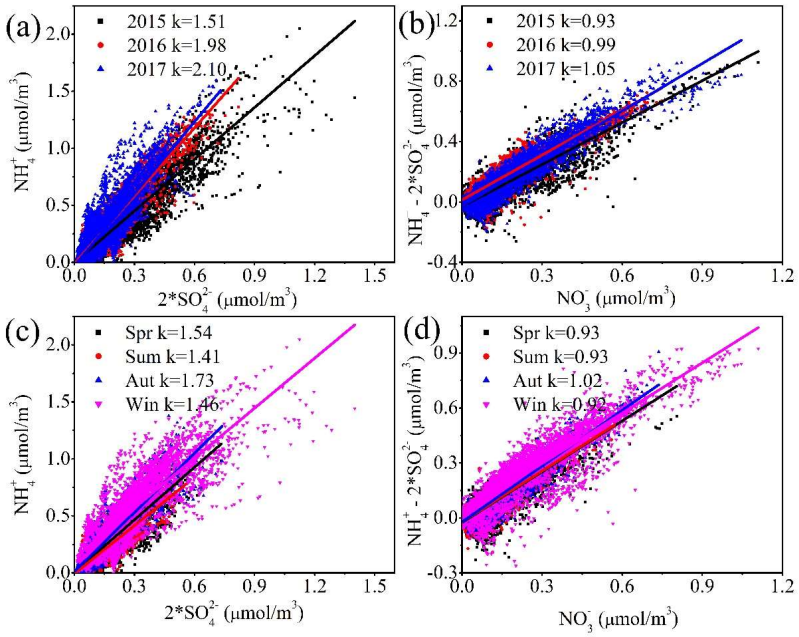


Fig. 7. Effects of RH on the chemical conversion of NSA (nitrate, sulfate and ammonium). (a) ~~SOR~~NOR. (b) ~~NOR~~SOR. ~~(c) AWC. (d) pH of $PM_{2.5}$.~~ (~~e~~c) Temperature (T). (~~f~~d) ~~Atmospheric pressure (P)~~AWC (aerosol water content). ~~(g) NO_3^- gas-particle phase partitioning. (h) SO_4^{2-} gas-particle phase partitioning.~~

3.4.3 Sensitivity analysis

The molar ratio analysis of NSA shown in Fig. 8 was used to analyse the chemical relationships among NSA. $(NH_4)_2SO_4$ and NH_4NO_3 are mainly composed of NH_4^+, SO_4^{2-} and NO_3^- in particulate matter (Malm and Hand, 2007;Meier et al., 2009). Because $(NH_4)_2SO_4$ has better stability than NH_4NO_3, NH_4^+ will first combine with SO_4^{2-} and then with NO_3^- (Liu et al., 2012). The annual average molar ratio of NH_4^+ to $2*SO_4^{2-}$ was more than 1, which indicates that SO_4^{2-} can be completely neutralized by NH_4^+ (Fig. 8a). The molar ratios of residual NH_4^+ ($NH_4^+ - 2*SO_4^{2-}$) to NO_3^- were 0.~~93~~85, 0.~~99~~ 96 and 1.~~05~~ 04 in 2015, 2016 and 2017, respectively. As shown in Fig. 8a and b, the gradual increase in the ratio (slope k) from 2015 to 2017 indicates that there is an increase in NH_4^+ in aerosol compared with SO_4^{2-} and NO_3^-~~indicates an increase in ammonia emissions from aerosols~~, especially in 2017, with a ratio of 1.0~~5~~,4, indicating the presence of other forms of NH_4^+~~ammonium salts~~, such as NH_4Cl and $(NH_4)_2C_2O_4$ (Sun et al., 2006). Seasonal variations in NH_4^+, SO_4^{2-} and NO_3^- are shown in Fig. 8c and d. The higher molar ratio in autumn indicates that the intensity of ammonia

emission in autumn was higher than that in other seasons. This result also shows that the proportion of NH_4^+ relative to NO_3^- and SO_4^{2-} in the $PM_{2.5}$ has increased. Therefore, while currently controlling NOx and SO_2 emissions, it is also necessary to strengthen NH_3 emissions control. ~~This finding also shows that the problem of atmospheric ammonia-rich environments in Chengdu in 2017 and autumn was more prominent.~~



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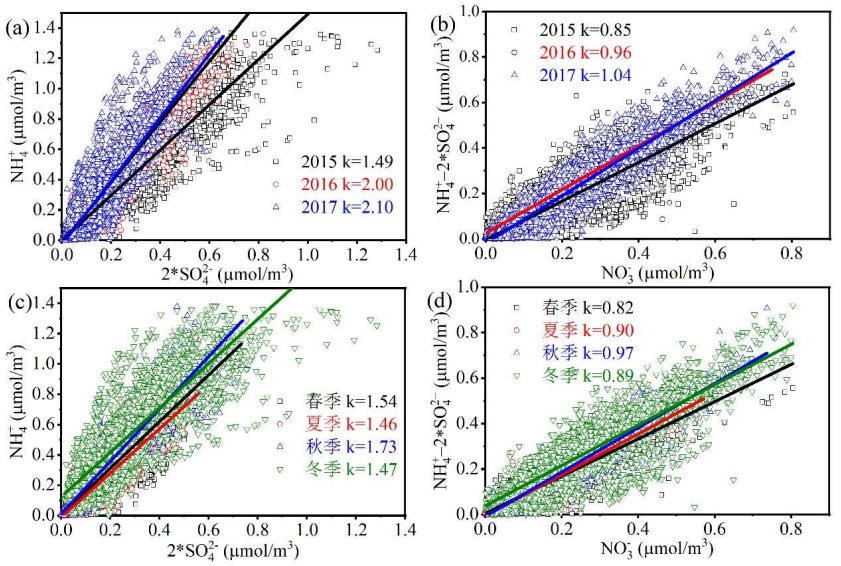

Fig. 8. Molar ratio analysis of NSA (nitrate, sulfate and ammonium). (a) Interannual variation in the molar ratio of SO_4^{2-} and NH_4^+. (b) Interannual variation in the molar ratio of NO_3^- and NH_4^+. (c) Seasonal variation in the molar ratio of SO_4^{2-} and NH_4^+. (d) Seasonal variation in the molar ratio of NO_3^- and NH_4^+. k: Fitting slope of linear regression.

Table 3 shows the sensitivity analysis of the concentration variations in SO_4^{2-}, NO_3^- and NH_4^+. ISORROPIA-II thermodynamic equilibrium model sensitivity analysis is described in detail in the Supplementary Materials. The coefficient of variance represents the response of the species to variations in other components. The coefficients of variance for NH_4^+ and NO_3^- produced by SO_4^{2-} changes were 52.62 22 and 51.3870, respectively. Similarly, the coefficients of variance for NH_4^+ and SO_4^{2-} produced by NO_3^- nitrate changes are 4951.27 42 and 0.0020005, respectively. The large coefficient of variance for NH_4^+ indicates that the changes in SO_4^{2-} NO_3^- and SO_4^{2-} NO_3^- can affect the presence of NH_4^+, which also indicates that $(NH_4)_2SO_4$ NH_4NO_3 and $(NH_4)_2SO_4NH_4NO_3$ were the main states of NH_4^+ (Liu et al., 2012). The coefficients of variance for SO_4^{2-} and NO_3^- produced by TNH3 (NH_3+NH_4^+) changes are 20.48 47 and 3115.3076, respectively, and the effect of TNH_3 on SO_4^{2-} is less than that of NO_3^-,

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which indicates that NH_4^+ was excessive to SO_4^{2-} ~~sulfate~~ and that NH_4^+ first combines with s SO_4^{2-} ~~ulfate~~ to form stable $(NH_4)_2SO_4$, and the remaining NH_4^+ and NO_3^- will combine to form NH_4NO_3. ~~From 2015 to 2017, the coefficient of variance for NH_4^+ and NO_3^- caused by the changes in SO_4^{2-} gradually decreased, which may be attributed to the decrease in SO_4^{2-} concentration in $PM_{2.5}$ (Table 2). The coefficients of variance for NO_3^- caused by the changes in NH_3 in 2015 and 2016 were 32.83 and 38.24, respectively. At this time, NO_3^- can completely neutralize NH_4^+. In 2017, the coefficient of variance was 21.88, and the ammonia was a surplus (Fig. 8b); thus, the coefficient of variance may be affected by the thermal instability of NH_4NO_3 during this time . In terms of seasonal variation, the changes in sulfate and nitrate can cause larger coefficients of variance for NH_4^+. When NH_3 changes, the coefficient of variance for NO_3^- was greater than that of SO_4^{2-}. In summer, the coefficient of variance for NO_3^- and NH_4^+ caused by the changes in SO_4^{2-} were obviously higher than those in other seasons. On the one hand, this may be due to the relatively low concentrations of NO_3^- and NH_4^+ in $PM_{2.5}$ due to lower gas-particle phase partitioning in summer (Fig. S10c and d). On the other hand, the stronger photochemical reaction may also lead to a greater change in the concentrations of NO_3^- and NH_4^+ . The coefficients of variance for NO_3^- and SO_4^{2-} in winter were 6.13 and 0.005, respectively, which are higher than those in spring and autumn, most likely due to higher NOR and SOR (Fig. S10a and b). Previous studies have shown that the conversion of SO_2 to sulfate in the aqueous phase not only increases the conversion of sulfate but also enhances the formation of nitrate particles in the aqueous phase . Therefore, sulfate emission reduction may play a key role in the process of controlling emission reduction in NSA pollution, as it not only reduces the presence of NH_4^+ ($(NH_4)_2SO_4$) in particulate matter but also affects the formation of NH_4NO_3 by influencing the formation of nitrate. NO_2 and NH_3 can also promote the conversion of SO_2 to sulfate through an aqueous phase environment . Therefore, in the current ammonia-rich environment, priority control of SO_2 and NO_2 emissions is an important way to reduce NSA in particulate matter.~~

Table 3. Sensitivity analysis of SNA (nitrate, sulfate and ammonium) concentration variations during the different observation periods.

Period	Variation	Coefficients of variance		
		NO_3^-	NH_4^+	SO_4^{2-}
2015-2017	NO_3^-		51.42	0.0005
	TNH_3	15.76		0.47
	SO_4^{2-}	1.70	52.22	

Coefficients of variance:Standard deviation/average *100%;

Variation TNH_3:NH_3+NH_4^+ (μg/m^3);

Variation SO_4^{2-} and NO_3^- units: μg/m^3

Through the implementation of the Air Pollution Prevention and Control Action Plan, the reduction in SO_4^{2-} ~~sulfate~~ emissions has achieved good results. Therefore, while continuing to promote "electricity instead of coal" and "natural gas instead of coal" to reduce coal combustion pollution, more stringent control measures should be added for NO_3^- and NH_4^+ ~~nitrate and ammonia~~ emissions. To further improve air quality, the Chinese government launched a “Three-Year Action Plan for Winning the Blue Sky Defense Battle” in 2018 (the Sate Council, 2018, last access: June 17, 2020~~http://www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm~~) and proposed emission reduction targets for ~~SO_2~~NOx and ~~NOx~~ SO_2 emissions, which will be 15% lower in 2020 than in 2015 (theSateCouncil, 2018). The results of~~By~~ using the ISORROPIA-II thermodynamic equilibrium model to simulate NO_3^-, SO_4^{2-}, ~~NO_3^-~~ and TNH_3 emission reduction control effects of 5%, 10%, 15% and 20% respectively, t~~he results we~~are shown in Table S3, showing that controlling the concentration of NO_3^- and SO_4^{2-} is also helpful to reduce the concentration of NH_4^+ and indicating that controlling its precursor NOx and SO_2 is of great significance to reduce the secondary inorganic aerosol in $PM_{2.5}$ (the detailed results are described in the supplementary materials). ~~the results show that a better effect can be achieved by controlling the SO_4^{2-} and NO_3^- emissions reduction, especially the effects of synergistic emissions reduction.~~ Previous studies have also shown that the conversion of SO_2 to SO_4^{2-} in the aqueous phase not only increases the conversion of SO_4^{2-} but also enhances the formation of NO_3^- particles in the aqueous phase (Wang et al., 2016). Therefore, SO_2 emission

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reduction may play a key role in the process of controlling emission reduction in NSA pollution, as it not only reduces the presence of NH_4^+ ($(NH_4)_2SO_4$) in particulate matter but also affects the formation of NH_4NO_3 by influencing the formation of NO_3^-. NO_2 and NH_3 can also promote the conversion of SO_2 to SO_4^{2-} through an aqueous phase environment (Wang et al., 2016). Therefore, priority control of NOx and SO_2 emissions is an important way to reduce NSA in particulate matter.

In addition, NSA can increase the hygroscopicity properties of aerosols, and more AWC can increase the pH by diluting the hydrogen ion concentration (Kong et al., 2020;Ding et al., 2019). ~~Higher sulfates, nitrates and AWC correspond to a lower pH, indicating that higher sulfates and nitrates have a greater effect on increasing aerosol acidity than AWC dilution (Fig. S11a and b).~~ Previous studies have also shown that SO_4^{2-} ~~sulfate~~ formation reduces aerosol pH (Sun et al., 2014). ~~The same increase in ammonia emissions can increase the aerosol pH (Fig. S11c). Table S3 also shows the impacts of SO_2^-, NO_3^- and NH_3 emissions reduction control on pH, such as sulfate and nitrate emissions reduction increasing pH~~ The effect of NO_3^-, SO_4^{2-} and TNH_3 on pH when using the ISORROPIA-II thermodynamic equilibrium model to simulate pollutant concentration reduction are shown in Table S3. With the decrease of NO_3^- and SO_4^{2-}, the pH value increases, but NO_3^- has no obvious effect on the pH value, SO_4^{2-} has obvious effect on the pH value, which indicates that the formation of SO_4^{2-} in the aerosol can increase the acidity (Sun et al., 2014). The greater the reduction of TNH_3, the lower the pH value is, which shows that the presence of NH_3 as an alkaline gas can alleviate some of the acidity produced by SO_4^{2-} (Cheng et al., 2016). When the synergistic control of pollutants is reduced, it also has a certain effect on pH, increasing from 4.07 to 4.16. Some studies believe that if the ammonia emissions are reduced significantly, the risk of acid rain may increase (Liu et al., 2019c). As shown in Fig. S13, the acid rain problem in China is mainly concentrated in southern China, especially in southwestern China, southern China, and the Yangtze River Delta in eastern China. Therefore, how to adjust the emission reduction ratio in combination with the characteristics of regional air pollution and energy consumption and thus help reduce the problem of aerosol acidity changes caused by air pollution reduction is a problem

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worthy of in-depth study. Therefore, when controlling NOx, SO_2 and NH_3 emissions, it is necessary to consider the aerosol acid and alkali changes caused by emission reduction.

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~~and NH3 emissions reduction reducing pH, and synergistic emission reduction has the least impact on pH changes, so controlling the emissions reduction ratio in the air pollutant emission reduction scheme to reduce the impacts of aerosol pH is worth further study. Acid rain is mostly concentrated in southern China, and there are also important acid rain problems in the Sichuan Basin (Fig. S12). Therefore, while controlling NSA emissions, especially controlling ammonia emissions, the potential environmental problems of acid rain are worth comprehensive assessment and analysis.~~

~~Table 3. Sensitivity analysis of SNA concentration variations during the different observation periods.~~

~~Periods~~	~~Variables~~	~~Coefficient of variance~~		
		~~SO_4^{2-}~~	~~NH_4^+~~	~~NO_3^-~~
~~2015-2017~~	~~SO_4^{2-}~~		~~52.62~~	~~5.38~~
	~~NH_3~~	~~2.48~~		~~31.30~~
	~~NO_3^-~~	~~0.002~~	~~49.27~~	
~~2015~~	~~SO_4^{2-}~~		~~58.37~~	~~11.27~~
	~~NH_3~~	~~2.69~~		~~32.83~~
	~~NO_3^-~~	~~0.005~~	~~43.34~~	
~~2016~~	~~SO_4^{2-}~~		~~45.15~~	~~5.55~~
	~~NH_3~~	~~3.09~~		~~38.24~~
	~~NO_3^-~~	~~0.004~~	~~46.27~~	
~~2017~~	~~SO_4^{2-}~~		~~43.64~~	~~2.56~~
	~~NH_3~~	~~1.23~~		~~21.88~~
	~~NO_3^-~~	~~0.001~~	~~58.22~~	
~~Spring~~	~~SO_4^{2-}~~		~~49.72~~	~~3.50~~
	~~NH_3~~	~~2.49~~		~~26.57~~
	~~NO_3^-~~	~~0.001~~	~~40.55~~	
~~Summer~~	~~SO_4^{2-}~~		~~86.23~~	~~27.85~~
	~~NH_3~~	~~1.74~~		~~58.29~~
	~~NO_3^-~~	~~0.002~~	~~34.69~~	
~~Autumn~~	~~SO_4^{2-}~~		~~49.18~~	~~2.71~~
	~~NH_3~~	~~1.87~~		~~32.30~~
	~~NO_3^-~~	~~0.002~~	~~47.34~~	
	~~SO_4^{2-}~~		~~36.08~~	~~6.13~~

Winter	NH_3	1.56		26.76
	NO_3^-	0.005	35.94	

Coefficient of variance: Standard deviation/Mean value*100

3.5 Characteristics of local emissions and regional transport

3.5.1 Local emissions

The concentration of pollutants is obviously affected by meteorological conditions; for example, WS and WD can affect the accumulation and removal of pollutants (Li et al., 2016). Figs. S12S14-14 16 show the annual variation characteristics of SNA and gas precursors affected by the WS and WD using CPF. Overall, the higher WS was accompanied by a lower pollutant concentration. As the WS decreases, the pollution becomes serious, and the pollution hot spots were gradually concentrated. On the whole, when the WS was usually greater than 2 m/s, the pollution was light (pollutant concentration percentile was between 0-25). When WS was usually less than 1 m/s, the pollution was heavy (pollutant concentration percentile was between 75-100), the which also reflects the distance and orientation between the emission source and the observation station, indicating that when the pollution was serious, the contribution of local source emissions was more prominent.

NO_3^-Nitrate and NOx have similar distributions of pollution hot spots in the polar plot diagram (Fig. S13S14), and when the concentration percentile was between 0-25, the hot spotsy were concentrated in the northeast and southeast directions and widely distributed. When the concentration percentile was between 25-75, the sources of NO_3^-nitrate and NOx were distributed west, southwest and northeast of the observation siteobservatory, and there were important contribution sources in the northwest direction (WS was approximately 3-4 m/s) in 2017. When the WS was approximately 1-2 m/s and the concentration percentile was between 50-75, the important NOx source was in the northwest direction. When the accumulation of pollution concentration was high (concentration percentile was between 75-100), the NO_3^-nitrate source was mainly concentrated in the east and southeast of the observation stationsite, and NOx was distributed in the south and southeast, with WSs of less than 1 m/s;—.

~~additionally~~Additionally, the distribution of pollution hot spots was relatively wide in 2016 (the annual mean values of NOx were 42.15, 43.99 and 39.63 (ppb) in 2015, 2016 and 2017, respectively) , indicating that the source was relatively wide, which may be one of the reasons for the relatively high concentration. The SO_4^{2-} ~~sulfate~~ and SO_2 pollution sources affected by meteorological conditions also have similar distribution characteristics (Fig. ~~S14~~S15). At a higher concentration of pollutants, the pollution hot spots of SO_4^{2-} ~~sulfate~~ were distributed in the east and southeast of the observation ~~station~~site, and SO_2 was distributed in the northeast, southeast and west. ~~The concentrations of SO_2 were 5.44, 4.15 and 3.68 (ppb) in 2015, 2016 and 2017, respectively.~~ Compared with 2017 and 2016, the distribution of SO_2 pollution sources in 2016 was also more extensive, mainly in the west and northeast. The NH_3 emissions were slightly different from those of NOx and SO_2~~SO_2 and NOx~~ (Fig. ~~S15~~S16). Under conditions of high pollution concentration (concentration percentile was between 75-100), the pollution hot spots were distributed in the west in 2015 (WS was approximately 2-3 m/s), in the north in 2016 (WS was approximately ~~3~~ 1 and 3 m/s), and in the near distance in 2017 (WS was approximately 0.5 m/s). The higher pollution concentration was accompanied by a relatively higher WS (2015 and 2016), which indicates that the NH_3 emission transport in the surrounding area was more obvious, which may come from the surrounding agricultural source distribution area (Liu et al., 2019b;Liu et al., 2013b). The annual mean value of NH_3 emissions in 2017 was 27.91 ppb, which is significantly higher than those in 2015 and 2016 at 17.93 ppb and 16.55 ppb, respectively. During the 25-50 concentration percentile period of the NH_3, there was a WS of approximately 2 m/s east of the observation site, and during the 50-75 concentration percentile period, there was an obvious source northwest of the observation site, with a WS of approximately 4 m/s. During the 75-100 concentration percentile periods, the pollution sources were mainly local. This shows that in 2017, in addition to the pollution sources being distributed in the east and southeast, the higher NH_3 emissions were also contributed by the surrounding emission sources northwest of Chengdu.

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3.5.2 Gaseous precursors of NSA regional transport

The PSCF is used to analyse the potential source distribution of pollutants to determine the regional transport characteristics of pollutants (Ji et al., 2019). In addition, considering the aerosol lifetime, SO_2 (about 9.6 d) and NOx (about 1 d) are also very different (Guo et al., 2014), and the research also shows that NH_3 is significantly contributed by local source emissions (Walker et al., 2004). Therefore, we comprehensively consider to select 24-hour backward trajectory to carry out PSCF simulation in Chengdu region. Fig. 9 shows the PSCF analysis of NOx, SO_2 and NH_3, with significant differences in their potential source distributions. The higher PSCF value of NOx was mainly distributed ~~within 300 km~~ west, northwest and southwest of Chengdu in 2015 ~~(Ya'an, Meishan, Leshan and western Chengdu)~~, northwest and south of Chengdu in 2016 ~~(Deyang, Meishan, Leshan and northwestern Chengdu)~~, and south~~-~~, west and northeast of Chengdu in 2017 ~~(Deyang, Mianyang, Meishan, Leshan and western Chengdu)~~. The PSCF of NO_2 and NO (Fig. S17) also reflects that their potential sources are mainly influenced by the interior of Sichuan Province, especially in the cities around Chengdu. Chengdu is located along the western margin of the Sichuan Basin. It was also observed through satellite remote sensing data that the higher NO_2 emissions in the Sichuan Basin are distributed in Chengdu and Chongqing (Fig. ~~S16~~S18a). ~~As shown in Fig. 9, the higher PSCF values were concentrated in the surrounding areas of Chengdu, indicating that the Chengdu NOx was mainly from local emissions.~~ The SO_2 emissions were widely distributed, mainly in the Sichuan Basin. Among them, Leshan city and Meishan city south of Chengdu have higher SO_2 emissions, and another higher emission source was distributed in Chongqing (Fig. ~~S16~~S18b). The PSCF analysis of SO_2 shows that the higher PSCF values were distributed in the ~~western~~southern, western ~~southern~~ and southwestern parts of Chengdu, ~~and the western, southern and southwestern marginal regions of the Sichuan Basin were also important potential distribution areas.~~ Therefore, a comparison Figs. 9 and ~~S16~~ S18b shows that the main source of SO_2 may be distributed in the southern ~~western,~~ , western ~~southern~~ and southwestern margin region~~edge areas~~ of the Sichuan Basin. In particular, Leshan, Ya'an and Meishan were important potential sources. As

shown in the PSCF analysis of NH_3 in Fig. 9, the higher PSCF was also concentrated in the inner Sichuan Basin, especially in the urban agglomeration around Chengdu~~There were different sources of NH_3 emissions from 2015 to 2017, mainly distributed in the Sichuan Province~~. In 2015, the potential source of NH_3 was mainly distributed in the southwest and northeast of Chengdu, with higher PSCF in Nanchong and other regions. In the southwest, it was concentrated in Ya'an, Meishan and Leshan. In 2016, potential sources were mainly distributed in the southwest of Ya'an, Meishan, Leshan and the southern part of the Ganzi Tibetan Autonomous Prefecture. There were two characteristics of potential sources in 2017. A relatively light source was relatively close to Chengdu, and the high PSCF was in Chengdu, which can be considered the contribution of local emissions. The other contribution is obvious as a long-distance potential source contribution, mainly in some cities in the northeast, Nanchong, Guangyuan and Mianyang, and to a certain extent at the junction of Shaanxi, Gansu and Sichuan. In 2017, in addition to the contribution of local emissions, the contribution of regional transport in the northeast may also be an important reason for the higher NH_3 concentration. ~~In 2015, the potential sources were mainly west of the Sichuan Basin, southwest of Chengdu city, which is approximately 100 km away from an important source. A higher PSCF was mainly distributed in Ya'an, Leshan, Meishan and Yibin in 2016. In 2017, the higher PSCF was mainly distributed in the western and northern areas of the Sichuan Basin, as well as Meishan and Leshan, which were close to Chengdu and contributed significantly. Northwest of Chengdu, Deyang, Mianyang and Guangyuan were important potential sources, and a small part also comes from south of Gansu and Shaanxi.~~ Fig. S19 shows satellite remote sensing data of NH_3, overall, the higher NH_3 column concentration is distributed in the Sichuan Basin, mainly concentrated in the region near Chengdu, showing that NH_3 is more discharged in the Sichuan Basin, especially in the surrounding areas of Chengdu. In addition, through the analysis of the Multiresolution Emission Inventory for China (MEIC), it is also found that the higher NOx, SO_2 and NH_3 emissions in Sichuan Basin are mainly concentrated in Sichuan Basin, as shown in Fig. S20. It can also be seen that NOx is mainly concentrated in more-developed Chengdu and Chongqing, SO_2 emissions are

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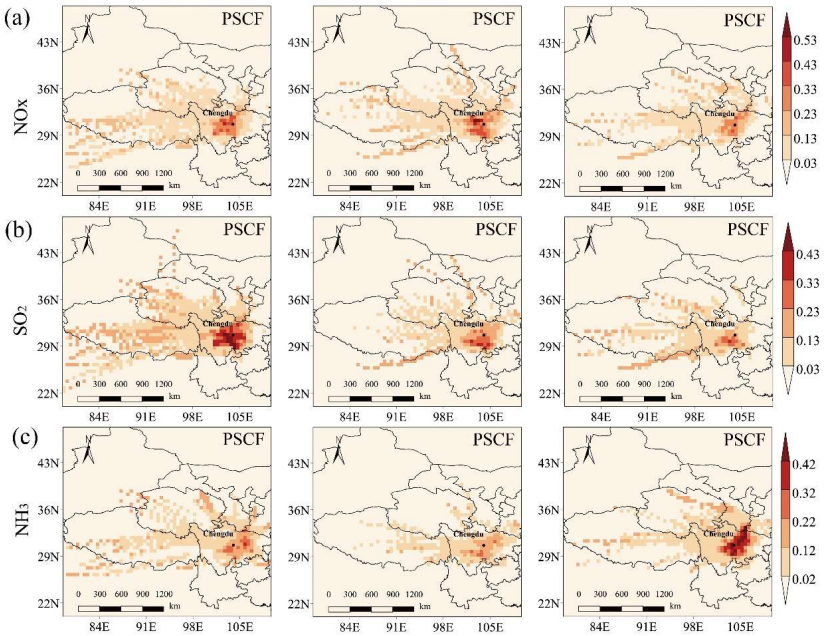
obvious in Chengdu and Western Chongqing, NH_3 emissions are widely distributed, and there are higher emission characteristics in Chengdu and its surrounding areas. Therefore, according to the analysis of pollution emissions and PSCF in Chengdu, it is necessary to strengthen regional air pollution control and take regional joint prevention and control measures to reduce the impact of air pollutant regional transport.

~~Fig. S17 shows the Multiresolution Emission Inventory for China (MEIC) Gridded emissions of NH_3 in 2016 (www.meicmodel.org). The higher NH_3 emissions were mainly concentrated in the interior of the Sichuan Basin, especially near Chengdu, the western edge of the basin. In comparison with Figs. 9 and S17, the regions with potential impacts on NH_3 concentration in Chengdu were mainly distributed in the Sichuan Basin, especially south and northeast of Chengdu.~~



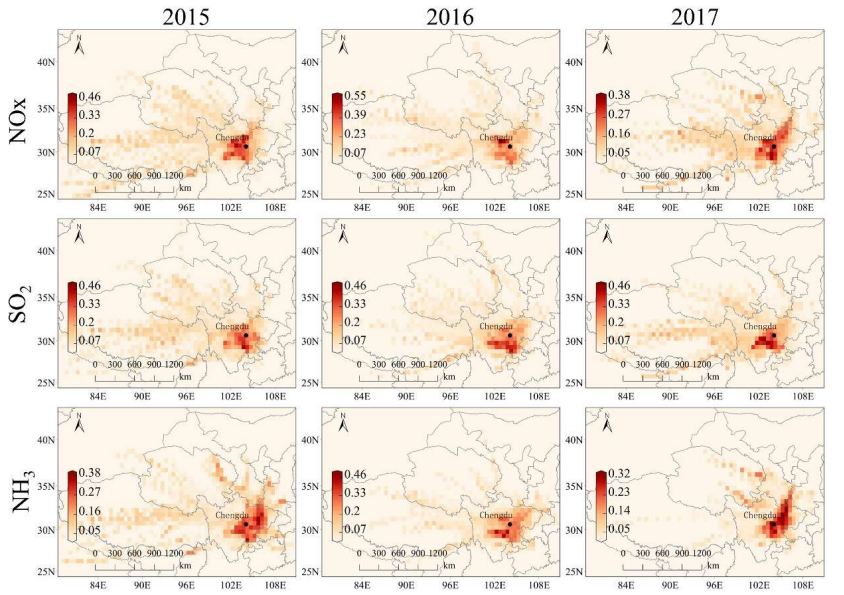


Fig. 9. PSCF (potential source contribution function) of NOx, SO_2 and NH_3 ~~(ppb)~~ in Chengdu from 2015 to 2017.

4 Conclusions

The ~~long-term~~three-year observation experiment with hourly resolution of NSA from January 1, 2015 to December 31, 2017 was carried out in Chengdu in southwest China, which is in the Sichuan Basin. The pollution characteristics of ~~SNA's~~ NSA's annual, monthly, seasonal, diurnal and weekly variations were demonstrated. The characteristics of chemical conversion~~, the relationship with carbonaceous aerosols,~~ and the sensitivity of emission reduction control were analysed. Finally, combined with meteorological ~~factors~~ parameters and PSCF simulation, the local emission and regional transport characteristics of NSA gaseous precursors were also illustrated. The main conclusions were as follows:

(1) ~~Compared with 2015, the concentration of NO_3^- in 2017 did not decrease significantly, while the concentrations of SO_4^{2-} and NH_4^+ decreased.~~ With the increase in $PM_{2.5}$ concentration, the NSA mass concentration increased, accounting for 31.45-

37.78% of $PM_{2.5}$, and the contribution of NSA is higher than that of carbon aerosol (OM and EC).~~but there was a downward trend, indicating that the contribution of other unknown components to $PM_{2.5}$ may significantly increase with the aggravation of pollution.~~ From 2015 to 2017, the contribution of NO_3^- to $PM_{2.5}$ increased, and in 2017, it became the main contribution component of NSA, and it plays an important role in the concentration accumulation of $PM_{2.5}$. Higher and lower NSA concentrations were seen in winter and summer, respectively, and higher concentrations were seen more during the day than at night. Although the NSA concentration on weekdays was slightly higher than that on weekends, the mean difference between them was nonsignificant.

(2) With the increase in $PM_{2.5}$ concentration, there is an increasing trend of NOR and SOR, which indicates that the formation of NO_3^- and SO_4^{2-} increases obviously, and the increase of RH will promote the formation of NO_3^- and SO_4^{2-}. Using ISORROPIA-II thermodynamic equilibrium model, it is found that NSA in aerosols is more likely to combine with AWC, which indicates that the aqueous environment of aerosols plays an important role in promoting the formation of NSA. The analysis of the interaction between NSA also confirmed that the NH_4^+ will first combine with SO_4^{2-} to form $(NH_4)_2SO_4$, and the remaining NH_4^+ will combine with NO_3^- to form NH_4NO_3. The sensitivity analysis of NSA concentration shows that reducing NOx and SO_2 is beneficial to reducing NSA contribution in $PM_{2.5}$, but their changes also have an important impact on the pH of aerosols.

~~_aqueous phase oxidation was an important method of NSA chemical conversion. The ammonia-rich environment in Chengdu became increasingly obvious. Under this condition, the main strategy to reduce the concentration of NSA was to continue to promote sulfate reduction and to strengthen the control of nitrate and ammonium reductions. When controlling the decrease in sulfate and nitrate, the decrease in ammonium will be obvious. SNA Synergistic emissions reduction control implementation can achieve a better emission reduction effect. Regulation of the emission reduction ratio of NSA and reduction of the impact on aerosol pH was also a problem worth further consideration.~~

(3) Local emissions and regional transport of NSA gaseous precursors have an

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important impact on air pollution in Chengdu. When the pollution is aggravated, the contribution of NOx and SO_2 to the local emission is relatively obvious. In addition to the local emission of NH_3, the contribution of pollution sources around Chengdu is also relatively obvious. PSCF analysis shows that the potential sources of pollution transmission in Chengdu are mainly distributed in Sichuan Province, and the most prominent contribution is made in Sichuan Basin, especially among the cities around Chengdu.~~In particular, NOx was the most obvious contribution from the western, southern and southwestern margins of the Sichuan Basin and local emissions in Chengdu. Northeast and west of Chengdu, there were high local SO_2 emission sources, and combined with satellite remote sensing data and PSCF analyses, within the Sichuan Basin, the cities of Leshan and Meishan south of Chengdu may be important sources of SO_2 regional transport. The potential sources of NH_3 were widely distributed, and the internal emissions of the Sichuan Basin may be important potential contribution sources. Southwest, south and southeast of Chengdu, the contribution was obvious.~~ The analysis of local emissions and regional transport shows that it is necessary to implement joint prevention and control of air pollution in the Sichuan Basin.~~The analysis of local emissions and regional transport shows that implementing regional joint prevention, controlling emissions reduction working mechanisms and simultaneously promoting pollutant emission control are important implementation plans.~~

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Data availability

The data are available on request to the corresponding author.

Author contribution

XL, QT and LK designed and led this study. QT and MF ~~was~~ were responsible for the observations. LK, MF, YL, YZ, CZ, CL analyzed the data. LK, YQ, JA, NC, YD, RZ and ZW discussed the results. LK and XL wrote the paper. All authors commented on

the paper.

Competing interests

The authors declare that they have no conflicts of interest.

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