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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal characteristics, formation mechanisms and source origins of $PM_{2.5}$ in two megacities in Sichuan Basin, China" *by* Huanbo Wang et al.

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We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed all of the comments carefully, as detailed below. Our responses start with "R:".

General Comments:

This manuscript elucidates the seasonal variations, the formation mechanism, and the sources of PM2.5 in two megacities in Sichuan Basin. The concentrations of major chemical components of PM2.5 in different seasons are investigated. The chemical

characteristics of clean days and polluted days are presented to study the formation of key chemical species as well as transportation pathway of secondary aerosols. In general, the research results provide meaningful information on both formation mechanism and environmental control strategies of PM2.5. However, there are still some key issues which need to be addressed before possible publication.

R: We have carefully studied the several key issues raised by this and another reviewer and revised the paper accordingly. We hope the reviewer will find that the quality of the paper has been improved significantly.

1. The factors contributing to the different temporal patterns of sulfate and nitrate should be further discussed. Specific heterogeneous reaction which may play important roles in polluted days and its major contributing components can be clearly pointed out.

R: We have added information of the gaseous precursors and meteorological parameters to facilitate the more in-depth discussion on the temporal patterns of SO42- and NO3- in the revised manuscript. We agree with the reviewer that heterogeneous reactions likely contributed to the formation of secondary aerosols considering the high relative humidity (RH) (60-88%) conditions during the study campaign. In the revised paper, the formation of secondary aerosols through heterogeneous reactions was discussed in detail based on the relationships between SOR, NOR and SOC/OC and RH. Note that RH exhibited no significant difference between clean and polluted periods in our study, suggesting RH was not the driving force for the polluted episodes in Sichuan Basin. This phenomenon was different from what was found in eastern coastal China and North China Plain, where sharp RH increase was observed during polluted episodes.

2. The discussion on PM2.5 formation process can be combined with the analysis on the variation of gaseous precursors, including SO2 and NO2.

R: As mentioned above, information of gaseous precursors has been added in the re-

vised paper. Temporal variations of PM2.5 and major gaseous precursors were then discussed together. For example, gaseous precursors (SO2 and NO2) and secondary inorganic aerosols (SO42- and NO3-) increased by a factor of 1.5-3.7 during the polluted periods than clean periods. To exclude the influence of atmospheric physical processes on these variations, CO-scaled variations were also provided. The CO-scaled SO42- was 40-70% higher and that of NO3- was 80-120% higher during the polluted periods, while their respective gaseous precursors were no more than 30% higher. These numbers suggested stronger chemical transformation from gaseous precursors to PM2.5 during the polluted periods, as well as the significant contribution of meteorological condition to the high PM2.5 levels.

3. More discussion on geographical sources of different chemical components of PM2.5 is recommended. I also suggest the analysis on the different topography of these two megacities, which will help to better explain the impact of local emission and regional transportation.

R: We have plotted several PSCF maps to illustrate the geographical sources of the different chemical components in PM2.5 and their gaseous precursors. Generally, similar spatial distributions of potential sources for PM2.5 and its chemical components were observed. At the CD site, high concentrations of the pollutants were mostly associated with sources broadly located in the southeast of the basin, covering Neijiang, Zigong, Yibin, Luzhou and east part of Chongqing. At the CQ site, the northeast area of Chongqing was identified as the major sources.

The impact of the special topography and meteorological conditions on PM2.5 levels at the two sampling sites were also discussed in detail. Firstly, CD and CQ sampling sites are both located in the Sichuan Basin, which is surrounded by high mountains. Such topography often forms a barrier for the dispersion of pollutants and causes air stagnation within the basin, therefor facilitates regional-scale pollution episodes inside the Basin. Back trajectory analysis showed that air masses reaching CD and CQ only traveled for short distances, primarily within the Sichuan Basin. This highlighted

the impact of the special topography on PM2.5 pollution at the two sites. Secondly, Chongqing is a famous mountain city with the majority of population using motorcycle instead of bicycle as the main daily travel tools. This contributed more VOCs emissions and high OC concentrations at CQ than CD.

Besides, I have some specific comments on the manuscript as follows:

1. Section 2.5: I'd like to recommend adding detailed equations of PSCF analysis for better understanding.

R: Information added.

2. Line 170-171: Citation format error: "Tao et al. (2013, 2014)" should be corrected into "(Tao et al., 2013, 2014)".

R: Corrected.

3. Line 244: The authors claimed the concentration of NO3- decreased on the polluted days in the warm season of CQ. But in Figure 6(d), the concentration of NO3- is higher in the polluted days. There seems to be contradictory.

R: Thanks for pointing out this negligence, which has been corrected in the revised paper.

4. Section 3.4.2: The authors applied CO-scaled PM2.5 and major components to isolate the impact of meteorological conditions. Specific scaling approach or related references should be provided.

R: The CO-scaled pollutant concentration was calcualetd as the ratio of concentrations between a pollutant of interest and CO (e.g. PM2.5/CO, SO42-/CO, OC/CO). We have added explanaiton and related references (Zheng et al., 2015, Zhang et al., 2014) in the revised paper.

5. Section 3.4.3: I'd like to recommend adding a graph containing RH levels and NO3- concentration between the two sites. Also, a correlation between [NO3-]/[SO42-] and

[NH4+]/[SO42-] is suggested to investigate the difference of NO3- formation between CQ and CD.

R: NO3- concentrations at CD and CQ were compared for all the seasons. NO3- was 58% lower at CQ than CD in summer, but was at similar levels in the other seasons. NO2 and most meteorological parameters (except RH) were comparable at both sites in summer. Thus, the lower NO3- at CQ in summer was likely mainly caused by the lower RH, which inhibited the formation of NH4NO3. We have added a figure in the supplemental information document showing the temporal variations of ambient RH and deliquescence relative humidity (DRH), which was used to explain the different NO3- concentrations between CD and CQ in summer.

We have also added correlation analysis between [NO3-]/[SO42-] and [NH4+]/[SO42-] and related in-depth discussion on the formation mechanism of NO3- in winter in the revised paper.

6. Figure 3: The black dots which indicate the average values should be stated in the figure caption.

R: We have added the description of the legend in this figure.

Reference cited above:

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimoto, T., Chang, D., Poschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, Atmos Chem Phys, 15, 2969-2983, 10.5194/acp-15-2969-2015, 2015.

Zhang, Q., Quan, J. N., Tie, X. X., Li, X., Liu, Q., Gao, Y., and Zhao, D. L.: Effects of meteorology and secondary particle formation on visibility during heavy haze events in Beijing, China, Sci Total Environ, 502, 578-584, 10.1016/j.scitotenv.2014.09.079, 2015.

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