Contribution of Secondary Particles to Wintertime PM$_{2.5}$ During 2015–2018 in a Major Urban Area of the Sichuan Basin, Southwest China

X. X. Du$^{1,2}$, G. M. Shi$^2$, T. L. Zhao$^1$, F. M. Yang$^2$, X. B. Zheng$^3$, Y. J. Zhang$^4$, and Q. W. Tan$^5$

$^1$Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters, Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science and Technology, Nanjing, China; $^2$National Engineering Research Center for Flue Gas Desulfurization, School of Architecture and Environment, Sichuan University, Chengdu, China; $^3$Guizhou Institute of Mountainous Environment and Climate, Guiyang, China; $^4$LAPC, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China; $^5$Chengdu Research Academy of Environmental Protection Science, Chengdu, China

Abstract A novel method, noted as approximate envelope method (AEM), was developed to estimate the secondary PM$_{2.5}$ concentrations based on the air quality monitoring data. This approach made it possible to obtain the long-term characteristics of the secondary PM$_{2.5}$ using only conventional observations. The secondary PM$_{2.5}$ concentrations and their variation characteristics under different PM$_{2.5}$ pollution levels during wintertime of 2015 to 2018 in Chengdu, a major urban area of Sichuan Basin over Southwest China, were analyzed. The results showed that the secondary PM$_{2.5}$ concentrations ranged from 52.3 to 122.8 $\mu$g·m$^{-3}$ and accounted for 56.3%, 63.6%, and 67.4% of the total PM$_{2.5}$ in slight, moderate, and heavy pollution status, respectively. Additionally, the concentrations of secondary PM$_{2.5}$ increased year by year, while the concentrations of primary PM$_{2.5}$ performed significantly decreasing trends from 2015 to 2018. The diurnal variations of primary and secondary PM$_{2.5}$ presented a significantly unimodal patterns but with maximum at morning and noon, respectively. Specific meteorological conditions such as high (low) relative humidity and temperature (wind speed and air pressure) aggravated the secondary particulate pollution in wintertime of Chengdu, reflecting an influence of regional climate change over the Sichuan Basin of Southwest China on the urban air pollution.

1. Introduction

Atmospheric aerosol refers to a heterogeneous system composed of solid and liquid particles suspended in the atmosphere and gas carriers (Sheng et al., 2013), including primary particles emitted directly from various sources and secondary particles generated by homogeneous and heterogeneous atmospheric chemical reactions of gaseous precursor (Lin et al., 2018; Seinfeld & Pandis, 2016). Previous studies showed that the secondary component accounted for 33% to 77% of PM$_{2.5}$ in China (Behera & Sharma, 2010; Huang et al., 2014; Zhang et al., 2018). Specifically, the rapid growth of secondary inorganic and organic aerosol is an important factor causing the accumulation of PM$_{2.5}$ concentration during heavily polluted episodes (Huang et al., 2014).

Chengdu is located at the western edge of Sichuan Basin, which is the central city of southwestern China. Influenced by basin topography, Sichuan Basin was confronted with meteorological conditions favoring the accumulation of atmospheric pollutions, such as obvious calm winds, frequent temperature inversion, and high humidity (Lin et al., 2018; Shi et al., 2019). Consequently, Sichuan Basin was acknowledged as one of the most polluted regions of China (Tao et al., 2017; Zhao et al., 2016). In Chengdu, PM$_{2.5}$ pollution occurred mainly in winter (Zhao et al., 2018), and the secondary inorganic aerosol accounted for nearly half of PM$_{2.5}$ mass concentration with increasing importance of nitrate in recent years (Liao et al., 2017; Tian et al., 2019; Wang et al., 2018).

The concentration of secondary inorganic components could be quantified by measuring the concentration of inorganic water-soluble ions, such as SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ (Wang et al., 2018). These components could be measured by ion chromatograph, thermal carbon analyzer, and other instruments after the particulate matter is collected through filter membrane sampling. The concentration of secondary organic aerosol is...
usually estimated using elemental carbon (EC) tracer method (Castro et al., 1999) or source apportionment method (Huang et al., 2014). Conventional sampling technique was implemented at relatively low time resolution and usually in short sampling period. Although automatic sampling observations with high time resolution have been realized (He et al., 2011; Sun et al., 2010), such methods were often limited by the relatively high cost and high equipment maintenance difficulty (Behera & Sharma, 2010). In contrast, the estimation of secondary aerosols using conventional environment-meteorological observation data can be applied in the analysis of regional long-term climate characteristics (Cui et al., 2013).

The recent studies about the secondary particles in Chengdu generally focus on specific short periods (Liao et al., 2017; Tian et al., 2019; Wang et al., 2018), and the characteristics and variations of secondary aerosols in long-term were seldomly analyzed. In this study, we developed a novel method for estimating the concentration of secondary particles based on the approach of Chang and Lee (2007). And the characteristics of secondary particles during the wintertime of 2015–2018 in Chengdu were articulated under different PM$_{2.5}$ pollution levels.

2. Method and Data Description

2.1. Air Quality and Meteorological Data

The hourly air quality monitoring data, including mass concentrations of PM$_{2.5}$, ozone (O$_3$), carbon monoxide (CO), and nitrogen dioxide (NO$_2$), were provided by the National Urban Air Quality Real-Time Publishing Platform. The air quality data in winter (January, February, and December) of 2015–2018 at seven sites in Chengdu were used in this study. These sites were distributed in the urban area, so the monitored data represented the air quality of urban Chengdu. Moreover, meteorological parameters in Wenjiang station (latitude: 30.4°N, longitude: 103.5°E), such as temperature, relative humidity (RH), air pressure, precipitation, wind speed (WS), and wind direction (WD), were analyzed to explore the relationship between air quality and meteorological conditions. Some observation data of PM$_{2.5}$ components, including inorganic water-soluble ions (SO$_2$$^-$, NO$_3^-$, and NH$_4^+$), EC, and organic carbon (OC), from short-term offline sampling in urban Chengdu (Li et al., 2017; Wu et al., 2019) and online continuous monitoring during winter of 2018 at the Chengdu Academy of Environmental Sciences (CAES) were used to evaluate the estimated secondary PM$_{2.5}$ contribution.

2.2. Method

Chang and Lee (2007) proposed a method to estimate the contribution of primary and secondary aerosols to PM$_{2.5}$. In that approach, CO was selected to be the tracer of primary pollutants emitted by motor vehicles, combustion, and other natural particles. And the observed hourly PM$_{2.5}$/CO ratio under clean conditions, noted as (PM$_{2.5}$/CO)$_C$, was used to represent the ratio of primary aerosol and CO concentrations under polluted conditions. Therefore, the concentration of primary aerosol could be estimated as (PM$_{2.5}$/CO)$_C$ × CO.

In Chang and Lee’s method, an essential assumption was made that the primary sources were the main contributors to the observed PM$_{2.5}$ concentration under clean conditions. However, it is not obvious that this assumption will hold in the real atmosphere. Even under clean conditions, the formation of secondary particles is not ignorable (Levy, 1971).

To minimize the impact of secondary components, we proposed a new method called approximate envelope method (AEM) to estimate the mass ratio of primary PM$_{2.5}$ to ambient CO, noted as R$_p$, instead of using (PM$_{2.5}$/CO)$_C$ to represent this ratio. R$_p$ is related to the emission conditions and should maintain stable in certain emission scenario. Because PM$_{2.5}$ are composed of both primarily emitted and secondarily formed particles, the mass ratio of observed PM$_{2.5}$ to CO, noted as R$_A$, changes with varying oxidation and humidity conditions. The existence of secondary particles makes R$_A$ always larger than R$_p$. Therefore, the lower envelope of the CO-PM$_{2.5}$ scatter plot diagram represents the cases with minimum impact of secondary particles and provides a robust estimation of R$_p$. Only the CO and PM$_{2.5}$ data under clean conditions (PM$_{2.5}$ concentration less than 75 μg·m$^{-3}$) were included in the scatter plot while determining the envelope mentioned above. And considering the varying emission characteristics during different periods of the day, the envelope was separately estimated for each 3-hr period, as shown in Figure 1. For every 3-hr period, PM$_{2.5}$ concentrations were grouped according to CO concentration bins (0.5–0.7, 0.7–0.9, 0.9–1.1, 1.1–1.3, 1.3–1.5, and 1.5–
The three smallest PM$_{2.5}$ concentrations and corresponding CO concentrations in each group were averaged to generate CO–PM$_{2.5}$ pairs. The mean PM$_{2.5}$/CO ratio of these pairs was taken as $R_p$.

According to ambient air quality standards of China (GB 3095–2012), PM$_{2.5}$ were categorized into four groups based on daily mean concentration: less than 75 μg·m$^{-3}$, between 75 and 115 μg·m$^{-3}$, between 115 and 150 μg·m$^{-3}$, and greater than 150 μg·m$^{-3}$, representing the clean air quality and the light, moderate, and heavy levels of PM$_{2.5}$ pollution, respectively. For three polluted conditions, the primary PM$_{2.5}$ ($PM_{2.5}^P$) is estimated by using equation 1.

$$PM_{2.5}^P (x, h) = CO_x \times (R_p)_h,$$

where $x$ could be L, M, and H, representing the light, moderate, and heavy pollution, respectively. And $h$ refers to different times of the day. Hence, the secondary PM$_{2.5}$ concentrations are taken as the difference between the observed mean PM$_{2.5}$ ($PM_{2.5}^O$) and the estimated primary PM$_{2.5}$ ($PM_{2.5}^P$).

$$PM_{2.5}^S (x, h) = (PM_{2.5}^O)_{x, h} - (PM_{2.5}^P)_{x, h}.$$  

To evaluate this newly developed method, we compared the contributions of secondary PM$_{2.5}$ concentrations estimated using AEM and observed PM$_{2.5}$ components. Li et al. (2017) and Wu et al. (2019) measured the PM$_{2.5}$ components, including SO$_2^{2-}$, NO$_3^-$, NH$_4^+$, OC, and EC, in Chengdu by offline sampling during 6–16 January 2015 and 1–20 January 2017, respectively. The secondary inorganic PM$_{2.5}$ concentration was estimated by summing the concentrations of SO$_2^{2-}$, NO$_3^-$, and NH$_4^+$. The secondary organic PM$_{2.5}$ concentration was calculated by Turpin’s empirical formula (Turpin et al., 1990) based on OC and EC concentrations. Relevant concentrations in these two observation periods were listed in Table 1. Because of different sampling locations and instruments, there was a slight difference in the total PM$_{2.5}$ concentration. The proportions of the secondary PM$_{2.5}$ estimated using measured components were 43.0% and 46.0% during two periods, respectively. The results from AEM, 63.3% and 61.2%, were much higher. There are two main reasons for the difference. First, the contribution of some long-range transported primary particles, such as dust, were identified as secondary particles in AEM. This effect causes the overestimation of secondary PM$_{2.5}$ concentrations. Second, the PM$_{2.5}$ pollution during these two sampling periods was extremely severe with average PM$_{2.5}$ concentration of 149.6 μg·m$^{-3}$ and 127.1 μg·m$^{-3}$, respectively. The minimum OC/EC ratio might be overestimated, which made the underestimation of secondary organic aerosol in the Turpin’s empirical formula. More observations under clean condition were available in the relatively longer online monitoring at CAES. As shown in Table 1, the mean secondary PM$_{2.5}$ concentration estimated from
observations accounted for 56.4% of the total PM$_{2.5}$ under lightly polluted conditions. The estimated contribution of secondary PM$_{2.5}$ from AEM, 61.4%, was comparable to the observations. Hence, it could be considered that the results from AEM were reasonable.

3. Results

3.1. Meteorological Conditions for Different Pollution Levels

Meteorological conditions are the driving factors of atmospheric pollution levels (Jacob & Winner, 2009). The diurnal changes of WS, temperature, RH, and air pressure in 2015–2018 for different pollution levels were showed in Figure 2. The PM$_{2.5}$ pollution became more severe as the WS decreases, which indicates that the strong wind was conducive to the diffusion and transport of atmospheric pollutants. Especially, persistent calm winds (WS less than 1.5 m·s$^{-1}$) were observed in heavily polluted cases. The relatively lower temperature in clean conditions shows that the rise of temperature may facilitate the gas–to–particle conversion processes. Meanwhile, the RH in clean conditions was lower as shown in Figure 2c. This implies that low PM$_{2.5}$ concentrations were always accompanied with cold-dry air masses in Chengdu. The PM$_{2.5}$

Table 1

| Period                           | Category          | PM$_{2.5}$ | SO$_2$ | NO$_x$ | NH$_4$ | SOC$^a$ | PM$_{2.5,S}$ | PM$_{2.5,S}$/PM$_{2.5}$ |
|----------------------------------|-------------------|------------|--------|--------|--------|---------|--------------|------------------------|
| 6–16 January 2015                | Li et al. (2017)  | 149.6      | 22.6   | 19.3   | 13.5   | 9.0     | 64.4         | 43.0%                  |
|                                  | This study        | 120.6      | —      | —      | —      | —       | 76.3         | 63.3%                  |
| 1–20 January 2017                | Wu et al., 2019   | 127.1      | 13.6   | 21.4   | 13.3   | 10.2    | 58.5         | 46.0%                  |
|                                  | This study        | 119.4      | —      | —      | —      | —       | 73.1         | 61.2%                  |
| Light pollution cases in 2018    | CAES              | 92.8       | 11.4   | 17.3   | 11.8   | 11.7    | 52.3         | 56.4%                  |
|                                  | This study        | 94.2       | —      | —      | —      | —       | 57.9         | 61.4%                  |

$^a$SOC was calculated by Turpin’s empirical formula.

Figure 2. (a–d) Diurnal variations of meteorological parameters in different polluted conditions in Chengdu during the wintertime of 2015–2018.
concentrations increased along with RH. This phenomenon could be partially interpreted by the accelerated hygroscopic growth of aerosol in higher ambient RH. Conversely, the air pressure in polluted conditions was lower than that in clean conditions (Figure 2d). In summary, the diurnal variations of the meteorological parameters show that higher (lower) temperature and RH (WS and air pressure) aggravated PM2.5 pollution in wintertime of Chengdu.

The frequency and mean WS from 16 WDs for different pollution levels are presented in Figure 3. The distribution of WS at different directions were similar for all pollution levels. The mean WSs were quite low (less than 2.0 m·s$^{-1}$) at all directions and relatively higher for northeasterly and southeasterly winds. Moreover, the mean WSs were slightly higher under clean conditions, which facilitated the accumulation of pollutants under polluted conditions. The prevailing winds were from northeast and northwest under clean and moderately pollution conditions. More northerly winds occurred under slightly polluted conditions. Under heavily polluted conditions, the northwesterly winds with highest WS were almost vanished while more northwesterly winds with lowest WS became dominant. In addition, though the mean WS from southeast was high, the frequency of southeast wind only accounted for 2.7% to 3.6%.

In order to quantify the relationship between meteorological factors and the PM$_{2.5}$ concentrations, correlation analysis was performed to obtain the Pearson correlation coefficients. As shown in Table 2, PM$_{2.5}$ was positively correlated with temperature and RH for all pollution levels while negatively correlated with air pressure. The correlation coefficients in heavy pollution condition were higher than those in other pollution levels for temperature and RH. This showed that the effects of temperature and RH on PM$_{2.5}$ concentrations

Figure 3. (a–d) The frequency and mean wind speed at different wind directions for four air quality levels in Chengdu during the wintertime of 2015–2018.
3.2. Primary and Secondary PM$_{2.5}$ Concentrations in Polluted Conditions

The primary and secondary PM$_{2.5}$ concentrations in three polluted conditions were shown in Table 3. The primary (secondary) PM$_{2.5}$ concentrations were 40.5 (52.3) μg·m$^{-3}$, 48.7 (85.1) μg·m$^{-3}$, and 59.5 (122.8) μg·m$^{-3}$ in lightly, moderately, and heavily polluted conditions, respectively. The primary PM$_{2.5}$ concentrations varied slightly in different pollution conditions, but the secondary PM$_{2.5}$ concentrations increased dramatically with aggravation of pollution. Especially, the concentration of secondary PM$_{2.5}$ for heavy pollution was 2.3 times higher than that for light pollution. As a result, the contribution of secondary PM$_{2.5}$ increased from 56.3% in lightly polluted condition to 67.4% in heavily polluted condition. The pollution was caused by unfavorable diffusion conditions, such as shallow planetary boundary layer (PBL) and stagnant weather, and conducive gas-to-particle conversion conditions, such as high RH (An et al., 2019). The facts that the RH was higher under heavier pollution conditions (Figure 3) and the PM$_{2.5}$ concentrations were positively correlated to RH (Table 2) indicated possible facilitated secondary aerosol formation in polluted cases. The relatively larger discrepancy of secondary PM$_{2.5}$ between different pollution conditions verified that the formation of secondary aerosol was more prominent than unfavorable diffusion conditions.

3.3. Patterns of Primary and Secondary PM$_{2.5}$ in Polluted Conditions

As shown in Figure 4, the diurnal variations of primary and secondary PM$_{2.5}$ presented different characteristics but kept similar in different polluted conditions. The diurnal variations of primary and secondary PM$_{2.5}$ presented unimodal characteristics with peak concentrations at morning and noon, respectively. The primary PM$_{2.5}$ concentrations were obviously influenced by human activity and PBL development. The increase during the morning and evening rush was due to the increase of emissions from traffic peak. The decrease that started at 10:00 a.m. was responsive to the increase of PBL height (PBLH) (Miao et al., 2017). The secondary PM$_{2.5}$ concentration started increasing at 08:00 a.m. and reached the maximum at 13:00 p.m. This might attribute to the accelerated photochemical reaction rate after sunrise. Similarly, the development of PBL led to the following decrease of secondary PM$_{2.5}$ concentration (Wu et al., 2019; Xue et al., 2016).

Figure 5 presented the variations of the wintertime primary and secondary PM$_{2.5}$ concentrations year by year during 2015–2018. The seasonal mean PM$_{2.5}$ concentrations in winter of 2015 was 116.0 μg·m$^{-3}$, the highest value among these 4 years. The PM$_{2.5}$ pollution in Chengdu got effectively controlled after 2015. Despite the concentration in 2017 (105 μg·m$^{-3}$) being close to that in 2015, the concentrations in 2016 and 2018 were significantly lower, about 85.5 and 74.4 μg·m$^{-3}$, respectively. As shown in Figures 5b to 5d, the wintertime primary PM$_{2.5}$ concentrations in Chengdu presented a decreasing trend from 2015 to 2018 in three polluted conditions. This can be attributed to the implementation of emission reduction measures in Chengdu. Conversely, the secondary PM$_{2.5}$ concentrations increased gradually. From 2015 to 2018, the contribution of secondary PM$_{2.5}$ to the total concentrations varied from 52.2% to 61.4% and from 63.9% to 70.8% in lightly and moderately polluted conditions, respectively. The heavy pollution occurred only in 2015 and 2017, and the contribution of secondary PM$_{2.5}$ rose from 64.0% in 2015 to 70.9% in 2017. In

Table 2

| Air quality level        | Sample size | T   | RH   | WS   | P     |
|--------------------------|-------------|-----|------|------|-------|
| Clean air quality        | 92          | 0.13| 0.10 | −0.18| −0.09 |
| Light pollution          | 76          | 0.04| 0.02 | 0.14 | −0.13 |
| Moderate pollution       | 35          | 0.02| 0.04 | −0.22| −0.17 |
| Heavy pollution          | 33          | 0.25| 0.38 | −0.08| −0.12 |

Significantly related at the 0.05 level (both sides).

Table 3

| PM$_{2.5}$ pollution degree | PM$_{2.5}$ | Primary PM$_{2.5}$ | Secondary PM$_{2.5}$ | PM$_{2.5,S}$/PM$_{2.5}$ |
|-----------------------------|------------|--------------------|----------------------|------------------------|
| Light pollution             | 92.8       | 40.5               | 52.3                 | 56.3%                  |
| Moderate pollution          | 133.8      | 48.7               | 85.1                 | 63.6%                  |
| Heavy pollution             | 182.4      | 59.5               | 122.8                | 67.4%                  |
summary, the total wintertime PM$_{2.5}$ concentrations in Chengdu were decreasing while the contribution of secondary particles to the total PM$_{2.5}$ was significantly expanding from 2015 to 2018. This suggests that strict emission control measures of primary PM and PM precursors are of equal importance.

4. Conclusions

Based on the air quality and meteorological parameters during wintertime of 2015–2018, the secondary PM$_{2.5}$ concentrations and their variation characteristics in Chengdu were evaluated. The diurnal variations of primary and secondary PM$_{2.5}$ concentrations both presented unimodal patterns but with maximum at morning and noon, respectively. The contribution of secondary PM$_{2.5}$ to the total PM$_{2.5}$ concentration in Chengdu were 56.3%, 63.6%, and 67.4% in slightly, moderately, and heavily polluted conditions, respectively. The secondary PM$_{2.5}$ concentration varied dramatically when the PM$_{2.5}$ pollution deteriorated, whereas the primary PM$_{2.5}$ concentrations kept relatively stable. This indicated that the formation of secondary particles was more important than the unfavorable diffusion conditions in the accumulation processes of PM$_{2.5}$ in

![Figure 4](image-url)

**Figure 4.** Diurnal variations of primary (a) and secondary (b) PM$_{2.5}$ in different polluted conditions in Chengdu during the wintertime of 2015–2018.

![Figure 5](image-url)

**Figure 5.** Annual variations of seasonal mean PM$_{2.5}$ (a), primary and secondary PM$_{2.5}$ and the contribution of secondary PM$_{2.5}$ in lightly (b), moderately (c), and heavily (d) polluted conditions in Chengdu during the wintertime of 2015–2018.

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Chengdu. In all polluted conditions, the contribution of secondary PM$_{2.5}$ holds increasing trends from 2015 to 2018, reaching 61.4% and 70.8% in slightly and moderately polluted conditions in 2018, respectively. This revealed that the mitigation of secondary PM$_{2.5}$ precursors should be the focus of pollution control in Chengdu.

The newly developed AEM method was implemented to estimate the ratio between primary PM$_{2.5}$ and CO concentrations at different times of the day. And the primary and secondary PM$_{2.5}$ concentrations could be calculated further. Comparison between the estimated secondary PM$_{2.5}$ from AEM and PM$_{2.5}$ component observations indicated that this method overestimated the contribution of secondary PM$_{2.5}$. But it still provided reasonable estimation. More importantly, it requires only conventional air-quality monitoring data, which are available at more locations and during longer periods than PM$_{2.5}$ component observations. Therefore, this method has the potential to be widely applied to acquire the contribution of secondary PM$_{2.5}$ in long-term perspective.

Data Availability Statement

The data presented in this study were archived at https://figshare.com/s/dda9b69fe83080ccbb7d (DOI: 10.6084/m9.figshare.12135945).

References

An, Z. S., Huang, R. J., Zhang, R. Y., Tie, X. X., Li, G. H., Cao, J. J., et al. (2019). Severe haze in northern China: A synergy of anthropogenic emissions and atmospheric processes. *Proceedings of the National Academy of Sciences of the United States of America*, 116(18), 8657–8666. https://doi.org/10.1073/pnas.1900125116

Behera, S. N., & Sharma, M. (2010). Reconstructing primary and secondary components of PM$_{2.5}$ composition for an urban atmosphere. *Aerosol Science and Technology*, 44(11), 983–992. https://doi.org/10.1080/0001873X.2010.504245

Castro, L. M., Pio, C. A., Harrison, R. M., & Smith, D. J. T. (1999). Carbonaceous aerosol in urban and rural European atmospheres: Estimation of secondary organic carbon concentrations. *Atmospheric Environment*, 33(17), 2771–2781. https://doi.org/10.1016/S1352-2310(98)00331-8

Chang, S. C., & Lee, C. T. (2007). Secondary aerosol formation through photochemical reactions estimated by using air quality monitoring data in Taipei City from 1994 to 2003. *Atmospheric Environment*, 41(19), 4002–4017. https://doi.org/10.1016/j.atmosenv.2007.01.040

Cui, H. X., Wu, Y. M., Duan, Y. S., Fu, Q. Y., Zhang, Y. H., Wang, D. F., & Wang, Q. (2013). Secondary aerosol formation through photochemical reactions estimated by using air quality monitoring data in the downtown of Pudong, Shanghai (in Chinese). *Environmental Science*, 34(5), 2003. https://doi.org/10.13227/j.hjxk.2013.05.041

He, K. B., Yang, F. M., Duan, F. K., & Ma, Y. L. (2011). *Atmospheric particulate matter and regional complex pollution (in Chinese)*. Beijing: Science Press.

Huang, R. J., Zhang, Y., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y., et al. (2014). High secondary aerosol contribution to particulate pollution during haze events in China. *Nature*, 514(7521), 218–222. https://doi.org/10.1038/nature13774

Jacob, D. J., & Winner, D. A. (2009). Effect of climate change on air quality. *Atmospheric Environment*, 43(1), 51–63. https://doi.org/10.1016/j.atmosenv.2008.09.051

Levy, H. (1971). Normal atmosphere—Large radical and formaldehyde concentrations predicted. *Science*, 173(3992), 141–143. https://doi.org/10.1126/science.173.3992.141

Li, J. L., Tan, Q. W., Zhang, Y. H., Feng, M., Qu, Y., An, J. L., & Liu, X. G. (2017). Characteristics and source apportionment of PM$_{2.5}$ during persistent extreme haze events in Chengdu, Southwest China. *Environmental Pollution*, 230, 718–729. https://doi.org/10.1016/j.envpol.2017.07.029

Liao, T. T., Wang, S., Ai, J., Gui, K., Duan, B., Zhao, Q., et al. (2017). Heavy pollution episodes, transport pathways and potential sources of PM$_{2.5}$ during the winter of 2013 in Chengdu (China). *Science of the Total Environment*, 584-585, 1056–1065. https://doi.org/10.1016/j.scitotenv.2017.01.160

Lin, Y. L., Zou, J. L., Yang, W., & Li, C. Q. (2018). A review of recent advances in research on PM$_{2.5}$ in China. *International Journal of Environmental Research and Public Health*, 15(3). https://doi.org/10.3390/ijerph150303438

Miao, Y. C., Guo, J. P., Liu, S. H., Liu, H., Li, Z. Q., Zhang, W. C., & Zhai, P. M. (2017). Classification of summertime synoptic patterns in Beijing and their associations with boundary layer structure affecting aerosol pollution. *Atmospheric Chemistry and Physics*, 17(4), 3997–4010. https://doi.org/10.5194/acp-2016-1048

Seinfeld, J. H., & Pandis, S. N. (2016). *Atmospheric chemistry and physics: From air pollution to climate change*, (3rd ed.). Hoboken, NJ: John Wiley & Sons, Inc.

Sheng, P. X., Mao, J. T., Li, J. G., Zhang, A. C., Sang, J. G., & Pan, N. X. (2013). *Atmosphere physics (in Chinese)*. (2nd ed.). Beijing: Peking University Press.

Shi, G. M., Yang, F. M., Zhang, L. M., Zhao, T. L., & Hu, J. (2019). Impact of atmospheric circulation and meteorological parameters on wintertime atmospheric extinction in Chengdu and Chongqing of Southwest China during 2001–2016. *Aerosol and Air Quality Research*, 19(7), 1538–1554. https://doi.org/10.4209/aaqr.2018.09.0336

Sun, J. Y., Zhang, Q., Canagaratna, M. R., Zhang, Y., Ng, N. L., Sun, Y., et al. (2010). Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an aerodyne aerosol mass spectrometer. *Atmospheric Environment*, 44(1), 131–140. https://doi.org/10.1016/j.atmosenv.2009.03.020

Tao, J., Zhang, L. M., Cao, J. J., & Zhang, R. J. (2017). A review of current knowledge concerning PM$_{2.5}$ chemical composition, aerosol optical properties and their relationships across China. *Atmospheric Chemistry and Physics*, 17(15), 9485–9518. https://doi.org/10.5194/acp-17-9485-2017
Tian, M., Liu, Y., Yang, F. M., Zhang, L. M., Peng, C., Chen, Y., et al. (2019). Increasing importance of nitrate formation for heavy aerosol pollution in two megacities in Sichuan Basin, Southwest China. *Environmental Pollution*, 250, 898–905. https://doi.org/10.1016/j.envpol.2019.04.098

Turpin, B. J., Cary, R. A., & Huntzicker, J. J. (1990). An in situ, time-resolved analyzer for aerosol organic and elemental carbon. *Aerosol Science and Technology*, 12(1), 161–171. https://doi.org/10.1080/02786829008959336

Wang, H. B., Tian, M., Chen, Y., Shi, G., Liu, Y., Yang, F., et al. (2018). Seasonal characteristics, formation mechanisms and source origins of PM2.5 in two megacities in Sichuan Basin, China. *Atmospheric Chemistry and Physics*, 18(2), 865–881. https://doi.org/10.5194/acp-18-865-2018

Wu, M., Wu, D., Xia, J. R., Zhao, T. L., & Yang, Q. J. (2019). Analysis of pollution characteristics and sources of PM2.5 chemical components in Chengdu in winter (in Chinese). *Environmental Science*, 40(1), 76–85. https://doi.org/10.13227/j.hjkx.201805035

Xue, L. K., Gu, R., Wang, T., Wang, X., Saunders, S., Blake, D., et al. (2016). Oxidative capacity and radical chemistry in the polluted atmosphere of Hong Kong and pearl river delta region: Analysis of a severe photochemical smog episode. *Atmospheric Chemistry and Physics*, 16(15), 9891–9903. https://doi.org/10.5194/acp-16-9891-2016

Zhang, Y. Y., Lang, J., Cheng, S., Li, S., Zhou, Y., Chen, D., et al. (2018). Chemical composition and sources of PM1 and PM2.5 in Beijing in autumn. *Science of the Total Environment*, 630, 72–82. https://doi.org/10.1016/j.scitotenv.2018.02.151

Zhao, S. P., Yu, Y., Yin, D. Y., He, J. J., Liu, N., Qu, J. J., & Xiao, J. H. (2016). Annual and diurnal variations of gaseous and particulate pollutants in 31 provincial capital cities based on in situ air quality monitoring data from China National Environmental Monitoring Center. *Environment International*, 44(1), 131–140. https://doi.org/10.1016/j.atmosenv.2009.03.020

Zhao, S. P., Yu, Y., Yin, D. Y., Qin, D. H., He, J. J., & Dong, L. X. (2018). Spatial patterns and temporal variations of six criteria air pollutants during 2015 to 2017 in the city clusters of Sichuan Basin, China. *Science of the Total Environment*, 624, 540–557. https://doi.org/10.1016/j.scitotenv.2017.12.172