



A review of current knowledge concerning PM_{2.5} chemical composition, aerosol optical properties and their relationships across China

Jun Tao¹, Leiming Zhang², Junji Cao³, and Renjian Zhang⁴

¹South China Institute of Environmental Sciences, Ministry of Environmental Protection, Guangzhou, China

²Air Quality Research Division, Science and Technology Branch, Environment and Climate Change Canada, Toronto, Canada

³Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China

⁴Key Laboratory of Regional Climate-Environment for Temperate East Asia, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China

Correspondence to: Leiming Zhang (leiming.zhang@canada.ca)

Received: 23 March 2017 – Discussion started: 18 April 2017

Revised: 30 June 2017 – Accepted: 3 July 2017 – Published: 8 August 2017

Abstract. To obtain a thorough knowledge of PM_{2.5} chemical composition and its impact on aerosol optical properties across China, existing field studies conducted after the year 2000 are reviewed and summarized in terms of geographical, interannual and seasonal distributions. Annual PM_{2.5} was up to 6 times the National Ambient Air Quality Standards (NAAQS) in some megacities in northern China. Annual PM_{2.5} was higher in northern than southern cities, and higher in inland than coastal cities. In a few cities with data longer than a decade, PM_{2.5} showed a slight decrease only in the second half of the past decade, while carbonaceous aerosols decreased, sulfate (SO₄²⁻) and ammonium (NH₄⁺) remained at high levels, and nitrate (NO₃⁻) increased. The highest seasonal averages of PM_{2.5} and its major chemical components were typically observed in the cold seasons. Annual average contributions of secondary inorganic aerosols to PM_{2.5} ranged from 25 to 48 %, and those of carbonaceous aerosols ranged from 23 to 47 %, both with higher contributions in southern regions due to the frequent dust events in northern China. Source apportionment analysis identified secondary inorganic aerosols, coal combustion and traffic emission as the top three source factors contributing to PM_{2.5} mass in most Chinese cities, and the sum of these three source factors explained 44 to 82 % of PM_{2.5} mass on annual average across China. Biomass emission in most cities, industrial emission in industrial cities, dust emission in northern cities and ship

emission in coastal cities are other major source factors, each of which contributed 7–27 % to PM_{2.5} mass in applicable cities.

The geographical pattern of scattering coefficient (b_{sp}) was similar to that of PM_{2.5}, and that of aerosol absorption coefficient (b_{ap}) was determined by elemental carbon (EC) mass concentration and its coating. b_{sp} in ambient condition of relative humidity (RH) = 80 % can be amplified by about 1.8 times that under dry conditions. Secondary inorganic aerosols accounted for about 60 % of aerosol extinction coefficient (b_{ext}) at RH greater than 70 %. The mass scattering efficiency (MSE) of PM_{2.5} ranged from 3.0 to 5.0 m² g⁻¹ for aerosols produced from anthropogenic emissions and from 0.7 to 1.0 m² g⁻¹ for natural dust aerosols. The mass absorption efficiency (MAE) of EC ranged from 6.5 to 12.4 m² g⁻¹ in urban environments, but the MAE of water-soluble organic carbon was only 0.05 to 0.11 m² g⁻¹. Historical emission control policies in China and their effectiveness were discussed based on available chemically resolved PM_{2.5} data, which provides the much needed knowledge for guiding future studies and emissions policies.

1 Introduction

Knowledge of spatiotemporal variations in chemical and optical properties of atmospheric aerosols is needed in addressing regional and global air quality and climate issues (Fuzzi et al., 2015; Ginoux et al., 2012; Li et al., 2016c; Liao et al., 2015; Monks et al., 2009; Qian et al., 2015). Aerosol concentrations across China have been at extremely high levels in the recent two decades, largely caused by rapidly increased energy consumption (Chan and Yao, 2008; Fang et al., 2009; Guan et al., 2014; Wang and Hao, 2012; Zhang et al., 2013b). The frequency of haze has also been increased significantly due to light extinction of atmospheric aerosols, especially PM_{2.5} (Li and Zhang, 2014; Pui et al., 2014; Watson, 2002). The Ministry of Environmental Protection of China thus promulgated NAAQS to include daily and annual PM_{2.5} standards starting in early 2012. As a result, real-time PM_{2.5} data in 74 Chinese cities have been recorded since 2013.

Light extinction, the sum of light scattering and absorption, is controlled by not only PM_{2.5} levels, but also its chemical composition, size distribution and hygroscopic potential of its major components, and meteorological conditions (Hand and Malm, 2007a; Malm et al., 2003; Pitchford et al., 2007; Zhang et al., 2014a). High humidity combined with large fractions of hygroscopic chemical components (e.g., sulfate, nitrate, ammonium and some organic matter) can enhance light extinction and haze intensity (Liu et al., 2011, 2013b; Zhang et al., 2015b; Zieger et al., 2013). A large number of studies has been conducted in China in recent years investigating PM_{2.5} composition, aerosol optical properties, aerosol hygroscopic properties and haze formation mechanisms (Guo et al., 2014; Jing et al., 2015; Liang et al., 2016; Liu et al., 2011, 2012; Pan et al., 2009; Tao et al., 2014b; Wang et al., 2015b; Yan et al., 2008, 2009; Yang et al., 2011b; Zheng et al., 2016). However, knowledge of long-term trends of PM_{2.5} concentration, especially its major chemical components, is still limited (Fontes et al., 2017), and few studies have focused on the geographical pattern of PM_{2.5} composition across China and its impact on aerosol optical properties (Li et al., 2017b). The present study aims to gain such knowledge through a thorough review of available studies.

Considering the large number of publications, only ground measurement data of chemical composition of PM_{2.5}, aerosol scattering and absorption coefficients, and aerosol hygroscopic properties published after the year 2000 in scientific papers of Science Citation Index (SCI) journals are reviewed and summarized in this study. A total of about 150 articles met the above criteria including 100 articles on PM_{2.5} chemical composition and source apportionment, 40 articles on aerosol optical properties and 10 articles on aerosol hygroscopic properties. Many of these articles focused on several of the biggest cities, such as Beijing, Shanghai, Guangzhou and Hong Kong, while other studies focused on cities such as Tianjin, Shijiazhuang, Jinan, Nanjing, Hangzhou, Fuzhou, Xiamen, Shenzhen, Chengdu, Chongqing, Xi'an, Lanzhou,

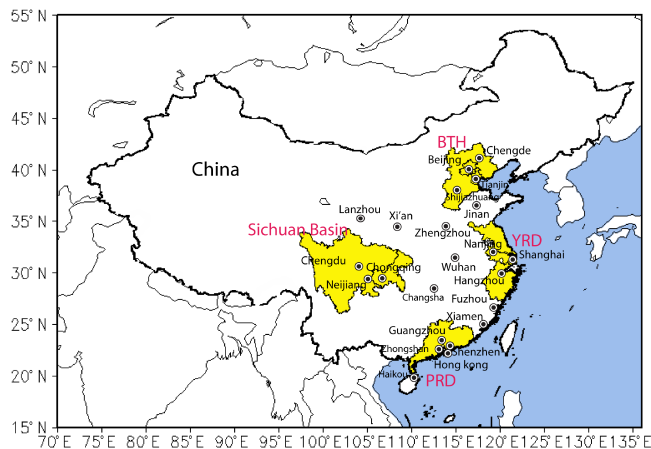


Figure 1. Geographical regions and location of cities with measurements.

Zhengzhou, Wuhan, Changsha, Haikou and several background sites (Fig. 2). Geographical and temporal patterns of PM_{2.5} and its major chemical components including SO₄²⁻, NO₃⁻, NH₄⁺, organic carbon (OC) and EC, and aerosol optical properties are generated, source-apportionment analysis results are summarized and relationships between aerosol optical properties and PM_{2.5} chemical composition are explored. Recommendations are also provided for alleviating PM_{2.5} levels and reducing haze occurrence.

2 Spatiotemporal patterns of PM_{2.5} and its major chemical components

In this section, available measurements of chemically resolved PM_{2.5} are reviewed and summarized in terms of geographical distributions, interannual variations and seasonal patterns. Measurements are grouped based on geographical regions, such as the Beijing–Tianjin–Hebei (BTH) in the North China Plain, the Yangtze River Delta (YRD), the Pearl River Delta (PRD), the Sichuan Basin and other regions (Fig. 1). Five dominant chemical components of PM_{2.5} (SO₄²⁻, NO₃⁻, NH₄⁺, OC and EC) are discussed in detail. Data reviewed in this section are all listed in Table S1 in the Supplement.

2.1 PM_{2.5} mass

Filter-based measurements of PM_{2.5} were mainly carried out in urban cities of BTH (Beijing, Tianjin, Shijiazhuang and Chengde), YRD (Shanghai, Nanjing and Hangzhou), PRD (Guangzhou, Hong Kong, Zhongshan and Shenzhen), Sichuan Basin (Chongqing, Chengdu and Neijiang), and other cities (e.g., Jinan, Xi'an, Lanzhou, Zhengzhou, Wuhan, Changsha, Fuzhou, Xiamen and Haikou). Geographical characteristics of annual PM_{2.5} are first discussed followed by interannual variations and seasonal patterns.

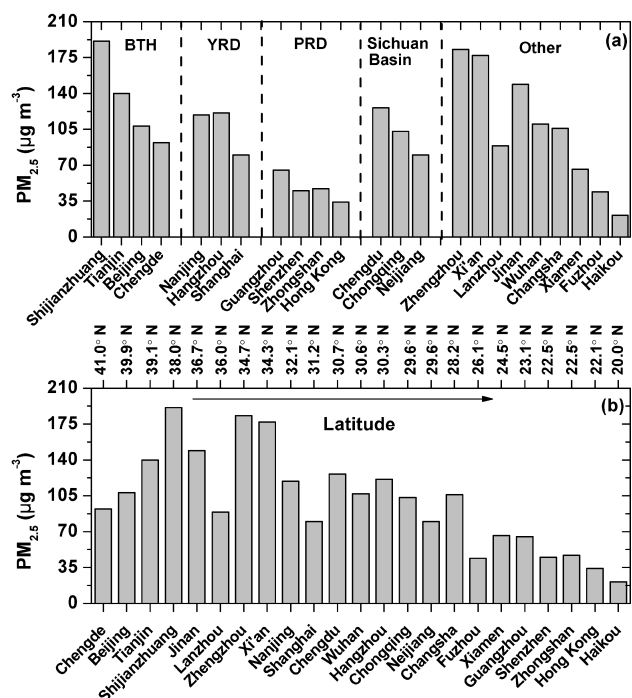


Figure 2. Annual PM_{2.5} mass concentration in various Chinese cities with filter-based measurements: (a) categorized into regions and (b) lined with latitude.

2.1.1 Geographical distributions

Annual mean PM_{2.5} mass concentrations in major cities in different regions are plotted in Fig. 2a. Regional annual mean and standard deviation (SD) values were calculated using annual mean data of all the cities where data are available. Regional annual mean PM_{2.5} was 115 ± 29 , 96 ± 28 , 50 ± 16 and $100 \pm 35 \mu\text{g m}^{-3}$ in BTH (Y. Chen et al., 2014a; Duan et al., 2006; He et al., 2001, 2012; Song et al., 2006a; Tian et al., 2016; Wang et al., 2005; Yang et al., 2011a, b; Zhang et al., 2013a; Zhao et al., 2013c; Zhou et al., 2015a; Ziková et al., 2016), YRD (Feng et al., 2009; Li et al., 2015a, 2016a; Liu et al., 2015; Ming et al., 2017; Wang et al., 2006, 2016b; Ye et al., 2003; Zhao et al., 2015b), PRD (Hagler et al., 2006; Huang et al., 2013; Louie et al., 2005a; Tao et al., 2014c, 2017) and Sichuan Basin (Y. Chen et al., 2014b; Tao et al., 2013a, 2014a; Wang et al., 2017a; Yang et al., 2011b), respectively, which was 3 to 6 times, 2 to 3 times, 1 to 2 times and 3 to 6 times NAAQS, respectively.

Within each region, the highest annual average PM_{2.5} concentration was observed in Shijiazhuang ($191 \mu\text{g m}^{-3}$), Hangzhou ($121 \mu\text{g m}^{-3}$), Guangzhou ($65 \mu\text{g m}^{-3}$) and Chengdu ($111 \mu\text{g m}^{-3}$) in BTH, YRD, PRD and Sichuan Basin, respectively. Outside the above-mentioned four regions, annual PM_{2.5} at individual cities was $183 \pm 25 \mu\text{g m}^{-3}$ (Geng et al., 2013; Wang et al., 2015a, 2017b), $177 \pm 15 \mu\text{g m}^{-3}$ (Shen et al., 2009; Wang et al.,

2015c; Zhang et al., 2011b), $89 \mu\text{g m}^{-3}$ (Wang et al., 2016c), $149 \mu\text{g m}^{-3}$ (Yang et al., 2012), $110 \pm 4 \mu\text{g m}^{-3}$ (Xiong et al., 2017; Zhang et al., 2015a), $106 \mu\text{g m}^{-3}$ (Tang et al., 2017), $66 \pm 22 \mu\text{g m}^{-3}$ (Zhang et al., 2011a, 2012a, 2016), $44 \mu\text{g m}^{-3}$ (Xu et al., 2012c) and $21 \mu\text{g m}^{-3}$ (Liu et al., 2017a) in Zhengzhou, Xi'an, Lanzhou, Jinan, Wuhan, Changsha, Xiamen, Fuzhou and Haikou, respectively. These PM_{2.5} levels were comparable to some of the cities within the four regions, e.g., Zhengzhou, Xi'an and Jinan to Shijiazhuang, Wuhan to Nanjing and Chengde, and Fuzhou and Xiamen of Fujian Province to Guangzhou. Cities in Fig. 2a are rearranged in Fig. 2b from northern to southern latitudes. Except for a few cities, such as Chengde and Beijing, there was a decreasing trend in annual PM_{2.5} mass concentration with decreasing latitude. Moreover, annual PM_{2.5} mass concentrations in western or inland cities were higher than those in eastern or coastal cities along the same latitudes. The geographical patterns of the filter-based PM_{2.5} measurements agreed well with the online monitoring of PM_{2.5} in 31 provincial capital cities in China (Wang et al., 2014c).

Filter-based measurements of PM_{2.5} at rural sites in China were limited and mainly conducted at Shangdianzi of Beijing, Conghua and Tianhu of Guangzhou, and Hok Tsui of Hong Kong (Hagler et al., 2006; Lai et al., 2016; Louie et al., 2005a; Zhao et al., 2013c). Rural PM_{2.5} was around half of that in the cities of the same region. A similar geographical pattern was seen in rural PM_{2.5} as in the urban, e.g., annual PM_{2.5} at the rural site of BTH (Shangdianzi) was $72 \mu\text{g m}^{-3}$, which was 2 times that ($35 \mu\text{g m}^{-3}$) at the rural sites of PRD.

2.1.2 Interannual variations

Data collected in most cities were within a 3-year time window, except in Beijing, Shanghai and Guangzhou, where PM_{2.5} data spanned for more than a decade (1999–2014) (Fig. 3). Interannual variations in PM_{2.5} in Beijing were small, ranging from 100 to $128 \mu\text{g m}^{-3}$, similar to the trends in the online data, which ranged from 65 to $83 \mu\text{g m}^{-3}$ during 2004–2012 (Liu et al., 2014b). The lower concentrations of the online than filter PM_{2.5} data is likely caused by volatilization loss of nitrate and organic matters from the tapered element oscillating microbalances (TEOMs), which operated at 50° during the online sampling. These results suggested that there was no evidence that PM_{2.5} pollution has been significantly improved in Beijing during the 15-year study period despite the many control measures that have been implemented. The impact of local pollution controls in Beijing has likely been offset by regional pollutant transport (Li et al., 2015b). In Shanghai, PM_{2.5} in 2003–2006 ($94 \mu\text{g m}^{-3}$) (Feng et al., 2009; Wang et al., 2006) and 2009 ($94 \mu\text{g m}^{-3}$) (Zhao et al., 2015b) was nearly 50 % higher than earlier years (e.g., $65 \mu\text{g m}^{-3}$ in 1999–2000) (Ye et al., 2003); although it decreased slightly to $58 \mu\text{g m}^{-3}$ in 2011–2013 (Wang et al., 2016b; Zhao et al., 2015b), it increased rapidly back to

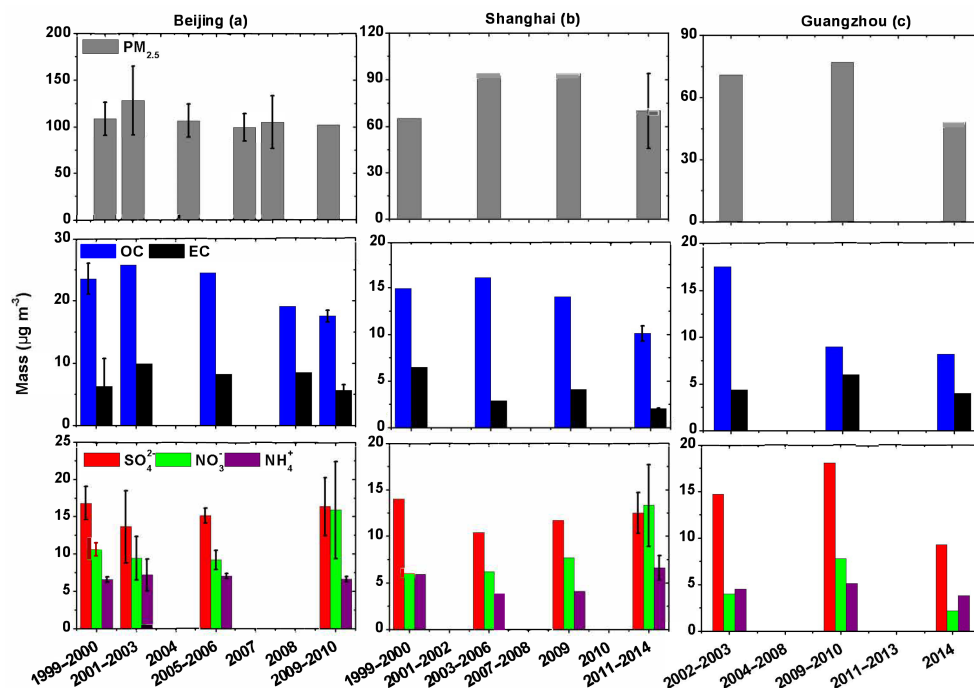


Figure 3. Interannual variations in PM_{2.5} and dominant chemical components in Beijing (a), Shanghai (b) and Guangzhou (c).

95 $\mu\text{g m}^{-3}$ in 2013–2014 (Ming et al., 2017). In Guangzhou, PM_{2.5} in 2002–2003 (71 $\mu\text{g m}^{-3}$) (Hagler et al., 2006) and in 2009–2010 (77 $\mu\text{g m}^{-3}$) (Tao et al., 2014c) were kept at stable levels and then decreased to 48 $\mu\text{g m}^{-3}$ in 2014 (Tao et al., 2017).

2.1.3 Seasonal patterns

In BTH, the highest seasonal average PM_{2.5} concentrations were observed in winter and the lowest in summer in all the cities with seasonal variations up to factors of 1.7, 1.5, 1.6 and 1.8 in Beijing (Cao et al., 2012b; Chan et al., 2005; J. Chen et al., 2014a; Dan et al., 2004; Duan et al., 2006; Han et al., 2014; He et al., 2001; Huang et al., 2014b; Lin et al., 2016; Okuda et al., 2011; Pathak et al., 2011; Song et al., 2006a, 2007; Sun et al., 2004, 2006; Tan et al., 2016a; Tao et al., 2016a, 2015a; Tian et al., 2015; Wang et al., 2005; Yang et al., 2005a, 2016; Zhang et al., 2013a; Zhao et al., 2013c), Tianjin (Cao et al., 2012b; Gu et al., 2010, 2011; Li et al., 2009; Tian et al., 2016; Zhao et al., 2013c), Shijiazhuang (Zhao et al., 2013c) and Chengde (Zhao et al., 2013c), respectively. It is noted that major pollutant sources in BTH were located south of Hebei Province and the prevailing winds in BTH were from the north in winter and from the south in summer (Li et al., 2016b; Lu et al., 2010, 2011; Wang et al., 2013; Xu et al., 2011). The location and distribution of major industrial sources, intensity of local minor sources such as winter heating, and prevailing wind directions together caused the slightly different magnitudes of seasonal variations among the four cities discussed above.

Moreover, extreme weather events such as weakening monsoon circulation, depression of strong cold air activities, strong temperature inversion and descending air masses in the planetary boundary layer also played important roles in the strong PM_{2.5} pollution during winter (Niu et al., 2010; Wang et al., 2014b; Zhao et al., 2013d). Several extreme wintertime air pollution events in recent years covered vast areas of northern China and were all correlated to some extent with extreme weather conditions (Zou et al., 2017).

In YRD, the highest seasonal average PM_{2.5} concentrations were also observed in winter and the lowest in summer with seasonal variations up to factors of 2.3, 1.9 and 2.0 in Nanjing (Li et al., 2015a, 2016a; Shen et al., 2014; Yang et al., 2005b), Shanghai (Cao et al., 2012b, 2013; Feng et al., 2009, 2012a; Hou et al., 2011; Huang et al., 2014a, b; Ming et al., 2017; Pathak et al., 2011; Wang et al., 2006, 2016b; Ye et al., 2003; Zhao et al., 2015b) and Hangzhou (Cao et al., 2012b; Liu et al., 2015), respectively. In PRD, most urban site PM_{2.5} studies were also accompanied with rural site studies (Andreae et al., 2008; Cao et al., 2004, 2012b; Cui et al., 2015; Duan et al., 2007; Fu et al., 2014; Ho et al., 2006a; Huang et al., 2007, 2014b; Jahn et al., 2013; Jung et al., 2009; Lai et al., 2007, 2016; Liu et al., 2014a; Louie et al., 2005a; Tan et al., 2009, 2016c; Tao et al., 2009, 2014c, 2015b, 2017; Wang et al., 2012; Yang et al., 2011b). Although the highest seasonal average PM_{2.5} was also observed in winter, the season with the lowest concentration was not consistent between the sites, e.g., in summer in Guangzhou and in spring in Hong Kong. This was

likely caused by warm/hot temperatures in this region and frequent precipitation in warm seasons, and thus small differences between spring and summer, e.g., PM_{2.5} concentration of 32 $\mu\text{g m}^{-3}$ in summer in Guangzhou (Cao et al., 2004, 2012b; Duan et al., 2007; Ho et al., 2006a; Lai et al., 2007; Louie et al., 2005a) and 29 $\mu\text{g m}^{-3}$ in spring in Hong Kong (Louie et al., 2005a). Seasonal variations were up to a factor of 1.9 in both cities. PM_{2.5} at rural sites in PRD generally doubled during dry seasons (autumn and winter) compared to wet seasons (spring and summer) due to frequent precipitation scavenging of aerosols in wet seasons (Cheung et al., 2005; Dai et al., 2013; Fu et al., 2014; Griffith et al., 2015; Hu et al., 2008; Lai et al., 2016).

Similar seasonal patterns as above were also observed in cities of other regions in China, such as Chengdu (Tao et al., 2013a, 2014a), Zhengzhou (Geng et al., 2013), Jinan (Yang et al., 2012) and Fuzhou (Xu et al., 2012b), with seasonal variations between a factor of 1.8 and 2.5. In conclusion, the highest seasonal average PM_{2.5} observed in winter at all urban sites in China was likely due to high emissions from winter heating and/or poor pollutant dispersion.

2.2 Major chemical components of PM_{2.5}

It is well known that OC, EC, SO_4^{2-} , NO_3^- and NH_4^+ are the dominant chemical components in PM_{2.5}. Thus, only studies having synchronous measurements of PM_{2.5} and the above-mentioned five major components were discussed below. Note that for most cities only short-term measurements were available; however, for Beijing, Shanghai and Guangzhou, existing studies span a period of 15 years (2000–2014).

To ensure the comparability of the data collected using different instruments, measurement uncertainties were first briefly discussed here. Most studies in China analyzed OC and EC using a DRI carbon analyzer or Sunset carbon analyzer. IMPROVE is the most widely used thermal–optical protocol for OC and EC analysis for the DRI analyzer, while NIOSH is the one used for the Sunset analyzer. OC and EC measured by the two analyzers are comparable if the same analysis protocol is used. For example, Wu et al. (2011) showed that OC from the Sunset analyzer was only 8 % lower than that from the DRI analyzer, while EC was only 5 % higher. However, when different protocols were used by the two analyzers, the differences were much larger, e.g., EC from NIOSH was almost 50 % lower than that from IMPROVE (Chow et al., 2010; Yang et al., 2011a). Note that OC and EC were also measured using a CHN elemental analyzer in 2001–2002 in Beijing, which uses a similar protocol to NIOSH (Duan et al., 2006). In any case, the measurement uncertainties of total carbon (TC, the sum of OC and EC) were less than 10 % (Chow et al., 2010; Wu et al., 2011).

The ions including SO_4^{2-} , NO_3^- and NH_4^+ were measured by ion chromatography. Measurement uncertainties should be less than 15 % in most cases under strict QA/QC procedures (Orsini et al., 2003; Trebs et al., 2004; Weber et al.,

2003), but could be larger for ammonium nitrate (NH_4NO_3) since it can evaporate from the filters before chemical analysis under high temperature and low RH conditions, and this applies to both quartz fiber filter and Teflon filter (Keck and Wittmaack, 2005; Weber et al., 2003). The loss of NO_3^- due to evaporation was found to range from 4 to 84 % depending on the ambient temperature (Chow et al., 2005). Although the exact magnitudes of measurement uncertainties cannot be determined for NO_3^- and NH_4^+ , they are not expected to significantly affect the interannual variations discussed below for the three cities (Beijing, Shanghai and Guangzhou) considering the small year-to-year temperature changes.

2.2.1 The Beijing–Tianjin–Hebei region

Interannual variations in Beijing

Chemically resolved PM_{2.5} data in BTH covering multiple years are only available in Beijing and the interannual variations are discussed for this city below (Duan et al., 2006; He et al., 2001; Song et al., 2006a; Yang et al., 2011b; Zhang et al., 2013a; Zhao et al., 2013c). Interannual variations in OC and EC were generally small, e.g., a factor of 1.5 for OC and 1.8 for EC (Fig. 3a). OC decreased from 23.6–25.8 $\mu\text{g m}^{-3}$ in earlier years (1999–2006) to below 17.6 $\mu\text{g m}^{-3}$ after 2008. EC increased from 6.3 $\mu\text{g m}^{-3}$ in 1999–2000 to 9.9 $\mu\text{g m}^{-3}$ in 2001–2002, and then gradually decreased to 5.7 $\mu\text{g m}^{-3}$ in 2009–2010. TC increased from 29.8 $\mu\text{g m}^{-3}$ in 1999–2000 to 32.7–35.7 $\mu\text{g m}^{-3}$ in 2001–2006, and then decreased to 23.3 $\mu\text{g m}^{-3}$ in 2009–2010. The nearly 30 % reduction in TC in recent years in Beijing can be taken as a real trend since measurement uncertainties were believed to be around 10 % as mentioned above. OC is produced from both primary emissions and secondary formation, while EC (also known as black carbon or BC) is mainly from primary emissions. The anthropogenic emission for OC and BC over the entire China showed an increasing trend in 1996–2010 (Lu et al., 2011), while BC emissions showed a slightly decreasing trend in Beijing and Tianjin in 2005–2009 (Qin and Xie, 2012). Meanwhile, BC emissions sharply increased in Hebei Province in 2005–2009. The amount of BC emissions in Hebei Province was much higher than the sum of those in Beijing and Tianjin (Qin and Xie, 2012). Thus, the decrease in EC concentration in Beijing was likely dominated by local emission reduction instead of regional transport from Hebei province.

Annual SO_4^{2-} concentration increased slightly during 1999–2010 and ranged from 10.2 to 16.4 $\mu\text{g m}^{-3}$ in Beijing. SO_2 emission in China increased by about 60 % during 2000–2006 and then decreased about 9 % during 2006–2010 due to the compulsory flue-gas desulfurization equipment applied in power plants (Lu et al., 2011). However, the sum of the SO_2 emissions in BTH (including Beijing, Tianjin and Hebei Province) increased sharply from 2097 Gg yr⁻¹ in 2000 to 2916 Gg yr⁻¹ in 2004, and further slightly in-

creased to 2998 Gg yr⁻¹ in 2007 before sharply decreased to 1821 Gg yr⁻¹ in 2010 (Lu et al., 2010; Zhao et al., 2013a). A continued increase in SO₂ emission was found in Hebei Province, which accounted for more than 50 % of the total SO₂ emission in BTH. In contrast, SO₂ emissions in Beijing continued decreasing. Surface annual SO₂ concentrations in Beijing gradually decreased from 56 to 35 μg m⁻³ during 2006–2009 (<http://www.zhb.gov.cn/>). Thus, the persistently high concentrations of SO₄²⁻ in Beijing was largely due to regional transport from Hebei Province, noting that the lifetime of SO₄²⁻ is longer than that of SO₂.

NO₃⁻ concentrations were relatively steady (7.4–10.9 μg m⁻³) during 1999–2006, but sharply increased to 15.9 μg m⁻³ in 2009–2010 in Beijing. Both NO_x (NO₂ + NO) emissions and satellite NO₂ vertical column densities synchronously increased during 2000–2010 in China (Zhang et al., 2012b; Zhao et al., 2013b). Different from those of SO₂ emissions, NO_x emissions in all the cities and provinces in BTH showed increasing trends in 2005–2010. NO_x emissions in Beijing slightly increased from 410 Gg yr⁻¹ in 2005 to 480 Gg yr⁻¹ in 2010 (Zhao et al., 2013b). However, annual average surface NO₂ concentrations in Beijing showed a decreasing trend and fluctuated in the range of 49–66 μg m⁻³ during 2006–2009 (<http://www.zhb.gov.cn/>). There were some inconsistencies between the trends of surface NO₂ concentrations and column NO₂ or NO_x emissions, likely due to the impact of photochemical reactions on surface NO₂ concentrations in urban areas. To some extent, the increasing trend of NO₃⁻ in Beijing was likely related to the increases in NO_x emissions in both Beijing and the surrounding cities or provinces.

Considering the potential large uncertainties in NH₄⁺ measurements, its trends should only be discussed qualitatively. NH₄⁺ concentrations were relatively steady in Beijing during 1999–2006, ranging from 5.7 to 7.3 μg m⁻³. NH₃ emissions changed little (13 400–13 600 Gg yr⁻¹) before 2005 in China, and increased slightly in BTH region during 2003–2010 (Zhou et al., 2015b). The small increase in NH₄⁺ in 2009–2010 in Beijing was consistent with the NH₃ emission trend in this region (Zhang et al., 2013a; Zhao et al., 2013c). Moreover, the increase in NO₃⁻ in Beijing was also an important factor contributing to the increase in NH₄⁺.

In summary, a decreasing trend was identified in TC and increasing trends were found for SO₄²⁻, NO₃⁻ and NH₄⁺ in Beijing. The interannual variations in EC agreed with the local emission trends in Beijing, but those in SO₄²⁻, NO₃⁻ and NH₄⁺ agreed more with the regional scale emission trends of their respective gaseous precursors in BTH rather than the local emission trends in Beijing. Nonlinear responses of concentration changes of these aerosol components to their respective emission trends were found, demonstrating other important factors potentially affecting aerosol formation. It is worth to note that several recent studies have highlighted the important role NO₂ might play in sulfate formation in

the polluted environment in China (Cheng et al., 2016; Wang et al., 2016a; Xie et al., 2015a). Nevertheless, the aqueous SO₂ + H₂O₂/O₃ oxidation should still be the dominant mechanism in most cases, especially at a background site (Lin et al., 2017). The aqueous SO₂ + oxygen (catalyzed by Fe (III)) reaction can also be important under heavy haze conditions in north China (Li et al., 2017a). Extensive measurements of stable oxygen are needed to confirm the relative contributions of different sulfate formation mechanisms.

Relative contributions to PM_{2.5}

To investigate the relative contributions of dominant chemical components to PM_{2.5} mass, the measured PM_{2.5} mass was reconstructed based on SO₄²⁻, NO₃⁻, NH₄⁺, OM (organic matter) and EC. The conversion factor between OC and OM was 1.8 considering the prevailing biomass burning in BTH (Cheng et al., 2013a; Du et al., 2014a).

Data collected in 2009–2010 were first discussed since multiple cities in BTH have data during this period (Zhang et al., 2013a; Zhao et al., 2013c). Secondary inorganic aerosols (the sum of sulfate, nitrate and ammonium) contributed 36–39 % of PM_{2.5} annually in the majority of the cities, but only 25 % in Chengde, a tourist city located in the north-east part of BTH and 200 km away from Beijing. Generally, the percentage contribution of secondary inorganic aerosols to PM_{2.5} decreased with decreasing PM_{2.5} level, e.g., from Shijiazhuang to Tianjin, Beijing and then Chengde, a phenomenon that is consistent with what was found within the same city but for different pollution levels in a winter season (Tao et al., 2015a). Carbonaceous aerosols (OM + EC) contributed 29–32 % to PM_{2.5} in most cities, but as high as 45 % in Chengde, and had an opposite trend to that of secondary inorganic aerosols in terms of city-to-city variations. At the rural site Shangdianzi near Beijing, secondary inorganic aerosols and carbonaceous aerosols accounted for 42 and 32 %, respectively, of the PM_{2.5} mass, which were not significantly different from those in cities located south of Yan Mountains. The sum of secondary inorganic aerosols and carbonaceous aerosols accounted for 65–70 % of the PM_{2.5} mass in cities of BTH.

In Beijing, where data are available for more than a decade, secondary inorganic aerosols accounted for 28 % of PM_{2.5} on average and ranged from 23 to 31 % from year to year. Carbonaceous aerosols accounted for 43 % of PM_{2.5} and ranged from 29 to 55 %. Seasonal average contributions of secondary inorganic aerosols were generally higher in warm seasons than in cold seasons in most cities, and an opposite trend was found for carbonaceous aerosols (Fig. 4). For example, secondary inorganic aerosols contributed 32, 41, 28 and 32 % in spring, summer, autumn and winter, respectively, to PM_{2.5} in Beijing, while carbonaceous aerosols contributed 35, 30, 44 and 45 % (Cao et al., 2012b; Duan et al., 2006; He et al., 2001; Huang et al., 2014b; Lin et al., 2016; Pathak et al., 2011; Song et al., 2006a, 2007; Sun et al.,

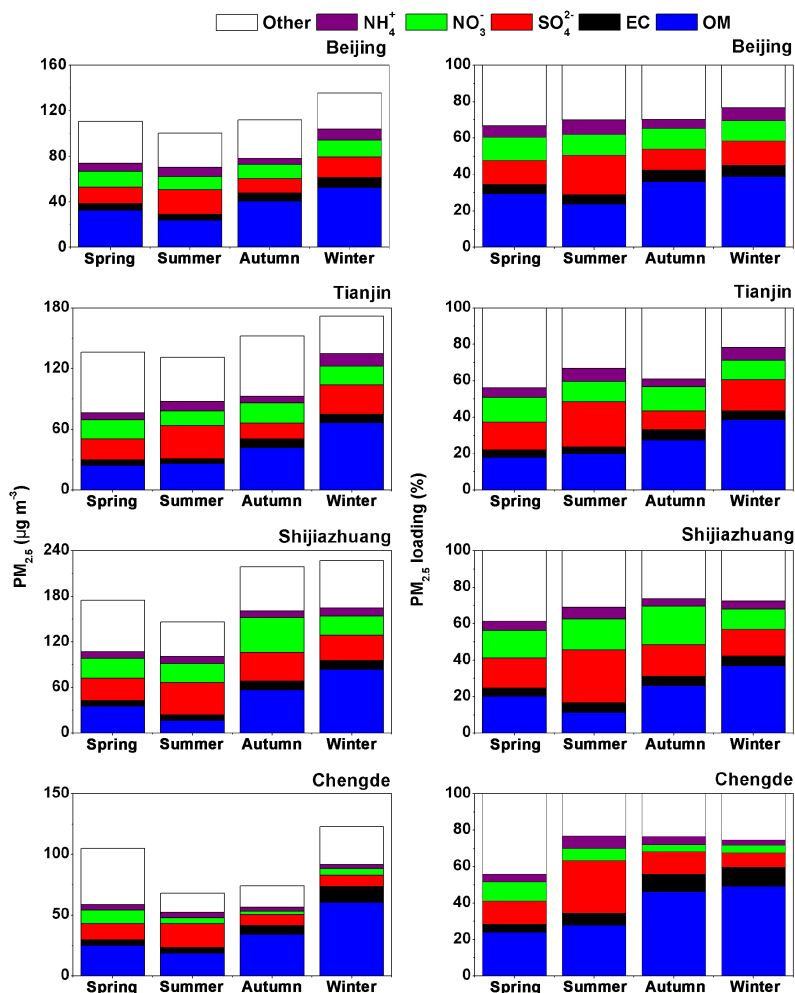


Figure 4. Seasonal PM_{2.5} and dominant chemical components in BTH.

2004; Tao et al., 2015a; Tian et al., 2015; Zhang et al., 2013a; Zhao et al., 2013c). A similar seasonal trend was also observed in other BTH cities, e.g., secondary inorganic aerosols accounted for 42–53 % of PM_{2.5} mass in summer and only 15–35 % in winter, while carbonaceous aerosols accounted for 16–34 % in summer and 42–60 % in winter in Tianjin (Cao et al., 2012b; Gu et al., 2011; Li et al., 2009; Zhao et al., 2013c), Shijiazhuang (Zhao et al., 2013c) and Chengde (Zhao et al., 2013c). Higher carbonaceous aerosols in winter should be related to heating activities and biomass burning in this region (Cheng et al., 2013a; Duan et al., 2004; Tao et al., 2016b; Wang et al., 2007; Yang et al., 2016).

2.2.2 The Yangtze River Delta region

Interannual variations in Shanghai

Chemically resolved PM_{2.5} data in YRD covering multiple years are only available in Shanghai (Ming et al., 2017; Wang et al., 2016b; Ye et al., 2003; Zhao et al., 2015b). Interannual

variations in OC in this city were within a factor of 1.6 for OC and a factor of 4.1 for EC (Fig. 3b). OC concentrations were relatively steady (14.0–14.9 µg m⁻³) during 1999–2009, but sharply decreased to 9.9–10.1 µg m⁻³ in 2011–2014. EC varied in the range of 4.1 to 6.5 µg m⁻³ during 1999–2009, and also sharply decreased to 1.6–2.1 µg m⁻³ in 2011–2014. TC decreased from 19.5 µg m⁻³ during 1999–2009 to 11.9 µg m⁻³ in 2011–2014, or nearly 40 % reduction, which was much higher than the known measurement uncertainties. Noticeable reduction of OC and EC occurred after the 2010 Shanghai World Expo, which resulted in a decrease in TC after 2010. BC emission slightly decreased in Shanghai in 2005–2009, but increased in the adjacent Zhejiang and Jiangsu provinces (Qin and Xie, 2012). BC emissions in Jiangsu Province were much higher than the sum of those in Shanghai and Zhejiang. Thus, the decreased EC concentration in Shanghai was largely a result of local emission reductions.

Annual SO₄²⁻ concentration decreased from 14.0 µg m⁻³ in 1999–2000 to the range of 11.7 µg m⁻³ to 12.5 µg m⁻³

during 2009–2014. The trend of SO₂ emission in YRD generally agreed with that across the entire China, which showed an increasing trend during 2000–2006 and a decreasing one during 2006–2010 (Lu et al., 2011). The annual variations in SO₂ emission in YRD (including Shanghai, Jiangsu and Zhejiang) were relative small, ranging 3171 Gg yr⁻¹ in 2000, 3506 Gg yr⁻¹ in 2004, 3376 Gg yr⁻¹ in 2007, and 3397 Gg yr⁻¹ in 2010 (Lu et al., 2010; Zhao et al., 2013a). Annual average SO₂ concentrations in Shanghai were in the range of 45–61 μg m⁻³ during 2000–2005 and decreased by around 50 % to 24–29 μg m⁻³ during 2010–2013 (<http://www.zhb.gov.cn/>). Note that SO₂ emissions in Shanghai only accounted for less than 20 % of the total SO₂ emissions in YRD and with small annual variations. The high concentrations of SO₄²⁻ observed in Shanghai were also closely related to regional transport from north China (e.g., BTH and Shandong Province) (Li et al., 2011; Wang et al., 2016b).

Annual NO₃⁻ concentrations in Shanghai were relatively steady (6.0–7.7 μg m⁻³) during 1999–2009, but sharply increased to 13.3 μg m⁻³ in 2011–2014. NO_x emissions in YRD also showed an increasing trend during these years, consistent with satellite retrieved vertical column NO₂ densities during 2000–2010 (Zhang et al., 2012b; Zhao et al., 2013b). In contrast, surface-level annual NO₂ concentrations in Shanghai sharply decreased from 90 μg m⁻³ in 2000 to a range of 48–61 μg m⁻³ during 2003–2013 (<http://www.zhb.gov.cn/>). The inconsistency in the trends between emissions and gaseous and particulate surface air concentrations was also found in Beijing. Photochemistry and regional transport of related pollutants should be the major causes of this phenomenon.

Annual NH₄⁺ concentrations decreased from 5.9 μg m⁻³ in 1999–2000 to the levels of 4.1 μg m⁻³ in 2009 and then increased to 6.6 μg m⁻³ in 2011–2014. NH₃ emissions increased in 2000–2005 in east China (including BTH, YRD and PRD) and possibly also increased in 2006–2010 due to the lack of control measures for NH₃ in China (Wang et al., 2011). The recently increased NH₄⁺ concentrations in Shanghai were likely due to the concurrent increases of NH₃ emissions and NO₃⁻ concentrations.

In summary, a decreasing trend was identified in TC, increasing ones for NO₃⁻ and NH₄⁺, and a stable one for SO₄²⁻ in Shanghai. The interannual variations in EC agreed with its local emission trends in Shanghai rather than regional emission trends. In contrast, interannual variations in SO₄²⁻, NO₃⁻ and NH₄⁺ agreed more with the regional scale emission trends of their respective gaseous precursors in YRD. Similar to Beijing, nonlinear responses of concentration changes of these aerosol components to their respective emission trends were also found in Shanghai.

Relative contributions to PM_{2.5}

The chemical compositions in PM_{2.5} between the cities in YRD were compared between Shanghai and Nanjing due

to the lack of continuous annual data in Hangzhou. A conversion factor of 1.6 between OC and OM was chosen for YRD, slightly smaller than that (1.8) chosen for BTH considering the lower impact of biomass burning on PM_{2.5} in this region (Feng et al., 2006; Li et al., 2016a). Secondary inorganic aerosols contributed 25–54 % of PM_{2.5} annually in Shanghai and Nanjing, while carbonaceous aerosols contributed 28–47 % (Li et al., 2016a; Wang et al., 2016b; Ye et al., 2003; Zhao et al., 2015b). The sum of secondary inorganic aerosols and carbonaceous aerosols accounted for 76 and 66 % of PM_{2.5} mass in Shanghai and Nanjing, respectively, which was comparable with that (65–70 %) in BTH.

Seasonal variations in secondary inorganic aerosols contributions to PM_{2.5} were small in both cities, e.g., 41–49 % in Shanghai and 32–40 % in Nanjing (Fig. 5). Larger seasonal variations were found for carbonaceous aerosols than secondary inorganic aerosols, e.g., 47 % in summer and 33–39 % in other seasons in Shanghai, and ranged from 27 % (spring) to 65 % (autumn) in Nanjing (Cao et al., 2012b; Huang et al., 2014a, b; Li et al., 2016a; Ming et al., 2017; Pathak et al., 2011; Shen et al., 2014; Wang et al., 2016b; Yang et al., 2005b; Ye et al., 2003; Zhao et al., 2015a).

In Hangzhou, seasonal contributions can only be estimated for summer and winter 2003 (Cao et al., 2012b). Seasonal contribution of secondary inorganic aerosols in winter was 44 %, which was evidently higher than that in summer (34 %), while carbonaceous aerosols contributed 33–35 %. At the rural sites (Ningbo and Lin'an) in Zhejiang Province, seasonal contributions of carbonaceous aerosols varied within a small range (28–34 %) in four seasons in 2008–2009, which were comparable with those in Hangzhou (Feng et al., 2015; Liu et al., 2013a).

In summary, the different seasonal average contributions of secondary inorganic aerosols and carbonaceous aerosols in Shanghai and Nanjing were likely due to different local sources in YRD. The seasonal patterns of these chemical components in Shanghai were a result of both local emissions and regional transport, but in Nanjing, the seasonal pattern was mainly determined by local emissions because it is an inland city surrounded by many industrial enterprises, including power plants, petrochemical plants and steel plants.

2.2.3 The Pearl River Delta region

Interannual variations in Guangzhou

Interannual variations for dominant chemical components were only discussed for Guangzhou in PRD since data for this city were available during 2002–2003, 2009–2010 and 2014 (Hagler et al., 2006; Tao et al., 2014c, 2017). Data for Shenzhen were only available during 2002–2003 and 2009 (Hagler et al., 2006; Huang et al., 2013) and for Hong Kong during 2000–2001 and 2002–2003 (Hagler et al., 2006; Louie et al., 2005b). Annual OC concentrations decreased significantly from 17.6 μg m⁻³ in 2002–2003 to

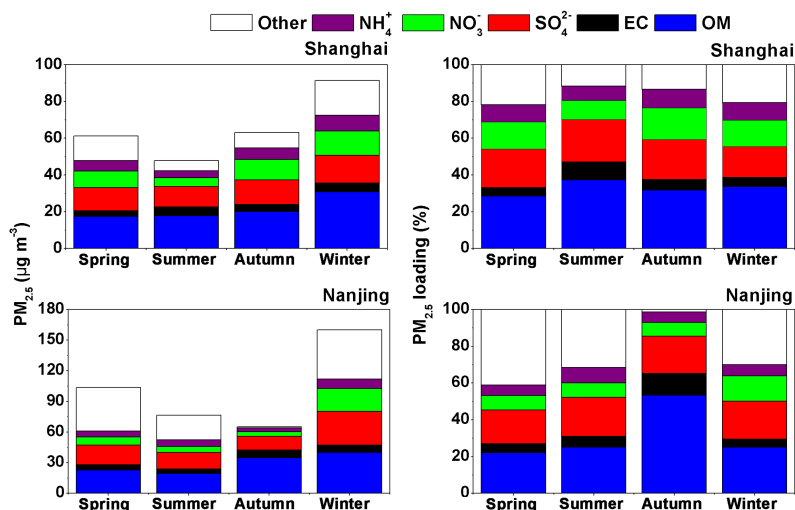


Figure 5. Seasonal PM_{2.5} and dominant chemical components in YRD.

9.0 µg m⁻³ in 2009–2010, and then to 8.2 µg m⁻³ in 2014 in Guangzhou, while EC slightly increased from 4.4 to 6.0 µg m⁻³ and then decreased to 4.0 µg m⁻³ during the same periods. Similar to Guangzhou, annual OC concentrations decreased significantly from 11.1 µg m⁻³ in 2002–2003 to 8.3 µg m⁻³ in 2009–2010 in Shenzhen, while EC slightly increased from 2.3 to 2.7 µg m⁻³. Apparently, the trends of EC in Guangzhou and Shenzhen were inconsistent with the BC emission trends in Guangdong Province during 2005–2009, which showed a slightly decreasing trend (Qin and Xie, 2012). As a result, TC concentrations gradually decreased from 22.0 to 15.0 µg m⁻³ in Guangzhou and from 15.0 to 13.0 µg m⁻³ in Shenzhen before 2010, similar to Beijing and Shanghai. The reduction of TC was significant in Guangzhou (32%). The same phenomenon was also observed at a suburban site of Guangzhou (Hagler et al., 2006; Lai et al., 2016).

In contrast to the TC trend, annual SO₄²⁻, NO₃⁻ and NH₄⁺ concentrations in Guangzhou increased from 14.7, 4.0 and 4.5 µg m⁻³ in 2002–2003 to 18.1, 7.8 and 5.1 µg m⁻³ in 2009–2010 and decreased to 9.3, 2.2 and 3.8 µg m⁻³ in 2014, respectively. Increases in the concentrations were also found in Shenzhen, e.g., from 10.0, 2.3 and 3.2 µg m⁻³ in 2002–2003 to 11.7, 2.7 and 3.5 µg m⁻³ in 2009, respectively (Hagler et al., 2006; Huang et al., 2013), and in the suburban areas of Guangzhou, e.g., from 10.4, 0.3 and 2.4 µg m⁻³ in 2002–2003 to 12.2, 2.0 and 5.2 µg m⁻³ in 2012–2013, respectively (Hagler et al., 2006; Lai et al., 2016). SO₂ emissions in Guangdong Province gradually increased in the previous decade, e.g., 964, 1150, 1177 and 1258 Ggyr⁻¹ in 2000, 2004, 2007 and 2010, respectively (Lu et al., 2010; Zhao et al., 2013a). However, SO₂ emissions in PRD decreased by more than 40% between 2005 and 2009, due to flue gas desulfurization facilities in power plants and large industrial boilers installed in this region (Lu et al., 2013). Annual average SO₂ concentrations in Guangzhou gradually

increased from 45 µg m⁻³ in 2000 to 77 µg m⁻³ in 2004, and then decreased to 17 µg m⁻³ in 2014 (<http://www.gzepb.gov.cn/>). Thus, the increased SO₄²⁻ concentration before 2010 in Guangzhou was largely due to regional transport of pollutants from outside of PRD. The decreased SO₄²⁻ concentration in 2014 in Guangzhou was likely due to flue gas desulfurization facilities in power plants and large industrial boilers implemented across the entire Guangdong Province (<http://www.gdep.gov.cn/>).

Meanwhile, NO_x emissions increased in Guangdong Province as well as across the entire PRD, similar to the trends in BTH and YRD (Lu et al., 2013; Zhang et al., 2012b; Zhao et al., 2013b). However, annual average surface NO₂ concentrations in Guangzhou fluctuated from 61 to 73 µg m⁻³ during 2000–2007 and from 48 to 56 µg m⁻³ during 2008–2014 (<http://www.gzepb.gov.cn/>). An opposite trend was also found between NO₂ and NO_x emissions with the former persistently decreased, while the latter increased in Guangzhou, although NO₃⁻ concentrations also increased. Thus, emissions as well as chemical processes both affected these ion concentrations in air. Annual NH₄⁺ concentrations slightly increased about 10% before 2010 in Guangzhou and Shenzhen although NH₃ emissions changed little during 2002–2006 in PRD (Zheng et al., 2012). Thus, the slightly increased NH₄⁺ concentrations, if not caused by measurement uncertainties, in Guangzhou and Shenzhen during 2002–2010 were largely due to the increased SO₄²⁻ and NO₃⁻, which enhanced the conversion of NH₃ to NH₄⁺.

In summary, a decreasing trend was identified in TC and increasing trends were found for SO₄²⁻, NO₃⁻ and NH₄⁺ in Guangzhou and Shenzhen before 2010, while all chemical components decreased after 2010 in Guangzhou. The interannual variations in EC were inconsistent with BC emission trends in Guangdong Province. In contrast, interannual variations in SO₄²⁻, NO₃⁻ and NH₄⁺ agreed with regional scale

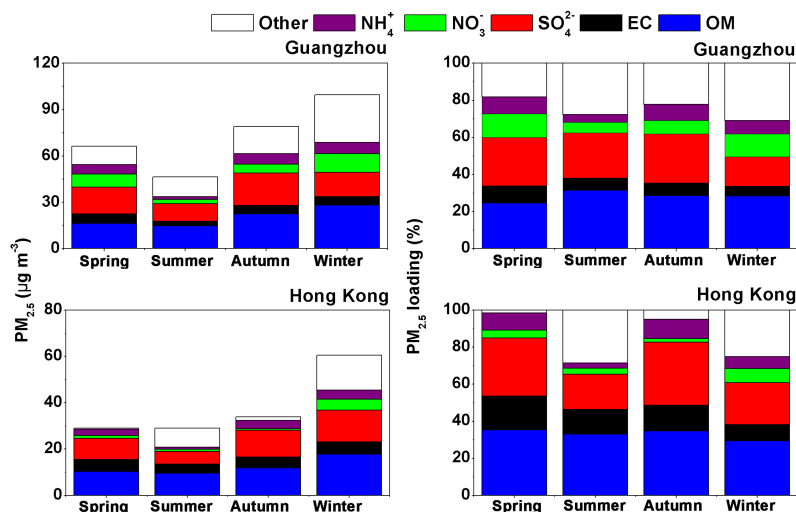


Figure 6. Seasonal PM_{2.5} and dominant chemical components in PRD.

emission trends of their respective gaseous precursors in Guangdong Province rather than PRD. Similar to Beijing and Shanghai, nonlinear responses of concentration changes of these aerosol components to their respective emission trends were also found in Guangzhou and Shenzhen.

Relative contributions to PM_{2.5}

Data collected in 2002–2003 were discussed since multiple cities (e.g., Guangzhou, Conghua, Zhongshan, Shenzhen and Hong Kong) in PRD have data during this period (Hagler et al., 2006). The conversion factor between OC and OM was chosen to be the same as in YRD (1.6). Secondary inorganic aerosols contributed 33–38 %, depending on location, of PM_{2.5} annually, while carbonaceous aerosols contributed 37–46 %. It is noted that PM_{2.5} in Guangzhou was much higher than those in other coastal cities (including Zhongshan, Shenzhen and Hong Kong), but the contributions of secondary inorganic aerosols and carbonaceous aerosols were not significantly different between these cities. At rural sites (Tianhu and Conghua near Guangzhou and Hok Tsui near Hong Kong), secondary inorganic aerosols and carbonaceous aerosols accounted for 35–48 and 24–43 %, respectively, of the PM_{2.5} mass, which were similar to those obtained in the cities in PRD (Hagler et al., 2006; Lai et al., 2016; Louie et al., 2005b). Thus, the sum of secondary inorganic aerosols and carbonaceous aerosols accounted for 68–83 % of the PM_{2.5} mass in the PRD region, similar to Shanghai (YRD).

Although many studies have been conducted in PRD, many of them were short term. Studies covering all four seasons were mainly carried out in Guangzhou and Hong Kong (Fig. 6) (Andreae et al., 2008; Cao et al., 2004, 2012b; Cui et al., 2015; Ho et al., 2006a; Huang et al., 2014b; Jung et al., 2009; Lai et al., 2007; Liu et al., 2014a; Louie et al.,

2005a; Tan et al., 2009; Tao et al., 2009, 2014c, 2015b, 2017; Yang et al., 2011b). Seasonal average contributions of secondary inorganic aerosols were generally higher in spring and autumn than in summer and winter in both Guangzhou and Hong Kong. If all the years of data were averaged together, secondary inorganic aerosols contributed 43, 31, 38 and 33 % in spring, summer, autumn and winter, respectively, to PM_{2.5} in Guangzhou and 45, 25, 46 and 37 %, respectively, in Hong Kong. However, different seasonal patterns were found between Guangzhou and Hong Kong for carbonaceous aerosols. Carbonaceous aerosols contributed 34, 37, 35 and 34 % in spring, summer, autumn and winter, respectively, to PM_{2.5} in Guangzhou and 54, 47, 49 and 38 %, respectively, in Hong Kong. Seasonal variations in OC / EC ratios ranged from 1.6 to 3.4 in Guangzhou and from 1.2 to 2.1 in Hong Kong, suggesting coal combustion and vehicle exhaust were the dominant sources in Guangzhou, while vehicle exhaust was the dominant source in Hong Kong (He et al., 2008; Watson et al., 2001).

2.2.4 Other cities

Besides the cities in BTH, YRD and PRD, synchronous measurements of PM_{2.5} and the dominant chemical components have also been conducted in several cities in other regions of China, mainly in the capital city of a province (e.g., Zhengzhou of Henan Province: Geng et al., 2013; Xi'an of Shaanxi Province: Wang et al., 2015c; Lanzhou of Gansu Province: Wang et al., 2016c; Jinan of Shandong Province: Yang et al., 2012; Chengdu of Sichuan Province: Tao et al., 2013a, 2014b; Chongqing of Chongqing municipality: Yang et al., 2011b; Changsha of Hunan Province: Tang et al., 2017; Xiamen and Fuzhou of Fujian Province: Xu et al., 2012b; Zhang et al., 2016; and Haikou of Hainan Province: Liu et al., 2017a). A conversion factor of 1.6 between OC and OM

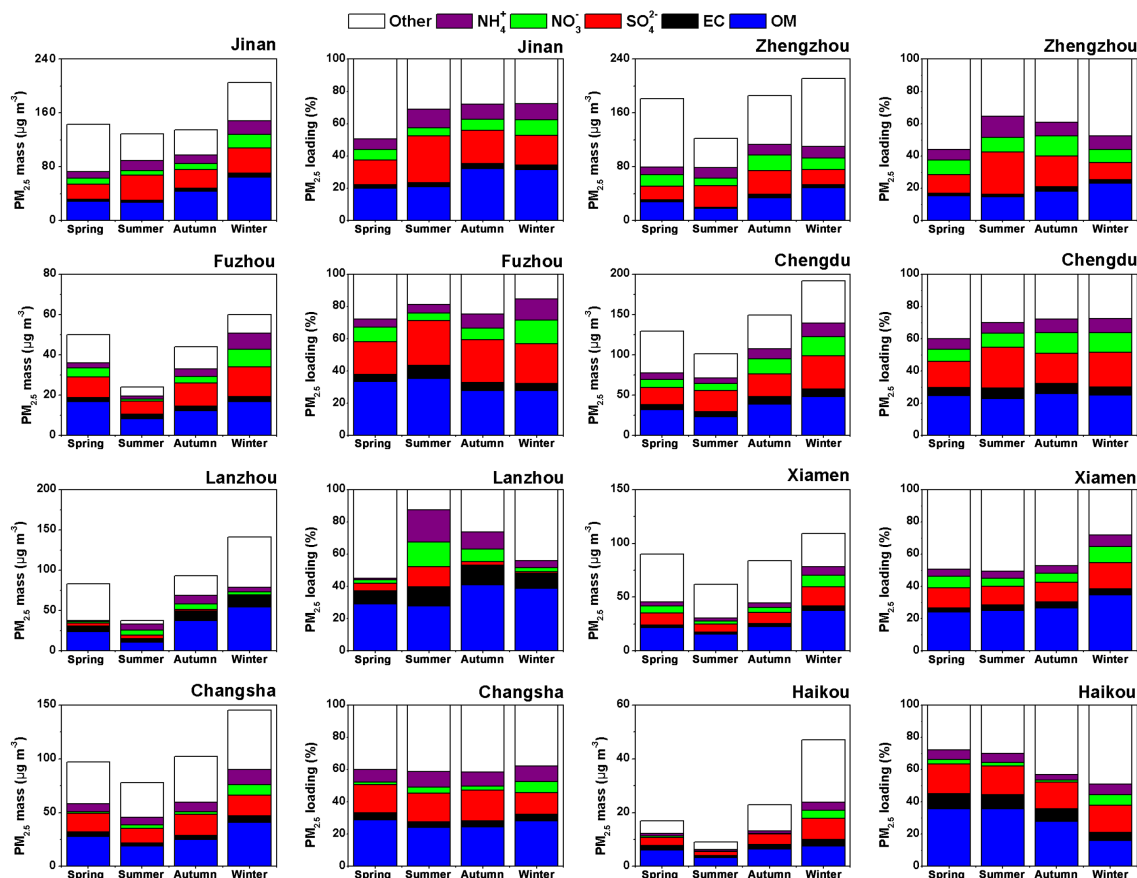


Figure 7. Seasonal PM_{2.5} and dominant chemical components in other cities.

was chosen for Fuzhou, Xiamen and Haikou and 1.8 for other cities based on their geographical locations.

Annual average contributions of secondary inorganic aerosols and carbonaceous aerosols to PM_{2.5} were 30 and 36 %, respectively, in the island city Haikou, similar to what was found in Beijing, and were 43–46 and 29–36 %, respectively, in the coastal cities Fuzhou and Xiamen, similar to what was found in Shanghai. Annual contributions of secondary inorganic aerosols ranged from 29 to 39 % in inland cities (Zhengzhou, Xi'an, Jinan, Chengdu, Chongqing and Changsha) except Lanzhou (15 %), which were comparable with those observed in PRD (33–41 %). In contrast, large differences were found in the annual contributions of carbonaceous aerosols, ranging from 23 % in Zhengzhou to 47 % in Chongqing. The sum of secondary inorganic aerosols and carbonaceous aerosols accounted for 56–79 % of the PM_{2.5} mass in these cities.

At an Asian continental outflow site (Penglai in Shandong Province), annual average contribution of secondary inorganic aerosols to PM_{2.5} reached 54 % (Feng et al., 2012b), evidently higher than those in urban and inland rural sites in China, while that of carbonaceous aerosols was 31 %, close to those in BTH. This finding suggested that intensive

emissions of SO₂ and NO_x in China enhanced the downward transport of secondary inorganic aerosols to the Pacific Ocean.

Seasonal average contributions are only shown here for Jinan (Yang et al., 2012), Zhengzhou (Geng et al., 2013), Fuzhou (Xu et al., 2012b), Chengdu (Tao et al., 2013a, 2014b), Lanzhou (Tan et al., 2016b; Wang et al., 2016c), Xiamen (Zhang et al., 2012a), Changsha (Tang et al., 2017) and Haikou (Liu et al., 2017a) due to the incomplete data in Xi'an and Chongqing (Fig. 7). Seasonal contributions of secondary inorganic aerosols were evidently higher in summer than in other seasons in Zhengzhou, Jinan and Lanzhou (typical northern cities), similar to what was seen in BTH. In the southwest city Chengdu and the central city Changsha, seasonal contribution of secondary inorganic aerosols in spring was only 30 and 27 %, respectively, lower than other seasons (40–42 and 30–31 %, respectively). In the two southern coastal cities Fuzhou and Xiamen, the highest seasonal average contribution of secondary inorganic aerosols was observed in winter (53 and 33 %, respectively), much higher than in other seasons (34–42 and 21–24 %, respectively). In the southern island city Haikou, seasonal contributions of secondary inorganic aerosols were also slightly higher in

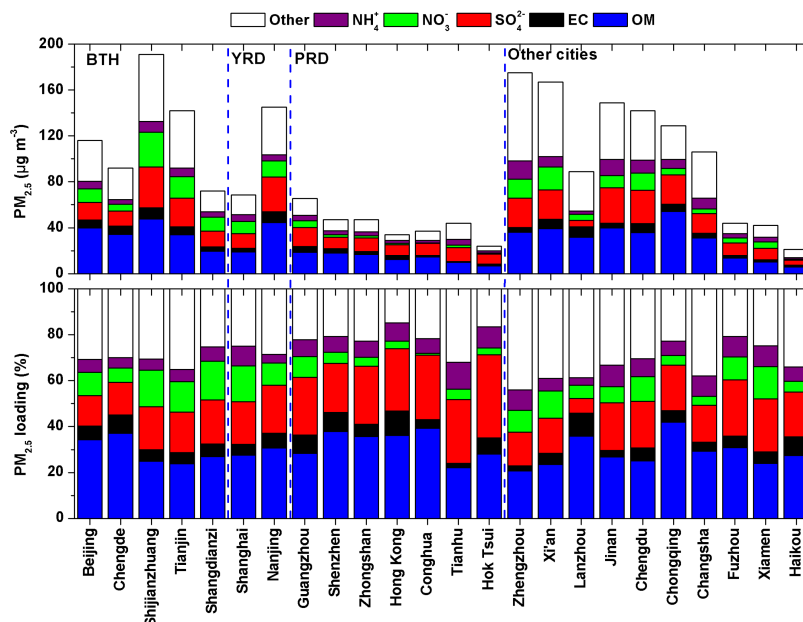


Figure 8. Annual PM_{2.5} and dominant chemical components in China.

winter (30 %) than in other seasons (21–27 %), similar to the coastal cities Fuzhou and Xiamen.

Seasonal average contributions of carbonaceous aerosols were evidently higher in cold seasons than in warm seasons in the three northern cities (Zhengzhou and Jinan and Lanzhou) due to heating activities and biomass burning, similar to BTH. Surprisingly, a similar seasonal pattern was also found in one coastal city Xiamen, e.g., 38 % in winter versus 27–30 % in other seasons. In contrast, higher seasonal contributions were found in warm season than in cold seasons in the southern coastal city (Fuzhou) and the southern island city (Haikou). No seasonal variations were found in the southwest inland city Chengdu (29–32 %) and the central inland city Changsha (28–33 %). The summed contributions of secondary inorganic aerosols and carbonaceous aerosols were evidently lower in spring than in other seasons in most of the northern cities (e.g., Jinan, Lanzhou, Zhengzhou and BTH), likely due to the frequent spring dust storm events in northern China.

2.2.5 Summary of PM_{2.5} chemical properties

Carbonaceous aerosols showed decreasing trends over the last 10 years (2000–2010) in Beijing, Shanghai and Guangzhou, consistent with BC emission trends in these cities and surrounding areas. SO₄²⁻ and NH₄⁺ remained at high levels with no significant trends in Beijing and Shanghai, but with an increasing trend in Guangzhou. NO₃⁻ showed increasing trends in all of the above-mentioned megacities. Annual mass concentrations of PM_{2.5}, secondary inorganic aerosols and carbonaceous aerosols showed similar spatial gradients decreasing from high- to low-latitude regions.

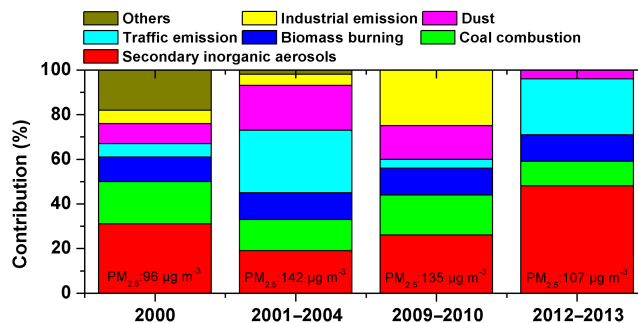


Figure 9. Interannual variations in PM_{2.5} and the dominant six sources in Beijing.

Annual average contributions of secondary inorganic aerosols to PM_{2.5} ranged from 25 to 48 % with higher values in southern regions, and those of carbonaceous aerosols ranged from 23 to 47 %, also with higher values in southern regions (Fig. 8). The percentage contributions of the sum of secondary inorganic aerosols and carbonaceous aerosols were higher in southern cities than in northern cities due to the frequent dust events in the north.

The highest seasonal average contributions of secondary inorganic aerosols to PM_{2.5} were observed in summer in most of the northern cities, but can occur in different seasons in southern cities. In contrast, the highest seasonal contributions of carbonaceous aerosols were observed in cold seasons in most of the northern cities, and in warm seasons in most of the southern cities. The different seasonal patterns were largely caused by heating and biomass burning in cold seasons in north China.

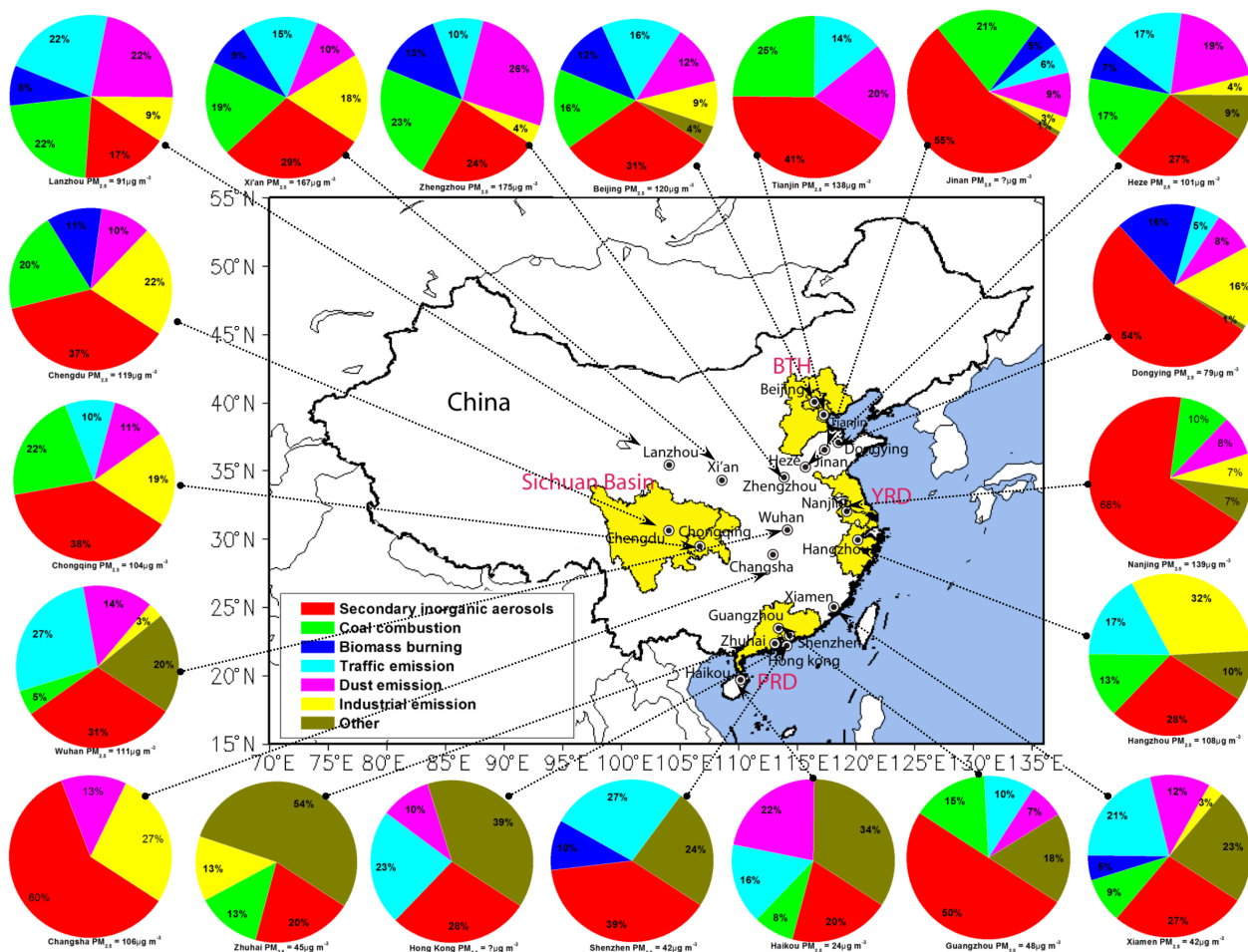


Figure 10. Annual contributions of PM_{2.5} dominant sources across China.

2.3 Source apportionment of PM_{2.5}

Advantages of receptor-based methods used for source apportionment analysis for various pollutants were discussed in I. Cheng et al. (2015) and Hopke (2016). Source apportionment studies of PM_{2.5} in China using receptor models have also been reviewed recently covering a wide range of topics (Liang et al., 2016; Lv et al., 2016; Pui et al., 2014; Zhang et al., 2017b). However, a general summary of spatial–temporal patterns of PM_{2.5} source factors and their relative contributions is still lacking, which is the focus of the discussion below. Data collected in this section are listed in Table S2 in the Supplement.

Commonly used receptor models in source apportionment of PM_{2.5} in China include Principal component analysis/absolute principal component scores (PCA/APCS), chemical mass balance receptor (CMB), positive matrix factorization (PMF) and UNMIX and the Multilinear Engine-2 model (ME-2). Among these, PMF and CMB models were the most widely used in China. Quantitative assessments of the uncertainties in using these methods are rare; studies us-

ing the same dataset collected in 2000 in Beijing and applying the above-mentioned models suggested that, while the models still identified the same dominant source factors, the relative contributions from these source factors differed by as much as 30 % between the different models (Song et al., 2006a, b). Similar magnitudes of uncertainties could also be caused by using different biomass burning tracers despite using the same receptor model (Tao et al., 2016b).

Major source factors identified for PM_{2.5} in most Chinese cities include secondary inorganic aerosols (SIA), coal combustion (COAL), biomass burning (BIOM), traffic emission (TRAF), dust emission (DUST) and industrial emission (INDU). Other source factors were also identified (and sometimes due to using more specific source names), such as metal manufacturing (including iron and steel industry, copper smelting) in industrial cities (e.g., Dongying and Tai'an of Shandong Province, Nanjing, Hangzhou, Lanzhou, Chengdu, Chongqing and Changsha), and sea salt and ship emissions in coastal cities (e.g., Longkou of Shandong Province, Nanjing, Guangzhou, Zhuhai and Hong Kong).

Contributions of dominant source factors to PM_{2.5} are discussed below in detail on a regional basis.

2.3.1 The Beijing–Tianjin–Hebei region

Studies in Beijing covered multiple years and mostly used the PMF model (Fig. 9). If averaging the results from the years in 2000 (Song et al., 2006b), 2001–2004 (Zhang et al., 2007), 2009–2010 (Zhang et al., 2013a), and 2012–2013 (Zíková et al., 2016), the six source factors (SIA, COAL, BIOM, TRAF, DUST and INDU) accounted for 31 ± 12 , 16 ± 4 , 12 ± 1 , 16 ± 13 , 12 ± 7 and $9 \pm 11\%$, respectively, of the PM_{2.5} mass in Beijing. There was an increasing trend for SIA contributions (from 19 to 48 %), a decreasing trend for COAL (from 19 to 11 %) and a stable trend for BIOM (11–12 %) during 2000–2013. There was more uncertainties in identifying TRAF and INDU than other source factors due to the differences in the source profiles.

A study in Tianjin in 2013–2014 only identified four dominant sources (SIA, COAL, TRAF and DUST) using the Multilinear Engine-2 model (ME-2), which accounted for 41, 25, 14 and 20 %, respectively, of the annual average PM_{2.5} mass (Tian et al., 2016). Compared with results in 2012–2013 in Beijing (Zíková et al., 2016), the contributions of SIA were comparable in the two cities, but those of COAL and DUST were much higher in Tianjin than Beijing. However, the results from an earlier study in Tianjin in 2009–2010 were much more comparable to those in Beijing during the same years in terms of PM_{2.5} levels and source attributions (Zhao et al., 2013c), implying faster decrease in COAL contribution in Beijing than Tianjin.

Seasonal results of source apportionment analysis are also available for Beijing (Huang et al., 2014b; Song et al., 2007; Wu et al., 2014; Zheng et al., 2005; Zhang et al., 2013a; Zíková et al., 2016). In most cases, SIA was the largest contributor in spring, summer and autumn, accounting for 26–61 % of the PM_{2.5} mass, while COAL was the largest contributor in winter, accounting for 13–57 % of the PM_{2.5} mass. The contributions of the other sources were lower than those of SIA and COAL, but subject to seasonal variations. For example, the largest seasonal contribution of BIOM was in autumn and of DUST in spring.

2.3.2 The Yangtze River Delta region

Studies for 1 year or longer were only made in Nanjing (Li et al., 2016a) and Hangzhou (Liu et al., 2015). Metal manufacturing was identified as a source factor in both Nanjing and Hangzhou, while ship emissions were also identified in Nanjing. Annual contributions of SIA to PM_{2.5} mass reached 68 % in Nanjing, while all the other sources (COAL, DUST, sea salt and ship emissions, and metal manufacturing) each contributed 10 % or less. In contrast, metal manufacturing, SIA, TRAF and COAL accounted for 32, 28, 17 and 13 %, respectively, of the PM_{2.5} mass in Hangzhou. Evidently, the

contributions of SIA in Nanjing were much higher than those in Hangzhou and cities in BTH. The contributions of COAL in Nanjing and Hangzhou were similar, but were evidently lower than those in cities in BTH.

Similar to the cities in BTH, the largest seasonal average contribution of SIA in Nanjing was in summer and of COAL in winter. Only winter data were available in Shanghai (Huang et al., 2014b), and the contributions of SIA and DUST to PM_{2.5} were similar between Shanghai and Nanjing.

2.3.3 The Pearl River Delta region

Studies covering 1 year or longer were available in Guangzhou (Tao et al., 2017), Shenzhen (Huang et al., 2013), Hong Kong (Guo et al., 2009a), and suburban Zhuhai (Tao et al., 2017) and suburban Hong Kong (Huang et al., 2014c). On an annual basis, SIA contributed 50 % to PM_{2.5} mass in Guangzhou, while other sources (ship emissions, COAL, TRAF and DUST) each contributed 7–17 %. In Shenzhen, SIA, TRAF and BIOM accounted for 39, 27 and 10 %, respectively, of the PM_{2.5} mass. In Hong Kong, SIA, TRAF, oil residue (related to Ni and V, or ship emissions), DUST, and sea salt accounted for 28, 23, 19, 10, and 7 %, respectively, of the PM_{2.5} mass.

Slightly different sources factors were identified in suburban studies. Annual contributions from mixed source (from regional transport), secondary nitrate and chloride, ship emissions, COAL, and electronic industries accounted for 36, 20, 18, 13, and 13 %, respectively, of the PM_{2.5} mass in suburban Zhuhai, while SIA, BIOM, sea salt, residual oil combustion (related to Ni and V, or ship emission), DUST, and TRAF accounted for 39, 20, 17, 12, 7, and 5 %, respectively, of the PM_{2.5} mass in suburban Hong Kong.

Despite the slightly different source factors identified between urban and suburban sites in PRD, SIA was the largest contributor to PM_{2.5} mass in this region. Ship emissions were identified in this region, but not in northern China, and this source factor contributed more than 10 % of the PM_{2.5} mass in all the studies except the one for Shenzhen. Similar to the cities in northern China, a high contribution from coal combustion was also found in Guangzhou and suburban Zhuhai.

Seasonal results of source apportionment analysis were available for four seasons in suburban Hong Kong (Huang et al., 2014c), winter in Guangzhou (Huang et al., 2014b), and summer and winter in Foshan (Tan et al., 2016c) and Hong Kong (Ho et al., 2006b). SIA was the largest contributor to PM_{2.5} among all the identified source factors in every season in suburban Hong Kong (30–45 %) and in winter in Guangzhou (59 %). In contrast, INDU was the largest contributor in winter in Foshan (39 %), a typical industrial city in PRD (Tan et al., 2016c). In suburban Hong Kong, seasonal average contribution of SIA was the lowest in summer, different from what was found for cities in BTH and YRD, while that of sea salt and ship emissions were the highest in sum-

mer due to the prevailing air masses from the South China Sea (Huang et al., 2014c).

2.3.4 Other cities

Studies covering 1 year or longer were mostly conducted for provincial capital cities, including Jinan (Yang et al., 2013), Zhengzhou (Geng et al., 2013), Xi'an (Wang et al., 2015c), Lanzhou (Wang et al., 2016c), Chengdu (Tao et al., 2014a), Chongqing (Chen et al., 2017), Changsha (Tang et al., 2017), Wuhan (Xiong et al., 2017), Xiamen (Zhang et al., 2016) and Haikou (Liu et al., 2017a), and for an inland city Heze (Liu et al., 2017b) and a regional background site (located in Yellow River Delta National Nature Reserve in the city of Dongying) (Yao et al., 2016), both in Shandong Province. Annual results were available from most studies, but were aggregated from seasonal results for Wuhan and Haikou. All the sites were grouped into four regions for easy discussion, i.e., northwest China (Lanzhou and Xi'an), southwest China or Sichuan Basin (Chengdu and Chongqing), eastern and central China (Jinan, Zhengzhou, Heze, Dongying, Wuhan and Changsha), and south coastal cities (Xiamen and Haikou).

The two northwest cities showed the same top four dominant source factors, although with slightly different percentage contributions to PM_{2.5} mass, e.g., 29 % from SIA, 19 % from COAL, 17 % from DUST and 15 % from TRAF in Xi'an, and 17 % from SIA and 22 % from the other three sources in Lanzhou. The lower SIA contribution in Lanzhou was likely due to the dry climate inhibiting formation of SIA. Similar results to those in Xi'an were also obtained in rural Xi'an, with SIA, COAL, DUST and TRAF contributing 31, 16, 20 and 13 %, respectively, to the PM_{2.5} mass (Wang et al., 2015c). The two southwest cities (Chengdu and Chongqing) showed nearly the same source-apportionment analysis results with SIA contributing just below 40 % and COAL and INDU each contributing around 20 % to PM_{2.5} mass.

The same top four dominant source factors (SIA, COAL, DUST and TRAF) found in the two northwest cities were also found in other capital cities (Jinan, Zhengzhou and Wuhan, except Changsha) and a medium-sized city (Heze) in eastern and central China, which accounted for 24–55, 14–23, 5–26 and 5–27 % of the PM_{2.5} mass, depending on location. In Changsha, SIA, a mixed source of INDU and BIOM, and DUST accounted for 60, 27 and 13 %, respectively, of PM_{2.5} mass. SIA, BIOM and INDU were the most important sources, accounting for 54, 16 and 16 %, respectively, of PM_{2.5} mass in a regional background site in Dongying. Similar source-apportionment results were found between the two south coastal cities (Xiamen and Haikou) with SIA, TRAF, DUST, COAL and sea salt accounting for 20–27, 16–21, 12–22, 8–9 and 6–10 %, respectively, of the PM_{2.5} mass.

Seasonal results of source apportionment analysis are available for four seasons in Jinan (Yang et al., 2013), a regional site in Dongying (Yao et al., 2016), Chengdu (Tao et al., 2014a), Chongqing (Chen et al., 2017), Wuhan (Xiong et

al., 2017) and Haikou (Liu et al., 2017a), for summer and autumn in Tai'an of Shandong Province (Liu et al., 2016a) and Xi'an (Xu et al., 2016), for summer and winter in Lanzhou (Tan et al., 2017), and for winter in Longkou (a coastal site in Shandong Province) (Zong et al., 2016).

In most seasons, SIA was the largest contributor to PM_{2.5} mass, e.g., in Jinan (30–45 %), Tai'an (27 %), a regional site in Dongying (35–72 %), Chengdu (33–44 %), Chongqing (24–52 %), Wuhan (23–41 %), Lanzhou (15–33 %) and Haikou (11–26 %), except during spring in Wuhan and summer in Haikou, when DUST was the largest contributor; during winter in Longkou and Xi'an, when COAL was the largest contributor; and during summer in Lanzhou, when smelting industry was the largest contributor. Only winter data were available in Longkou, and ship emissions contributed 9 % to PM_{2.5} mass, similar to what was found in the cities of PRD.

2.3.5 Summary of PM_{2.5} source apportionment studies

SIA, COAL and TRAF were the dominant source factors in most cities in China (Fig. 10). On an annual average, the sum of these three factors accounted for 63–80 % of PM_{2.5} mass in the cities of BTH region, 58–78 % in the cities of YRD region, 51–67 % in the cities of PRD region, 51–61 % in the northwest cities, 57–60 % in the southwest cities, 57–82 % in the eastern and central cities, and 44–57 % in the south coastal cities. The contributions of DUST were significant (7–26 %) in northern cities and a central city (Zhengzhou), of INDU significant (19–27 %) in typical industrial cities (e.g., Chengdu, Chongqing, Changsha), and of ship emission significant (7–19 %) in coastal and river cities (e.g., Longkou, Nanjing, Guangzhou, Zhuhai, Hong Kong). High seasonal contributions were found for SIA in summer, COAL in winter, DUST in spring and ship emission in summer in applicable cities.

It should be noted that SIA chemical compounds are formed by reactions in the atmosphere involving primary emissions of gaseous precursors that can be produced from any of the identified sources factors discussed above as well as from sources seldom mentioned in source apportionment studies such as agricultural emissions and many natural sources. However, sources of the gaseous precursors are often undetermined in source apportionment studies resulting in a large proportion of PM_{2.5} that cannot be explained (Karagulian et al., 2015). If the SIA contributions can be allocated to specific types of primary emissions, the overall percentage contributions from each of the identified source factor should be much higher, especially for COAL, TRAF, INDU and BIOM due to their high emission rates of primary pollutants of gaseous species. To identify the various types of primary emission sources, datasets containing trace element and other chemical markers need to be included in source apportionment models (Lee and Hopke, 2006). Combining receptor-based analysis results with source-based studies us-

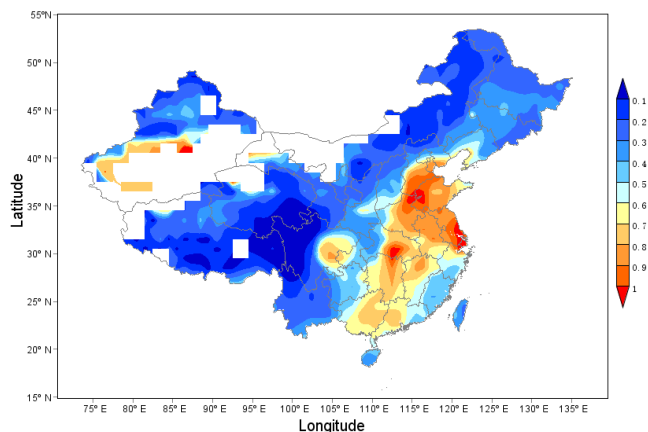


Figure 11. Spatial distribution of annual average aerosol optical depth (AOD) across China in 2014.

ing chemical transport models can provide a more complete picture by quantifying contributions of dominant emission sources to PM_{2.5} pollution.

3 Aerosol optical properties

There were much fewer measurements of aerosol optical properties than chemically resolved PM_{2.5} data in China. Data reviewed in this section are all listed in Table S3 in the Supplement. Measurements were available at (1) urban sites, including Beijing in BTH (Bergin et al., 2001; Garland et al., 2009; Han et al., 2014; He et al., 2009; Jing et al., 2015; Liu et al., 2009; Tian et al., 2015; Tao et al., 2015a; Wu et al., 2016; Zhao et al., 2011), Shanghai (T. Cheng et al., 2015; Feng et al., 2014; Han et al., 2015; Huang et al., 2014a; Li et al., 2013a; Xu et al., 2012a; Zha et al., 2014), Nanjing (Kang et al., 2013) and Shouxian (Anhui Province) in YRD (Fan et al., 2010), Guangzhou, Shenzhen and Hong Kong in PRD (Andreae et al., 2008; Cheng et al., 2006a, b, 2008a; Gao et al., 2015; Garland et al., 2008; Jung et al., 2009; Lan et al., 2013; Man and Shih, 2001; Tao et al., 2014c; Verma et al., 2010; Wu et al., 2009, 2013), Chengdu in southwest China (Tao et al., 2014b; Wang et al., 2017a) and Xi'an in northwest China (Cao et al., 2012a; Zhu et al., 2015); (2) rural sites, including rural Beijing (Shangdianzi) and rural Tianjin (Wuqing) in BTH (Ma et al., 2011; Yan et al., 2008; Zhao et al., 2011); (3) and remote sites in north and northwest China (Li et al., 2010; Xu et al., 2004; Yan, 2007). Sites with 1 year or longer data included Beijing, rural Beijing, Shanghai, Guangzhou, Chengdu, Xi'an and Shouxian.

Aerosol optical depth (AOD), representing the integrated light extinction coefficient in a vertical column, can be obtained from MODerate-resolution Imaging Spectroradiometer (MODIS) data. Satellite retrievals of AOD have been widely applied to estimate surface PM_{2.5} concentrations using statistical models (Liu et al., 2005; Hu et al., 2013; Ma et

al., 2014; Wang and Christopher, 2003). Although the correlation between AOD and PM_{2.5} mass concentration depends on many factors, such as aerosol size distribution, refractive index, single-scattering albedo and meteorological factors (Che et al., 2009; Guo et al., 2009b, 2017), the predicted PM_{2.5} mass from satellite AOD data compared well with ground-level measurements (Ma et al., 2014; Xie et al., 2015b). Moreover, the spatial distributions of AOD measured using sun photometers mostly agreed with those retrieved from satellite data (Che et al., 2014, 2015; Liu et al., 2016b; Pan et al., 2010). Spatial distributions of annual average AOD in 2014 are shown in Fig. 11. The spatial distributions of PM_{2.5} shown in Fig. 2 are similar to the patterns of AOD shown in Fig. 11. Differences in fine structures of their patterns were due to surface PM_{2.5} versus column AOD comparison and spatial variations in PM_{2.5} chemical composition.

In this section, geographical patterns of the aerosol optical properties including b_{sp} and b_{ap} measured on ground base in major Chinese cities are first discussed (Sect. 3.1). Temporal patterns of b_{sp} and b_{ap} on annual and seasonal scales are then discussed for major regions (Sect. 3.2). Fewer studies were available for b_{ap} than b_{sp} ; however, the measured BC concentrations (at 880 nm wavelength) can be converted to b_{ap} (at 532 nm wavelength) by a factor of $8.28 \text{ m}^2 \text{ g}^{-1}$ (Wu et al., 2009).

3.1 Geographical patterns

Annual average b_{sp} and b_{ap} from ground measurements in major cities in China are plotted in Fig. 12. Most b_{sp} measurements were conducted using a nephelometer under $\text{RH} < 60\%$. The highest annual b_{sp} was in Xi'an (525 Mm^{-1} , $\text{RH} < 60\%$) (Cao et al., 2012a), followed by Chengdu (456 Mm^{-1} , $\text{RH} < 40\%$; 421 Mm^{-1} , ambient RH) (Tao et al., 2014b; Wang et al., 2017a), Guangzhou (326 Mm^{-1} , $\text{RH} < 70\%$) (Tao et al., 2014c), Beijing (309 Mm^{-1} , $\text{RH} < 60\%$) (He et al., 2009; Jing et al., 2015; Zhao et al., 2011) and Shanghai (217 Mm^{-1} , $\text{RH} < 60\%$) (T. Cheng et al., 2015). Such spatial patterns were mainly due to the spatial patterns of annual PM_{2.5} mass, i.e., Xi'an ($177 \mu\text{g m}^{-3}$) > Chengdu ($111 \mu\text{g m}^{-3}$) > Beijing ($108 \mu\text{g m}^{-3}$) > Shanghai ($77 \mu\text{g m}^{-3}$) > Guangzhou ($65 \mu\text{g m}^{-3}$), and partly due to humidity conditions, e.g., Beijing versus Guangzhou. Noticeably, b_{sp} in Shouxian County was higher than those in several megacities (e.g., Beijing, Shanghai and Guangzhou), suggesting hazy weather also frequently occurred even in small cities in China (Fan et al., 2010). b_{sp} in rural Beijing was 179 Mm^{-1} (Yan et al., 2008; Zhao et al., 2011), which was much lower than that in urban Beijing, but was close to the level in Shanghai.

Annual average b_{ap} ranged from 37 to 96 Mm^{-1} with higher values observed in Chengdu and Xi'an (likely due to popular biomass burning besides large amount of coal burning) (Cao et al., 2012a; Tao et al., 2014a, b; Wang et al., 2017a; Zhang et al., 2014b), and lower values in Shouxian

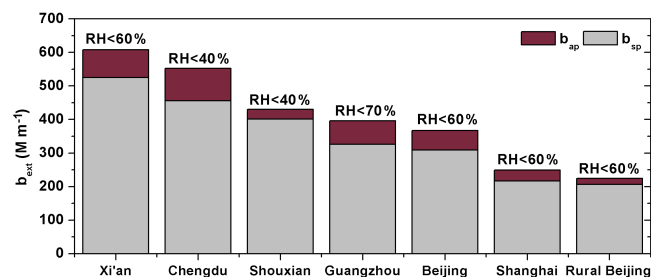


Figure 12. Annual b_{sp} and b_{ap} in China.

and rural Beijing (Fan et al., 2010; Yan et al., 2008; Zhao et al., 2011). b_{ap} in Guangzhou was higher than that in Beijing and Shanghai despite their similar PM_{2.5} EC levels, likely due to the different coating of EC in Guangzhou than in other cities. For example, the mass absorption of EC in Guangzhou was $8.5 \text{ m}^2 \text{ g}^{-1}$ (at 532 nm) in autumn 2004 (Andreae et al., 2008), which was higher than that ($4.2 \text{ m}^2 \text{ g}^{-1}$ at 870 nm, equivalent to $7.2 \text{ m}^2 \text{ g}^{-1}$ at 532 nm) in winter 2013 in Beijing (Wu et al., 2016).

3.2 Temporal patterns

3.2.1 The Beijing–Tianjin–Hebei region

b_{sp} measurements in BTH longer than 1 year were only available in Beijing, including the years of 2005, 2006, 2008–2009 and 2009–2010 (He et al., 2009; Jing et al., 2015; Zhao et al., 2011). Annual b_{sp} in Beijing increased by 36 % from 264 Mm^{-1} in 2005 to 360 Mm^{-1} in 2009–2010, while PM_{2.5} increased by 20 % from 107 to $129 \mu\text{g m}^{-3}$ during the same period. However, annual b_{ap} in 2009–2010 was 64 Mm^{-1} , which was slightly higher than 56 Mm^{-1} in 2005–2006, although the annual EC in 2009–2010 was evidently lower than that in 2005–2006. Meanwhile, annual secondary inorganic aerosols in 2009–2010 were evidently lower than that in 2005–2006. The coating by secondary inorganic aerosols likely enhanced the absorption of EC (Bond et al., 2006; Cheng et al., 2009; Yu et al., 2010).

b_{sp} measurements in rural Beijing included the years 2003–2005 (175 Mm^{-1}) and 2008–2009 (182 Mm^{-1}), while b_{ap} only included the years of 2003–2005 (18 Mm^{-1}) (Yan et al., 2008; Zhao et al., 2011). Considering all of the above-mentioned data together, we can conclude that b_{sp} and b_{ap} showed slightly increasing tendencies in urban and rural Beijing in recent years.

Seasonal variations in b_{sp} and b_{ap} at urban and rural sites in Beijing are plotted in Fig. 13. The highest seasonal average b_{sp} in Beijing was observed in winter and the lowest in spring with seasonal variations up to a factor of 1.7 (Bergin et al., 2001; Garland et al., 2009; Han et al., 2014; He et al., 2009; Jing et al., 2015; Li et al., 2013b; Liu et al., 2009; Tao et al., 2015a; Tian et al., 2015; Zhao et al., 2011). A different seasonal pattern was seen at the rural site located north

of Beijing, which showed 10–26 % higher values in summer than in the other seasons (Yan et al., 2008; Zhao et al., 2011). The highest seasonal b_{sp} in winter in Beijing was consistent with the highest seasonal PM_{2.5} mass. However, in rural Beijing the highest PM_{2.5} mass was observed in spring due to the frequent dust storm events, and the second highest seasonal average PM_{2.5} mass in summer, which corresponded to the highest seasonal b_{sp} . This is because scattering efficiency of dust aerosols was lower than that of anthropogenic aerosols (Zhao et al., 2011).

The highest seasonal b_{ap} in Beijing appeared in autumn and the lowest in spring with seasonal variations up to a factor of 2.0 (Bergin et al., 2001; Garland et al., 2009; He et al., 2009; Jing et al., 2015; Li et al., 2013c; Liu et al., 2009; Tian et al., 2015; Wu et al., 2016). Seasonal variations in b_{ap} were different from those of b_{sp} due to their dependence on different chemical compounds, i.e., b_{sp} mainly on PM mass while b_{ap} mainly on EC mass in PM and its coating. In rural Beijing b_{ap} was lower by 19–57 % in summer than in other seasons, and with similar seasonal variations to b_{sp} , suggesting aerosols in rural Beijing mainly came from regional transport (Yan et al., 2008).

At the rural site in Tianjin (Wuqing) located between Beijing and Tianjin, only spring and summer 2009 and winter 2010 data were available, which gave a seasonal average of 280 Mm^{-1} in spring, 379 Mm^{-1} in summer and 485 Mm^{-1} in winter for b_{sp} , and 47 Mm^{-1} in spring and 43 Mm^{-1} in summer for b_{ap} (Fig. 13) (J. Chen et al., 2014b; Ma et al., 2011). These seasonal values in Wuqing were higher than those observed at the rural sites near Beijing, likely because Wuqing is close to and downwind of Tianjin and Hebei Province, where major pollutant sources are located.

3.2.2 The Yangtze River Delta and Pearl River Delta region

No multi-year b_{sp} measurement data were available for exploring interannual variations, although multi-year measurements of BC or b_{ap} were made in Shanghai (YRD) and Guangzhou (PRD). Annual b_{ap} in 2011–2012 (19 Mm^{-1}) was evidently lower than that in 2010 (31 Mm^{-1}) in Shanghai (Feng et al., 2014; Zha et al., 2014), consistent with the trend of EC, e.g., annual concentration of EC in 2012 ($2.0 \mu\text{g m}^{-3}$) was only half of that in 2009 ($4.1 \mu\text{g m}^{-3}$) (Wang et al., 2016b; Zhao et al., 2015b). In Guangzhou, annual b_{ap} in 2007 (51 Mm^{-1}) was also evidently lower than that in 2004 (90 Mm^{-1}) (Wu et al., 2009), while EC in 2006–2007 ($4.0 \mu\text{g m}^{-3}$) was similar or slightly lower than that in 2002–2003 ($4.4 \mu\text{g m}^{-3}$) (Hagler et al., 2006; Huang et al., 2012). Thus, the interannual variations in b_{ap} were mainly determined by EC trends in the same cities.

b_{sp} and b_{ap} in winter were evidently higher than those in spring in Shanghai, consistent with the seasonal patterns of PM_{2.5} and EC, respectively (Fig. 13) (Cao et al., 2012b; T. Cheng et al., 2015; Feng et al., 2014; Han et al., 2015;

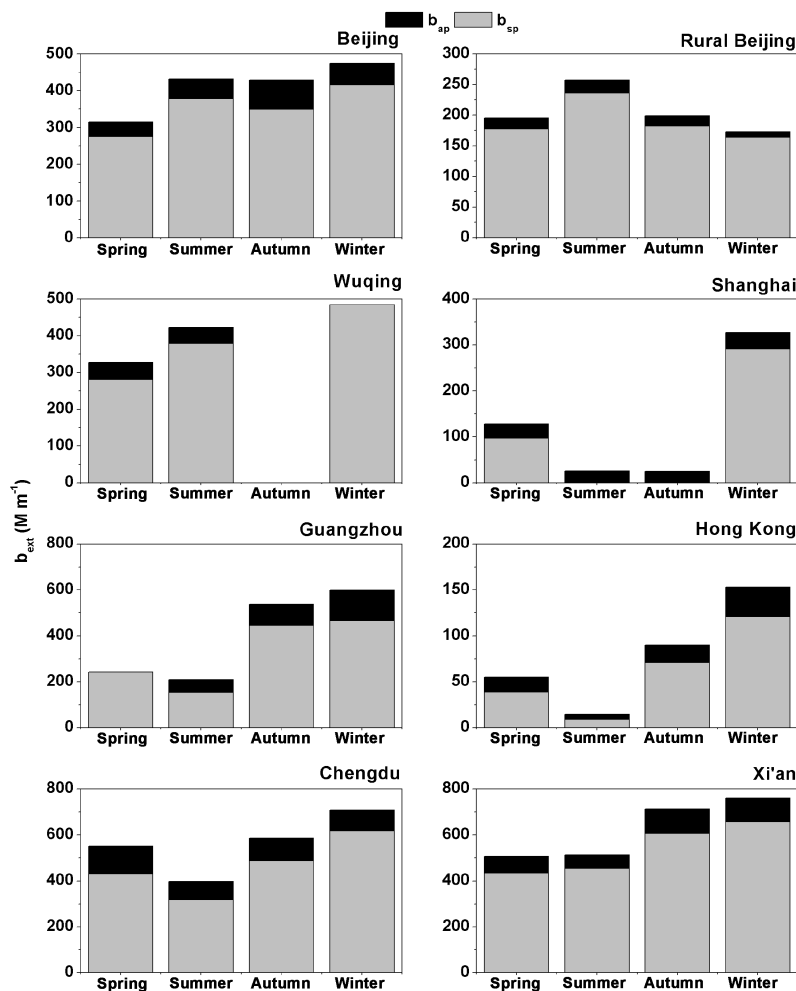


Figure 13. Seasonal b_{sp} and b_{ap} in cities with measurements.

Huang et al., 2014a; Li et al., 2013a; Pathak et al., 2011; Wang et al., 2016b; Xu et al., 2012a; Ye et al., 2003; Zha et al., 2014; Zhao et al., 2015a). Similar seasonal variations were found for b_{sp} and b_{ap} in the two PRD cities (Guangzhou and Hong Kong), which also agreed with the patterns of PM_{2.5} and EC (Andreae et al., 2008; Cao et al., 2004, 2012b; Cui et al., 2015; Gao et al., 2015; Huang et al., 2014b; Jung et al., 2009; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tao et al., 2009, 2014c, 2015b, 2017; Verma et al., 2010; Wu et al., 2009, 2013). The highest b_{sp} and b_{ap} appeared in winter and the lowest in summer with seasonal variations up to a factor of 3.1 and 17.1 for b_{sp} , 2.3 and 5.9 for b_{ap} , in Guangzhou and Hong Kong, respectively.

3.2.3 Other cities

In Chengdu of southwest China, the highest b_{sp} appeared in winter and the lowest in summer with seasonal variations up to a factor of 1.9, which was consistent with the seasonal pattern of PM_{2.5} (Tao et al., 2014a, b). However, the highest

b_{ap} appeared in spring despite the highest EC in winter (Tao et al., 2014b). One explanation could be the large amount of OC emitted from biomass burning in spring, which enhanced the absorption of EC (Schneider et al., 2005; Tao et al., 2013b). b_{sp} and b_{ap} in winter were evidently higher than those in summer in Xi'an in northwest China, consistent with the seasonal patterns of PM_{2.5} and EC, respectively (Cao et al., 2009, 2012a; Wang et al., 2015c).

Seasonal measurements of b_{sp} and b_{ap} were also made at remote sites (Dunhuang, Yulin and Zhangye of Gansu Province, Dongsheng of Inner Mongolia) focusing on dust aerosols and only covered spring and winter (Li et al., 2010; Xu et al., 2004; Yan, 2007). b_{sp} in winter ranged from 303 to 304 Mm⁻¹, which is double that in spring (126 to 183 Mm⁻¹).

4 Relationships between aerosol optical properties and PM_{2.5} mass concentrations

4.1 Mass scattering efficiency of PM_{2.5}

b_{sp} and PM_{2.5} mass concentration have been found to correlate well in numerous field studies (Andreae et al., 2008; Han et al., 2015; Hand and Malm, 2007b; Jung et al., 2009; Pu et al., 2015; Tao et al., 2014b, c, 2015a, 2016a; Tian et al., 2015; Wang et al., 2012b; Zhao et al., 2011). A parameter describing their relationship is defined as mass scattering efficiency (MSE), which is the slope of the linear regression of b_{sp} against PM_{2.5} mass. MSE was found to vary with location and season due to the variations in PM_{2.5} chemical composition. Some of the variations may be due to different sampling conditions, e.g., ambient (controlled RH < 60 %) versus dry condition (controlled RH < 40 %), online versus filter-based PM_{2.5} sampling. Available MSE data are discussed here, although uncertainties from measurements will not be addressed in this study.

In BTH, annual average PM_{2.5} MSE was higher in Beijing (5.9 m² g⁻¹) than in rural Beijing (4.8 m² g⁻¹) based on online PM_{2.5} mass (Zhao et al., 2011). In urban Beijing in winter of 2013, PM_{2.5} MSE increased to 4.9 m² g⁻¹ during a heavily pollution episode and decreased to 3.6 m² g⁻¹ during clean days, due to a large fraction of soluble inorganic components (e.g., (NH₄)₂SO₄ and NH₄NO₃) in PM_{2.5} under heavy polluted conditions (Tao et al., 2015a). In rural Beijing in 2005–2010, dust episodes had lower PM_{2.5} MSE (0.7 m² g⁻¹) and anthropogenic pollution episodes had higher PM_{2.5} MSE (4.3 m² g⁻¹) (Pu et al., 2015).

In YRD, annual average PM_{2.5} MSE ranged from 3.8 m² g⁻¹ in Ningbo to 5.3 m² g⁻¹ in Hangzhou with a regional urban average (including the cities of Nanjing, Shanghai, Suzhou, Hangzhou and Ningbo) of 4.1 m² g⁻¹ in 2011–2012 (Cheng et al., 2013b). PM_{2.5} MSE in Lin'an (4.0 m² g⁻¹), a rural site of YRD, was close to the regional urban average value in YRD (Xu et al., 2002). PM_{2.5} MSE in Shanghai reached 5.6 m² g⁻¹ in winter of 2012 (Han et al., 2015), which was higher than that in Beijing in the same season (Tao et al., 2015a).

In PRD, annual average PM_{2.5} MSE in Guangzhou was 3.5 m² g⁻¹ with seasonal average ranging from 2.3 m² g⁻¹ in summer to 4.5 m² g⁻¹ in autumn in 2009–2010 (Tao et al., 2014c). These values were close to 4.2 m² g⁻¹ (Andreae et al., 2008) and 2.7 m² g⁻¹ (Jung et al., 2009) measured in the same city in autumn of 2004. However, PM_{2.5} MSE in rural Guangzhou (Wanqingsha, south of Guangzhou) was 5.3 m² g⁻¹ (Wang et al., 2012), which was evidently higher than that in Guangzhou in the same season (Tao et al., 2014c).

In southwest China, seasonal average PM_{2.5} MSE ranged from 3.5 to 4.4 m² g⁻¹ in Chengdu in 2011 (Tao et al., 2014b). In northwest China, PM_{2.5} MSE was 3.0 m² g⁻¹ for anthropogenic pollution and 1.0 m² g⁻¹ for dust pollution at

a remote site (Yulin, located at the interface of the desert and loess regions, Shanxi Province), which was similar to rural Beijing (Xu et al., 2004).

In summary, annual PM_{2.5} MSE typically ranged from 3.5 to 5.9 m² g⁻¹ in urban areas in China with higher values in north China and lower values in south China. Seasonal average PM_{2.5} MSE typically ranged from 2.3 to 5.6 m² g⁻¹ with higher values in winter and autumn and lower values in spring and summer. Generally, PM_{2.5} MSE typically ranged from 3.0 to 5.0 m² g⁻¹ for anthropogenic pollution and from 0.7 to 1.0 m² g⁻¹ for natural dust aerosols.

4.2 Mass absorption efficiency of EC and organic matter

EC is the dominant absorption species in PM_{2.5}. Similar to PM_{2.5} MSE, the slope between b_{ap} and EC mass was defined as mass absorption efficiency (MAE) of EC. Various instruments have been used to measure b_{ap} including an Aethalometer, multi-angle absorption photometer (MAAP), Radiance Research particle soot absorption photometer (PSAP) and photoacoustic spectrometer (PAS), with the first two instruments measuring attenuation of the sample on the filter for estimating BC mass concentration, and the last two measuring b_{ap} directly. Most studies in China used an Aethalometer and MAAP. BC mass concentrations (880 nm) were converted to b_{ap} (532 nm) using an empirical constant of 8.28 m² g⁻¹, which was obtained by the regression between BC mass and b_{ap} synchronously measured in autumn in Guangzhou, keeping in mind that application of an empirical constant obtained from one specific study to other cases may cause large uncertainties (Wu et al., 2009).

EC MAE was 7.5–8.5 m² g⁻¹ in winter and 9.4 m² g⁻¹ in summer in Beijing (632 nm) (Cheng et al., 2011; Wu et al., 2016). The higher EC MAE in summer was likely due to more coating of EC in higher ambient humidity (Wu et al., 2016). BC MAE was 6.5 m² g⁻¹ at 532 nm in autumn in Shenzhen of PRD (Lan et al., 2013). However, BC MAE was 12.4 m² g⁻¹ at 532 nm in winter in Xi'an (Wang et al., 2014a). Moreover, EC MAE of diesel was 8.4 m² g⁻¹ (632 nm), which was higher than those (3.0–6.8 m² g⁻¹) of biomass burning sources (e.g., crop residual and wood) (Cheng et al., 2011).

Organic matter or brown carbon is also a strong light absorbing material at short wavelengths. Available MAE values of OC include 0.76 m² g⁻¹ (532 nm) in autumn in 2008 in Guangzhou (Andreae et al., 2008). Moreover, available MAE values of water-soluble organic carbon (WSOC) include 1.79 and 0.71 m² g⁻¹ (365 nm) in winter and summer, respectively, in Beijing (Cheng et al., 2011). The WSOC MAEs of wood, grass, corn and diesel tractors were 0.97, 0.90, 1.05 and 1.33 m² g⁻¹ (365 nm), respectively, which were much higher than that of gasoline motorcycles (0.20 m² g⁻¹, 365 nm) (Du et al., 2014b). Evidently, the MAEs of OC or

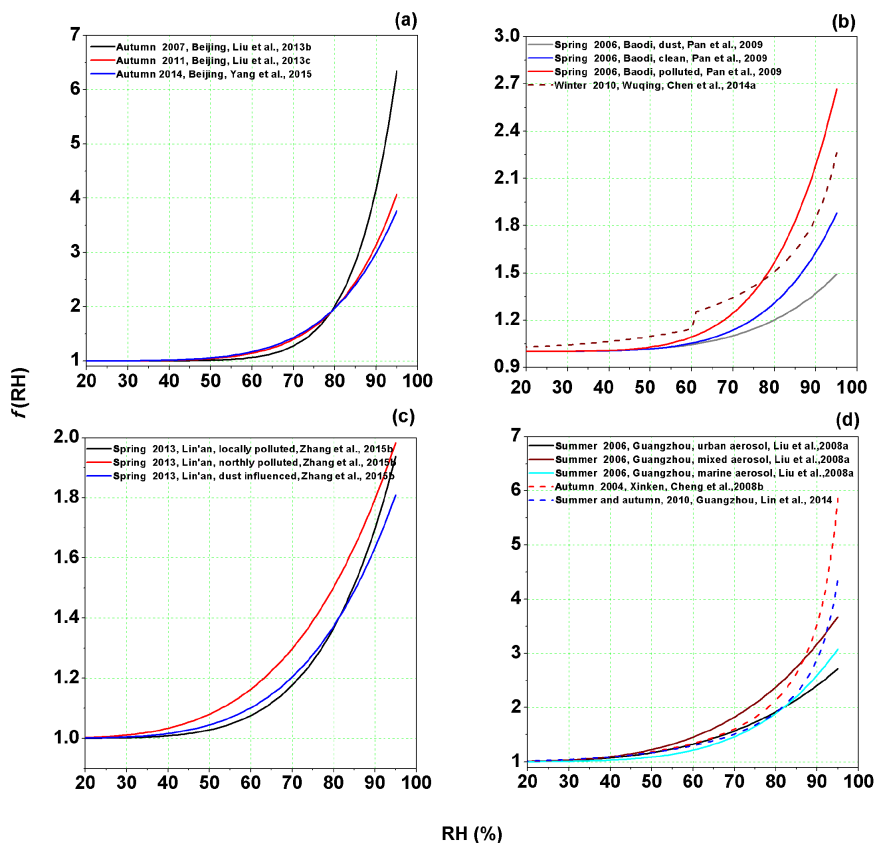


Figure 14. The hygroscopic growth curves in different sites in China.

WSOC should not be neglected for short wavelength absorption.

4.3 Aerosol hygroscopic properties

b_{sp} under ambient condition can differ significantly from dry conditions due to hygroscopic properties of soluble aerosol chemical components. A relationship between ambient and dry b_{sp} is thus developed for estimating ambient b_{sp} from measured dry b_{sp} , which is often described by the hygroscopic growth curve ($f(\text{RH})$) as a function of RH: $f(\text{RH}) = 1 + a \times (\text{RH}/100)^b$. Here, a and b are empirical fitting parameters. Only a few studies conducted in Beijing, Wuqing, Lin'an and Guangzhou provided the aerosol hygroscopic curves (Table S4 in the Supplement). Three different methods have been used to obtain $f(\text{RH})$. The first one measures simultaneously dry and wet b_{sp} using a nephelometer and visibility meter, respectively. The second one measures wet b_{sp} by integrating a nephelometer equipped with a humidifier, and the third one estimates dry and wet b_{sp} based on Mie theory with size-resolved chemical components.

Available $f(\text{RH})$ curves in China are summarized in Fig. 14. The three $f(\text{RH})$ curves in autumn of 2007, 2011 and 2014 in urban Beijing were all measured using the first method (Fig. 14a) (Liu et al., 2013b, c; Yang et al., 2015).

The two $f(\text{RH})$ curves measured in 2011 and 2014 were quite close, but the one in 2007 was lower under $\text{RH} < 80\%$ and higher under $\text{RH} > 80\%$, likely due to aerosol chemical composition and size distribution changes in these years.

The $f(\text{RH})$ curves at four rural sites were measured using the second method, including Baodi of Tianjin in spring, Wuqing of Tianjin in winter (Fig. 14b) (J. Chen et al., 2014b; Pan et al., 2009), Raoyang of Hebei Province in summer (Wu et al., 2017) and Lin'an of Zhejiang Province in spring (Fig. 14c) (Zhang et al., 2015b). It is known that the hygroscopic chemical components are mostly water-soluble inorganic salts (e.g., $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3), while mineral dust and organic matter are mostly hydrophobic. In Baodi in the spring season, the concentrations of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 and their mass fractions in fine-mode particles were higher during polluted episodes than during clean periods or dust storm episodes, resulting in higher $f(\text{RH})$ values during the polluted episode. $f(\text{RH})$ values measured in winter in Wuqing were evidently higher than those measured in spring in Baodi under $\text{RH} < 80\%$, likely due to more hygroscopic chemical components in winter in Wuqing. In Raoyang, a different fitting curve of $f(\text{RH})$ was obtained with a much higher $f(\text{RH} = 80\%)$ value (2.3) than in other rural sites in BTH mentioned above, likely due to higher frac-

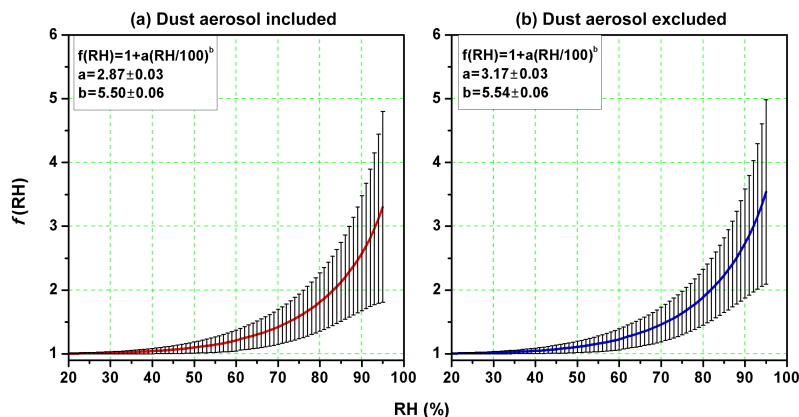


Figure 15. Distribution of the hygroscopic growth curves in China.

tions of hydrophilic components in PM_{2.5} (> 56 %). In all the BTH sites, $f(\text{RH})$ value increased continuously with increasing RH. However, in a different study an abrupt increase in $f(\text{RH})$ at RH values of 73–81 % was observed in summer in Wangdu of Hebei Province due to the deliquescence of ammonium sulfate (Kuang et al., 2016). Similar to what was found in Baodi, $f(\text{RH})$ values during polluted episodes were also higher than those during dust episodes in Lin'an, but the differences between polluted and dust periods were smaller in Lin'an than in Baodi. Noticeably, the $f(\text{RH})$ values during polluted episodes were similar in Lin'an and Baodi, e.g., $f(\text{RH} = 80\%)$ was 1.5 and 1.6, respectively.

The $f(\text{RH})$ curves (solid lines) in summer in urban Guangzhou were measured by the first method, while those (dotted lines) in autumn in rural Guangzhou and in summer and autumn seasons in urban Guangzhou were measured by the third method (Fig. 14d) (Cheng et al., 2008b; Lin et al., 2014; Liu et al., 2008b). $f(\text{RH} = 80\%)$ values were 2.04 and 2.68, respectively, for urban aerosols originating from the north and marine aerosols originating from the South China Sea. $f(\text{RH} < 80\%)$ curves were similar in urban and rural Guangzhou; however, $f(80\% < \text{RH} < 90\%)$ values in rural Guangzhou were evidently higher than those in urban Guangzhou, likely due to the much higher fraction of secondary inorganic aerosols in fine-mode particles in rural Guangzhou than urban Guangzhou in the dry season (Lin et al., 2014; Liu et al., 2008a).

If averaging all available $f(\text{RH})$ curves shown in Fig. 15, the empirical fitting parameters a and b were found to be 2.87 ± 0.03 , 5.50 ± 0.06 , respectively (Fig. 15a). But if excluding dust episodes in Baodi and Lin'an (Fig. 15b), the empirical fitting parameters a and b were 3.17 ± 0.03 , 5.54 ± 0.06 , respectively (Fig. 15b). Based on the average $f(\text{RH})$ curve, b_{sp} under ambient condition (RH = 80 %) can be amplified by about 1.8 times that under dry conditions in China. This suggests that reducing inorganic water-soluble salts is critical in alleviating hazy weather in China.

4.4 Source apportionment of haze in China

To investigate the contributions of PM_{2.5} chemical components to b_{sp} , a revised formula developed by the original IMPROVE method is applied in this section (Pitchford et al., 2007). The revised IMPROVE formula can be simplified as follows:

$$b_{\text{ext}} \approx 2.2 \times f_{\text{S}} \times [\text{Small}(\text{NH}_4)_2\text{SO}_4] + 4.8 \times f_{\text{L}} \times [\text{Large}(\text{NH}_4)_2\text{SO}_4] + 2.4 \times f_{\text{S}} \times [\text{Small NH}_4\text{NO}_3] + 5.1 \times f_{\text{L}} \times [\text{Large NH}_4\text{NO}_3] + 2.8 \times [\text{Small OM}] + 6.1 \times [\text{Large OM}] + 1.0 \times [\text{Other}] + 10 \times [\text{EC}], \quad (1)$$

$$[\text{Large X}] = [\text{Total X}]^2/20, \text{ for } [\text{Total X}] < 20, \quad (2)$$

$$[\text{Large X}] = [\text{Total X}], \text{ for } [\text{Total X}] \geq 20, \quad (3)$$

$$[\text{Small X}] = [\text{Total X}] - [\text{Large X}], \quad (4)$$

where X represents $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and OM. RH growth curves of f_{S} and f_{L} for $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 can be found in Pitchford et al. (2007).

Using the chemical composition data shown in Fig. 8 and annual average RH values in major cities in China as input (<http://data.cma.cn/>), the estimated annual b_{ext} and its load percentages under dry and ambient conditions are plotted in Fig. 16. For b_{ext} under dry conditions, carbonaceous aerosols had similar percentage contributions to secondary inorganic aerosols in Shijiazhuang, Tianjin, Shangdianzi, Shanghai, Hok Tsui, Zhengzhou, Xi'an, Jinan, Chengdu, Fuzhou and Xiamen, but the percentage contributions were 11–65 % higher in other urban and rural sites. However, under ambient conditions the contributions of secondary inorganic aerosols were evidently higher (by 2–54 %) than those of carbonaceous aerosols in most cities except in Beijing, Chengde, Lanzhou and Chongqing. Noticeably, the contributions of secondary inorganic aerosols for b_{ext} sharply increased by about 18–25 % under ambient conditions than dry conditions in humid (RH > 70 %) cities (e.g., Haikou, Changsha, Xiamen, Nanjing, cities in PRD, and Chengdu).

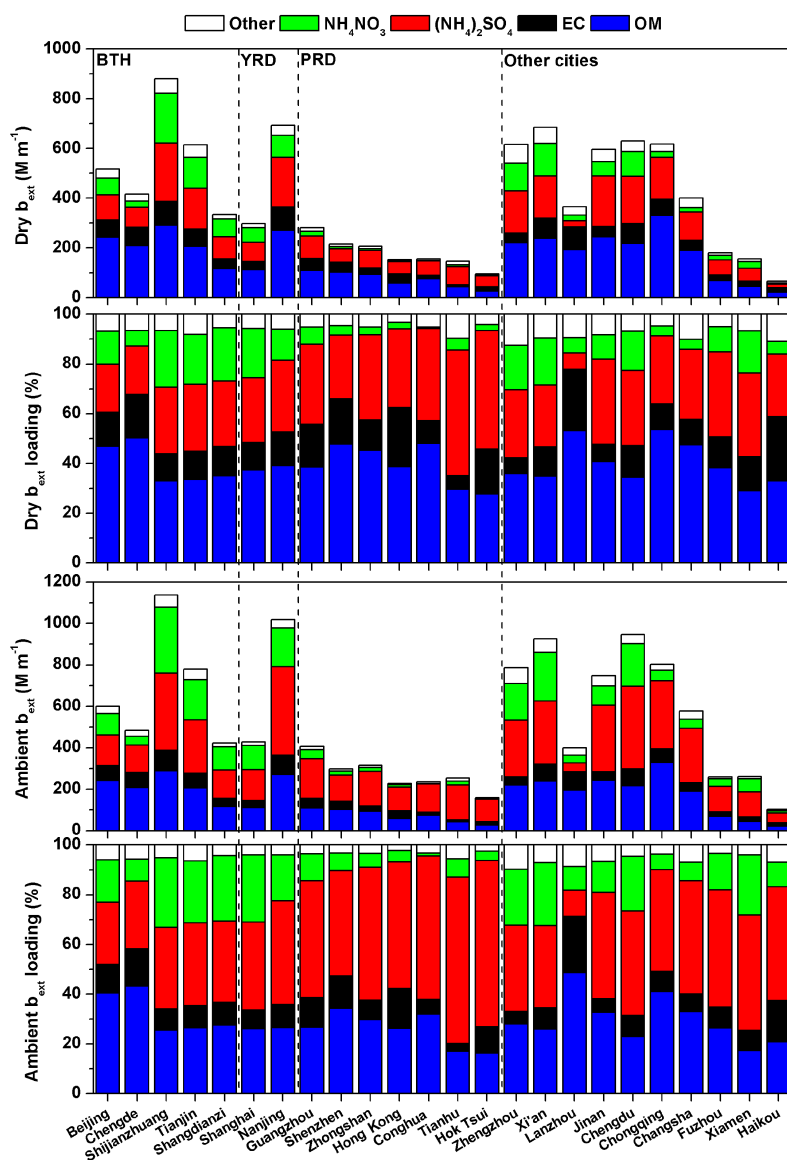


Figure 16. Annual b_{ext} percentage loading under dry and ambient conditions at urban sites in China.

5 Implications for aerosol pollution controls

There is no doubt that reduction of PM_{2.5} will be the ultimate approach for improving visibility and alleviating hazy weather. Industrial emission contributions to secondary inorganic aerosols were the dominant sources of PM_{2.5} in urban areas in China (Liang et al., 2016). Aerosols produced from traffic emissions, biomass burning and soil dust were also important sources in north China. Secondary inorganic aerosols were formed from atmospheric reactions involving SO₂ and NO_x, which were mainly emitted from coal combustion, the major energy source in China for decades.

A series of regulations controlling coal combustion emissions have been made since the first version of the NAAQS was promulgated in 1982. The Air Pollution Prevention law

of the People's Republic of China was promulgated in 1987, which was the milestone in air pollution prevention history in China. It also marked the beginning of a new era for preventing air pollution based on the national law, followed by a series of regulations for controlling coal combustion. During 1990–2000, most of the control measures or technologies (e.g., desulfurization and dedusting for coal combustion) were focused on reducing SO₂ emissions. The measure for gross control of SO₂ emissions had been enforced since 1996. Despite these efforts, the amount of SO₂ emissions increased by about 28% from 2000 to 2005 (<http://www.zhb.gov.cn/>). The amount of SO₂ emission began to decrease in 2006 and gradually reduced to the emission level of 2000 in 2010 (<http://www.zhb.gov.cn/>). Meanwhile, am-

bient annual SO₂ concentration in urban cities in China also decreased from 57 μg m⁻³ in 2005 to 40 μg m⁻³ in 2010 (<http://www.zhb.gov.cn/>). Apparently, the emission control efforts for reducing SO₂ emissions since 2006 have been effective.

The control measures for NO₂ only began with the control of vehicular emissions in 1995, but the inclusion of NO₂ in the gross control indexes did not happen until 2010. New coal power plants were also required to denitrate after 2010. The emissions of NO_x actually increased from 1996 to 2010, as seen in the vertical column NO₂ derived from satellite data (Zhang et al., 2012b). Although annual average ambient NO₂ at the surface fluctuated from 30 to 40 μg m⁻³ during 2000–2010 in China (<http://www.zhb.gov.cn/>), annual average ambient NO₂ in megacities (e.g., Beijing, Shanghai and Guangzhou) slowly increased. Evidently, the control of emissions of nitrate gaseous precursors was not very effective during 2000–2010.

Despite the above-mentioned control measures, sulfate concentrations remained high and nitrate concentrations even gradually increased in megacities in China. More recently, the Clean Air Action Plan (CAAP) for improving air quality was promulgated and implemented by the State Council of the People's Republic of China in 2013 (<http://www.gov.cn/>). This plan aims to reduce the PM_{2.5} annual mass concentrations by 25, 20 and 15 % of the 2012 levels in BTH, YRD and PRD, respectively. Key industries, including power plants, iron and steel smelting, petroleum chemical, cement, non-ferrous metals smelting and chemical production, were required to implement stricter emission standards in key regions including most megacities in China (<http://www.zhb.gov.cn/>). Accordingly, annual average PM_{2.5} in China from online monitored data at 74 cities gradually decreased from 72 μg m⁻³ in 2013 to 50 μg m⁻³ in 2015, showing some promising results from the series of control measures.

One factor that needs to be considered in future pollution reduction is the nonlinearity of chemistry (Cheng et al., 2016). For example, a model sensitivity study suggested a potential increase in NO₃⁻ mass concentrations due to the increased atmospheric oxidizing capacity, even with decreasing NO_x emissions (Zhao et al., 2013a). Furthermore, increased atmospheric oxidizing capacity may also enhance the conversion of volatile organic compounds to OM. In fact, the contribution of secondary organic aerosols to PM_{2.5} was also high and could increase further in typical megacities in China (He et al., 2011; Huang et al., 2014b; Sun et al., 2013). Another factor that requires more attention is ammonia emissions from agricultural activities in rural areas and human activities in cities. Ammonia emissions can enhance PM_{2.5} pollution substantially, especially in ammonia-limited (acid aerosols) areas (Wang et al., 2011). This topic needs further investigation through both modeling simulation and field observations.

To improve the air quality across China, the following recommendations are provided based on the major chemi-

cal components contributing to PM_{2.5} and their impact on aerosol optical properties. Emissions produced from coal combustion, in both the industrial sectors and in residential areas, need to be further reduced. While advanced pollution control technologies should be adopted in the medium term in major industrial sectors consuming coal, cleaner energy sources should be considered a long-term goal (Cao et al., 2016). Providing cleaner energy to the vast rural and urban areas in north China for heating and cooking can reduce not only coal combustion emissions but also biomass burning emissions. Efficient use of fertilizers in agriculture is needed to reduce nitrogen emissions especially ammonia (Behera et al., 2013). Educating the public to reduce meat consumption in their daily lives, especially in the more affluent developed regions, can reduce the nitrogen footprint substantially and thus nitrogen emissions (Galloway et al., 2014), in addition to the potential benefits to human health. Traffic emissions in megacities may also need to be constrained, such as developing more efficient public transportation systems and limiting the use of personal automobiles. Having more vegetation coverage is especially important in arid or semi-arid areas as well as urban areas in reducing dust emissions (Baldauf, 2017), aside from the biological benefits. The continued expansion of the three northern region shelter forests in north China can potentially reduce dust emissions by increasing the dry deposition removal of aerosols (Zhang et al., 2017a).

Data availability. Data used in this study are from literature surveys and are available in the Supplement of this paper.

The Supplement related to this article is available online at <https://doi.org/10.5194/acp-17-9485-2017-supplement>.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This study was supported by the National Natural Science Foundation of China (no. 41475119).

Edited by: Yun Qian

Reviewed by: three anonymous referees

References

- Andreae, M. O., Schmid, O., Yang, H., Chand, D., Zhen Yu, J., Zeng, L. M., and Zhang, Y. H.: Optical properties and chemical composition of the atmospheric aerosol in urban Guangzhou, China, *Atmos. Environ.*, 42, 6335–6350, 2008.

- Baldauf, R.: Roadside vegetation design characteristics that can improve local, near-road air quality, *Transport. Res. D-Tr. E.*, 52A, 354–361, 2017.
- Behera, S. N., Sharma, M., Aneja, V. P., and Balasubramanian, R.: Ammonia in the atmosphere: A review on emission sources, atmospheric chemistry and deposition on terrestrial bodies, *Environ. Sci. Pollut. Research*, 20, 8092–8131, 2013.
- Bergin, M. H., Cass, G. R., Xu, J., Fang, C., Zeng, L., Yu, T., Salmon, L. G., Kiang, C. S., Tang, X. Y., and Zhang, Y.: Aerosol radiative, physical, and chemical properties in Beijing during June 1999, *J. Geophys. Res.*, 106, 17969–17980, 2001.
- Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, *J. Geophys. Res.*, 111, D20211, <https://doi.org/10.1029/2006JD007315>, 2006.
- Cao, J., Lee, S., Ho, K., Zou, S., Fung, K., Li, Y., Watson, J. G., and Chow, J. C.: Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, *Atmos. Environ.*, 38, 4447–4456, 2004.
- Cao, J., Zhu, C., Chow, J. C., Watson, J. G., Han, Y., Wang, G., Shen, Z., and An, Z.: Black carbon relationships with emissions and meteorology in Xi'an, China, *Atmos. Res.*, 94, 194–202, 2009.
- Cao, J., Wang, Q., Chow, J. C., Watson, J. G., Tie, X., Shen, Z., Wang, P., and An, Z.: Impacts of aerosol compositions on visibility impairment in Xi'an, China, *Atmos. Environ.*, 59, 559–566, 2012a.
- Cao, J., Shen, Z., Chow, J. C., Watson, J. G., Lee, S., Tie, X., Ho, K., Wang, G., and Han, Y.: Winter and summer PM_{2.5} chemical compositions in fourteen Chinese cities, *J. Air Waste Manage.*, 62, 1214–1226, 2012b.
- Cao, J., Cohen, A. M., Hansen, J. E., Lester, R. K., Peterson, P. F., and Xu, H.: China-U.S. cooperation to advance nuclear power, *Science*, 353, 547–548, 2016.
- Cao, J.-J., Zhu, C.-S., Tie, X.-X., Geng, F.-H., Xu, H.-M., Ho, S. S. H., Wang, G.-H., Han, Y.-M., and Ho, K.-F.: Characteristics and sources of carbonaceous aerosols from Shanghai, China, *Atmos. Chem. Phys.*, 13, 803–817, <https://doi.org/10.5194/acp-13-803-2013>, 2013.
- Chan, C. K. and Yao, X.: Air pollution in mega cities in China, *Atmos. Environ.*, 42, 1–42, 2008.
- Chan, C. Y., Xu, X., Li, Y. S., Wong, K. H., Ding, G. A., Chan, L. Y., and Cheng, X.: Characteristics of vertical profiles and sources of PM_{2.5}, PM₁₀ and carbonaceous species in Beijing, *Atmos. Environ.*, 39, 5113–5124, 2005.
- Che, H., Yang, Z., Zhang, X., Zhu, C., Ma, Q., Zhou, H., and Wang, P.: Study on the aerosol optical properties and their relationship with aerosol chemical compositions over three regional background stations in China, *Atmos. Environ.*, 43, 1093–1099, 2009.
- Che, H., Xia, X., Zhu, J., Li, Z., Dubovik, O., Holben, B., Goloub, P., Chen, H., Estelles, V., Cuevas-Agulló, E., Blarel, L., Wang, H., Zhao, H., Zhang, X., Wang, Y., Sun, J., Tao, R., Zhang, X., and Shi, G.: Column aerosol optical properties and aerosol radiative forcing during a serious haze-fog month over North China Plain in 2013 based on ground-based sun-photometer measurements, *Atmos. Chem. Phys.*, 14, 2125–2138, <https://doi.org/10.5194/acp-14-2125-2014>, 2014.
- Che, H., Zhang, X.-Y., Xia, X., Goloub, P., Holben, B., Zhao, H., Wang, Y., Zhang, X. C., Wang, H., Blarel, L., Damiri, B., Zhang, R., Deng, X., Ma, Y., Wang, T., Geng, F., Qi, B., Zhu, J., Yu, J., Chen, Q., and Shi, G.: Ground-based aerosol climatology of China: aerosol optical depths from the China Aerosol Remote Sensing Network (CARSNET) 2002–2013, *Atmos. Chem. Phys.*, 15, 7619–7652, <https://doi.org/10.5194/acp-15-7619-2015>, 2015.
- Chen, J., Qiu, S., Shang, J., Wilfrid, O. M. F., Liu, X., Tian, H., and Boman, J.: Impact of relative humidity and water soluble constituents of PM_{2.5} on visibility impairment in Beijing, China, *Aerosol Air Qual. Res.*, 14, 260–268, 2014a.
- Chen, J., Zhao, C. S., Ma, N., and Yan, P.: Aerosol hygroscopicity parameter derived from the light scattering enhancement factor measurements in the North China Plain, *Atmos. Chem. Phys.*, 14, 8105–8118, <https://doi.org/10.5194/acp-14-8105-2014>, 2014b.
- Chen, Y., Schleicher, N., Chen, Y., Chai, F., and Norra, S.: The influence of governmental mitigation measures on contamination characteristics of PM_{2.5} in Beijing, *Sci. Total Environ.*, 490, 647–658, <https://doi.org/10.1016/j.scitotenv.2014.05.049>, 2014a.
- Chen, Y., Xie, S., Luo, B., and Zhai, C.: Characteristics and origins of carbonaceous aerosol in the Sichuan Basin, China, *Atmos. Environ.*, 94, 215–223, <https://doi.org/10.1016/j.atmosenv.2014.05.037>, 2014b.
- Chen, Y., Xie, S., Luo, B., and Zhai, C.: Particulate pollution in urban Chongqing of southwest China: Historical trends of variation, chemical characteristics and source apportionment, *Sci. Total Environ.*, 584–585, 523–534, <https://doi.org/10.1016/j.scitotenv.2017.01.060>, 2017.
- Cheng, I., Xu, X., and Zhang, L.: Overview of receptor-based source apportionment studies for speciated atmospheric mercury, *Atmos. Chem. Phys.*, 15, 7877–7895, <https://doi.org/10.5194/acp-15-7877-2015>, 2015.
- Cheng, T., Xu, C., Duan, J., Wang, Y., Leng, C., Tao, J., Che, H., He, Q., Wu, Y., and Zhang, R.: Seasonal variation and difference of aerosol optical properties in columnar and surface atmospheres over Shanghai, *Atmos. Environ.*, 123, 315–326, 2015.
- Cheng, Y., Lee, S. C., Ho, K. F., Wang, Y. Q., Cao, J. J., Chow, J. C., and Watson, J. G.: Black carbon measurement in a coastal area of South China, *J. Geophys. Res.*, 111, 1–11, 2006a.
- Cheng, Y., Wiedensohler, A., Eichler, H., Su, H., Gnauk, T., Brüggemann, E., Herrmann, H., Heintzenberg, J., Slanina, J., and Tuch, T.: Aerosol optical properties and related chemical apportionment at Xinken in Pearl River Delta of China, *Atmos. Environ.*, 42, 6351–6372, 2008a.
- Cheng, Y., He, K.-B., Zheng, M., Duan, F.-K., Du, Z.-Y., Ma, Y.-L., Tan, J.-H., Yang, F.-M., Liu, J. M., Zhang, X. L., Weber, R. J., Bergin, M. H., and Russell, A. G.: Mass absorption efficiency of elemental carbon and water-soluble organic carbon in Beijing, China, *Atmos. Chem. Phys.*, 11, 11497–11510, <https://doi.org/10.5194/acp-11-11497-2011>, 2011.
- Cheng, Y., Engling, G., He, K.-B., Duan, F. K., Ma, Y. L., Du, Z. Y., Liu, J. M., Zheng, M., and Weber, R. J.: Biomass burning contribution to Beijing aerosol, *Atmos. Chem. Phys.*, 13, 7765–7781, <https://doi.org/10.5194/acp-13-7765-2013>, 2013a.
- Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Pöschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, *Sci. Adv.*, 2, e1601530, <https://doi.org/10.1126/sciadv.1601530>, 2016.

- Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H., Zeng, L. M., Liu, S., and Gnauk, T.: Mixing state of elemental carbon and non-light-absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, *J. Geophys. Res.*, 111, D20204, <https://doi.org/10.1029/2005JD006929>, 2006b.
- Cheng, Y. F., Wiedensohler, A., Eichler, H., Heintzenberg, J., Tesche, M., Ansmann, A., Wendisch, M., Su, H., Althausen, D., and Herrmann, H.: Relative humidity dependence of aerosol optical properties and direct radiative forcing in the surface boundary layer at Xinken in Pearl River Delta of China: An observation based numerical study, *Atmos. Environ.*, 42, 6373–6397, 2008b.
- Cheng, Y. F., Berghof, M., Garland, R. M., Wiedensohler, A., Wehner, B., Muller, T., Su, H., Zhang, Y., Achtert, P., and Nowak, A.: Influence of soot mixing state on aerosol light absorption and single scattering albedo during air mass aging at a polluted regional site in northeastern China, *J. Geophys. Res.*, 114, D00G10, <https://doi.org/10.1029/2008JD010883>, 2009.
- Cheng, Z., Wang, S., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Fu, X., and Hao, J.: Long-term trend of haze pollution and impact of particulate matter in the Yangtze River Delta, China, *Environ. Pollut.*, 182, 101–110, 2013b.
- Cheung, H. C., Wang, T., Baumann, K., and Guo, H.: Influence of regional pollution outflow on the concentrations of fine particulate matter and visibility in the coastal area of southern China, *Atmos. Environ.*, 39, 6463–6474, 2005.
- Chow, J. C., Watson, J. G., Lowenthal, D. H., and Magliano, K. L.: Loss of PM_{2.5} nitrate from filter samples in central California, *Journal of The Air & Waste Management Association*, 55, 1158–1168, 2005.
- Chow, J. C., Watson, J. G., Crow, D., Lowenthal, D. H., and Merrifield, T.: Comparison of IMPROVE and NIOSH carbon measurements, *Aerosol Sci. Tech.*, 34, 23–34, 2010.
- Cui, H., Chen, W., Dai, W., Liu, H., Wang, X., and He, K.: Source apportionment of PM_{2.5} in Guangzhou combining observation data analysis and chemical transport model simulation, *Atmos. Environ.*, 116, 262–271, <https://doi.org/10.1016/j.atmosenv.2015.06.054>, 2015.
- Dai, W., Gao, J., Cao, G., and Ouyang, F.: Chemical composition and source identification of PM_{2.5} in the suburb of Shenzhen, China, *Atmos. Res.*, 122, 391–400, <https://doi.org/10.1016/j.atmosres.2012.12.004>, 2013.
- Dan, M., Zhuang, G., Li, X., Tao, H., and Zhuang, Y.: The characteristics of carbonaceous species and their sources in PM_{2.5} in Beijing, *Atmos. Environ.*, 38, 3443–3452, 2004.
- Du, Z., He, K., Cheng, Y., Duan, F., Ma, Y., Liu, J., Zhang, X., Zheng, M., and Weber, R.: A yearlong study of water-soluble organic carbon in Beijing I: Sources and its primary vs. secondary nature, *Atmos. Environ.*, 92, 514–521, 2014a.
- Du, Z., He, K., Cheng, Y., Duan, F., Ma, Y., Liu, J., Zhang, X., Zheng, M., and Weber, R.: A yearlong study of water-soluble organic carbon in Beijing II: Light absorption properties, *Atmos. Environ.*, 89, 235–241, 2014b.
- Duan, F., Liu, X., Yu, T., and Cachier, H.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, *Atmos. Environ.*, 38, 1275–1282, 2004.
- Duan, F., He, K., Ma, Y., Yang, F., Yu, X., Cadle, S., Chan, T., and Mulawa, P.: Concentration and chemical characteristics of PM_{2.5} in Beijing, China: 2001–2002, *Sci. Total Environ.*, 355, 264–275, 2006.
- Duan, J., Tan, J., Cheng, D., Bi, X., Deng, W., Sheng, G., Fu, J., and Wong, M. H.: Sources and characteristics of carbonaceous aerosol in two largest cities in Pearl River Delta Region, China, *Atmos. Environ.*, 41, 2895–2903, 2007.
- Fan, X., Chen, H., Xia, X., Li, Z., and Cribb, M.: Aerosol optical properties from the Atmospheric Radiation Measurement Mobile Facility at Shouxian, China, *J. Geophys. Res.*, 115, D00K33, <https://doi.org/10.1029/2010JD014650>, 2010.
- Fang, M., Chan, C. K., and Yao, X.: Managing air quality in a rapidly developing nation: China, *Atmos. Environ.*, 43, 79–86, 2009.
- Feng, J., Chan, C. K., Fang, M., Hu, M., He, L., and Tang, X.: Characteristics of organic matter in PM_{2.5} in Shanghai, *Chemosphere*, 64, 1393–1400, 2006.
- Feng, J., Sun, P., Hu, X., Zhao, W., Wu, M., and Fu, J.: The chemical composition and sources of PM_{2.5} during the 2009 Chinese New Year's holiday in Shanghai, *Atmos. Res.*, 118, 435–444, <https://doi.org/10.1016/j.atmosres.2012.08.012>, 2012a.
- Feng, J., Zhong, M., Xu, B., Du, Y., Wu, M., Wang, H., and Chen, C.: Concentrations, seasonal and diurnal variations of black carbon in PM_{2.5} in Shanghai, China, *Atmos. Res.*, 147, 1–9, 2014.
- Feng, J., Hu, J., Xu, B., Hu, X., Sun, P., Han, W., Gu, Z., Yu, X., and Wu, M.: Characteristics and seasonal variation of organic matter in PM_{2.5} at a regional background site of the Yangtze River Delta region, China, *Atmos. Environ.*, 123, 288–297, <https://doi.org/10.1016/j.atmosenv.2015.08.019>, 2015.
- Feng, J. L., Guo, Z. G., Zhang, T. R., Yao, X., Chan, C. K., and Fang, M.: Source and formation of secondary particulate matter in PM_{2.5} in Asian continental outflow, *J. Geophys. Res.*, 117, D03302, <https://doi.org/10.1029/2011JD016400>, 2012b.
- Feng, Y., Chen, Y., Guo, H., Zhi, G., Xiong, S., Li, J., Sheng, G., and Fu, J.: Characteristics of organic and elemental carbon in PM_{2.5} samples in Shanghai, China, *Atmos. Res.*, 92, 434–442, 2009.
- Fontes, T., Li, P., Barros, N., and Zhao, P.: Trends of PM_{2.5} concentrations in China: A long term approach, *J. Environ. Manage.*, 196, 719–732, 2017.
- Fu, X., Wang, X., Guo, H., Cheung, K., Ding, X., Zhao, X., He, Q., Gao, B., Zhang, Z., Liu, T., and Zhang, Y.: Trends of ambient fine particles and major chemical components in the Pearl River Delta region: Observation at a regional background site in fall and winter, *Sci. Total Environ.*, 497–498, 274–281, <https://doi.org/10.1016/j.scitotenv.2014.08.008>, 2014.
- Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., Denier van der Gon, H., Facchini, M. C., Fowler, D., Koren, I., Langford, B., Lohmann, U., Nemitz, E., Pandis, S., Riipinen, I., Rudich, Y., Schaap, M., Slowik, J. G., Spracklen, D. V., Vignati, E., Wild, M., Williams, M., and Gilardoni, S.: Particulate matter, air quality and climate: lessons learned and future needs, *Atmos. Chem. Phys.*, 15, 8217–8299, <https://doi.org/10.5194/acp-15-8217-2015>, 2015.
- Gao, Y., Lai, S., Lee, S., Yau, P. S., Huang, Y., Cheng, Y., Wang, T., Xu, Z., Yuan, C., and Zhang, Y.: Optical properties of size-resolved particles at a Hong Kong urban site during winter, *Atmos. Res.*, 155, 1–12, 2015.

- Galloway, J. N., Winiwarter, W., Leip, A., Leach, A. M., Bleeker, A., and Erisman, J. W.: Nitrogen footprints: Past, present and future, *Environ. Res. Lett.*, 9, 115003, <https://doi.org/10.1088/1748-9326/9/11/115003>, 2014.
- Garland, R. M., Yang, H., Schmid, O., Rose, D., Nowak, A., Achtert, P., Wiedensohler, A., Takegawa, N., Kita, K., Miyazaki, Y., Kondo, Y., Hu, M., Shao, M., Zeng, L. M., Zhang, Y. H., Andreae, M. O., and Pöschl, U.: Aerosol optical properties in a rural environment near the mega-city Guangzhou, China: implications for regional air pollution, radiative forcing and remote sensing, *Atmos. Chem. Phys.*, 8, 5161–5186, <https://doi.org/10.5194/acp-8-5161-2008>, 2008.
- Garland, R. M., Schmid, O., Nowak, A., Achtert, P., Wiedensohler, A., Gunthe, S. S., Takegawa, N., Kita, K., Kondo, Y., and Hu, M.: Aerosol optical properties observed during Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Characteristic differences between the inflow and outflow of Beijing city air, *J. Geophys. Res.*, 114, D00G04, <https://doi.org/10.1029/2008JD010780>, 2009.
- Geng, N., Wang, J., Xu, Y., Zhang, W., Chen, C., and Zhang, R.: PM_{2.5} in an industrial district of Zhengzhou, China: Chemical composition and source apportionment, *Particuology*, 11, 99–109, 2013.
- Ginoux, P., Prospero, J. M., Gill, T. E., Hsu, N. C., and Zhao, M.: Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products, *Rev. Geophys.*, 50, RG3005, <https://doi.org/10.1029/2012RG000388>, 2012.
- Griffith, S. M., Huang, X. H., Louie, P. K. K., and Yu, J. Z.: Characterizing the thermodynamic and chemical composition factors controlling PM_{2.5} nitrate: Insights gained from two years of on-line measurements in Hong Kong, *Atmos. Environ.*, 122, 864–875, <https://doi.org/10.1016/j.atmosenv.2015.02.009>, 2015.
- Gu, J., Bai, Z., Liu, A., Wu, L., Xie, Y., Li, W., Dong, H., and Zhang, X.: Characterization of Atmospheric Organic Carbon and Element Carbon of PM_{2.5} and PM₁₀ at Tianjin, China, *Aerosol Air Qual. Res.*, 10, 167–176, 2010.
- Gu, J., Bai, Z., Li, W., Wu, L., Liu, A., Dong, H., and Xie, Y.: Chemical composition of PM_{2.5} during winter in Tianjin, China, *Particuology*, 9, 215–221, 2011.
- Guan, D., Su, X., Zhang, Q., Peters, G. P., Liu, Z., Lei, Y., and He, K.: The socioeconomic drivers of China's primary PM_{2.5} emissions, *Environ. Res. Lett.*, 9, 024010, <https://doi.org/10.1088/1748-9326/9/2/024010>, 2014.
- Guo, H., Ding, A. J., So, K. L., Ayoko, G. A., Li, Y., and Hung, W. T.: Receptor modeling of source apportionment of Hong Kong aerosols and the implication of urban and regional contribution, *Atmos. Environ.*, 43, 1159–1169, 2009a.
- Guo, J., Zhang, X., Che, H., Gong, S., An, X., Cao, C., Guang, J., Zhang, H., Wang, Y., and Zhang, X.: Correlation between PM concentrations and aerosol optical depth in eastern China, *Atmos. Environ.*, 43, 5876–5886, 2009b.
- 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, *Environ. Pollut.*, 221, 94–104, <https://doi.org/10.1016/j.envpol.2016.11.043>, 2017.
- Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., and Zeng, L.: Elucidating severe urban haze formation in China, *P. Natl. Acad. Sci. USA*, 111, 17373–17378, 2014.
- Hagler, G. S. W., Bergin, M. H., Salmon, L. G., Yu, J. Z., Wan, E. C. H., Zheng, M., Zeng, L. M., Kiang, C. S., Zhang, Y. H., Lau, A. K. H., and Schauer, J. J.: Source areas and chemical composition of fine particulate matter in the Pearl River Delta region of China, *Atmos. Environ.*, 40, 3802–3815, <https://doi.org/10.1016/j.atmosenv.2006.02.032>, 2006.
- Han, T., Liu, X., Zhang, Y., Qu, Y., Gu, J., Ma, Q., Lu, K., Tian, H., Chen, J., and Zeng, L.: Characteristics of aerosol optical properties and their chemical apportionments during CAREBeijing 2006, *Aerosol Air Qual. Res.*, 14, 1431–1442, 2014.
- Han, T., Qiao, L., Zhou, M., Qu, Y., Du, J., Liu, X., Lou, S., Chen, C., Wang, H., and Zhang, F.: Chemical and optical properties of aerosols and their interrelationship in winter in the megacity Shanghai of China, *J. Environ. Sci.*, 27, 59–69, 2015.
- Hand, J. L. and Malm, W. C.: Review of the IMPROVE equation for estimating ambient light extinction coefficients, CIRA, Colorado State University, 2007a.
- Hand, J. L. and Malm, W. C.: Review of aerosol mass scattering efficiencies from ground-based measurements since 1990, *J. Geophys. Res.*, 112, D16203, <https://doi.org/10.1029/2007JD008484>, 2007b.
- He, K., Yang, F., Ma, Y., Zhang, Q., Yao, X., Chan, C. K., Cadle, S., Chan, T., and Mulawa, P.: The characteristics of PM_{2.5} in Beijing, China, *Atmos. Environ.*, 35, 4959–4970, 2001.
- He, K., Zhao, Q., Ma, Y., Duan, F., Yang, F., Shi, Z., and Chen, G.: Spatial and seasonal variability of PM_{2.5} acidity at two Chinese megacities: insights into the formation of secondary inorganic aerosols, *Atmos. Chem. Phys.*, 12, 1377–1395, <https://doi.org/10.5194/acp-12-1377-2012>, 2012.
- He, L. Y., Hu, M., Zhang, Y. H., Huang, X. F., and Yao, T. T.: Fine particle emissions from on-road vehicles in the Zhujiang Tunnel, China, *Environ. Sci. Technol.*, 42, 4461–4466, 2008.
- He, L. Y., Huang, X. F., Xue, L., Hu, M., Lin, Y., Zheng, J., Zhang, R., and Zhang, Y. H.: Submicron aerosol analysis and organic source apportionment in an urban atmosphere in Pearl River Delta of China using high-resolution aerosol mass spectrometry, *J. Geophys. Res.-Atmos.*, 116, D12304, <https://doi.org/10.1029/2010JD014566>, 2011.
- He, X., Li, C. C., Lau, A. K. H., Deng, Z. Z., Mao, J. T., Wang, M. H., and Liu, X. Y.: An intensive study of aerosol optical properties in Beijing urban area, *Atmos. Chem. Phys.*, 9, 8903–8915, <https://doi.org/10.5194/acp-9-8903-2009>, 2009.
- Ho, K. F., Lee, S. C., Cao, J. J., Chow, J. C., Watson, J. G., and Chan, C. K.: Seasonal variations and mass closure analysis of particulate matter in Hong Kong, *Sci. Total Environ.*, 355, 276–287, 2006a.
- Ho, K. F., Cao, J. J., Lee, S. C., and Chan, C. K.: Source apportionment of PM_{2.5} in urban area of Hong Kong, *J. Hazard. Mater.*, 138, 73–85, 2006b.
- Hopke, P. K.: Review of receptor modeling methods for source apportionment, *J. Air Waste Manage.*, 66, 237–259, 2016.
- Hou, B., Zhuang, G., Zhang, R., Liu, T., Guo, Z., and Chen, Y.: The implication of carbonaceous aerosol to the formation of haze: Revealed from the characteristics and sources of OC/EC over a mega-city in China, *J. Hazard. Mater.*, 190, 529–536, <https://doi.org/10.1016/j.jhazmat.2011.03.072>, 2011.

- Hu, M., Wu, Z., Slanina, J., Lin, P., Liu, S., and Zeng, L.: Acidic gases, ammonia and water-soluble ions in PM_{2.5} at a coastal site in the Pearl River Delta, China, *Atmos. Environ.*, 42, 6310–6320, 2008.
- Hu, X., Waller, L. A., Alhamdan, M. Z., Crosson, W. L., Estes, M. G., Estes, S., Quattrocchi, D. A., Sarnat, J. A., and Liu, Y.: Estimating ground-level PM_{2.5} concentrations in the southeastern U.S. using geographically weighted regression, *Environ. Res.*, 121, 1–10, 2013.
- Huang, G., Cheng, T., Zhang, R., Tao, J., Leng, C., Zhang, Y., Zha, S., Zhang, D., Li, X., and Xu, C.: Optical properties and chemical composition of PM_{2.5} in Shanghai in the spring of 2012, *Particuology*, 13, 52–59, 2014a.
- Huang, H., Lee, S. C., Cao, J. J., Zou, C. W., Chen, X. G., and Fan, S. J.: Characteristics of indoor/outdoor PM_{2.5} and elemental components in generic urban, roadside and industrial plant areas of Guangzhou city, China, *J. Environ. Sci.*, 19, 35–43, 2007.
- Huang, H., Ho, K. F., Lee, S. C., Tsang, P. K., Ho, S. S. H., Zou, C. W., Zou, S. C., Cao, J. J., and Xu, H. M.: Characteristics of carbonaceous aerosol in PM_{2.5}: Pearl Delta River Region, China, *Atmos. Res.*, 104–105, 227–236, <https://doi.org/10.1016/j.atmosres.2011.10.016>, 2012.
- Huang, R. J., Zhang, Y., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., and Canonaco, F.: High secondary aerosol contribution to particulate pollution during haze events in China, *Nature*, 514, 218–222, 2014b.
- Huang, X., Yun, H., Gong, Z., Li, X., He, L., Zhang, Y., and Hu, M.: Source apportionment and secondary organic aerosol estimation of PM_{2.5} in an urban atmosphere in China, *Sci. China-Earth Sci.*, 57, 1352–1362, 2013.
- Huang, X. H. H., Bian, Q., Ng, W. M., Louie, P. K. K., and Yu, J. Z.: Characterization of PM_{2.5} major components and source Investigation in suburban Hong Kong: a one year monitoring study, *Aerosol Air Qual. Res.*, 14, 237–250, 2014c.
- Jahn, H. J., Kraemer, A., Chen, X. C., Chan, C. Y., Engling, G., and Ward, T. J.: Ambient and personal PM_{2.5} exposure assessment in the Chinese megacity of Guangzhou, *Atmos. Environ.*, 74, 402–411, 2013.
- Jing, J., Wu, Y., Tao, J., Che, H., Xia, X., Zhang, X., Yan, P., Zhao, D., and Zhang, L.: Observation and analysis of near-surface atmospheric aerosol optical properties in urban Beijing, *Particuology*, 18, 144–154, 2015.
- Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Gu, J., and Fan, S.: Aerosol chemistry and the effect of aerosol water content on visibility impairment and radiative forcing in Guangzhou during the 2006 Pearl River Delta campaign, *J. Environ. Manage.*, 90, 3231–3244, 2009.
- Kang, H., Zhu, B., Su, J., Wang, H., Zhang, Q., and Wang, F.: Analysis of a long-lasting haze episode in Nanjing, China, *Atmos. Res.*, 120, 78–87, 2013.
- Karagulian, F., Belis, C. A., Dora, C. F. C., Prüss-Ustün, A. M., Bonjour, S., Adair-Rohani, H., and Amann, M.: Contributions to cities' ambient particulate matter (PM): A systematic review of local source contributions at global level, *Atmos. Environ.*, 120, 475–483, 2015.
- Keck, L. and Wittmaack, K.: Effect of filter type and temperature on volatilisation losses from ammonium salts in aerosol matter, *Atmos. Environ.*, 39, 4093–4100, 2005.
- Kuang, Y., Zhao, C. S., Ma, N., Liu, H. J., Bian, Y., Tao, J. C., and Hu, M.: Deliquescent phenomena of ambient aerosols on the North China Plain, *Geophys. Res. Lett.*, 43, 8744–8750, <https://doi.org/10.1002/2016GL070273>, 2016.
- Lai, S., Zhao, Y., Ding, A., Zhang, Y., Song, T., Zheng, J., Ho, K. F., Lee, S., and Zhong, L.: Characterization of PM_{2.5} and the major chemical components during a 1-year campaign in rural Guangzhou, Southern China, *Atmos. Res.*, 167, 208–215, 2016.
- Lai, S. C., Zou, S. C., Cao, J. J., Lee, S. C., and Ho, K. F.: Characterizing ionic species in PM_{2.5} and PM₁₀ in four Pearl River Delta cities, South China, *J. Environ. Sci.*, 19, 939–947, 2007.
- Lan, Z., Huang, X., Yu, K., Sun, T., Zeng, L., and Hu, M.: Light absorption of black carbon aerosol and its enhancement by mixing state in an urban atmosphere in South China, *Atmos. Environ.*, 69, 118–123, 2013.
- Lee, J. H. and Hopke, P. K.: Apportioning sources of PM_{2.5} in St. Louis, MO using speciation trends network data, *Atmos. Environ.*, 40, S360–S377, 2006.
- Li, B., Zhang, J., Zhao, Y., Yuan, S., Zhao, Q., Shen, G., and Wu, H.: Seasonal variation of urban carbonaceous aerosols in a typical city Nanjing in Yangtze River Delta, China, *Atmos. Environ.*, 106, 223–231, <https://doi.org/10.1016/j.atmosenv.2015.01.064>, 2015a.
- Li, C., Tsay, S., Fu, J. S., Dickerson, R. R., Ji, Q., Bell, S. W., Gao, Y., Zhang, W., Huang, J., and Li, Z.: Anthropogenic air pollution observed near dust source regions in northwestern China during springtime 2008, *J. Geophys. Res.*, 115, D00K22, <https://doi.org/10.1029/2009JD013659>, 2010.
- Li, G., Bei, N., Cao, J., Huang, R., Wu, J., Feng, T., Wang, Y., Liu, S., Zhang, Q., Tie, X., and Molina, L. T.: A possible pathway for rapid growth of sulfate during haze days in China, *Atmos. Chem. Phys.*, 17, 3301–3316, <https://doi.org/10.5194/acp-17-3301-2017>, 2017a.
- Li, H., Wang, Q., Yang, M., Li, F., Wang, J., Sun, Y., Wang, C., Wu, H., and Qian, X.: Chemical characterization and source apportionment of PM_{2.5} aerosols in a megacity of Southeast China, *Atmos. Res.*, 181, 288–299, <https://doi.org/10.1016/j.atmosres.2016.07.005>, 2016a.
- Li, L., Chen, C. H., Fu, J. S., Huang, C., Streets, D. G., Huang, H. Y., Zhang, G. F., Wang, Y. J., Jang, C. J., Wang, H. L., Chen, Y. R., and Fu, J. M.: Air quality and emissions in the Yangtze River Delta, China, *Atmos. Chem. Phys.*, 11, 1621–1639, <https://doi.org/10.5194/acp-11-1621-2011>, 2011.
- Li, L., Chen, J., Wang, L., Melluki, W., and Zhou, H.: Aerosol single scattering albedo affected by chemical composition An investigation using CRDS combined with MARGA, *Atmos. Res.*, 124, 149–157, 2013a.
- Li, M. and Zhang, L.: Haze in China: Current and future challenges, *Environ. Pollut.*, 189, 85–86, 2014.
- Li, P., Yan, R., Yu, S., Wang, S., Liu, W., and Bao, H.: Reinstatement regional transport of PM_{2.5} as a major cause of severe haze in Beijing, *P. Natl. Acad. Sci. USA*, 112, E2739–E2740, <https://doi.org/10.1073/pnas.1502596112>, 2015b.
- Li, W., Bai, Z., Liu, A., Chen, J., and Chen, L.: Characteristics of Major PM_{2.5} Components during Winter in Tianjin, China, *Aerosol Air Qual. Res.*, 9, 105–119, 2009.
- Li, X., He, K., Li, C., Yang, F., Zhao, Q., Ma, Y., Cheng, Y., Ouyang, W., and Chen, G.: PM_{2.5} mass, chemical composition, and light

- extinction before and during the 2008 Beijing Olympics, *J. Geophys. Res.*, 118, 12158–12167, 2013b.
- Li, X., He, K., Li, C., Yang, F., Zhao, Q., Ma, Y., Cheng, Y., Ouyang, W., and Chen, G.: PM_{2.5} mass, chemical composition, and light extinction before and during the 2008 Beijing Olympics, *J. Geophys. Res.-Atmos.*, 118, 12158–12167, 2013c.
- Li, Y., Meng, J., Liu, J., Xu, Y., Guan, D., Tao, W., Huang, Y., and Tao, S.: Interprovincial Reliance for Improving Air Quality in China: A Case Study on Black Carbon Aerosol, *Environ. Sci. Technol.*, 50, 4118–4126, 2016b.
- Li, Y. J., Sun, Y., Zhang, Q., Li, X., Li, M., Zhou, Z., and Chan, C. K.: Real-time chemical characterization of atmospheric particulate matter in China: A review, *Atmos. Environ.*, 158, 270–304, <https://doi.org/10.1016/j.atmosenv.2017.02.027>, 2017b.
- Li, Z., Lau, W. K., Ramanathan, V., Wu, G., Ding, Y., Manoj, M. G., Liu, J., Qian, Y., Li, J., Zhou, T., Fan, J., Rosenfeld, D., Ming, Y., Wang, Y., Huang, J., Wang, B., Xu, X., Lee, S. S., Cribb, M., Zhang, F., Yang, X., Takemura, T., Wang, K., Xia, X., Yin, Y., Zhang, H., Guo, J., Zhai, P. M., Sugimoto, N., Babu, S. S., and Brasseur, G. P.: Aerosol and Monsoon Climate Interactions over Asia, *Rev. Geophys.*, 54, 866–929, <https://doi.org/10.1002/2015rg000500>, 2016c.
- Liang, C. S., Duan, F. K., He, K. B., and Ma, Y. L.: Review on recent progress in observations, source identifications and countermeasures of PM_{2.5}, *Environ. Int.*, 86, 150–170, <https://doi.org/10.1016/j.envint.2015.10.016>, 2016.
- Liao, H., Chang, W., and Yang, Y.: Climatic effects of air pollutants over China: A review, *Adv. Atmos. Sci.*, 32, 115–139, 2015.
- Lin, M., Biglari, S., Zhang, Z., Crocker, D., Tao, J., Su, B., Liu, L., and Thiemens, M. H.: Vertically uniform formation pathways of tropospheric sulfate aerosols in East China detected from triple stable oxygen and radiogenic sulfur isotopes, *Geophys. Res. Lett.*, 44, <https://doi.org/10.1002/2017GL073637>, 2017.
- Lin, Y., Hsu, S., Chou, C. C. K., Zhang, R., Wu, Y., Kao, S., Luo, L., Huang, C., Lin, S., and Huang, Y.: Wintertime haze deterioration in Beijing by industrial pollution deduced from trace metal fingerprints and enhanced health risk by heavy metals, *Environ. Pollut.*, 208, 284–293, 2016.
- Lin, Z. J., Zhang, Z. S., Zhang, L., Tao, J., Zhang, R. J., Cao, J. J., Fan, S. J., and Zhang, Y. H.: An alternative method for estimating hygroscopic growth factor of aerosol light-scattering coefficient: a case study in an urban area of Guangzhou, South China, *Atmos. Chem. Phys.*, 14, 7631–7644, <https://doi.org/10.5194/acp-14-7631-2014>, 2014.
- Liu, B., Song, N., Dai, Q., Mei, R., Sui, B., Bi, X., and Feng, Y.: Chemical composition and source apportionment of ambient PM_{2.5} during the non-heating period in Taian, China, *Atmos. Res.*, 170, 23–33, <https://doi.org/10.1016/j.atmosres.2015.11.002>, 2016a.
- Liu, B., Li, T., Yang, J., Wu, J., Wang, J., Gao, J., Bi, X., Feng, Y., Zhang, Y., and Yang, H.: Source apportionment and a novel approach of estimating regional contributions to ambient PM_{2.5} in Haikou, China, *Environ. Pollut.*, 223, 334–345, <https://doi.org/10.1016/j.envpol.2017.01.030>, 2017a.
- Liu, B., Wu, J., Zhang, J., Wang, L., Yang, J., Liang, D., Dai, Q., Bi, X., Feng, Y., Zhang, Y., and Zhang, Q.: Characterization and source apportionment of PM_{2.5} based on error estimation from EPA PMF 5.0 model at a medium city in China, *Environ. Pollut.*, 222, 10–22, <https://doi.org/10.1016/j.envpol.2017.01.005>, 2017b.
- Liu, D., Li, J., Zhang, Y., Xu, Y., Liu, X., Ding, P., Shen, C., Chen, Y., Tian, C., and Zhang, G.: The use of levoglucosan and radiocarbon for source apportionment of PM_{2.5} carbonaceous aerosols at a background site in east China, *Environ. Sci. Technol.*, 47, 10454, <https://doi.org/10.1021/es401250k>, 2013a.
- Liu, G., Li, J., Wu, D., and Xu, H.: Chemical composition and source apportionment of the ambient PM_{2.5} in Hangzhou, China, *Particuology*, 18, 135–143, <https://doi.org/10.1016/j.partic.2014.03.011>, 2015.
- Liu, J., Li, J., Zhang, Y., Liu, D., Ding, P., Shen, C., Shen, K., He, Q., Ding, X., and Wang, X.: Source apportionment using radiocarbon and organic tracers for PM_{2.5} carbonaceous aerosols in Guangzhou, south China: contrasting local- and regional-scale haze events, *Environ. Sci. Technol.*, 48, 12002–12011, 2014a.
- Liu, P. F., Zhao, C. S., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W. Y., Deng, Z. Z., Ma, N., Mildenerberger, K., Henning, S., Stratmann, F., and Wiedensohler, A.: Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain, *Atmos. Chem. Phys.*, 11, 3479–3494, <https://doi.org/10.5194/acp-11-3479-2011>, 2011.
- Liu, S., Hu, M., Slanina, S., He, L., Niu, Y., Bruegemann, E., Gnauk, T., and Herrmann, H.: Size distribution and source analysis of ionic compositions of aerosols in polluted periods at Xinken in Pearl River Delta (PRD) of China, *Atmos. Environ.*, 42, 6284–6295, 2008a.
- Liu, X., Chen, Q., Che, H., Zhang, R., Gui, K., Zhang, H., and Zhao, T.: Spatial distribution and temporal variation of aerosol optical depth in the Sichuan basin, China, the recent ten years, *Atmos. Environ.*, 147, 434–445, <https://doi.org/10.1016/j.atmosenv.2016.10.008>, 2016b.
- Liu, X., Cheng, Y., Zhang, Y., Jung, J., Sugimoto, N., Chang, S. Y., Kim, Y. J., Fan, S., and Zeng, L.: Influences of relative humidity and particle chemical composition on aerosol scattering properties during the 2006 PRD campaign, *Atmos. Environ.*, 42, 1525–1536, 2008b.
- Liu, X., Zhang, Y., Jung, J., Gu, J., Li, Y., Guo, S., Chang, S., Yue, D., Lin, P., and Kim, Y. J.: Research on the hygroscopic properties of aerosols by measurement and modeling during CAREBeijing-2006, *J. Geophys. Res.*, 114, D00G16, <https://doi.org/10.1029/2008JD010805>, 2009.
- Liu, X., Zhang, Y., Cheng, Y., Hu, M., and Han, T.: Aerosol hygroscopicity and its impact on atmospheric visibility and radiative forcing in Guangzhou during the 2006 PRIDE-PRD campaign, *Atmos. Environ.*, 60, 59–67, 2012.
- Liu, X., Gu, J., Li, Y., Cheng, Y., Qu, Y., Han, T., Wang, J., Tian, H., Chen, J., and Zhang, Y.: Increase of aerosol scattering by hygroscopic growth: Observation, modeling, and implications on visibility, *Atmos. Res.*, 132–133, 91–101, <https://doi.org/10.1016/j.atmosres.2013.04.007>, 2013b.
- Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., Gu, J., Chen, C., Yang, Y., Liu, X., Yang, T., Zhang, Y., Tian, H., and Hu, M.: Formation and evolution mechanism of regional haze: a case study in the megacity Beijing, China, *Atmos. Chem. Phys.*, 13, 4501–4514, <https://doi.org/10.5194/acp-13-4501-2013>, 2013c.
- Liu, Y., Sarnat, J. A., Kilaru, V., Jacob, D. J., and Koutrakis, P.: Estimating ground-level PM_{2.5} in the eastern United States using

- satellite remote sensing, *Environ. Sci. Technol.*, 39, 3269–3278, 2005.
- Liu, Z., Hu, B., Wang, L., Wu, F., Gao, W., and Wang, Y.: Seasonal and diurnal variation in particulate matter (PM₁₀ and PM_{2.5}) at an urban site of Beijing: analyses from a 9-year study, *Environ. Sci. Pollut. R.*, 22, 627–642, 2014b.
- Louie, P. K., Watson, J. G., Chow, J. C., Chen, A., Sin, D. W., and Lau, A. K.: Seasonal characteristics and regional transport of PM_{2.5} in Hong Kong, *Atmos. Environ.*, 39, 1695–1710, 2005a.
- Louie, P. K. K., Chow, J. C., Chen, L. W. A., Watson, J. G., Leung, G., and Sin, D. W. M.: PM_{2.5} chemical composition in Hong Kong: urban and regional variations, *Sci. Total Environ.*, 338, 267–281, <https://doi.org/10.1016/j.scitotenv.2004.07.021>, 2005b.
- Lu, Q., Zheng, J., Ye, S., Shen, X., Yuan, Z., and Yin, S.: Emission trends and source characteristics of SO₂, NO_x, PM₁₀ and VOCs in the Pearl River Delta region from 2000 to 2009, *Atmos. Environ.*, 76, 11–20, 2013.
- Lu, Z., Streets, D. G., Zhang, Q., Wang, S., Carmichael, G. R., Cheng, Y. F., Wei, C., Chin, M., Diehl, T., and Tan, Q.: Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, *Atmos. Chem. Phys.*, 10, 6311–6331, <https://doi.org/10.5194/acp-10-6311-2010>, 2010.
- Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, *Atmos. Chem. Phys.*, 11, 9839–9864, <https://doi.org/10.5194/acp-11-9839-2011>, 2011.
- Lv, B., Zhang, B., and Bai, Y.: A systematic analysis of PM_{2.5} in Beijing and its sources from 2000 to 2012, *Atmos. Environ.*, 124, 98–108, <https://doi.org/10.1016/j.atmosenv.2015.09.031>, 2016.
- Ma, N., Zhao, C. S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y. F., Deng, Z. Z., Liu, P. F., Xu, W. Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildnerberger, K., Henning, S., Yu, J., Chen, L. L., Zhou, X. J., Stratmann, F., and Wiedensohler, A.: Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study, *Atmos. Chem. Phys.*, 11, 5959–5973, <https://doi.org/10.5194/acp-11-5959-2011>, 2011.
- Ma, Z., Hu, X., Huang, L., Bi, J., and Liu, Y.: Estimating ground-level PM_{2.5} in China using satellite remote sensing, *Environ. Sci. Technol.*, 48, 7436–7444, 2014.
- Malm, W. C., Day, D. E., Kreidenweis, S. M., Collett, J. L., and Lee, T.: Humidity-dependent optical properties of fine particles during the Big Bend Regional Aerosol and Visibility Observational Study, *J. Geophys. Res.*, 108, 4279, <https://doi.org/10.1029/2002JD002998>, 2003.
- Man, C. K. and Shih, M. Y.: Light scattering and absorption properties of aerosol particles in Hong Kong, *J. Aerosol Sci.*, 32, 795–804, 2001.
- Ming, L., Jin, L., Li, J., Fu, P., Yang, W., Liu, D., Zhang, G., Wang, Z., and Li, X.: PM_{2.5} in the Yangtze River Delta, China: Chemical compositions, seasonal variations, and regional pollution events, *Environ. Pollut.*, 223, 200–212, <https://doi.org/10.1016/j.envpol.2017.01.013>, 2017.
- Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akiyama, H., Amann, M., Baklanov, A., Baltensperger, U., and Bey, I.: Atmospheric composition change – Global and regional air quality, *Atmos. Environ.*, 43, 5268–5350, 2009.
- Niu, F., Li, Z., Li, C., Lee, K., and Wang, M.: Increase of wintertime fog in China: Potential impacts of weakening of the Eastern Asian monsoon circulation and increasing aerosol loading, *J. Geophys. Res.*, 115, D00K20, <https://doi.org/10.1029/2009JD013484>, 2010.
- Okuda, T., Matsuura, S., Yamaguchi, D., Umemura, T., Hanada, E., Orihara, H., Tanaka, S., He, K., Ma, Y., and Cheng, Y.: The impact of the pollution control measures for the 2008 Beijing Olympic Games on the chemical composition of aerosols, *Atmos. Environ.*, 45, 2789–2794, 2011.
- Orsini, D. A., Ma, Y., Sullivan, A., Sierau, B., Baumann, K., and Weber, R. J.: Refinements to the particle-into-liquid sampler (PILS) for ground and airborne measurements of water soluble aerosol composition, *Atmos. Environ.*, 37, 1243–1259, [https://doi.org/10.1016/S1352-2310\(02\)01015-4](https://doi.org/10.1016/S1352-2310(02)01015-4), 2003.
- Pan, L., Che, H., Geng, F., Xia, X., Wang, Y., Zhu, C., Chen, M., Gao, W., and Guo, J.: Aerosol optical properties based on ground measurements over the Chinese Yangtze Delta Region, *Atmos. Environ.*, 44, 2587–2596, 2010.
- Pan, X. L., Yan, P., Tang, J., Ma, J. Z., Wang, Z. F., Gbaguidi, A., and Sun, Y. L.: Observational study of influence of aerosol hygroscopic growth on scattering coefficient over rural area near Beijing mega-city, *Atmos. Chem. Phys.*, 9, 7519–7530, <https://doi.org/10.5194/acp-9-7519-2009>, 2009.
- Pathak, R. K., Wang, T., Ho, K., and Lee, S.: Characteristics of summertime PM_{2.5} organic and elemental carbon in four major Chinese cities: implications of high acidity for water-soluble organic carbon (WSOC), *Atmos. Environ.*, 45, 318–325, 2011.
- Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., and Hand, J.: Revised algorithm for estimating light extinction from IMPROVE particle speciation data, *J. Air Waste Manage.*, 57, 1326–1336, 2007.
- Pu, W., Zhao, X., Shi, X., Ma, Z., Zhang, X., and Yu, B.: Impact of long-range transport on aerosol properties at a regional background station in Northern China, *Atmos. Res.*, 153, 489–499, 2015.
- Pui, D. Y., Chen, S.-C., and Zuo, Z.: PM_{2.5} in China: Measurements, sources, visibility and health effects, and mitigation, *Particulateology*, 13, 1–26, 2014.
- Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K. M., Ming, J., Wang, H., Wang, M., Warren, S. G., and Zhang, R.: Light-absorbing Particles in Snow and Ice: Measurement and Modeling of Climatic and Hydrological Impact, *Adv. Atmos. Sci.*, 32, 64–91, 2015.
- Qin, Y. and Xie, S. D.: Spatial and temporal variation of anthropogenic black carbon emissions in China for the period 1980–2009, *Atmos. Chem. Phys.*, 12, 4825–4841, <https://doi.org/10.5194/acp-12-4825-2012>, 2012.
- Schnaiter, M., Linke, C., Mohler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic aerosol, *J. Geophys. Res.*, 110, D19204, <https://doi.org/10.1029/2005JD006046>, 2005.
- Shen, G., Xue, M., Yuan, S., Zhang, J., Zhao, Q., Li, B., Wu, H., and Ding, A.: Chemical compositions and reconstructed light extinction coefficients of particulate matter in a mega-city in the western Yangtze River Delta, China, *Atmos. Environ.*, 83, 14–20, <https://doi.org/10.1016/j.atmosenv.2013.10.055>, 2014.
- Shen, Z., Cao, J., Arimoto, R., Han, Z., Zhang, R., Han, Y., Liu, S., Okuda, T., Nakao, S., and Tanaka, S.: Ionic composition of

- TSP and PM_{2.5} during dust storms and air pollution episodes at Xi'an, China, *Atmos. Environ.*, 43, 2911–2918, 2009.
- Song, Y., Xie, S., Zhang, Y., Zeng, L., Salmon, L. G., and Zheng, M.: Source apportionment of PM_{2.5} in Beijing using principal component analysis/absolute principal component scores and UNMIX, *Sci. Total Environ.*, 372, 278–286, 2006a.
- Song, Y., Zhang, Y., Xie, S., Zeng, L., Zheng, M., Salmon, L. G., Shao, M., and Slanina, S.: Source apportionment of PM_{2.5} in Beijing by positive matrix factorization, *Atmos. Environ.*, 40, 1526–1537, 2006b.
- Song, Y., Tang, X., Xie, S., Zhang, Y., Wei, Y., Zhang, M., Zeng, L., and Lu, S.: Source apportionment of PM_{2.5} in Beijing in 2004, *J. Hazard. Mater.*, 146, 124–130, 2007.
- Sun, Y., Zhuang, G., Wang, Y., Han, L., Guo, J., Dan, M., Zhang, W., Wang, Z., and Hao, Z.: The air-borne particulate pollution in Beijing – concentration, composition, distribution and sources, *Atmos. Environ.*, 38, 5991–6004, 2004.
- Sun, Y., Zhuang, G., Tang, A., Wang, Y., and An, Z.: Chemical Characteristics of PM_{2.5} and PM₁₀ in Haze-Fog Episodes in Beijing, *Environ. Sci. Technol.*, 40, 3148, <https://doi.org/10.1021/es051533g>, 2006.
- Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.: Aerosol composition, sources and processes during wintertime in Beijing, China, *Atmos. Chem. Phys.*, 13, 4577–4592, <https://doi.org/10.5194/acp-13-4577-2013>, 2013.
- Tan, J., Duan, J., He, K., Ma, Y., Duan, F., Chen, Y., and Fu, J.: Chemical characteristics of PM_{2.5} during a typical haze episode in Guangzhou, *J. Environ. Sci.*, 21, 774–781, 2009.
- Tan, J., Duan, J., Zhen, N., He, K., and Hao, J.: Chemical characteristics and source of size-fractionated atmospheric particle in haze episode in Beijing, *Atmos. Res.*, 167, 24–33, <https://doi.org/10.1016/j.atmosres.2015.06.015>, 2016a.
- Tan, J., Xiang, P., Zhou, X., Duan, J., Ma, Y., He, K., Cheng, Y., Yu, J., and Querol, X.: Chemical characterization of humic-like substances (HULIS) in PM_{2.5} in Lanzhou, China, *Sci. Total Environ.*, 573, 1481–1490, <https://doi.org/10.1016/j.scitotenv.2016.08.025>, 2016b.
- Tan, J., Duan, J., Ma, Y., He, K., Cheng, Y., Deng, S., Huang, Y., and Si-Tu, S.: Long-term trends of chemical characteristics and sources of fine particle in Foshan City, Pearl River Delta: 2008–2014, *Sci. Total Environ.*, 565, 519–528, <https://doi.org/10.1016/j.scitotenv.2016.05.059>, 2016c.
- Tan, J., Zhang, L., Zhou, X., Duan, J., Li, Y., Hu, J., and He, K.: Chemical characteristics and source apportionment of PM_{2.5} in Lanzhou, China, *Sci. Total Environ.*, 601–602, 1743–1752, 2017.
- Tang, X., Chen, X., and Tian, Y.: Chemical composition and source apportionment of PM_{2.5} – A case study from one year continuous sampling in the Chang-Zhu-Tan urban agglomeration, *Atmospheric Pollution Research*, 8, 885–899, <https://doi.org/10.1016/j.apr.2017.02.004>, 2017.
- Tao, J., Ho, K., Chen, L., Zhu, L., Han, J., and Xu, Z.: Effect of chemical composition of PM_{2.5} on visibility in Guangzhou, China, 2007 spring, *Particuology*, 7, 68–75, 2009.
- Tao, J., Cheng, T., Zhang, R., Cao, J., Zhu, L., Wang, Q., Luo, L., and Zhang, L.: Chemical composition of PM_{2.5} at an urban site of Chengdu in southwestern China, *Adv. Atmos. Sci.*, 30, 1070–1084, 2013a.
- Tao, J., Zhang, L., Engling, G., Zhang, R., Yang, Y., Cao, J., Zhu, C., Wang, Q., and Luo, L.: Chemical composition of PM_{2.5} in an urban environment in Chengdu, China: Importance of springtime dust storms and biomass burning, *Atmos. Res.*, 122, 270–283, <https://doi.org/10.1016/j.atmosres.2012.11.004>, 2013b.
- Tao, J., Gao, J., Zhang, L., Zhang, R., Che, H., Zhang, Z., Lin, Z., Jing, J., Cao, J., and Hsu, S.-C.: PM_{2.5} pollution in a megacity of southwest China: source apportionment and implication, *Atmos. Chem. Phys.*, 14, 8679–8699, <https://doi.org/10.5194/acp-14-8679-2014>, 2014a.
- Tao, J., Zhang, L., Cao, J., Hsu, S. C., Xia, X., Zhang, Z., Lin, Z., Cheng, T., and Zhang, R.: Characterization and source apportionment of aerosol light extinction in Chengdu, southwest China, *Atmos. Environ.*, 95, 552–562, <https://doi.org/10.1016/j.atmosenv.2014.07.017>, 2014b.
- Tao, J., Zhang, L., Ho, K., Zhang, R., Lin, Z., Zhang, Z., Lin, M., Cao, J., Liu, S., and Wang, G.: Impact of PM_{2.5} chemical compositions on aerosol light scattering in Guangzhou - the largest megacity in South China, *Atmos. Res.*, 135–136, 48–58, <https://doi.org/10.1016/j.atmosres.2013.08.015>, 2014c.
- Tao, J., Zhang, L., Gao, J., Wang, H., Chai, F., and Wang, S.: Aerosol chemical composition and light scattering during a winter season in Beijing, *Atmos. Environ.*, 110, 36–44, <https://doi.org/10.1016/j.atmosenv.2015.03.037>, 2015a.
- Tao, J., Zhang, L., Zhang, Z., Huang, R., Wu, Y., Zhang, R., Cao, J., and Zhang, Y.: Control of PM_{2.5} in Guangzhou during the 16th Asian Games period: Implication for haze weather prevention, *Sci. Total Environ.*, 508, 57–66, <https://doi.org/10.1016/j.scitotenv.2014.11.074>, 2015b.
- Tao, J., Gao, J., Zhang, L., Wang, H., Qiu, X., Zhang, Z., Wu, Y., Chai, F., and Wang, S.: Chemical and optical characteristics of atmospheric aerosols in Beijing during the Asia-Pacific Economic Cooperation China 2014, *Atmos. Environ.*, 144, 8–16, <https://doi.org/10.1016/j.atmosenv.2016.08.067>, 2016a.
- Tao, J., Zhang, L., Zhang, R., Wu, Y., Zhang, Z., Zhang, X., Tang, Y., Cao, J., and Zhang, Y.: Uncertainty assessment of source attribution of PM_{2.5} and its water-soluble organic carbon content using different biomass burning tracers in positive matrix factorization analysis – a case study in Beijing, China, *Sci. Total Environ.*, 543, 326–335, 2016b.
- Tao, J., Zhang, L., Cao, J., Zhong, L., Chen, D., Yang, Y., Chen, D., Chen, L., Zhang, Z., Wu, Y., Xia, Y., Ye, S., and Zhang, R.: Source apportionment of PM_{2.5} at urban and suburban areas of the Pearl River Delta region, south China – With emphasis on ship emissions, *Sci. Total Environ.*, 574, 1559–1570, <https://doi.org/10.1016/j.scitotenv.2016.08.175>, 2017.
- Tian, P., Wang, G., Zhang, R., Wu, Y., and Yan, P.: Impacts of aerosol chemical compositions on optical properties in urban Beijing, China, *Particuology*, 18, 155–164, <https://doi.org/10.1016/j.partic.2014.03.014>, 2015.
- Tian, Y. Z., Chen, G., Wang, H. T., Huang-Fu, Y. Q., Shi, G. L., Han, B., and Feng, Y. C.: Source regional contributions to PM_{2.5} in a megacity in China using an advanced source regional apportionment method, *Chemosphere*, 147, 256–263, <https://doi.org/10.1016/j.chemosphere.2015.12.132>, 2016.
- Trebs, I., Meixner, F. X., Slanina, J., Otjes, R., Jongejan, P., and Andreae, M. O.: Real-time measurements of ammonia, acidic trace gases and water-soluble inorganic aerosol species at a rural site in the Amazon Basin, *Atmos. Chem. Phys.*, 4, 967–987, <https://doi.org/10.5194/acp-4-967-2004>, 2004.

- Verma, R. L., Sahu, L. K., Kondo, Y., Takegawa, N., Han, S., Jung, J. S., Kim, Y. J., Fan, S., Sugimoto, N., Shammaa, M. H., Zhang, Y. H., and Zhao, Y.: Temporal variations of black carbon in Guangzhou, China, in summer 2006, *Atmos. Chem. Phys.*, 10, 6471–6485, <https://doi.org/10.5194/acp-10-6471-2010>, 2010.
- Wang, J. and Christopher, S. A.: Intercomparison between satellite-derived aerosol optical thickness and PM_{2.5} mass: Implications for air quality studies, *Geophys. Res. Lett.*, 30, 2095–2099, 2003.
- Wang, G., Zhang, R., Gomez, M. E., Yang, L., Zamora, M. L., Hu, M., Lin, Y., Peng, J., Guo, S., and Meng, J.: Persistent sulfate formation from London Fog to Chinese haze, *P. Natl. Acad. Sci.*, 113, 13630–13635, 2016a.
- Wang, H., Shi, G., Tian, M., Zhang, L., Chen, Y., Yang, F., and Cao, X.: Aerosol optical properties and chemical composition apportionment in Sichuan Basin, China, *Sci. Total Environ.*, 577, 245–257, <https://doi.org/10.1016/j.scitotenv.2016.10.173>, 2017a.
- Wang, H. L., Qiao, L. P., Lou, S. R., Zhou, M., Ding, A. J., Huang, H. Y., Chen, J. M., Wang, Q., Tao, S. K., Chen, C. H., Li, L., and Huang, C.: Chemical composition of PM_{2.5} and meteorological impact among three years in urban Shanghai, China, *J. Clean. Prod.*, 112, 1302–1311, <https://doi.org/10.1016/j.jclepro.2015.04.099>, 2016b.
- Wang, J., Li, X., Jiang, N., Zhang, W., Zhang, R., and Tang, X.: Long term observations of PM_{2.5}-associated PAHs: Comparisons between normal and episode days, *Atmos. Environ.*, 104, 228–236, <https://doi.org/10.1016/j.atmosenv.2015.01.026>, 2015a.
- Wang, L., Zhou, X., Ma, Y., Cao, Z., Wu, R., and Wang, W.: Carbonaceous aerosols over China—review of observations, emissions, and climate forcing, *Environ. Sci. Pollut. R.*, 23, 1671–1680, 2015b.
- Wang, P., Cao, J., Shen, Z., Han, Y., Lee, S., Huang, Y., Zhu, C., Wang, Q., Xu, H., and Huang, R.: Spatial and seasonal variations of PM_{2.5} mass and species during 2010 in Xi'an, China, *Sci. Total Environ.*, 508, 477–487, <https://doi.org/10.1016/j.scitotenv.2014.11.007>, 2015c.
- Wang, Q., Shao, M., Liu, Y., William, K., Paul, G., Li, X., Liu, Y., and Lu, S.: Impact of biomass burning on urban air quality estimated by organic tracers: Guangzhou and Beijing as cases, *Atmos. Environ.*, 41, 8380–8390, 2007.
- Wang, Q., Huang, R. J., Cao, J., Han, Y., Wang, G., Li, G., Wang, Y., Dai, W., Zhang, R., and Zhou, Y.: Mixing state of black carbon aerosol in a heavily polluted urban area of China: Implications for light absorption enhancement, *Aerosol Sci. Tech.*, 48, 689–697, <https://doi.org/10.1080/02786826.2014.917758>, 2014a.
- Wang, Q., Jiang, N., Yin, S., Li, X., Yu, F., Guo, Y., and Zhang, R.: Carbonaceous species in PM_{2.5} and PM₁₀ in urban area of Zhengzhou in China: Seasonal variations and source apportionment, *Atmos. Res.*, 191, 1–11, <https://doi.org/10.1016/j.atmosres.2017.02.003>, 2017b.
- Wang, S. and Hao, J.: Air quality management in China: Issues, challenges, and options, *J. Environ. Sci.*, 24, 2–13, 2012.
- Wang, S., Xing, J., Jang, C., Zhu, Y., Fu, J. S., and Hao, J.: Impact assessment of ammonia emissions on inorganic aerosols in East China using response surface modeling technique, *Environ. Sci. Technol.*, 45, 9293–9300, 2011.
- Wang, X., Ding, X., Fu, X., He, Q., Wang, S., Bernard, F., Zhao, X., and Wu, D.: Aerosol scattering coefficients and major chemical compositions of fine particles observed at a rural site in the central Pearl River Delta, south China, *J. Environ. Sci.*, 24, 72–77, 2012.
- Wang, Y., Jia, C., Tao, J., Zhang, L., Liang, X., Ma, J., Gao, H., Huang, T., and Zhang, K.: Chemical characterization and source apportionment of PM_{2.5} in a semi-arid and petrochemical – industrialized city, Northwest China, *Sci. Total Environ.*, 573, 1031–1040, <https://doi.org/10.1016/j.scitotenv.2016.08.179>, 2016c.
- Wang, Y., Zhuang, G., Tang, A., Yuan, H., Sun, Y., Chen, S., and Zheng, A.: The ion chemistry and the source of PM_{2.5} aerosol in Beijing, *Atmos. Environ.*, 39, 3771–3784, 2005.
- Wang, Y., Zhuang, G., Zhang, X. Y., Huang, K., Xu, C., Tang, A. H., Chen, J. M., and An, Z.: The ion chemistry, seasonal cycle, and sources of PM_{2.5} and TSP aerosol in Shanghai, *Atmos. Environ.*, 40, 2935–2952, 2006.
- Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, *Sci. China-Earth Sci.*, 57, 14–25, 2014b.
- Wang, Y., Ying, Q., Hu, J., and Zhang, H.: Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014, *Environ. Int.*, 73, 413–422, <https://doi.org/10.1016/j.envint.2014.08.016>, 2014c.
- Wang, Z., Li, J., Wang, Z., Yang, W., Tang, X., Ge, B., Yan, P., Zhu, L., Chen, X., and Chen, H.: Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control, *Sci. China-Earth Sci.*, 57, 3–13, 2013.
- Watson, J. G.: Visibility: Science and regulation, *J. Air Waste Manage.*, 52, 628–713, 2002.
- Watson, J. G., Chow, J. C., and Houck, J. E.: PM_{2.5} chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995, *Chemosphere*, 43, 1141–1151, 2001.
- Weber, R., Orsini, D., Duan, Y., Baumann, K., Kiang, C. S., Chameides, W. L., Lee, Y. N., Brechtel, F. J., Klotz, P. J., and Jongejan, P.: Intercomparison of near real time monitors of PM_{2.5} nitrate and sulfate at the U.S. Environmental Protection Agency Atlanta Supersite, *J. Geophys. Res.*, 108, 8421, <https://doi.org/10.1029/2001JD001220>, 2003.
- Wu, C., Ng, W. M., Huang, J., Wu, D., and Yu, J. Z.: Determination of Elemental and Organic Carbon in PM_{2.5} in the Pearl River Delta Region: Inter-Instrument (Sunset vs. DRI Model 2001 Thermal/Optical Carbon Analyzer) and Inter-Protocol Comparisons (IMPROVE vs. ACE-Asia Protocol), *Aerosol Sci. Tech.*, 46, 610–621, 2011.
- Wu, D., Mao, J., Deng, X., Tie, X., Zhang, Y., Zeng, L., Li, F., Tan, H., Bi, X., and Huang, X.: Black carbon aerosols and their radiative properties in the Pearl River Delta region, *Sci. China Ser. D*, 52, 1152–1163, 2009.
- Wu, D., Wu, C., Liao, B., Chen, H., Wu, M., Li, F., Tan, H., Deng, T., Li, H., Jiang, D., and Yu, J. Z.: Black carbon over the South China Sea and in various continental locations in South China, *Atmos. Chem. Phys.*, 13, 12257–12270, <https://doi.org/10.5194/acp-13-12257-2013>, 2013.
- Wu, S., Deng, F., Wei, H., Huang, J., Wang, X., Hao, Y., Zheng, C., Qin, Y., Lv, H., Shima, M., and Guo, X.: Association of Cardiopulmonary Health Effects with Source-Appointed Ambient Fine Particulate in Beijing, China: A Com-

- bined Analysis from the Healthy Volunteer Natural Relocation (HVNR) Study, *Environ. Sci. Technol.*, 48, 3438–3448, <https://doi.org/10.1021/es404778w>, 2014.
- Wu, Y., Zhang, R., Tian, P., Tao, J., Hsu, S. C., Yan, P., Wang, Q., Cao, J., Zhang, X., and Xia, X.: Effect of ambient humidity on the light absorption amplification of black carbon in Beijing during January 2013, *Atmos. Environ.*, 124, 217–223, 2016.
- Wu, Y., Wang, X., Yan, P., Zhang, L., Tao, J., Liu, X., Tian, P., Han, Z., and Zhang, R.: Investigation of hygroscopic growth effect on aerosol scattering coefficient at a rural site in the southern North China Plain, *Sci. Total Environ.*, 599–600, 76–84, <https://doi.org/10.1016/j.scitotenv.2017.04.194>, 2017.
- Xie, Y., Ding, A., Nie, W., Mao, H., Qi, X., Huang, X., Xu, Z., Kerminen, V. M., Petaja, T., and Chi, X.: Enhanced sulfate formation by nitrogen dioxide: Implications from in situ observations at the SORPES station, *J. Geophys. Res.*, 120, 12679–12694, 2015a.
- Xie, Y., Wang, Y., Zhang, K., Dong, W., Lv, B., and Bai, Y.: Daily Estimation of Ground-Level PM_{2.5} Concentrations over Beijing Using 3 km Resolution MODIS AOD, *Environ. Sci. Technol.*, 49, 12280–12288, 2015b.
- Xiong, Y., Zhou, J., Schauer, J. J., Yu, W., and Hu, Y.: Seasonal and spatial differences in source contributions to PM_{2.5} in Wuhan, China, *Sci. Total Environ.*, 577, 155–165, <https://doi.org/10.1016/j.scitotenv.2016.10.150>, 2017.
- Xu, H., Cao, J., Chow, J. C., Huang, R. J., Shen, Z., Chen, L. W. A., Ho, K. F., and Watson, J. G.: Inter-annual variability of wintertime PM_{2.5} chemical composition in Xi'an, China: Evidences of changing source emissions, *Sci. Total Environ.*, 545, 546–555, 2016.
- Xu, J., Bergin, M., Yu, X., Liu, G., Zhao, J., Carrico, C., and Baumann, K.: Measurement of aerosol chemical, physical and radiative properties in the Yangtze delta region of China, *Atmos. Environ.*, 36, 161–173, 2002.
- Xu, J., Bergin, M. H., Greenwald, R., Schauer, J. J., Shafer, M. M., Jaffrezou, J. L., and Aymoz, G.: Aerosol chemical, physical, and radiative characteristics near a desert source region of northwest China during ACE-Asia, *J. Geophys. Res.*, 109, D19S03, <https://doi.org/10.1029/2003JD004239>, 2004.
- Xu, J., Tao, J., Zhang, R., Cheng, T., Leng, C., Chen, J., Huang, G., Li, X., and Zhu, Z.: Measurements of surface aerosol optical properties in winter of Shanghai, *Atmos. Res.*, 109, 25–35, 2012a.
- Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical compositions of PM_{2.5} aerosol in the urban area of Fuzhou, China, *Atmos. Res.*, 104–105, 264–272, <https://doi.org/10.1016/j.atmosres.2011.10.017>, 2012b.
- Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical compositions of PM_{2.5} aerosol in the urban area of Fuzhou, China, *Atmos. Res.*, 104, 264–272, 2012c.
- Xu, W. Y., Zhao, C. S., Ran, L., Deng, Z. Z., Liu, P. F., Ma, N., Lin, W. L., Xu, X. B., Yan, P., He, X., Yu, J., Liang, W. D., and Chen, L. L.: Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain, *Atmos. Chem. Phys.*, 11, 4353–4369, <https://doi.org/10.5194/acp-11-4353-2011>, 2011.
- Yan, H.: Aerosol scattering properties in northern China, *Atmos. Environ.*, 41, 6916–6922, 2007.
- Yan, P., Tang, J., Huang, J., Mao, J. T., Zhou, X. J., Liu, Q., Wang, Z. F., and Zhou, H. G.: The measurement of aerosol optical properties at a rural site in Northern China, *Atmos. Chem. Phys.*, 8, 2229–2242, <https://doi.org/10.5194/acp-8-2229-2008>, 2008.
- Yan, P., Pan, X., Tang, J., Zhou, X., Zhang, R., and Zeng, L.: Hygroscopic growth of aerosol scattering coefficient: A comparative analysis between urban and suburban sites at winter in Beijing, *Particuology*, 7, 52–60, <https://doi.org/10.1016/j.partic.2008.11.009>, 2009.
- Yang, F., He, K., Ye, B., Chen, X., Cha, L., Cadle, S. H., Chan, T., and Mulawa, P. A.: One-year record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai, *Atmos. Chem. Phys.*, 5, 1449–1457, <https://doi.org/10.5194/acp-5-1449-2005>, 2005a.
- Yang, F., Huang, L., Duan, F., Zhang, W., He, K., Ma, Y., Brook, J. R., Tan, J., Zhao, Q., and Cheng, Y.: Carbonaceous species in PM_{2.5} at a pair of rural/urban sites in Beijing, 2005–2008, *Atmospheric Chemistry and Physics*, 11, 7893–7903, <https://doi.org/10.5194/acp-11-7893-2011>, 2011a.
- Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., Chen, G., and Zhao, Q.: Characteristics of PM_{2.5} speciation in representative megacities and across China, *Atmos. Chem. Phys.*, 11, 5207–5219, <https://doi.org/10.5194/acp-11-5207-2011>, 2011b.
- Yang, H., Yu, J. Z., Ho, S. S. H., Xu, J., Wu, W., Wan, C. H., Wang, X., Wang, X., and Wang, L.: The chemical composition of inorganic and carbonaceous materials in PM_{2.5} in Nanjing, China, *Atmos. Environ.*, 39, 3735–3749, 2005b.
- Yang, H., Chen, J., Wen, J., Tian, H., and Liu, X.: Composition and sources of PM_{2.5} around the heating periods of 2013 and 2014 in Beijing: Implications for efficient mitigation measures, *Atmos. Environ.*, 124, 378–386, <https://doi.org/10.1016/j.atmosenv.2015.05.015>, 2016.
- Yang, L., Zhou, X., Wang, Z., Zhou, Y., Cheng, S., Xu, P., Gao, X., Nie, W., Wang, X., and Wang, W.: Airborne fine particulate pollution in Jinan, China: concentrations, chemical compositions and influence on visibility impairment, *Atmos. Environ.*, 55, 506–514, 2012.
- Yang, L., Cheng, S., Wang, X., Nie, W., Xu, P., Gao, X., Yuan, C., and Wang, W.: Source identification and health impact of PM_{2.5} in a heavily polluted urban atmosphere in China, *Atmos. Environ.*, 75, 265–269, <https://doi.org/10.1016/j.atmosenv.2013.04.058>, 2013.
- Yang, Y. R., Liu, X. G., Qu, Y., An, J. L., Jiang, R., Zhang, Y. H., Sun, Y. L., Wu, Z. J., Zhang, F., Xu, W. Q., and Ma, Q. X.: Characteristics and formation mechanism of continuous hazes in China: a case study during the autumn of 2014 in the North China Plain, *Atmos. Chem. Phys.*, 15, 8165–8178, <https://doi.org/10.5194/acp-15-8165-2015>, 2015.
- Yao, L., Yang, L., Yuan, Q., Yan, C., Dong, C., Meng, C., Sui, X., Yang, F., Lu, Y., and Wang, W.: Sources apportionment of PM_{2.5} in a background site in the North China Plain, *Sci. Total Environ.*, 541, 590–598, <https://doi.org/10.1016/j.scitotenv.2015.09.123>, 2016.
- Ye, B., Ji, X., Yang, H., Yao, X., Chan, C. K., Cadle, S. H., Chan, T., and Mulawa, P. A.: Concentration and chemical composition of PM_{2.5} in Shanghai for a 1-year period, *Atmos. Environ.*, 37, 499–510, 2003.
- Yu, H., Wu, C., Wu, D., and Yu, J. Z.: Size distributions of elemental carbon and its contribution to light extinction in urban

- and rural locations in the pearl river delta region, China, *Atmos. Chem. Phys.*, 10, 5107–5119, <https://doi.org/10.5194/acp-10-5107-2010>, 2010.
- Zha, S., Cheng, T., Tao, J., Zhang, R., Chen, J., Zhang, Y., Leng, C., Zhang, D., and Du, J.: Characteristics and relevant remote sources of black carbon aerosol in Shanghai, *Atmos. Res.*, 135, 159–171, 2014.
- Zhang, F., Zhao, J., Chen, J., Xu, Y., and Xu, L.: Pollution characteristics of organic and elemental carbon in PM_{2.5} in Xiamen, China, *J. Environ. Sci.*, 23, 1342–1349, 2011a.
- Zhang, F., Xu, L., Chen, J., Yu, Y., Niu, Z., and Yin, L.: Chemical compositions and extinction coefficients of PM_{2.5} in peri-urban of Xiamen, China, during June 2009–May 2010, *Atmos. Res.*, 106, 150–158, 2012a.
- Zhang, F., Wang, Z., Cheng, H., Lv, X., Gong, W., Wang, X., and Zhang, G.: Seasonal variations and chemical characteristics of PM_{2.5} in Wuhan, central China, *Sci. Total Environ.*, 518–519, 97–105, <https://doi.org/10.1016/j.scitotenv.2015.02.054>, 2015a.
- Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, *Atmos. Chem. Phys.*, 15, 8439–8454, <https://doi.org/10.5194/acp-15-8439-2015>, 2015b.
- Zhang, N., Zhuang, M., Tian, J., Tian, P., Zhang, J., Wang, Q., Zhou, Y., Huang, R., Zhu, C., Zhang, X., and Cao, J.: Development of source profiles and their application in source apportionment of PM_{2.5} in Xiamen, China, *Front. Environ. Sci. Eng.*, 10, 17, <https://doi.org/10.1007/s11783-016-0879-1>, 2016.
- Zhang, Q., Geng, G., Wang, S., Richter, A., and He, K.: Satellite remote sensing of changes in NO_x emissions over China during 1996–2010, *Chinese Sci. Bull.*, 57, 2857–2864, <https://doi.org/10.1007/s11434-012-5015-4>, 2012b.
- Zhang, R., Jing, J., Tao, J., Hsu, S.-C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM_{2.5} in Beijing: seasonal perspective, *Atmos. Chem. Phys.*, 13, 7053–7074, <https://doi.org/10.5194/acp-13-7053-2013>, 2013a.
- Zhang, R., Li, Q., and Zhang, R.: Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013, *Sci. China-Earth Sci.*, 57, 26–35, <https://doi.org/10.1007/s11430-013-4774-3>, 2014a.
- Zhang, T., Cao, J., Tie, X., Shen, Z., Liu, S., Ding, H., Han, Y., Wang, G., Ho, K., and Qiang, J.: Water-soluble ions in atmospheric aerosols measured in Xi'an, China: seasonal variations and sources, *Atmos. Res.*, 102, 110–119, 2011b.
- Zhang, T., Cao, J., Chow, J. C., Shen, Z., Ho, K., Ho, S. S. H., Liu, S., Han, Y., Watson, J. G., and Wang, G.: Characterization and seasonal variations of levoglucosan in fine particulate matter in Xi'an, China, *J. Air Waste Manage.*, 64, 1317–1327, 2014b.
- Zhang, W., Guo, J., Sun, Y., Yuan, H., Zhuang, G., Zhuang, Y., and Hao, Z.: Source apportionment for urban PM₁₀ and PM_{2.5} in the Beijing area, *Chinese Sci. Bull.*, 52, 608–615, <https://doi.org/10.1007/s11434-007-0076-5>, 2007.
- Zhang, X., Wu, L., Zhang, R., Deng, S., Zhang, Y., Wu, J., Li, Y., Lin, L., Li, L., Wang, Y., and Wang, L.: Evaluating the relationships among economic growth, energy consumption, air emissions and air environmental protection investment in China, *Renewable and Sustainable Energy Reviews*, 18, 259–270, <https://doi.org/10.1016/j.rser.2012.10.029>, 2013b.
- Zhang, X., Du, J., Huang, T., Zhang, L., Gao, H., Zhao, Y., and Ma, J.: Atmospheric removal of PM_{2.5} by man-made three northern regions shelter forest in northern China estimated using satellite retrieved PM_{2.5} concentrations, *Sci. Total Environ.*, 593–594, 713–721, 2017a.
- Zhang, Y., Cai, J., Wang, S., He, K., and Zheng, M.: Review of receptor-based source apportionment research of fine particulate matter and its challenges in China, *Sci. Total Environ.*, 586, 917–929, <https://doi.org/10.1016/j.scitotenv.2017.02.071>, 2017b.
- Zhao, B., Wang, S., Wang, J., Fu, J. S., Liu, T., Xu, J., Fu, X., and Hao, J.: Impact of national NO_x and SO₂ control policies on particulate matter pollution in China, *Atmos. Environ.*, 77, 453–463, 2013a.
- Zhao, B., Wang, S. X., Liu, H., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NO_x emissions in China: historical trends and future perspectives, *Atmos. Chem. Phys.*, 13, 9869–9897, <https://doi.org/10.5194/acp-13-9869-2013>, 2013b.
- Zhao, M., Huang, Z., Qiao, T., Zhang, Y., Xiu, G., and Yu, J.: Chemical characterization, the transport pathways and potential sources of PM_{2.5} in Shanghai: Seasonal variations, *Atmos. Res.*, 158, 66–78, 2015a.
- Zhao, M., Qiao, T., Huang, Z., Zhu, M., Xu, W., Xiu, G., Tao, J., and Lee, S.: Comparison of ionic and carbonaceous compositions of PM_{2.5} in 2009 and 2012 in Shanghai, China, *Sci. Total Environ.*, 536, 695–703, <https://doi.org/10.1016/j.scitotenv.2015.07.100>, 2015b.
- Zhao, P. S., Dong, F., He, D., Zhao, X. J., Zhang, X. L., Zhang, W. Z., Yao, Q., and Liu, H. Y.: Characteristics of concentrations and chemical compositions for PM_{2.5} in the region of Beijing, Tianjin, and Hebei, China, *Atmos. Chem. Phys.*, 13, 4631–4644, <https://doi.org/10.5194/acp-13-4631-2013>, 2013c.
- Zhao, X., Zhang, X., Pu, W., Meng, W., and Xu, X.: Scattering properties of the atmospheric aerosol in Beijing, China, *Atmos. Res.*, 101, 799–808, 2011.
- Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., and Shi, Q. F.: Analysis of a winter regional haze event and its formation mechanism in the North China Plain, *Atmos. Chem. Phys.*, 13, 5685–5696, <https://doi.org/10.5194/acp-13-5685-2013>, 2013d.
- Zheng, J., Hu, M., Peng, J., Wu, Z., Kumar, P., Li, M., Wang, Y., and Guo, S.: Spatial distributions and chemical properties of PM_{2.5} based on 21 field campaigns at 17 sites in China, *Chemosphere*, 159, 480–487, 2016.
- Zheng, J. Y., Yin, S. S., Kang, D. W., Che, W. W., and Zhong, L. J.: Development and uncertainty analysis of a high-resolution NH₃ emissions inventory and its implications with precipitation over the Pearl River Delta region, China, *Atmos. Chem. Phys.*, 12, 7041–7058, <https://doi.org/10.5194/acp-12-7041-2012>, 2012.
- Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., and Cass, G. R.: Seasonal trends in PM_{2.5} source contributions in Beijing, China, *Atmos. Environ.*, 39, 3967–3976, 2005.
- Zhou, B., Shen, H., Huang, Y., Li, W., Chen, H., Zhang, Y., Su, S., Chen, Y., Lin, N., Zhuo, S., Zhong, Q., Liu, J., Li, B., and Tao, S.: Daily variations of size-segregated ambient particulate matter in Beijing, *Environ. Pollut.*, 197, 36–42, <https://doi.org/10.1016/j.envpol.2014.11.029>, 2015a.

- Zhou, Y., Shuiyuan, C., Lang, J., Chen, D., Zhao, B., Liu, C., Xu, R., and Li, T.: A comprehensive ammonia emission inventory with high-resolution and its evaluation in the Beijing–Tianjin–Hebei (BTH) region, China, *Atmos. Environ.*, 106, 305–317, <https://doi.org/10.1016/j.atmosenv.2015.01.069>, 2015b.
- Zhu, C., Cao, J., Ho, K., Chen, L. W. A., Huang, R., Wang, Y., Li, H., Shen, Z., Chow, J. C., and Watson, J. G.: The optical properties of urban aerosol in northern China: A case study at Xi'an, *Atmos. Res.*, 160, 59–67, 2015.
- Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, *Atmos. Chem. Phys.*, 13, 10609–10631, <https://doi.org/10.5194/acp-13-10609-2013>, 2013.
- Zíková, N., Wang, Y., Yang, F., Li, X., Tian, M., and Hopke, P. K.: On the source contribution to Beijing PM_{2.5} concentrations, *Atmos. Environ.*, 134, 84–95, <https://doi.org/10.1016/j.atmosenv.2016.03.047>, 2016.
- Zong, Z., Wang, X., Tian, C., Chen, Y., Qu, L., Ji, L., Zhi, G., Li, J., and Zhang, G.: Source apportionment of PM_{2.5} at a regional background site in North China using PMF linked with radiocarbon analysis: insight into the contribution of biomass burning, *Atmos. Chem. Phys.*, 16, 11249–11265, <https://doi.org/10.5194/acp-16-11249-2016>, 2016.
- Zou, Y., Wang, Y., Zhang, Y., and Koo, J. H.: Arctic sea ice, Eurasia snow, and extreme winter haze in China, *Sci. Adv.*, 3, e1602751, <https://doi.org/10.1126/sciadv.1602751>, 2017.