Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

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

Jun Tao[1], Leiming Zhang[2,*], Junji Cao[3], Renjian Zhang[4]

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

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

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

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

*Corresponds to: leiming.zhang@canada.ca

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

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 of 2000 are reviewed and summarized in terms of geographical, inter-annual, and seasonal distributions. Annual $PM_{2.5}$ was up to six times of the national air quality standard 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 (SO_4^{2-}) and ammonium (NH_4^+) remained at high levels, and nitrate (NO_3^-) increased. The highest seasonal averages of $PM_{2.5}$ and its major chemical components were mostly 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 values in southern regions due to the frequent dust events in northern China.

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 RH=80% can be amplified about 1.8 times of that under dry condition. Secondary inorganic aerosols accounted for about 60% of aerosol extinction coefficient (b_{ext}) under ambient conditions in megacities with RH higher than 70%. The mass scattering efficiency (MSE) of $PM_{2.5}$ ranged from 3.0 to 5.0 $m^2\ g^{-1}$ for aerosols

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

produced from anthropogenic emissions and from 0.7 to 1.0 m^2 g^{-1} for natural dust aerosols. The mass absorption efficiency (MAE) of EC ranged from 6.5 to 12.4 m^2 g^{-1} in urban environments, but the MAE of water-soluble organic carbon (WSOC) was only 0.05 to 0.11 m^2 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 emission policy making.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

30 # Contents

46

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

1. Introduction

Knowledge of spatiotemporal variations of 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 weather occurrence 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 the National Ambient Air Quality Standards (NAAQS) to include $PM_{2.5}$ daily and annual 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 $PM_{2.5}$ level, 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 matters) can enhance light extinction and haze intensity (Liu et al., 2011; Liu et al., 2013b; Zhang et al., 2015b; Zieger et al., 2013). A large number of studies has been conducted in China in recent

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

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; Liu et al., 2012; Pan et al., 2009; Tao et al., 2014b; Wang et al., 2015b; Yan et al., 2008; Yan et al., 2009; Yang et al., 2011b; Zheng et al., 2016). However, few studies looking at the geographical pattern of $PM_{2.5}$ composition across China and its impact on aerosol optical properties (Li et al., 2017). The present study aims to gain such knowledge through a thorough review of available studies.

Considering the large number of available 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 of 2000 in scientific papers of Science Citation Index (SCI) journals are reviewed and summarized in this study. A total of about 130 articles met the above criteria including 80 articles on $PM_{2.5}$ chemical composition, 40 articles on aerosol optical properties, and 10 articles on aerosol hygroscopic properties. Many of these articles focused on the several biggest cities such as Beijing, Shanghai, Guangzhou and Hong Kong, while other studies focused on cities including Tianjin, Jinan, Nanjing, Hangzhou, Fuzhou, Xiamen, Shenzhen, Chengdu, Chongqing, Xi'an, Lanzhou, Zhengzhou, Wuhan and several background sites (Fig. 2). Geographical and temporal patterns of $PM_{2.5}$ and its major chemical components including (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic carbon (OC), and elemental carbon (EC), and aerosol optical properties are generated. Relationships between aerosol optical

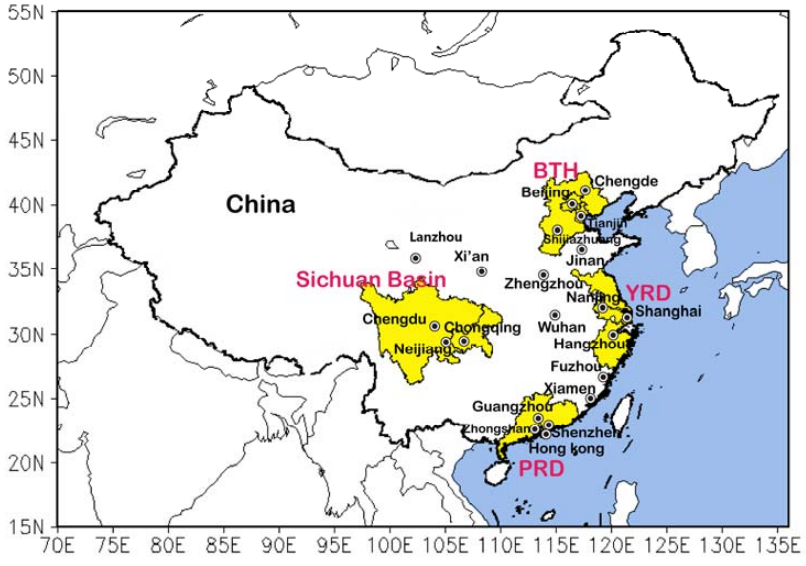
Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

91 properties and $PM_{2.5}$ chemical composition are explored. Recommendations are also
92 provided for alleviating $PM_{2.5}$ level and reducing haze occurrence frequency.

93 2. Spatiotemporal patterns of $PM_{2.5}$ and its major chemical
94 components

95 In this section, available measurements of chemically resolved $PM_{2.5}$ are
96 reviewed and summarized in terms of geographical distributions, inert-annual
97 variations, and seasonal patterns. Measurements are grouped based on geographical
98 regions, such as the Beijing-Tianjin-Hebei (BTH) in North China Plain, the Yangtze
99 River Delta (YRD), the Pearl River Delta (PRD), the Sichuan Basin, and other regions
100 (Fig. 1). Five dominant chemical components of $PM_{2.5}$ (SO_4^{2-}, NO_3^-, NH_4^+, OC, and
101 EC) are discussed in detail. Data reviewed in this section are all listed in Table S1 of
102 the supplement document.



103

104 Fig. 1. Geographical regions and location of cities with measurements.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

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, Fuzhou and Xiamen). Geographical characteristics of annual $PM_{2.5}$ are first discussed followed by internal annual variations and seasonal patterns.

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±30, 92±29, 50±16, and 100±35 μg m^{-3} in BTH (Chen et al., 2014b; Duan et al., 2006; He et al., 2001; He et al., 2012; Song et al., 2006; Wang et al., 2005; Yang et al., 2011a; Yang et al., 2011b; Zhang et al., 2013a; Zhao et al., 2013c; Zhou et al., 2015a), YRD (Feng et al., 2009; Li et al., 2015a; Li et al., 2016a; Liu et al., 2015; Wang et al., 2006; Wang et al., 2016a; 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; Tao et al., 2017), and Sichuan basin (Chen et al., 2014c; Tao et al., 2013a; Tao et al., 2014a; Wang et al., 2017; Yang et al., 2011b), respectively, which was 3 to 6 times, 2 to 3 times, 1 to 2 times, and 3 to 6 times of NAAQS, respectively.

Within each region, the highest annual average $PM_{2.5}$ concentration was

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

observed in Shijiazhuang (191 μg m^{-3}), Nanjing (130 μg m^{-3}), Guangzhou (65 μg m^{-3}) and Chengdu (111 μg m^{-3}) in BTH, YRD, PRD and Sichuan basin, respectively. Outside the above-mentioned four regions, annual $PM_{2.5}$ at individual cities was 192±17 μg m^{-3} (Geng et al., 2013; Wang et al., 2015a), 172±15 μg m^{-3} (Shen et al., 2009; Wang et al., 2015c; Zhang et al., 2011b), 89 μg m^{-3} (Wang et al., 2016b), 149 μg m^{-3} (Yang et al., 2012), 107 μg m^{-3} (Zhang et al., 2015a), 78 μg m^{-3} (Zhang et al., 2011a; Zhang et al., 2012a) and 44 μg m^{-3} (Xu et al., 2012c) in Zhengzhou, Xi'an, Lanzhou, Jinan, Wuhan, Xiamen and Fuzhou, 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 Chengdu, and Fuzhou and Xiamen of Fujian province to Guangzhou. Cities in Fig. 2a are rearranged in Fig. 2b based on their latitude from north to south. 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 at 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., 2014b).

Filter-based measurements of $PM_{2.5}$ at rural sites in China were limited, and were 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

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

region. A similar geographical patter 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 μg m^{-3}, which was 2 times of that (35 μg m^{-3}) at the rural sites of PRD.

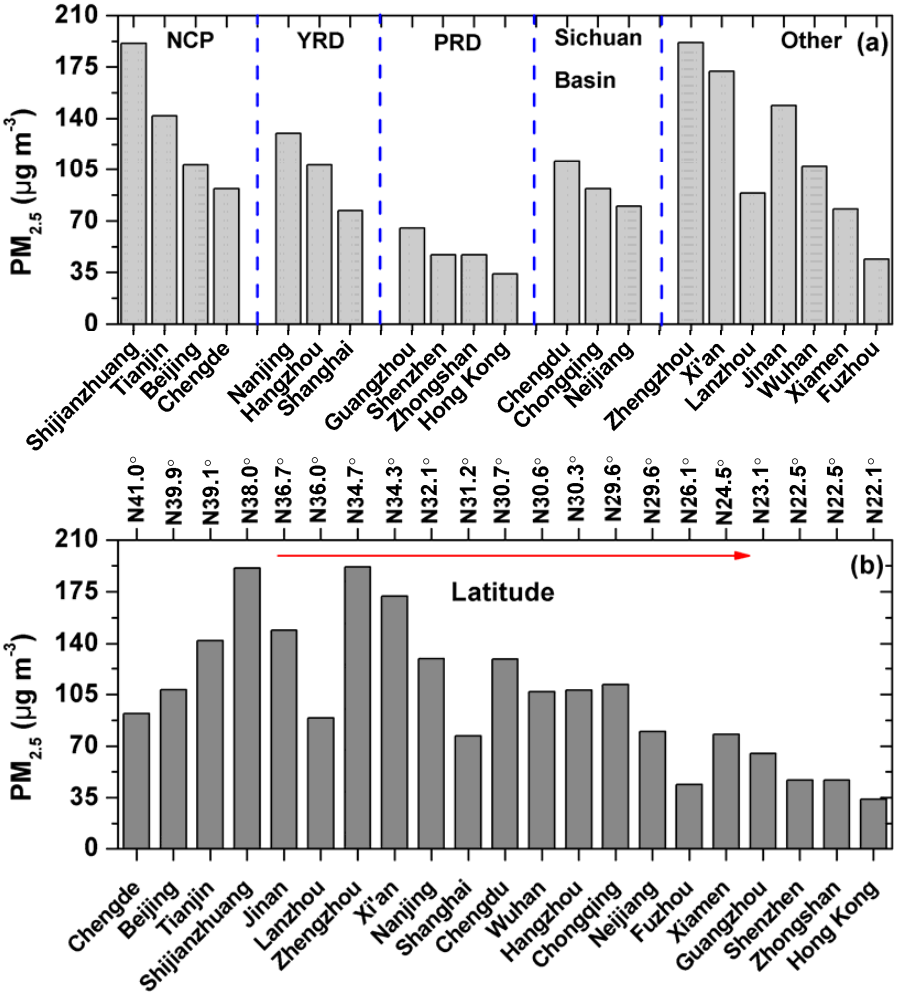


Fig. 2. Annual $PM_{2.5}$ mass concentration in various Chinese cities having filter-based measurements: (a) categorized into regions, and (b) lined with latitude.

2.1.2 Inter-annual variations

Data collected in most cities were within a three-year time window, except in

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Beijing, Shanghai and Guangzhou where $PM_{2.5}$ data spanned for more than a decade (1999-2014) (Fig. 3). Inter-annual variations in $PM_{2.5}$ in Beijing were small, ranging from 100 to 128 μg m^{-3}, similar to the trend of online data, which ranged from 65 to 83 μg m^{-3} during 2004-2012 (Liu et al., 2014b). The lower concentrations of the online than filter $PM_{2.5}$ data should be caused by volatilization loss of nitrate and organic matters from the tapered element oscillating microbalances (TEOM) working at the 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 excised. The impact of local effort of pollution control in Beijing has likely been offset by regional pollutant transport (Li et al., 2015b). In Shanghai, $PM_{2.5}$ in 2003-2006 (94 μg m^{-3}) (Feng et al., 2009; Wang et al., 2006) and 2009 (94 μg m^{-3}) (Zhao et al., 2015b) was nearly 50% higher than earlier years (e.g., 65 μg m^{-3} in 1999-2000) (Ye et al., 2003), but has then been decreased substantially and reached a level of 58 μg m^{-3} in 2011-2013 (Wang et al., 2016a; Zhao et al., 2015b). In Guangzhou, $PM_{2.5}$ in 2002-2003 (71 μg m^{-3}) (Hagler et al., 2006) and in 2009-2010 (77 μg m^{-3}) (Tao et al., 2014c) kept the stable levels and then decreased to 48 μg m^{-3} in 2014 (Tao et al., 2017).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

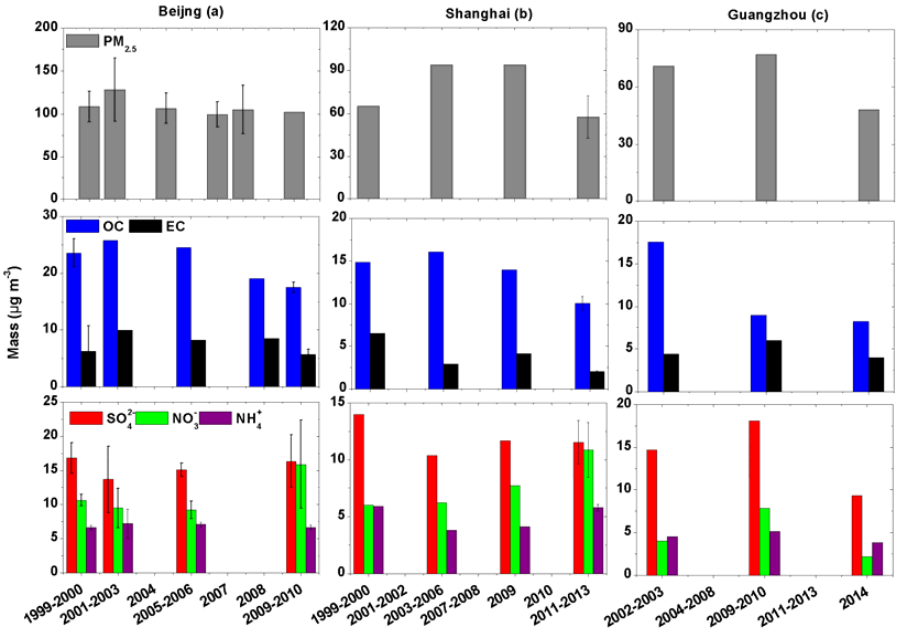


Fig. 3. Inter-annual variations in $PM_{2.5}$ and dominant chemical components in Beijing (a), Shanghai (b) and Guangzhou(c).

2.1.3 Season 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 the factors of 1.7, 1.4, 1.6 and 1.8 in Beijing (Cao et al., 2012b; Chan et al., 2005; Dan et al., 2004; Duan et al., 2006; He et al., 2001; Huang et al., 2014b; Ji et al., 2014; Jung et al., 2009b; Lin et al., 2016; Okuda et al., 2011; Pathak et al., 2011; Song et al., 2006; Song et al., 2007; Sun et al., 2004; Sun et al., 2006; Tan et al., 2016; Tao et al., 2016a; Tao et al., 2015a; Tian et al., 2015; Wang et al., 2005; Yang et al., 2005a; Yang et al., 2016; Zhao et al., 2013c), Tianjin (Cao et al., 2012b; Gu et al., 2010; Gu et al., 2011; Li et al., 2009; Zhao et al., 2013c), Shijiazhuang (Zhao et al., 2013c),

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

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; Lu et al., 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.

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 the factors of 2.3, 1.9 and 2.0 in Nanjing (Li et al., 2015a; Li et al., 2016a; Shen et al., 2014; Yang et al., 2005b), Shanghai (Cao et al., 2012b; Cao et al., 2013; Feng et al., 2009; Feng et al., 2012; Huang et al., 2014b; Pathak et al., 2011; Wang et al., 2006; Wang et al., 2016a; 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., 2003; Cao et al., 2012b; Cui et al., 2015; Duan et al., 2007; Huang et al., 2007; Huang et al., 2013; Huang et al., 2014b; Jahn et al., 2013; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tan et al., 2009; Tan et al., 2014; Tao et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Yang et al., 2011b). Although the highest seasonal average $PM_{2.5}$ was also observed in winter, the lowest season 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

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

thus small differences between spring and summer, e.g., $PM_{2.5}$ concentration of 32 μg m^{-3} in summer (Cao et al., 2003; Cao et al., 2012b; Duan et al., 2007; Ho et al., 2006; Lai et al., 2007; Louie et al., 2005a) and 29 μg m^{-3} in spring (Louie et al., 2005a) in Hong Kong. Seasonal variations were up to a factor of 1.9 at both cities. $PM_{2.5}$ at rural sites in PRD was 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 season patterns as above were also observed in cities of other regions in China, such as Chengdu (Tao et al., 2013a; Tao et al., 2014a), Zhengzhou (Geng et al., 2013), Jinan (Yang et al., 2012) and Fuzhou (Xu et al., 2012b), with seasonal variations between the factors of 1.8 to 2.5. In conclusion, the highest seasonal average $PM_{2.5}$ was observed in winter in all the urban sites in China likely due to more emissions from winter heating and/or poor pollutant diffusion conditions.

2.2 Major chemical components of $PM_{2.5}$

It is well known that OC, EC, SO_4^{2-}, NO_3^- and NH_4^+ were 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).

2.2.1 The Beijing-Tianjin-Hebei region

2.2.1.1 Inter-annul variations in Beijing

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Chemically-resolved $PM_{2.5}$ data in BTH covering multiple-years are only available in Beijing and the inert-annual variations are discussed for this city below (Duan et al., 2006; He et al., 2001; Song et al., 2006; Yang et al., 2011b; Zhang et al., 2013a; Zhao et al., 2013c). Inter-annual variations of 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 μg m^{-3} in earlier years (1999-2006) to below 17.6 μg m^{-3} after 2008. EC increased from 6.3 μg m^{-3} in 1999-2000 to 9.9 μg m^{-3} in 2001-2002, and then gradually decreased to 5.7 μg m^{-3} in 2009-2010. Despite using different analysis methods (e.g. CHN elemental analyzer, thermal-optical carbon analyzer) and protocols (e.g. TOT and TOR) for obtaining carbon fraction, total carbon (TC, the sum of OC and EC) showed slightly decrease. OC is produced from both primary emissions and secondary formation and EC (also known as black carbon or BC) is mainly from primary emissions. The anthropogenic emission for OC and BC in the whole 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 of 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 μg m^{-3} to 16.4 μg 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 SO_2 emission in BTH (including Beijing, Tianjin, and Hebei province) increased sharply from 2097 Gg year^{-1} in 2000 to 2916 Gg year^{-1} in 2004, and further slightly increased to 2998 Gg year^{-1} in 2007 before sharply decreased to 1821 Gg year^{-1} in 2010 (Lu et al., 2010; Zhao et al., 2013a). A continued increase in SO_2 emission was found in Hebei province, which accounted for more than 50% of the total SO_2 emission in BTH. In contrast, SO_2 emission in Beijing continued decreasing. Surface annual SO_2 concentration in Beijing gradually decreased from 56 μg m^{-3} to 35 μg m^{-3} during 2006-2009 (http://www.zhb.gov.cn/). Thus, the persistent high concentrations of SO_4^{2-} in Beijing was largely due to regional transport from Hebei province, noting that the lifetime of SO_4^{2-} is longer than that of SO_2.

NO_3^- 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 (NO_2+NO) emissions and satellite NO_2 vertical column densities synchronously increased during 2000-2010 in China (Zhang et al., 2012b; Zhao et al., 2013b). Different from those of SO_2 emissions, NO_x emissions in all the cities and provinces in BTH showed increasing trends in 2005-2010. NO_x emission in Beijing slightly increased from 410 Gg year^{-1} in 2005 to 480 Gg year^{-1} in 2010 (Zhao et al., 2013b). However, annual average surface NO_2 concentration 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/).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

There were some inconsistences between the trends of surface NO_2 concentration and column NO_2 or NO_x emission, likely due to the impact of photochemical reaction on surface NO_2 concentration in urban areas. To some extent, the increasing trend of NO_3^- in Beijing was likely related to the increases in NO_x emissions in both Beijing and the surrounding cities or provinces.

NH_4^+ concentrations were also relatively steady in Beijing during 1999-2006, ranging from 5.7 to 7.3 μg m^{-3}. NH_3 emissions changed little (13400-13600 Gg $year^{-1}$) before 2005 in China, and increased slightly in BTH region during 2003-2010 (Zhou et al., 2015b). The small increase of NH_4^+ in 2009-2010 in Beijing was consistent with the NH_3 emission trend in this region (Zhang et al., 2013a; Zhao et al., 2013c). Moreover, the increase of NO_3^- in Beijing was also an important factor for the increase of NH_4^+.

In summary, an decreasing trend was identified in TC and increasing ones for SO_4^{2-}, NO_3^- and NH_4^+ in Beijing. The inter-annual variations in EC agreed with the its local emission trend in Beijing, but those in SO_4^{2-}, NO_3^- and NH_4^+ 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 the other important factors affecting aerosol formation, such as meteorological-dependent chemical reaction mechanisms.

2.2.1.2 Relative contributions to $PM_{2.5}$

To investigate the relative contributions of dominant chemical components to

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

298 $PM_{2.5}$ mass, the measured $PM_{2.5}$ mass was reconstructed based on SO_4^{2-}, NO_3^-, NH_4^+,
299 OM (organic matter), and EC. The converting factor between OC and OM was 1.8
300 considering the prevailing biomass burning in BTH (Cheng et al., 2013a; Du et al.,
301 2014a).

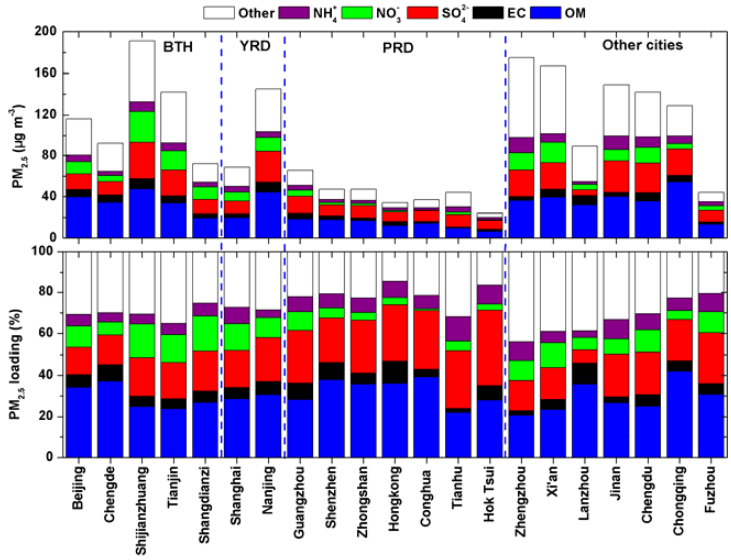
302 Data collected in 2009-2010 were first discussed since multiple cities in BTH
303 have data during this period (Fig. 4) (Zhang et al., 2013a; Zhao et al., 2013c).
304 Secondary inorganic aerosols (the sum of sulfate, nitrate and ammonium) contributed
305 36-39% of $PM_{2.5}$ annually in the majority of the cities having measurements, but
306 only 25% in Chengde, a tourist city located in the northeast part of BTH and 200
307 kilometer away from Beijing. Generally, the percentage contribution of secondary
308 inorganic aerosols to $PM_{2.5}$ decreased with decreasing $PM_{2.5}$ level, e.g., from
309 Shijiazhuang to Tianjin, Beijing, and then Chengde, a phenomenon that is consistent
310 with what was found within the same city but for different pollution levels in a
311 winter season (Tao et al., 2015a). Carbonaceous aerosols contributed 29-32% to
312 $PM_{2.5}$ in most cities, but as high as 45% in Chengde, and had an opposite trend to
313 secondary inorganic aerosols in terms of city-to-city variations. At the rural site
314 Shangdianzi near Beijing, secondary inorganic aerosols and carbonaceous aerosols
315 accounted for 42% and 32%, respectively, of $PM_{2.5}$ mass, which were not
316 significantly different from those in cities located south of Yanshan Mountain. The
317 sum of secondary inorganic aerosols and carbonaceous aerosols accounted for
318 65%-70% of $PM_{2.5}$ mass in cities of BTH.

319 In Beijing where data are available for more than a decade, secondary inorganic

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

320 aerosols accounted for 28% of $PM_{2.5}$ on average and ranged from 23% to 31% from
321 year to year. Carbonaceous aerosols accounted for 43% of $PM_{2.5}$ and ranged from
322 29% to 55%. Seasonal average contributions of secondary inorganic aerosols were
323 generally higher in warm seasons than in cold seasons in most cities, and an opposite
324 trend was found for carbonaceous aerosols (Fig.5). For example, secondary inorganic
325 aerosols contributed 32%, 41%, 28% and 32% in spring, summer, autumn and winter,
326 respectively, to $PM_{2.5}$ in Beijing, while carbonaceous aerosols contributed 35%, 30%,
327 44% and 45% (Cao et al., 2012b; Duan et al., 2006; He et al., 2001; Huang et al.,
328 2014b; Pathak et al., 2011; Song et al., 2007; Song et al., 2006; Sun et al., 2004; Tao
329 et al., 2015a; Tian et al., 2015; Zhang et al., 2013a; Zhao et al., 2013c). Higher
330 carbonaceous aerosols in winter should be related to heating activities and biomass
331 burning in this region (Cheng et al., 2013a; Duan et al., 2004; Tao et al., 2016b;
332 Wang et al., 2007; Yang et al., 2016).



333

334 Fig. 4. Annual $PM_{2.5}$ and dominant chemical components in China.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

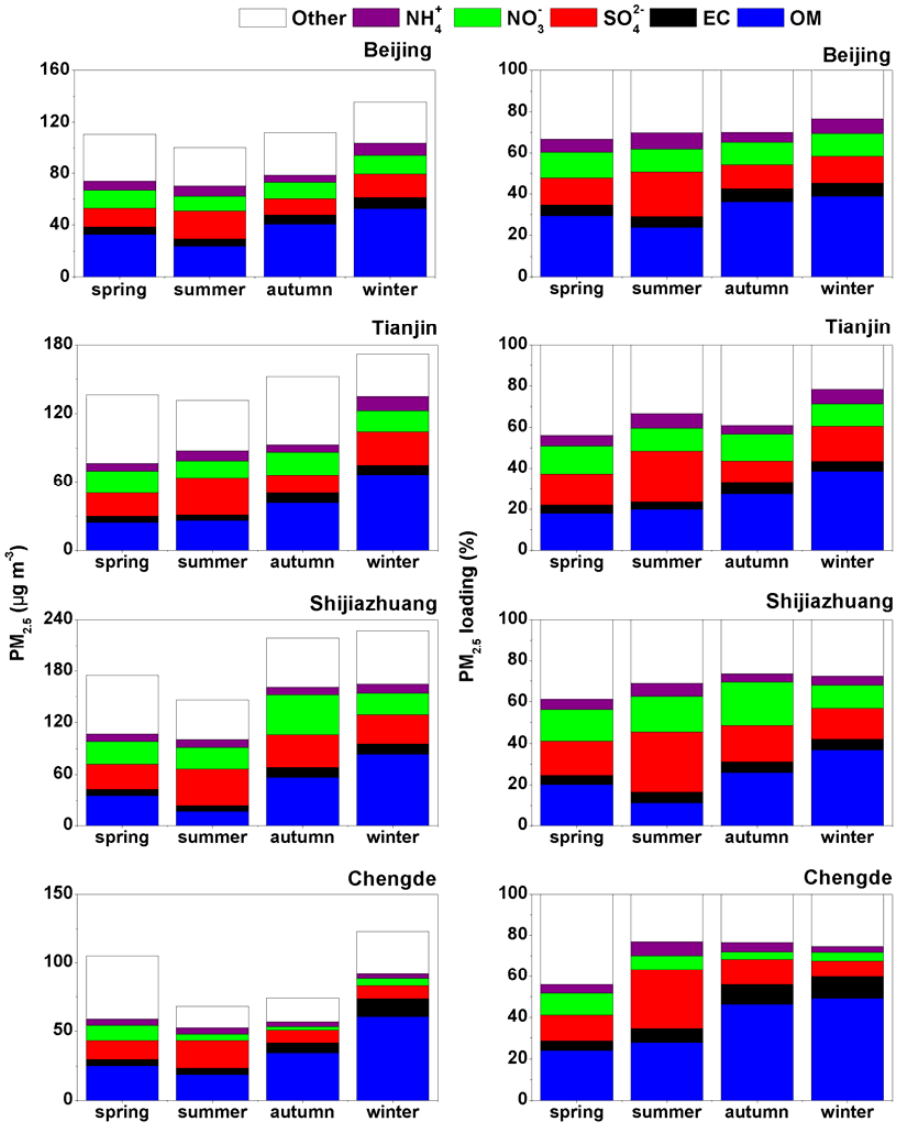


Fig. 5. Seasonal $PM_{2.5}$ and dominant chemical components in BTH.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

2.2.2 The Yangtze River Delta region

2.2.2.1 Inter-annul variations in Shanghai

Chemically-resolved $PM_{2.5}$ data in YRD covering multiple-years are only available in Shanghai (Wang et al., 2016a; Ye et al., 2003; Zhao et al., 2015b). Inter-annual variations of OC in this city were within a factor of 1.6 for OC and a factor of 3.3 for EC (Fig. 3b). OC concentrations were relatively steady (14.0-14.9 μg m^{-3}) during 1999-2009, but sharply decreased to 10.1 μg m^{-3} in 2011-2013. EC varied in the range of 4.1 to 6.5 μg m^{-3} during 1999-2009, and also sharply decreased to 2.1 μg m^{-3} in 2011-2013. Noticeable reduction of OC and EC occurred after 2010 Shanghai World Expo, which resulted in evident decrease of 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). Especially, BC emission in Jiangsu province was much higher than the sum of those in Shanghai and Zhejiang. Thus, the decreased EC concentration in Shanghai was mostly resulted from local emission reduction.

Annual SO_4^{2-} concentration decreased from 14.0 μg m^{-3} in 1999-2000 to the range of 10.2 μg m^{-3} to 12.9 μg m^{-3} during 2009-2013. The trend of SO_2 emission in YRD generally agreed with that in the whole China, which showed an increasing trend during 2000-2006 and a decrease one during 2006-2010 (Lu et al., 2011). The annual variations in SO_2 emission in YRD (including Shanghai, Jiangsu, and Zhejiang) were relative small, ranging from 3171 Gg $year^{-1}$ in 2000, 3506 Gg $year^{-1}$ in 2004, 3376 Gg $year^{-1}$ in 2007, and to 3397 Gg $year^{-1}$ in 2010 (Lu et al., 2010;

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

363 Zhao et al., 2013a). Annual average SO_2 concentrations in Shanghai were in the
364 range of 45-61 μg m^{-3} during 2000-2005 and decreased by around 50% to 24-29 μg
365 m^{-3} during 2010-2013 (http://www.zhb.gov.cn/). Note that SO_2 emissions in
366 Shanghai only accounted for less than 20% of the total SO_2 emissions in YRD and
367 with small annual variations. The high concentrations of SO_4^{2-} observed in Shanghai
368 were also closely related to regional transport from north China (e.g. BTH and
369 Shandong province) (Li et al., 2011; Wang et al., 2016a).

370 Annual NO_3^- concentrations in Shanghai were relatively steady (6.0-7.7 μg m^{-3})
371 during 1999-2009, but sharply increased to 10.9 μg m^{-3} in 2011-2013. NO_x
372 emissions in YRD also showed an increasing trend during these years, consistent
373 with satellite retrieved vertical column NO_2 density during 2000-2010 (Zhang et al.,
374 2012b; Zhao et al., 2013b). In contrast, surface-level annual NO_2 concentration in
375 Shanghai sharply decreased from 90 μg m^{-3} in 2000 to a range of 48-61 μg m^{-3}
376 during 2003-2013 (http://www.zhb.gov.cn/). The inconsistence in the trends between
377 emissions and gaseous and particulate matters surface air concentrations was similar
378 to that found in Beijing. Photochemistry and regional transport of related pollutants
379 should be the major causes of this phenomenon.

380 Annual NH_4^+ concentrations decreased from 5.9 μg m^{-3} in 1999-2000 to the
381 levels of 4.1 μg m^{-3} in 2009 and then recovered to 5.8 μg m^{-3} in 2011-2013. NH_3
382 emission increased in 2000-2005 in east China (including BTH, YRD and PRD) and
383 possibly also increased in 2006-2010 due to the lack of control measures for NH_3 in
384 China (Wang et al., 2011). The recently increased NH_4^+ concentrations in Shanghai

were likely due to the concurrent increases of NH_3 emissions and NO_3^- concentrations.

In summary, a decreasing trend was identified in TC, increasing ones for NO_3^- and NH_4^+, and a stable one for SO_4^{2-} in Shanghai. The inter-annual variations in EC agreed with the its local emission trend in Shanghai rather than the regional transport. In contrast, inter-annual variations in SO_4^{2-}, NO_3^- and NH_4^+ agreed more with the regional scale emission trends of their respective gaseous precursors in YRD. Similar to what was found for Beijing, nonlinear responses of concentration changes of these aerosol components to their respective emission trends were also found in Shanghai.

2.2.2.2 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 converting factor of 1.6 between OC and OM was chosen for YRD, slight smaller than that (1.8) chosen for BTH considering the less impact of biomass burning to $PM_{2.5}$ 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., 2016a; Ye et al., 2003; Zhao et al., 2015b). The sum of secondary inorganic aerosols (sulfate, nitrate and ammonium) and carbonaceous aerosols (OM and EC) accounted for 76% and 66% of $PM_{2.5}$ mass in Shanghai and Nanjing, respectively, which was comparable with those (65%-70%) in BTH.

Seasonal variations of secondary inorganic aerosols contributions to $PM_{2.5}$ were

small in both cities, e.g., 41-49% in Shanghai and 32-40% in Nanjing. Larger seasonal variations were found for carbonaceous aerosols than secondary inorganic aerosols, e.g., 47% in summer and 33%-39% in the other seasons in Shanghai, and ranged from 27% (spring) to 65% (autumn) in Nanjing (Cao et al., 2012b; Huang et al., 2014a; Huang et al., 2014b; Li et al., 2016a; Pathak et al., 2011; Shen et al., 2014; Wang et al., 2016a; 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 the different local sources in YRD. The seasonal patterns of these chemical components in Shanghai were resulted from both local emissions and regional transport, but in Nanjing mainly determined by local emissions because Nanjing is an inland city surrounded by many industrial enterprises including power plants, petrochemical plants, and steel plants.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

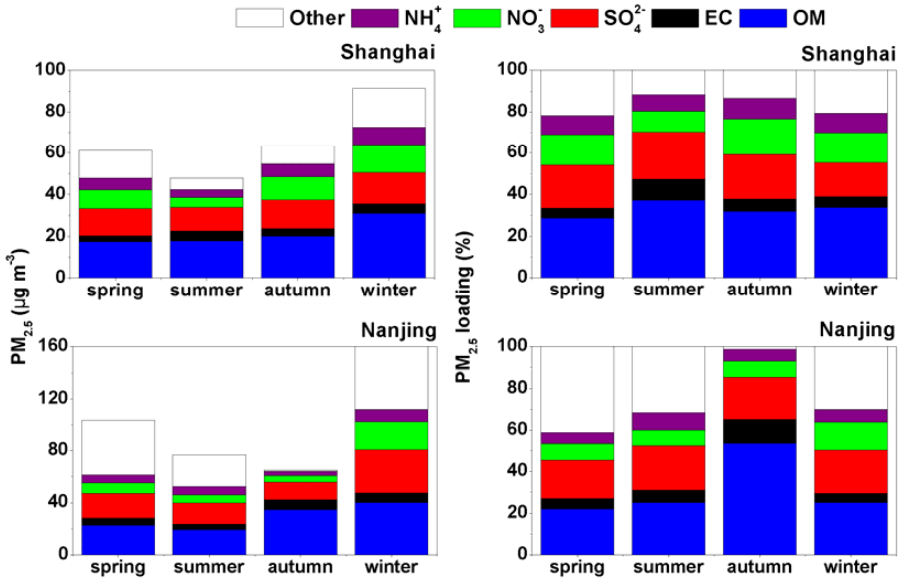


Fig. 6. Seasonal $PM_{2.5}$ and dominant chemical components in YRD.

2.2.3 The Pearl River Delta region

2.2.3.1 Inter-annul variations in Guangzhou

Inter-annual 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; Tao et al., 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. Annual OC concentration decreased significantly from 17.6 μg m^{-3} in 2002-2003 to 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 μg m^{-3} to 6.0 μg m^{-3} and then decreased to 4.0 μg m^{-3} during the same periods. Similar to Guangzhou, annual OC concentration decreased

441 significantly from 11.1 μg m^{-3} in 2002-2003 to 8.3 μg m^{-3} in 2009-2010 in Shenzhen,
442 while EC slightly increased from 2.3 μg m^{-3} to 2.7 μg m^{-3}. Apparently, the trends of
443 EC in Guangzhou and Shenzhen was inconsistent with the BC emission trend in
444 Guangdong province during 2005-2009, which showed slightly decrease (Qin and
445 Xie, 2012). As a result, TC concentrations gradually decreased from 22.0 μg m^{-3} to
446 15.0 μg m^{-3} in Guangzhou and from 15.0 μg m^{-3} to 13.0 μg m^{-3} in Shenzhen before
447 2010, similar to what was found in Beijing and Shanghai. The same phenomenon
448 was also observed at a suburban site of Guangzhou (Hagler et al., 2006; Lai et al.,
449 2016).

450 Contrast to the TC trend, annual SO_4^{2-}, NO_3^- and NH_4^+ concentrations in
451 Guangzhou increased from 14.7, 4.0 and 4.5 μg m^{-3} in 2002-2003 to 18.1, 7.8 and
452 5.1 μg m^{-3} in 2009-2010 and evidently decreased to 9.3, 2.2 and 3.8 μg m^{-3} in 2014,
453 respectively. The similar increases were also found in Shenzhen, e.g., from 10.0, 2.3
454 and 3.2 μg m^{-3} in 2002-2003 to 11.7, 2.7 and 3.5 μg m^{-3} in 2009, respectively
455 (Hagler et al., 2006; Huang et al., 2013), and in the suburban of Guangzhou, e.g.,
456 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,
457 respectively (Hagler et al., 2006; Lai et al., 2016). SO_2 emissions in Guangdong
458 province gradually increased in the previous decade, e.g., 964, 1150, 1177 and 1258
459 Gg $year^{-1}$ in 2000, 2004, 2007 and 2010, respectively (Lu et al., 2010; Zhao et al.,
460 2013a). However, SO_2 emissions in PRD decreased more than 40% in 2009
461 compared with that in 2005, due to flue gas desulfurization facilities in power plants
462 and large industrial boilers installed in this region (Lu et al., 2013). Annual average

463 SO_2 concentrations in Guangzhou gradually increased from 45 μg m^{-3} in 2000 to 77
464 μg m^{-3} in 2004, and then decreased to 17 μg m^{-3} in 2014 (http://www.gzepb.gov.cn/).
465 Thus, the increased SO_4^{2-} concentration before 2010 in Guangzhou was largely due
466 to the regional transport of pollutants from outside of PRD. The decreased SO_4^{2-}
467 concentration in 2014 in Guangzhou was likely due to flue gas desulfurization
468 facilities in power plants and large industrial boilers extended to the whole
469 Guangdong province (http://www.gdep.gov.cn/).

470 Meanwhile, NO_x emissions increased in Guangdong province as well as in the
471 whole PRD, similar to the trends in BTH and YRD (Lu et al., 2013; Zhang et al.,
472 2012b; Zhao et al., 2013b). However, annual average surface NO_2 concentration in
473 Guangzhou fluctuated in the range of 61 - 73 μg m^{-3} during 2000-2007 and 48 - 56
474 μg m^{-3} during 2008-2014 (http://www.gzepb.gov.cn/). An opposite trend was also
475 found between NO_2 and NO_x emission with the former persistently decreased while
476 the latter increased in Guangzhou, although NO_3^- concentration was also increased.
477 Thus, emission as well as chemical processes both affect these ions concentrations in
478 air. Annual NH_4^+ concentrations slightly increased about 10% before 2010 in
479 Guangzhou and Shenzhen although NH_3 emissions changed little during 2002-2006
480 in PRD (Zheng et al., 2012). Thus, the increased NH_4^+ concentrations in Guangzhou
481 and Shenzhen during 2002-2010 were largely due to the increased SO_4^{2-} and NO_3^-,
482 which enhanced the conversion of NH_4^+ from NH_3.

483 In summary, a decreasing trend was identified in TC and increasing ones for
484 SO_4^{2-}, NO_3^- and NH_4^+ in Guangzhou and Shenzhen before 2010, while all chemical

485 components decreased after 2010 in Guangzhou. The inter-annual variations in EC
486 was inconsistent with BC emission trend in Guangdong province. In contrast,
487 inter-annual variations in SO_4^{2-}, NO_3^- and NH_4^+ agreed with the regional scale
488 emission trends of their respective gaseous precursors in Guangdong province rather
489 than PRD. Similar to what was found for Beijing and Shanghai, nonlinear responses
490 of concentration changes of these aerosol components to their respective emission
491 trends were also found in Guangzhou and Shenzhen.

492 **2.2.3.2 Relative contributions to $PM_{2.5}$**

493 Data collected in 2002-2003 were discussed since multiple cities (e.g.
494 Guangzhou, Conghua, Zhongshan, Shenzhen and Hong Kong) in PRD have data
495 during this period (Fig. 4) (Hagler et al., 2006). The converting factor between OC
496 and OM was chosen to be the same as in YRD (1.6). Secondary inorganic aerosols
497 contributed 33-38%, depending on location, of $PM_{2.5}$ annually, while carbonaceous
498 aerosols contributed 37-46%. It is noted that $PM_{2.5}$ in Guangzhou was much higher
499 than those in the other coastal cities (including Zhongshan, Shenzhen and Hong
500 Kong), but the contributions of secondary inorganic aerosols and carbonaceous
501 aerosols were not significantly different between these cities. At rural sites (Tianhu
502 and Conghua near Guangzhou and Hok Tsui near Hong Kong), secondary inorganic
503 aerosols and carbonaceous aerosols accounted for 35-48% and 24-43%, respectively,
504 of $PM_{2.5}$ mass, which were similar to those obtained in the cities in PRD (Hagler et
505 al., 2006; Lai et al., 2016; Louie et al., 2005b). Thus, the sum of secondary inorganic
506 aerosols and carbonaceous aerosols accounted for 68%-83% of $PM_{2.5}$ mass in the

PRD region, similar to what was found in Shanghai (YRD).

Although many studies have been conducted in PRD, most studies were short period studies. Studies covering the full four seasons were mainly carried out in Guangzhou and Hong Kong (Fig.7) (Andreae et al., 2008; Cao et al., 2003; Cao et al., 2012b; Cui et al., 2015; Ho et al., 2006; Huang et al., 2014b; Jung et al., 2009a; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Tan et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 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 averaging all the years data 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 of OC/EC ratios ranged from 1.6 to 3.4 in Guangzhou and ranged from 1.2 to 2.1 in Hong Kong, suggesting coal and vehicle exhaust as dominant sources in Guangzhou while vehicle exhaust as dominant source in Hong Kong (He et al., 2008; Watson et al., 2001).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

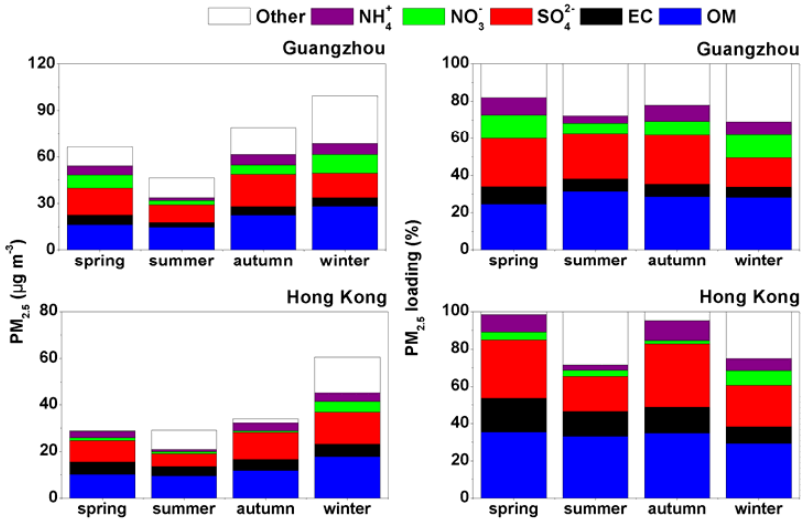


Fig. 7. Seasonal $PM_{2.5}$ and dominant chemical components in PRD.

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 of the other regions in China, mostly the capital city of a province (e.g. Zhengzhou of Henan province, Xi'an of Shaanxi province, Lanzhou of Gansu province, Jinan of Shandong province, Chengdu of Sichuan province, Chongqing of Chongqing municipality, and Fuzhou of Fujian province) (Fig.4) (Geng et al., 2013; Tao et al., 2013a; Tao et al., 2014b; Wang et al., 2015c; Wang et al., 2016b; Wang et al., 2017; Xu et al., 2012b; Yang et al., 2011b; Yang et al., 2012). A converting factor of 1.6 between OC and OM was chosen for Fuzhou and 1.8 for other cities based on their geographical locations.

Annual average contributions of secondary inorganic aerosols and carbonaceous

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

aerosols to $PM_{2.5}$ were 43% and 36%, respectively, in the coastal city Fuzhou, similar to what was found in Shanghai. Annual contributions of secondary inorganic aerosols ranged from 30% to 39% in inland cities (Zhengzhou, Xi'an, Jinan, Chengdu, Chongqing) except Lanzhou (12%), which were comparable with those observed in PRD (33-38%). In contrast, large differences were found in the annual contributions of carbonaceous aerosols, ranging from 23% in Zhengzhou to 51% in Lanzhou. The sum of secondary inorganic aerosols and carbonaceous aerosols accounted for 56%-79% of $PM_{2.5}$ mass in these cities.

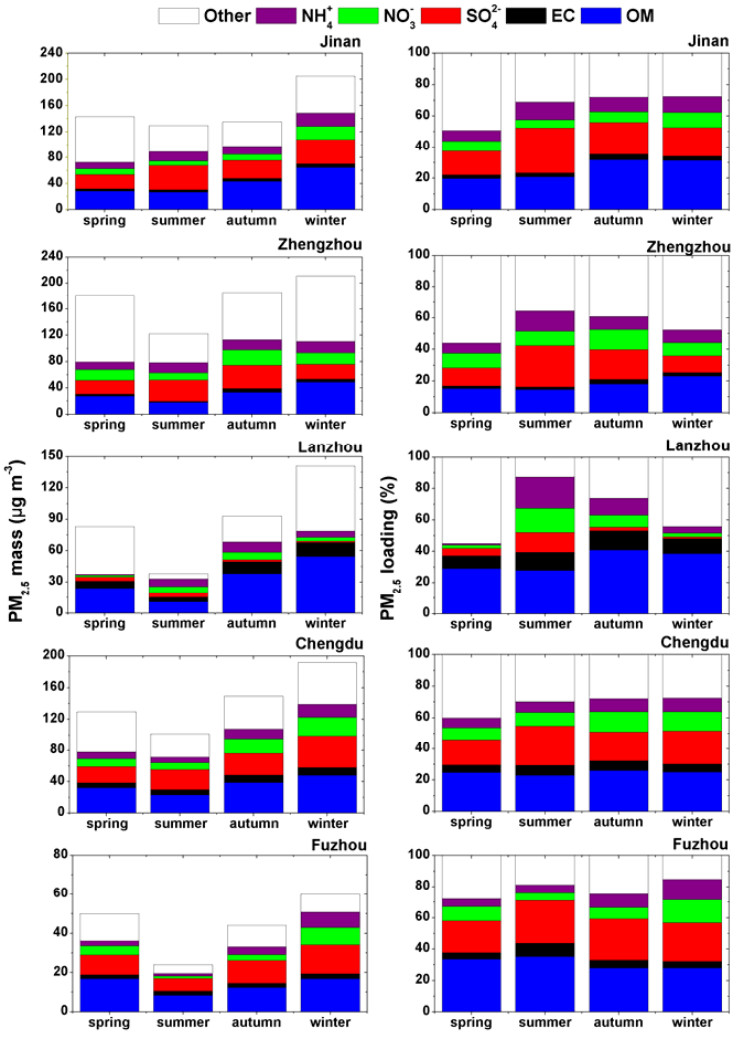
Seasonal average contributions are only shown for Chengdu, Zhengzhou, Jinan Lanzhou, and Fuzhou due to the incomplete data in Xi'an and Chongqing (Fig. 8). Seasonal contributions of secondary inorganic aerosols were evidently higher in summer than in other seasons in Zhengzhou, Jinan and Lanzhou (typical northern cities), similar to what was seen in BTH. Interestingly, in a southwest city Chengdu, seasonal contribution of secondary inorganic aerosols was only 30% in spring compared to the much higher values in the other seasons (40-42%). Moreover, in a southern coastal city Fuzhou, the highest seasonal average contribution of secondary inorganic aerosols was observed in winter (53%), much higher than in other seasons (34-42%).

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 what was observed in BTH. In contrast, higher seasonal contributions were found in warm

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

563 season than in cold seasons in the southern coastal city (Fuzhou), and flat seasonal
564 variations (29%-32%) in the southwest inland city (Chengdu). The sum contributions
565 of secondary inorganic aerosols and carbonaceous aerosols were evidently lower in
566 spring than in other seasons in most of the northern cities (e.g. Jinan, Lanzhou,
567 Zhengzhou, and BTH), likely due to the frequent spring dust storm events in
568 northern China.



569

570 Fig. 8. Seasonal $PM_{2.5}$ and dominant chemical components in other cities.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

2.2.5 Summary of $PM_{2.5}$ chemical properties

Carbonaceous aerosols showed decreasing trends over the last ten years (2000-2010) in Beijing, Shanghai and Guangzhou, consistent with BC emission trends in these cities and surrounding areas. SO_4^{2-} and NH_4^+ remained at high levels with no significant trends in Beijing and Shanghai, but with an increasing trend in Guangzhou. NO_3^- showed increase 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.

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. 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 be 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.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

3. Aerosol optical properties

There were 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 S2 of the supplement document. Measurements were available at 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 (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; Cheng et al., 2006b; Cheng et al., 2008a; Gao et al., 2015; Garland et al., 2008; Jung et al., 2009a; Lan et al., 2013; Man and Shih, 2001; Tao et al., 2014c; Verma et al., 2010; Wu et al., 2009; Wu et al., 2013), Chengdu in southwest China (Tao et al., 2014b; Wang et al., 2017), and Xi'an in northwest China (Cao et al., 2012a; Zhu et al., 2015), rural sites including rural Beijing (Shangdianzi) and rural Tianjin (Wuqing) in BTH (Ma et al., 2011; Yan et al., 2008; Zhao et al., 2011), and remote sites in north and northwest China (Li et al., 2010; Xu et al., 2004; Yan, 2007). Sites with one year or longer data included Beijing, rural Beijing, Shanghai, Guangzhou, Chengdu, Xi'an and Shouxian.

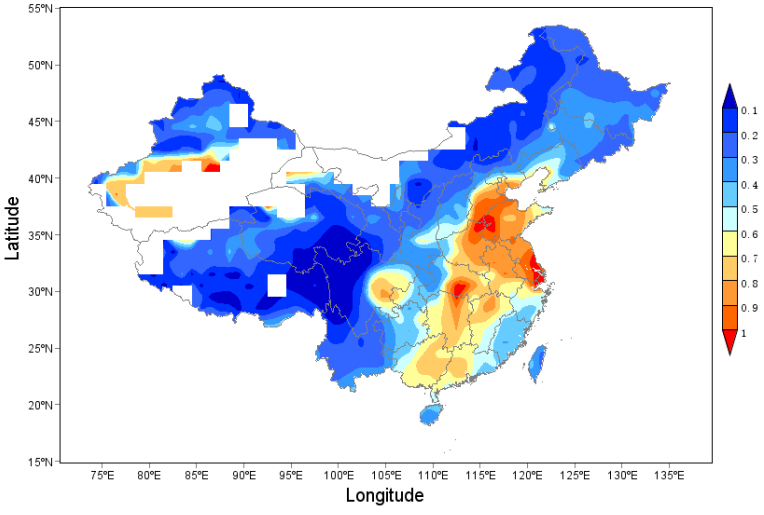
Aerosol optical depth (AOD), representing the integrated light extinction coefficient in a vertical column, can be achieved from MODerate-resolution Imaging Spectroradiometer (MODIS) data. Spatial distributions of annual average AOD in

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

614 2014 are shown in Fig. 9. Compared with spatial distributions of $PM_{2.5}$ shown in Fig.
615 2, similar general patterns were found between these two variables likely due to the
616 dominant role $PM_{2.5}$ played on light extinction. Differences in fine structures of their
617 patterns were due to surface $PM_{2.5}$ versus column AOD comparison and spatial
618 variations in $PM_{2.5}$ chemical composition.

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



625

626 Fig. 9. Spatial distribution of annual average AOD across China in 2014.

627 **3.1 Geographical patterns**

628 Annual average b_{sp} and b_{ap} from ground measurements in major cities in China

629 are plotted in Fig. 10. Most b_{sp} measurements were conducted using the
630 nephelometer under RH<60%. The highest annual b_{sp} was in Xi'an (525 Mm^{-1},
631 RH<60%) (Cao et al., 2012a), followed by Chengdu (456 Mm^{-1}, RH<40%; 421
632 Mm^{-1}, ambient RH) (Tao et al., 2014b; Wang et al., 2017), Guangzhou (326 Mm^{-1},
633 RH<70%) (Tao et al., 2014c), Beijing (309 Mm^{-1}, RH<60%) (He et al., 2009; Jing et
634 al., 2015; Zhao et al., 2011), and Shanghai (217 Mm^{-1}, RH<60%) (Cheng et al.,
635 2015). Such a spatial pattern was mostly due to the spatial pattern of annual $PM_{2.5}$
636 mass, i.e. Xi'an (177 μg m^{-3}) > Chengdu (111 μg m^{-3}) > Beijing (108 μg m^{-3}) >
637 Shanghai (77 μg m^{-3}) > Guangzhou (65 μg m^{-3}), and partly due to humidity
638 condition, e.g., Beijing versus Guangzhou. Noticeably, b_{sp} in Shouxian County was
639 higher than those in several megacities (e.g. Beijing, Shanghai and Guangzhou),
640 suggesting hazy weather also frequently occurred even in small cities in China (Fan
641 et al., 2010). b_{sp} in rural Beijing was 179 Mm^{-1} (Yan et al., 2008; Zhao et al., 2011),
642 which was much lower than that in urban Beijing, but was close to the level in
643 Shanghai.

644 Annual average b_{ap} ranged from 37 to 96 Mm^{-1} with higher values observed in
645 Chengdu and Xi'an (likely due to popular biomass burning besides large amount of
646 coal burning) (Cao et al., 2012a; Tao et al., 2014a; Tao et al., 2014b; Wang et al.,
647 2017; Zhang et al., 2014b), and lower values in Shouxian and rural Beijing (Fan et
648 al., 2010; Yan et al., 2008; Zhao et al., 2011). b_{ap} in Guangzhou was higher than that
649 in Beijing and Shanghai despite their similar $PM_{2.5}$ EC levels, likely due to the
650 different coating of EC in Guangzhou than in other cities. For example, the mass

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

absorption of EC in Guangzhou was 8.5 m^2 g^{-1} (at 532 nm) in autumn 2004 (Andreae et al., 2008), which was higher than that (4.2 m^2 g^{-1} at 870 nm, equivalent to 7.2 m^2 g^{-1} at 532 nm) in winter 2013 in Beijing (Wu et al., 2016).

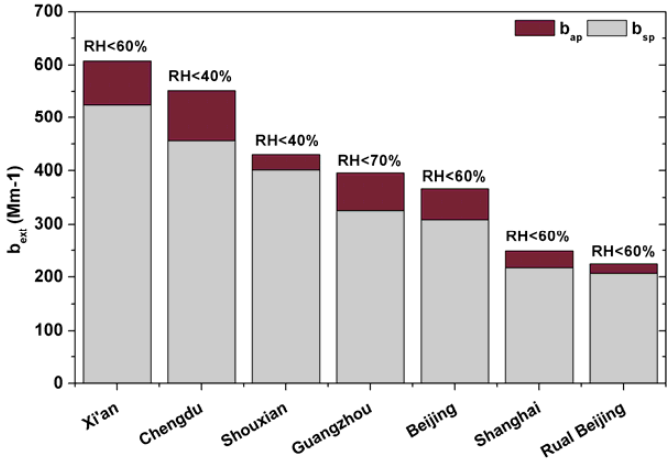


Fig. 10. Annual b_{sp} and b_{ap} in China.

3.2 Temporal patterns

3.2.1 The Beijing-Tianjin-Hebei region

b_{sp} measurements in BTH longer than one 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, when $PM_{2.5}$ increased by 20% from 107 to 129 μg 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 evident lower than that in 2005-2006. Meanwhile, annual secondary inorganic aerosols in 2009-2010 were evident lower than that in 2005-2006. The

666 coating by secondary inorganic aerosols likely enhanced the absorption of EC (Bond
667 et al., 2006; Cheng et al., 2009; Yu et al., 2010).

668 b_{sp} measurements in rural Beijing included the years of 2003-2005 and
669 2008-2009, while b_{ap} only included the years of 2003-2005. Generally, annual b_{sp}
670 and b_{ap} in rural Beijing changed little, which ranged from 175 to 182 Mm^{-1} and from
671 18 to 18 Mm^{-1}, respectively (Yan et al., 2008; Zhao et al., 2011). In conclusion, b_{sp}
672 and b_{ap} showed slightly increasing tendencies in urban and rural Beijing in recent
673 years.

674 Seasonal variations of b_{sp} and b_{ap} at urban and rural sites in Beijing are plotted in
675 Fig. 13. The highest seasonal average b_{sp} in Beijing was observed in winter and the
676 lowest in spring with seasonal variations up to a factor of 1.7 (Bergin et al., 2001;
677 Garland et al., 2009; Han et al., 2014; He et al., 2009; Jing et al., 2015; Li et al.,
678 2013b; Liu et al., 2009; Tao et al., 2015a; Tian et al., 2015; Zhao et al., 2011). A
679 different seasonal pattern was seen at the rural site located north of Beijing, which
680 showed 10-26% higher values in summer than in the other seasons (Yan et al., 2008;
681 Zhao et al., 2011). The highest seasonal b_{sp} in winter in Beijing was consistent with
682 the highest seasonal $PM_{2.5}$ mass. However, in rural Beijing the highest $PM_{2.5}$ mass
683 was observed in spring due to the frequent dust storm events, and the second highest
684 seasonal average $PM_{2.5}$ mass in summer which corresponded to the highest seasonal
685 b_{sp}. This is because scattering efficiency of dust aerosols was lower than that of
686 anthropogenic aerosols (Zhao et al., 2011).

687 The highest seasonal b_{ap} in Beijing appeared in autumn and the lowest in spring

688 with seasonal variations up to a factor of 2.0 (Bergin et al., 2001; Garland et al., 2009;
689 He et al., 2009; Jing et al., 2015; Li et al., 2013c; Liu et al., 2009; Tian et al., 2015;
690 Wu et al., 2016). Seasonal variations of b_{ap} were different from those of b_{sp} due to
691 their dependence on different chemical compounds, i.e. b_{sp} mainly on PM mass
692 while b_{ap} mainly on EC mass in PM and its coating. In rural Beijing b_{ap} was lower by
693 19%~57% in summer than in other seasons, and with similar seasonal variations to
694 b_{sp}, suggesting aerosols in rural Beijing mainly came from regional transport (Yan et
695 al., 2008).

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

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

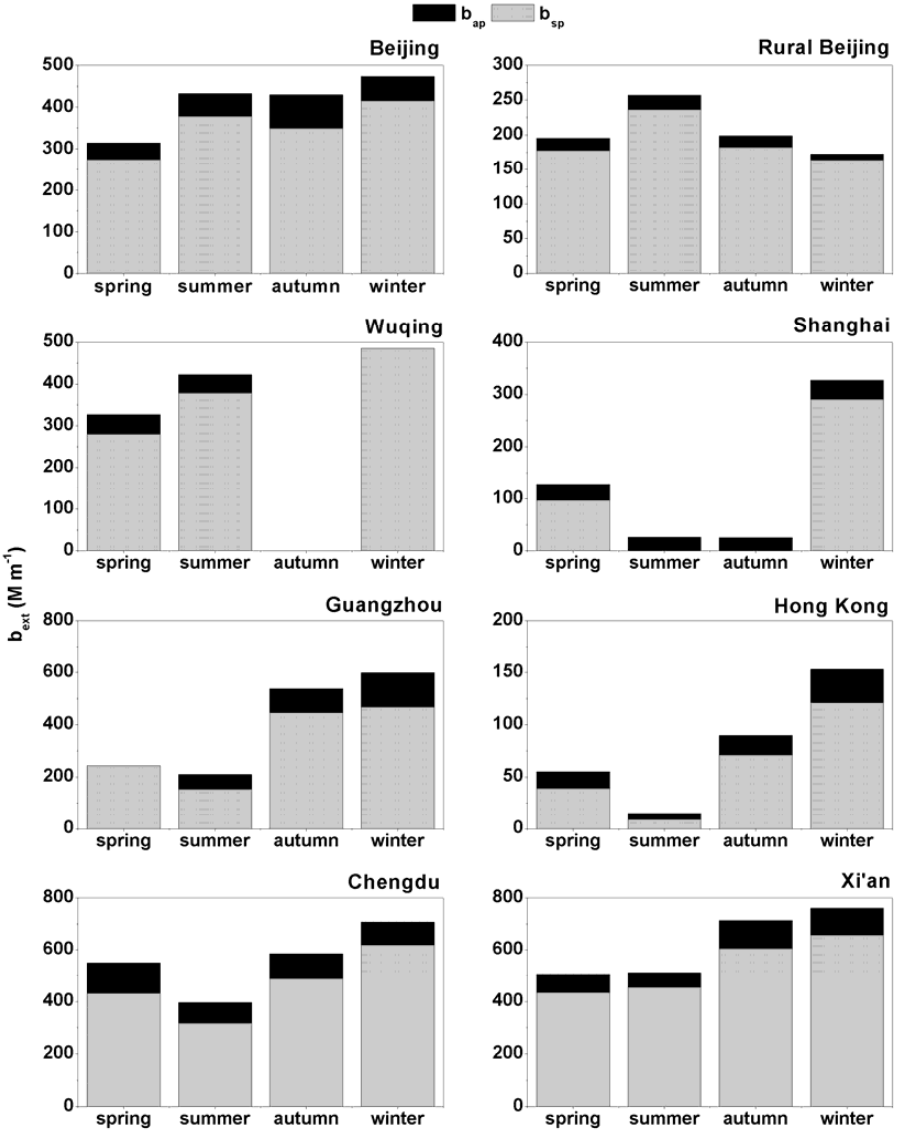


Fig. 11. Seasonal b_{sp} and b_{ap} in cities with measurements.

3.2.2 The Yangtze River Delta and Pearl River Delta region

No multi-year b_{sp} measurement data were available for exploring inter-annual 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 $\mu g\ m^{-3}$) was only half of that in 2009 (4.1 $\mu g\ m^{-3}$) (Wang et al., 2016a; 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 $\mu g\ m^{-3}$) was similar or slightly lower than that in 2002-2003 (4.4 $\mu g\ m^{-3}$) (Hagler et al., 2006; Huang et al., 2012). Thus, the inter-annual 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; Cheng et al., 2015; Feng et al., 2014; Han et al., 2015; Huang et al., 2014a; Li et al., 2013a; Pathak et al., 2011; Wang et al., 2016a; 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., 2003; Cao et al., 2004; Cao et al., 2012b; Cui et al., 2015; Gao et al., 2015; Huang et al., 2014b; Jung et al., 2009a; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tao et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Verma et al., 2010; Wu et al., 2009; Wu et al., 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.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

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 due to the large amount of OC emitted from biomass burning in spring season, which enhanced the absorption of EC (Schnaiter 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; Cao et al., 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 doubled those in spring of 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., 2009a; Pu et al., 2015; Tao et al., 2014b; Tao et al., 2014c; Tao et al., 2015a; Tao et al., 2016a; Tian et al., 2015; Wang et al., 2012; Zhao et al., 2011). A parameter

752 describing their relationship is defined as mass scattering efficiency (MSE), which is
753 the slop of the linear regression of b_{sp} against $PM_{2.5}$ mass. MSE was found to vary
754 with location and season due to the variations in $PM_{2.5}$ chemical composition. Some
755 of the variations may due to different sampling conditions, e.g., ambient (controlled
756 RH<60%) versus dry condition (controlled RH<40%), online versus filter-based
757 $PM_{2.5}$ sampling. Available MSE data are discussed here, although uncertainties from
758 measurements will not be addressed in this study.

759 In BTH, annual average $PM_{2.5}$ MSE was higher in Beijing (5.9 m^2 g^{-1}) than in
760 rural Beijing (4.8 m^2 g^{-1}) based on online $PM_{2.5}$ mass (Zhao et al., 2011). In urban
761 Beijing in winter of 2013, $PM_{2.5}$ MSE increased to 4.9 m^2 g^{-1} during the heavy
762 pollution episode and deceased to 3.6 m^2 g^{-1} during clean days, due to the large
763 fraction of soluble inorganic components (e.g. $(NH_4)_2SO_4$ and NH_4NO_3) in $PM_{2.5}$
764 under heavy polluted condition (Tao et al., 2015a). In rural Beijing in 2005-2010,
765 dust episodes had lower $PM_{2.5}$ MSE (0.7 m^2 g^{-1}) and anthropogenic pollution
766 episodes had higher $PM_{2.5}$ MSE (4.3 m^2 g^{-1}) (Pu et al., 2015).

767 In YRD, annual average $PM_{2.5}$ MSE ranged from 3.8 m^2 g^{-1} in Ningbo to 5.3 m^2
768 g^{-1} in Hangzhou with a regional urban average (including cities of Nanjing, Shanghai,
769 Suzhou, Hangzhou and Ningbo) of 4.1 m^2 g^{-1} in 2011-2012 (Cheng et al., 2013b).
770 $PM_{2.5}$ MSE in Lin'an (4.0 m^2 g^{-1}), a rural site of YRD, was close to the regional
771 urban average value in YRD (Xu et al., 2002). $PM_{2.5}$ MSE in Shanghai reached 5.6
772 m^2 g^{-1} in winter of 2012 (Han et al., 2015), which was higher than that in Beijing in
773 the same season (Tao et al., 2015a).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

774 In PRD, annual average $PM_{2.5}$ MSE in Guangzhou was 3.5 m^2 g^{-1} with seasonal
775 average ranged from 2.3 m^2 g^{-1} in summer to 4.5 m^2 g^{-1} in autumn in 2009-2010 (Tao
776 et al., 2014c). These values were close to 4.2 m^2 g^{-1} (Andreae et al., 2008) and 2.7 m^2
777 g^{-1} (Jung et al., 2009a) measured in the same city in autumn of 2004. However, $PM_{2.5}$
778 MSE in rural Guangzhou (Wanqingsha, south of Guangzhou) was 5.3 m^2 g^{-1} (Wang
779 et al., 2012), which was evidently higher than that in Guangzhou in the same season
780 (Tao et al., 2014c).

781 In southwest China, seasonal average $PM_{2.5}$ MSE ranged from 3.5 to 4.4 m^2 g^{-1}
782 in Chengdu in 2011 (Tao et al., 2014b). In Northwest China, $PM_{2.5}$ MSE was 3.0 m^2
783 g^{-1} for anthropogenic pollution and 1.0 m^2 g^{-1} for dust pollution at a remote site
784 (Yulin, located at the interface of the desert and loess regions, Shanxi province),
785 which was similar to what was observed in rural Beijing (Xu et al., 2004).

786 In summary, annual $PM_{2.5}$ MSE mostly ranged from 3.5 to 5.9 m^2 g^{-1} in urban
787 areas in China with higher values in north China and lower values in south China.
788 Seasonal average $PM_{2.5}$ MSE mostly ranged from 2.3 to 5.6 m^2 g^{-1} with higher
789 values in winter and autumn and lower values in spring and summer. Generally,
790 $PM_{2.5}$ MSE mostly ranged from 3.0 to 5.0 m^2 g^{-1} for anthropogenic pollution and
791 from 0.7 to 1.0 m^2 g^{-1} for natural dust aerosols.

792 **4.2 Mass absorption efficiency of EC and organic matter**

793 EC is the dominant absorption species in $PM_{2.5}$. Similar to $PM_{2.5}$ MSE, the slope
794 between b_{ap} and EC mass was defined as mass scattering efficiency (MAE) of EC.
795 Various instruments have been used to measure b_{ap} including Aethalometer,

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

multi-angle absorption photometer (MAAP), Radiance Research Particle Soot Absorption Photometer (PSAP), and Photoacoustic Spectrometer (PAS), with the former two instruments measuring attenuation of the sample on the filter for estimating BC mass concentration, and the latter two measuring b_{ap} directly. Most studies in China used Aethalometer and MAAP. BC mass concentrations (880nm) were converted to b_{ap} (532nm) by an empirical constant of 8.28 m^2 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 m^2 g^{-1} in winter and 9.4 m^2 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 the higher ambient humidity (Wu et al., 2016). BC MAE was 6.5 m^2 g^{-1} at 532 nm in autumn in Shenzhen of PRD (Lan et al., 2013). However, BC MAE was 12.4 m^2 g^{-1} at 532 nm in winter in Xi'an (Wang et al., 2014a). Moreover, EC MAE of diesel was 8.4 m^2 g^{-1} (632 nm), which was higher than those (3.0-6.8 m^2 g^{-1}) of biomass burning sources (e.g. crop residual and wood) (Cheng et al., 2011).

Organic matter or brown carbon has also been found to be strong light absorption materials at the short wavelength. Available MAE values of OC include 0.76 m^2 g^{-1} (532 nm) in autumn in 2008 in Guangzhou. Moreover, available MAE values of WSOC include 1.79 and 0.71 m^2 g^{-1} (365nm) in winter and summer, respectively, in

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Beijing (Cheng et al., 2011). The WSOC MAEs of wood, grass, corn, and diesel tractor were 0.97, 0.90, 1.05, and 1.33 m^2 g^{-1} (365nm), respectively, which were much higher than that of gasoline motorcycle (0.20 m^2 g^{-1}, 365nm) (Du et al., 2014b). Evidently, the MAEs of OC or WSOC should not be neglected for short wavelength absorption.

4.3 Aerosol hygroscopic properties

b_{sp} under ambient condition can differ significantly from that under dry condition 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(RH)) as a function of RH: $f(RH)=1+a\times(RH/100)^b$. 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 S3 of the supplement document). Three different methods have been used to obtain f(RH). The first one measures simultaneously dry and wet b_{sp} using nephelometer and visibility meter, respectively. The second one measures wet b_{sp} by integrating 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(RH) curves in China are summarized in Fig. 12. The three f(RH) curves in autumn of 2007, 2011 and 2014 in urban Beijing were all measured using the first method (Fig.12 a) (Liu et al., 2013b; Liu et al., 2013c; Yang et al., 2015). The two f(RH) curves measured in 2011 and 2014 were quite close, but the one in

2007 was lower under RH< 80% and higher under RH>80%, likely due to aerosol chemical composition changes in these years. The *f*(RH) curves in spring in two Tianjin rural sites (Baodi and Wuqing) were measured using the second method (Fig.12 b) (Chen et al., 2014a; Pan et al., 2009). The hygroscopic chemical components are mostly water-soluble inorganic salts (e.g. $(NH_4)_2SO_4$, NH_4NO_3), while mineral dust and organic matter are mostly hydrophobic. The concentrations of $(NH_4)_2SO_4$ and NH_4NO_3 were higher during the polluted episode than during the clean period or dust storm episodes, which resulted in higher *f*(RH) values during the polluted episode in spring in Baodi. *f*(RH) values measured in winter in Wuqing were evidently higher than those measured in spring in Baodi under RH<80% likely due to more hygroscopic chemical components in winter in Wuqing.

The *f*(RH) curves in spring at a rural site Lin'an of Zhejiang province were also measured using the second method (Fig.12 c) (Zhang et al., 2015b). Similar to what was found in Baodi as discussed above, *f*(RH) values during the polluted episodes were also higher than those during the dust influenced episode in Lin'an, but differences between polluted and dust periods were smaller in Lin'an than in Baodi. Noticeably, the *f*(RH) values during the polluted episode were similar in Lin'an and Baodi, e.g. *f*(RH=80%) was 1.5 and 1.6, respectively, in Lin'an and Baodi.

The *f*(RH) curves (solid lines) in summer in urban Guangzhou were measured by the first method, while those (dot lines) in autumn in rural Guangzhou and in summer and autumn seasons in urban Guangzhou were measured by the third method (Fig.12 d) (Cheng et al., 2008b; Lin et al., 2014; Liu et al., 2008). *f*(RH=80%)

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

862 values were 2.04 and 2.68, respectively, for urban aerosols originated from air
863 masses in the north and marine aerosols originated from the South China Sea.
864 *f*(RH<80%) curves were similar in urban and rural Guangzhou; however,
865 *f*(80%<RH<90%) values in rural Guangzhou were evidently higher than those in
866 urban Guangzhou.

867 If averaging all available *f*(RH) curves shown in Figure 13, the empirical fitting
868 parameters *a* and *b* were found to be 2.87±0.03, 5.50±0.06, respectively (Fig 13a).
869 But if excluding dust episodes in Baodi and Lin'an (Fig 13 b), the empirical fitting
870 parameters *a* and *b* were 3.17±0.03, 5.54±0.06, respectively (Figure 15b). Based on
871 the average *f*(RH) curve, b_{sp} under ambient condition (RH=80%) can be amplified by
872 about 1.8 times of that under dry conditions in China. This suggests that reducing
873 inorganic water-soluble salts is critical in alleviating hazy weather in China.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

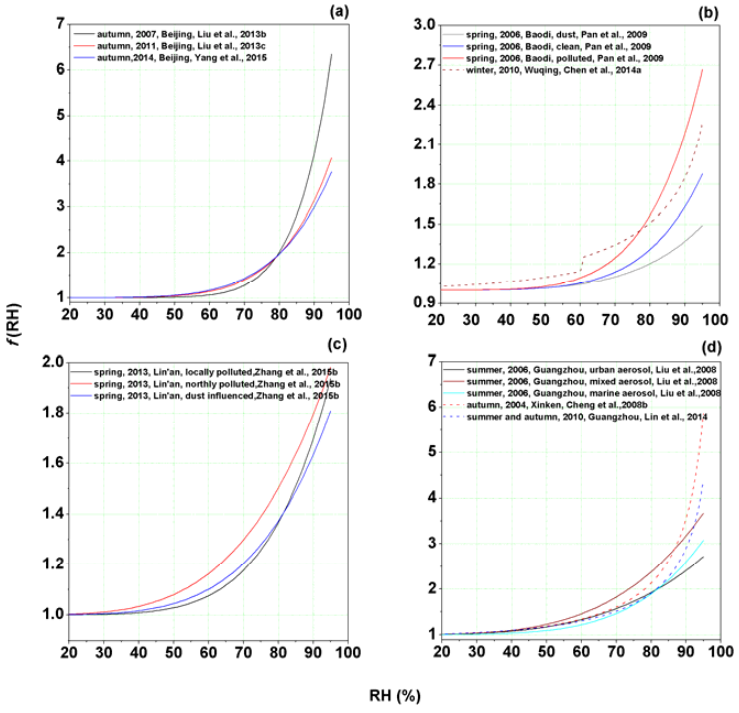


Fig. 12. The hygroscopic growth curves in different sites in China.

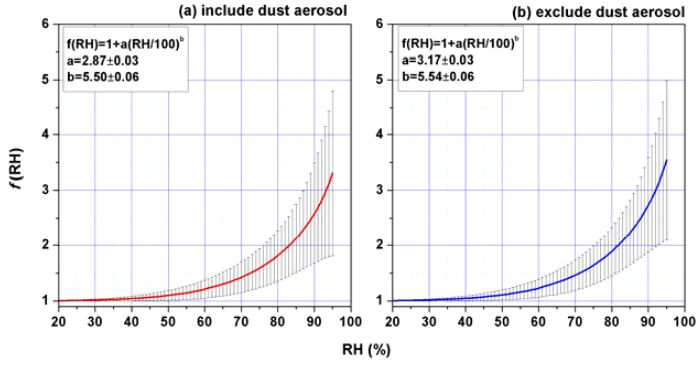


Fig. 13. Distribution of the hygroscopic growth curves in China.

4.4 Source apportionment of haze in China

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

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

883 $b_{ext} \approx 2.2\times f_S \times$ [Small $(NH_4)_2SO_4$] + 4.8 × f_L × [Large $(NH_4)_2SO_4$] + 2.4 × f_S ×
884 [Small NH_4NO_3] + 5.1 × f_L ×[Large NH_4NO_3] + 2.8 × [Small OM] + 6.1 × [Large OM]
885 + 1.0 × [Other]+ 10 × [EC] (1)

886 [Large X] = [Total X] ^{2} / 20, for [Total X] < 20 (2)

887 [Large X] = [Total X], for [Total X] ≥ 20 (3)

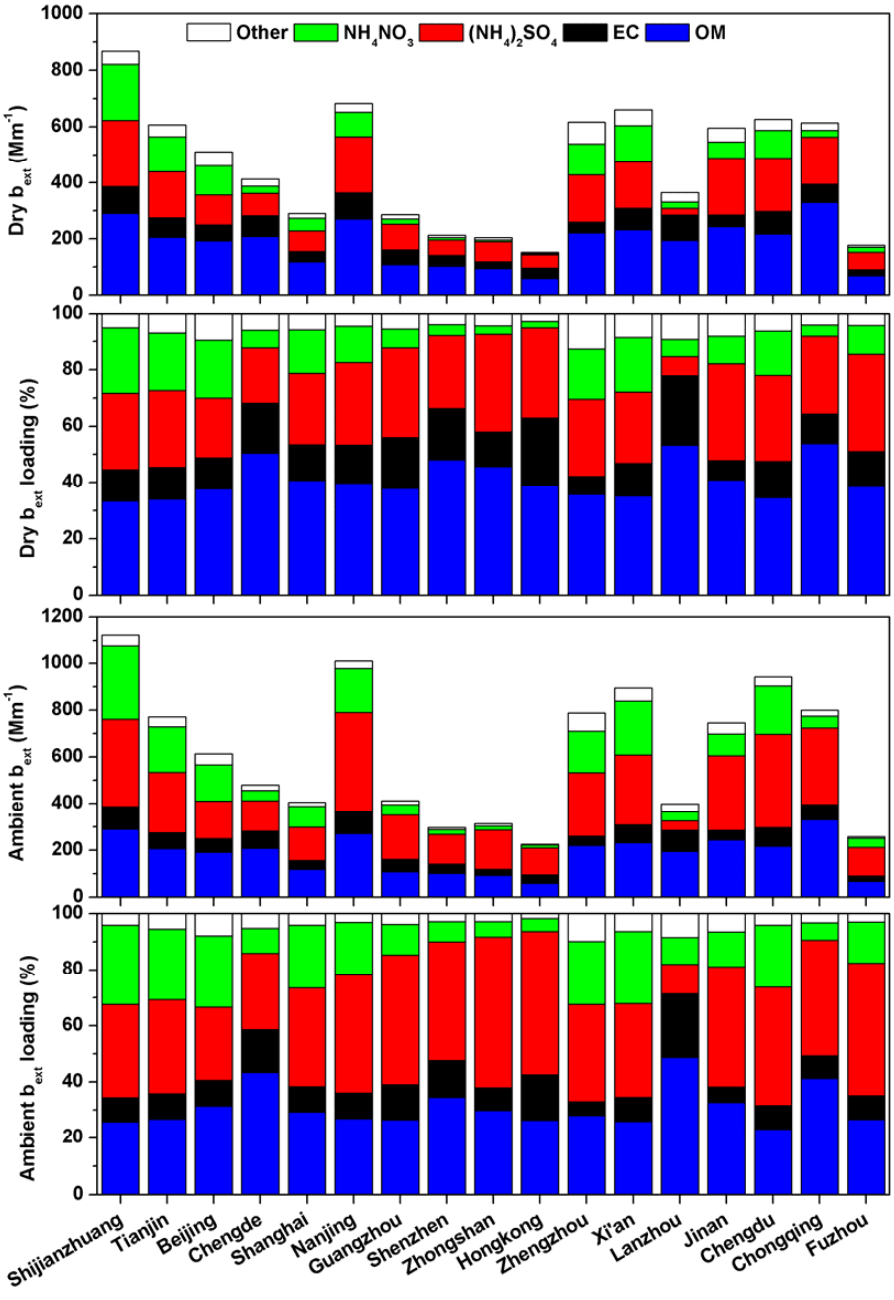
888 [Small X] = [Total X] - [Large X] (4)

889 Here, RH growth curves of f_S and f_L of $(NH_4)_2SO_4$ and NH_4NO_3 can be referred to
890 (Pitchford et al., 2007).

891 Using the chemical composition data shown in Fig. 4 and annual average RH
892 values in major cities in China as input (http://data.cma.cn/), the estimated annual b_{ext}
893 and its load percentages under dry and ambient conditions are plotted in Fig. 14. For
894 b_{ext} under dry condition, carbonaceous aerosols had similar percentage contributions
895 to secondary inorganic aerosols in Shijiazhuang, Tianjin, Beijing, Zhengzhou, Xi'an,
896 Jinan, Chengdu and Fuzhou, but higher by 11-65% in other cities. However, under
897 ambient conditions the contributions of secondary inorganic aerosols were evidently
898 higher (by 2-33%) than of carbonaceous aerosols in most cities except in Chengde,
899 Lanzhou and Chongqing. Noticeably, the contributions of secondary inorganic
900 aerosols for b_{ext} sharply increased by about 18-22% under ambient conditions than dry
901 condition in humid (RH>70%) cities (e.g. Nanjing, cities in PRD, and Chengdu).

902

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017



903

904 Fig. 14. Annual b_{ext} percentage loading under dry and ambient conditions at

905 urban sites in China.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-271, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 18 April 2017

Atmospheric Chemistry and Physics Discussions

5. Implications for aerosol pollution controls

There is no doubt that reduction of $PM_{2.5}$ mass concentration will be the ultimate approach for improving visibility and alleviating hazy weather. Industrial emission induced secondary inorganic aerosols were the most dominant sources of $PM_{2.5}$ in urban areas in China (Liang et al., 2016). Aerosols produced from biomass burning and soil dust were also important sources in north China in all the seasons except summer. Secondary inorganic aerosols were formed from SO_2 and NO_x, which were mainly emitted from coal combustion, as coal has been the major energy source in China for decades.

A series of regulations controlling coal combustion has been made since the first version NAAQS promulgated in 1982. The Air Pollution Prevention law of PRC 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 SO_2 emissions. The measure for gross control of SO_2 emissions was enforced since 1996. Despite these efforts, the amount of SO_2 emissions increased about 28% in 2005 compared with that in 2000 (http://www.zhb.gov.cn/). The amount of SO_2 emission began to decrease in 2006 and gradually reduced to the emission level of 2000 in 2010 (http://www.zhb.gov.cn/). Meanwhile, ambient annual SO_2 concentration in urban cities in China also decreased from 57 μg m^{-3} in 2005 to 40 μg m^{-3} in 2010

928 (http://www.zhb.gov.cn/). Apparently, the emission controlling efforts for reducing
929 SO_2 emissions were effective after 2006.

930 The control measures for NO_2 only began with the control of vehicle emissions
931 in 1995, but the inclusion of NO_2 in the gross control indexes did not happen until
932 2010. New coal power plants were also required to denitrate after 2010. The
933 emissions of NO_x actually increased from 1996 to 2010, as is also seen in vertical
934 column NO_2 derived from satellite data (Zhang et al., 2012b). Although annual
935 average ambient NO_2 at surface level fluctuated from 30-40 μg m^{-3} during
936 2000-2010 in China (http://www.zhb.gov.cn/), annual average ambient NO_2 in
937 megacities (e.g. Beijing, Shanghai and Guangzhou) slowly increased. Evidently, the
938 control of emissions of nitrate gaseous precursors was not very effective during
939 2000-2010.

940 Despite the above-mentioned control measures, sulfate remained at high levels
941 and nitrate even gradually increased in megacities in China. More recently, the Clean
942 Air Action Plan (CAAP) for improving the air quality was promulgated and
943 implemented by the State Council of the People's Republic of China in 2013
944 (http://www.gov.cn). This plan aims to reduce $PM_{2.5}$ annual mass concentrations by
945 25%, 20%, and 15% of the 2012 levels in BTH, YRD, and PRD, respectively. The
946 key industries including power plant, iron and steel smelting industry, petroleum
947 chemical industry, cement industry, nonferrous metals smelting industry, and
948 chemical industry were required to execute stricter emission standards in the key
949 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 non-linearity of chemistry (Cheng et al., 2016). For example, a modeling sensitivity study suggested potential increase in NO_3^- mass concentrations due to the increased atmospheric oxidizability, even under NO_x emissions decreasing conditions (Zhao et al., 2013a). Furthermore, increased atmospheric oxidizability may also enhance the conversion of VOCs 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 intension is the ammonia emissions from agricultural activities in rural areas and human activities in cities. Ammonia emission can enhance $PM_{2.5}$ pollution substantially, especially in ammonia-limited (acid aerosols) areas (Wang et al., 2011), and this topic needs further investigation through both modeling simulation and field observations.

For cleaning the atmosphere across China, the following recommendations are provided based on the major chemical components contributing to $PM_{2.5}$ and their impact of aerosol optical properties. Emissions produced from coal combustion, both in industrial sectors and in residential areas, need to be further reduced. While advanced technology should be adopted in the medium term in major industrial sectors consuming coal, cleaner energy sources should be considered for the

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 not only reduce coal emissions but also biomass burning emissions. Improving fertilizer use efficiency in agriculture is needed in reducing nitrogen emissions especially ammonia gas (Behera et al., 2013). Educating public to reduce meat consumption in daily life, especially in the developed regions with high living standard, can reduce substantially nitrogen footprint and thus nitrogen emission (Galloway et al., 2014), besides gaining human body health benefits. Traffic emissions in megacities may also need to be constrained such as developing more efficient public transportation systems and limit personal automobiles. Planting more trees and other vegetation such as the continued expansion of the three northern region shelter forests in north China can reduce dust emissions and increase atmospheric removal of aerosols through dry deposition process (Zhang et al., 2017). Having more vegetation coverage is especially important for arid or semi-arid areas as well as for urban areas in reducing dust emissions (Baldauf, 2017), besides biological benefits.

Acknowledgements

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

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, Atmospheric Environment, 42, 6335-6350, 2008.

Baldauf, R.: Roadside vegetation design characteristics that can improve local, near-road air quality, Transportation Research Part D: Transport and Environment, 52A, 354-361, 2017.

Behera, S. N., Sharma, M., Aneja, V. P., Balasubramanian, R.: Ammonia in the atmosphere: A review

997 on emission sources, atmospheric chemistry and deposition on terrestrial bodies, Environmental
998 Science and Pollution Research, 20, 8092-8131, 2013.
999 Bergin, M. H., Cass, G. R., Xu, J., Fang, C., Zeng, L., Yu, T., Salmon, L. G., Kiang, C. S., Tang, X. Y.,
1000 and Zhang, Y.: Aerosol radiative, physical, and chemical properties in Beijing during June 1999,
1001 Journal of Geophysical Research, 106, 17969-17980, 2001.
1002 Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light
1003 absorption due to mixing state, Journal of Geophysical Research, 111, 2006.
1004 Cao, J., Wang, Q., Chow, J. C., Watson, J. G., Tie, X., Shen, Z., Wang, P., and An, Z.: Impacts of
1005 aerosol compositions on visibility impairment in Xi'an, China, Atmospheric Environment, 59,
1006 559-566, 2012a.
1007 Cao, J., Lee, S., Ho, K., Zhang, X., Zou, S., Fung, K., Chow, J. C., and Watson, J. G.: Characteristics of
1008 carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period, Atmospheric
1009 Environment, 37, 1451-1460, 2003.
1010 Cao, J., Lee, S., Ho, K., Zou, S., Fung, K., Li, Y., Watson, J. G., and Chow, J. C.: Spatial and seasonal
1011 variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China,
1012 Atmospheric Environment, 38, 4447-4456, 2004.
1013 Cao, J., Zhu, C., Chow, J. C., Watson, J. G., Han, Y., Wang, G., Shen, Z., and An, Z.: Black carbon
1014 relationships with emissions and meteorology in Xi'an, China, Atmospheric Research, 94, 194-202,
1015 2009.
1016 Cao, J., Shen, Z., Chow, J. C., Watson, J. G., Lee, S., Tie, X., Ho, K., Wang, G., and Han, Y.: Winter and
1017 Summer $PM_{2.5}$ Chemical Compositions in Fourteen Chinese Cities, Journal of The Air & Waste
1018 Management Association, 62, 1214-1226, 2012b.
1019 Cao, J. J., Zhu, C. S., Tie, X., Geng, F., Xu, H., Ho, S., Wang, G. H., Han, Y. M., and Ho, K. F.:
1020 Characteristics and sources of carbonaceous aerosols from Shanghai, China, Atmospheric
1021 Chemistry and Physics, 13, 803-817, 2013.
1022 Cao, J., Cohen, A. M., Hansen, J. E., Lester, R. K., Peterson, P. F., and Xu, H.: China-U.S. cooperation
1023 to advance nuclear power, Science, 353, 547-548, 2016.
1024 Chan, C. K., and Yao, X.: Air pollution in mega cities in China, Atmospheric environment, 42, 1-42,
1025 2008.
1026 Chan, C. Y., Xu, X., Li, Y. S., Wong, K. H., Ding, G. A., Chan, L. Y., and Cheng, X.: Characteristics of
1027 vertical profiles and sources of $PM_{2.5}$, PM_{10} and carbonaceous species in Beijing, Atmospheric
1028 Environment, 39, 5113-5124, 2005.
1029 Chen, J., Zhao, C. S., Ma, N., and Yan, P.: Aerosol hygroscopicity parameter derived from the light
1030 scattering enhancement factor measurements in the North China Plain, Atmospheric Chemistry
1031 and Physics, 14, 8105-8118, 10.5194/acp-14-8105-2014, 2014a.
1032 Chen, Y., Schleicher, N., Chen, Y., Chai, F., and Norra, S.: The influence of governmental mitigation
1033 measures on contamination characteristics of $PM_{2.5}$ in Beijing, Science of The Total Environment,
1034 490, 647-658, http://dx.doi.org/10.1016/j.scitotenv.2014.05.049, 2014b.
1035 Chen, Y., Xie, S., Luo, B., and Zhai, C.: Characteristics and origins of carbonaceous aerosol in the
1036 Sichuan Basin, China, Atmospheric Environment, 94, 215-223,
1037 http://dx.doi.org/10.1016/j.atmosenv.2014.05.037, 2014c.
1038 Cheng, T., Xu, C., Duan, J., Wang, Y., Leng, C., Tao, J., Che, H., He, Q., Wu, Y., and Zhang, R.:
1039 Seasonal variation and difference of aerosol optical properties in columnar and surface
1040 atmospheres over Shanghai, Atmospheric Environment, 123, 315-326, 2015.

1041 Cheng, Y., Lee, S. C., Ho, K. F., Wang, Y. Q., Cao, J. J., Chow, J. C., and Watson, J. G.: Black carbon
1042 measurement in a coastal area of South China, Journal of Geophysical Research, 111, 1-11, 2006a.
1043 Cheng, Y., Wiedensohler, A., Eichler, H., Su, H., Gnauk, T., Brüggemann, E., Herrmann, H.,
1044 Heintzenberg, J., Slanina, J., and Tuch, T.: Aerosol optical properties and related chemical
1045 apportionment at Xinken in Pearl River Delta of China, Atmospheric Environment, 42, 6351-6372,
1046 2008a.
1047 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.,
1048 and Zhang, X. L.: Mass absorption efficiency of elemental carbon and water-soluble organic
1049 carbon in Beijing, China, Atmospheric Chemistry and Physics, 11, 11497-11510, 2011.
1050 Cheng, Y., Engling, G., He, K.-B., Duan, F. K., Ma, Y. L., Du, Z. Y., Liu, J. M., Zheng, M., and Weber,
1051 R. J.: Biomass burning contribution to Beijing aerosol, Atmospheric Chemistry and Physics, 13,
1052 7765-7781, 2013a.
1053 Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H.,
1054 Zeng, L. M., Liu, S., and Gnauk, T.: Mixing state of elemental carbon and non‐light‐absorbing
1055 aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta
1056 of China, Journal of Geophysical Research, 111, 2006b.
1057 Cheng, Y. F., Wiedensohler, A., Eichler, H., Heintzenberg, J., Tesche, M., Ansmann, A., Wendisch, M.,
1058 Su, H., Althausen, D., and Herrmann, H.: Relative humidity dependence of aerosol optical
1059 properties and direct radiative forcing in the surface boundary layer at Xinken in Pearl River Delta
1060 of China: An observation based numerical study, Atmospheric Environment, 42, 6373-6397,
1061 2008b.
1062 Cheng, Y. F., Berghof, M., Garland, R. M., Wiedensohler, A., Wehner, B., Muller, T., Su, H., Zhang, Y.,
1063 Achtert, P., and Nowak, A.: Influence of soot mixing state on aerosol light absorption and single
1064 scattering albedo during air mass aging at a polluted regional site in northeastern China, Journal of
1065 Geophysical Research, 114, 2009.
1066 Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael,
1067 G., Pöschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate
1068 during haze events in China, Science Advances, 2, 10.1126/sciadv.1601530, 2016.
1069 Cheng, Z., Wang, S., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Fu, X., and Hao, J.: Long-term trend of
1070 haze pollution and impact of particulate matter in the Yangtze River Delta, China, Environmental
1071 Pollution, 182, 101-110, 2013b.
1072 Cheung, H. C., Wang, T., Baumann, K., and Guo, H.: Influence of regional pollution outflow on the
1073 concentrations of fine particulate matter and visibility in the coastal area of southern China,
1074 Atmospheric Environment, 39, 6463-6474, 2005.
1075 Cui, H., Chen, W., Dai, W., Liu, H., Wang, X., and He, K.: Source apportionment of $PM_{2.5}$ in
1076 Guangzhou combining observation data analysis and chemical transport model simulation,
1077 Atmospheric Environment, 116, 262-271, http://dx.doi.org/10.1016/j.atmosenv.2015.06.054, 2015.
1078 Dai, W., Gao, J., Cao, G., and Ouyang, F.: Chemical composition and source identification of $PM_{2.5}$ in
1079 the suburb of Shenzhen, China, Atmospheric Research, 122, 391-400,
1080 http://dx.doi.org/10.1016/j.atmosres.2012.12.004, 2013.
1081 Dan, M., Zhuang, G., Li, X., Tao, H., and Zhuang, Y.: The characteristics of carbonaceous species and
1082 their sources in $PM_{2.5}$ in Beijing, Atmospheric Environment, 38, 3443-3452, 2004.
1083 Du, Z., He, K., Cheng, Y., Duan, F., Ma, Y., Liu, J., Zhang, X., Zheng, M., and Weber, R.: A yearlong
1084 study of water-soluble organic carbon in Beijing I: Sources and its primary vs. secondary nature,

Atmospheric Environment, 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, Atmospheric Environment, 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, Atmospheric Environment, 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, Science of the Total Environment, 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, Atmospheric Environment, 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, Journal of Geophysical Research, 115, 2010.

Fang, M., Chan, C. K., and Yao, X.: Managing air quality in a rapidly developing nation: China, Atmospheric Environment, 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, Atmospheric Research, 118, 435-444, http://dx.doi.org/10.1016/j.atmosres.2012.08.012, 2012.

Feng, J., Zhong, M., Xu, B., Du, Y., Wu, M., Wang, H., and Chen, C.: Concentrations, seasonal and diurnal variations of black carbon in PM2.5 in Shanghai, China, Atmospheric Research, 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, Atmospheric Environment, 123, Part B, 288-297, http://dx.doi.org/10.1016/j.atmosenv.2015.08.019, 2015.

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, Atmospheric Research, 92, 434-442, 2009.

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, Science of The Total Environment, 497–498, 274-281, http://dx.doi.org/10.1016/j.scitotenv.2014.08.008, 2014.

Fuzzi, S., Baltensperger, U., Carslaw, K. S., Decesari, S., Der Gon, H. A. C. D. V., Facchini, M. C., Fowler, D., Koren, I., Langford, B., and Lohmann, U.: Particulate matter, air quality and climate: lessons learned and future needs, Atmospheric Chemistry and Physics, 15, 8217-8299, 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, Atmospheric Research, 155, 1-12, 2015.

Galloway, J. N., Winiwarter, W., Leip, A., Leach, A. M., Bleeker, A., Erisman, J. W.: Nitrogen

footprints: Past, present and future, Environmental Research Letters, 9, 115003, 2014.

Garland, R. M., Yang, H., Schmid, O., Rose, D., Nowak, A., Achtert, P., Wiedensohler, A., Takegawa, N., Kita, K., and Miyazaki, Y.: Aerosol optical properties in a rural environment near the mega-city Guangzhou, China: implications for regional air pollution, radiative forcing and remote sensing, Atmospheric Chemistry and Physics, 8, 5161-5186, 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, Journal of Geophysical Research, 114, 2009.

Geng, N., Wang, J., Xu, Y., Zhang, W., Chen, C., and Zhang, R.: PM2.5 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, Reviews of Geophysics, 50, 2012.

Griffith, S. M., Huang, X. H. 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 online measurements in Hong Kong, Atmospheric Environment, 122, 864-875, http://dx.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 PM_{10} at Tianjin, China, Aerosol and Air Quality Research, 10, 167-176, 2010.

Gu, J., Bai, Z., Li, W., Wu, L., Liu, A., Dong, H., and Xie, Y.: Chemical composition of PM2.5 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, Environmental Research Letters, 9, 024010, 2014.

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, Proceedings of the National Academy of Sciences of the United States of America, 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, Atmospheric Environment, 40, 3802-3815, http://dx.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 and Air Quality Research, 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, Journal of Environmental Sciences, 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, Journal of Geophysical Research, 112, 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, Atmospheric Environment, 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, Atmospheric Chemistry and Physics, 12, 1377-1395, 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, Environmental Science & Technology, 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, Journal of Geophysical Research: Atmospheres (1984–2012), 116, 2011.

He, X., Li, C., Lau, A., Deng, Z., Mao, J., Wang, M., and Liu, X.: An intensive study of aerosol optical properties in Beijing urban area, Atmospheric Chemistry and Physics, 9, 8903-8915, 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, Science of The Total Environment, 355, 276-287, 2006.

Hu, M., Wu, Z., Slanina, J., Lin, P., Liu, S., and Zeng, L.: Acidic gases, ammonia and water-soluble ions in PM2.5 at a coastal site in the Pearl River Delta, China, Atmospheric Environment, 42, 6310-6320, 2008.

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, Journal of Environmental Sciences, 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, Atmospheric Research, 104–105, 227-236, http://dx.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, Science China-earth Sciences, 57, 1352-1362, 2013.

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, Atmospheric Environment, 74, 402-411, 2013.

Ji, D., Li, L., Wang, Y., Zhang, J., Cheng, M., Sun, Y., Liu, Z., Wang, L., Tang, G., Hu, B., Chao, N., Wen, T., and Miao, H.: The heaviest particulate air-pollution episodes occurred in northern China in January, 2013: Insights gained from observation, Atmospheric Environment, 92, 546-556, http://dx.doi.org/10.1016/j.atmosenv.2014.04.048, 2014.

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, Journal of environmental management, 90, 3231-3244, 2009a.

Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Hu, M., and Sugimoto, N.: Optical properties of atmospheric aerosols obtained by in situ and remote measurements during 2006 Campaign of Air Quality Research in Beijing (CAREBeijing-2006), Journal of Geophysical Research: Atmospheres (1984–2012), 114, 2009b.

Kang, H., Zhu, B., Su, J., Wang, H., Zhang, Q., and Wang, F.: Analysis of a long-lasting haze episode in Nanjing, China, Atmospheric Research, 120, 78-87, 2013.

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, Atmospheric Research, 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 PM_{10} in four Pearl River Delta cities,South China, Journal of Environmental Sciences, 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, Atmospheric Environment, 69, 118-123, 2013.

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, Atmospheric Environment, 106, 223-231, http://dx.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, Journal of Geophysical Research, 115, 2010.

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, Atmospheric Research, 181, 288-299, http://dx.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, Atmospheric Chemistry and Physics, 11, 1621-1639, 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, Atmospheric Research, 124, 149-157, 2013a.

Li, M., and Zhang, L.: Haze in China: Current and future challenges, Environmental Pollution, 189, 85-86, 2014.

Li, P., Yan, R., Yu, S., Wang, S., Liu, W., and Bao, H.: Reinstate regional transport of $PM_{2.5}$ as a major cause of severe haze in Beijing, Proceedings of the National Academy of Sciences of the United States of America, 112, 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 and Air Quality Research, 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, Journal of Geophysical Research, 118, 12158-12167, 2013b.

1261 Li, X., He, K., Li, C., Yang, F., Zhao, Q., Ma, Y., Cheng, Y., Ouyang, W., and Chen, G.: $PM_{2.5}$ mass,
1262 chemical composition, and light extinction before and during the 2008 Beijing Olympics, Journal
1263 of Geophysical Research: Atmospheres, 118, 12,158-112,167, 2013c.
1264 Li, Y., Meng, J., Liu, J., Xu, Y., Guan, D., Tao, W., Huang, Y., and Tao, S.: Interprovincial Reliance for
1265 Improving Air Quality in China: A Case Study on Black Carbon Aerosol, Environmental Science
1266 & Technology, 50, 4118-4126, 2016b.
1267 Li, Y. J., Sun, Y., Zhang, Q., Li, X., Li, M., Zhou, Z., and Chan, C. K.: Real-time chemical
1268 characterization of atmospheric particulate matter in China: A review, Atmospheric Environment,
1269 http://dx.doi.org/10.1016/j.atmosenv.2017.02.027, 2017, in press.
1270 Li, Z., Lau, W. K., Ramanathan, V., Wu, G., Ding, Y., Manoj, M. G., Liu, J., Qian, Y., Li, J., Zhou, T.,
1271 Fan, J., Rosenfeld, D., Ming, Y., Wang, Y., Huang, J., Wang, B., Xu, X., Lee, S. S., Cribb, M.,
1272 Zhang, F., Yang, X., Takemura, T., Wang, K., Xia, X., Yin, Y., Zhang, H., Guo, J., Zhai, P. M.,
1273 Sugimoto, N., Babu, S. S., and Brasseur, G. P.: Aerosol and Monsoon Climate Interactions over
1274 Asia, Reviews of Geophysics, 54, doi:10.1002/2015rg000500, 2016c.
1275 Liang, C. S., Duan, F.-K., He, K. B., and Ma, Y. L.: Review on recent progress in observations, source
1276 identifications and countermeasures of $PM_{2.5}$, Environment International, 86, 150-170,
1277 http://dx.doi.org/10.1016/j.envint.2015.10.016, 2016.
1278 Liao, H., Chang, W., and Yang, Y.: Climatic effects of air pollutants over China: A review, Advances in
1279 Atmospheric Sciences, 32, 115-139, 2014.
1280 Lin, Y., Hsu, S., Chou, C. C. K., Zhang, R., Wu, Y., Kao, S., Luo, L., Huang, C., Lin, S., and Huang, Y.:
1281 Wintertime haze deterioration in Beijing by industrial pollution deduced from trace metal
1282 fingerprints and enhanced health risk by heavy metals, Environmental Pollution, 208, 284, 2016.
1283 Lin, Z., Zhang, Z., Zhang, L., Tao, J., Zhang, R., Cao, J., Fan, S., and Zhang, Y.: An alternative method
1284 for estimating hygroscopic growth factor of aerosol light-scattering coefficient: a case study in an
1285 urban area of Guangzhou, South China, Atmospheric Chemistry and Physics, 14, 7631-7644,
1286 2014.
1287 Liu, D., Li, J., Zhang, Y., Xu, Y., Liu, X., Ding, P., Shen, C., Chen, Y., Tian, C., and Zhang, G.: The Use
1288 of Levoglucosan and Radiocarbon for Source Apportionment of $PM_{2.5}$ Carbonaceous Aerosols at a
1289 Background Site in East China, Environmental Science & Technology, 47, 10454, 2013a.
1290 Liu, G., Li, J., Wu, D., and Xu, H.: Chemical composition and source apportionment of the ambient
1291 $PM_{2.5}$ in Hangzhou, China, Particuology, 18, 135-143,
1292 http://dx.doi.org/10.1016/j.partic.2014.03.011, 2015.
1293 Liu, J., Li, J., Zhang, Y., Liu, D., Ding, P., Shen, C., Shen, K., He, Q., Ding, X., and Wang, X.: Source
1294 Apportionment Using Radiocarbon and Organic Tracers for $PM_{2.5}$ Carbonaceous Aerosols in
1295 Guangzhou, South China: Contrasting Local- and Regional-Scale Haze Events, Environmental
1296 Science & Technology, 48, 12002-12011, 2014a.
1297 Liu, P., Zhao, C., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W., Deng, Z., Ma, N., and
1298 Mildenberger, K.: Hygroscopic properties of aerosol particles at high relative humidity and their
1299 diurnal variations in the North China Plain, Atmospheric Chemistry and Physics, 11, 3479-3494,
1300 2011.
1301 Liu, X., Cheng, Y., Zhang, Y., Jung, J., Sugimoto, N., Chang, S.-Y., Kim, Y. J., Fan, S., and Zeng, L.:
1302 Influences of relative humidity and particle chemical composition on aerosol scattering properties
1303 during the 2006 PRD campaign, Atmospheric Environment, 42, 1525-1536, 2008.
1304 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, Journal of Geophysical Research, 114, 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, Atmospheric Environment, 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, Atmospheric Research, 132–133, 91-101, http://dx.doi.org/10.1016/j.atmosres.2013.04.007, 2013b.

Liu, X., Li, J., Qu, Y., Han, T., Hou, L., Gu, J., Chen, C., Yang, Y., Liu, X., and Yang, T.: Formation and evolution mechanism of regional haze: a case study in the megacity Beijing, China, Atmospheric Chemistry and Physics, 13, 4501-4514, 2013c.

Liu, Z., Hu, B., Wang, L., Wu, F., Gao, W., and Wang, Y.: Seasonal and diurnal variation in particulate matter (PM10 and PM2.5) at an urban site of Beijing: analyses from a 9-year study, Environmental Science and Pollution Research, 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, Atmospheric Environment, 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, Science of The Total Environment, 338, 267-281, http://dx.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 SO_2, NO_x, PM_{10} and VOCs in the Pearl River Delta region from 2000 to 2009, Atmospheric environment, 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, Atmospheric Chemistry and Physics, 10, 2010.

Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, Atmospheric Chemistry and Physics, 11, 9839-9864, 2011.

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., Mildenberger, 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, Atmospheric Chemistry and Physics, 11, 5959-5973, 10.5194/acp-11-5959-2011, 2011.

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, Journal of Geophysical Research, 108, 4279, 2003.

Man, C. K., and Shih, M. Y.: Light scattering and absorption properties of aerosol particles in Hong Kong, Journal of Aerosol Science, 32, 795-804, 2001.

Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., and Bey, I.: Atmospheric composition change - Global and regional air quality, Atmospheric Environment, 43, 5268-5350, 2009.

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, Atmospheric Environment, 45, 2789-2794, 2011.

1349 Pan, X. L., Yan, P., Tang, J., Ma, J. Z., Wang, Z. F., Gbaguidi, A., and Sun, Y. L.: Observational study
1350 of influence of aerosol hygroscopic growth on scattering coefficient over rural area near Beijing
1351 mega-city, Atmospheric Chemistry and Physics, 9, 7519-7530, 10.5194/acp-9-7519-2009, 2009.
1352 Pathak, R. K., Wang, T., Ho, K., and Lee, S.: Characteristics of summertime $PM_{2.5}$ organic and
1353 elemental carbon in four major Chinese cities: implications of high acidity for water-soluble
1354 organic carbon (WSOC), Atmospheric Environment, 45, 318-325, 2011.
1355 Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., and Hand, J.: Revised algorithm for
1356 estimating light extinction from IMPROVE particle speciation data, Journal of the Air & Waste
1357 Management Association, 57, 1326-1336, 2007.
1358 Pu, W., Zhao, X., Shi, X., Ma, Z., Zhang, X., and Yu, B.: Impact of long-range transport on aerosol
1359 properties at a regional background station in Northern China, Atmospheric Research, 153,
1360 489-499, 2015.
1361 Pui, D. Y., Chen, S.-C., and Zuo, Z.: $PM_{2.5}$ in China: Measurements, sources, visibility and health
1362 effects, and mitigation, Particuology, 13, 1-26, 2014.
1363 Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K. M., Ming, J., Wang, H., Wang, M.,
1364 Warren, S. G., and Zhang, R.: Light-absorbing Particles in Snow and Ice: Measurement and
1365 Modeling of Climatic and Hydrological Impact, Advances in Atmospheric Sciences, 32, 64-91,
1366 2015.
1367 Qin, Y., and Xie, S.: Spatial and temporal variation of anthropogenic black carbon emissions in China
1368 for the period 1980–2009, Atmospheric Chemistry and Physics, 12, 4825-4841, 2012.
1369 Schnaiter, M., Linke, C., Mohler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and
1370 Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic
1371 aerosol, Journal of Geophysical Research, 110, 2005.
1372 Shen, G., Xue, M., Yuan, S., Zhang, J., Zhao, Q., Li, B., Wu, H., and Ding, A.: Chemical compositions
1373 and reconstructed light extinction coefficients of particulate matter in a mega-city in the western
1374 Yangtze River Delta, China, Atmospheric Environment, 83, 14-20,
1375 http://dx.doi.org/10.1016/j.atmosenv.2013.10.055, 2014.
1376 Shen, Z., Cao, J., Arimoto, R., Han, Z., Zhang, R., Han, Y., Liu, S., Okuda, T., Nakao, S., and Tanaka,
1377 S.: Ionic composition of TSP and $PM_{2.5}$ during dust storms and air pollution episodes at Xi'an,
1378 China, Atmospheric Environment, 43, 2911-2918, 2009.
1379 Song, Y., Xie, S., Zhang, Y., Zeng, L., Salmon, L. G., and Zheng, M.: Source apportionment of $PM_{2.5}$ in
1380 Beijing using principal component analysis/absolute principal component scores and UNMIX,
1381 Science of the Total Environment, 372, 278-286, 2006.
1382 Song, Y., Tang, X., Xie, S., Zhang, Y., Wei, Y., Zhang, M., Zeng, L., and Lu, S.: Source apportionment
1383 of PM2.5 in Beijing in 2004, Journal of Hazardous Materials, 146, 124-130, 2007.
1384 Sun, Y., Zhuang, G., Wang, Y., Han, L., Guo, J., Dan, M., Zhang, W., Wang, Z., and Hao, Z.: The
1385 air-borne particulate pollution in Beijing - concentration, composition, distribution and sources,
1386 Atmospheric Environment, 38, 5991-6004, 2004.
1387 Sun, Y., Zhuang, G., Tang, A., Wang, Y., and An, Z.: Chemical Characteristics of $PM_{2.5}$ and PM_{10} in
1388 Haze-Fog Episodes in Beijing, Environmental Science & Technology, 40, 3148, 2006.
1389 Sun, Y. L., Wang, Z. F., Fu, P., Yang, T., Jiang, Q., Dong, H., Li, J., and Jia, J.: Aerosol composition,
1390 sources and processes during wintertime in Beijing, China, Atmospheric Chemistry and Physics,
1391 13, 4577-4592, 2013.
1392 Tan, J. H., Duan, J. C., Ma, Y. L., Yang, F. M., Cheng, Y., He, K. B., Yu, Y. C., and Wang, J. W.: Source

of atmospheric heavy metals in winter in Foshan, China, Science of The Total Environment, 493, 262-270, http://dx.doi.org/10.1016/j.scitotenv.2014.05.147, 2014.

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, Journal of Environmental Sciences, 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, Atmospheric Research, 167, 24-33, http://dx.doi.org/10.1016/j.atmosres.2015.06.015, 2016.

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, Advances in Atmospheric Sciences, 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 PM2.5 in an urban environment in Chengdu, China: Importance of springtime dust storms and biomass burning, Atmospheric Research, 122, 270-283, http://dx.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, Atmospheric Chemistry and Physics, 14, 8679-8699, 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, Atmospheric Environment, 95, 552-562, http://dx.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, Atmospheric Research, 135–136, 48-58, http://dx.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, Atmospheric Environment, 110, 36-44, http://dx.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 hazy weather prevention, Science of The Total Environment, 508, 57-66, http://dx.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, Atmospheric Environment, 144, 8-16, http://dx.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, Science of The Total Environment, 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, Science of The Total Environment, 574, 1559-1570, http://dx.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, http://dx.doi.org/10.1016/j.partic.2014.03.014, 2015.

Verma, R. L., Sahu, L. K., Kondo, Y., Takegawa, N., Han, S., Jung, J., Kim, Y. J., Fan, S. J., Sugimoto, N., and Shammaa, M. H.: Temporal variations of black carbon in Guangzhou, China, in summer 2006, Atmospheric Chemistry and Physics, 10, 6471-6485, 2010.

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, Science of The Total Environment, 577, 245-257, http://dx.doi.org/10.1016/j.scitotenv.2016.10.173, 2017.

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, Journal of Cleaner Production, 112, Part 2, 1302-1311, http://dx.doi.org/10.1016/j.jclepro.2015.04.099, 2016a.

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, Atmospheric Environment, 104, 228-236, http://dx.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, Environmental Science and Pollution Research, 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, Science of The Total Environment, 508, 477-487, http://dx.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, Atmospheric Environment, 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 Science and Technology, 48, 689-697, 10.1080/02786826.2014.917758, 2014a.

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, Environmental Science & Technology, 45, 9293-9300, 2011.

Wang, S., and Hao, J.: Air quality management in China: Issues, challenges, and options, Journal of Environmental Sciences, 24, 2-13, 2012.

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, Journal of Environmental Sciences, 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, Science of The Total Environment, 573, 1031-1040, http://dx.doi.org/10.1016/j.scitotenv.2016.08.179, 2016b.

1481 Wang, Y., Zhuang, G., Tang, A., Yuan, H., Sun, Y., Chen, S., and Zheng, A.: The ion chemistry and the
1482 source of $PM_{2.5}$ aerosol in Beijing, Atmospheric Environment, 39, 3771-3784, 2005.
1483 Wang, Y., Zhuang, G., Zhang, X. Y., Huang, K., Xu, C., Tang, A. H., Chen, J. M., and An, Z.: The ion
1484 chemistry，seasonal cycle，and sources of $PM_{2.5}$ and TSP aerosol in Shanghai, Atmospheric
1485 Environment, 40, 2935-2952, 2006.
1486 Wang, Y., Ying, Q., Hu, J., and Zhang, H.: Spatial and temporal variations of six criteria air pollutants
1487 in 31 provincial capital cities in China during 2013–2014, Environment International, 73, 413-422,
1488 http://dx.doi.org/10.1016/j.envint.2014.08.016, 2014b.
1489 Wang, Z., Li, J., Wang, Z., Yang, W., Tang, X., Ge, B., Yan, P., Zhu, L., Chen, X., and Chen, H.:
1490 Modeling study of regional severe hazes over mid-eastern China in January 2013 and its
1491 implications on pollution prevention and control, Science China-earth Sciences, 57, 3-13, 2013.
1492 Watson, J. G., Chow, J. C., and Houck, J. E.: $PM_{2.5}$ chemical source profiles for vehicle exhaust,
1493 vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995,
1494 Chemosphere, 43, 1141-1151, 2001.
1495 Watson, J. G.: Visibility: Science and regulation, Journal of the Air & Waste Management Association,
1496 52, 628-713, 2002.
1497 Wu, D., Mao, J., Deng, X., Tie, X., Zhang, Y., Zeng, L., Li, F., Tan, H., Bi, X., and Huang, X.: Black
1498 carbon aerosols and their radiative properties in the Pearl River Delta region, Science in China
1499 Series D: Earth Sciences, 52, 1152-1163, 2009.
1500 Wu, D., Wu, C., Liao, B., Chen, H., Wu, M., Li, F., Tan, H., Deng, T., Li, H., and Jiang, D.: Black
1501 carbon over the South China Sea and in various continental locations in South China, Atmospheric
1502 Chemistry and Physics, 13, 12257-12270, 2013.
1503 Wu, Y., Zhang, R., Tian, P., Tao, J., Hsu, S. C., Yan, P., Wang, Q., Cao, J., Zhang, X., and Xia, X.:
1504 Effect of ambient humidity on the light absorption amplification of black carbon in Beijing during
1505 January 2013, Atmospheric Environment, 124, 217-223, 2016.
1506 Xu, J., Bergin, M., Yu, X., Liu, G., Zhao, J., Carrico, C., and Baumann, K.: Measurement of aerosol
1507 chemical, physical and radiative properties in the Yangtze delta region of China, Atmospheric
1508 Environment, 36, 161-173, 2002.
1509 Xu, J., Bergin, M. H., Greenwald, R., Schauer, J. J., Shafer, M. M., Jaffrezo, J. L., and Aymoz, G.:
1510 Aerosol chemical, physical, and radiative characteristics near a desert source region of northwest
1511 China during ACE-Asia, Journal of Geophysical Research, 109, 2004.
1512 Xu, J., Tao, J., Zhang, R., Cheng, T., Leng, C., Chen, J., Huang, G., Li, X., and Zhu, Z.: Measurements
1513 of surface aerosol optical properties in winter of Shanghai, Atmospheric Research, 109, 25-35,
1514 2012a.
1515 Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical
1516 compositions of $PM_{2.5}$ aerosol in the urban area of Fuzhou, China, Atmospheric Research,
1517 104–105, 264-272, http://dx.doi.org/10.1016/j.atmosres.2011.10.017, 2012b.
1518 Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical
1519 compositions of $PM_{2.5}$ aerosol in the urban area of Fuzhou, China, Atmospheric Research, 104,
1520 264-272, 2012c.
1521 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.,
1522 Yu, J., Liang, W. D., and Chen, L. L.: Characteristics of pollutants and their correlation to
1523 meteorological conditions at a suburban site in the North China Plain, Atmospheric Chemistry and
1524 Physics, 11, 4353-4369, 10.5194/acp-11-4353-2011, 2011.

1525 Yan, H.: Aerosol scattering properties in northern China, Atmospheric Environment, 41, 6916-6922,
1526 2007.
1527 Yan, P., Tang, J., Huang, J., Mao, J., Zhou, X., Liu, Q., Wang, Z., and Zhou, H.: The measurement of
1528 aerosol optical properties at a rural site in Northern China, Atmospheric Chemistry and Physics, 8,
1529 2229-2242, 2008.
1530 Yan, P., Pan, X., Tang, J., Zhou, X., Zhang, R., and Zeng, L.: Hygroscopic growth of aerosol scattering
1531 coefficient: A comparative analysis between urban and suburban sites at winter in Beijing,
1532 Particuology, 7, 52-60, http://dx.doi.org/10.1016/j.partic.2008.11.009, 2009.
1533 Yang, F., He, K. B., Ye, B., Chen, X., Cha, L. Z., Cadle, S. H., Chan, T., and Mulawa, P. A.: One-year
1534 record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai,
1535 Atmospheric Chemistry and Physics, 5, 1449-1457, 2005a.
1536 Yang, F., Huang, L., Duan, F., Zhang, W., He, K., Ma, Y., Brook, J. R., Tan, J., Zhao, Q., and Cheng, Y.:
1537 Carbonaceous species in $PM_{2.5}$ at a pair of rural/urban sites in Beijing, 2005–2008, Atmospheric
1538 Chemistry and Physics, 11, 7893-7903, 10.5194/acp-11-7893-2011, 2011a.
1539 Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., and Chen, G.: Characteristics of $PM_{2.5}$
1540 speciation in representative megacities and across China, Atmospheric Chemistry and Physics, 11,
1541 5207-5219, 2011b.
1542 Yang, H., Yu, J. Z., Ho, S. S. H., Xu, J., Wu, W., Wan, C. H., Wang, X., Wang, X., and Wang, L.: The
1543 chemical composition of inorganic and carbonaceous materials in $PM_{2.5}$ in Nanjing, China,
1544 Atmospheric Environment, 39, 3735-3749, 2005b.
1545 Yang, H., Chen, J., Wen, J., Tian, H., and Liu, X.: Composition and sources of $PM_{2.5}$ around the heating
1546 periods of 2013 and 2014 in Beijing: Implications for efficient mitigation measures, Atmospheric
1547 Environment, 124, Part B, 378-386, http://dx.doi.org/10.1016/j.atmosenv.2015.05.015, 2016.
1548 Yang, L., Zhou, X., Wang, Z., Zhou, Y., Cheng, S., Xu, P., Gao, X., Nie, W., Wang, X., and Wang, W.:
1549 Airborne fine particulate pollution in Jinan, China: concentrations, chemical compositions and
1550 influence on visibility impairment, Atmospheric Environment, 55, 506-514, 2012.
1551 Yang, Y., Liu, X., Qu, Y. Z., An, J., Jiang, R., Zhang, Y., Sun, Y., Wu, Z. J., Zhang, F., and Xu, W. Q.:
1552 Characteristics and formation mechanism of continuous hazes in China: a case study during the
1553 autumn of 2014 in the North China Plain, Atmospheric Chemistry and Physics, 15, 8165-8178,
1554 2015.
1555 Ye, B., Ji, X., Yang, H., Yao, X., Chan, C. K., Cadle, S. H., Chan, T., and Mulawa, P. A.: Concentration
1556 and chemical composition of $PM_{2.5}$ in Shanghai for a 1-year period, Atmospheric Environment, 37,
1557 499-510, 2003.
1558 Yu, H., Wu, C., Wu, D., and Yu, J.: Size distributions of elemental carbon and its contribution to light
1559 extinction in urban and rural locations in the pearl river delta region, China, Atmospheric
1560 Chemistry and Physics, 10, 5107-5119, 2010.
1561 Zha, S., Cheng, T., Tao, J., Zhang, R., Chen, J., Zhang, Y., Leng, C., Zhang, D., and Du, J.:
1562 Characteristics and relevant remote sources of black carbon aerosol in Shanghai, Atmospheric
1563 Research, 135, 159-171, 2014.
1564 Zhang, F., Zhao, J., Chen, J., Xu, Y., and Xu, L.: Pollution characteristics of organic and elemental
1565 carbon in $PM_{2.5}$ in Xiamen, China, Journal of Environmental Sciences, 23, 1342-1349, 2011a.
1566 Zhang, F., Xu, L., Chen, J., Yu, Y., Niu, Z., and Yin, L.: Chemical compositions and extinction
1567 coefficients of $PM_{2.5}$ in peri-urban of Xiamen, China, during June 2009-May 2010, Atmospheric
1568 Research, 106, 150-158, 2012a.

1569 Zhang, F., Wang, Z., Cheng, H., Lv, X., Gong, W., Wang, X., and Zhang, G.: Seasonal variations and
1570 chemical characteristics of $PM_{2.5}$ in Wuhan, central China, Science of The Total Environment,
1571 518–519, 97-105, http://dx.doi.org/10.1016/j.scitotenv.2015.02.054, 2015a.
1572 Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and
1573 Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze
1574 River Delta of China, Atmospheric Chemistry and Physics, 15, 8439-8454,
1575 10.5194/acp-15-8439-2015, 2015b.
1576 Zhang, Q., Geng, G., Wang, S., Richter, A., and He, K.: Satellite remote sensing of changes in NO_x
1577 emissions over China during 1996–2010, Chinese Science Bulletin, 57, 2857-2864,
1578 10.1007/s11434-012-5015-4, 2012b.
1579 Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and
1580 Shen, Z.: Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal
1581 perspective, Atmospheric Chemistry and Physics, 13, 7053-7074, 10.5194/acp-13-7053-2013,
1582 2013a.
1583 Zhang, R., Li, Q., and Zhang, R.: Meteorological conditions for the persistent severe fog and haze
1584 event over eastern China in January 2013, Science China Earth Sciences, 57, 26-35,
1585 10.1007/s11430-013-4774-3, 2014a.
1586 Zhang, T., Cao, J., Tie, X., Shen, Z., Liu, S., Ding, H., Han, Y., Wang, G., Ho, K., and Qiang, J.:
1587 Water-soluble ions in atmospheric aerosols measured in Xi'an, China: seasonal variations and
1588 sources, Atmospheric Research, 102, 110-119, 2011b.
1589 Zhang, T., Cao, J., Chow, J. C., Shen, Z., Ho, K., Ho, S. S. H., Liu, S., Han, Y., Watson, J. G., and Wang,
1590 G.: Characterization and seasonal variations of levoglucosan in fine particulate matter in Xi'an,
1591 China, Journal of The Air & Waste Management Association, 64, 1317-1327, 2014b.
1592 Zhang, X., Wu, L., Zhang, R., Deng, S., Zhang, Y., Wu, J., Li, Y., Lin, L., Li, L., Wang, Y., and Wang,
1593 L.: Evaluating the relationships among economic growth, energy consumption, air emissions and
1594 air environmental protection investment in China, Renewable and Sustainable Energy Reviews, 18,
1595 259-270, http://dx.doi.org/10.1016/j.rser.2012.10.029, 2013b.
1596 Zhang X., Du J., Huang T., Zhang L., Gao H., Zhao Y., and Ma J.: Atmospheric removal of $PM_{2.5}$ by
1597 man-made Three Northern Regions Shelter Forest in Northern China estimated using satellite
1598 retrieved $PM_{2.5}$ concentrations, Science of the Total Environment, 593-594, 713-721, 2017.
1599 Zhao, B., Wang, S., Wang, J., Fu, J. S., Liu, T., Xu, J., Fu, X., and Hao, J.: Impact of national NO_x and
1600 SO_2 control policies on particulate matter pollution in China, Atmospheric Environment, 77,
1601 453-463, 2013a.
1602 Zhao, B., Wang, S. X., Liu, H., Xu, J., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and
1603 Amann, M.: NO_x emissions in China: historical trends and future perspectives, Atmospheric
1604 Chemistry and Physics, 13, 9869-9897, 2013b.
1605 Zhao, M., Huang, Z., Qiao, T., Zhang, Y., Xiu, G., and Yu, J.: Chemical characterization, the transport
1606 pathways and potential sources of $PM_{2.5}$ in Shanghai: Seasonal variations, Atmospheric Research,
1607 158, 66-78, 2015a.
1608 Zhao, M., Qiao, T., Huang, Z., Zhu, M., Xu, W., Xiu, G., Tao, J., and Lee, S.: Comparison of ionic and
1609 carbonaceous compositions of $PM_{2.5}$ in 2009 and 2012 in Shanghai, China, Science of The Total
1610 Environment, 536, 695-703, http://dx.doi.org/10.1016/j.scitotenv.2015.07.100, 2015b.
1611 Zhao, P., Dong, F., He, D., Zhao, X., Zhang, X., Zhang, W., Yao, Q., and Liu, H.: Characteristics of
1612 concentrations and chemical compositions for $PM_{2.5}$ in the region of Beijing, Tianjin, and Hebei,

China, Atmospheric Chemistry and Physics, 13, 4631-4644, 2013c.

Zhao, X., Zhang, X., Pu, W., Meng, W., and Xu, X.: Scattering properties of the atmospheric aerosol in Beijing, China, Atmospheric Research, 101, 799-808, 2011.

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 NH_3 emissions inventory and its implications with precipitation over the Pearl River Delta region, China, Atmospheric Chemistry and Physics, 12, 7041-7058, 10.5194/acp-12-7041-2012, 2012.

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, Environmental Pollution, 197, 36-42, http://dx.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, Atmospheric Environment, 106, 305-317, http://dx.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, Atmospheric Research, 160, 59-67, 2015.

Zieger, P., Fierzschmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, Atmospheric Chemistry and Physics, 13, 10609-10631, 2013.