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

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Abstract

To obtain a thorough knowledge of $PM_{2.5}$ chemical composition and its impact on aerosol optical properties across China, existing field studies conducted after the year 2000 are reviewed and summarized in terms of geographical, inter-annual, and seasonal distributions. Annual $PM_{2.5}$ was up to six times of the National Ambient Air Quality Standards (NAAQS) in some megacities in northern China. Annual $PM_{2.5}$ was higher in northern than southern cities, and higher in inland than coastal cities. In a few cities with data longer than a decade, $PM_{2.5}$ showed a slight decrease only in the second half of the past decade, while carbonaceous aerosols decreased, sulfate (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 typically observed in the cold seasons. Annual average contributions of secondary inorganic aerosols to $PM_{2.5}$ ranged from 25% to 48%, and those of carbonaceous aerosols ranged from 23% to 47%, both with higher contributions in southern regions due to the frequent dust events in northern China. Source apportionment analysis identified secondary inorganic aerosols, coal combustion, and traffic emission as the top three source factors contributing to $PM_{2.5}$ mass in most Chinese cities, and the sum of these three source factors explained 44% to 82% of $PM_{2.5}$ mass on annual average across China. Biomass emission in most cities, industrial emission in industrial cities, dust emission in northern cities, and ship emission in coastal cities are other major source factors, each of which contributed 7-27% to $PM_{2.5}$ mass in applicable cities.

The geographical pattern of scattering coefficient (b_{sp}) was similar to that of $PM_{2.5}$, and that of aerosol absorption coefficient (b_{ap}) was determined by elemental carbon (EC) mass concentration and its coating. b_{sp} in ambient condition of RH=80% can be amplified by about 1.8 times of that under dry condition. Secondary inorganic aerosols accounted for about 60% of aerosol extinction coefficient (b_{ext}) at relative humidity (RH) greater than 70%. The mass scattering efficiency (MSE) of $PM_{2.5}$ ranged from 3.0 to 5.0 $m^2\ g^{-1}$ for aerosols 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 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 emissions policies.

Contents

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 has also been increased significantly due to light extinction of atmospheric aerosols, especially $PM_{2.5}$ (Li and Zhang, 2014; Pui et al., 2014; Watson, 2002). The Ministry of Environmental Protection of China thus promulgated NAAQS to include daily and annual $PM_{2.5}$ standards starting in early 2012. As a result, real-time $PM_{2.5}$ data in 74 Chinese cities have been recorded since 2013.

Light extinction, the sum of light scattering and absorption, is controlled by not only $PM_{2.5}$ levels, but also its chemical composition, size-distribution and hygroscopic potential of its major components, and meteorological conditions (Hand and Malm, 2007a; Malm et al., 2003; Pitchford et al., 2007; Zhang et al., 2014a). High humidity combined with large fractions of hygroscopic chemical components (e.g. sulfate, nitrate, ammonium, and some organic matter) can enhance light extinction and haze intensity (Liu et al., 2011; Liu et al., 2013b; Zhang et al., 2015b; Zieger et al., 2013). A large number of studies has been conducted in China in recent years investigating $PM_{2.5}$ composition, aerosol optical properties, aerosol

hygroscopic properties, and haze formation mechanisms (Guo et al., 2014; Jing et al., 2015; Liang et al., 2016; Liu et al., 2011; 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, knowledge of long-term trends of $PM_{2.5}$ concentration, especially its major chemical components, is still limited (Fontes, 2017), and few studies have focused on the geographical pattern of $PM_{2.5}$ composition across China and its impact on aerosol optical properties (Li et al., 2017a). The present study aims to gain such knowledge through a thorough review of available studies.

Considering the large number of publications, only ground measurement data of chemical composition of $PM_{2.5}$, aerosol scattering and absorption coefficients, and aerosol hygroscopic properties published after the year 2000 in scientific papers of Science Citation Index (SCI) journals are reviewed and summarized in this study. A total of about 150 articles met the above criteria including 100 articles on $PM_{2.5}$ chemical composition and source apportionment, 40 articles on aerosol optical properties, and 10 articles on aerosol hygroscopic properties. Many of these articles focused on several of the biggest cities such as, Beijing, Shanghai, Guangzhou and Hong Kong, while other studies focused on cities including Tianjin, Shijiazhuang, Jinan, Nanjing, Hangzhou, Fuzhou, Xiamen, Shenzhen, Chengdu, Chongqing, Xi'an, Lanzhou, Zhengzhou, Wuhan, Changsha, Haikou and several background sites (Fig. 2). Geographical and temporal patterns of $PM_{2.5}$ and its major chemical components including SO_4^{2-}, NO_3^-, NH_4^+, organic carbon (OC), and EC, and aerosol optical properties are generated, source-apportionment analysis results are summarized, and

relationships between aerosol optical properties and $PM_{2.5}$ chemical composition are explored. Recommendations are also provided for alleviating $PM_{2.5}$ levels and reducing haze occurrence.

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

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

2.1 $PM_{2.5}$ mass

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

2.1.1 Geographical distributions

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

Within each region, the highest annual average $PM_{2.5}$ concentration was observed in Shijiazhuang (191 μg m^{-3}), Hangzhou (121 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 183±25 μg m^{-3} (Geng et al., 2013; Wang et al., 2015a; Wang et al., 2017b), 177±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), 110±4 μg m^{-3} (Xiong et al., 2017; Zhang et al., 2015a), 106 μg m^{-3} (Tang et al., 2017), 66±22 μg m^{-3} (Zhang et al., 2011a; Zhang et al.,

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

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

2.1.2 Inter-annual variations

Data collected in most cities were within a three-year time window, except in 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 trends in the 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 is likely caused by volatilization loss of nitrate and organic matters from the tapered element oscillating microbalances (TEOM), which operated at 50℃ during the online sampling. These results suggested that there was no evidence that $PM_{2.5}$ pollution has been significantly improved in Beijing during the 15 year study period despite the many control measures that have been implemented. The impact of local pollution controls in Beijing has likely been offset by regional pollutant transport (Li et al., 2015b). In Shanghai, $PM_{2.5}$ in 2003-2006 (94 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); although it decreased slightly to 58 μg m^{-3} in 2011-2013 (Wang et al., 2016a; Zhao et al., 2015b), it increased rapidly back to 95 μg m^{-3} in 2013-2014 (Ming et al., 2017). 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) were kept at stable levels and then decreased to 48 μg m^{-3} in 2014 (Tao et al., 2017).

2.1.3 Seasonal patterns

In BTH, the highest seasonal average $PM_{2.5}$ concentrations were observed in winter and the lowest in summer in all the cities with seasonal variations up to factors of 1.7, 1.5, 1.6 and 1.8 in Beijing (Cao et al., 2012b; Chan et al., 2005; Chen et al., 2014a; Dan et al., 2004; Duan et al., 2006; Han et al.,2014; He et al., 2001;

Huang et al., 2014b; Lin et al., 2016; Okuda et al., 2011; Pathak et al., 2011; Song et al., 2006a; Song et al., 2007; Sun et al., 2004; Sun et al., 2006; Tan et al., 2016a; Tao et al., 2016a; Tao et al., 2015a; Tian et al., 2015; Wang et al., 2005; Yang et al., 2005a; Yang et al., 2016; Zhang et al., 2013a; Zhao et al., 2013c), Tianjin (Cao et al., 2012b; Gu et al., 2010; Gu et al., 2011; Li et al., 2009; Tian et al., 2016; Zhao et al., 2013c), Shijiazhuang (Zhao et al., 2013c), and Chengde (Zhao et al., 2013c), respectively. It is noted that major pollutant sources in BTH were located south of Hebei province and the prevailing winds in BTH were from the north in winter and from the south in summer (Li et al., 2016b; Lu et al., 2010; 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. Moreover, extreme weather events such as weakening monsoon circulation, depression of strong cold air activities, strong temperature inversion, and descending air masses in the planetary boundary layer also played important roles in the strong $PM_{2.5}$ pollution during winter (Niu et al., 2010; Wang et al., 2014c; Zhao et al., 2013d). Several extreme wintertime air pollution events in recent years covered vast areas of northern China and were all correlated to some extent with extreme weather conditions (Zou et al., 2017).

In YRD, the highest seasonal average $PM_{2.5}$ concentrations were also observed in winter and the lowest in summer with seasonal variations up to factors of 2.3, 1.9 and 2.0 in Nanjing (Li et al., 2015a; 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., 2012a; Hou et al., 2011; Huang et al., 2014a; Huang et al., 2014b; Ming et al., 2017; 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., 2004; Cao et al., 2012b; Cui et al., 2015; Duan et al., 2007; Fu et al., 2014; Ho et al., 2006a; Huang et al., 2007; Huang et al., 2014b; Jahn et al., 2013; Jung et al., 2009; Lai et al., 2007; Lai et al., 2016; Liu et al., 2014a; Louie et al., 2005a; Tan et al., 2009; Tan et al., 2016c; Tao et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Wang et al., 2012; Yang et al., 2011b). Although the highest seasonal average $PM_{2.5}$ was also observed in winter, the season with the lowest concentration was not consistent between the sites, e.g., in summer in Guangzhou and in spring in Hong Kong. This was likely caused by warm/hot temperatures in this region and frequent precipitation in warm seasons, and thus small differences between spring and summer, e.g., $PM_{2.5}$ concentration of 32 μg m^{-3} in summer in Guangzhou (Cao et al., 2004; Cao et al., 2012b; Duan et al., 2007; Ho et al., 2006a; Lai et al., 2007; Louie et al., 2005a) and 29 μg m^{-3} in spring in Hong Kong (Louie et al., 2005a). Seasonal variations were up to a factor of 1.9 in both cities. $PM_{2.5}$ at rural sites in PRD generally doubled during dry seasons (autumn and winter) compared to wet seasons (spring and summer) due to frequent precipitation scavenging of aerosols in wet seasons (Cheung et al., 2005; Dai et al., 2013; Fu et al., 2014; Griffith et al., 2015; Hu et al., 2008; Lai et al., 2016).

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

2.2 Major chemical components of $PM_{2.5}$

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

To ensure the comparability of the data collected using different instruments, measurement uncertainties were first briefly discussed here. Most studies in China analyzed OC and EC using DRI carbon analyzer or Sunset carbon analyzer. IMPROVE is the most widely used thermal/optical protocol for OC and EC analysis for DRI analyzer while NIOSH is the one for the Sunset analyzer. OC and EC measured by the two analyzers are comparable if the same analysis protocol is used. For example, Wu et al. (2011) showed that OC from the Sunset analyzer was only 8% lower than that from the DRI analyzer, while EC was only 5% higher. However, when different protocols were used by the two analyzers, the differences were much larger, e.g., EC from NIOSH was almost 50% lower than that from IMPROVE (Chow et al.,

2010; Yang et al., 2011a). Note that OC and EC were also measured using a CHN elemental analyzer in 2001-2002 in Beijing, which uses a similar protocol to NIOSH (Duan et al., 2006). In any case, the measurement uncertainties of total carbon (TC, the sum of OC and EC) were less than 10% (Chow et al., 2010; Wu et al., 2011).

The ions including SO_4^{2-}, NO_3^- and NH_4^+ were measured by ion chromatography. Measurement uncertainties should be less than 15% in most cases under strict QA/QC procedures (Orsini et al., 2003; Trebs et al., 2004; Weber et al., 2003), but could be larger for ammonium nitrate (NH_4NO_3) since it can evaporate from the filters before chemical analysis under high temperature and low RH conditions, and this applies to both quartz fiber filter and Teflon filter (Keck and Wittmaack, 2005; Weber et al., 2003). The loss of NO_3^- due to evaporation was found to range from 4% to 84% depending on the ambient temperature (Chow et al., 2005). Although the exact magnitudes of measurement uncertainties cannot be determined for NO_3^- and NH_4^+, they are not expected to affect significantly the inter-annual variations discussed below for the three cities (Beijing, Shanghai, and Guangzhou) considering the small year-to-year temperature changes.

2.2.1 The Beijing-Tianjin-Hebei region

2.2.1.1 Inter-annual variations in Beijing

Chemically-resolved $PM_{2.5}$ data in BTH covering multiple-years are only available in Beijing and the inter-annual variations are discussed for this city below (Duan et al., 2006; He et al., 2001; Song et al., 2006a; Yang et al., 2011b; Zhang et al., 2013a; Zhao et al., 2013c). 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. TC increased from 29.8 μg m^{-3} in 1999-2000 to 32.7-35.7 μg m^{-3} in 2001-2006, and then decreased to 23.3 μg m^{-3} in 2009-2010. The nearly 30% reduction in TC in recent years in Beijing can be taken as a real trend since measurement uncertainties were believed to be around 10% as mentioned above. OC is produced from both primary emissions and secondary formation, while EC (also known as black carbon or BC) is mainly from primary emissions. The anthropogenic emission for OC and BC over the entire China showed an increasing trend in 1996-2010 (Lu et al., 2011), while BC emissions showed a slightly decreasing trend in Beijing and Tianjin in 2005-2009 (Qin and Xie, 2012). Meanwhile, BC emissions sharply increased in Hebei province in 2005-2009. The amount of BC emissions in Hebei province was much higher than the sum of those in Beijing and Tianjin (Qin and Xie, 2012). Thus, the decrease 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 the SO_2 emissions 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 emissions in Beijing continued decreasing. Surface annual SO_2 concentrations in Beijing gradually decreased from 56 μg m^{-3} to 35 μg m^{-3} during 2006-2009 (http://www.zhb.gov.cn/). Thus, the persistently 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 emissions 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 concentrations in Beijing showed a decreasing trend and fluctuated in the range of 49 - 66 μg m^{-3} during 2006-2009 (http://www.zhb.gov.cn/). There were some inconsistences between the trends of surface NO_2 concentrations and column NO_2 or NO_x emissions, likely due to the impact of photochemical reactions on surface NO_2 concentrations in urban areas. To some extent, the

317 increasing trend of NO_3^- in Beijing was likely related to the increases in NO_x
318 emissions in both Beijing and the surrounding cities or provinces.

319 Considering the potential large uncertainties in NH_4^+ measurements, its trends
320 should only be discussed qualitatively. NH_4^+ concentrations were relatively steady in
321 Beijing during 1999-2006, ranging from 5.7 to 7.3 μg m^{-3}. NH_3 emissions changed
322 little (13400-13600 Gg $year^{-1}$) before 2005 in China, and increased slightly in BTH
323 region during 2003-2010 (Zhou et al., 2015b). The small increase of NH_4^+ in
324 2009-2010 in Beijing was consistent with the NH_3 emission trend in this region
325 (Zhang et al., 2013a; Zhao et al., 2013c). Moreover, the increase of NO_3^- in Beijing
326 was also an important factor contributing to the increase of NH_4^+.

327 In summary, a decreasing trend was identified in TC and increasing trends were
328 found for SO_4^{2-}, NO_3^- and NH_4^+ in Beijing. The inter-annual variations in EC agreed
329 with the local emission trends in Beijing, but those in SO_4^{2-}, NO_3^- and NH_4^+
330 agreed more with the regional scale emission trends of their respective gaseous
331 precursors in BTH rather than the local emission trends in Beijing. Nonlinear
332 responses of concentration changes of these aerosol components to their respective
333 emission trends were found, demonstrating other important factors potentially
334 affecting aerosol formation. It is worth to note that several recent studies have
335 highlighted the important role NO_2 might play in sulfate formation in the polluted
336 environment in China (Cheng et al., 2016; Wang et al., 2016c; Xie et al., 2015a).
337 Nevertheless, the aqueous SO_2 + H_2O_2/O_3 oxidation should still be the dominant
338 mechanism in most cases, especially at a background site (Lin et al., 2017). The

aqueous SO_2 + oxygen (catalyzed by Fe (III)) reaction can also be important under heavy haze conditions in north China (Li et al., 2017b). Extensive measurements of stable oxygen are needed to confirm the relative contributions of different sulfate formation mechanisms.

2.2.1.2 Relative contributions to $PM_{2.5}$

To investigate the relative contributions of dominant chemical components to $PM_{2.5}$ mass, the measured $PM_{2.5}$ mass was reconstructed based on SO_4^{2-}, NO_3^-, NH_4^+, OM (organic matter), and EC. The conversion factor between OC and OM was 1.8 considering the prevailing biomass burning in BTH (Cheng et al., 2013a; Du et al., 2014a).

Data collected in 2009-2010 were first discussed since multiple cities in BTH have data during this period (Zhang et al., 2013a; Zhao et al., 2013c). Secondary inorganic aerosols (the sum of sulfate, nitrate and ammonium) contributed 36-39% of $PM_{2.5}$ annually in the majority of the cities, but only 25% in Chengde, a tourist city located in the northeast part of BTH and 200 kilometers away from Beijing. Generally, the percentage contribution of secondary inorganic aerosols to $PM_{2.5}$ decreased with decreasing $PM_{2.5}$ level, e.g., from Shijiazhuang to Tianjin, Beijing, and then Chengde, a phenomenon that is consistent with what was found within the same city but for different pollution levels in a winter season (Tao et al., 2015a). Carbonaceous aerosols contributed 29-32% to $PM_{2.5}$ in most cities, but as high as 45% in Chengde, and had an opposite trend to that of secondary inorganic aerosols in terms of city-to-city variations. At the rural site Shangdianzi near Beijing, secondary

361 inorganic aerosols and carbonaceous aerosols accounted for 42% and 32%,
362 respectively, of the $PM_{2.5}$ mass, which were not significantly different from those in
363 cities located south of Yanshan Mountain. The sum of secondary inorganic aerosols
364 and carbonaceous aerosols accounted for 65%-70% of the $PM_{2.5}$ mass in cities of
365 BTH.

366 In Beijing where data are available for more than a decade, secondary inorganic
367 aerosols accounted for 28% of $PM_{2.5}$ on average and ranged from 23% to 31% from
368 year to year. Carbonaceous aerosols accounted for 43% of $PM_{2.5}$ and ranged from
369 29% to 55%. Seasonal average contributions of secondary inorganic aerosols were
370 generally higher in warm seasons than in cold seasons in most cities, and an opposite
371 trend was found for carbonaceous aerosols (Fig. 4). For example, secondary
372 inorganic aerosols contributed 32%, 41%, 28% and 32% in spring, summer, autumn
373 and winter, respectively, to $PM_{2.5}$ in Beijing, while carbonaceous aerosols
374 contributed 35%, 30%, 44% and 45% (Cao et al., 2012b; Duan et al., 2006; He et al.,
375 2001; Huang et al., 2014b; Lin et al., 2016; Pathak et al., 2011; Song et al., 2006a;
376 Song et al., 2007; Sun et al., 2004; Tao et al., 2015a; Tian et al., 2015; Zhang et al.,
377 2013a; Zhao et al., 2013c). A similar seasonal trend was also observed in other BTH
378 cities, e.g., secondary inorganic aerosols accounted for 42-53% of $PM_{2.5}$ mass in
379 summer and only 15-35% in winter, while carbonaceous aerosols accounted for
380 16-34% in summer and 42- 60% in winter in Tianjin (Cao et al., 2012b; Gu et al.,
381 2011; Li et al., 2009; Zhao et al., 2013c), Shijiazhuang (Zhao et al., 2013c) and
382 Chengde (Zhao et al., 2013c). Higher carbonaceous aerosols in winter should be

related to heating activities and biomass burning in this region (Cheng et al., 2013a; Duan et al., 2004; Tao et al., 2016b; Wang et al., 2007; Yang et al., 2016).

2.2.2 The Yangtze River Delta region

2.2.2.1 Inter-annual variations in Shanghai

Chemically-resolved $PM_{2.5}$ data in YRD covering multiple-years are only available in Shanghai (Ming et al., 2017; Wang et al., 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 4.1 for EC (Fig. 3b). OC concentrations were relatively steady (14.0-14.9 μg m^{-3}) during 1999-2009, but sharply decreased to 9.9-10.1 μg m^{-3} in 2011-2014. EC varied in the range of 4.1 to 6.5 μg m^{-3} during 1999-2009, and also sharply decreased to 1.6-2.1 μg m^{-3} in 2011-2014. TC decreased from 19.5 μg m^{-3} during 1999-2009 to 11.9 μg m^{-3} in 2011-2014, or nearly 40% reduction, which was much higher than the known measurement uncertainties. Noticeable reduction of OC and EC occurred after 2010 Shanghai World Expo, which resulted in a 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). BC emissions in Jiangsu province were much higher than the sum of those in Shanghai and Zhejiang. Thus, the decreased EC concentration in Shanghai was largely a result of local emission reductions.

Annual SO_4^{2-} concentration decreased from 14.0 μg m^{-3} in 1999-2000 to the range of 11.7 μg m^{-3} to 12.5 μg m^{-3} during 2009-2014. The trend of SO_2 emission in YRD generally agreed with that across the entire 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; Zhao et al., 2013a). Annual average SO_2 concentrations in Shanghai were in the range of 45-61 μg m^{-3} during 2000-2005 and decreased by around 50% to 24-29 μg m^{-3} during 2010-2013 (http://www.zhb.gov.cn/). Note that SO_2 emissions in Shanghai only accounted for less than 20% of the total SO_2 emissions in YRD and with small annual variations. The high concentrations of SO_4^{2-} observed in Shanghai were also closely related to regional transport from north China (e.g. BTH and Shandong province) (Li et al., 2011; Wang et al., 2016a).

Annual NO_3^- concentrations in Shanghai were relatively steady (6.0-7.7 μg m^{-3}) during 1999-2009, but sharply increased to 13.3 μg m^{-3} in 2011-2014. NO_x emissions in YRD also showed an increasing trend during these years, consistent with satellite retrieved vertical column NO_2 densities during 2000-2010 (Zhang et al., 2012b; Zhao et al., 2013b). In contrast, surface-level annual NO_2 concentrations in Shanghai sharply decreased from 90 μg m^{-3} in 2000 to a range of 48-61 μg m^{-3} during 2003-2013 (http://www.zhb.gov.cn/). The inconsistency in the trends between emissions and gaseous and particulate surface air concentrations was also found in Beijing. Photochemistry and regional transport of related pollutants should be the major causes of this phenomenon.

Annual NH_4^+ concentrations decreased from 5.9 μg m^{-3} in 1999-2000 to the

427 levels of 4.1 μg m^{-3} in 2009 and then increased to 6.6 μg m^{-3} in 2011-2014. NH_3
428 emissions increased in 2000-2005 in east China (including BTH, YRD and PRD) and
429 possibly also increased in 2006-2010 due to the lack of control measures for NH_3 in
430 China (Wang et al., 2011). The recently increased NH_4^+ concentrations in Shanghai
431 were likely due to the concurrent increases of NH_3 emissions and NO_3^-
432 concentrations.

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

440 **2.2.2.2 Relative contributions to $PM_{2.5}$**

441 The chemical compositions in $PM_{2.5}$ between the cities in YRD were compared
442 between Shanghai and Nanjing due to the lack of continuous annual data in
443 Hangzhou. A conversion factor of 1.6 between OC and OM was chosen for YRD,
444 slight smaller than that (1.8) chosen for BTH considering the lower impact of
445 biomass burning on $PM_{2.5}$ in this region (Feng et al., 2006; Li et al., 2016a).
446 Secondary inorganic aerosols contributed 25-54% of $PM_{2.5}$ annually in Shanghai and
447 Nanjing, while carbonaceous aerosols contributed 28-47% (Li et al., 2016a; Wang et
448 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 (Fig.5). Larger seasonal variations were found for carbonaceous aerosols than secondary inorganic aerosols, e.g., 47% in summer and 33%-39% in other seasons in Shanghai, and ranged from 27% (spring) to 65% (autumn) in Nanjing (Cao et al., 2012b; Huang et al., 2014a; Huang et al., 2014b; Li et al., 2016a; Ming et al., 2017; 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 different local sources in YRD. The seasonal patterns of these chemical components in Shanghai were a result of both local emissions and regional transport, but in

Nanjing, the seasonal pattern was mainly determined by local emissions because it is an inland city surrounded by many industrial enterprises including power plants, petrochemical plants, and steel plants.

2.2.3 The Pearl River Delta region

2.2.3.1 Inter-annual 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 (Hagler et al., 2006; Louie et al., 2005b). Annual OC concentrations 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 concentrations decreased significantly from 11.1 μg m^{-3} in 2002-2003 to 8.3 μg m^{-3} in 2009-2010 in Shenzhen, while EC slightly increased from 2.3 μg m^{-3} to 2.7 μg m^{-3}. Apparently, the trends of EC in Guangzhou and Shenzhen were inconsistent with the BC emission trends in Guangdong province during 2005-2009, which showed a slightly decreasing trend (Qin and Xie, 2012). As a result, TC concentrations gradually decreased from 22.0 μg m^{-3} to 15.0 μg m^{-3} in Guangzhou and from 15.0 μg m^{-3} to 13.0 μg m^{-3} in Shenzhen before 2010, similar to Beijing and Shanghai. The reduction of TC was significant in Guangzhou (32%). The same

phenomenon was also observed at a suburban site of Guangzhou (Hagler et al., 2006; Lai et al., 2016).

In contrast to the TC trend, annual SO_4^{2-}, NO_3^- and NH_4^+ concentrations in Guangzhou increased from 14.7, 4.0 and 4.5 μg m^{-3} in 2002-2003 to 18.1, 7.8 and 5.1 μg m^{-3} in 2009-2010 and decreased to 9.3, 2.2 and 3.8 μg m^{-3} in 2014, respectively. Increases in the concentrations were also found in Shenzhen, e.g., from 10.0, 2.3 and 3.2 μg m^{-3} in 2002-2003 to 11.7, 2.7 and 3.5 μg m^{-3} in 2009, respectively (Hagler et al., 2006; Huang et al., 2013), and in the suburban areas of Guangzhou, e.g., from 10.4, 0.3 and 2.4 μg m^{-3} in 2002-2003 to 12.2, 2.0 and 5.2 μg m^{-3} in 2012-2013, respectively (Hagler et al., 2006; Lai et al., 2016). SO_2 emissions in Guangdong province gradually increased in the previous decade, e.g., 964, 1150, 1177 and 1258 Gg $year^{-1}$ in 2000, 2004, 2007 and 2010, respectively (Lu et al., 2010; Zhao et al., 2013a). However, SO_2 emissions in PRD decreased by more than 40% between 2005 and 2009, due to flue gas desulfurization facilities in power plants and large industrial boilers installed in this region (Lu et al., 2013). Annual average SO_2 concentrations in Guangzhou gradually increased from 45 μg m^{-3} in 2000 to 77 μg m^{-3} in 2004, and then decreased to 17 μg m^{-3} in 2014 (http://www.gzepb.gov.cn/). Thus, the increased SO_4^{2-} concentration before 2010 in Guangzhou was largely due to regional transport of pollutants from outside of PRD. The decreased SO_4^{2-} concentration in 2014 in Guangzhou was likely due to flue gas desulfurization facilities in power plants and large industrial boilers implemented across the entire Guangdong province (http://www.gdep.gov.cn/).

Meanwhile, NO_x emissions increased in Guangdong province as well as across the entire PRD, similar to the trends in BTH and YRD (Lu et al., 2013; Zhang et al., 2012b; Zhao et al., 2013b). However, annual average surface NO_2 concentrations in Guangzhou fluctuated from 61 to 73 μg m^{-3} during 2000-2007 and from 48 to 56 μg m^{-3} during 2008-2014 (http://www.gzepb.gov.cn/). An opposite trend was also found between NO_2 and NO_x emissions with the former persistently decreased while the latter increased in Guangzhou, although NO_3^- concentrations also increased. Thus, emissions as well as chemical processes both affected these ions concentrations in air. Annual NH_4^+ concentrations slightly increased about 10% before 2010 in Guangzhou and Shenzhen although NH_3 emissions changed little during 2002-2006 in PRD (Zheng et al., 2012). Thus, the slightly increased NH_4^+ concentrations, if not caused by measurement uncertainties, in Guangzhou and Shenzhen during 2002-2010 were largely due to the increased SO_4^{2-} and NO_3^-, which enhanced the conversion of NH_3 to NH_4^+.

In summary, a decreasing trend was identified in TC and increasing trends were found for SO_4^{2-}, NO_3^- and NH_4^+ in Guangzhou and Shenzhen before 2010, while all chemical components decreased after 2010 in Guangzhou. The inter-annual variations in EC were inconsistent with BC emission trends in Guangdong province. In contrast, inter-annual variations in SO_4^{2-}, NO_3^- and NH_4^+ agreed with regional scale emission trends of their respective gaseous precursors in Guangdong province rather than PRD. Similar to Beijing and Shanghai, nonlinear responses of concentration changes of these aerosol components to their respective emission

trends were also found in Guangzhou and Shenzhen.

2.2.3.2 Relative contributions to $PM_{2.5}$

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

Although many studies have been conducted in PRD, many of them were short term. Studies covering all four seasons were mainly carried out in Guangzhou and Hong Kong (Fig.6) (Andreae et al., 2008; Cao et al., 2004; Cao et al., 2012b; Cui et al., 2015; Ho et al., 2006a; Huang et al., 2014b; Jung et al., 2009; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Tan et al., 2009; Tao et al., 2009; 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 all the years of data were averaged together, secondary inorganic aerosols contributed 43%, 31%, 38% and 33% in spring, summer, autumn and winter, respectively, to $PM_{2.5}$ in Guangzhou and 45%, 25%, 46% and 37%, respectively, in Hong Kong. However, different seasonal patterns were found between Guangzhou and Hong Kong for carbonaceous aerosols. Carbonaceous aerosols contributed 34%, 37%, 35% and 34% in spring, summer, autumn and winter, respectively, to $PM_{2.5}$ in Guangzhou and 54%, 47%, 49% and 38%, respectively, in Hong Kong. Seasonal variations of OC/EC ratios ranged from 1.6 to 3.4 in Guangzhou and from 1.2 to 2.1 in Hong Kong, suggesting coal combustion and vehicle exhaust were the dominant sources in Guangzhou while vehicle exhaust was the dominant source in Hong Kong (He et al., 2008; Watson et al., 2001).

2.2.4 Other cities

Besides the cities in BTH, YRD and PRD, synchronous measurements of $PM_{2.5}$ and the dominant chemical components have also been conducted in several cities in other regions of China, mainly in the capital city of a province (e.g. Zhengzhou of Henan province (Geng et al., 2013), Xi'an of Shaanxi province (Wang et al., 2015c), Lanzhou of Gansu province (Wang et al., 2016b), Jinan of Shandong province (Yang et al., 2012), Chengdu of Sichuan province (Tao et al., 2013a; Tao et al., 2014b), Chongqing of Chongqing municipality (Yang et al., 2011b), Changsha of Hunan

province (Tang et al., 2017), Xiamen and Fuzhou of Fujian province (Xu et al., 2012b; Zhang et al., 2016) and Haikou of Hainan province (Liu et al., 2017a)). A conversion factor of 1.6 between OC and OM was chosen for Fuzhou, Xiamen and Haikou and 1.8 for other cities based on their geographical locations.

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

At an Asian continental outflow site (Penglai in Shandong province), annual average contribution of secondary inorganic aerosols to $PM_{2.5}$ reached 54% (Feng et al., 2012b), evidently higher than those in urban and inland rural sites in China, while that of carbonaceous aerosols was 31%, close to those in BTH. This finding suggested that intensive emissions of SO_2 and NO_x in China enhanced the downward transport of secondary inorganic aerosols to the Pacific Ocean.

Seasonal average contributions are only shown here for Jinan (Yang et al., 2012),

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

Seasonal average contributions of carbonaceous aerosols were evidently higher in cold seasons than in warm seasons in the three northern cities (Zhengzhou and Jinan and Lanzhou) due to heating activities and biomass burning, similar to BTH. Surprisingly, a similar seasonal pattern was also found in one coastal city Xiamen, e.g., 38% in winter versus 27-30% in other seasons. In contrast, higher seasonal contributions were found in warm season than in cold seasons in the southern coastal city (Fuzhou) and the southern island city (Haikou). No seasonal variations were

found in the southwest inland city Chengdu (29%-32%) and the central inland city Changsha (28-33%). The summed contributions of secondary inorganic aerosols and carbonaceous aerosols were evidently lower in spring than in other seasons in most of the northern cities (e.g. Jinan, Lanzhou, Zhengzhou, and BTH), likely due to the frequent spring dust storm events in northern China.

2.2.5 Summary of $PM_{2.5}$ chemical properties

Carbonaceous aerosols showed decreasing trends over the last 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 increasing trends in all of the above-mentioned megacities. Annual mass concentrations of $PM_{2.5}$, secondary inorganic aerosols, and carbonaceous aerosols showed similar spatial gradients decreasing from high to low latitude regions.

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

The highest seasonal average contributions of secondary inorganic aerosols to $PM_{2.5}$ were observed in summer in most of the northern cities, but can occur in

different seasons in southern cities. In contrast, the highest seasonal contributions of carbonaceous aerosols were observed in cold seasons in most of the northern cities, and in warm seasons in most of the southern cities. The different seasonal patterns were largely caused by heating and biomass burning in cold seasons in north China.

2.3 Source apportionment of $PM_{2.5}$

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

Commonly used receptor models in source apportionment of $PM_{2.5}$ in China include Principal Component Analysis/Absolute Principal Component Scores (PCA/APCS), Chemical Mass Balance receptor (CMB), Positive Matrix Factorization (PMF), and UNMIX and Multilinear Engine-2 model (ME-2). Among these, PMF and CMB models were the most widely used in China. Quantitative assessments of the uncertainties in using these methods are rare; studies using the same dataset collected in 2000 in Beijing and applying the above-mentioned models suggested that, while the models still identified the same dominant source factors, the relative contributions from these source factors differed by as much as 30%

669 between the different models (Song et al., 2006a, b). Similar magnitudes of
670 uncertainties could also be caused by using different biomass burning tracers despite
671 using the same receptor model (Tao et al., 2016b).

672 Major source factors identified for $PM_{2.5}$ in most Chinese cities include
673 secondary inorganic aerosols (SIA), coal combustion (COAL), biomass burning
674 (BIOM), traffic emission (TRAF), dust emission (DUST), and industrial emission
675 (INDU). Other source factors were also identified (and sometimes due to using more
676 specific source names), such as metal manufacturing (including iron and steel
677 industry, Cu smelting) in industrial cities (e.g. Dongying and Tai'an of Shandong
678 province, Nanjing, Hangzhou, Lanzhou, Chengdu, Chongqing and Changsha), and
679 sea salt and ship emissions in coastal cities (e.g. Longkou of Shandong province,
680 Nanjing, Guangzhou, Zhuhai and Hong Kong). Contributions of dominant source
681 factors to $PM_{2.5}$ are discussed below in detail on a regional basis.

682 **2.3.1 The Beijing-Tianjin-Hebei region**

683 Studies in Beijing covered multiple years and mostly used the PMF model. If
684 averaging the results from the years in 2000 (Song et al., 2006b), 2001-2004 (Zhang
685 et al., 2007), 2009-2010 (Zhang et al., 2013a), and 2012-2013 (Zíková et al., 2016),
686 the six source factors (SIA, COAL, BIOM, TRAF, DUST, and INDU) accounted for
687 31±12%, 16±4%, 12±1%, 16±13%, 12±7%, and 9±11%, respectively, of the $PM_{2.5}$
688 mass in Beijing. There was an increasing trend for SIA contributions (from 19% to
689 48%), a decreasing trend for COAL (from 19% to 11%), and a stable trend for BIOM
690 (11-12%) during 2000-2013. There was more uncertainties in identifying TRAF and

INDU than other source factors due to the differences in the source profiles.

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

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

2.3.2 The Yangtze River Delta region

Studies for one year or longer were only made in Nanjing (Li et al., 2016a) and Hangzhou (Liu et al., 2015). Metal manufacturing was identified as a source factor in

both Nanjing and Hangzhou, while ship emissions were also identified in Nanjing. Annual contributions of SIA to $PM_{2.5}$ mass reached 68% in Nanjing while all the other sources (COAL, DUST, sea salt and ship emissions, and metal manufacturing) each contributed 10% or less. In contrast, metal manufacturing, SIA, TRAF, and COAL accounted for 32%, 28%, 17%, and 13%, respectively, of the $PM_{2.5}$ mass in Hangzhou. Evidently, the contributions of SIA in Nanjing were much higher than those in Hangzhou and cities in BTH. The contributions of COAL in Nanjing and Hangzhou were similar, but were evidently lower than those in cities in BTH.

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

2.3.3 The Pearl River Delta region

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

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

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

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

757 the highest in summer due to the prevailing air masses from the South China Sea
758 (Huang et al., 2014c).

759 **2.3.4 Other cities**

760 Studies covering one year or longer were mostly conducted for provincial capital
761 cities including Jinan (Yang et al, 2013), Zhengzhou (Geng et al., 2013), Xi'an
762 (Wang et al., 2015c), Lanzhou (Wang et al, 2016b), Chengdu (Tao et al., 2014a),
763 Chongqing (Chen et al., 2017), Changsha (Tang et al., 2017), Wuhan (Xiong et al.,
764 2017), Xiamen (Zhang et al., 2016) and Haikou (Liu et al., 2017a), and for an inland
765 city Heze (Liu et al., 2017b) and a regional background site (located in Yellow River
766 Delta National Nature Reserve in Dongying city) (Yao et al., 2016) both in Shandong
767 province. Annual results were available from most studies, but were aggregated from
768 seasonal results for Wuhan and Haikou. All the sites were grouped into four regions
769 for easy discussion, i.e., northwest China (Lanzhou and Xi'an), southwest China or
770 Sichuan basin (Chengdu and Chongqing), eastern and central China (Jinan,
771 Zhengzhou, Heze, Dongying, Wuhan and Changsha), and south coastal cities
772 (Xiamen and Haikou).

773 The two northwest cities showed the same top four dominant source factors,
774 although with slightly different percentage contributions to $PM_{2.5}$ mass, e.g., 29%
775 from SIA, 19% from COAL, 17% from DUST, and 15% from TRAF in Xi'an, and
776 17% from SIA and 22% from the other three sources in Lanzhou. The lower SIA
777 contribution in Lanzhou was likely due to the dry climate inhibiting formation of
778 SIA. Similar results to those in Xi'an were also obtained in rural Xi'an, with SIA,

779 COAL, DUST and TRAF contributing 31%, 16%, 20% and 13%, respectively, to the
780 $PM_{2.5}$ mass (Wang et al., 2015c). The two southwest cities (Chengdu and Chongqing)
781 showed nearly the same source-apportionment analysis results with SIA contributing
782 just below 40% and COAL and INDU each contributing around 20% to $PM_{2.5}$ mass.
783 The same top four dominant source factors (SIA, COAL, DUST and TRAF)
784 found in the two northwest cities were also found in other capital cities (Jinan,
785 Zhengzhou, and Wuhan, except Changsha) and a medium size city (Heze) in eastern
786 and central China, which accounted for 24-55%, 14-23%, 5-26% and 5-27% of the
787 $PM_{2.5}$ mass, depending on location. In Changsha, SIA, mixed source of INDU and
788 BIOM, and DUST accounted for 60%, 27%, and 13%, respectively, to $PM_{2.5}$ mass.
789 SIA, BIOM and INDU were the most important sources, accounting for 54%, 16%,
790 and 16%, respectively, of $PM_{2.5}$ mass in a regional background site in Dongying.
791 Similar source-apportionment results were found between the two south coastal
792 cities (Xiamen and Haikou) with SIA, TRAF, DUST, COAL, and sea salt accounting
793 for 20-27%, 16-21%, 12-22%, 8-9% and 6-10%, respectively, of the $PM_{2.5}$ mass.
794 Seasonal results of source apportionment analysis are available for four seasons
795 in Jinan (Yang et al., 2013), a regional site in Dongying (Yao et al., 2016), Chengdu
796 (Tao et al., 2014a), Chongqing (Chen et al., 2017), Wuhan (Xiong et al., 2017) and
797 Haikou (Liu et al., 2017a), for summer and autumn in Tai'an of Shandong province
798 (Liu et al., 2016a) and Xi'an (Xu et al., 2016), for summer and winter in Lanzhou
799 (Tan et al., 2017), and for winter in Longkou (a coastal site in Shandong province)
800 (Zong et al., 2016).

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

2.3.5 Summary of $PM_{2.5}$ source apportionment studies

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

It should be noted that SIA chemical compounds are formed by reactions in the atmosphere involving primary emissions of gaseous precursors that can be produced

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

3. Aerosol optical properties

There were much fewer measurements of aerosol optical properties than chemically resolved $PM_{2.5}$ data in China. Data reviewed in this section are all listed in Table S3 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., 2009; 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., 2017a), 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 obtained from MODerate-resolution Imaging Spectroradiometer (MODIS) data. Satellite retrievals of AOD have been widely applied to estimate surface $PM_{2.5}$ concentrations using statistical models (Liu et al., 2005; Hu et al., 2013; Ma et al., 2014; Wang and Christopher, 2003). Although the correlation between AOD and $PM_{2.5}$ mass concentration depends on many factors, such as aerosol size distribution, refractive index, single-scattering albedo, and meteorological factors (Che et al., 2009; Guo et al., 2009b; Guo et al., 2017), the predicted $PM_{2.5}$ mass from satellite AOD data compared well with ground-level measurements (Ma et al., 2014; Xie et al., 2015b). Moreover, the spatial distributions of AOD measured using sun photometers mostly agreed with those retrieved from

satellite data (Che et al., 2014; Che et al., 2015; Liu et al., 2016b; Pan et al., 2010). Spatial distributions of annual average AOD in 2014 are shown in Fig. 11. The spatial distributions of $PM_{2.5}$ shown in Fig. 2 are similar to the patterns of AOD shown in Fig. 11. Differences in fine structures of their patterns were due to surface $PM_{2.5}$ versus column AOD comparison and spatial variations in $PM_{2.5}$ chemical composition.

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

3.1 Geographical patterns

Annual average b_{sp} and b_{ap} from ground measurements in major cities in China are plotted in Fig. 12. Most b_{sp} measurements were conducted using a nephelometer under RH<60%. The highest annual b_{sp} was in Xi'an (525 Mm^{-1}, RH<60%) (Cao et al., 2012a), followed by Chengdu (456 Mm^{-1}, RH<40%; 421 Mm^{-1}, ambient RH) (Tao et al., 2014b; Wang et al., 2017a), Guangzhou (326 Mm^{-1}, RH<70%) (Tao et al., 2014c), Beijing (309 Mm^{-1}, RH<60%) (He et al., 2009; Jing et al., 2015; Zhao et al., 2011), and Shanghai (217 Mm^{-1}, RH<60%) (Cheng et al., 2015b). Such spatial patterns were mainly due to the spatial patterns of annual $PM_{2.5}$ mass, i.e. Xi'an (177 μg m^{-3}) > Chengdu (111 μg m^{-3}) > Beijing (108 μg m^{-3}) > Shanghai (77 μg m^{-3}) >

Guangzhou (65 μg m^{-3}), and partly due to humidity conditions, e.g., Beijing versus Guangzhou. Noticeably, b_{sp} in Shouxian County was higher than those in several megacities (e.g. Beijing, Shanghai and Guangzhou), suggesting hazy weather also frequently occurred even in small cities in China (Fan et al., 2010). b_{sp} in rural Beijing was 179 Mm^{-1} (Yan et al., 2008; Zhao et al., 2011), which was much lower than that in urban Beijing, but was close to the level in Shanghai.

Annual average b_{ap} ranged from 37 to 96 Mm^{-1} with higher values observed in Chengdu and Xi'an (likely due to popular biomass burning besides large amount of coal burning) (Cao et al., 2012a; Tao et al., 2014a; Tao et al., 2014b; Wang et al., 2017a; Zhang et al., 2014b), and lower values in Shouxian and rural Beijing (Fan et al., 2010; Yan et al., 2008; Zhao et al., 2011). b_{ap} in Guangzhou was higher than that in Beijing and Shanghai despite their similar $PM_{2.5}$ EC levels, likely due to the different coating of EC in Guangzhou than in other cities. For example, the mass absorption of EC in Guangzhou was 8.5 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).

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, while $PM_{2.5}$ 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 evidently lower than that in 2005-2006. Meanwhile, annual secondary inorganic aerosols in 2009-2010 were evidently lower than that in 2005-2006. The coating by secondary inorganic aerosols likely enhanced the absorption of EC (Bond et al., 2006; Cheng et al., 2009; Yu et al., 2010).

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

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

933 b_{sp}. This is because scattering efficiency of dust aerosols was lower than that of
934 anthropogenic aerosols (Zhao et al., 2011).

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

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

951 ### 3.2.2 The Yangtze River Delta and Pearl River Delta region

952 No multi-year b_{sp} measurement data were available for exploring inter-annual
953 variations, although multi-year measurements of BC or b_{ap} were made in Shanghai

954 (YRD) and Guangzhou (PRD). Annual b_{ap} in 2011-2012 (19 Mm^{-1}) was evidently
955 lower than that in 2010 (31 Mm^{-1}) in Shanghai (Feng et al., 2014; Zha et al., 2014),
956 consistent with the trend of EC, e.g. annual concentration of EC in 2012 (2.0 μg m^{-3})
957 was only half of that in 2009 (4.1 μg m^{-3}) (Wang et al., 2016a; Zhao et al., 2015b). In
958 Guangzhou, annual b_{ap} in 2007 (51 Mm^{-1}) was also evidently lower than that in 2004
959 (90 Mm^{-1}) (Wu et al., 2009), while EC in 2006-2007 (4.0 μg m^{-3}) was similar or
960 slightly lower than that in 2002-2003 (4.4 μg m^{-3}) (Hagler et al., 2006; Huang et al.,
961 2012). Thus, the inter-annual variations in b_{ap} were mainly determined by EC trends
962 in the same cities.

963 b_{sp} and b_{ap} in winter were evidently higher than those in spring in Shanghai,
964 consistent with the seasonal patterns of $PM_{2.5}$ and EC, respectively (Fig. 13) (Cao et
965 al., 2012b; Cheng et al., 2015; Feng et al., 2014; Han et al., 2015; Huang et al.,
966 2014a; Li et al., 2013a; Pathak et al., 2011; Wang et al., 2016a; Xu et al., 2012a; Ye
967 et al., 2003; Zha et al., 2014; Zhao et al., 2015a). Similar seasonal variations were
968 found for b_{sp} and b_{ap} in the two PRD cities (Guangzhou and Hong Kong), which also
969 agreed with the patterns of $PM_{2.5}$ and EC (Andreae et al., 2008; Cao et al., 2004; Cao
970 et al., 2012b; Cui et al., 2015; Gao et al., 2015; Huang et al., 2014b; Jung et al., 2009;
971 Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tao et al.,
972 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Verma et al., 2010; Wu et
973 al., 2009; Wu et al., 2013). The highest b_{sp} and b_{ap} appeared in winter and the lowest
974 in summer with seasonal variations up to a factor of 3.1 and 17.1 for b_{sp}, 2.3 and 5.9
975 for b_{ap}, in Guangzhou and Hong Kong, respectively.

3.2.3 Other cities

In Chengdu of southwest China, the highest b_{sp} appeared in winter and the lowest in summer with seasonal variations up to a factor of 1.9, which was consistent with the seasonal pattern of $PM_{2.5}$ (Tao et al., 2014a, b). However, the highest b_{ap} appeared in spring despite the highest EC in winter (Tao et al., 2014b). One explanation could be due to the large amount of OC emitted from biomass burning in spring, 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 (126 to 183 Mm^{-1}).

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

4.1 Mass scattering efficiency of $PM_{2.5}$

b_{sp} and $PM_{2.5}$ mass concentration have been found to correlate well in numerous field studies (Andreae et al., 2008; Han et al., 2015; Hand and Malm, 2007b; Jung et al., 2009; Pu et al., 2015; Tao et al., 2014b; Tao et al., 2014c; Tao et al., 2015a; Tao et al., 2016a; Tian et al., 2015; Wang et al., 2012b; Zhao et al., 2011). A parameter

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

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

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

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

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

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

4.2 Mass absorption efficiency of EC and organic matter

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

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 an aethalometer and MAAP. BC mass concentrations (880nm) were converted to b_{ap} (532nm) using 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 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 is also a strong light absorbing material at short wavelengths. Available MAE values of OC include 0.76 m^2 g^{-1} (532 nm) in autumn in 2008 in Guangzhou (Andreae et al., 2008). Moreover, available MAE values of WSOC include 1.79 and 0.71 m^2 g^{-1} (365nm) in winter and summer, respectively, in

Beijing (Cheng et al., 2011). The WSOC MAEs of wood, grass, corn, and diesel tractors were 0.97, 0.90, 1.05, and 1.33 m^2 g^{-1} (365nm), respectively, which were much higher than that of gasoline motorcycles (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 dry conditions due to hygroscopic properties of soluble aerosol chemical components. A relationship between ambient and dry b_{sp} is thus developed for estimating ambient b_{sp} from measured dry b_{sp}, which is often described by the hygroscopic growth curve (f(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 S4 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 a nephelometer and visibility meter, respectively. The second one measures wet b_{sp} by integrating a nephelometer equipped with a humidifier, and the third one estimates dry and wet b_{sp} based on Mie theory with size-resolved chemical components.

Available f(RH) curves in China are summarized in Fig. 14. The three f(RH) curves in autumn of 2007, 2011 and 2014 in urban Beijing were all measured using the first method (Fig.14 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 and size distribution changes in these years.

The *f*(RH) curves at four rural sites were measured using the second method, including Baodi of Tianjin in spring, Wuqing of Tianjin in winter (Fig.14 b) (Chen et al., 2014b; Pan et al., 2009), Raoyang of Hebei province in summer (Wu et al., 2017), and Lin'an of Zhejiang province in spring (Fig.14 c) (Zhang et al., 2015b). It is known that 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. In Baodi in spring season, the concentrations of $(NH_4)_2SO_4$ and NH_4NO_3 and their mass fractions in fine mode particles were higher during polluted episodes than during clean periods or dust storm episodes, resulting in higher *f*(RH) values during the polluted episode. *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. In Raoyang, a different fitting curve of *f*(RH) was obtained with a much higher *f*(RH=80%) value (2.3) than in other rural sites in BTH mentioned above, likely due to higher fractions of hydrophilic components in $PM_{2.5}$ (>56%). In all the BTH sites, *f*(RH) value increased continuously with increasing RH. However, in a different study an abrupt increase in *f*(RH) at RH values of 73-81% was observed in summer in Wangdu of Hebei province due to the deliquescence of ammonium sulfate (Kuang et al., 2016). Similar to what was found in Baodi, *f*(RH) values during polluted episodes were also higher than those during dust episodes in Lin'an, but the differences between

polluted and dust periods were smaller in Lin'an than in Baodi. Noticeably, the *f*(RH) values during polluted episodes were similar in Lin'an and Baodi, e.g. *f*(RH=80%) was 1.5 and 1.6, respectively.

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.14 d) (Cheng et al., 2008b; Lin et al., 2014; Liu et al., 2008a). *f*(RH=80%) values were 2.04 and 2.68, respectively, for urban aerosols originating from the north and marine aerosols originating from the South China Sea. *f*(RH<80%) curves were similar in urban and rural Guangzhou; however, *f*(80%<RH<90%) values in rural Guangzhou were evidently higher than those in urban Guangzhou, likely due to the much higher fraction of secondary inorganic aerosols in fine mode particles in rural Guangzhou than urban Guangzhou in the dry season (Lin et al., 2014; Liu et al., 2008b).

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

4.4 Source apportionment of haze in China

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

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

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

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

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

Where X represents $(NH_4)_2SO_4$, NH_4NO_3 and OM, respectively. RH growth curves of f_S and f_L for $(NH_4)_2SO_4$ and NH_4NO_3 can be found in Pitchford et al. (2007).

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

Changsha, Xiamen, Nanjing, cities in PRD, and Chengdu).

5. Implications for aerosol pollution controls

There is no doubt that reduction of $PM_{2.5}$ will be the ultimate approach for improving visibility and alleviating hazy weather. Industrial emission contributions to secondary inorganic aerosols were the dominant sources of $PM_{2.5}$ in urban areas in China (Liang et al., 2016). Aerosols produced from traffic emissions, biomass burning and soil dust were also important sources in north China. Secondary inorganic aerosols were formed from atmospheric reactions involving SO_2 and NO_x, which were mainly emitted from coal combustion, the major energy source in China for decades.

A series of regulations controlling coal combustion emissions has been made since the first version of the NAAQS was 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 by about 28% from 2000 to 2005 (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 (http://www.zhb.gov.cn/). Apparently, the emission control efforts for reducing SO_2 emissions since 2006 have been effective.

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

Despite the above-mentioned control measures, sulfate concentrations remained high and nitrate concentrations even gradually increased in megacities in China. More recently, the Clean Air Action Plan (CAAP) for improving air quality was promulgated and implemented by the State Council of the People's Republic of China in 2013 (http://www.gov.cn). This plan aims to reduce the $PM_{2.5}$ annual mass concentrations by 25%, 20%, and 15% of the 2012 levels in BTH, YRD, and PRD, respectively. The key industries including power plants, iron and steel smelting, petroleum chemical, cement, nonferrous metals smelting, and chemical production were required to execute stricter emission standards in the key regions including most megacities in China (http://www.zhb.gov.cn). Accordingly, annual average

1199 $PM_{2.5}$ in China from online monitored data at 74 cities gradually decreased from 72
1200 μg m^{-3} in 2013 to 50 μg m^{-3} in 2015, showing some promising results from the series
1201 of control measures.

1202 One factor that needs to be considered in future pollution reduction is the
1203 non-linearity of chemistry (Cheng et al., 2016). For example, a model sensitivity
1204 study suggested potential increase in NO_3^- mass concentrations due to the increased
1205 atmospheric oxidizing capacity, even with decreasing NO_x emissions (Zhao et al.,
1206 2013a). Furthermore, increased atmospheric oxidizing capacity may also enhance the
1207 conversion of VOCs to OM. In fact, the contribution of secondary organic aerosols
1208 to $PM_{2.5}$ was also high and could increase further in typical megacities in China (He
1209 et al., 2011; Huang et al., 2014b; Sun et al., 2013). Another factor that requires more
1210 attention is ammonia emissions from agricultural activities in rural areas and human
1211 activities in cities. Ammonia emissions can enhance $PM_{2.5}$ pollution substantially,
1212 especially in ammonia-limited (acid aerosols) areas (Wang et al., 2011). This topic
1213 needs further investigation through both modeling simulation and field observations.

1214 To improve the air quality across China, the following recommendations are
1215 provided based on the major chemical components contributing to $PM_{2.5}$ and their
1216 impact on aerosol optical properties. Emissions produced from coal combustion, in
1217 both the industrial sectors and in residential areas, need to be further reduced. While
1218 advanced pollution control technologies should be adopted in the medium term in
1219 major industrial sectors consuming coal, cleaner energy sources should be
1220 considered a long-term goal (Cao et al., 2016). Providing cleaner energy to the vast

rural and urban areas in north China for heating and cooking can not only reduce coal combustion emissions but also biomass burning emissions. Efficient use of fertilizers in agriculture is needed to reduce nitrogen emissions especially ammonia (Behera et al., 2013). Educating the public to reduce meat consumption in their daily lives, especially in the more affluent developed regions, can reduce the nitrogen footprint substantially and thus nitrogen emissions (Galloway et al., 2014), besides the potential benefits to human health. Traffic emissions in megacities may also need to be constrained, such as developing more efficient public transportation systems and limiting the use of personal automobiles. Having more vegetation coverage is especially important in arid or semi-arid areas as well as urban areas in reducing dust emissions (Baldauf, 2017), aside from the biological benefits. The continued expansion of the three northern region shelter forests in north China can potentially reduce dust emissions by increasing the dry deposition removal of aerosols (Zhang et al., 2017b).

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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 on emission sources, atmospheric chemistry and deposition on terrestrial bodies, Environmental Science and Pollution Research, 20, 8092-8131, 2013.

Bergin, M. H., Cass, G. R., Xu, J., Fang, C., Zeng, L., Yu, T., Salmon, L. G., Kiang, C. S., Tang, X. Y., and Zhang, Y.: Aerosol radiative, physical, and chemical properties in Beijing during June 1999, Journal of Geophysical Research, 106, 17969-17980, 2001.

Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, Journal of Geophysical Research, 111, 2006.

Cao, J., Wang, Q., Chow, J. C., Watson, J. G., Tie, X., Shen, Z., Wang, P., and An, Z.: Impacts of aerosol compositions on visibility impairment in Xi'an, China, Atmospheric Environment, 59, 559-566, 2012a.

Cao, J., Lee, S., Ho, K., Zou, S., Fung, K., Li, Y., Watson, J. G., and Chow, J. C.: Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, Atmospheric Environment, 38, 4447-4456, 2004.

Cao, J., Zhu, C., Chow, J. C., Watson, J. G., Han, Y., Wang, G., Shen, Z., and An, Z.: Black carbon relationships with emissions and meteorology in Xi'an, China, Atmospheric Research, 94, 194-202, 2009.

Cao, J., Shen, Z., Chow, J. C., Watson, J. G., Lee, S., Tie, X., Ho, K., Wang, G., and Han, Y.: Winter and summer $PM_{2.5}$ chemical compositions in fourteen Chinese cities, Journal of The Air & Waste Management Association, 62, 1214-1226, 2012b.

Cao, J. J., Zhu, C. S., Tie, X., Geng, F., Xu, H., Ho, S., Wang, G. H., Han, Y. M., and Ho, K. F.: Characteristics and sources of carbonaceous aerosols from Shanghai, China, Atmospheric Chemistry and Physics, 13, 803-817, 2013.

Cao, J., Cohen, A. M., Hansen, J. E., Lester, R. K., Peterson, P. F., and Xu, H.: China-U.S. cooperation to advance nuclear power, Science, 353, 547-548, 2016.

Chan, C. K., and Yao, X.: Air pollution in mega cities in China, Atmospheric environment, 42, 1-42, 2008.

Chan, C. Y., Xu, X., Li, Y. S., Wong, K. H., Ding, G. A., Chan, L. Y., and Cheng, X.: Characteristics of vertical profiles and sources of $PM_{2.5}$, PM_{10} and carbonaceous species in Beijing, Atmospheric Environment, 39, 5113-5124, 2005.

Che, H., Xia, X., Zhu, J., Li, Z., Dubovik, O., Holben, B., Goloub, P., Chen, H., Estelles, V., Cuevas-Agulló, E., Blarel, L., Wang, H., Zhao, H., Zhang, X., Wang, Y., Sun, J., Tao, R., Zhang, X., and Shi, G.: Column aerosol optical properties and aerosol radiative forcing during a serious haze-fog month over North China Plain in 2013 based on ground-based sunphotometer measurements, Atmospheric Chemistry and Physics, 14, 2125-2138, 10.5194/acp-14-2125-2014, 2014.

Che, H., Yang, Z., Zhang, X., Zhu, C., Ma, Q., Zhou, H., and Wang, P.: Study on the aerosol optical properties and their relationship with aerosol chemical compositions over three regional background stations in China, Atmospheric Environment, 43, 1093-1099, 2009.

Che, H., Zhang, X. Y., Xia, X. H., Goloub, P., Holben, B. N., Zhao, H., Wang, Y., Zhang, X., Wang, H., and Blarel, L.: Ground-based aerosol climatology of China: aerosol optical depths from the China Aerosol Remote Sensing Network (CARSNET) 2002–2013, Atmospheric Chemistry and Physics, 15, 7619-7652, 2015.

Chen, J., Qiu, S., Shang, J., Wilfrid, O. M. F., Liu, X., Tian, H., and Boman, J.: Impact of relative humidity and water soluble constituents of $PM_{2.5}$ on visibility impairment in Beijing, China, Aerosol and Air Quality Research, 14, 260-268, 2014a.Chen, J., Zhao, C. S., Ma, N., and Yan, P.: Aerosol hygroscopicity parameter derived from the light scattering enhancement factor

measurements in the North China Plain, Atmospheric Chemistry and Physics, 14, 8105-8118, 10.5194/acp-14-8105-2014, 2014b.
Chen, Y., Schleicher, N., Chen, Y., Chai, F., and Norra, S.: The influence of governmental mitigation measures on contamination characteristics of $PM_{2.5}$ in Beijing, Science of The Total Environment, 490, 647-658, http://dx.doi.org/10.1016/j.scitotenv.2014.05.049, 2014c.
Chen, Y., Xie, S., Luo, B., and Zhai, C.: Characteristics and origins of carbonaceous aerosol in the Sichuan Basin, China, Atmospheric Environment, 94, 215-223, http://dx.doi.org/10.1016/j.atmosenv.2014.05.037, 2014d.
Chen, Y., Xie, S., Luo, B., and Zhai, C.: Particulate pollution in urban Chongqing of southwest China: Historical trends of variation, chemical characteristics and source apportionment, Science of The Total Environment, 584–585, 523-534, http://doi.org/10.1016/j.scitotenv.2017.01.060, 2017.
Cheng, I., Xu, X., and Zhang, L.: Overview of receptor-based source apportionment studies for speciated atmospheric mercury, Atmospheric Chemistry and Physics, 15, 7877-7895, 2015.
Cheng, T., Xu, C., Duan, J., Wang, Y., Leng, C., Tao, J., Che, H., He, Q., Wu, Y., and Zhang, R.: Seasonal variation and difference of aerosol optical properties in columnar and surface atmospheres over Shanghai, Atmospheric Environment, 123, 315-326, 2015.
Cheng, Y., Lee, S. C., Ho, K. F., Wang, Y. Q., Cao, J. J., Chow, J. C., and Watson, J. G.: Black carbon measurement in a coastal area of South China, Journal of Geophysical Research, 111, 1-11, 2006a.
Cheng, Y., Wiedensohler, A., Eichler, H., Su, H., Gnauk, T., Brüggemann, E., Herrmann, H., Heintzenberg, J., Slanina, J., and Tuch, T.: Aerosol optical properties and related chemical apportionment at Xinken in Pearl River Delta of China, Atmospheric Environment, 42, 6351-6372, 2008a.
Cheng, Y., He, K. B., Zheng, M., Duan, F. K., Du, Z. Y., Ma, Y. L., Tan, J. H., Yang, F. M., Liu, J. M., and Zhang, X. L.: Mass absorption efficiency of elemental carbon and water-soluble organic carbon in Beijing, China, Atmospheric Chemistry and Physics, 11, 11497-11510, 2011.
Cheng, Y., Engling, G., He, K. B., Duan, F. K., Ma, Y. L., Du, Z. Y., Liu, J. M., Zheng, M., and Weber, R. J.: Biomass burning contribution to Beijing aerosol, Atmospheric Chemistry and Physics, 13, 7765-7781, 2013a.
Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H., Zeng, L. M., Liu, S., and Gnauk, T.: Mixing state of elemental carbon and non‐light‐absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, Journal of Geophysical Research, 111, 2006b.
Cheng, Y. F., Wiedensohler, A., Eichler, H., Heintzenberg, J., Tesche, M., Ansmann, A., Wendisch, M., Su, H., Althausen, D., and Herrmann, H.: Relative humidity dependence of aerosol optical properties and direct radiative forcing in the surface boundary layer at Xinken in Pearl River Delta of China: An observation based numerical study, Atmospheric Environment, 42, 6373-6397, 2008b.
Cheng, Y. F., Berghof, M., Garland, R. M., Wiedensohler, A., Wehner, B., Muller, T., Su, H., Zhang, Y., Achtert, P., and Nowak, A.: Influence of soot mixing state on aerosol light absorption and single scattering albedo during air mass aging at a polluted regional site in northeastern China, Journal of Geophysical Research, 114, 2009.
Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Pöschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, 10.1126/sciadv.1601530, 2016.

Cheng, Z., Wang, S., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Fu, X., and Hao, J.: Long-term trend of haze pollution and impact of particulate matter in the Yangtze River Delta, China, Environmental Pollution, 182, 101-110, 2013b.

Cheung, H. C., Wang, T., Baumann, K., and Guo, H.: Influence of regional pollution outflow on the concentrations of fine particulate matter and visibility in the coastal area of southern China, Atmospheric Environment, 39, 6463-6474, 2005.

Chow, J. C., Watson, J. G., Crow, D., Lowenthal, D. H., and Merrifield, T.: Comparison of IMPROVE and NIOSH carbon measurements, Aerosol Science and Technology, 34, 23-34, 2010.

Chow, J. C., Watson, J. G., Lowenthal, D. H., and Magliano, K. L.: Loss of $PM_{2.5}$ nitrate from filter samples in central California, Journal of The Air & Waste Management Association, 55, 1158-1168, 2005.

Cui, H., Chen, W., Dai, W., Liu, H., Wang, X., and He, K.: Source apportionment of $PM_{2.5}$ in Guangzhou combining observation data analysis and chemical transport model simulation, Atmospheric Environment, 116, 262-271, http://dx.doi.org/10.1016/j.atmosenv.2015.06.054, 2015.

Dai, W., Gao, J., Cao, G., and Ouyang, F.: Chemical composition and source identification of $PM_{2.5}$ in the suburb of Shenzhen, China, Atmospheric Research, 122, 391-400, http://dx.doi.org/10.1016/j.atmosres.2012.12.004, 2013.

Dan, M., Zhuang, G., Li, X., Tao, H., and Zhuang, Y.: The characteristics of carbonaceous species and their sources in $PM_{2.5}$ in Beijing, Atmospheric Environment, 38, 3443-3452, 2004.

Du, Z., He, K., Cheng, Y., Duan, F., Ma, Y., Liu, J., Zhang, X., Zheng, M., and Weber, R.: A yearlong study of water-soluble organic carbon in Beijing I: Sources and its primary vs. secondary nature, 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. L., Guo, Z. G., Zhang, T. R., Yao, X., Chan, C. K., and Fang, M.: Source and formation of secondary particulate matter in $PM_{2.5}$ in Asian continental outflow, Journal of Geophysical Research, 117, 2012b.

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, 2012a.

Feng, J., Zhong, M., Xu, B., Du, Y., Wu, M., Wang, H., and Chen, C.: Concentrations, seasonal and diurnal variations of black carbon in $PM_{2.5}$ in Shanghai, China, 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.

Fontes, T., Li, P., Barros, N., and Zhao, P.: Trends of $PM_{2.5}$ concentrations in China: A long term approach, Journal of Environmental Management, 196, 719-732, 2017.

Fu, X., Wang, X., Guo, H., Cheung, K., Ding, X., Zhao, X., He, Q., Gao, B., Zhang, Z., Liu, T., and Zhang, Y.: Trends of ambient fine particles and major chemical components in the Pearl River Delta region: Observation at a regional background site in fall and winter, 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.: $PM_{2.5}$ in an industrial district of Zhengzhou, China: Chemical composition and source apportionment, Particuology, 11, 99-109, 2013.

Ginoux, P., Prospero, J. M., Gill, T. E., Hsu, N. C., and Zhao, M.: Global‐scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products, 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, H., Ding, A. J., So, K. L., Ayoko, G. A., Li, Y., and Hung, W. T.: Receptor modeling of source apportionment of Hong Kong aerosols and the implication of urban and regional contribution, Atmospheric Environment, 43, 1159-1169, 2009a.

Guo, J., Zhang, X., Che, H., Gong, S., An, X., Cao, C., Guang, J., Zhang, H., Wang, Y., and Zhang, X.: Correlation between PM concentrations and aerosol optical depth in eastern China, Atmospheric Environment, 43, 5876-5886, 2009b.

Guo, J., Xia, F., Zhang, Y., Liu, H., Li, J., Lou, M., He, J., Yan, Y., Wang, F., Min, M., and Zhai, P.: Impact of diurnal variability and meteorological factors on the $PM_{2.5}$ - AOD relationship: Implications for $PM_{2.5}$ remote sensing, Environmental Pollution, 221, 94-104, https://doi.org/10.1016/j.envpol.2016.11.043, 2017.

Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., and Zeng, L.: Elucidating severe urban haze formation in China, 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, 2006a.

Ho, K. F., Cao, J. J., Lee, S. C., and Chan, C. K.: Source apportionment of $PM_{2.5}$ in urban area of Hong Kong, Journal of Hazardous Materials, 138, 73-85, 2006b.

Hopke, P. K.: Review of receptor modeling methods for source apportionment, Journal of The Air & Waste Management Association, 66, 237-259, 2016.

Hou, B., Zhuang, G., Zhang, R., Liu, T., Guo, Z., and Chen, Y.: The implication of carbonaceous aerosol to the formation of haze: Revealed from the characteristics and sources of OC/EC over a mega-city in China, Journal of Hazardous Materials, 190, 529-536, http://dx.doi.org/10.1016/j.jhazmat.2011.03.072, 2011.

Hu, M., Wu, Z., Slanina, J., Lin, P., Liu, S., and Zeng, L.: Acidic gases, ammonia and water-soluble ions in $PM_{2.5}$ at a coastal site in the Pearl River Delta, China, Atmospheric Environment, 42, 6310-6320, 2008.

Hu, X., Waller, L. A., Alhamdan, M. Z., Crosson, W. L., Estes, M. G., Estes, S., Quattrochi, D. A., Sarnat, J. A., and Liu, Y.: Estimating ground-level $PM_{2.5}$ concentrations in the southeastern U.S. using geographically weighted regression, Environmental Research, 121, 1-10, 2013.

Huang, G., Cheng, T., Zhang, R., Tao, J., Leng, C., Zhang, Y., Zha, S., Zhang, D., Li, X., and Xu, C.: Optical properties and chemical composition of $PM_{2.5}$ in Shanghai in the spring of 2012, Particuology, 13, 52-59, 2014a.

Huang, H., Lee, S. C., Cao, J. J., Zou, C. W., Chen, X. G., and Fan, S. J.: Characteristics of indoor/outdoor $PM_{2.5}$ and elemental components in generic urban, roadside and industrial plant areas of Guangzhou city, China, 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.

Huang, X. H. H., Bian, Q., Ng, W. M., Louie, P. K. K., and Yu, J. Z.: Characterization of $PM_{2.5}$ major components and source Investigation in suburban Hong Kong: a one year monitoring study, Aerosol and Air Quality Research, 14, 237-250, 2014c.

Jahn, H. J., Kraemer, A., Chen, X. C., Chan, C. Y., Engling, G., and Ward, T. J.: Ambient and personal $PM_{2.5}$ exposure assessment in the Chinese megacity of Guangzhou, Atmospheric Environment, 74, 402-411, 2013.

Jing, J., Wu, Y., Tao, J., Che, H., Xia, X., Zhang, X., Yan, P., Zhao, D., and Zhang, L.: Observation and analysis of near-surface atmospheric aerosol optical properties in urban Beijing, Particuology, 18, 144-154, 2015.

Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Gu, J., and Fan, S.: Aerosol chemistry and the effect of aerosol water content on visibility impairment and radiative forcing in Guangzhou during the 2006 Pearl River Delta campaign, Journal of environmental management, 90, 3231-3244, 2009.

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

Karagulian, F., Belis, C. A., Dora, C. F. C., Prüss-Ustün, A. M., Bonjour, S., Adair-Rohani, H., and Amann, M.: Contributions to cities' ambient particulate matter (PM): A systematic review of local source contributions at global level, Atmospheric Environment, 120, 475-483, 2015.

Keck, L., and Wittmaack, K.: Effect of filter type and temperature on volatilisation losses from ammonium salts in aerosol matter, Atmospheric Environment, 39, 4093-4100, 2005.

Kuang, Y., Zhao, C. S., Ma, N., Liu, H. J., Bian, Y., Tao, J. C., and Hu, M.: Deliquescent phenomena of ambient aerosols on the North China Plain, Geophysical Research Letters, 43, doi:10.1002/2016GL070273, 2016.

Lai, S., Zhao, Y., Ding, A., Zhang, Y., Song, T., Zheng, J., Ho, K. F., Lee, S., and Zhong, L.: Characterization of $PM_{2.5}$ and the major chemical components during a 1-year campaign in rural Guangzhou, Southern China, 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.

Lee, J. H. and Hopke, P. K.: Apportioning sources of $PM_{2.5}$ in St. Louis, MO using speciation trends network data, Atmospheric Environment, 40, S360–S377, 2006.

Li, B., Zhang, J., Zhao, Y., Yuan, S., Zhao, Q., Shen, G., and Wu, H.: Seasonal variation of urban carbonaceous aerosols in a typical city Nanjing in Yangtze River Delta, China, 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, G., Bei, N., Cao, J., Huang, R., Wu, J., Feng, T., Wang, Y., Liu, S., Zhang, Q., Tie, X., and Molina, L. T.: A possible pathway for rapid growth of sulfate during haze days in China, Atmospheric Chemistry and Physics, 17, 3301-3316, 10.5194/acp-17-3301-2017, 2017b.

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.
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: Atmospheres, 118, 12,158-112,167, 2013c.
Li, Y., Meng, J., Liu, J., Xu, Y., Guan, D., Tao, W., Huang, Y., and Tao, S.: Interprovincial Reliance for Improving Air Quality in China: A Case Study on Black Carbon Aerosol, Environmental Science & Technology, 50, 4118-4126, 2016b.
Li, Y. J., Sun, Y., Zhang, Q., Li, X., Li, M., Zhou, Z., and Chan, C. K.: Real-time chemical characterization of atmospheric particulate matter in China: A review, Atmospheric Environment, 158, 270-304, https://doi.org/10.1016/j.atmosenv.2017.02.027, 2017a.
Li, Z., Lau, W. K., Ramanathan, V., Wu, G., Ding, Y., Manoj, M. G., Liu, J., Qian, Y., Li, J., Zhou, T., Fan, J., Rosenfeld, D., Ming, Y., Wang, Y., Huang, J., Wang, B., Xu, X., Lee, S. S., Cribb, M., Zhang, F., Yang, X., Takemura, T., Wang, K., Xia, X., Yin, Y., Zhang, H., Guo, J., Zhai, P. M., Sugimoto, N., Babu, S. S., and Brasseur, G. P.: Aerosol and Monsoon Climate Interactions over Asia, Reviews of Geophysics, 54, doi:10.1002/2015rg000500, 2016c.
Liang, C. S., Duan, F. K., He, K. B., and Ma, Y. L.: Review on recent progress in observations, source identifications and countermeasures of $PM_{2.5}$, Environment International, 86, 150-170, http://dx.doi.org/10.1016/j.envint.2015.10.016, 2016.
Liao, H., Chang, W., and Yang, Y.: Climatic effects of air pollutants over China: A review, Advances in Atmospheric Sciences, 32, 115-139, 2015.
Lin, M., Biglari, S., Zhang, Z., Crocker, D., Tao, J., Su, B., Liu, L., and Thiemens, M. H.: Vertically uniform formation pathways of tropospheric sulfate aerosols in East China detected from triple stable oxygen and radiogenic sulfur isotopes, Geophysical Research Letters, 44, doi:10.1002/2017GL073637, 2017.
Lin, Y., Hsu, S., Chou, C. C. K., Zhang, R., Wu, Y., Kao, S., Luo, L., Huang, C., Lin, S., and Huang, Y.: Wintertime haze deterioration in Beijing by industrial pollution deduced from trace metal fingerprints and enhanced health risk by heavy metals, Environmental Pollution, 208, 284-293, 2016.
Lin, Z., Zhang, Z., Zhang, L., Tao, J., Zhang, R., Cao, J., Fan, S., and Zhang, Y.: An alternative method for estimating hygroscopic growth factor of aerosol light-scattering coefficient: a case study in an urban area of Guangzhou, South China, Atmospheric Chemistry and Physics, 14, 7631-7644, 2014.
Liu, B., Song, N., Dai, Q., Mei, R., Sui, B., Bi, X., and Feng, Y.: Chemical composition and source apportionment of ambient $PM_{2.5}$ during the non-heating period in Taian, China, Atmospheric

Research, 170, 23-33, http://doi.org/10.1016/j.atmosres.2015.11.002, 2016a.
Liu, B., Li, T., Yang, J., Wu, J., Wang, J., Gao, J., Bi, X., Feng, Y., Zhang, Y., and Yang, H.: Source apportionment and a novel approach of estimating regional contributions to ambient $PM_{2.5}$ in Haikou, China, Environmental Pollution, 223, 334-345, http://doi.org/10.1016/j.envpol.2017.01.030, 2017a.
Liu, B., Wu, J., Zhang, J., Wang, L., Yang, J., Liang, D., Dai, Q., Bi, X., Feng, Y., Zhang, Y., and Zhang, Q.: Characterization and source apportionment of $PM_{2.5}$ based on error estimation from EPA PMF 5.0 model at a medium city in China, Environmental Pollution, 222, 10-22, http://doi.org/10.1016/j.envpol.2017.01.005, 2017b.
Liu, D., Li, J., Zhang, Y., Xu, Y., Liu, X., Ding, P., Shen, C., Chen, Y., Tian, C., and Zhang, G.: The use of levoglucosan and radiocarbon for source apportionment of $PM_{2.5}$ carbonaceous aerosols at a background site in east China, Environmental Science & Technology, 47, 10454, 2013a.
Liu, G., Li, J., Wu, D., and Xu, H.: Chemical composition and source apportionment of the ambient $PM_{2.5}$ in Hangzhou, China, Particuology, 18, 135-143, http://dx.doi.org/10.1016/j.partic.2014.03.011, 2015.
Liu, J., Li, J., Zhang, Y., Liu, D., Ding, P., Shen, C., Shen, K., He, Q., Ding, X., and Wang, X.: Source apportionment using radiocarbon and organic tracers for $PM_{2.5}$ carbonaceous aerosols in Guangzhou, south China: contrasting local- and regional-scale haze events, Environmental Science & Technology, 48, 12002-12011, 2014a.
Liu, P., Zhao, C., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W., Deng, Z., Ma, N., and Mildenberger, K.: Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain, Atmospheric Chemistry and Physics, 11, 3479-3494, 2011.
Liu, S., Hu, M., Slanina, S., He, L., Niu, Y., Bruegemann, E., Gnauk, T., and Herrmann, H.: Size distribution and source analysis of ionic compositions of aerosols in polluted periods at Xinken in Pearl River Delta (PRD) of China, Atmospheric Environment, 42, 6284-6295, 2008b.
Liu, X., Chen, Q., Che, H., Zhang, R., Gui, K., Zhang, H., and Zhao, T.: Spatial distribution and temporal variation of aerosol optical depth in the Sichuan basin, China, the recent ten years, Atmospheric Environment, 147, 434-445, https://doi.org/10.1016/j.atmosenv.2016.10.008, 2016b.
Liu, X., Cheng, Y., Zhang, Y., Jung, J., Sugimoto, N., Chang, S. Y., Kim, Y. J., Fan, S., and Zeng, L.: Influences of relative humidity and particle chemical composition on aerosol scattering properties during the 2006 PRD campaign, Atmospheric Environment, 42, 1525-1536, 2008a.
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, Y., Sarnat, J. A., Kilaru, V., Jacob, D. J., and Koutrakis, P.: Estimating ground-level $PM_{2.5}$ in the eastern United States using satellite remote sensing, Environmental Science & Technology, 39, 3269-3278, 2005.
Liu, Z., Hu, B., Wang, L., Wu, F., Gao, W., and Wang, Y.: Seasonal and diurnal variation in particulate matter (PM_{10} and $PM_{2.5}$) 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.
Lv, B., Zhang, B., and Bai, Y.: A systematic analysis of $PM_{2.5}$ in Beijing and its sources from 2000 to 2012, Atmospheric Environment, 124, Part B, 98-108, https://doi.org/10.1016/j.atmosenv.2015.09.031, 2016.
Ma, N., Zhao, C. S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y. F., Deng, Z. Z., Liu, P. F., Xu, W. Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., 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.
Ma, Z., Hu, X., Huang, L., Bi, J., and Liu, Y.: Estimating ground-level $PM_{2.5}$ in China using satellite remote sensing, Environmental Science & Technology, 48, 7436-7444, 2014.
Malm, W. C., Day, D. E., Kreidenweis, S. M., Collett, J. L., and Lee, T.: Humidity-dependent optical properties of fine particles during the Big Bend Regional Aerosol and Visibility Observational Study, 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.
Ming, L., Jin, L., Li, J., Fu, P., Yang, W., Liu, D., Zhang, G., Wang, Z., and Li, X.: $PM_{2.5}$ in the Yangtze River Delta, China: Chemical compositions, seasonal variations, and regional pollution events, Environmental Pollution, 223, 200-212, https://doi.org/10.1016/j.envpol.2017.01.013, 2017.
Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., 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.
Niu, F., Li, Z., Li, C., Lee, K., and Wang, M.: Increase of wintertime fog in China: Potential impacts of weakening of the Eastern Asian monsoon circulation and increasing aerosol loading, Journal of Geophysical Research, 115, 2010.

Okuda, T., Matsuura, S., Yamaguchi, D., Umemura, T., Hanada, E., Orihara, H., Tanaka, S., He, K., Ma, Y., and Cheng, Y.: The impact of the pollution control measures for the 2008 Beijing Olympic Games on the chemical composition of aerosols, Atmospheric Environment, 45, 2789-2794, 2011.

Orsini, D. A., Ma, Y., Sullivan, A., Sierau, B., Baumann, K., and Weber, R. J.: Refinements to the particle-into-liquid sampler (PILS) for ground and airborne measurements of water soluble aerosol composition, Atmospheric Environment, 37, 1243-1259, http://dx.doi.org/10.1016/S1352-2310(02)01015-4, 2003.

Pan, L., Che, H., Geng, F., Xia, X., Wang, Y., Zhu, C., Chen, M., Gao, W., and Guo, J.: Aerosol optical properties based on ground measurements over the Chinese Yangtze Delta Region, Atmospheric Environment, 44, 2587-2596, 2010.

Pan, X. L., Yan, P., Tang, J., Ma, J. Z., Wang, Z. F., Gbaguidi, A., and Sun, Y. L.: Observational study of influence of aerosol hygroscopic growth on scattering coefficient over rural area near Beijing mega-city, Atmospheric Chemistry and Physics, 9, 7519-7530, 10.5194/acp-9-7519-2009, 2009.

Pathak, R. K., Wang, T., Ho, K., and Lee, S.: Characteristics of summertime $PM_{2.5}$ organic and elemental carbon in four major Chinese cities: implications of high acidity for water-soluble organic carbon (WSOC), Atmospheric Environment, 45, 318-325, 2011.

Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., and Hand, J.: Revised algorithm for estimating light extinction from IMPROVE particle speciation data, Journal of the Air & Waste Management Association, 57, 1326-1336, 2007.

Pu, W., Zhao, X., Shi, X., Ma, Z., Zhang, X., and Yu, B.: Impact of long-range transport on aerosol properties at a regional background station in Northern China, Atmospheric Research, 153, 489-499, 2015.

Pui, D. Y., Chen, S.-C., and Zuo, Z.: $PM_{2.5}$ in China: Measurements, sources, visibility and health effects, and mitigation, Particuology, 13, 1-26, 2014.

Qian, Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K. M., Ming, J., Wang, H., Wang, M., Warren, S. G., and Zhang, R.: Light-absorbing Particles in Snow and Ice: Measurement and Modeling of Climatic and Hydrological Impact, Advances in Atmospheric Sciences, 32, 64-91, 2015.

Qin, Y., and Xie, S.: Spatial and temporal variation of anthropogenic black carbon emissions in China for the period 1980–2009, Atmospheric Chemistry and Physics, 12, 4825-4841, 2012.

Schnaiter, M., Linke, C., Mohler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic aerosol, Journal of Geophysical Research, 110, 2005.

Shen, G., Xue, M., Yuan, S., Zhang, J., Zhao, Q., Li, B., Wu, H., and Ding, A.: Chemical compositions and reconstructed light extinction coefficients of particulate matter in a mega-city in the western Yangtze River Delta, China, Atmospheric Environment, 83, 14-20, http://dx.doi.org/10.1016/j.atmosenv.2013.10.055, 2014.

Shen, Z., Cao, J., Arimoto, R., Han, Z., Zhang, R., Han, Y., Liu, S., Okuda, T., Nakao, S., and Tanaka, S.: Ionic composition of TSP and $PM_{2.5}$ during dust storms and air pollution episodes at Xi'an, China, Atmospheric Environment, 43, 2911-2918, 2009.

Song, Y., Xie, S., Zhang, Y., Zeng, L., Salmon, L. G., and Zheng, M.: Source apportionment of $PM_{2.5}$ in Beijing using principal component analysis/absolute principal component scores and UNMIX, Science of the Total Environment, 372, 278-286, 2006a.

Song, Y., Zhang, Y., Xie, S., Zeng, L., Zheng, M., Salmon, L. G., Shao, M., and Slanina, S.: Source

apportionment of $PM_{2.5}$ in Beijing by positive matrix factorization, Atmospheric Environment, 40, 1526-1537, 2006b.

Song, Y., Tang, X., Xie, S., Zhang, Y., Wei, Y., Zhang, M., Zeng, L., and Lu, S.: Source apportionment of $PM_{2.5}$ in Beijing in 2004, Journal of Hazardous Materials, 146, 124-130, 2007.

Sun, Y., Zhuang, G., Wang, Y., Han, L., Guo, J., Dan, M., Zhang, W., Wang, Z., and Hao, Z.: The air-borne particulate pollution in Beijing - concentration, composition, distribution and sources, Atmospheric Environment, 38, 5991-6004, 2004.

Sun, Y., Zhuang, G., Tang, A., Wang, Y., and An, Z.: Chemical Characteristics of $PM_{2.5}$ and PM_{10} in Haze-Fog Episodes in Beijing, Environmental Science & Technology, 40, 3148, 2006.

Sun, Y. L., Wang, Z. F., Fu, P., Yang, T., Jiang, Q., Dong, H., Li, J., and Jia, J.: Aerosol composition, sources and processes during wintertime in Beijing, China, Atmospheric Chemistry and Physics, 13, 4577-4592, 2013.

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, 2016a.

Tan, J., Xiang, P., Zhou, X., Duan, J., Ma, Y., He, K., Cheng, Y., Yu, J., and Querol, X.: Chemical characterization of humic-like substances (HULIS) in $PM_{2.5}$ in Lanzhou, China, Science of The Total Environment, 573, 1481-1490, https://doi.org/10.1016/j.scitotenv.2016.08.025, 2016b.

Tan, J., Duan, J., Ma, Y., He, K., Cheng, Y., Deng, S., Huang, Y., and Si-Tu, S.: Long-term trends of chemical characteristics and sources of fine particle in Foshan City, Pearl River Delta: 2008–2014, Science of The Total Environment, 565, 519-528, http://doi.org/10.1016/j.scitotenv.2016.05.059, 2016c.

Tan, J., Zhang, L., Zhou, X., Duan, J., Li, Y., Hu, J., and He, K.: Chemical characteristics and source apportionment of PM2.5 in Lanzhou, China, Science of the Total Environment, 601-602, 1743-1752, 2017.

Tang, X., Chen, X., and Tian, Y.: Chemical composition and source apportionment of $PM_{2.5}$ – A case study from one year continuous sampling in the Chang-Zhu-Tan urban agglomeration, Atmospheric Pollution Research, 8, 885-899, https://doi.org/10.1016/j.apr.2017.02.004, 2017.

Tao, J., Ho, K., Chen, L., Zhu, L., Han, J., and Xu, Z.: Effect of chemical composition of $PM_{2.5}$ on visibility in Guangzhou, China, 2007 spring, Particuology, 7, 68-75, 2009.

Tao, J., Cheng, T., Zhang, R., Cao, J., Zhu, L., Wang, Q., Luo, L., and Zhang, L.: Chemical composition of $PM_{2.5}$ at an urban site of Chengdu in southwestern China, 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 $PM_{2.5}$ 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.

Trebs, I., Meixner, F. X., Slanina, J., Otjes, R. P., Jongejan, P., and Andreae, M. O.: Real-time measurements of ammonia, acidic trace gases and water-soluble inorganic aerosol species at a rural site in the Amazon Basin, Atmospheric Chemistry and Physics, 4, 967-987, 2004.

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.

Tian, Y. Z., Chen, G., Wang, H. T., Huang-Fu, Y.Q., Shi, G. L., Han, B., and Feng, Y. C.: Source regional contributions to $PM_{2.5}$ in a megacity in China using an advanced source regional apportionment method, Chemosphere, 147, 256-263, https://doi.org/10.1016/j.chemosphere.2015.12.132, 2016.

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, J., and Christopher, S. A.: Intercomparison between satellite‐derived aerosol optical thickness and $PM_{2.5}$ mass: Implications for air quality studies, Geophysical Research Letters, 30, 2095-2099,

2003.
Wang, G., Zhang, R., Gomez, M. E., Yang, L., Zamora, M. L., Hu, M., Lin, Y., Peng, J., Guo, S., and Meng, J.: Persistent sulfate formation from London Fog to Chinese haze, Proceedings of the National Academy of Sciences, 113, 13630-13635, 2016c.
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, 2017a.
Wang, H. L., Qiao, L. P., Lou, S. R., Zhou, M., Ding, A. J., Huang, H. Y., Chen, J. M., Wang, Q., Tao, S. K., Chen, C. H., Li, L., and Huang, C.: Chemical composition of $PM_{2.5}$ and meteorological impact among three years in urban Shanghai, China, 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, Q., Jiang, N., Yin, S., Li, X., Yu, F., Guo, Y., and Zhang, R.: Carbonaceous species in $PM_{2.5}$ and PM_{10} in urban area of Zhengzhou in China: Seasonal variations and source apportionment, Atmospheric Research, 191, 1-11, https://doi.org/10.1016/j.atmosres.2017.02.003, 2017b.
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.
Wang, Y., Zhuang, G., Tang, A., Yuan, H., Sun, Y., Chen, S., and Zheng, A.: The ion chemistry and the source of $PM_{2.5}$ aerosol in Beijing, Atmospheric Environment, 39, 3771-3784, 2005.

Wang, Y., Zhuang, G., Zhang, X. Y., Huang, K., Xu, C., Tang, A. H., Chen, J. M., and An, Z.: The ion chemistry, seasonal cycle, and sources of $PM_{2.5}$ and TSP aerosol in Shanghai, Atmospheric Environment, 40, 2935-2952, 2006.

Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Science China Earth Sciences, 57, 14-25, 2014c.

Wang, Y., Ying, Q., Hu, J., and Zhang, H.: Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014, Environment International, 73, 413-422, http://dx.doi.org/10.1016/j.envint.2014.08.016, 2014b.

Wang, Z., Li, J., Wang, Z., Yang, W., Tang, X., Ge, B., Yan, P., Zhu, L., Chen, X., and Chen, H.: Modeling study of regional severe hazes over mid-eastern China in January 2013 and its implications on pollution prevention and control, Science China-earth Sciences, 57, 3-13, 2013.

Watson, J. G., Chow, J. C., and Houck, J. E.: $PM_{2.5}$ chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995, Chemosphere, 43, 1141-1151, 2001.

Watson, J. G.: Visibility: Science and regulation, Journal of the Air & Waste Management Association, 52, 628-713, 2002.

Weber, R., Orsini, D., Duan, Y., Baumann, K., Kiang, C. S., Chameides, W. L., Lee, Y. N., Brechtel, F. J., Klotz, P. J., and Jongejan, P.: Intercomparison of near real time monitors of $PM_{2.5}$ nitrate and sulfate at the U.S. Environmental Protection Agency Atlanta Supersite, Journal of Geophysical Research, 108, 2003.

Wu, C., Ng, W. M., Huang, J., Wu, D., and Yu, J. Z.: Determination of Elemental and Organic Carbon in $PM_{2.5}$ in the Pearl River Delta Region: Inter-Instrument (Sunset vs. DRI Model 2001 Thermal/Optical Carbon Analyzer) and Inter-Protocol Comparisons (IMPROVE vs. ACE-Asia Protocol), Aerosol Science and Technology, 46, 610-621, 2011.

Wu, D., Mao, J., Deng, X., Tie, X., Zhang, Y., Zeng, L., Li, F., Tan, H., Bi, X., and Huang, X.: Black carbon aerosols and their radiative properties in the Pearl River Delta region, Science in China Series D: Earth Sciences, 52, 1152-1163, 2009.

Wu, D., Wu, C., Liao, B., Chen, H., Wu, M., Li, F., Tan, H., Deng, T., Li, H., and Jiang, D.: Black carbon over the South China Sea and in various continental locations in South China, Atmospheric Chemistry and Physics, 13, 12257-12270, 2013.

Wu, S., Deng, F., Wei, H., Huang, J., Wang, X., Hao, Y., Zheng, C., Qin, Y., Lv, H., Shima, M., and Guo, X.: Association of Cardiopulmonary Health Effects with Source-Appointed Ambient Fine Particulate in Beijing, China: A Combined Analysis from the Healthy Volunteer Natural Relocation (HVNR) Study, Environmental Science & Technology, 48, 3438-3448, 10.1021/es404778w, 2014.

Wu, Y., Wang, X., Yan, P., Zhang, L., Tao, J., Liu, X., Tian, P., Han, Z., and Zhang, R.: Investigation of hygroscopic growth effect on aerosol scattering coefficient at a rural site in the southern North China Plain, Science of The Total Environment, 599–600, 76-84, https://doi.org/10.1016/j.scitotenv.2017.04.194, 2017.

Wu, Y., Zhang, R., Tian, P., Tao, J., Hsu, S. C., Yan, P., Wang, Q., Cao, J., Zhang, X., and Xia, X.: Effect of ambient humidity on the light absorption amplification of black carbon in Beijing during January 2013, Atmospheric Environment, 124, 217-223, 2016.

Xie, Y., Ding, A., Nie, W., Mao, H., Qi, X., Huang, X., Xu, Z., Kerminen, V. M., Petaja, T., and Chi, X.:

Enhanced sulfate formation by nitrogen dioxide: Implications from in situ observations at the SORPES station, Journal of Geophysical Research, 120, 12679-12694, 2015a.

Xie, Y., Wang, Y., Zhang, K., Dong, W., Lv, B., and Bai, Y.: Daily Estimation of Ground-Level $PM_{2.5}$ Concentrations over Beijing Using 3 km Resolution MODIS AOD, Environmental Science & Technology, 49, 12280-12288, 2015b.

Xiong, Y., Zhou, J., Schauer, J. J., Yu, W., and Hu, Y.: Seasonal and spatial differences in source contributions to $PM_{2.5}$ in Wuhan, China, Science of The Total Environment, 577, 155-165, http://doi.org/10.1016/j.scitotenv.2016.10.150, 2017.

Xu, H., Cao, J., Chow, J. C., Huang, R. J., Shen, Z., Chen, L. W. A., Ho, K. F., and Watson, J. G.: Inter-annual variability of wintertime $PM_{2.5}$ chemical composition in Xi'an, China: Evidences of changing source emissions, Science of The Total Environment, 545, 546-555, 2016.

Xu, J., Bergin, M., Yu, X., Liu, G., Zhao, J., Carrico, C., and Baumann, K.: Measurement of aerosol chemical, physical and radiative properties in the Yangtze delta region of China, Atmospheric Environment, 36, 161-173, 2002.

Xu, J., Bergin, M. H., Greenwald, R., Schauer, J. J., Shafer, M. M., Jaffrezo, J. L., and Aymoz, G.: Aerosol chemical, physical, and radiative characteristics near a desert source region of northwest China during ACE-Asia, Journal of Geophysical Research, 109, 2004.

Xu, J., Tao, J., Zhang, R., Cheng, T., Leng, C., Chen, J., Huang, G., Li, X., and Zhu, Z.: Measurements of surface aerosol optical properties in winter of Shanghai, Atmospheric Research, 109, 25-35, 2012a.

Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical compositions of $PM_{2.5}$ aerosol in the urban area of Fuzhou, China, Atmospheric Research, 104–105, 264-272, http://dx.doi.org/10.1016/j.atmosres.2011.10.017, 2012b.

Xu, L., Chen, X., Chen, J., Zhang, F., He, C., Zhao, J., and Yin, L.: Seasonal variations and chemical compositions of $PM_{2.5}$ aerosol in the urban area of Fuzhou, China, Atmospheric Research, 104, 264-272, 2012c.

Xu, W. Y., Zhao, C. S., Ran, L., Deng, Z. Z., Liu, P. F., Ma, N., Lin, W. L., Xu, X. B., Yan, P., He, X., Yu, J., Liang, W. D., and Chen, L. L.: Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain, Atmospheric Chemistry and Physics, 11, 4353-4369, 10.5194/acp-11-4353-2011, 2011.

Yan, H.: Aerosol scattering properties in northern China, Atmospheric Environment, 41, 6916-6922, 2007.

Yan, P., Tang, J., Huang, J., Mao, J., Zhou, X., Liu, Q., Wang, Z., and Zhou, H.: The measurement of aerosol optical properties at a rural site in Northern China, Atmospheric Chemistry and Physics, 8, 2229-2242, 2008.

Yan, P., Pan, X., Tang, J., Zhou, X., Zhang, R., and Zeng, L.: Hygroscopic growth of aerosol scattering coefficient: A comparative analysis between urban and suburban sites at winter in Beijing, Particuology, 7, 52-60, http://dx.doi.org/10.1016/j.partic.2008.11.009, 2009.

Yang, F., He, K. B., Ye, B., Chen, X., Cha, L. Z., Cadle, S. H., Chan, T., and Mulawa, P. A.: One-year record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai, Atmospheric Chemistry and Physics, 5, 1449-1457, 2005a.

Yang, F., Huang, L., Duan, F., Zhang, W., He, K., Ma, Y., Brook, J. R., Tan, J., Zhao, Q., and Cheng, Y.: Carbonaceous species in $PM_{2.5}$ at a pair of rural/urban sites in Beijing, 2005–2008, Atmospheric Chemistry and Physics, 11, 7893-7903, 10.5194/acp-11-7893-2011, 2011a.

Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., and Chen, G.: Characteristics of $PM_{2.5}$ speciation in representative megacities and across China, Atmospheric Chemistry and Physics, 11, 5207-5219, 2011b.

Yang, H., Yu, J. Z., Ho, S. S. H., Xu, J., Wu, W., Wan, C. H., Wang, X., Wang, X., and Wang, L.: The chemical composition of inorganic and carbonaceous materials in $PM_{2.5}$ in Nanjing, China, Atmospheric Environment, 39, 3735-3749, 2005b.

Yang, H., Chen, J., Wen, J., Tian, H., and Liu, X.: Composition and sources of $PM_{2.5}$ around the heating periods of 2013 and 2014 in Beijing: Implications for efficient mitigation measures, Atmospheric Environment, 124, Part B, 378-386, http://dx.doi.org/10.1016/j.atmosenv.2015.05.015, 2016.

Yang, L., Cheng, S., Wang, X., Nie, W., Xu, P., Gao, X., Yuan, C., and Wang, W.: Source identification and health impact of $PM_{2.5}$ in a heavily polluted urban atmosphere in China, Atmospheric Environment, 75, 265-269, http://doi.org/10.1016/j.atmosenv.2013.04.058, 2013.

Yang, L., Zhou, X., Wang, Z., Zhou, Y., Cheng, S., Xu, P., Gao, X., Nie, W., Wang, X., and Wang, W.: Airborne fine particulate pollution in Jinan, China: concentrations, chemical compositions and influence on visibility impairment, Atmospheric Environment, 55, 506-514, 2012.

Yang, Y., Liu, X., Qu, Y. Z., An, J., Jiang, R., Zhang, Y., Sun, Y., Wu, Z. J., Zhang, F., and Xu, W. Q.: Characteristics and formation mechanism of continuous hazes in China: a case study during the autumn of 2014 in the North China Plain, Atmospheric Chemistry and Physics, 15, 8165-8178, 2015.

Yao, L., Yang, L., Yuan, Q., Yan, C., Dong, C., Meng, C., Sui, X., Yang, F., Lu, Y., and Wang, W.: Sources apportionment of $PM_{2.5}$ in a background site in the North China Plain, Science of The Total Environment, 541, 590-598, http://doi.org/10.1016/j.scitotenv.2015.09.123, 2016.

Ye, B., Ji, X., Yang, H., Yao, X., Chan, C. K., Cadle, S. H., Chan, T., and Mulawa, P. A.: Concentration and chemical composition of $PM_{2.5}$ in Shanghai for a 1-year period, Atmospheric Environment, 37, 499-510, 2003.

Yu, H., Wu, C., Wu, D., and Yu, J.: Size distributions of elemental carbon and its contribution to light extinction in urban and rural locations in the pearl river delta region, China, Atmospheric Chemistry and Physics, 10, 5107-5119, 2010.

Zha, S., Cheng, T., Tao, J., Zhang, R., Chen, J., Zhang, Y., Leng, C., Zhang, D., and Du, J.: Characteristics and relevant remote sources of black carbon aerosol in Shanghai, Atmospheric Research, 135, 159-171, 2014.

Zhang, F., Zhao, J., Chen, J., Xu, Y., and Xu, L.: Pollution characteristics of organic and elemental carbon in $PM_{2.5}$ in Xiamen, China, Journal of Environmental Sciences, 23, 1342-1349, 2011a.

Zhang, F., Xu, L., Chen, J., Yu, Y., Niu, Z., and Yin, L.: Chemical compositions and extinction coefficients of $PM_{2.5}$ in peri-urban of Xiamen, China, during June 2009-May 2010, Atmospheric Research, 106, 150-158, 2012a.

Zhang, F., Wang, Z., Cheng, H., Lv, X., Gong, W., Wang, X., and Zhang, G.: Seasonal variations and chemical characteristics of $PM_{2.5}$ in Wuhan, central China, Science of The Total Environment, 518–519, 97-105, http://dx.doi.org/10.1016/j.scitotenv.2015.02.054, 2015a.

Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, Atmospheric Chemistry and Physics, 15, 8439-8454, 10.5194/acp-15-8439-2015, 2015b.

Zhang, N., Zhuang, M., Tian, J., Tian, P., Zhang, J., Wang, Q., Zhou, Y., Huang, R., Zhu, C., Zhang, X., and Cao, J.: Development of source profiles and their application in source apportionment of $PM_{2.5}$ in Xiamen, China, Frontiers of Environmental Science & Engineering, 10, 17, 10.1007/s11783-016-0879-1, 2016.

Zhang, Q., Geng, G., Wang, S., Richter, A., and He, K.: Satellite remote sensing of changes in NO_x emissions over China during 1996–2010, Chinese Science Bulletin, 57, 2857-2864, 10.1007/s11434-012-5015-4, 2012b.

Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective, Atmospheric Chemistry and Physics, 13, 7053-7074, 10.5194/acp-13-7053-2013, 2013a.

Zhang, R., Li, Q., and Zhang, R.: Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013, Science China Earth Sciences, 57, 26-35, 10.1007/s11430-013-4774-3, 2014a.

Zhang, T., Cao, J., Tie, X., Shen, Z., Liu, S., Ding, H., Han, Y., Wang, G., Ho, K., and Qiang, J.: Water-soluble ions in atmospheric aerosols measured in Xi'an, China: seasonal variations and sources, Atmospheric Research, 102, 110-119, 2011b.

Zhang, T., Cao, J., Chow, J. C., Shen, Z., Ho, K., Ho, S. S. H., Liu, S., Han, Y., Watson, J. G., and Wang, G.: Characterization and seasonal variations of levoglucosan in fine particulate matter in Xi'an, China, Journal of The Air & Waste Management Association, 64, 1317-1327, 2014b.

Zhang, W., Guo, J., Sun, Y., Yuan, H., Zhuang, G., Zhuang, Y., and Hao, Z.: Source apportionment for urban PM_{10} and $PM_{2.5}$ in the Beijing area, Chinese Science Bulletin, 52, 608-615, 10.1007/s11434-007-0076-5, 2007.

Zhang, X., Wu, L., Zhang, R., Deng, S., Zhang, Y., Wu, J., Li, Y., Lin, L., Li, L., Wang, Y., and Wang, L.: Evaluating the relationships among economic growth, energy consumption, air emissions and air environmental protection investment in China, Renewable and Sustainable Energy Reviews, 18, 259-270, http://dx.doi.org/10.1016/j.rser.2012.10.029, 2013b.

Zhang, X., Du J., Huang T., Zhang L., Gao H., Zhao Y., and Ma J.: Atmospheric removal of $PM_{2.5}$ by man-made three northern regions shelter forest in northern China estimated using satellite retrieved $PM_{2.5}$ concentrations, Science of the Total Environment, 593-594, 713-721, 2017b.

Zhang, Y., Cai, J., Wang, S., He, K., and Zheng, M.: Review of receptor-based source apportionment research of fine particulate matter and its challenges in China, Science of The Total Environment, 586, 917-929, https://doi.org/10.1016/j.scitotenv.2017.02.071, 2017a.

Zhao, B., Wang, S., Wang, J., Fu, J. S., Liu, T., Xu, J., Fu, X., and Hao, J.: Impact of national NO_x and SO_2 control policies on particulate matter pollution in China, Atmospheric Environment, 77, 453-463, 2013a.

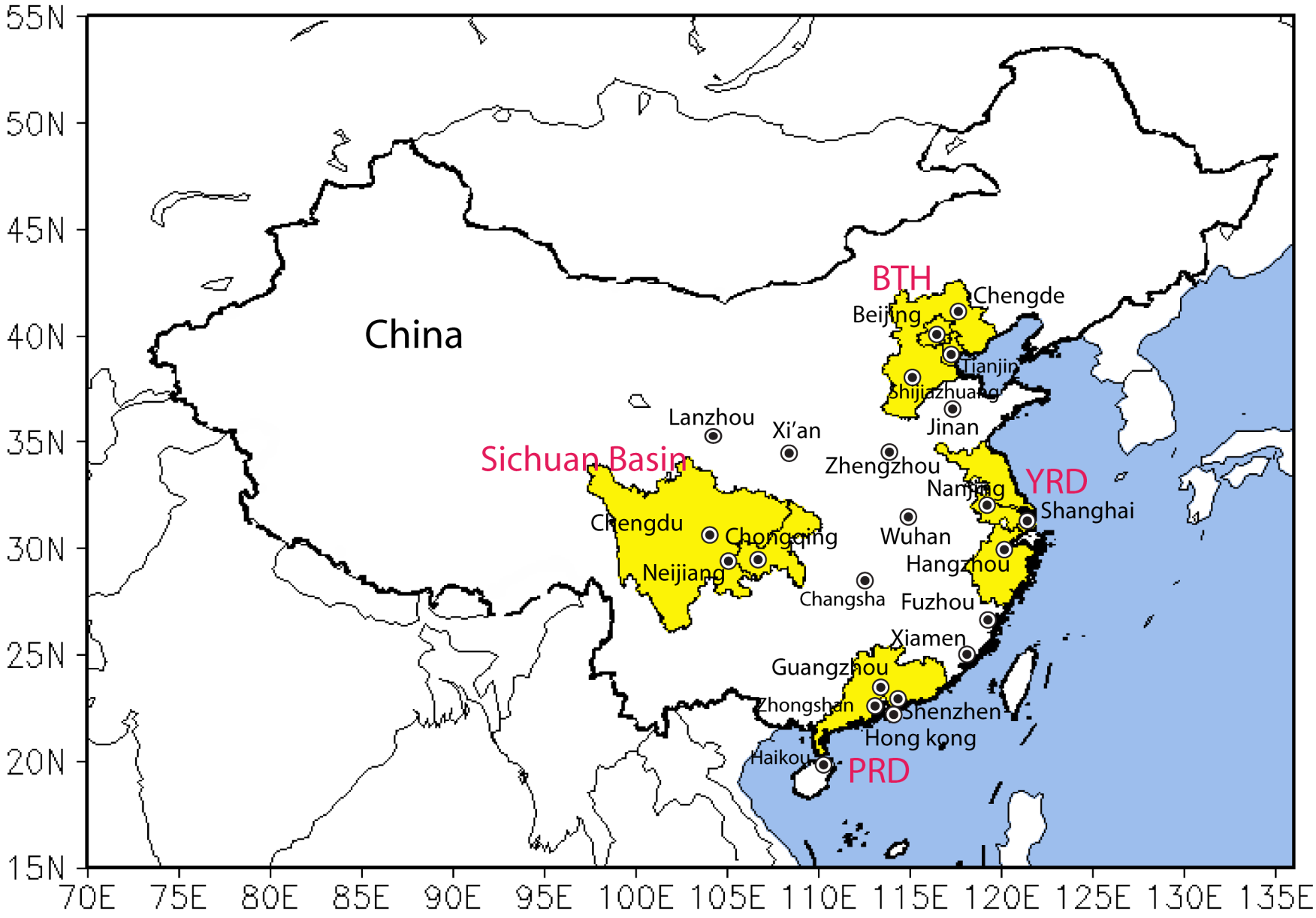
Zhao, B., Wang, S. X., Liu, H., Xu, J., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NO_x emissions in China: historical trends and future perspectives, Atmospheric Chemistry and Physics, 13, 9869-9897, 2013b.

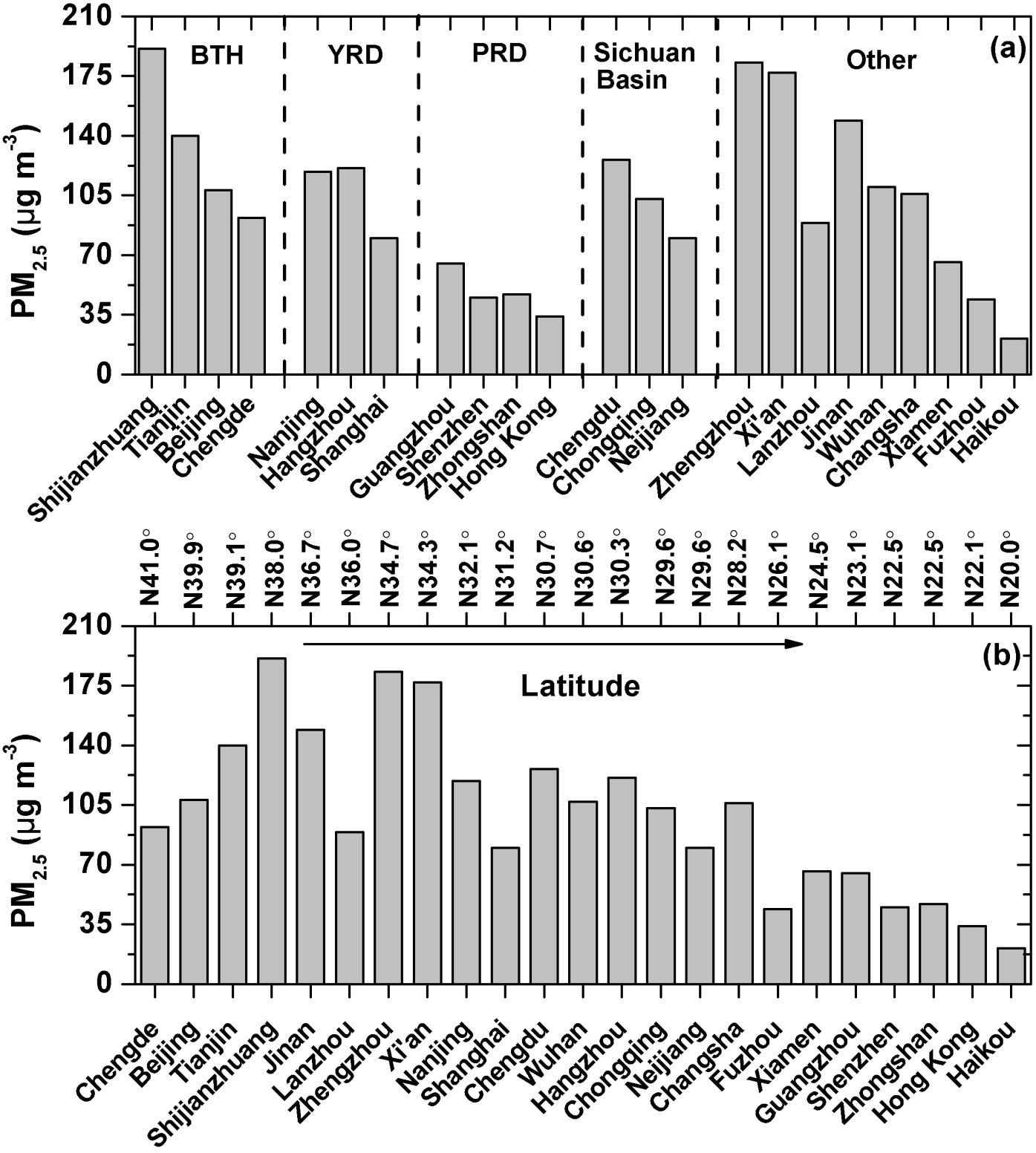
Zhao, M., Huang, Z., Qiao, T., Zhang, Y., Xiu, G., and Yu, J.: Chemical characterization, the transport pathways and potential sources of $PM_{2.5}$ in Shanghai: Seasonal variations, Atmospheric Research, 158, 66-78, 2015a.

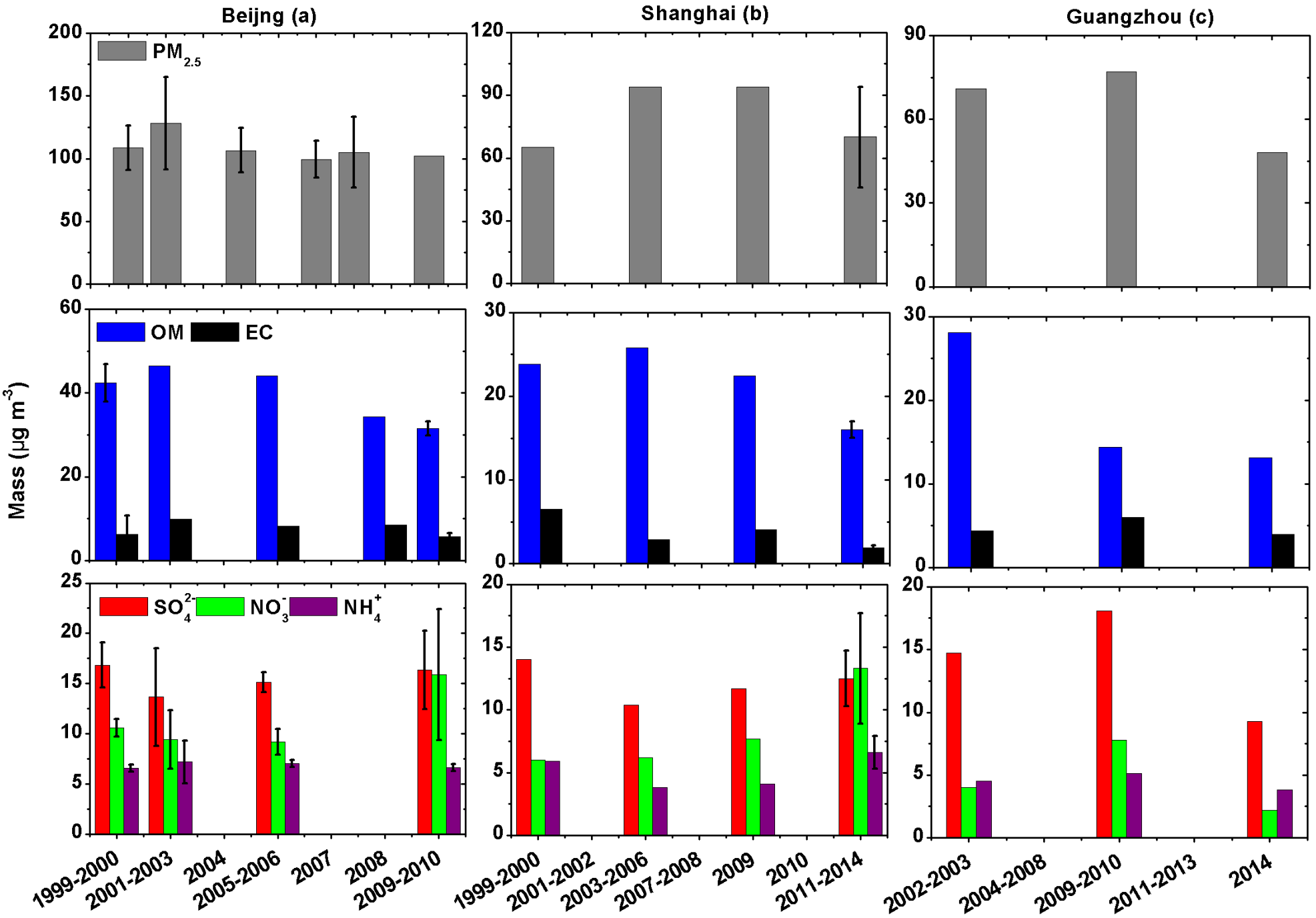
Zhao, M., Qiao, T., Huang, Z., Zhu, M., Xu, W., Xiu, G., Tao, J., and Lee, S.: Comparison of ionic and carbonaceous compositions of $PM_{2.5}$ in 2009 and 2012 in Shanghai, China, Science of The Total

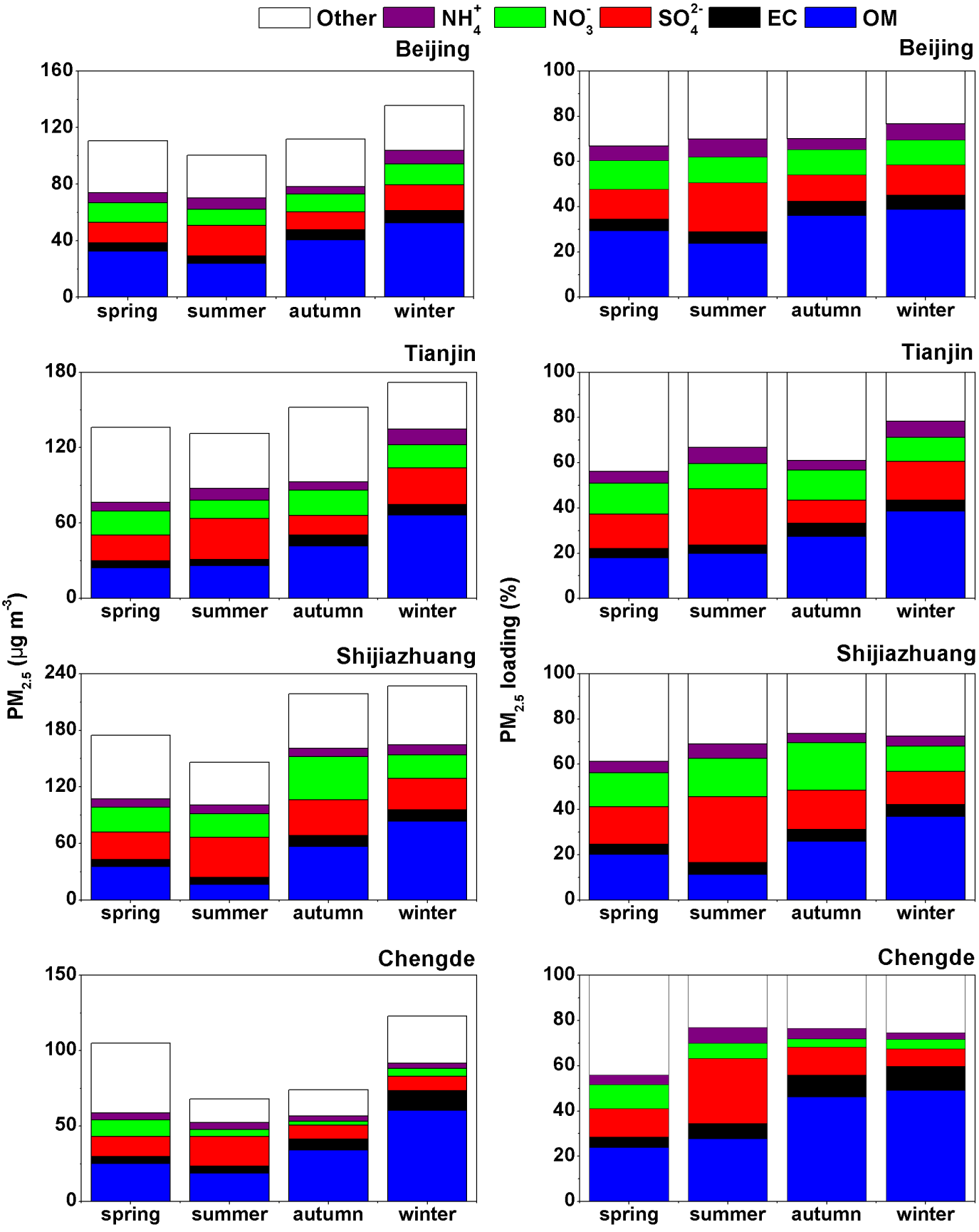
Environment, 536, 695-703, http://dx.doi.org/10.1016/j.scitotenv.2015.07.100, 2015b.
Zhao, P., Dong, F., He, D., Zhao, X., Zhang, X., Zhang, W., Yao, Q., and Liu, H.: Characteristics of concentrations and chemical compositions for $PM_{2.5}$ in the region of Beijing, Tianjin, and Hebei, China, Atmospheric 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.
Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W., Dong, F., He, D., and Shi, Q. F.: Analysis of a winter regional haze event and its formation mechanism in the North China Plain, Atmospheric Chemistry and Physics, 13, 5685-5696, 2013d.
Zheng, J., Hu, M., Peng, J., Wu, Z., Kumar, P., Li, M., Wang, Y., and Guo, S.: Spatial distributions and chemical properties of $PM_{2.5}$ based on 21 field campaigns at 17 sites in China, Chemosphere, 159, 480-487, 2016.
Zheng, J. Y., Yin, S. S., Kang, D. W., Che, W. W., and Zhong, L. J.: Development and uncertainty analysis of a high-resolution 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.
Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., and Cass, G. R.: Seasonal trends in $PM_{2.5}$ source contributions in Beijing, China, Atmospheric Environment, 39, 3967-3976, 2005.
Zhou, B., Shen, H., Huang, Y., Li, W., Chen, H., Zhang, Y., Su, S., Chen, Y., Lin, N., Zhuo, S., Zhong, Q., Liu, J., Li, B., and Tao, S.: Daily variations of size-segregated ambient particulate matter in Beijing, 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.
Zíková, N., Wang, Y., Yang, F., Li, X., Tian, M., and Hopke, P. K.: On the source contribution to Beijing $PM_{2.5}$ concentrations, Atmospheric Environment, 134, 84-95, http://doi.org/10.1016/j.atmosenv.2016.03.047, 2016.
Zong, Z., Wang, X., Tian, C., Chen, Y., Qu, L., Ji, L., Zhi, G., Li, J., and Zhang, G.: Source apportionment of $PM_{2.5}$ at a regional background site in North China using PMF linked with radiocarbon analysis: insight into the contribution of biomass burning, Atmospheric Chemistry and Physics, 16, 11249-11265, 10.5194/acp-16-11249-2016, 2016.
Zou, Y., Wang, Y., Zhang, Y., and Koo, J.H.: Arctic sea ice, Eurasia snow, and extreme winter haze in China, Science Advances, 3, 10.1126/sciadv.1602751, 2017.

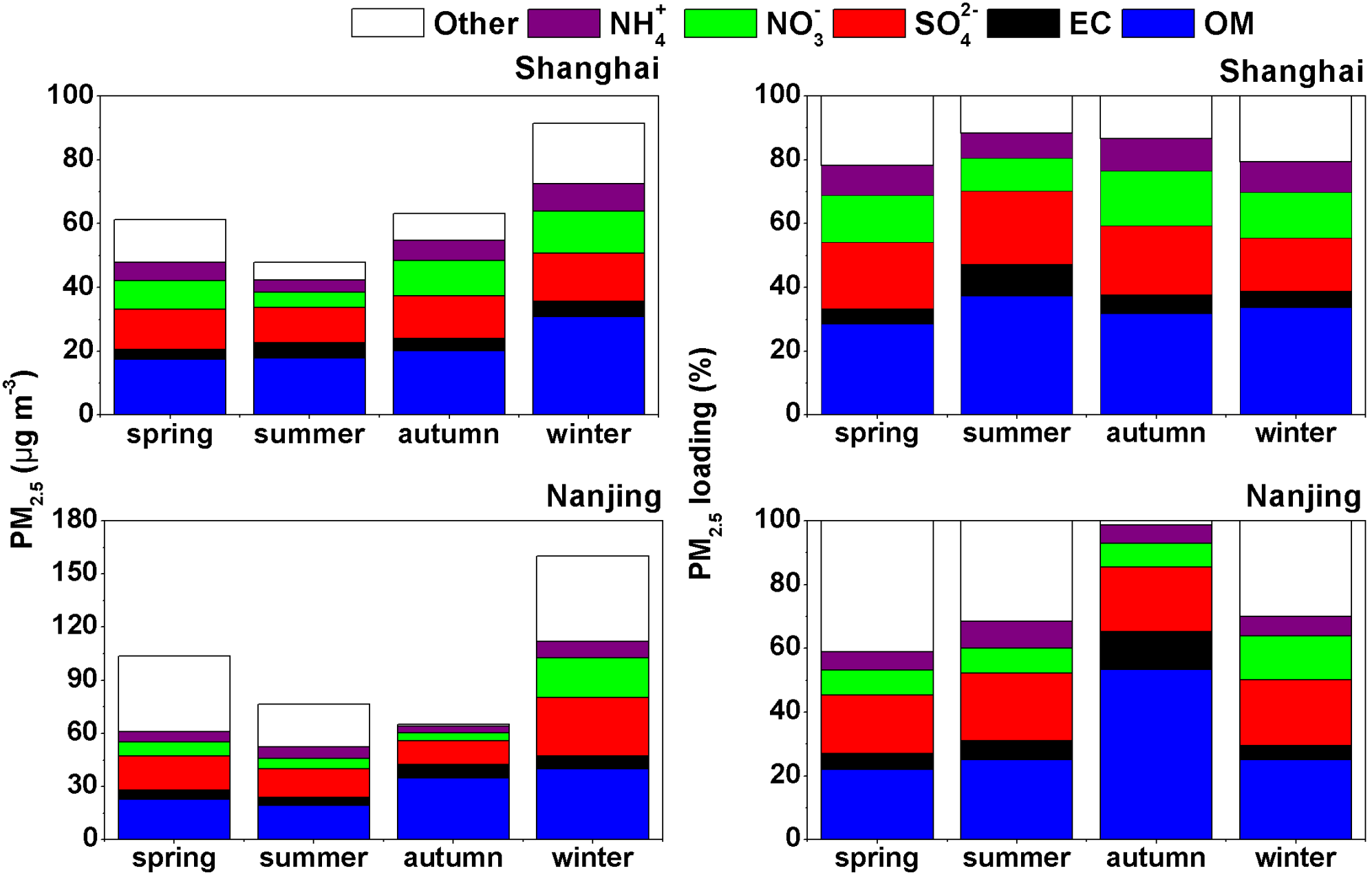
List of Figures

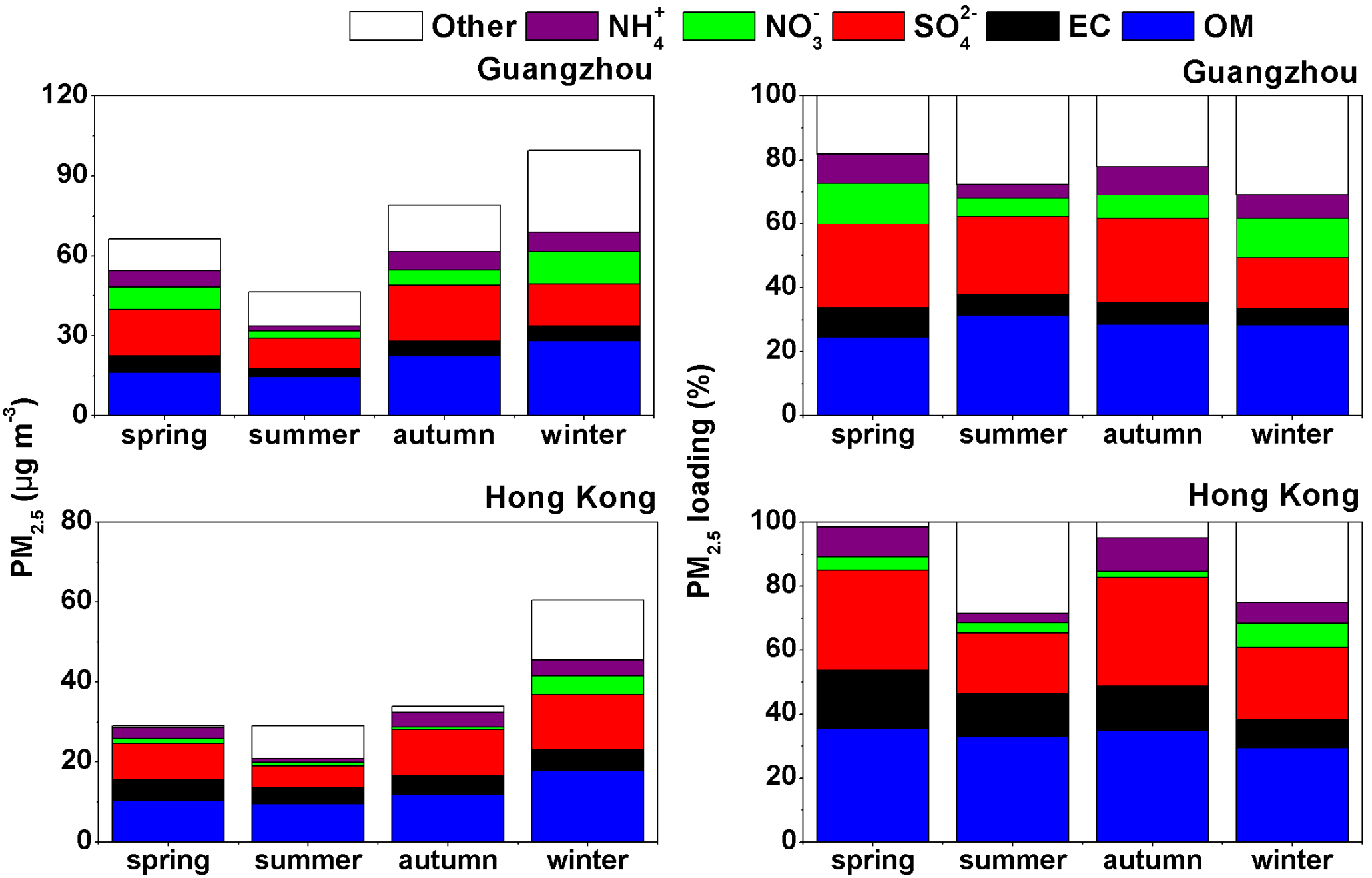

55N
50N
45N
40N
35N
30N
25N
20N
15N
70E
75E
80E
85E
90E
95E
100E
105E
110E
115E
120E
125E
130E
135E
China
BTH
Chengde
Beijing
Tianjin
Shijiazhuang
Lanzhou
Xi'an
Jinan
Sichuan Basin
Zhengzhou
YRD
Nanjing
Shanghai
Chengdu
Chongqing
Wuhan
Hangzhou
Neijiang
Changsha
Fuzhou
Xiamen
Guangzhou
Zhongshan
Shenzhen
Hong kong
Haikou
PRD

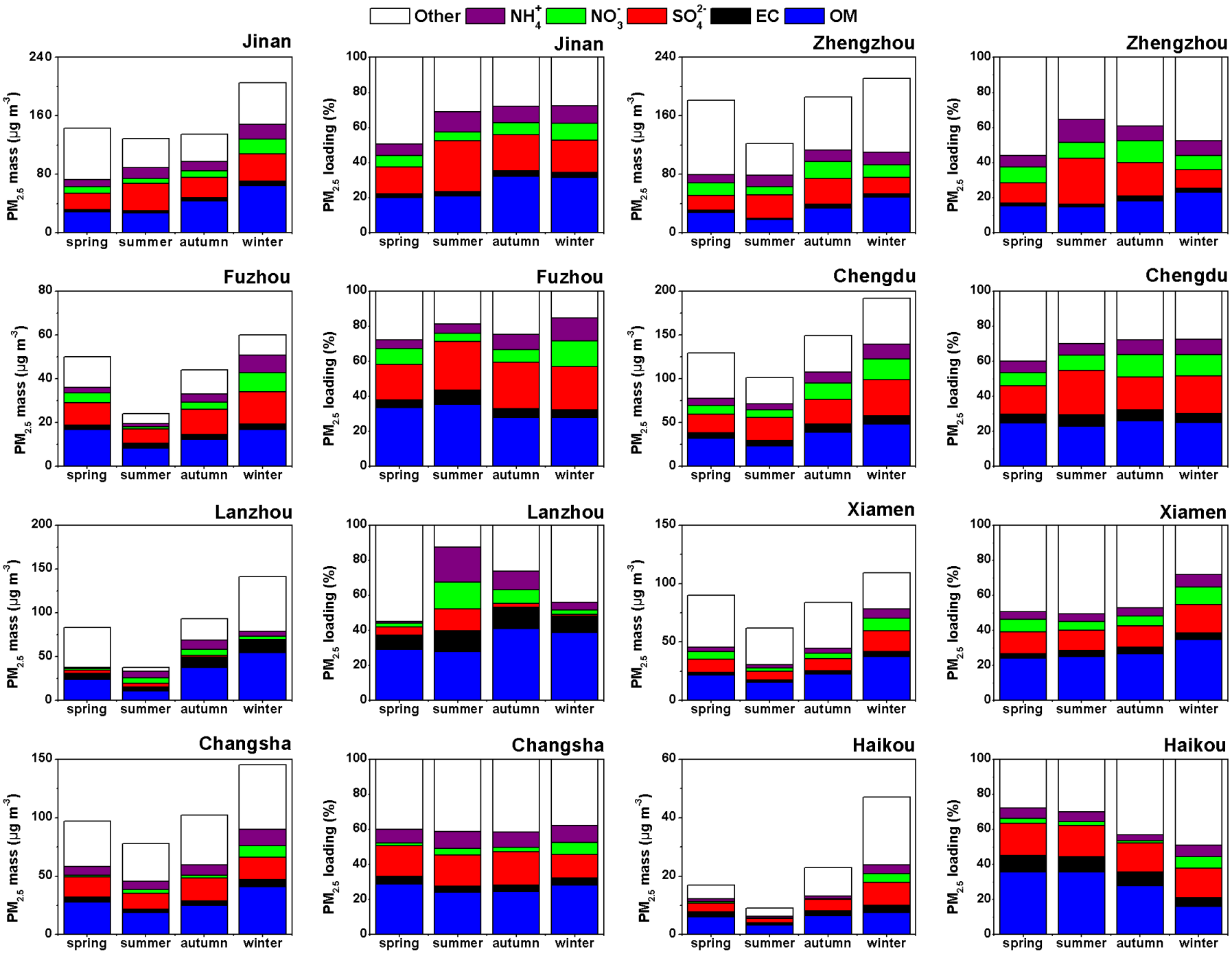

(a)
BTH
YRD
PRD
Sichuan Basin
Other
PM2.5 (μg m-3)
210
175
140
105
70
35
0
Shijianzhuang
Tianjin
Beijing
Chengde
Nanjing
Hangzhou
Shanghai
Guangzhou
Shenzhen
Zhongshan
Hong Kong
Chengdu
Chongqing
Neijiang
Zhengzhou
Xi'an
Lanzhou
Jinan
Wuhan
Changsha
Xiamen
Fuzhou
Haikou
(b)
Latitude
N41.0°
N39.9°
N39.1°
N38.0°
N36.7°
N36.0°
N34.7°
N34.3°
N32.1°
N31.2°
N30.7°
N30.6°
N30.3°
N29.6°
N29.6°
N28.2°
N26.1°
N24.5°
N23.1°
N22.5°
N22.5°
N22.1°
N20.0°
Chengde
Beijing
Tianjin
Shijianzhuang
Jinan
Lanzhou
Zhengzhou
Xi'an
Nanjing
Shanghai
Chengdu
Wuhan
Hangzhou
Chongqing
Neijiang
Changsha
Fuzhou
Xiamen
Guangzhou
Shenzhen
Zhongshan
Hong Kong
Haikou

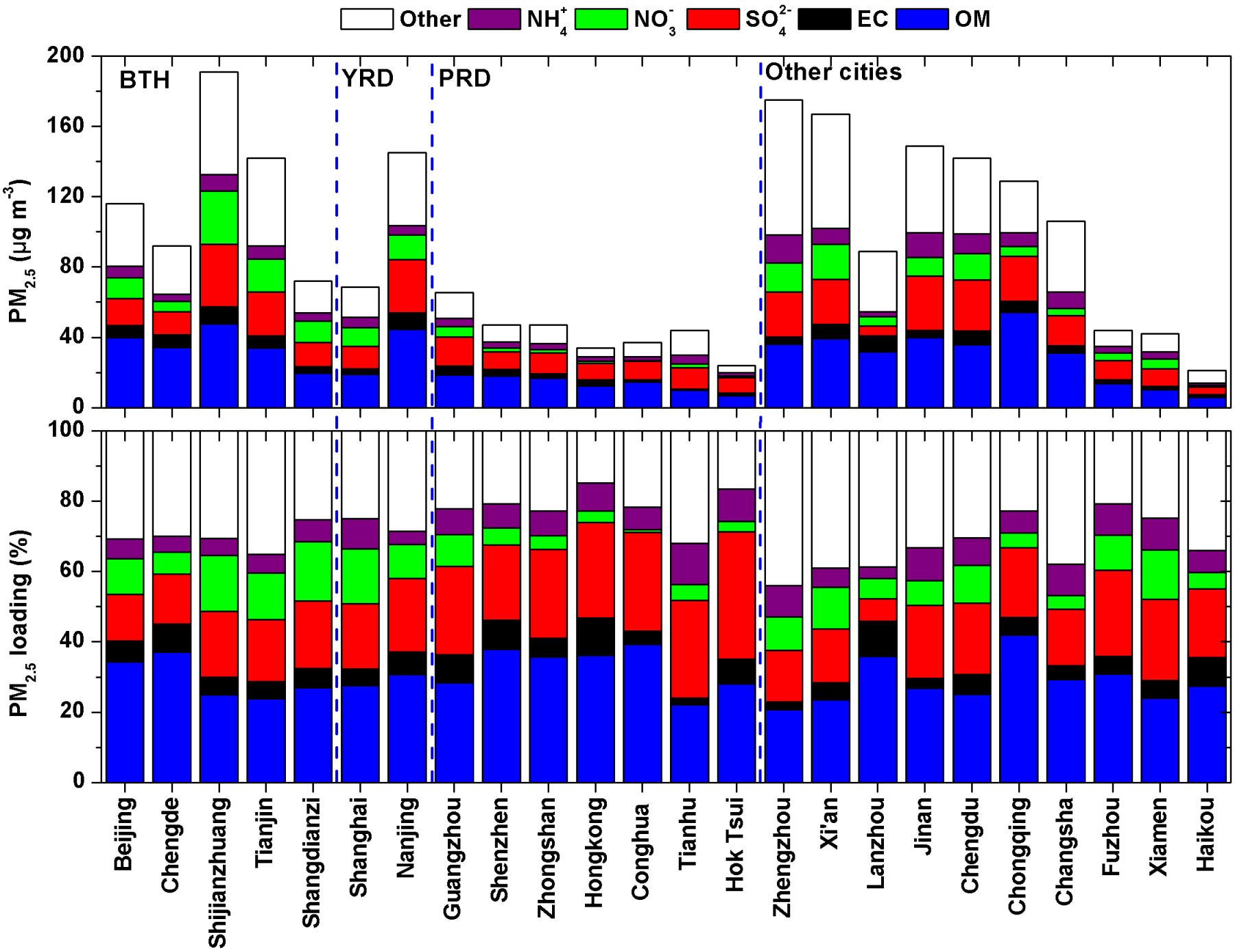

Beijng (a)
Shanghai (b)
Guangzhou (c)
PM2.5
OM
EC
SO4 2-
NO3 -
NH4 +
Mass (μg m-3)
1999-2000
2001-2003
2004
2005-2006
2007
2008
2009-2010
1999-2000
2001-2002
2003-2006
2007-2008
2009
2010
2011-2014
2002-2003
2004-2008
2009-2010
2011-2013
2014

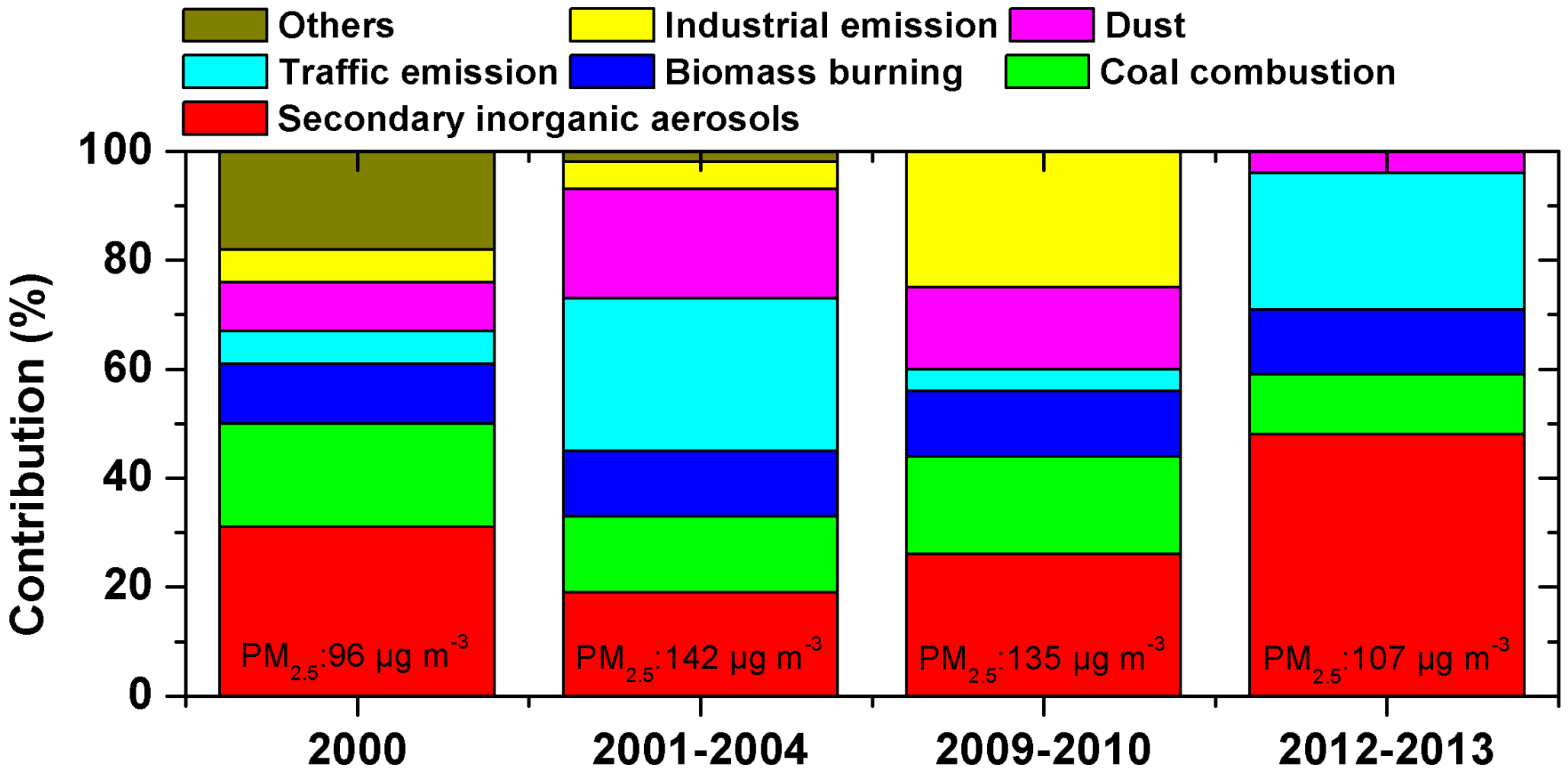

Other
NH4+
NO3-
SO42-
EC
OM
Beijing
Tianjin
Shijiazhuang
Chengde
spring
summer
autumn
winter
PM2.5 (μg m-3)
PM2.5 loading (%)

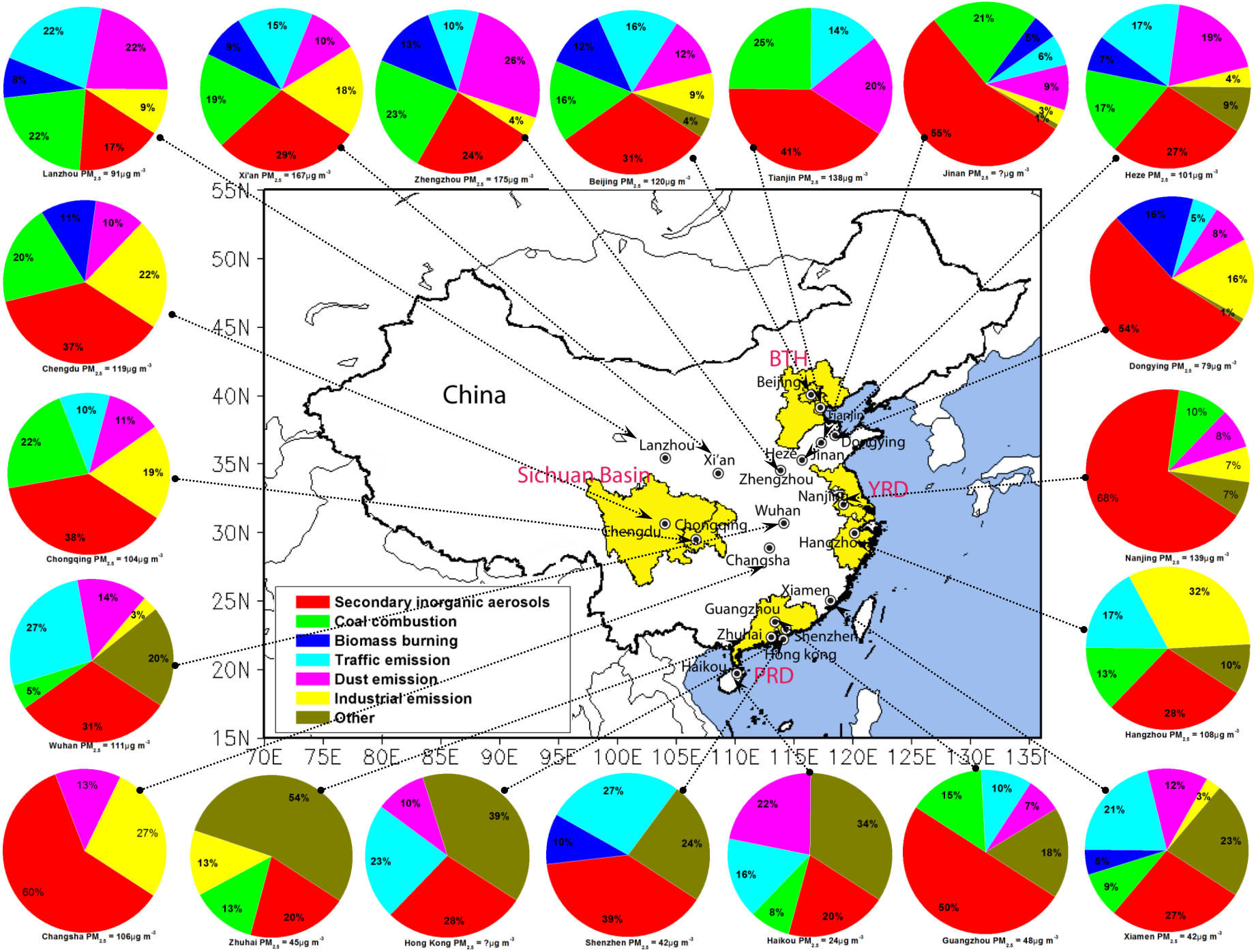

Other
NH4+
NO3-
SO42-
EC
OM
Shanghai
Nanjing
PM2.5 (μg m-3)
PM2.5 loading (%)
spring
summer
autumn
winter
0
20
40
60
80
100
30
90
120
150
180

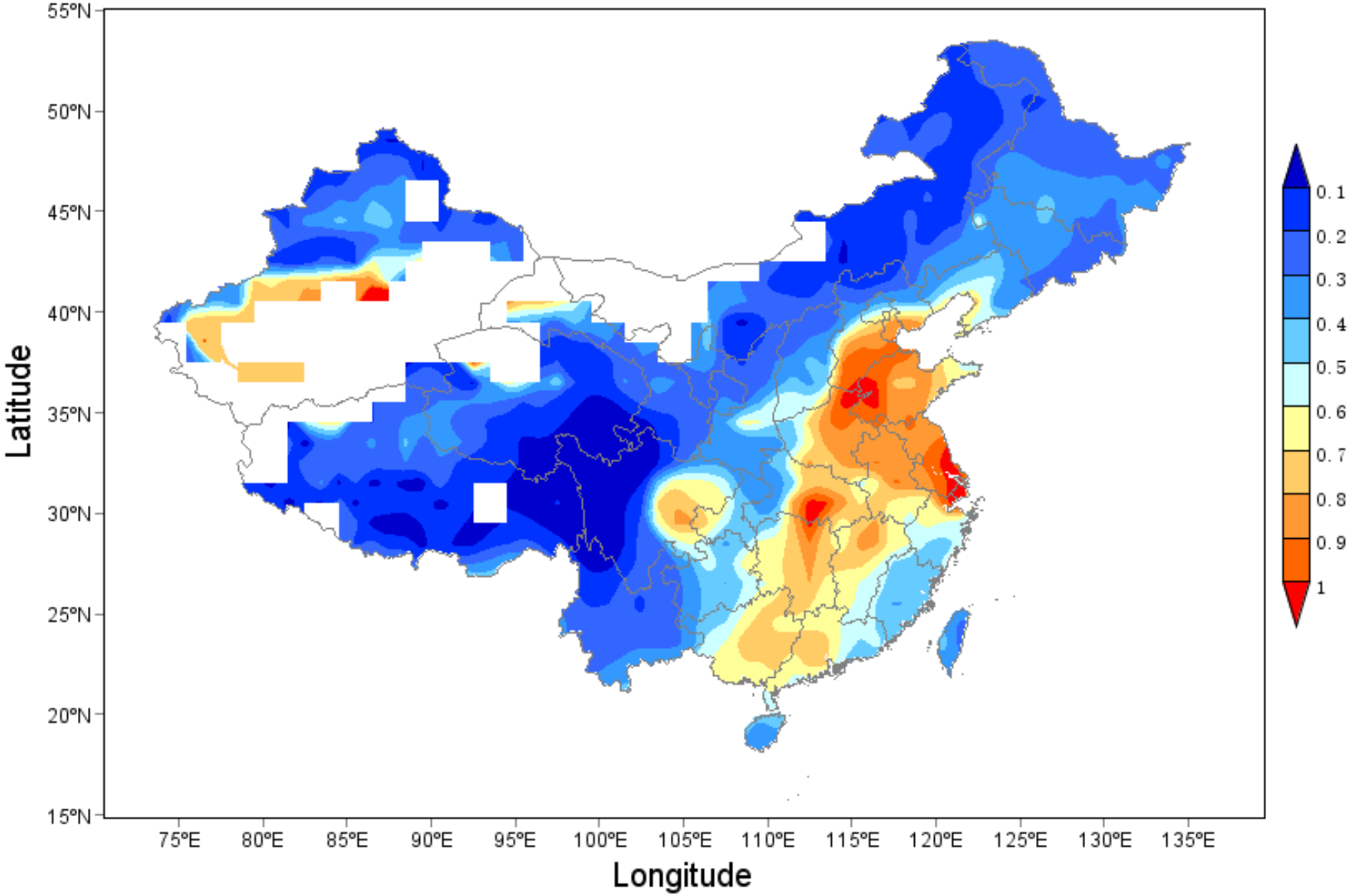

Other
NH4+
NO3-
SO42-
EC
OM
Guangzhou
Hong Kong
PM2.5 (μg m-3)
PM2.5 loading (%)
spring
summer
autumn
winter
0
20
30
40
60
80
90
100
120

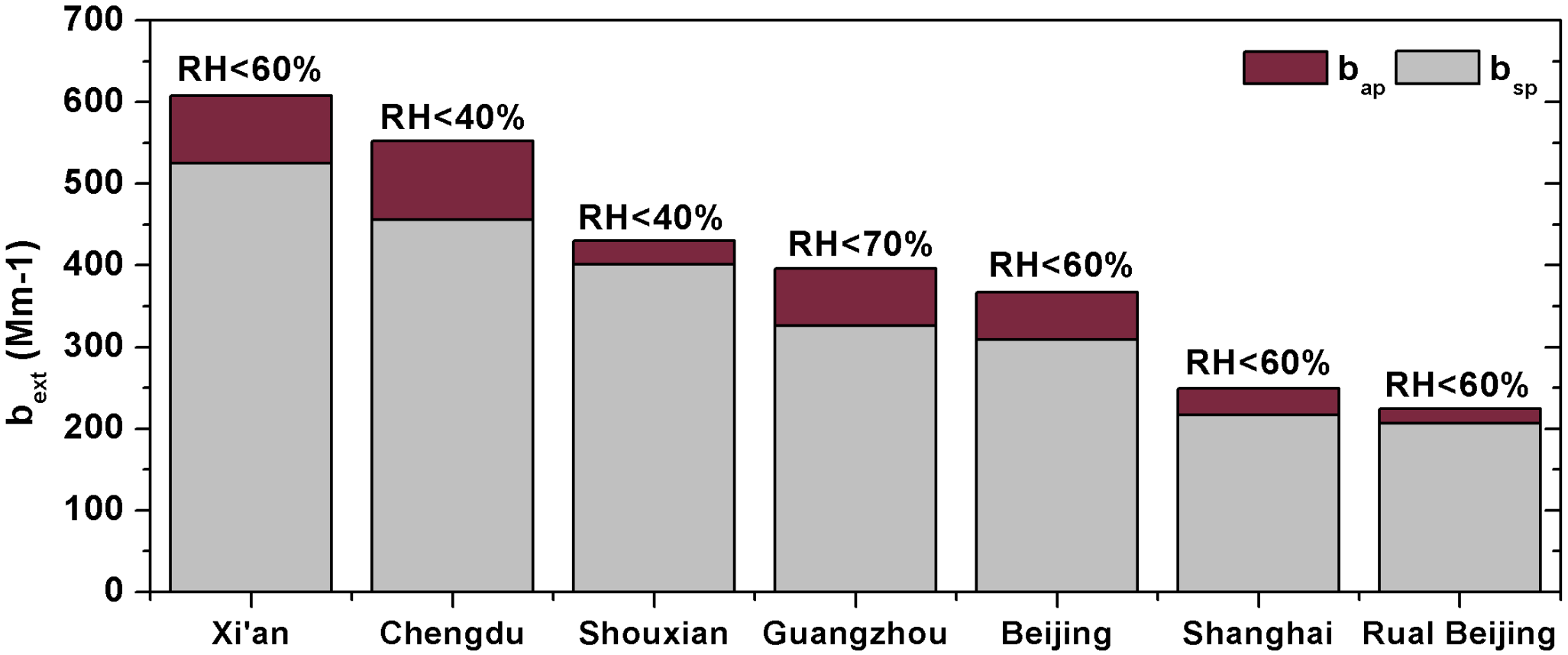

Other
NH4+
NO3-
SO42-
EC
OM
Jinan
Jinan
Zhengzhou
Zhengzhou
Fuzhou
Fuzhou
Chengdu
Chengdu
Lanzhou
Lanzhou
Xiamen
Xiamen
Changsha
Changsha
Haikou
Haikou
PM2.5 mass (μg m-3)
PM2.5 loading (%)
spring
summer
autumn
winter

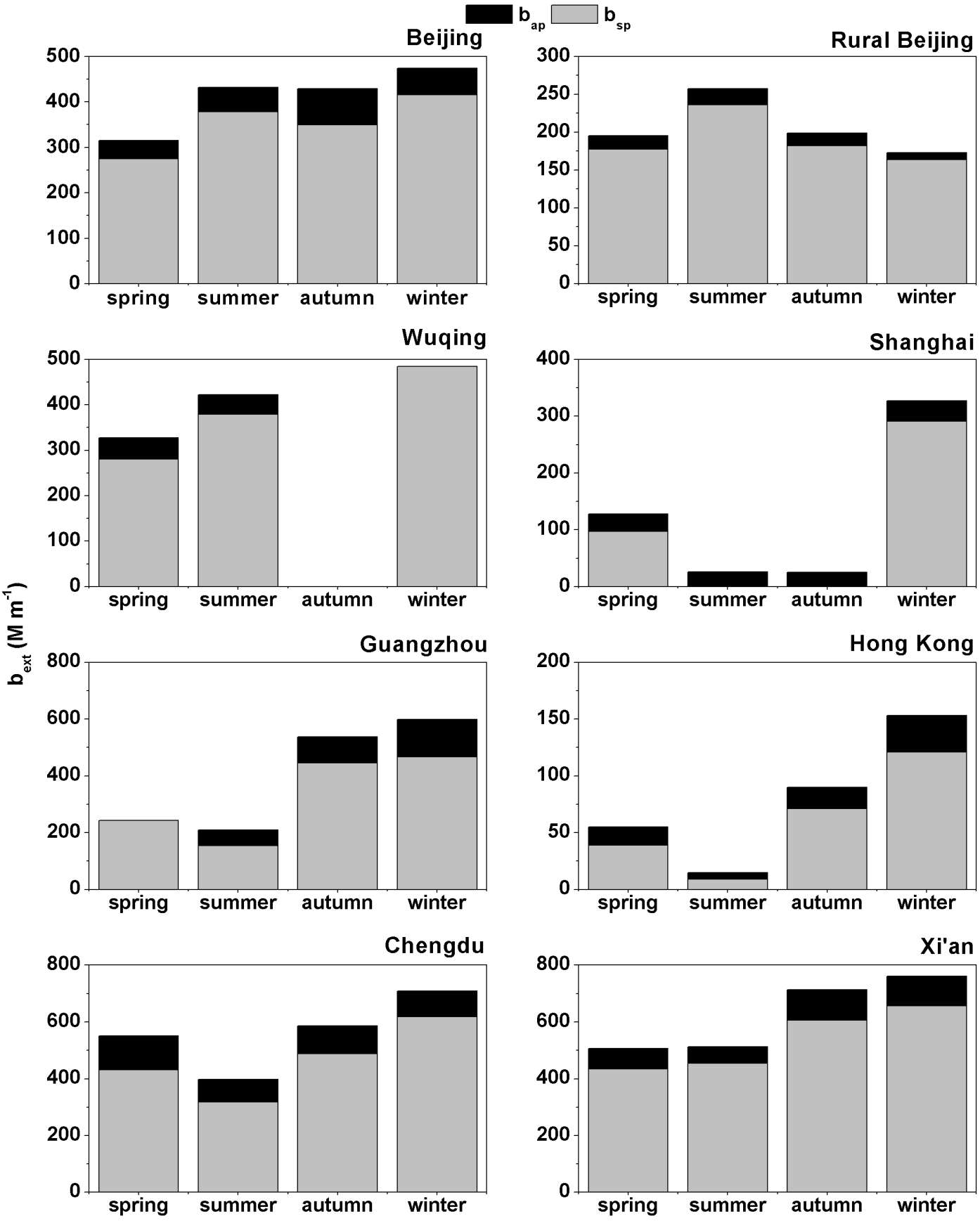

Other
NH4+
NO3-
SO42-
EC
OM
BTH
YRD
PRD
Other cities
PM2.5 (μg m-3)
PM2.5 loading (%)
0
40
80
120
160
200
0
20
40
60
80
100
Beijing
Chengde
Shijianzhuang
Tianjin
Shangdianzi
Shanghai
Nanjing
Guangzhou
Shenzhen
Zhongshan
Hongkong
Conghua
Tianhu
Hok Tsui
Zhengzhou
Xi'an
Lanzhou
Jinan
Chengdu
Chongqing
Changsha
Fuzhou
Xiamen
Haikou

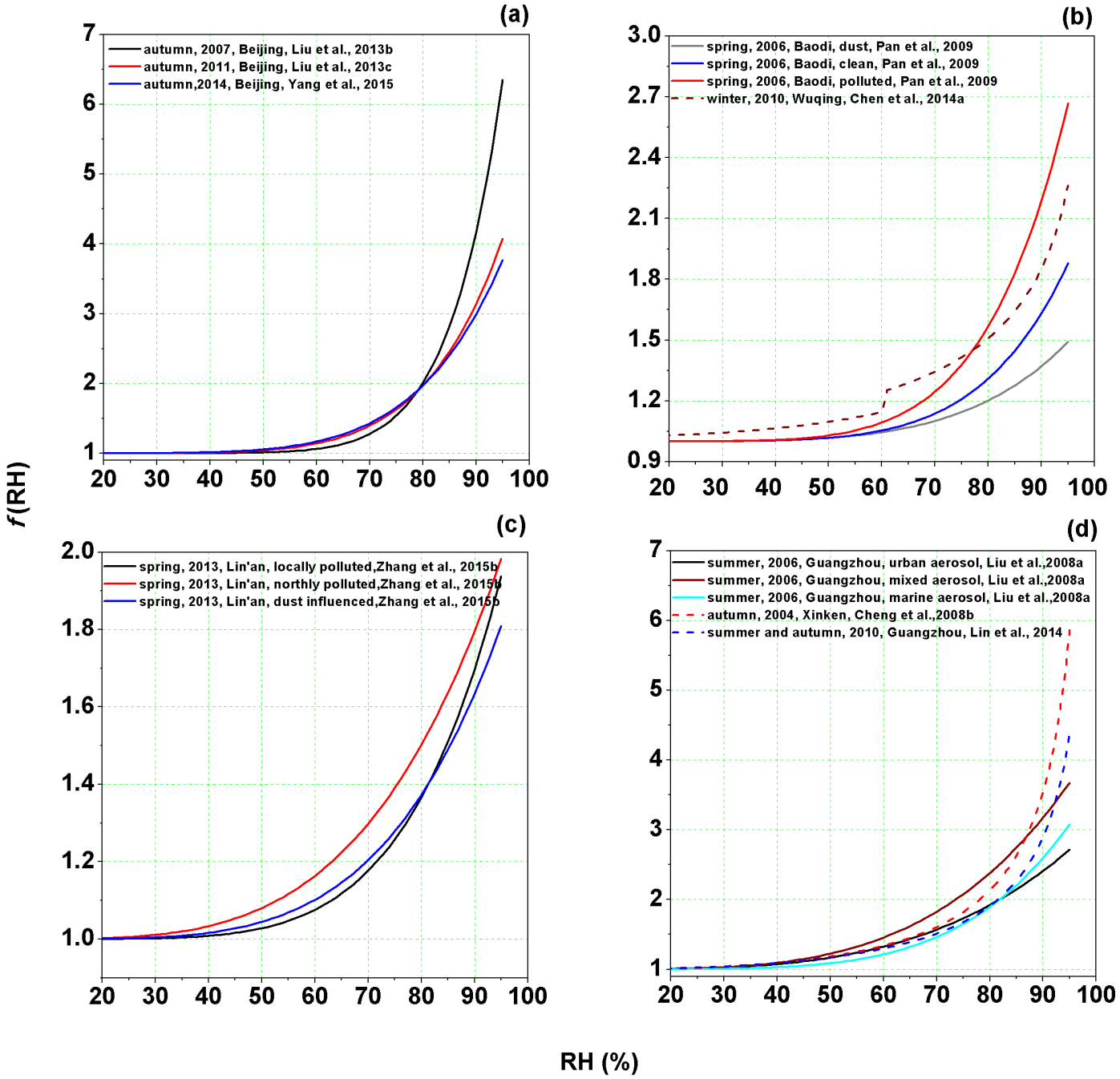

Others
Industrial emission
Dust
Traffic emission
Biomass burning
Coal combustion
Secondary inorganic aerosols
Contribution (%)
100
80
60
40
20
0
PM2.5:96 μg m-3
PM2.5:142 μg m-3
PM2.5:135 μg m-3
PM2.5:107 μg m-3
2000
2001-2004
2009-2010
2012-2013

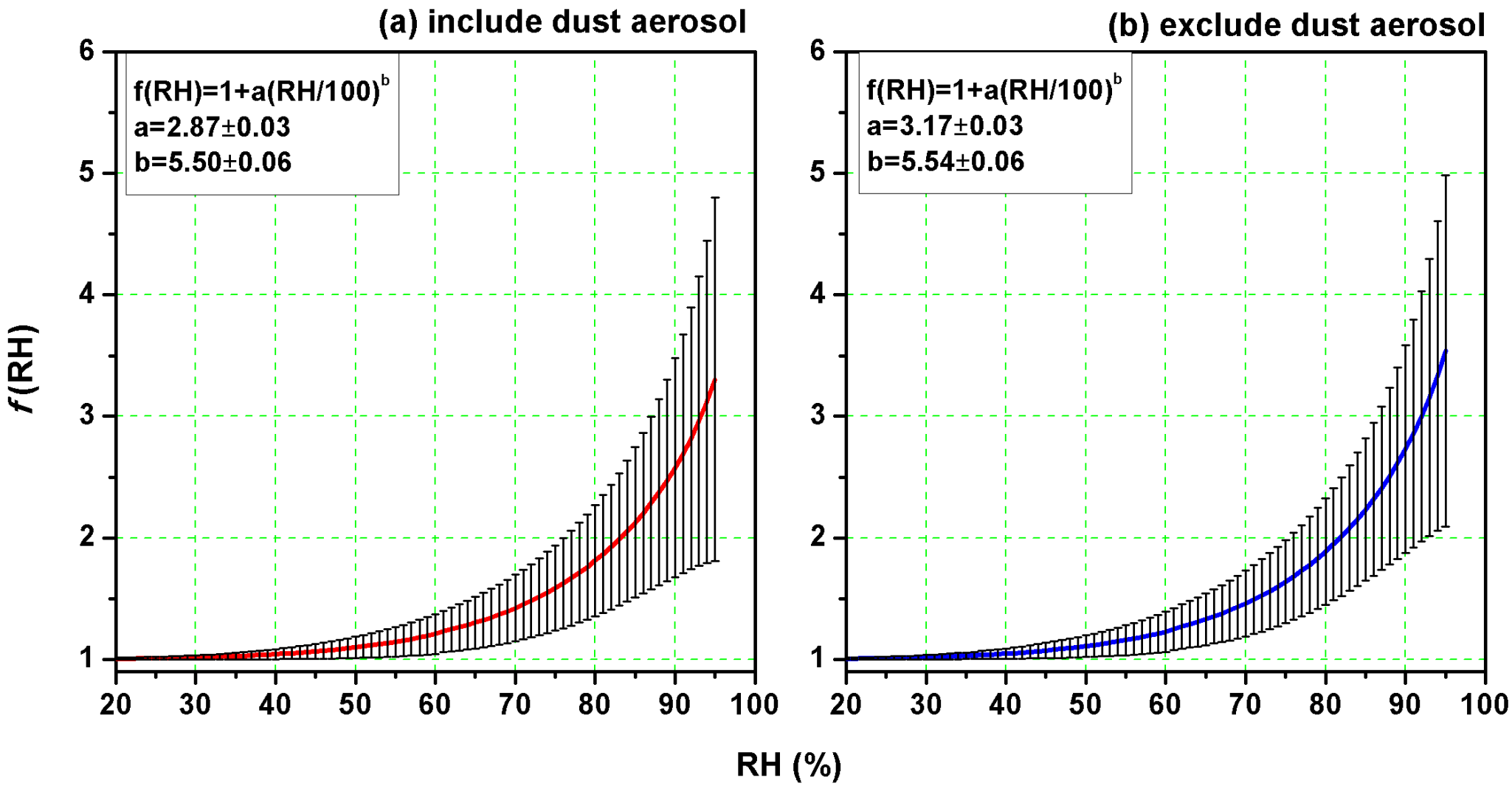

Lanzhou PM2.5 = 91μg m-3
Xi'an PM2.5 = 167μg m-3
Zhengzhou PM2.5 = 175μg m-3
Beijing PM2.5 = 120μg m-3
Tianjin PM2.5 = 138μg m-3
Jinan PM2.5 = ?μg m-3
Heze PM2.5 = 101μg m-3
Chengdu PM2.5 = 119μg m-3
Dongying PM2.5 = 79μg m-3
Chongqing PM2.5 = 104μg m-3
Nanjing PM2.5 = 139μg m-3
Wuhan PM2.5 = 111μg m-3
Hangzhou PM2.5 = 108μg m-3
Changsha PM2.5 = 106μg m-3
Zhuhai PM2.5 = 45μg m-3
Hong Kong PM2.5 = ?μg m-3
Shenzhen PM2.5 = 42μg m-3
Haikou PM2.5 = 24μg m-3
Guangzhou PM2.5 = 48μg m-3
Xiamen PM2.5 = 42μg m-3
China
Sichuan Basin
BTH
YRD
PRD
Beijing
Tianjin
Dongying
Heze
Jinan
Zhengzhou
Lanzhou
Xi'an
Nanjing
Wuhan
Chongqing
Chengdu
Hangzhou
Changsha
Xiamen
Guangzhou
Zhuhai
Shenzhen
Hong kong
Haikou
Secondary inorganic aerosols
Coal combustion
Biomass burning
Traffic emission
Dust emission
Industrial emission
Other
55N
50N
45N
40N
35N
30N
25N
20N
15N
70E
75E
80E
85E
90E
95E
100E
105E
110E
115E
120E
125E
130E
135E

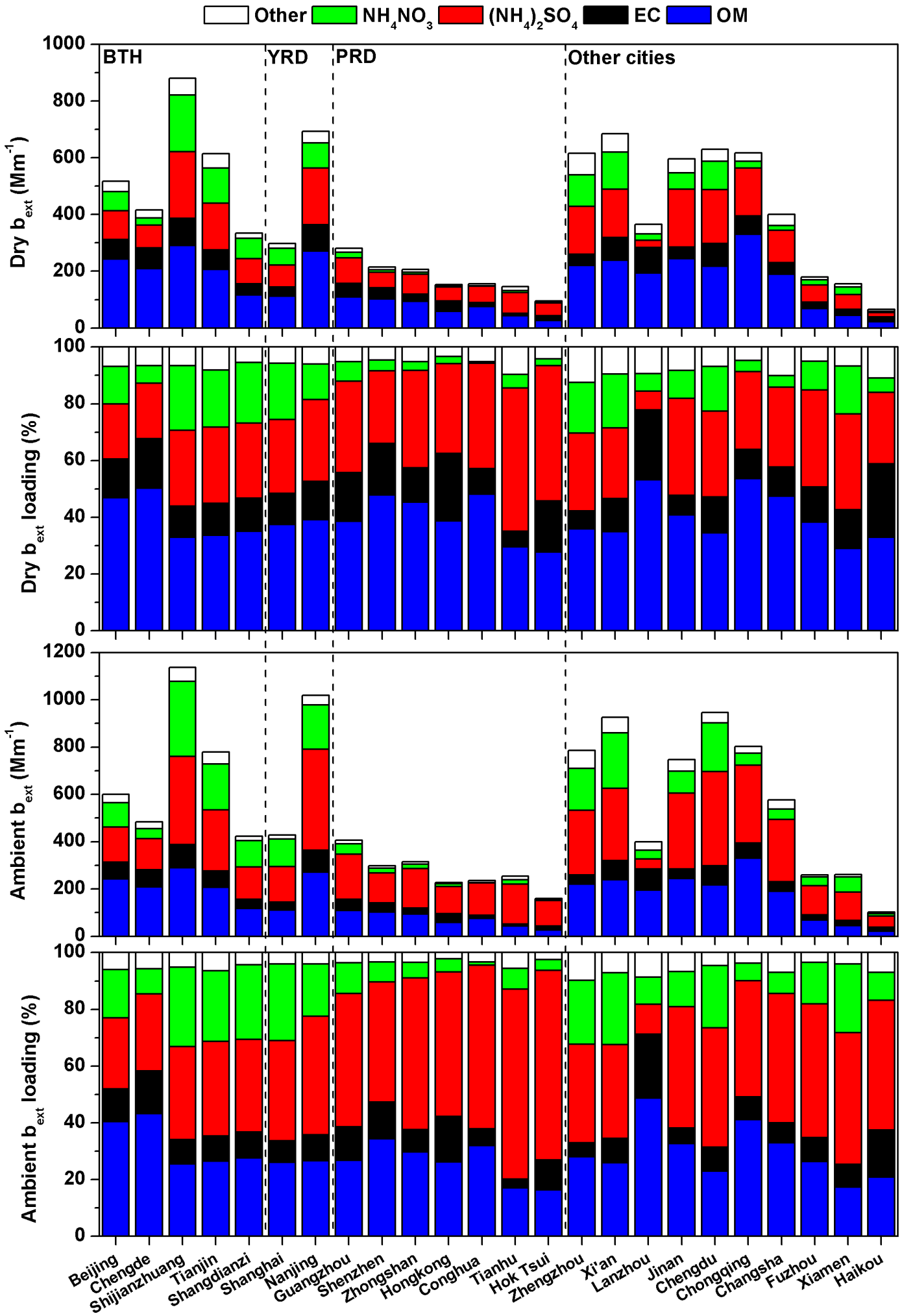

55°N
50°N
45°N
40°N
35°N
30°N
25°N
20°N
15°N
Latitude
75°E
80°E
85°E
90°E
95°E
100°E
105°E
110°E
115°E
120°E
125°E
130°E
135°E
Longitude
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1


700
600
500
400
300
200
100
0
b_ext (Mm-1)
b_ap
b_sp
RH<60%
RH<40%
RH<40%
RH<70%
RH<60%
RH<60%
RH<60%
Xi'an
Chengdu
Shouxian
Guangzhou
Beijing
Shanghai
Rual Beijing


b_{ap}
b_{sp}
Beijing
Rural Beijing
Wuqing
Shanghai
Guangzhou
Hong Kong
Chengdu
Xi'an
b_{ext} (M m^{-1})
spring
summer
autumn
winter


(a)
autumn, 2007, Beijing, Liu et al., 2013b
autumn, 2011, Beijing, Liu et al., 2013c
autumn,2014, Beijing, Yang et al., 2015
(b)
spring, 2006, Baodi, dust, Pan et al., 2009
spring, 2006, Baodi, clean, Pan et al., 2009
spring, 2006, Baodi, polluted, Pan et al., 2009
winter, 2010, Wuqing, Chen et al., 2014a
(c)
spring, 2013, Lin'an, locally polluted,Zhang et al., 2015b
spring, 2013, Lin'an, northly polluted,Zhang et al., 2015b
spring, 2013, Lin'an, dust influenced,Zhang et al., 2015b
(d)
summer, 2006, Guangzhou, urban aerosol, Liu et al.,2008a
summer, 2006, Guangzhou, mixed aerosol, Liu et al.,2008a
summer, 2006, Guangzhou, marine aerosol, Liu et al.,2008a
autumn, 2004, Xinken, Cheng et al.,2008b
summer and autumn, 2010, Guangzhou, Lin et al., 2014
f(RH)
RH (%)


(a) include dust aerosol
f(RH)=1+a(RH/100)^b
a=2.87±0.03
b=5.50±0.06
(b) exclude dust aerosol
f(RH)=1+a(RH/100)^b
a=3.17±0.03
b=5.54±0.06
f(RH)
RH (%)
1
2
3
4
5
6
20
30
40
50
60
70
80
90
100


Other
NH4NO3
(NH4)2SO4
EC
OM
BTH
YRD
PRD
Other cities
Dry bext (Mm-1)
Dry bext loading (%)
Ambient bext (Mm-1)
Ambient bext loading (%)
Beijing
Chengde
Shijianzhuang
Tianjin
Shangdianzi
Shanghai
Nanjing
Guangzhou
Shenzhen
Zhongshan
Hongkong
Conghua
Tianhu
Hok Tsui
Zhengzhou
Xi'an
Lanzhou
Jinan
Chengdu
Chongqing
Changsha
Fuzhou
Xiamen
Haikou