Dear Editor:

We have addressed all of the comments provided by the three reviewers. The details can be found in our enclosed responses to the reviewers' comments. For your and the reviewers' convenience in reviewing the changes, a copy of the paper with highlighted major changes is also attached below.

Thank you for taking care of the review process for this paper.

Sincerely,

Jun Tao, Leiming Zhang and co-authors

Response to Referee #1

We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed the comments carefully, as detailed below.

1. An urgent task facing the Chinese government and the scientific community is to quantify the sources and formation mechanisms causing episodic events of high $PM_{2.5}$ mass concentrations and sever haze. This paper provides a summary of source appointment studies on haze events, but not on $PM_{2.5}$ mass concentrations. It is recommended to also include a review of source factors identified for $PM_{2.5}$ in various regions of China.

Response: We have found more than 40 SCI articles on $PM_{2.5}$ source-appointment studies published during 2000-2017. We have provided a summary table (Table S2) of these studies in the SI document, and added a new section (2.3) in the revised paper focusing on source-apportionment studies. In this section, we first briefly summarized common receptor models used in $PM_{2.5}$ source-appointment studies and common source factors found in Chines cities. We then discussed annual and seasonal contributions of dominant source factors to $PM_{2.5}$ mass region by region.

In the abstract, we have also provided a summary of major findings based on the review of these studies, which reads: "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 across China. Biomass emission in any 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.

2. For a few cities such as Beijing, Shanghai and Guangzhou, inter-annual variations are discussed based on field measurements conducted by different researchers (and likely using different instruments and/or QA/QC methods). How much confidence do you have on these inter-annual variations compared to measurement uncertainties?

Response: We have carefully collected the information about the measurement and analysis methods used in literature and identified potential measurement uncertainties for the dominant chemical components (OC/EC and water soluble inorganic ions). We have added this information in section 2.2 in the revised paper:

"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 Sunset analyzer. OC and EC measured

by the two analyzers are comparable if using the same analysis protocol. For example, Wu et al. (2011) showed that OC from Sunset analyzer was only 8% lower than that from DRI analyzer, while EC was only 5% higher. However, when using different protocols 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 protocol was similar 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 chromatograph. 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 relative humidity (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 ambient temperature (Chow et al., 2005). Although the exact magnitudes of measurement uncertainties cannot be determined for NO_3^- and NH_4^+, they are expected not 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."

We have taken into account the above information when discussing the trends of measured species through this section.

3. A related question to question 2 above: is it possible to compare the trends identified in this study to other sources such as the online $PM_{2.5}$ data, the AOD trend analysis data, or available literature?

Response: As noted in a recent paper by Fontes (2017): "The long trends of $PM_{2.5}$ concentrations were not fully investigated in China, in particular the year-to-year trends and the seasonal and daily cycles." They analyzed $PM_{2.5}$ data from 1999-2008 at five megacities in China. We have added this reference in the revised paper. The data set we collected in this review paper covered much longer periods and all the sites across China.

We have added a brief discussion on the relationship between AOD and $PM_{2.5}$ at the beginning of Section 3, which reads: "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)."

4. Please also add sub-section titles in the content lists.

Response: We have added sub-section titles in the contents list.

Response to Referee #2

We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed the comments carefully, as detailed below.

The review paper jumps to the hot topics on PM2.5 pollution and aerosol optical property in China. It is well-written and very helpful for understanding the current situations and challenges ahead for alleviating severe PM2.5 pollution in China. This reviewer has a few minor comments for authors considering before publishing in ACP

1. The authors are encouraged to use either OC or OM through the manuscript.

Response: After a careful consideration, we feel both OC and OM are needed in the discussions in various places. For example, OC is measured directly and needed to be discussed in the measurement data as well as in comparing related emission inventories. OM is converted from OC using different conversion factors in different regions and is needed in assessing $PM_{2.5}$ mass distributions among different chemical components.

2. As a rural site downwind of BTH, the study by Feng et al. (JRG, 117, D03302, doi:10.1029/2011JD016400, 2012) is worthy of inclusion for comparison.

Response: We have included this reference and the following discussion in the revised paper: "At an Asian continental outflow site (Penglai in Shandong province), annual average contribution of secondary inorganic aerosols to $PM_{2.5}$ reached to 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 Pacific Ocean."

3. Lines 191-194, temperature effect should be included. A few very recent studies suggest that extreme weather could also be important factors for heavy $PM_{2.5}$ pollution in winter. The authors may have no time to read, but these studies are really worthy of inclusion for a complete review.

Response: We have added the following discussion in the revised paper**: "**Moreover, extreme weather events such as weakening monsoon circulation, depression of strong cold air activities, strong temperature inversion, and descending air motions in the planetary boundary layer also played important roles in wintertime heavy $PM_{2.5}$ pollution (Niu et al., 2010; Wang et al., 2014c; Zhao et al., 2013). 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)."

4. Lines 288-295, the reviewer suggest to include these contentious studies for sulfate formation in atmospheric particles published in 2016 and add a few arguments as well. It is helpful for students and young scientists.

Response: We have added the following discussion in the revised paper**: "**It is worth to note that several recent studies have highlighted the important role NO_2 might play in sulfate formation in the polluted environment in China (Cheng et al., 2016; Wang et al., 2016c; Xie et al., 2015a). Nevertheless, the aqueous SO_2 + H_2O_2/O_3 oxidation should still be the dominant 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 condition in north China (Li et al., 2017b). Extensive measurements of stable oxygen are needed to confirm the relative contributions of different sulfate formation mechanisms."

5. Lines 616-624, relative humidity is also important factor to determine spatial variation of AOD.

Response: We have included meteorological factors (which cover RH) in AOD discussion in the revised paper, which reads: "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)."

6. In Section "4.3 Aerosol gyroscopic properties", the authors are encouraged to include aerosol particle size information if possible.

Response: We agree that particle size distribution is an important factor affecting *f*(RH) curves. However, size distributions of the dominant chemical components (NH_4^+, SO_4^{2-}, NO_3^-, OC, and EC) were only available in autumn of 2007 in urban Beijing. Thus, it is difficult to investigate the impact of different size distributions on the differences in *f*(RH) curves in different years (autumns of 2007, 2011 and 2014) in urban Beijing. For most of the other cities, only one study was available for *f*(RH) curves and particle size distribution data were also very limited. Thus, we chose not to go to the details on size-distribution related impacts. We, however, simply pointed out the potential influence of size distribution on *f*(RH) curves, which reads: "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).”

Additional references added in the revised paper adding the comments from this reviewer are listed below:

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.

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 PM2.5 in Asian continental outflow, Journal of Geophysical Research, 117, 2012b.

Li, G., Bei, N., Cao, J., Huang, R., Wu, J., Feng, T., Wang, Y., Liu, S., Zhang, Q., Tie, X., and Molina, L. T.: A possible pathway for rapid growth of sulfate during haze days in China, Atmos. Chem. Phys., 17, 3301-3316, 10.5194/acp-17-3301-2017, 2017b.

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.

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.

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, 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.

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.

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, 2013.

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.

Response to Referee #3

We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed the comments carefully, as detailed below.

General Comments

Aerosol is very important to impact atmospheric cycle and climate system by direct and indirect effects, a hot issue of scientific researches internationally. The paper summarizes the recent published on Chinese PM2.5 and reviews the tempo-spatial distribution of PM2.5, chemical composition, aerosol optical properties, and reveals their relation across the whole country, based on ground-based filter measurements of particles, gases (e.g. SO2, NO2, CO). In fact, high aerosol burden regions such as areas in Asia are still not well characterized in terms of particle chemical and microphysical properties and long-term variation trend. The topic of this paper is of common interest within the scientific community. Although the manuscript includes some important data, however, the quality is not sufficient in the current state to be directly published. The authors should take the suggestions made here into consideration for revision.

Specific suggestions

1. The paper mainly presents the $PM_{2.5}$ measurements in urban sites, especially in eastern areas and other areas with relatively strong human activities (Figure 1). In addition, the variation of $PM_{2.5}$ is very different in the North, the Middle and the South, so the authors should address it clearly (Figure 3). This paper somewhat provides more efforts to give a long-term trend of $PM_{2.5}$. However, it lacks some remote sites such as in northeastern, Xinjiang, Yunnan areas etc, maybe it is better to select one typical year to focus on these sites and compare with the sites in the paper.

Response: As explained at the beginning of section 2, the purpose of the study is to summarize chemically-resolved $PM_{2.5}$ data across China. Thus, only data sets have synchronous measurements of $PM_{2.5}$ and its major chemical components (inorganic ions, OC and EC) are included in this review. We are aware that they are many other studies concerning $PM_{2.5}$ pollution in many regions of China, however, most of these studies do not have information on $PM_{2.5}$ chemical composition and thus cannot be included in this review. We have gone through a more careful literature survey and found a few additional studies conducted in medium-sized cities and remote sites. These studies have been added in the SI tables and numbers in the paper due to the addition of these studies have been updated.

2. In lines of 620-624, AOD can reflect the column amount of aerosol in the whole atmosphere, while $PM_{2.5}$ is only the mass of particles at the surface. The differences in fine structures of $PM_{2.5}$ and AOD are related to $PM_{2.5}$-AOD comparison and spatial variations of chemical composition, the size, number, vertical distribution and transport of aerosol are also responsible for these differences. The authors should

address them clearly.

Response: We have added some materials and relevant references explaining the relationship between AOD and $PM_{2.5}$ in section 3, which reads: “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).”

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

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

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

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

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

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

*Corresponds to: leiming.zhang@canada.ca

Abstract

To obtain a thorough knowledge of $PM_{2.5}$ chemical composition and its impact on aerosol optical properties across China, existing field studies conducted after the year of 2000 are reviewed and summarized in terms of geographical, inter-annual, and seasonal distributions. Annual $PM_{2.5}$ was up to six times of the national air quality standard in some megacities in northern China. Annual $PM_{2.5}$ was higher in northern than southern cities, and higher in inland than coastal cities. In a few cities with data longer than a decade, $PM_{2.5}$ showed a slight decrease only in the second half of the past decade, while carbonaceous aerosols decreased, sulfate (SO_4^{2-}) and ammonium (NH_4^+) remained at high levels, and nitrate (NO_3^-) increased. The highest seasonal averages of $PM_{2.5}$ and its major chemical components were mostly observed in the cold seasons. Annual average contributions of secondary inorganic aerosols to $PM_{2.5}$ ranged from 25% to 48%, and those of carbonaceous aerosols ranged from 23% to 47%, both with higher values in southern regions due to the frequent dust events in northern China. 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 across China. Biomass emission in any 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 about 1.8 times of that under dry condition. Secondary inorganic aerosols accounted for about 60% of aerosol extinction coefficient (b_{ext}) under ambient conditions in megacities with RH higher than 70%. The mass scattering efficiency (MSE) of $PM_{2.5}$ ranged from 3.0 to 5.0 m^2 g^{-1} for aerosols produced from anthropogenic emissions and from 0.7 to 1.0 m^2 g^{-1} for natural dust aerosols. The mass absorption efficiency (MAE) of EC ranged from 6.5 to 12.4 m^2 g^{-1} in urban environments, but the MAE of water-soluble organic carbon (WSOC) was only 0.05 to 0.11 m^2 g^{-1}. Historical emission control policies in China and their effectiveness were discussed based on available chemically resolved $PM_{2.5}$ data, which provides the much-needed knowledge for guiding future studies and emission policy making.

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

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

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 available publications, only ground measurement data of chemical composition of $PM_{2.5}$, aerosol scattering and absorption coefficients, and aerosol hygroscopic properties published after the year of 2000 in scientific papers of Science Citation Index (SCI) journals are reviewed and summarized in this study. A total of about 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 the several 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-}), nitrate (NO_3^-),

ammonium (NH_4^+), organic carbon (OC), and elemental carbon (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 $PM_{2.5}$ level and reducing haze occurrence frequency.

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, inert-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.

Insert Fig. 1.

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 internal annual variations and seasonal patterns.

2.1.1 Geographical distributions

Annual mean $PM_{2.5}$ mass concentrations in major cities in different regions are plotted in Fig. 2a. Regional annual mean and standard deviation (SD) values were calculated using annual mean data of all the cities where data are available. Regional annual mean $PM_{2.5}$ was 115±29, 96±28, 50±16, and 100±35 μg m^{-3} in BTH (Chen et al., 2014b; Duan et al., 2006; He et al., 2001; He et al., 2012; Song et al., 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., 2014c; 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} (Zhang et al., 2015a; Xiong et al., 2017), 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 based on their latitude from north to south. Except for a few cities, such as Chengde and Beijing, there was a decreasing trend in annual $PM_{2.5}$ mass concentration with decreasing latitude. Moreover, annual $PM_{2.5}$ mass concentrations in western or inland cities were higher than those in eastern or coastal cities at the same latitudes. The geographical patterns of the filter based $PM_{2.5}$ measurements agreed well with the online monitoring of $PM_{2.5}$ in 31 provincial capital cities in China (Wang et al., 2014b).

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

of that (35 μg m^{-3}) at the rural sites of PRD.

Insert Fig. 2.

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 trend of online data, which ranged from 65 to 83 μg m^{-3} during 2004-2012 (Liu et al., 2014b). The lower concentrations of the online than filter $PM_{2.5}$ data should be caused by volatilization loss of nitrate and organic matters from the tapered element oscillating microbalances (TEOM) working at the 50℃ during the online sampling. These results suggested that there was no evidence that $PM_{2.5}$ pollution has been significantly improved in Beijing during the 15 year study period despite the many control measures that have been excised. The impact of local effort of pollution control in Beijing has likely been offset by regional pollutant transport (Li et al., 2015b). In Shanghai, $PM_{2.5}$ in 2003-2006 (94 μg m^{-3}) (Feng et al., 2009; Wang et al., 2006) and 2009 (94 μg m^{-3}) (Zhao et al., 2015b) was nearly 50% higher than earlier years (e.g., 65 μg m^{-3} in 1999-2000) (Ye et al., 2003); although it decreased slightly to 58 μg m^{-3} in 2011-2013 (Wang et al., 2016a; Zhao et al., 2015b), it increased rapidly and back to the level of 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) kept the stable levels and then decreased to 48 μg m^{-3} in 2014 (Tao et al., 2017).

Insert Fig. 3.

2.1.3 Season patterns

In BTH, the highest seasonal average $PM_{2.5}$ concentrations were observed in winter and the lowest in summer in all the cities with seasonal variations up to the factors of 1.7, 1.5, 1.6 and 1.8 in Beijing (Cao et al., 2012b; Chan et al., 2005; Dan et al., 2004; Duan et al., 2006; He et al., 2001; Huang et al., 2014b; Ji et al., 2014; Jung et al., 2009b; Lin et al., 2016; Okuda et al., 2011; Pathak et al., 2011; Song et al., 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; 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 motions in the planetary boundary layer also played important roles in wintertime heavy $PM_{2.5}$ pollution (Niu et al., 2010; Wang et al., 2014c; Zhao et al., 2013). 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 the factors of 2.3, 1.9 and 2.0 in Nanjing (Li et al., 2015a; Li et al., 2016a; Shen et al., 2014; Yang et al., 2005b), Shanghai (Cao et al., 2012b; Cao et al., 2013; Feng et al., 2009; Feng et al., 2012a; 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., 2003; Cao et al., 2012b; Cui et al., 2015; Duan et al., 2007; Huang et al., 2007; Huang et al., 2013; Huang et al., 2014b; Jahn et al., 2013; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tan et al., 2009; Tan et al., 2014; Tao et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Yang et al., 2011b). Although the highest seasonal average $PM_{2.5}$ was also observed in winter, the lowest season was not consistent between the sites, e.g., in summer in Guangzhou and in spring in Hong

Kong. This was likely caused by warm/hot temperatures in this region and frequent precipitation in warm seasons, and thus small differences between spring and summer, e.g., $PM_{2.5}$ concentration of 32 μg m^{-3} in summer (Cao et al., 2003; Cao et al., 2012b; Duan et al., 2007; Ho et al., 2006a; Lai et al., 2007; Louie et al., 2005a) and 29 μg m^{-3} in spring (Louie et al., 2005a) in Hong Kong. Seasonal variations were up to a factor of 1.9 at both cities. $PM_{2.5}$ at rural sites in PRD generally doubled during dry seasons (autumn and winter) compared to wet seasons (spring and summer) due to frequent precipitation scavenging of aerosols in wet seasons (Cheung et al., 2005; Dai et al., 2013; Fu et al., 2014; Griffith et al., 2015; Hu et al., 2008; Lai et al., 2016).

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

2.2 Major chemical components of $PM_{2.5}$

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

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 Sunset analyzer. OC and EC measured by the two analyzers are comparable if using the same analysis protocol. For example, Wu et al. (2011) showed that OC from Sunset analyzer was only 8% lower than that from DRI analyzer, while EC was only 5% higher. However, when using different protocols 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 protocol was similar 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 chromatograph. 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 relative humidity (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 ambient temperature (Chow et al., 2005). Although the exact

magnitudes of measurement uncertainties cannot be determined for NO_3^- and NH_4^+, they are expected not 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-annul variations in Beijing

Chemically-resolved $PM_{2.5}$ data in BTH covering multiple-years are only available in Beijing and the inert-annual variations are discussed for this city below (Duan et al., 2006; He et al., 2001; Song et al., 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 and EC (also known as black carbon or BC) is mainly from primary emissions. The anthropogenic emission for OC and BC in the whole China showed an increasing trend in 1996-2010 (Lu et al., 2011), while BC emissions showed a slightly decreasing trend in Beijing and Tianjin in 2005-2009 (Qin and Xie, 2012).

296 Meanwhile, BC emissions sharply increased in Hebei province in 2005-2009. The
297 amount of BC emissions in Hebei province was much higher than the sum of those
298 in Beijing and Tianjin (Qin and Xie, 2012). Thus, the decrease of EC concentration
299 in Beijing was likely dominated by local emission reduction instead of regional
300 transport from Hebei province.

301 Annual SO_4^{2-} concentration increased slightly during 1999-2010 and ranged
302 from 10.2 μg m^{-3} to 16.4 μg m^{-3} in Beijing. SO_2 emission in China increased by
303 about 60% during 2000-2006 and then decreased about 9% during 2006-2010 due to
304 the compulsory flue-gas desulfurization equipment applied in power plants (Lu et al.,
305 2011). However, the sum of SO_2 emission in BTH (including Beijing, Tianjin, and
306 Hebei province) increased sharply from 2097 Gg $year^{-1}$ in 2000 to 2916 Gg $year^{-1}$ in
307 2004, and further slightly increased to 2998 Gg $year^{-1}$ in 2007 before sharply
308 decreased to 1821 Gg $year^{-1}$ in 2010 (Lu et al., 2010; Zhao et al., 2013a). A
309 continued increase in SO_2 emission was found in Hebei province, which accounted
310 for more than 50% of the total SO_2 emission in BTH. In contrast, SO_2 emission in
311 Beijing continued decreasing. Surface annual SO_2 concentration in Beijing gradually
312 decreased from 56 μg m^{-3} to 35 μg m^{-3} during 2006-2009 (http://www.zhb.gov.cn/).
313 Thus, the persistent high concentrations of SO_4^{2-} in Beijing was largely due to
314 regional transport from Hebei province, noting that the lifetime of SO_4^{2-} is longer
315 than that of SO_2.

316 NO_3^- concentrations were relatively steady (7.4-10.9 μg m^{-3}) during 1999-2006,
317 but sharply increased to 15.9 μg m^{-3} in 2009-2010 in Beijing. Both NO_x (NO_2+NO)

318 emissions and satellite NO_2 vertical column densities synchronously increased
319 during 2000-2010 in China (Zhang et al., 2012b; Zhao et al., 2013b). Different from
320 those of SO_2 emissions, NO_x emissions in all the cities and provinces in BTH
321 showed increasing trends in 2005-2010. NO_x emission in Beijing slightly increased
322 from 410 Gg $year^{-1}$ in 2005 to 480 Gg $year^{-1}$ in 2010 (Zhao et al., 2013b). However,
323 annual average surface NO_2 concentration in Beijing showed a decreasing trend and
324 fluctuated in the range of 49 - 66 μg m^{-3} during 2006-2009 (http://www.zhb.gov.cn/).
325 There were some inconsistences between the trends of surface NO_2 concentration
326 and column NO_2 or NO_x emission, likely due to the impact of photochemical
327 reaction on surface NO_2 concentration in urban areas. To some extent, the increasing
328 trend of NO_3^- in Beijing was likely related to the increases in NO_x emissions in both
329 Beijing and the surrounding cities or provinces.

330 Considering the potential large uncertainties in NH_4^+ measurements, its trends
331 should only be discussed qualitatively. NH_4^+ concentrations were relatively steady in
332 Beijing during 1999-2006, ranging from 5.7 to 7.3 μg m^{-3}. NH_3 emissions changed
333 little (13400-13600 Gg $year^{-1}$) before 2005 in China, and increased slightly in BTH
334 region during 2003-2010 (Zhou et al., 2015b). The small increase of NH_4^+ in
335 2009-2010 in Beijing was consistent with the NH_3 emission trend in this region
336 (Zhang et al., 2013a; Zhao et al., 2013c). Moreover, the increase of NO_3^- in Beijing
337 was also an important factor for the increase of NH_4^+.

338 In summary, a decreasing trend was identified in TC and increasing ones for
339 SO_4^{2-}, NO_3^- and NH_4^+ in Beijing. The inter-annual variations in EC agreed with its

local emission trend in Beijing, but those in SO_4^{2-}, NO_3^- and NH_4^+ agreed more with the regional scale emission trends of their respective gaseous precursors in BTH rather than the local emission trends in Beijing. Nonlinear responses of concentration changes of these aerosol components to their respective emission trends were found, demonstrating the other important factors affecting aerosol formation. It is worth to note that several recent studies have highlighted the important role NO_2 might play in sulfate formation in the polluted environment in China (Cheng et al., 2016; Wang et al., 2016c; Xie et al., 2015a). Nevertheless, the aqueous SO_2 + H_2O_2/O_3 oxidation should still be the dominant 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 condition 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 converting 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 (Fig. 4) (Zhang et al., 2013a; Zhao et al., 2013c). Secondary inorganic aerosols (the sum of sulfate, nitrate and ammonium) contributed

362 36-39% of $PM_{2.5}$ annually in the majority of the cities having measurements, but
363 only 25% in Chengde, a tourist city located in the northeast part of BTH and 200
364 kilometer away from Beijing. Generally, the percentage contribution of secondary
365 inorganic aerosols to $PM_{2.5}$ decreased with decreasing $PM_{2.5}$ level, e.g., from
366 Shijiazhuang to Tianjin, Beijing, and then Chengde, a phenomenon that is consistent
367 with what was found within the same city but for different pollution levels in a
368 winter season (Tao et al., 2015a). Carbonaceous aerosols contributed 29-32% to
369 $PM_{2.5}$ in most cities, but as high as 45% in Chengde, and had an opposite trend to
370 that of secondary inorganic aerosols in terms of city-to-city variations. At the rural
371 site Shangdianzi near Beijing, secondary inorganic aerosols and carbonaceous
372 aerosols accounted for 42% and 32%, respectively, of $PM_{2.5}$ mass, which were not
373 significantly different from those in cities located south of Yanshan Mountain. The
374 sum of secondary inorganic aerosols and carbonaceous aerosols accounted for
375 65%-70% of $PM_{2.5}$ mass in cities of BTH.

376 In Beijing where data are available for more than a decade, secondary inorganic
377 aerosols accounted for 28% of $PM_{2.5}$ on average and ranged from 23% to 31% from
378 year to year. Carbonaceous aerosols accounted for 43% of $PM_{2.5}$ and ranged from
379 29% to 55%. Seasonal average contributions of secondary inorganic aerosols were
380 generally higher in warm seasons than in cold seasons in most cities, and an opposite
381 trend was found for carbonaceous aerosols (Fig.5). For example, secondary inorganic
382 aerosols contributed 32%, 41%, 28% and 32% in spring, summer, autumn and winter,
383 respectively, to $PM_{2.5}$ in Beijing, while carbonaceous aerosols contributed 35%, 30%,

44% and 45% (Cao et al., 2012b; Duan et al., 2006; He et al., 2001; Huang et al., 2014b; Pathak et al., 2011; Song et al., 2007; Song et al., 2006a; Sun et al., 2004; Tao et al., 2015a; Tian et al., 2015; Zhang et al., 2013a; 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).

Insert Fig. 4.

Insert Fig. 5.

2.2.2 The Yangtze River Delta region

2.2.2.1 Inter-annul variations in Shanghai

Chemically-resolved $PM_{2.5}$ data in YRD covering multiple-years are only available in Shanghai (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, much higher than the known measurement uncertainties. Noticeable reduction of OC and EC occurred after 2010 Shanghai World Expo, which resulted in evident decrease of

406 TC after 2010. BC emission slightly decreased in Shanghai in 2005-2009, but
407 increased in the adjacent Zhejiang and Jiangsu provinces (Qin and Xie, 2012).
408 Especially, BC emission in Jiangsu province was much higher than the sum of those
409 in Shanghai and Zhejiang. Thus, the decreased EC concentration in Shanghai was
410 mostly resulted from local emission reduction.

411 Annual SO_4^{2-} concentration decreased from 14.0 μg m^{-3} in 1999-2000 to the
412 range of 11.7 μg m^{-3} to 12.5 μg m^{-3} during 2009-2014. The trend of SO_2 emission in
413 YRD generally agreed with that in the whole China, which showed an increasing
414 trend during 2000-2006 and a decrease one during 2006-2010 (Lu et al., 2011). The
415 annual variations in SO_2 emission in YRD (including Shanghai, Jiangsu, and
416 Zhejiang) were relative small, ranging from 3171 Gg $year^{-1}$ in 2000, 3506 Gg $year^{-1}$
417 in 2004, 3376 Gg $year^{-1}$ in 2007, and to 3397 Gg $year^{-1}$ in 2010 (Lu et al., 2010;
418 Zhao et al., 2013a). Annual average SO_2 concentrations in Shanghai were in the
419 range of 45-61 μg m^{-3} during 2000-2005 and decreased by around 50% to 24-29 μg
420 m^{-3} during 2010-2013 (http://www.zhb.gov.cn/). Note that SO_2 emissions in
421 Shanghai only accounted for less than 20% of the total SO_2 emissions in YRD and
422 with small annual variations. The high concentrations of SO_4^{2-} observed in Shanghai
423 were also closely related to regional transport from north China (e.g. BTH and
424 Shandong province) (Li et al., 2011; Wang et al., 2016a).

425 Annual NO_3^- concentrations in Shanghai were relatively steady (6.0-7.7 μg m^{-3})
426 during 1999-2009, but sharply increased to 13.3 μg m^{-3} in 2011-2014. NO_x
427 emissions in YRD also showed an increasing trend during these years, consistent

with satellite retrieved vertical column NO_2 density during 2000-2010 (Zhang et al., 2012b; Zhao et al., 2013b). In contrast, surface-level annual NO_2 concentration 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 inconsistence in the trends between emissions and gaseous and particulate matters surface air concentrations was similar to that 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 levels of 4.1 μg m^{-3} in 2009 and then increased to 6.6 μg m^{-3} in 2011-2014. NH_3 emission increased in 2000-2005 in east China (including BTH, YRD and PRD) and possibly also increased in 2006-2010 due to the lack of control measures for NH_3 in China (Wang et al., 2011). The recently increased NH_4^+ concentrations in Shanghai were likely due to the concurrent increases of NH_3 emissions and NO_3^- concentrations.

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

2.2.2.2 Relative contributions to $PM_{2.5}$

The chemical compositions in $PM_{2.5}$ between the cities in YRD were compared between Shanghai and Nanjing due to the lack of continuous annual data in Hangzhou. A converting factor of 1.6 between OC and OM was chosen for YRD, slight smaller than that (1.8) chosen for BTH considering the less impact of biomass burning to $PM_{2.5}$ in this region (Feng et al., 2006; Li et al., 2016a). Secondary inorganic aerosols contributed 25-54% of $PM_{2.5}$ annually in Shanghai and Nanjing, while carbonaceous aerosols contributed 28-47% (Li et al., 2016a; Wang et al., 2016a; Ye et al., 2003; Zhao et al., 2015b). The sum of secondary inorganic aerosols (sulfate, nitrate and ammonium) and carbonaceous aerosols (OM and EC) accounted for 76% and 66% of $PM_{2.5}$ mass in Shanghai and Nanjing, respectively, which was comparable with those (65%-70%) in BTH.

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

In Hangzhou, seasonal contributions can only be estimated for summer and winter 2003 (Cao et al., 2012b). Seasonal contribution of secondary inorganic aerosols in winter was 44%, which was evidently higher than that in summer (34%), while carbonaceous aerosols contributed 33-35%. At the rural sites (Ningbo and

Lin'an) in Zhejiang province, seasonal contributions of carbonaceous aerosols varied within a small range (28%-34%) in four seasons in 2008-2009, which were comparable with those in Hangzhou (Feng et al., 2015; Liu et al., 2013a).

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

Insert Fig. 6.

2.2.3 The Pearl River Delta region

2.2.3.1 Inter-annul variations in Guangzhou

Inter-annual variations for dominant chemical components were only discussed for Guangzhou in PRD since data for this city were available during 2002-2003, 2009-2010 and 2014 (Hagler et al., 2006; Tao et al., 2014c; Tao et al., 2017). Data for Shenzhen were only available during 2002-2003 and 2009 (Hagler et al., 2006; Huang et al., 2013) and for Hong Kong during 2000-2001 and 2002-2003. Annual OC concentration decreased significantly from 17.6 μg m^{-3} in 2002-2003 to 9.0 μg m^{-3} in 2009-2010, and then to 8.2 μg m^{-3} in 2014 in Guangzhou, while EC slightly

494 increased from 4.4 μg m^{-3} to 6.0 μg m^{-3} and then decreased to 4.0 μg m^{-3} during the
495 same periods. Similar to Guangzhou, annual OC concentration decreased
496 significantly from 11.1 μg m^{-3} in 2002-2003 to 8.3 μg m^{-3} in 2009-2010 in Shenzhen,
497 while EC slightly increased from 2.3 μg m^{-3} to 2.7 μg m^{-3}. Apparently, the trends of
498 EC in Guangzhou and Shenzhen were inconsistent with the BC emission trend in
499 Guangdong province during 2005-2009, which showed slightly decrease (Qin and
500 Xie, 2012). As a result, TC concentrations gradually decreased from 22.0 μg m^{-3} to
501 15.0 μg m^{-3} in Guangzhou and from 15.0 μg m^{-3} to 13.0 μg m^{-3} in Shenzhen before
502 2010, similar to what was found in Beijing and Shanghai. The reduction of TC was
503 significant in Guangzhou (32%), suggesting TC also really decreased in recent years.
504 The same phenomenon was also observed at a suburban site of Guangzhou (Hagler
505 et al., 2006; Lai et al., 2016).

506 Contrast to the TC trend, annual SO_4^{2-}, NO_3^- and NH_4^+ concentrations in
507 Guangzhou increased from 14.7, 4.0 and 4.5 μg m^{-3} in 2002-2003 to 18.1, 7.8 and
508 5.1 μg m^{-3} in 2009-2010 and evidently decreased to 9.3, 2.2 and 3.8 μg m^{-3} in 2014,
509 respectively. The similar increases were also found in Shenzhen, e.g., from 10.0, 2.3
510 and 3.2 μg m^{-3} in 2002-2003 to 11.7, 2.7 and 3.5 μg m^{-3} in 2009, respectively
511 (Hagler et al., 2006; Huang et al., 2013), and in the suburban of Guangzhou, e.g.,
512 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,
513 respectively (Hagler et al., 2006; Lai et al., 2016). SO_2 emissions in Guangdong
514 province gradually increased in the previous decade, e.g., 964, 1150, 1177 and 1258
515 Gg $year^{-1}$ in 2000, 2004, 2007 and 2010, respectively (Lu et al., 2010; Zhao et al.,

516 2013a). However, SO_2 emissions in PRD decreased more than 40% in 2009
517 compared with that in 2005, due to flue gas desulfurization facilities in power plants
518 and large industrial boilers installed in this region (Lu et al., 2013). Annual average
519 SO_2 concentrations in Guangzhou gradually increased from 45 μg m^{-3} in 2000 to 77
520 μg m^{-3} in 2004, and then decreased to 17 μg m^{-3} in 2014 (http://www.gzepb.gov.cn/).
521 Thus, the increased SO_4^{2-} concentration before 2010 in Guangzhou was largely due
522 to the regional transport of pollutants from outside of PRD. The decreased SO_4^{2-}
523 concentration in 2014 in Guangzhou was likely due to flue gas desulfurization
524 facilities in power plants and large industrial boilers extended to the whole
525 Guangdong province (http://www.gdep.gov.cn/).

526 Meanwhile, NO_x emissions increased in Guangdong province as well as in the
527 whole PRD, similar to the trends in BTH and YRD (Lu et al., 2013; Zhang et al.,
528 2012b; Zhao et al., 2013b). However, annual average surface NO_2 concentration in
529 Guangzhou fluctuated in the range of 61 - 73 μg m^{-3} during 2000-2007 and 48 - 56
530 μg m^{-3} during 2008-2014 (http://www.gzepb.gov.cn/). An opposite trend was also
531 found between NO_2 and NO_x emissions with the former persistently decreased while
532 the latter increased in Guangzhou, although NO_3^- concentration was also increased.
533 Thus, emission as well as chemical processes both affect these ions concentrations in
534 air. Annual NH_4^+ concentrations slightly increased about 10% before 2010 in
535 Guangzhou and Shenzhen although NH_3 emissions changed little during 2002-2006
536 in PRD (Zheng et al., 2012). Thus, the slightly increased NH_4^+ concentrations, if not
537 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_4^+ from NH_3.

In summary, a decreasing trend was identified in TC and increasing ones 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 was inconsistent with BC emission trend in Guangdong province. In contrast, inter-annual variations in SO_4^{2-}, NO_3^- and NH_4^+ agreed with the regional scale emission trends of their respective gaseous precursors in Guangdong province rather than PRD. Similar to what was found for 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 (Fig. 4) (Hagler et al., 2006). The converting 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 the 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 $PM_{2.5}$ 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 $PM_{2.5}$ mass in the PRD region, similar to what was found in Shanghai (YRD).

Although many studies have been conducted in PRD, most of which were for short periods. Studies covering the full four seasons were mainly carried out in Guangzhou and Hong Kong (Fig.7) (Andreae et al., 2008; Cao et al., 2003; Cao et al., 2012b; Cui et al., 2015; Ho et al., 2006a; Huang et al., 2014b; Jung et al., 2009a; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Tan et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Yang et al., 2011b). Seasonal average contributions of secondary inorganic aerosols were generally higher in spring and autumn than in summer and winter in both Guangzhou and Hong Kong. If averaging all the years data together, secondary inorganic aerosols contributed 43%, 31%, 38% and 33% in spring, summer, autumn and winter, respectively, to $PM_{2.5}$ in Guangzhou and 45%, 25%, 46% and 37%, respectively, in Hong Kong. However, different seasonal patterns were found between Guangzhou and Hong Kong for carbonaceous aerosols. Carbonaceous aerosols contributed 34%, 37%, 35% and 34% in spring, summer, autumn and winter, respectively, to $PM_{2.5}$ in Guangzhou and 54%, 47%, 49% and 38%, respectively, in Hong Kong. Seasonal variations of OC/EC ratios ranged from 1.6 to 3.4 in Guangzhou and ranged from 1.2 to 2.1 in Hong Kong, suggesting coal and vehicle exhaust as dominant sources in Guangzhou while

582 vehicle exhaust as dominant source in Hong Kong (He et al., 2008; Watson et al.,
583 2001).
584
585 *Insert Fig. 7.*
586
587 **2.2.4 Other cities**
588 Besides the cities in BTH, YRD and PRD, synchronous measurements of $PM_{2.5}$
589 and the dominant chemical components have also been conducted in several cities of
590 the other regions in China, mostly the capital city of a province (e.g. Zhengzhou of
591 Henan province, Xi'an of Shaanxi province, Lanzhou of Gansu province, Jinan of
592 Shandong province, Chengdu of Sichuan province, Chongqing of Chongqing
593 municipality, Changsha of Hunan province, Xiamen and Fuzhou of Fujian province
594 and Haikou of Hainan province) (Fig.4) (Geng et al., 2013; Liu et al., 2017a; Tan et
595 al., 2016b; Tang et al., 2017; Tao et al., 2013a; Tao et al., 2014b; Wang et al., 2015c;
596 Wang et al., 2016b; Wang et al., 2017a; Xu et al., 2012b; Yang et al., 2011b; Yang et
597 al., 2012; Zhang et al., 2012a; Zhang et al., 2016). A converting factor of 1.6
598 between OC and OM was chosen for Fuzhou, Xiamen and Haikou and 1.8 for other
599 cities based on their geographical locations.
600 Annual average contributions of secondary inorganic aerosols and carbonaceous
601 aerosols to $PM_{2.5}$ were 30% and 36%, respectively, in the island city Haikou, similar
602 to what was found in Beijing, and were 43-46% and 29-36%, respectively, in the
603 coastal cities Fuzhou and Xiamen, similar to what was found in Shanghai. Annual

604 contributions of secondary inorganic aerosols ranged from 29% to 39% in inland
605 cities (Zhengzhou, Xi'an, Jinan, Chengdu, Chongqing and Changsha) except
606 Lanzhou (15%), which were comparable with those observed in PRD (33-41%). In
607 contrast, large differences were found in the annual contributions of carbonaceous
608 aerosols, ranging from 23% in Zhengzhou to 47% in Chongqing. The sum of
609 secondary inorganic aerosols and carbonaceous aerosols accounted for 56%-79% of
610 $PM_{2.5}$ mass in these cities.

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

617 Seasonal average contributions are only shown here for Jinan, Zhengzhou,
618 Fuzhou, Chengdu, Lanzhou, Xiamen, Changsha, and Haikou due to the incomplete
619 data in Xi'an and Chongqing (Fig. 8). Seasonal contributions of secondary inorganic
620 aerosols were evidently higher in summer than in other seasons in Zhengzhou, Jinan
621 and Lanzhou (typical northern cities), similar to what was seen in BTH. In the
622 southwest city Chengdu and the central city Changsha, seasonal contribution of
623 secondary inorganic aerosols in spring was only 30% and 27%, respectively, lower
624 than in the other seasons (40-42% and 30-31%, respectively). In the two southern
625 coastal cities Fuzhou and Xiamen, the highest seasonal average contribution of

626 secondary inorganic aerosols was observed in winter (53% and 33%, respectively),
627 much higher than in other seasons (34-42% and 21-24%, respectively). In the
628 southern island city Haikou, seasonal contributions of secondary inorganic aerosols
629 were also slightly higher in winter (30%) than in other seasons (21-27%), similar to
630 what was seen in the coastal cities Fuzhou and Xiamen.

631 Seasonal average contributions of carbonaceous aerosols were evidently higher
632 in cold seasons than in warm seasons in the three northern cities (Zhengzhou and
633 Jinan and Lanzhou) due to heating activities and biomass burning, similar to what
634 was observed in BTH. Surprisingly, a similar seasonal pattern was also found in one
635 coastal city Xiamen, e.g., 38% in winter versus 27-30% in the other seasons. In
636 contrast, higher seasonal contributions were found in warm season than in cold
637 seasons in the southern coastal city (Fuzhou) and the southern island city (Haikou).
638 Flat seasonal variations were found in the southwest inland city Chengdu (29%-32%)
639 and the central inland city Changsha (28-33%). The sum contributions of secondary
640 inorganic aerosols and carbonaceous aerosols were evidently lower in spring than in
641 other seasons in most of the northern cities (e.g. Jinan, Lanzhou, Zhengzhou, and
642 BTH), likely due to the frequent spring dust storm events in northern China.

643

644 *Insert Fig. 8.*

645

646 **2.2.5 Summary of $PM_{2.5}$ chemical properties**

647 Carbonaceous aerosols showed decreasing trends over the last ten years

(2000-2010) in Beijing, Shanghai and Guangzhou, consistent with BC emission trends in these cities and surrounding areas. SO_4^{2-} and NH_4^+ remained at high levels with no significant trends in Beijing and Shanghai, but with an increasing trend in Guangzhou. NO_3^- showed increase trends in all of the above-mentioned megacities. Annual mass concentrations of $PM_{2.5}$, secondary inorganic aerosols, and carbonaceous aerosols showed similar spatial gradients decreasing from high to low latitude regions.

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

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

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 ones 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 to most of 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% from different models (Song et al., 2006a, b). Similar magnitudes of uncertainties could also be caused by using different biomass burning tracers despite using the same receptor model (Tao et al., 2016b).

Major source factors identified for $PM_{2.5}$ in most Chinese cities include secondary inorganic aerosols (SIA), coal combustion (COAL), biomass burning (BIOM), traffic emission (TRAF), dust emission (DUST), and industrial emission (INDU). Other source factors were also identified (and sometimes due to using more

specific source names), such as metal manufacturing (including iron and steel industry, Cu smelt) in industrial cities (e.g. Dongying and Tai'an of Shandong province, Nanjing, Hangzhou, Lanzhou, Chengdu, Chongqing and Changsha), and sea salt and ship emissions in coastal cities (e.g. Longkou of Shandong province, Nanjing, Guangzhou, Zhuhai and Hong Kong). Contributions of dominant source factors to $PM_{2.5}$ are discussed below in detail on regional basis.

2.3.1 The Beijing-Tianjin-Hebei region

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

Insert Fig. 9.

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 $PM_{2.5}$ mass on annual

714 average (Tian et al., 2016). Compared with results in 2012-2013 in Beijing (Zíková
715 et al., 2016), the contributions of SIA were comparable in the two cities, but those of
716 COAL and DUST were much higher in Tianjin than Beijing. However, an earlier
717 study in Tianjin in 2009-2010 showed much closer results to those in Beijing during
718 the same years in terms of $PM_{2.5}$ level and source attributions (Zhao et al., 2013c),
719 implying faster decrease of COAL contribution in Beijing than Tianjin.

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

728 **2.3.2 The Yangtze River Delta region**

729 Studies for one year or longer were only made in Nanjing (Li et al., 2016a) and
730 Hangzhou (Liu et al., 2015). Metal manufacture was identified as a source factor in
731 both Nanjing and Hangzhou, while ship emission was also identified in Nanjing.
732 Annual contributions of SIA to $PM_{2.5}$ mass reached 68% in Nanjing while all the
733 other sources (COAL, DUST, sea salt and ship emissions, and metal manufacture)
734 each contributed 10% or less. In contrast, metal manufacture, SIA, TRAF, and
735 COAL accounted for 32%, 28%, 17%, and 13%, respectively, of $PM_{2.5}$ 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 close to each other, 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 of Zhuhai (Tao et al., 2017) and Hong Kong (Huang et al., 2014c). On 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 $PM_{2.5}$ mass. In Hong Kong, SIA, TRAF, oil residue (related with Ni and V, or ship emission), DUST, and sea salt accounted for 28%, 23%, 19%, 10%, and 7%, respectively, of $PM_{2.5}$ mass.

Slight different sources factors were defined in suburban studies. Annual contributions from mixed source (from regional transport), secondary nitrate and chloride, ship emissions, COAL, and electronic industry accounted for 36%, 20%, 18%, 13%, and 13%, respectively, of $PM_{2.5}$ 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 $PM_{2.5}$ mass in suburban Hong Kong.

Despite the slightly different source factors defined 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 $PM_{2.5}$ mass in all the studies except the one for Shenzhen. Similar to the cities in northern China, the high contribution of 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, sseasonal 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 emission was the highest in summer due to the prevailing air masses from South China Sea (Huang et al., 2014c).

2.3.4 Other cities

Studies covering one year or longer were mostly conducted for provincial capital

780 cities including Jinan (Yang et al, 2013), Zhengzhou (Geng et al., 2013), Xi'an
781 (Wang et al., 2015c), Lanzhou (Wang et al, 2016b), Chengdu (Tao et al., 2014a),
782 Chongqing (Chen et al., 2017), Changsha (Tang et al., 2017), Wuhan (Xiong et al.,
783 2017), Xiamen (Zhang et al., 2016) and Haikou (Liu et al., 2017a), and for an inland
784 city Heze (Liu et al., 2017b) and a regional background site (located in Yellow River
785 Delta National Nature Reserve in Dongying city) (Yao et al., 2016) both in Shandong
786 province. Annual results were available from most studies, but were aggregated from
787 seasonal results for Wuhan and Haikou. All the sites were grouped into four regions
788 for easy discussion, i.e., northwest China (Lanzhou and Xi'an), southwest China or
789 Sichuan basin (Chengdu and Chongqing), eastern and central China (Jinan,
790 Zhengzhou, Heze, Dongying, Wuhan and Changsha), and south coastal cities
791 (Xiamen and Haikou).

792 The two northwest cities showed the same top four dominate source factors,
793 although with slightly different percentage contributions to $PM_{2.5}$ mass, e.g., 29%
794 from SIA, 19% from COAL, 17% from DUST, and 15% from TRAF in Xi'an, and
795 17% from SIA and 22% from the other three sources in Lanzhou. The lower SIA
796 contribution in Lanzhou was likely due to the dry climate inhibiting formation of
797 SIA. Similar results to those in Xi'an were also obtained in rural Xi'an, with SIA,
798 COAL, DUST and TRAF contributed 31%, 16%, 20% and 13%, respectively, to
799 $PM_{2.5}$ mass (Wang et al., 2015c). The two southwest cities (Chengdu and Chongqing)
800 showed nearly the same source-apportionment analysis results with SIA contributed
801 just below 40% and COAL and INDU each contributed around 20% to $PM_{2.5}$ mass.

802 The same top four dominant source factors (SIA, COAL, DUST and TRAF)
803 found in the two northwest cities were also found in the other capital cities (Jinan,
804 Zhengzhou, and Wuhan, but excepting Changsha) and a medium size city (Heze) in
805 eastern and central China, which accounted for 24-55%, 14-23%, 5-26% and 5-27%
806 of $PM_{2.5}$ mass, depending on location. In Changsha, SIA, mixed source of INDU and
807 BIOM, and DUST accounted for 60%, 27%, and 13%, respectively, to $PM_{2.5}$ mass.
808 SIA, BIOM and INDU were the most important sources, accounted for 54%, 16%,
809 and 16%, respectively, of $PM_{2.5}$ mass in a regional background site in Dongying.
810 Similar source-apportionment results were found between the two south coastal
811 cities (Xiamen and Haikou) with SIA, TRAF, DUST, COAL, and sea salt accounted
812 for 20-27%, 16-21%, 12-22%, 8-9% and 6-10%, respectively, $PM_{2.5}$ mass.
813 Seasonal results of source apportionment analysis are available for four seasons
814 in Jinan (Yang et al., 2013), a regional site in Dongying (Yao et al., 2016), Chengdu
815 (Tao et al., 2014a), Chongqing (Chen et al., 2017), Wuhan (Xiong et al., 2017) and
816 Haikou (Liu et al., 2017a), and for summer and autumn in Tai'an of Shandong
817 province (Liu et al., 2016a) and Xi'an (Xu et al., 2016), and for winter in Longkou (a
818 coastal site in Shandong province) (Zong et al., 2016).
819 In most seasons, SIA was the largest contributor to $PM_{2.5}$ mass, e.g., in Jinan
820 (30-45%), Tai'an (27%), a regional site in Dongying (35-72%), Chengdu (33-44%),
821 Chongqing (24-52%), Wuhan (23-41%) and Haikou (11-26%), with exceptions of
822 spring in Wuhan and summer in Haikou when DUST was the largest contributor, and
823 winter in Longkou and Xi'an when COAL was the largest contributor. Only winter

824 data was available in Longkou, and ship emission contributed 9% to $PM_{2.5}$ mass,
825 similar to what was found in the cities of PRD.
826
827 *Insert Fig. 10.*
828
829 **2.3.5 Summary of $PM_{2.5}$ source apportionment studies**
830 SIA, COAL and TRAF were the dominant source factors in most cities in China.
831 On annual average, the sum of these three factors accounted for 63%-80% of $PM_{2.5}$
832 mass in the cities of BTH region, 58%-78% in the in the cities of YRD region,
833 51%-67% in the in the cities of PRD region, 51%-61% in the northwest cities,
834 57%-60% in the southwest cities, 57%-82% in the eastern and central cities, and
835 44%-57% in the south coastal cities. The contributions of DUST were significant
836 (7-26%) in northern cities and a central city (Zhengzhou), of INDU significant
837 (19-27%) in typical industrial cities (e.g. Chengdu, Chongqing, Changsha), and of
838 ship emission significant (7-19%) in coastal and river cities (e.g. Longkou, Nanjing,
839 Guangzhou, Zhuhai, Hong Kong). High seasonal contributions were found for SIA in
840 summer, COAL in winter, DUST in spring, and ship emission in summer in
841 applicable cities.
842 It should be noted that SIA chemical compounds are formed from primary
843 emissions of gaseous precursors that can be produced from any of the identified
844 sources factors discussed above as well as from sources seldom mentioned in
845 source-apportionment studies such as agricultural emissions and many natural

sources. If allocating SIA contributions to the source factors producing 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. Combining receptor-based analysis results with source-based studies using chemical transport models can provide a more complete picture qualifying 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., 2009a; Lan et al., 2013; Man and Shih, 2001; Tao et al., 2014c; Verma et al., 2010; Wu et al., 2009; Wu et al., 2013), Chengdu in southwest China (Tao et al., 2014b; Wang et al., 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 achieved 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.

Insert Fig. 11.

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}.

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 the 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., 2015). Such a spatial pattern was mostly due to the spatial pattern 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 condition, 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).

Insert Fig. 12.

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}

933 in 2005 to 360 Mm^{-1} in 2009-2010, when $PM_{2.5}$ increased by 20% from 107 to 129
934 $\mu g\ m^{-3}$ during the same period. However, annual b_{ap} in 2009-2010 was 64 Mm^{-1},
935 which was slightly higher than 56 Mm^{-1} in 2005-2006, although the annual EC in
936 2009-2010 was evidently lower than that in 2005-2006. Meanwhile, annual
937 secondary inorganic aerosols in 2009-2010 were evidently lower than that in
938 2005-2006. The coating by secondary inorganic aerosols likely enhanced the
939 absorption of EC (Bond et al., 2006; Cheng et al., 2009; Yu et al., 2010).

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

945 Seasonal variations of b_{sp} and b_{ap} at urban and rural sites in Beijing are plotted in
946 Fig. 13. The highest seasonal average b_{sp} in Beijing was observed in winter and the
947 lowest in spring with seasonal variations up to a factor of 1.7 (Bergin et al., 2001;
948 Garland et al., 2009; Han et al., 2014; He et al., 2009; Jing et al., 2015; Li et al.,
949 2013b; Liu et al., 2009; Tao et al., 2015a; Tian et al., 2015; Zhao et al., 2011). A
950 different seasonal pattern was seen at the rural site located north of Beijing, which
951 showed 10-26% higher values in summer than in the other seasons (Yan et al., 2008;
952 Zhao et al., 2011). The highest seasonal b_{sp} in winter in Beijing was consistent with
953 the highest seasonal $PM_{2.5}$ mass. However, in rural Beijing the highest $PM_{2.5}$ mass
954 was observed in spring due to the frequent dust storm events, and the second highest

seasonal average $PM_{2.5}$ mass in summer which corresponded to the highest seasonal b_{sp}. This is because scattering efficiency of dust aerosols was lower than that of anthropogenic aerosols (Zhao et al., 2011).

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

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

Insert Fig. 13.

3.2.2 The Yangtze River Delta and Pearl River Delta region

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

b_{sp} and b_{ap} in winter were evidently higher than those in spring in Shanghai, consistent with the seasonal patterns of $PM_{2.5}$ and EC, respectively (Fig. 13) (Cao et al., 2012b; Cheng et al., 2015; Feng et al., 2014; Han et al., 2015; Huang et al., 2014a; Li et al., 2013a; Pathak et al., 2011; Wang et al., 2016a; Xu et al., 2012a; Ye et al., 2003; Zha et al., 2014; Zhao et al., 2015a). Similar seasonal variations were found for b_{sp} and b_{ap} in the two PRD cities (Guangzhou and Hong Kong), which also agreed with the patterns of $PM_{2.5}$ and EC (Andreae et al., 2008; Cao et al., 2003; Cao et al., 2004; Cao et al., 2012b; Cui et al., 2015; Gao et al., 2015; Huang et al., 2014b; Jung et al., 2009a; Lai et al., 2007; Liu et al., 2014a; Louie et al., 2005a; Pathak et al., 2011; Tao et al., 2009; Tao et al., 2014c; Tao et al., 2015b; Tao et al., 2017; Verma et

al., 2010; Wu et al., 2009; Wu et al., 2013). The highest b_{sp} and b_{ap} appeared in winter and the lowest in summer with seasonal variations up to a factor of 3.1 and 17.1 for b_{sp}, 2.3 and 5.9 for b_{ap}, in Guangzhou and Hong Kong, respectively.

3.2.3 Other cities

In Chengdu of southwest China, the highest b_{sp} appeared in winter and the lowest in summer with seasonal variations up to a factor of 1.9, which was consistent with the seasonal pattern of $PM_{2.5}$ (Tao et al., 2014a, b). However, the highest b_{ap} appeared in spring despite the highest EC in winter (Tao et al., 2014b). One explanation could be due to the large amount of OC emitted from biomass burning in spring season, which enhanced the absorption of EC (Schnaiter et al., 2005; Tao et al., 2013b). b_{sp} and b_{ap} in winter were evidently higher than those in summer in Xi'an in northwest China, consistent with the seasonal patterns of $PM_{2.5}$ and EC, respectively (Cao et al., 2009; Cao et al., 2012a; Wang et al., 2015c).

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

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

4.1 Mass scattering efficiency of $PM_{2.5}$

b_{sp} and $PM_{2.5}$ mass concentration have been found to correlate well in numerous field studies (Andreae et al., 2008; Han et al., 2015; Hand and Malm, 2007b; Jung et al., 2009a; Pu et al., 2015; Tao et al., 2014b; Tao et al., 2014c; Tao et al., 2015a; Tao et al., 2016a; Tian et al., 2015; Wang et al., 2012; Zhao et al., 2011). A parameter describing their relationship is defined as mass scattering efficiency (MSE), which is the slop 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 the heavy pollution episode and deceased to 3.6 m^2 g^{-1} during clean days, due to the large fraction of soluble inorganic components (e.g. $(NH_4)_2SO_4$ and NH_4NO_3) in $PM_{2.5}$ under heavy polluted condition (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 ranged 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., 2009a) 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 what was observed in rural Beijing (Xu et al., 2004).

In summary, annual $PM_{2.5}$ MSE mostly 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 mostly 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 mostly 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 scattering 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 Aethalometer and MAAP. BC mass concentrations (880nm) were converted to b_{ap} (532nm) by an empirical constant of 8.28 m^2 g^{-1}, which was obtained by the regression between BC mass and b_{ap} synchronously measured in autumn in Guangzhou, keeping in mind that application of an empirical constant obtained from one specific study to other cases may cause large uncertainties (Wu et al., 2009).

EC MAE was 7.5-8.5 m^2 g^{-1} in winter and 9.4 m^2 g^{-1} in summer in Beijing (632 nm) (Cheng et al., 2011; Wu et al., 2016). The higher EC MAE in summer was likely due to more coating of EC in the higher ambient humidity (Wu et al., 2016). BC MAE was 6.5 m^2 g^{-1} at 532 nm in autumn in Shenzhen of PRD (Lan et al., 2013). However, BC MAE was 12.4 m^2 g^{-1} at 532 nm in winter in Xi'an (Wang et al.,

2014a). Moreover, EC MAE of diesel was 8.4 m^2 g^{-1} (632 nm), which was higher than those (3.0-6.8 m^2 g^{-1}) of biomass burning sources (e.g. crop residual and wood) (Cheng et al., 2011).

Organic matter or brown carbon has also been found to be strong light absorption materials at the short wavelength. Available MAE values of OC include 0.76 m^2 g^{-1} (532 nm) in autumn in 2008 in Guangzhou. Moreover, available MAE values of WSOC include 1.79 and 0.71 m^2 g^{-1} (365nm) in winter and summer, respectively, in Beijing (Cheng et al., 2011). The WSOC MAEs of wood, grass, corn, and diesel tractor were 0.97, 0.90, 1.05, and 1.33 m^2 g^{-1} (365nm), respectively, which were much higher than that of gasoline motorcycle (0.20 m^2 g^{-1}, 365nm) (Du et al., 2014b). Evidently, the MAEs of OC or WSOC should not be neglected for short wavelength absorption.

4.3 Aerosol hygroscopic properties

b_{sp} under ambient condition can differ significantly from that under dry condition due to hygroscopic properties of soluble aerosol chemical components. A relationship between ambient and dry b_{sp} is thus developed for estimating ambient b_{sp} from measured dry b_{sp}, which is often described by the hygroscopic growth curve (f(RH)) as a function of RH: $f(RH)=1+a\times(RH/100)^b$. Here, a and b are empirical fitting parameters. Only a few studies conducted in Beijing, Wuqing, Lin'an and Guangzhou provided the aerosol hygroscopic curves (Table 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 nephelometer and visibility meter,

1105 respectively. The second one measures wet b_{sp} by integrating nephelometer equipped
1106 with a humidifier. And the third one estimates dry and wet b_{sp} based on Mie theory
1107 with size-resolved chemical components.

1108 Available *f*(RH) curves in China are summarized in Fig. 14. The three *f*(RH)
1109 curves in autumn of 2007, 2011 and 2014 in urban Beijing were all measured using
1110 the first method (Fig.14 a) (Liu et al., 2013b; Liu et al., 2013c; Yang et al., 2015).
1111 The two *f*(RH) curves measured in 2011 and 2014 were quite close, but the one in
1112 2007 was lower under RH< 80% and higher under RH>80%, likely due to aerosol
1113 chemical composition and size distribution changes in these years.

1114 The *f*(RH) curves at four rural sites were measured using the second method,
1115 including Baodi of Tianjin in spring, Wuqing of Tianjin in winter (Fig.14 b) (Chen et
1116 al., 2014a; Pan et al., 2009), Raoyang of Hebei province in summer (Wu et al., 2017),
1117 and Lin'an of Zhejiang province in spring (Fig.14 c) (Zhang et al., 2015b). It is
1118 known that the hygroscopic chemical components are mostly water-soluble inorganic
1119 salts (e.g. $(NH_4)_2SO_4$, NH_4NO_3), while mineral dust and organic matter are mostly
1120 hydrophobic. In Baodi in spring season, the concentrations of $(NH_4)_2SO_4$ and
1121 NH_4NO_3 and their mass fractions in fine mode particles were higher during the
1122 polluted episode than during the clean period or dust storm episode, resulting in
1123 higher *f*(RH) values during the polluted episode. *f*(RH) values measured in winter in
1124 Wuqing were evidently higher than those measured in spring in Baodi under
1125 RH<80% likely due to more hygroscopic chemical components in winter in Wuqing.
1126 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 the polluted episodes were also higher than those during the dust influenced episode in Lin'an, but differences between polluted and dust periods were smaller in Lin'an than in Baodi. Noticeably, the *f*(RH) values during the polluted episode were similar in Lin'an and Baodi, e.g. *f*(RH=80%) was 1.5 and 1.6, respectively.

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 originated from air masses in the north and marine aerosols originated 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.

Insert Fig. 14.

Insert Fig. 15.

4.4 Source apportionment of haze in China

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

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

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

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

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

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

Using the chemical composition data shown in Fig. 4 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 condition, carbonaceous aerosols had similar percentage contributions to secondary inorganic aerosols in Shijiazhuang, Tianjin, Shangdianzi, Shanghai, Hok Tsui, Zhengzhou, Xi'an, Jinan, Chengdu, Fuzhou and Xiamen, but higher by 11-65% 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 condition in humid (RH>70%) cities (e.g. Haikou, Changsha, Xiamen, Nanjing, cities in PRD, and Chengdu).

Insert Fig. 16.

5. Implications for aerosol pollution controls

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

A series of regulations controlling coal combustion has been made since the first

version NAAQS promulgated in 1982. The Air Pollution Prevention law of PRC was promulgated in 1987, which was the milestone in air pollution prevention history in China. It also marked the beginning of a new era for preventing air pollution based on the national law, followed by a series of regulations for controlling coal combustion. During 1990-2000, most of the control measures or technologies (e.g., desulfurization and dedusting for coal combustion) were focused on reducing SO_2 emissions. The measure for gross control of SO_2 emissions was enforced since 1996. Despite these efforts, the amount of SO_2 emissions increased about 28% in 2005 compared with that in 2000 (http://www.zhb.gov.cn/). The amount of SO_2 emission began to decrease in 2006 and gradually reduced to the emission level of 2000 in 2010 (http://www.zhb.gov.cn/). Meanwhile, ambient annual SO_2 concentration in urban cities in China also decreased from 57 μg m^{-3} in 2005 to 40 μg m^{-3} in 2010 (http://www.zhb.gov.cn/). Apparently, the emission controlling efforts for reducing SO_2 emissions were effective after 2006.

The control measures for NO_2 only began with the control of vehicle 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 is also seen in vertical column NO_2 derived from satellite data (Zhang et al., 2012b). Although annual average ambient NO_2 at surface level 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 remained at high levels and nitrate even gradually increased in megacities in China. More recently, the Clean Air Action Plan (CAAP) for improving the 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 $PM_{2.5}$ annual mass concentrations by 25%, 20%, and 15% of the 2012 levels in BTH, YRD, and PRD, respectively. The key industries including power plant, iron and steel smelting industry, petroleum chemical industry, cement industry, nonferrous metals smelting industry, and chemical industry were required to execute stricter emission standards in the key regions including most megacities in China (http://www.zhb.gov.cn). Accordingly, annual average $PM_{2.5}$ in China from online monitored data at 74 cities gradually decreased from 72 μg m^{-3} in 2013 to 50 μg m^{-3} in 2015, showing some promising results from the series of control measures.

One factor that needs to be considered in future pollution reduction is the non-linearity of chemistry (Cheng et al., 2016). For example, a modeling sensitivity study suggested potential increase in NO_3^- mass concentrations due to the increased atmospheric oxidizability, even under NO_x emissions decreasing conditions (Zhao et al., 2013a). Furthermore, increased atmospheric oxidizability may also enhance the conversion of VOCs to OM. In fact, the contribution of secondary organic aerosols to $PM_{2.5}$ was also high and could increase further in typical megacities in China (He

et al., 2011; Huang et al., 2014b; Sun et al., 2013). Another factor that requires more intension is the ammonia emissions from agricultural activities in rural areas and human activities in cities. Ammonia emission can enhance $PM_{2.5}$ pollution substantially, especially in ammonia-limited (acid aerosols) areas (Wang et al., 2011), and this topic needs further investigation through both modeling simulation and field observations.

For cleaning the atmosphere across China, the following recommendations are provided based on the major chemical components contributing to $PM_{2.5}$ and their impact of aerosol optical properties. Emissions produced from coal combustion, both in industrial sectors and in residential areas, need to be further reduced. While advanced technology should be adopted in the medium term in major industrial sectors consuming coal, cleaner energy sources should be considered for the long-term goal (Cao et al., 2016). Providing cleaner energy to the vast rural and urban areas in north China for heating and cooking can not only reduce coal emissions but also biomass burning emissions. Improving fertilizer use efficiency in agriculture is needed in reducing nitrogen emissions especially ammonia gas (Behera et al., 2013). Educating public to reduce meat consumption in daily life, especially in the developed regions with high living standard, can reduce substantially nitrogen footprint and thus nitrogen emission (Galloway et al., 2014), besides gaining human body health benefits. Traffic emissions in megacities may also need to be constrained such as developing more efficient public transportation systems and limit personal automobiles. Planting more trees and other vegetation such as the continued

expansion of the three northern region shelter forests in north China can reduce dust emissions and increase atmospheric removal of aerosols through dry deposition process (Zhang et al., 2017b). Having more vegetation coverage is especially important for arid or semi-arid areas as well as for urban areas in reducing dust emissions (Baldauf, 2017), besides biological benefits.

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