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Review article

Sources of particulate matter in China: Insights from source apportionment studies published in 1987–2017

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ABSTRACT

Particulate matter (PM) in the atmosphere has adverse effects on human health, ecosystems, and visibility. It also plays an important role in meteorology and climate change. A good understanding of its sources is essential for effective emission controls to reduce PM and to protect public health. In this study, a total of 239 PM source apportionment studies in China published during 1987–2017 were reviewed. The documents studied include peer-reviewed papers in international and Chinese journals, as well as degree dissertations. The methods applied in these studies were summarized and the main sources in various regions of China were identified. The trends of source contributions at two major cities with abundant studies over long-time periods were analyzed. The most frequently used methods for PM source apportionment in China are receptor models, including chemical mass balance (CMB), positive matrix factorization (PMF), and principle component analysis (PCA). Dust, fossil fuel combustion, transportation, biomass burning, industrial emission, secondary inorganic aerosol (SIA) and secondary organic aerosol (SOA) are the main source categories of fine PM identified in China. Even though the sources of PM vary among seven different geographical areas of China, SIA, industrial, and dust emissions are generally found to be the top three source categories in 2007–2016. A number of studies investigated the sources of SIA and SOA in China using air quality models and indicated that fossil fuel combustion and industrial emissions were the most important sources of SIA (total contributing 63.5%–88.1% of SO_4^{2-}, and 47.3%–70% NO_3^-), and agriculture emissions were the dominant source of NH_4^+ (contributing 53.9%–90%). Biogenic emissions were the most important source of SOA in China in summer, while residential and industrial emissions were important in winter. Long-term changes of PM sources at two megacities of Beijing and Nanjing indicated that the contributions of fossil fuel and industrial sources have been declining after stricter emission controls in recent years. In general, dust and industrial contributions decreased and transportation contributions increased after 2000. $PM_{2.5}$ emissions are predicted to decline in most regions during 2005–2030, even though the energy consumptions except biomass burning are predicted to continue to increase. Industrial, residential, and biomass burning sources will become more important in the future in the businuess-as-usual senarios. This review provides valuable information about main sources of PM and their trends in China. A few recommendations are suggested to further improve our understanding the sources and to develop effective PM control strategies in various regions of China.

1. Introduction

Airborne particulate matter (PM) is a complex mixture of inorganic and organic compounds that exist in either the solid or liquid state. PM can cause atmospheric visibility impairment by scattering and absorbing light (Hyslop, 2009). It also can influence climate directly by scattering and absorbing solar radiation and indirectly by modifying clouds microphysical properties of albedo and lifetime (Solomon et al., 2007). Furthermore, exposure to high levels of PM can cause various human health problems, such as respiratory diseases (Hacon et al.,

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2007; Willers et al., 2013) and cardiovascular diseases (Franchini and Mannucci, 2009; Langrish et al., 2012; Ostro et al., 2015).

China has been experiencing severe regional haze pollution in recent two decades, especially in a few key regions such as the North China Plain (NCP) and the Yangtze River Delta (YRD), characterized by extremely low visibility ranges and high $PM_{2.5}$ (PM with aerodynamic diameter equal to or less than 2.5 μm) concentrations (Chan and Yao, 2008; Fu and Chen, 2017; Hu et al., 2014a; Wang et al., 2014f; Zhuang et al., 2014). Recognizing the severity of the haze pollution situation, the Chinese government has set a target in 2013 to reduce $PM_{2.5}$ level by up to 25% in these regions by 2017 (State Council of PRC, 2013). Accurate identification and quantification of $PM_{2.5}$ source contributions are the basis to establish effective control strategies. Therefore, source apportionment studies of $PM_{2.5}$ have become one of the core contents of atmospheric environment research in China.

Accurate source apportionment of PM is complex and difficult, because PM has many primary and secondary components and different components of PM are from different sources. The directly emitted PM is called primary PM, including elemental carbon, primary organic aerosols, minerals, and etc.. In addition, part of PM is formed in the atmosphere through chemical reactions that convert gaseous pollutants (including NO_x, SO_x, and volatile organic compounds (VOCs)) to semi-volatile products that partition to the particle phase. PM formed through atmospheric chemistry processes is called secondary PM, including nitrate, sulfate, ammonium, and secondary organic aerosols (SOA). A variety of sources release PM directly to the atmosphere, including combustion sources (stationary and mobile), food cooking, activities that create dust (road travel and agriculture), and natural sources such as wind-blown dust and sea spray. Ambient measurements have revealed that high primary PM emissions and high secondary PM formation were responsible for severe haze pollution in China (Huang et al., 2014b).

Many source apportionment studies have been conducted in various locations in the United States and Europe since 1960s (Hopke, 2016). Review studies have been conducted to summarize the methods that can be used for source apportionment (Hopke, 2016; Viana et al., 2008) and the sources of regional PM (Belis et al., 2013; Viana et al., 2008). Viana et al. (2008) reviewed 30 years' European publications dealing with source apportionment of PM during 1987 to 2007. The analysis showed that four main source types (PM_{10} and $PM_{2.5}$): a vehicular source, a crustal source, a sea-salt source, a mixed industrial/fuel-oil combustion and a secondary aerosol source (the latter two probably representing the same source type). Their contributions to bulk PM levels varied widely at different monitoring sites, and showed clear spatial patterns in the cases of the crustal and sea-salt sources. Another review by Belis et al. (2013) was conducted focusing on the published literature on source apportionment of PM in Europe using receptor models. Six major source categories for PM were apportioned in Europe: atmospheric formation of secondary inorganic aerosol (SIA) (SO_4^{2-}, NO_3^-, and NH_4^+), traffic, re-suspension of crustal/mineral dust, biomass burning, (industrial) point sources, and sea/road salt. These reviews provide comprehensive understanding of contributions of different sources to PM in these regions.

The first source apportionment study in China was reported in 1987 (Zhang et al., 1987), and since then source apportionment studies have made significant progress in various regions of China due to increasing concerns of severe air pollution problems. Recently, Zheng et al. (2014) summarized $PM_{2.5}$ source apportionment methods and techniques previously and currently applied in China, including sampling preparation, sampler selection, chemical speciation analysis, and source apportionment tools. Zhang et al. (2015b) introduced the development history and characteristics of three main kinds of source apportionment methods (i.e., emission inventory, source-oriented models and receptor models), and discussed the performance differences of the methods using $PM_{2.5}$ chemical components data in Atlanta, USA. More recently, Zhang et al. (2017) reviewed the source apportionment results from 21 cities using the receptor-based method in China, and discussed chemical components and sources of $PM_{2.5}$ in these cities. These review studies provide valuable information of the source apportionment methods that can be applied in China and the sources of $PM_{2.5}$ in multiple locations of China based on receptor models. However, source apportionment studies using air quality models have not been included. Moreover, secondary PM accounts for a large fraction of total PM mass in China, but its sources typically cannot be resolved by the receptor models. Reviews on the sources of secondary PM are necessary to gain a more comprehensive understanding of PM sources in China.

In this study, we reviewed 239 PM source apportionment studies in China published during 1987–2017. We searched China National Knowledge Infrastructure (CNKI) for papers and dissertations in Chinese, and Elsevier ScienceDirect for papers in English, respectively. The key words included sources/source contributions/source apportionment, air quality/particulate matter, and China. The studies included 185 peer-reviewed literatures in Chinese (113) and in English (74), 43 master's theses, and 9 doctoral dissertations. The methods and results in these studies were compiled, and a meta-analysis of the source contributions for PM was conducted to provide a quantitative estimation of the important source types and their contributions in various regions in China. Changes in the source contributions of PM over long term periods were investigated and discussed in locations where such long term data were available.

This review is organized as follows. In Section 2, we reviewed the chronological changes in source apportionment studies and the methods used. In Section 3, we compiled the source apportionment results and identified the major sources of PM in various regions of China. The results of $PM_{2.5}$ are shown in the manuscript and the results of PM_{10} are included in the supplemental materials. In Section 4, we summarized the source contributions of PM in two cities where multiple studies have been taken in different periods over more than a decade time period, and the changes of source contributions were investigated. We also discussed future trends of $PM_{2.5}$ emissions, energy consumption and their sources upon different energy and emission scenarios. In Section 5, we discussed a few issues in the source apportionment studies reviewed in this study. Finally in Section 6, we summarized the major findings and discussed the needs for future PM source apportionment studies in China.

2. Source apportionment studies in China

2.1. Chronological changes in source apportionment studies and methods

Table 1 summarizes the number of PM source apportionment studies in China during different periods. Only a few source apportionment (SA) studies were conducted before 2000. There has been an explosive growth on the number of studies on SA of PM in China since 2010. The SA methods used in these studies are also summarized and listed in Table 1. Some studies utilized multiple methods, so the total number of the methods may be greater than the total number of studies. The methods applied in China can be mainly divided into three general types: (1) receptor models, including chemical mass balance (CMB),

Table 1
Number of particulate matter source apportionment studies in China by applying various methods during different periods compiled in this review in 1980–2016 (This is the study years. The publication year is 1987–2017, the same as Table 2, Table S1, Fig. 1, Fig. 2, and Fig. 4).

Period	Number of studies	CMB	PMF	EF	PCA	FA	AQM	Others
1980–1994	5	4	0	0	0	1	0	0
1995–1999	4	3	1	0	0	0	0	0
2000–2004	39	19	8	2	4	2	7	4
2005–2009	57	15	18	11	19	10	2	9
2010–2016	145	33	57	12	30	13	12	27

positive matrix factorization (PMF), enrichment factor (EF), principle component analysis (PCA), and factor analysis (FA); (2) air quality models (AQM), such as the Community Multi-scale Air Quality model (CMAQ), the Comprehensive Air Quality Model with Extensions (CAMx), the Atmospheric Dispersion Modeling System (ADMS), etc.; and (3) other methods (Others), including isotope ratio, single particle aerosol mass spectrometry, scanning electron microscopes (SEM), ratio analysis, cluster analysis, etc.. Details about the theory of the methods have been provided in previous reviews (Zhang et al., 2015b; Zheng et al., 2014), therefore are not repeated in this study.

Overall, receptor models have been the most popular method for source apportionment studies in China. FA (Zhang et al., 1987) and CMB (Chen et al., 1994) were the early choices for source apportionment studies. The earliest dataset apportioned with PMF was 1998 (Huang et al., 2009), and gradually replaced CMB and became the most popular receptor method. Since 2000, methods used for source apportionment studies become more diverse. PMF, CMB, and PCA have been the most frequently used source apportionment methods in China, accounting for 31.0%, 17.9%, and 16.3% of the studies after 2010. However, AQM accounted for 15.2% more than PCA, becoming the third frequently used SA method during 2000–2004. A few AQM studies have been carried out for regional SA of PM, but the total number (21) is still small compared to the receptor method (262), accounting for only 6.5% of all the studies. Each method has its own features. Results from the CMB model have relatively clear physical meaning as CMB requires detailed profiles for all sources to estimate. Therefore, profile testing for some local sources becomes a first step in order to accurately estimate their contributions. The PMF model does not require known profiles and easy for application. However, it is sometimes difficult to determine the actual sources of the factors identified by PMF. In addition, both CMB and PMF have difficulties to apportion contributions of different sources to secondary pollutants because of the high nonlinearity of the chemical processes. AQM methods can apportion the sources of secondary pollutants and also can determine the contributions from different sources/regions to target locations. However, AQM results are highly affected by the accuracy of emission inventories and the method is technically more difficult for application.

2.2. Source apportionment studies in different regions of China

China has a vast territory with distinct landscape and climate in different parts of China. Studies have revealed substantial variability of PM pollution in different regions of China (Chan and Yao, 2008; Hu et al., 2014a; Hu et al., 2015; Wang et al., 2014f; Zhang and Cao, 2015). The source regions have been grouped into seven parts in this review, following the same definitions in a previous study (Ying et al., 2014): (1) North China, (2) Northeast China, (3) East China, (4) Central China, (5) South China, (6) Southwest China, and (7) Northwest China, as shown in Fig. 1. 86 and 71 studies have been conducted in East and North China, respectively. Relative fewer studies have been conducted in other regions (28, 26, 21, 17, and 16 studies in South, Southwest, Northwest, Central, and Northeast China, respectively). Despite the substantial difference in the number of studies among the regions, the percentages of source apportionment methods used in these studies are similar, as shown in Fig. 1. In general, CMB, PMF, and PCA were the main techniques in all regions, accounting for a total of 44–76.7%. In comparison, AQM method only accounts for 4.5–16.7% of the studies.

Table S1 shows the number of the studies in the five key air pollution regions of China: (1) Jing-Jin-Ji (JJJ) in North China, (2) Yangtze River Delta (YRD) in East China, (3) Pearl River Delta (PRD) in South China, (4) Guanzhong Plain (GZP) in Northwest China, and (5) Sichuan Basin (SCB) in Southwest China. The five regions are characterized by high pollution levels and therefore have been the focus of the SA studies in China. The number of studies for JJJ accounts for 95.8% of the studies in North China, and the fraction is 75.6% for YRD in East China, 75% for PRD in South China, 42.9% for GZP in Northwest China, and 88.5% for SCB in Southwest China, respectively. Table S1 also shows that most of the source apportionment studies using AQM have been carried out in these five key regions, although CMB, PMF, and PCA are still the most frequently used techniques.

3. Major sources of particulate matter in different regions of China

3.1. Sources of bulk PM mass

The source apportionment results in individual studies were extracted and compiled to identify the major sources of PM in various regions of China. Even though direct comparison of different methods applied for different episodes in different studies may lead to some uncertainties in the source contribution estimation, it still provides valuable information about the relative importance of different sources. Seven major source categories were frequently observed in the studies: (1) dust, (2) fossil fuel combustion, (3) transportation emission, (4) biomass burning, (5) industrial emission, (6) SIA (including sulfate, nitrate, and ammonia), and (7) SOA. While SIA and SOA are due to gaseous precursor emissions from many different sources, they are listed as separate source categories here as source contributions to these secondary components were not determined in receptor-based studies. In total, there are 87 studies that have source contributions of secondary aerosols. 69 studies refer to SIA only while 1 study refers to SOA only. 10 studies have estimated the contributions of SIA and SOA from detailed sources, while 7 studies reported SIA and SOA together as secondary sources. Therefore, we summarized the contributions of SIA and SOA using the data in the 80 studies that could count SIA and SOA contributions separately. Table S2 in the supplemental materials shows how different sources reported in the reviewed studies are mapped to the above sources. It should be noted that the above seven identified sources are general categories. Overlap of certain source categories in different studies may exist due to their estimate methods. More discussion of this problem is in Section 5. Fig. 2 illustrates the relative contribution of each source category in the seven regions of China during 1980–2006 and 2007–2016. We chose 2006 to separate the study time because SO_2 and total PM emissions peaked in 2006 and then reduced afterwards due to wide application of flue-gas desulfurization (FGD) devices in coal-fired power plants in response to a new policy of China's government in 2006 (Xu et al., 2009).

Contributions from these sources vary significantly in different regions. In North China, the top three sources are dust, SIA and fossil fuel in 1980–2006, with the average contribution of 25.4%, 23.8% and 18.7%, respectively. Industrial emission, SOA, transportation and biomass burning are also very important sources, accounting for 17.5%, 15%, 11.4%, and 11.0% of $PM_{2.5}$ in this region, respectively. After 2006, industrial emission increased and became the highest contributing source of $PM_{2.5}$, with an average contribution of 28.7% in 2007–2016. The other two top sources are SIA (26.4%) and dust (17.5%). Fossil fuel and transportation are also important sources, contributing to 11.7% and 10.2% of total $PM_{2.5}$ in this region. Biomass burning and SOA are the two least important sources, accounting for 8.7% and 8.5%.

No studies were reported in 1980–2006 in Northeast China. The major sources of $PM_{2.5}$ and their relative contributions in Northeast China in 2007–2016 are similar to those in North China. The top three sources are industrial (22.2%), dust (20.2%), and fossil fuel (18.1%), followed by biomass (18%), SIA (15.6%) and transportation (8.5%). There is only one study conducted in Northeast China for biomass burning during this period. The contribution from SOA is marginal.

In East China, the top three sources are dust, SIA and transportation in 1980–2006, with the average contribution of 33.2%, 22.4% and 19.5%, respectively. Fossil fuel and industrial emission are also very important sources, contributing 18.4%, 16.6%. Biomass and SOA were not identified in the SA studies. After 2006, SIA became the top one source accounting for 39.1% in 2007–2016. Dust, industrial and fossil

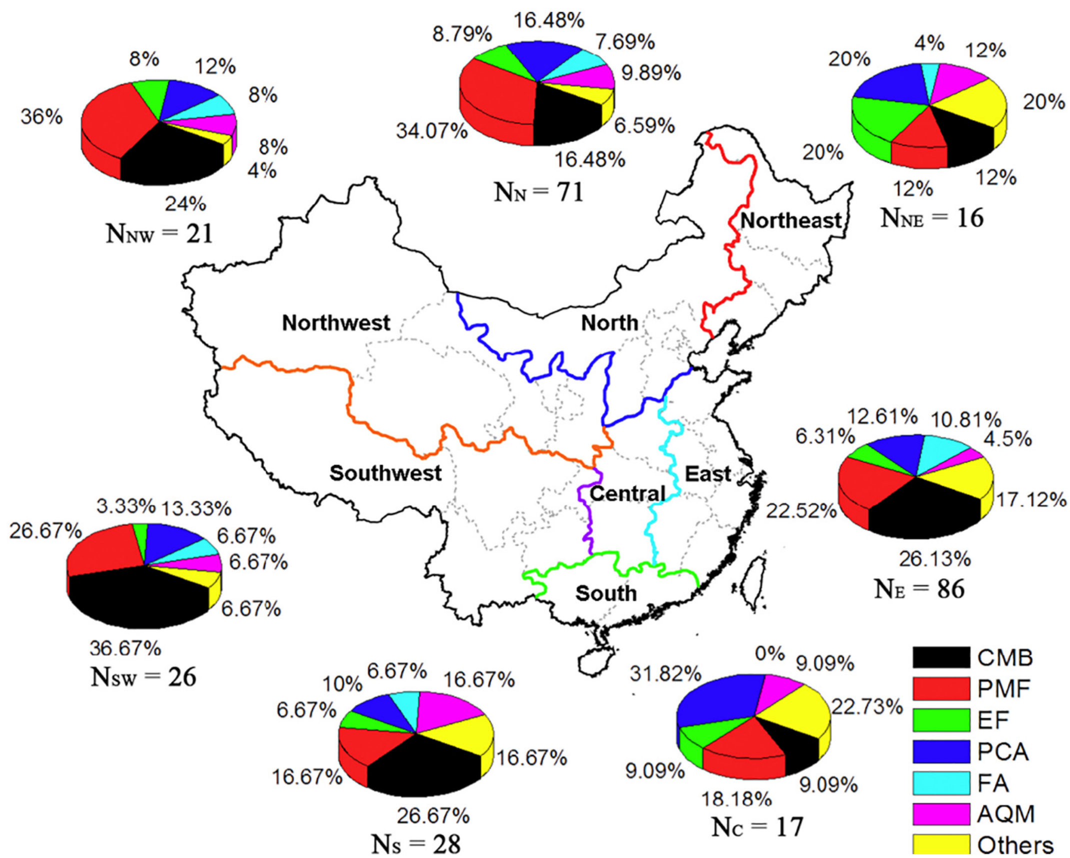

Fig. 1. Distribution of various particulate matter source apportionment methods applied in different regions of China during 1980–2016.

*The references reviewed in each region are as follows.

(1) North China (Cao, 2012; Chen et al., 2013a; Chen, 2007; Chen et al., 1994; Cheng et al., 2007; Cheng et al., 2014; Ding et al., 2010; Ding, 2012; Dong et al., 2013; Dong et al., 2012; Duan et al., 2009; Fang et al., 2016; Fu et al., 2016; Gao et al., 2016; Gao, 2012b; Gu et al., 2010; Gu et al., 2013; Guo, 2015; Han et al., 2016; He et al., 2002; Huang et al., 2014a; Huo et al., 2011; Ji et al., 2006; Kong et al., 2010; Li et al., 2010; Li et al., 2015b; Liang et al., 2015; Liu et al., 2015b; Ma, 2007; Ma et al., 2016a; Meng et al., 2016; Shi, 2010; Shi et al., 2016a; Shi et al., 2016b; Shi et al., 2016c; Shi et al., 2017b; Song et al., 2002; Song et al., 2007; Song et al., 2006a; Song et al., 2006b; Tan et al., 2016b; Tao et al., 2016; Tian et al., 2016b; Wang et al., 2009; Wang et al., 2008; Wang et al., 2016c; Wang et al., 2015a; Wang et al., 2014c; Wang et al., 2015c; Wang et al., 2012b; Wei, 2014; Wei et al., 2015; Wu et al., 2014b; Xu et al., 2007; Yang et al., 2016; Yang et al., 2001; Yang, 2007; Yu et al., 2013; Zhang et al., 2015a; Zhang et al., 2012; Zhang et al., 2016c; Zhang et al., 2013a; Zhang, 2016a; Zhang et al., 2007; Zhao et al., 2009; Zheng et al., 2005; Zhou et al., 2016a; Zhu et al., 1995; Zhu et al., 1996; Zhu et al., 2005)

(2) Northeast China (Fang et al., 2015; Huang and Wang, 2014; Jia, 2014; Liu, 2014; Liu et al., 2015d; Luan et al., 2016; Sha, 2007; Shao, 2004; Shi et al., 2017b; Wang, 2008; Wang et al., 2012a; Wang, 2016; Zhang, 2014a; Zhao et al., 2015; Zheng and Lv, 2015)

(3) East China (An et al., 2014; Bao et al., 2010; Cao et al., 2016; Chen et al., 2013a; Chen, 2011; Chen et al., 2016b; Chen et al., 2016c; Chen et al., 2015b; Chen et al., 2017; Chen et al., 2016e; Chuang et al., 2016; Cui, 2005; Ding et al., 2014; Ding, 2004; Fan et al., 2016; Fan et al., 2005; Feng et al., 2004; Gao, 2012a; Gao, 2012b; Gong, 2013; Gu, 2009; Han et al., 2009; Hou, 2012; Hu et al., 2013; Hu, 2016; Hu et al., 2014b; Huang et al., 2006; Huang et al., 2014a; Huang et al., 2014d; Jiang et al., 2015; Li et al., 2016; Li et al., 2015a; Li, 2013; Li and Li, 2016; Liu et al., 2016a; Liu et al., 2015a; Liu, 2006; Liu et al., 2001; Liu, 2016a; Liu and Gan, 2014; Lu et al., 2008; Peng, 2009; Qi et al., 2016; Qiao et al., 2016; Qin, 2015; Qiu, 2012; Shen et al., 2014; Shi, 2010; Shi et al., 2017b; Tang et al., 2014; Tang et al., 2015; Tao, 2016; Wang et al., 2013; Wang et al., 2016a; Wang et al., 2016b; Wang, 2015a; Wang et al., 2016d; Wang et al., 2015d; Wang et al., 2016e; Wang et al., 2015e; Wang et al., 2015f; Wu et al., 2013b; Wu et al., 2014a; Wu, 2014; Xiao, 2007; Xiao et al., 2012; Xu et al., 2016b; Yan, 2011; Yang, 2008; Yang et al., 2013; Yang et al., 2010; Yao et al., 2016; Yao et al., 2010; Ye, 2011; Yin, 2016; Yu et al., 2015; Yue et al., 2006; Zhang and Chen, 2015; Zhang et al., 2016b; Zhang et al., 2012; Zhang and Zhuang, 2007; Zhang et al., 2016d; Zhang, 2014b; Zhang et al., 2016e; Zhao et al., 2016)

(4) Central China (Chen et al., 2013b; Chen et al., 2015c; Gao, 2012b; Geng, 2012; Kang et al., 2015; Ke, 2015; Liu et al., 2016b; Shi, 2007; Shi et al., 2017b; Song et al., 2016; Wang et al., 2016h; Yang, 2010; Yu et al., 2016; Zeng, 2011; Zhang et al., 2016a; Zhou et al., 2015)

(5) South China (Chen et al., 2016a; Chen et al., 2016d; Cheng et al., 2009; Guo et al., 2009; He, 2006; Ho et al., 2006; Hu et al., 2009; Huang et al., 2014a; Huang et al., 2015; Huang et al., 2009; Huang et al., 2014c; Li, 2007; Li et al., 2013; Li et al., 2012; Liu et al., 2015c; Lu and Fung, 2016; Luo, 2006; Ma et al., 2015; Ma et al., 2016b; Shi et al., 2017b; Song et al., 2015; Tan et al., 2016a; Wu et al., 2013a; Wu et al., 2016; Zhang et al., 2012; Zhao, 2005; Zhou et al., 2016b)

(6) Southwest China (Chen et al., 2015a; Chen et al., 1996; Chen, 2009; Dai et al., 2009; Fan et al., 2015; Jiao et al., 2014; Liang, 2015; Lin et al., 2016; Ren et al., 2014; Shi, 2010; Shi et al., 2017a; Shi et al., 2017b; Sun, 2011; Tao, 2003; Tao et al., 2011; Tao et al., 2006; Tao et al., 2014; Tian et al., 2015; Tian et al., 2013; Wang, 2015b; Xiang et al., 2016; Zhang et al., 2014; Zhang et al., 2012; Zhang, 2016b; Zhang et al., 2013b)

(7) Northwest China (Cao et al., 2005; Dou et al., 2016; Feng et al., 2005; Gao, 2012a; Huang et al., 2014a; Li, 2010; Liu, 2016b; Luo, 2015; Ma et al., 2016a; Qiu et al., 2016; Shi, 2010; Shi et al., 2017b; Tian et al., 2016a; Wang et al., 2014a; Wang et al., 2014b; Wang et al., 2015b; Wang et al., 2016f; Wang et al., 2014e; Xu et al., 2016a; Zhang et al., 1987).

fuel are also very important sources, contributing 18.3%, 17.2%, and 15.9% of $PM_{2.5}$ in this region. Transportation, SOA and biomass account for 14.2%, 12.0% and 8.5%.

In the central region of China during 2007–2016, industrial, SIA and dust are the most important sources, with an average contribution of 28.0%, 26.0% and 23.9%, respectively. Transportation, fossil fuel and SOA account for 16.8%, 12.6% and 10.7%. Biomass burning is not identified as a source of $PM_{2.5}$ in Central China.

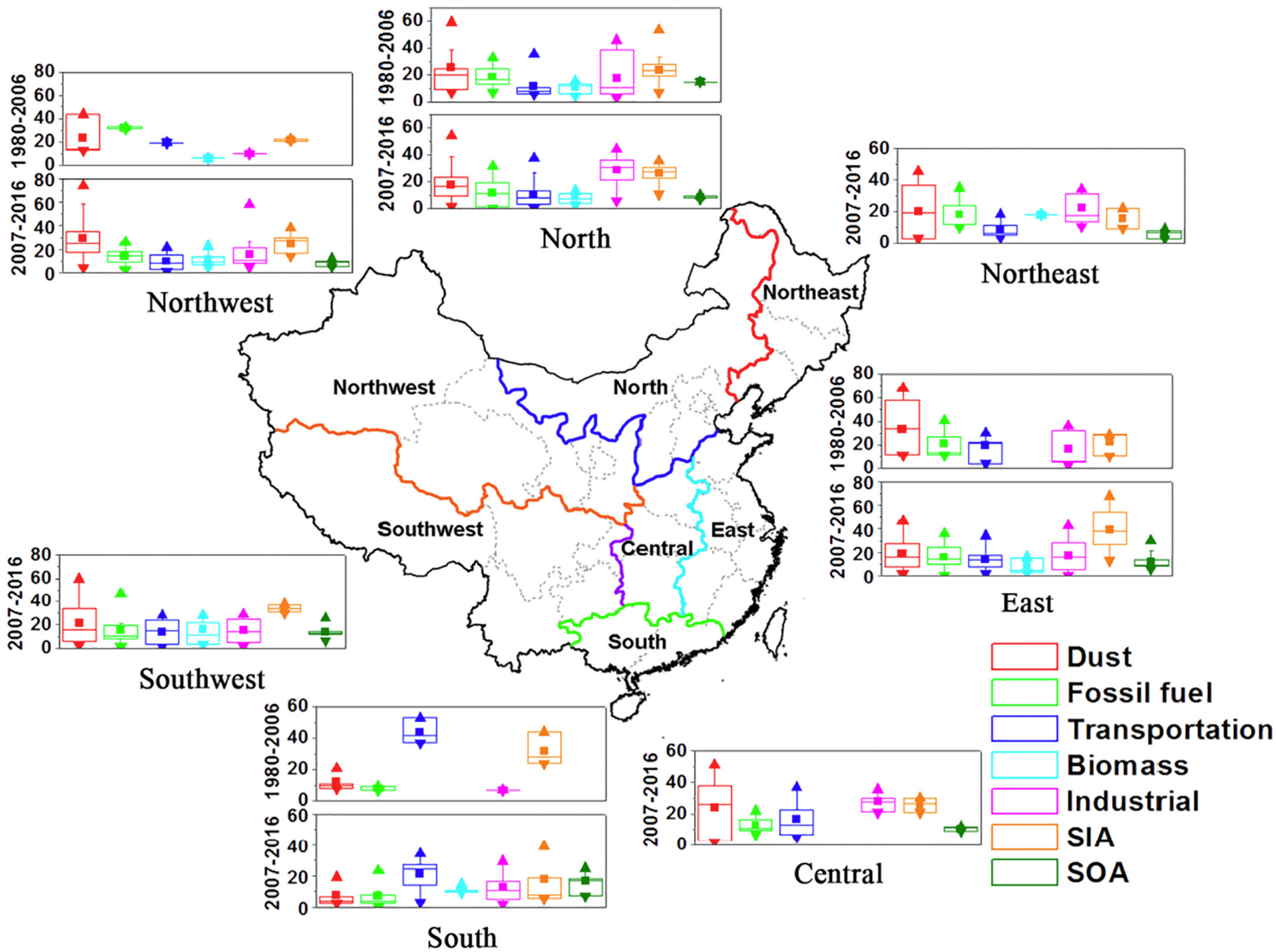

Fig. 2. Major sources of particulate matter in different regions of China during 1980–2016 compiled in this review. The y-axis in each box chart is the contribution (%) of each source type. The maximum, mean, and minimum value are represented by the top edge, middle line, and the bottom edge of each box. The central box represents the values from the lower to upper quartile (25th to 75th percentile). The vertical line extends from the 10th percentile to the 90th percentile. The middle solid line represents the median. The solid squares represent the arithmetic average. Outliers are plotted as triangles.
* The references that we used the data in each region are as follows.

(1) North	1980–2006	(Chen et al., 1994; Gao, 2012a; He et al., 2002; Li et al., 2010; Li et al., 2015b; Song et al., 2007; Song et al., 2006a; Song et al., 2006b; Wang et al., 2008; Yang, 2007; Zhang et al., 2007; Zheng et al., 2005; Zhu et al., 2005)
	2007–2016	(Gao et al., 2016; Gao, 2012a; Guo, 2015; Han et al., 2016; Kong et al., 2010; Li et al., 2015b; Ma et al., 2016a; Meng et al., 2016; Shi et al., 2016a; Shi et al., 2016c; Shi et al., 2017b; Tao et al., 2016; Tian et al., 2016b; Wang et al., 2016c; Wang et al., 2015a; Wang et al., 2015c; Wang et al., 2012b; Wei, 2014; Wei et al., 2015; Wu et al., 2014b; Yang et al., 2016; Yu et al., 2013; Zhang et al., 2016c; Zhang et al., 2013a; Zhang, 2016a)
(2) Northeast	2007–2016	(Huang and Wang, 2014; Jia, 2014; Shi et al., 2017b; Wang, 2016; Zheng and Lv, 2015)
(3) East	1980–2006	(Bao et al., 2010; Fan et al., 2005; Gao, 2012a; Huang et al., 2006; Liu et al., 2015a; Lu et al., 2008; Yue et al., 2006; Zhang and Zhuang, 2007)
	2007–2016	(An et al., 2014; Chen et al., 2016b; Chen et al., 2016c; Chen et al., 2015b; Chuang et al., 2016; Ding et al., 2014; Li et al., 2016; Li et al., 2015a; Liu et al., 2016a; Peng, 2009; Qi et al., 2016; Qiao et al., 2016; Shi et al., 2017b; Wang et al., 2016b; Wang, 2015a; Wang et al., 2015d; Wang et al., 2015e; Wu et al., 2013b; Wu, 2014; Xiao, 2007; Xiao et al., 2012; Xu et al., 2016b; Yan, 2011; Yang et al., 2013; Yang et al., 2010; Yao et al., 2016; Ye, 2011; Yu et al., 2015; Zhang, 2014b)
(4) Central	2007–2016	(Chen et al., 2015c; Geng, 2012; Kang et al., 2015; Shi et al., 2017b; Wang et al., 2016h; Yang, 2010; Zhou et al., 2015)
(5) South	1980–2006	(Guo et al., 2009; Ho et al., 2006)
	2007–2016	(Chen et al., 2016a; Huang et al., 2015; Huang et al., 2014c; Shi et al., 2017b; Song et al., 2015; Tan et al., 2016a; Wu et al., 2013a)
(6) Southwest	2007–2016	(Chen, 2009; Dai et al., 2009; Fan et al., 2015; Jiao et al., 2014; Liang, 2015; Lin et al., 2016; Ren et al., 2014; Shi et al., 2017a; Shi et al., 2017b; Tao et al., 2014; Tian et al., 2015; Tian et al., 2013; Wang, 2015b; Zhang et al., 2013b)
(7) Northwest	1980–2006	(Gao, 2012a; Xu et al., 2016a; Zhang et al., 1987)
	2007–2016	(Dou et al., 2016; Liu, 2016b; Luo, 2015; Qiu et al., 2016; Shi et al., 2017b; Wang et al., 2014a; Wang et al., 2014b; Wang et al., 2015b; Wang et al., 2016f; Wang et al., 2016g; Xu et al., 2016a)

Table 2
Top three sources in each region over China during 2007–2016.

Region	1st source	2nd source	3rd source
North	Industrial	SIA	Dust
Northeast	Industrial	Dust	Fossil fuel
East	SIA	Dust	Industrial
Central	Industrial	SIA	Dust
South	Transportation	SIA	SOA
Southwest	SIA	Dust	Biomass
Northwest	Dust	SIA	Industrial

In South China, the contributions from transportation and SIA are remarkably significant, with mean values of 44% and 32% in 1980–2006, and 21.5% and 17.8% in 2007–2016, respectively. Dust contributes to 12.4% of $PM_{2.5}$, followed by the fossil fuel contribution of 8% in 1980–2006. In 2007–2016, SOA, industrial and biomass are also very important sources, contributing to 16.8%, 12.9% and 11.4% of $PM_{2.5}$, followed by dust (7.7%) and fossil fuel (7.0%).

Southwest and Northwest China share the same top two sources of $PM_{2.5}$, which are SIA (33.4% vs. 24.5%), and dust (21.3% vs. 29.0%) in 2007–2016. In 1980–2006 these two sources in Northwest account for 21.6% and 23.3%, respectively. Biomass, fossil fuel, industrial emission, SOA and transportation are also important sources in Southwest, contributing to 16.1%, 15.7%, 15.4% 14.2% and 14.1% of $PM_{2.5}$ in 2007–2016. Fossil fuel (31.8%), transportation (19.3%) in 1980–2006 and industrial sources (15.6%) in 2007–2016 have significant contributions in Northwest.

The top three sources in each region over China during 2007–2016 are summarized in Table 2. It is obvious that SIA, dust and industrial emission are generally the most important three sources for $PM_{2.5}$ in almost all regions over China, due to a huge backer of infrastructure projects and large amount of industrial production. The situation is slightly different in South, Northeast and Southwest China. In South China, the influence of transportation and SOA source becomes more important due to relatively more motorization, while in Northeast/Southwest China, fossil fuel/biomass is a very important source due to large amount of coal combustion for residential heating/field burning of crop residue.

3.2. Sources of SIA and SOA

SIA and SOA account for a large fraction of $PM_{2.5}$ in China (Huang et al., 2014b), the contributions from different sources to SIA and SOA need to be estimated to design effective emission control programs. Source apportionment of SIA and SOA typically cannot be resolved by the receptor models, and AQM models are usually used to determine the source contributions of SIA and SOA. Source contributions of SIA in $PM_{2.5}$ are summarized from available AQM source apportionment studies in China. These studies used different AQM models including CMAQ (in 11 studies) and CAMx (in 5 studies). Other models used include the ADMS and AERMOD dispersion models and a nested air quality prediction modeling system (NAQPMS) (in 4 studies). Different source apportionment methods such as brute force (in 6 studies), non-reactive tracer method (in 8 studies), and source-oriented methods (in 6 studies) have been applied. It should be noted that due to the complex non-linear dependencies to precursor emissions in the SIA formation, negative or over 100% contributions may appear when using the brute force source apportionment method. Fig. 3 shows the results of SIA in different regions. Fig. 3a shows that fossil fuel and industrial sources combined account for 63.5%–88.1% of SO_4^{2-} in all regions. Residential emissions are also important, accounting for 9.3%–13.6%, except in the PRD and YRD regions. Contributions from transportation emissions to SO_4^{2-} are generally low except in one study in PRD (Lu and Fung, 2016) which estimated relatively large emissions of SO_2 from vehicles. For NO_3^-, as shown in Fig. 3b, fossil fuel and industrial emissions are still the most important sources (47.3%–70%), which are similar to their relative contributions to SO_4^{2-}. But different from SO_4^{2-}, transportation contributes to 22%–34% of NO_3^- in all regions. Agriculture is the primary source of NH_4^+ (53.9%–90%) in all regions, as shown in Fig. 3c. The sources and their contributions to SIA in some cities are shown in Fig. S1 in the supplemental materials.

Only a few source apportionment studies of SOA were reported in the literature. Cheng et al. (2009) estimated the source contributions to SOA in fall of 2004 in the PRD region using the two-dimensional model with the brute force method. The study found that biogenic, transportation, point, and solvent and oil paint sources accounted for 72.6%, 30.7%, 12% and 12% of $PM_{2.5}$, respectively (note that the total is over 100% by the brute force method). Wang et al. (2017) applied CMAQ model with a source-oriented SOA module and found that biogenic emissions contributed significantly to SOA in summer (68.7%) and industrial (39%) and residential (42.2%) sources were the main winter SOA contributors in China when using the Multi-resolution Emission Inventory for China (MEIC). However, the study also indicated that the source contributions to SOA could be substantially different when using the Regional Emission inventory in ASia v2.1 (REAS2), although the total predicted SOA concentrations were similar. More studies on the source contributions to SOA are needed to have a comprehensive understanding about the sources of SOA in China.

4. Changes in source contributions of particulate matter in two cities of China

Only three cities (Beijing, Nanjing, and Chongqing) were found to have multiple source apportionment studies conducted in different years over more than a decade. The results of Chongqing are of TSP and PM_{10} and only have 1 or 2 studies during the 5-year periods, therefore the data in Chongqing were not used. The results were compiled and shown in Fig. 4. Fig. 4a presents the $PM_{2.5}$ averaged concentrations and the relative contributions to $PM_{2.5}$ from each source during five five-year periods between 1985 and 2016 (i.e. 1985–1989, 1995–1999, 2000–2004, 2005–2009, 2010–2016; no studies for 1990–1994 were found) in Beijing. The averaged $PM_{2.5}$ concentrations were 78.5, 142 ± 4, 108.1 ± 19.4, 107.5 ± 31.8, and 86.8 ± 25.0 μg/m^3, respectively. The major sources were transportation emission, fossil fuel combustion and dust in 1985–1989. Source contribution of transportation decreased sharply from 35.7% in 1985–1989 to 8.7% ± 0.2% in 1995–1999, 10.3% ± 7.5% in 2000–2004 and 5.7% ± 2.6% after 2005–2009, but increased to 14.5% ± 8.9% after 2010. The contribution from fossil fuel combustion decreased to 12.3% ± 6.5% in 2005–2009 but increased slightly to 15.0% ± 6.6% after 2010. The contribution from dust source dropped down from 24.1% in 1985–1989 to 13.6% ± 7.4% in 2010–2016, except for an increase of 30.5% ± 18.7% in 2005–2009. SIA source has become the major source ever since 1995 and increased to 25.4% ± 1.9% after 2010. The percentage from industrial source was as low as 13.8% ± 0.7% in 1995–1999, and then prominently increased to 21.3% ± 15.0% in 2005–2009. The contribution from industrial sources decreased to 14.6% ± 12.8% after 2010 possibly due to emission controls. There is little change in the contribution of biomass burning (approximately 10% ± 3.4%) since 2000. The contribution of SOA was estimated in only a few studies during 2000–2004 and after 2010, and therefore the long-term trend of its contribution to $PM_{2.5}$ cannot be determined.

Fig. 4b shows the changes in percentage source contributions to $PM_{2.5}$ during 2000–2016 in Nanjing. No data were available before 2000 in Nanjing. The most significant change in Nanjing is that the contribution from SIA source had boomed from 10.5% to 43.9% ± 21.2% since 2000. Dust source contribution decreased from more than half in 2000–2004 to 10.2% ± 9.9% during 2010–2016. Fossil fuel combustion also exhibits a declining trend. It accounts for 22.4% ± 8.6%/27.2% of $PM_{2.5}$ in 2000–2004/2005–2009 but its contribution decreases to 10.7% ± 8.3% in 2010–2016. The highest

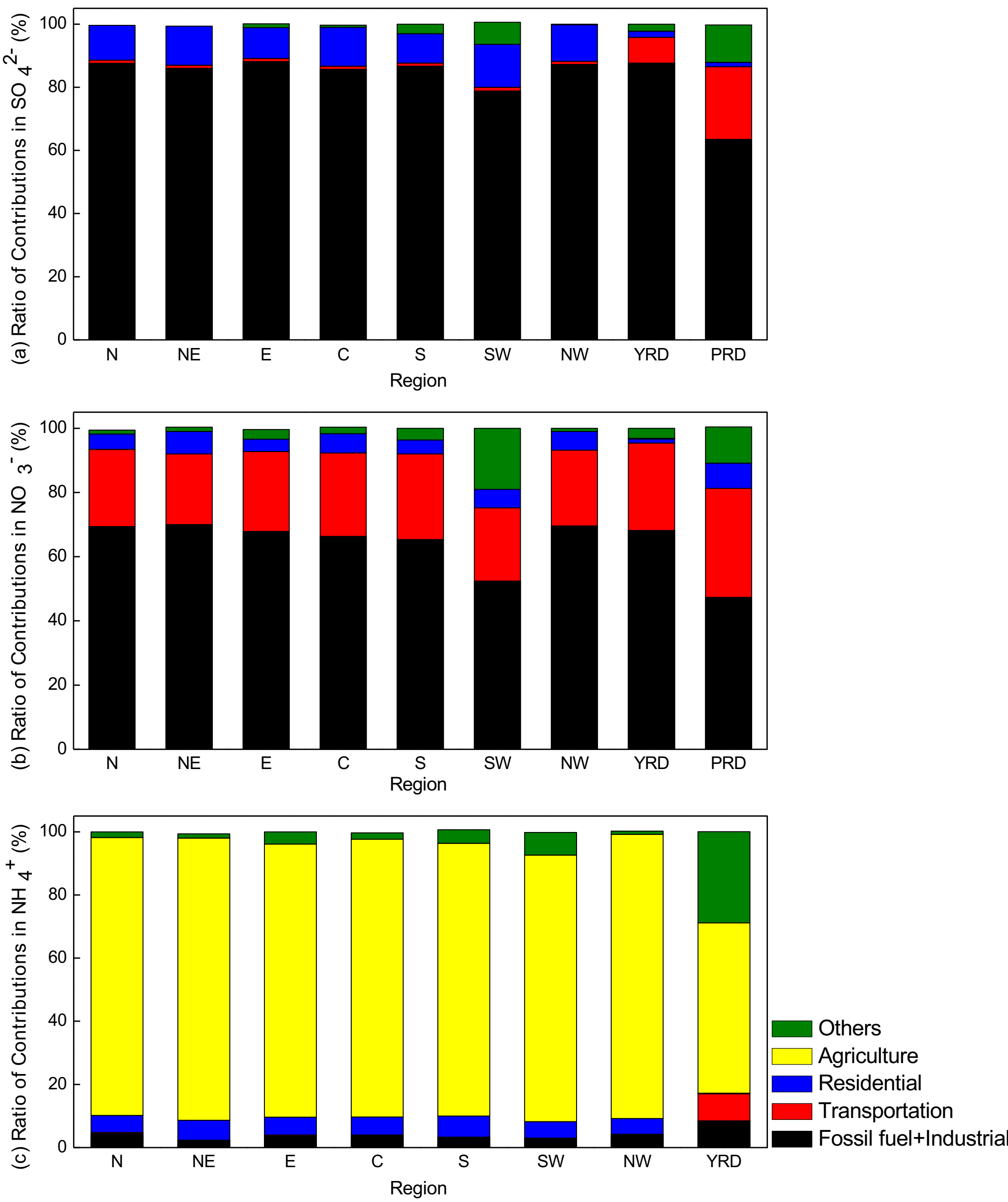

Fig. 3. Sources and contributions to $PM_{2.5}$ (a) SO_4^{2-}, (b) NO_3^- and (c) NH_4^+ in the regions of China using AQM.
*N = North China, NE = Northeast China, E = East China, C = Central China, S = South China, SW = Southwest China, NW = Northwest China, YRD = Yangtze River Delta, PRD = Pearl River Delta.
* The references that we used the data and their methods in each region are as follows.
(1) N, NE, E, C, S, SW, NW: (Shi et al., 2017b) CMAQ
(2) PRD: (Lu and Fung, 2016) WRF (Weather Research Forecast) and SMOKE (Sparse Matrix Operator Kernel Emission)-CAMx modeling system with PSAT (particulate source apportionment technology) module
(3) YRD: (Li et al., 2015a) PSAT method coupled within CAMx.

relative contribution of industrial source to $PM_{2.5}$ (20.2%) occurred during 2005–2009. This was significantly higher than the percentage contribution of 4.6% ± 1.3% in 2000–2004 and 9.6% ± 6.4% after 2010. Similar to Beijing, the contribution from transportation source showed an obvious increase from 4.0% in 2000–2004 to 17.8% ± 6.3% in 2010–2016.

Central and local Chinese governments recently have developed emission control policies aiming to improve air quality before 2030. Control policy contributing to reductions of $PM_{2.5}$ emissions include energy-saving measures, e.g., energy efficiency improvements, cogeneration of heat and power, fuel substitution, and end-of-pipe control measures such as installations of dust collectors and flue gas desulfurization systems. The policies have led to the source emission trends that partly have been observed in Fig. 4, but also will drive the future emission trends. Fig. 5 illustrates future trends of $PM_{2.5}$ emissions, energy consumption and $PM_{2.5}$ emission rate from the major sources in China during 2005–2030. The calculation of $PM_{2.5}$ emission rate is $PM_{2.5}$ emissions / energy consumption ∗ 100. The data are derived from the study of Wang et al. (2014d) and the references therein. Two energy scenarios business-as-usual (BAU) and alternative policy (PC) are discussed. The BAU scenario is based on that current regulations and implementation status will be continued during 2011–2030. The PC

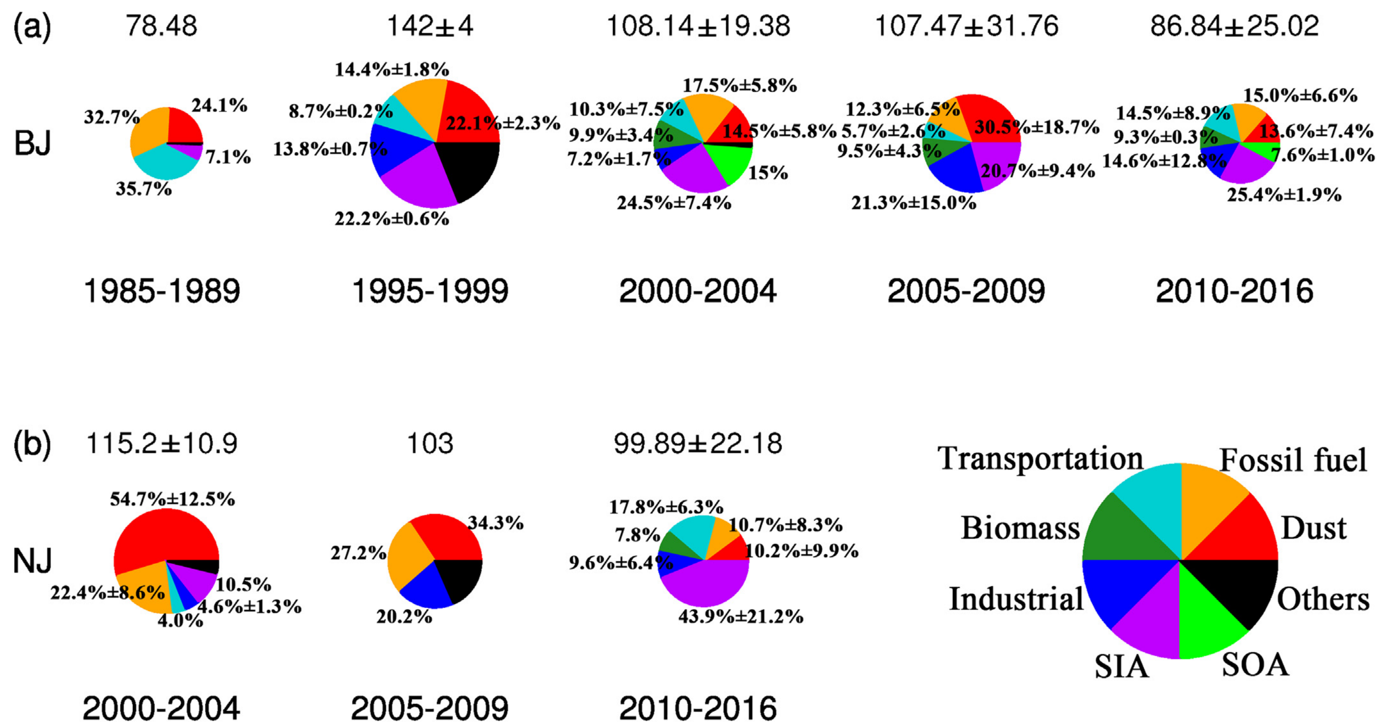

Fig. 4. Chronological changes in source contributions of $PM_{2.5}$ in (a) Beijing (BJ) and (b) Nanjing (NJ) considered in this review. Units are μg/m^3.
*The references that we used the data in each city are as follows.

(1)		1985–1989	(Chen et al., 1994)
	Beijing	1995–1999	(He et al., 2002)
		2000–2004	(Song et al., 2007; Song et al., 2006a; Song et al., 2006b; Wang et al., 2008; Zhang et al., 2007; Zheng et al., 2005; Zhu et al., 2005)
		2005–2009	(Gao, 2012a; Li et al., 2015b; Tao et al., 2016; Wang et al., 2009; Wang et al., 2008; Yang, 2007; Zhang et al., 2013a)
		2010–2016	(Gao et al., 2016; Han et al., 2016; Li et al., 2015b; Shi et al., 2017b; Wang et al., 2015c; Wang et al., 2012b; Wu et al., 2014b; Yang et al., 2016; Yu et al., 2013; Zhang et al., 2016c)
(2)		2000–2004	(Fan et al., 2005; Huang et al., 2006)
	Nanjing	2005–2009	(Yang et al., 2010)
		2010–2016	(Chen et al., 2015b; Ding et al., 2014; Hu et al., 2013; Li et al., 2016; Qi et al., 2016; Wang et al., 2016a; Wang et al., 2015d)

scenario assumes that new energy saving policies will be released and more strongly enforced starting from 2011, resulting in lifestyle changes, structural adjustment, and energy efficiency improvement. Only the data that correspond with main $PM_{2.5}$ emission sources in Fig. 5 were used. Fig. 5a shows that BAU and PC have the same effects on biomass, residential and transportation sources. Residential, biomass and transportation source increase from 2005 to 2010 and decrease then. Industrial source decreases all the time in 2005–2030. Power plant decreases after 2005 but increases a little in 2020. Fig. 5b shows the energy consumption of china in 2005–2030. All of energy consumptions except biomass burning increase during 2005–2030 in two energy scenarios. Fig. 5c shows that $PM_{2.5}$ emission rates of transportation and power plant are lower than other three sources. Residential and biomass source increase from 2005 to 2010 and then decrease while other sources decrease from 2005 to 2030. Biomass source with BAU has lower $PM_{2.5}$ emission rates than PC and residential source is just the opposite.

5. Discussion

Receptor models are the most common source apportionment methods used in China. However, major source categories and their source contributions estimated by different receptor models may be different even though the data come from the same place in the same time because many of the methods do not have a unique solution and many source tracers used to identify sources are not unique to a specific source (Zhang et al., 2015b). Zhang et al. (2015b) analyzed $PM_{2.5}$ and its chemical component at Atlanta in July, 2001 using various tracers and methods such as CMB-LGO (Lipschitz Generalized Optimization), CMB-MM (Molecular Marker), PMF, and CMAQ. The source categories were different, and therefore the contribution percentage of each source was different. Another comparative study also shows the difference in the source apportionment results using the PMF, CMB and FA models (Chen, 2011). Thus, the source contributions assembled in this study might have significant uncertainties by directly incorporating source apportionment results with different methods. However, the conclusion that dust, coal combustion, transportation emission, biomass burning, industrial emission, SIA and SOA were main sources of $PM_{2.5}$ in China is likely robust.

AQM studies can provide information of sources of SIA and SOA. However, the application of AQM in China is still relatively limited and has mainly focused on the five key air pollution regions. More AQM source apportionment studies should be conducted in China, especially in the less-studied regions. Different models and different source apportionment techniques have been used, leading to some uncertainties in the direct comparison of the results. Comparisons of different AQM models with different techniques should be conducted. In addition, the accuracy of AQM source apportionment results is often limited by the uncertainties in the emission inventories. Therefore, comparisons of AQM and receptor models should also be conducted in order to build confidence on the robustness of source apportionment results and reduce uncertainties in emission inventories.

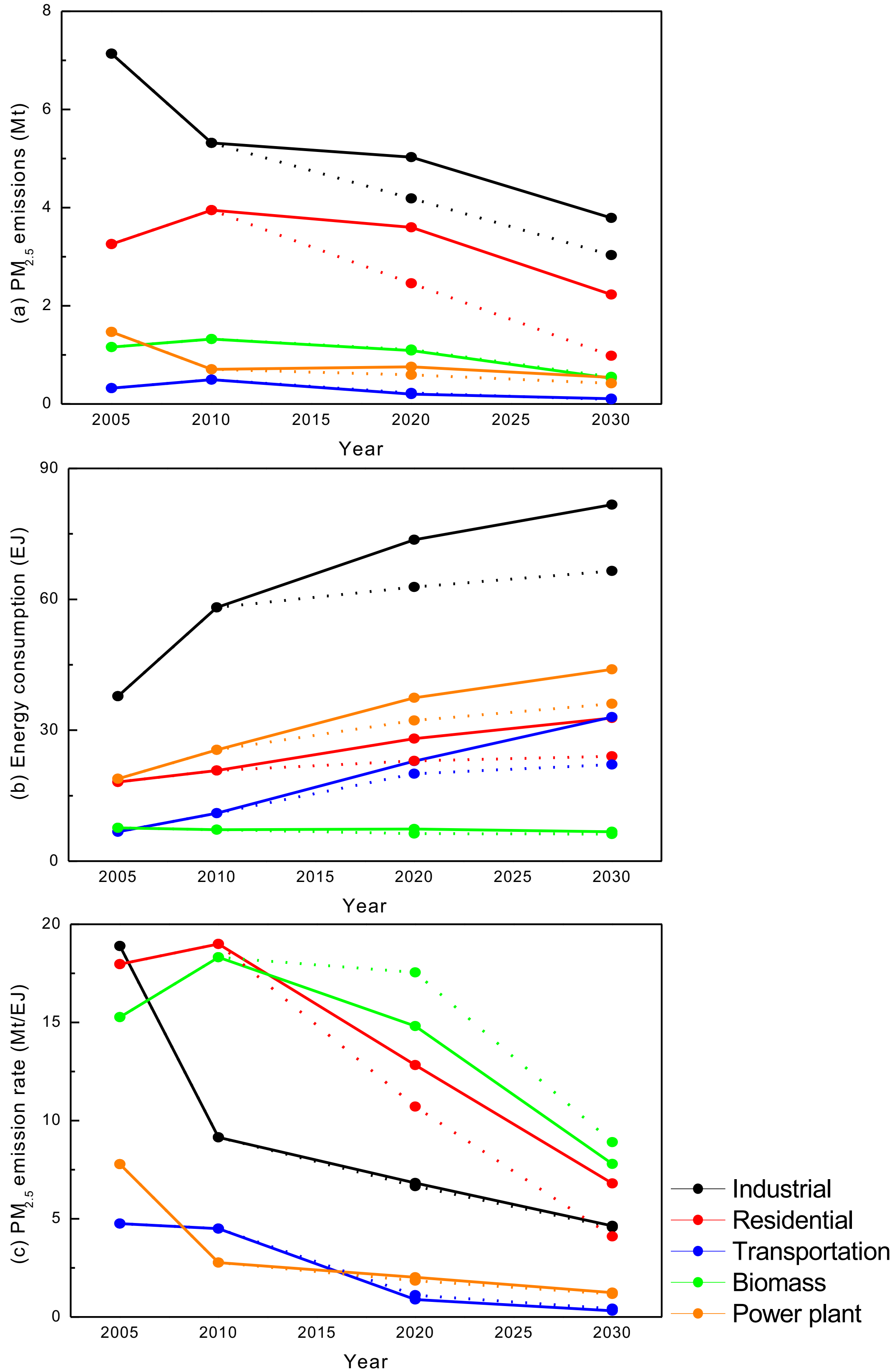

Fig. 5. Future trends of each source about (a) $PM_{2.5}$ emissions, (b) energy consumption, (c) $PM_{2.5}$ emission rate of China during 2005–2030. The data of Taiwan, Hong Kong and Macao are not included. Straight line represents BAU and dotted line represents PC.

China has been undergoing fast economy and society development. The analysis of major sources of $PM_{2.5}$ and their trends available at three cities indicated the sources have also undergone substantial changes. Long-term source apportionment studies are highly valuable for developing effective air pollution control strategies, evaluating the effectiveness of designed strategies as well as evaluating the adverse health effects due to exposure to air pollution. Such studies should be recommended in more cities/regions of China.

Most of the studies identified and estimated contributions from 6 to 8 source categories. The definitions of the source categories usually are general with many sub-source categories. For example, industrial source category includes many different industries, and transportation source category includes all vehicle types with all fuel types. Developing cost-effective emission control strategies requires more accurate analyses of the contributions from detailed sub-categories, many of them cannot be readily determined using receptor models due to collinearity and/or lack of source-specific tracers. Developing methods to resolve these sub-source categories should be a future research priority in China.

6. Conclusions

239 studies on source apportionment of $PM_{2.5}$ in China published during 1987–2017 were summarized to provide a better understanding of $PM_{2.5}$ sources for effective pollution abatement. By analyzing the methods that have been applied in these studies, identifying the main sources of particulate matter, and investigating the trends of the main sources over the years, the follow findings were made:

(1) Source apportionment studies have been conducted in various locations of China, mainly in the five key regions of JJJ in North China, YRD in East China, PRD in South China, GZP in Northwest China, and SCB in Southwest China. Sources and their contributions to $PM_{2.5}$ are still not well understood in many other regions.
(2) Up to date, the receptor models of CMB, PMF and PCA are the most frequently used methods for PM source apportionment studies in China. A few AQM source apportionment studies have been carried out, but mostly in the five key regions.
(3) Dust, fossil fuel combustion, transportation emission, biomass burning, industrial emission, SIA and SOA are identified to be the main sources of $PM_{2.5}$ in China, while SIA, dust and industrial emission were the most important sources of particulate matter in seven geographical areas during 2007–2016.
(4) A limited number of studies investigated the sources of SIA and SOA in China using regional air quality models. These studies indicated that the important sources of SIA include coal and industry (63.5%-88.1%) for SO_4^{2-}, coal and industry (47.3%-70%) and transportation (22%-34%) for NO_3^-, agriculture (53.9%-90%) for NH_4^+, respectively. Biogenic emissions are an important source of SOA in fall and summer, while residential and industrial sources are important in winter.
(5) Source contributions of PM have undergone substantial changes at Beijing and Nanjing. In general, dust and industrial contributions decreased and transportation contributions increased after 2000. The importance of the sources changes with different trends in various cities over the past 30 years.
(6) $PM_{2.5}$ emissions are predicted to decline in most regions during 2005–2030, even though the energy consumptions except biomass burning are predicted to continue increase. Industrial, residential, and biomass burning sources will become more important in the future in the businuess-as-usual senarios.

A few recommendations for future source apportionment studies in China can be made based on this comprehensive review of existing source apportionment studies:

(1) Inter-comparison of the different methods and evaluating the robustness of the models for applications in various regions of China are highly valuable and should be considered in future. The comparison studies include inter-comparisons among different receptor models, among different AQM models, as well as comparisons of receptor models and AQM models.
(2) More source apportionment studies using the AQM methods and other advanced methods such as isotope ratio and single particle aerosol mass spectrometry methods should be encouraged in China and more source apportionment studies on SIA and SOA should be conducted considering that SIA and SOA account for large fractions of PM. Accurate emission inventory are needed for AQM studies.
(3) Long-term source apportionment studies are recommended in more cities/regions to reflect the fast changing sources of China in different regions.
(4) Future source apportionment studies should identify and estimate contributions from more detailed sources other than broad source categories to provide quantitative information for accurate and effective emission control.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.03.037.

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