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Trends of $PM_{2.5}$ and Chemical Composition in Beijing, 2000–2015

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ABSTRACT

$PM_{2.5}$ is the major pollutant in most cities of China, especially contributed significantly to the poor air quality in Beijing. This study aimed to investigate the long-term trend (2000–2015) of $PM_{2.5}$, based on intensive observation and comprehensive literature investigation of $PM_{2.5}$ and its chemical components. Results showed that the annual average concentration of $PM_{2.5}$ generally decreased by 1.5 μg m^{-3} $year^{-1}$ from 2000 to 2015 under the implementation of 16 phases' air pollution control measures. In the most polluted season (winter), four change stages were found related with emission control effect and meteorological conditions: $PM_{2.5}$ decreased in 2000–2008 and 2010–2013, increased in 2008–2010, and was at a high level in 2013–2015. As for detailed chemical components, OC, soil dust and typical elements (Si, Ca, Fe, Mn, Cu, Pb and K) presented a downward trend generally. EC had almost no change before 2003, increased from 2003 to 2007, but decreased after 2007; this may be caused by the replacement of fossil fuel and control of biomass emission. The continuous rising of OC/EC and SOC/OC (secondary organic carbon, SOC) in recent years illustrated the secondary carbonaceous species pollution is becoming serious. SO_4^{2-} showed a slight increase from 2000–2013, but decreased obviously from 2013–2015. NO_3^- and NH_4^+ had an upward trend during the past decade. The proportion of secondary inorganic aerosol (SIA) increased at a rate of 0.7% yr^{-1}, and it has become the major composition of $PM_{2.5}$ in Beijing instead of carbonaceous component since 2009. In general, $PM_{2.5}$ change trend indicated the emission mitigation measures implemented in Beijing have reduced the primary $PM_{2.5}$ effectively. However, the control of secondary components should be paid special attention in order to further improve the air quality in Beijing effectively.

Keywords: Fine particle; Chemical components; Long-term trends; Beijing; Emission control effect.

INTRODUCTION

$PM_{2.5}$ (i.e., fine particles with an aerodynamic diameter smaller than 2.5 μm) is an important atmospheric pollutant, which was found to be responsible for adverse effect on human health (Jiang *et al.*, 2015; Lelieveld *et al.*, 2015; Powell *et al.*, 2015; Pedersen *et al.*, 2016), climate change (Thompson *et al.*, 2014; Wang *et al.*, 2014a; Klein *et al.*, 2016) and visibility degradation (Sun *et al.*, 2015; Zhang *et al.*, 2015b). The $PM_{2.5}$ pollution problem had been pushed to the front of public and attracted wide attention among scientists, government, media and the public in China even around the world, since the extremely severe haze pollution occurred in December 2011 in China (Pandolfi *et al.*, 2012; Yuan *et al.*, 2012; Herrmann *et al.*, 2014). $PM_{2.5}$ has been the major pollutant contributing significantly to the poor air quality in most regions of China (http://www.cnemc.cn/). Especially in the capital of China – Beijing, it is being faced with severe fine particle pollution. The annual average mass concentrations of $PM_{2.5}$ were 94.5 and 85.5 μg m^{-3} in 2013 and 2014, respectively, about 2.5 times of the National Ambient Air Quality Standards (NAAQS) (35 μg m^{-3} as class II annual average level) (BMEPB, 2015). More than 18.2 million people (98% of Beijing population) are exposed to harmful level of lone-term $PM_{2.5}$ pollution (Xie *et al.*, 2015b). As a result, the control of $PM_{2.5}$ pollution in Beijing is urgent. Beijing had just proposed the following $PM_{2.5}$ control target at the beginning of 2016: the $PM_{2.5}$ concentration in 2020 should be decreased about 15% on the basis of 2015. However, $PM_{2.5}$ was a compound pollutant with complex chemical composition, including tens of primary and secondary components (Huang *et al.*, 2014). This brought difficulty and challenge to lower the $PM_{2.5}$ concentration. Consequently, it is of great importance to clarify the $PM_{2.5}$ pollution characteristic, in order to provide scientific support for improving the air quality effectively.

Beijing is a typical city of China for $PM_{2.5}$ pollution control, not only due to its high $PM_{2.5}$ concentration accompanied

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with rapid development of economic and urbanization process, but also because it had implemented many emission mitigation measures earlier than other provinces/cities. In order to mitigate the air pollution in Beijing, the government has implemented 16 phases' air pollution control measures to reduce source emissions since 2000 (http://www.bjepb.gov.cn/). Analyzing the effect of the implemented measures on the $PM_{2.5}$ pollution mitigation could provide scientific basis and control experiences for the following $PM_{2.5}$ control policy formulating both in Beijing and other areas of China. How effectively and how to assess the implementation effectiveness of these measures under the control of $PM_{2.5}$? Trend analysis was an effective and necessary approach. Therefore, it is of necessity to investigate the change trend of $PM_{2.5}$ and its chemical components during the past decade.

A number of studies have been conducted to investigate the $PM_{2.5}$ pollution characteristics of Beijing. For example, it was found that carbonaceous aerosols were significant contributors to the observed $PM_{2.5}$ mass in Beijing (He *et al.*, 2001; Ji *et al.*, 2016). SO_4^{2-}, NO_3^-, and NH_4^+ were the dominant ions in water soluble ions, accounting for more than 40% of the total $PM_{2.5}$ mass (Wang *et al.*, 2015b). Gao *et al.* (2014) analyzed 19 trace elements from April 2011 to January 2012 and illustrated that high levels of some trace elements (As, Mn and Cd) revealed the pollution of trace heavy metals in Beijing could not be neglected. In addition, Han *et al* (2016) reconstructed a time series of annual $PM_{2.5}$ from 1973 to 2013, concluded that $PM_{2.5}$ increased significantly with the growth of population and Gross Domestic Product (GDP). Lv *et al* (2016) studied seasonal tendency of $PM_{2.5}$ in winter and summer during 2000–2012, and illustrated that concentrations of $PM_{2.5}$ and SIA decreased in winter, and increased in summer, while carbonaceous components barely changed. From the result of Liu *et al* (2015b), it was shown that autumn was the main season that contributed to the annual mean $PM_{2.5}$ decrease during 2004–2012.

Although a lot of research has been conducted to analyze the $PM_{2.5}$ pollution in Beijing, a more comprehensive long-term trend study of $PM_{2.5}$ and its detailed components is still needed in order to illustrate the $PM_{2.5}$ evolution characteristic and the implementation effect of air pollution control during the past decade. As a result, an intensive $PM_{2.5}$ observation (2011–2015) and a comprehensive literature investigation were conducted in this study to collect valid $PM_{2.5}$ data as many as possible. Then the long-term trend (2000–2015) of $PM_{2.5}$ and its chemical composition in Beijing was analyzed, based on $PM_{2.5}$ precursors' concentration, sources emissions, air pollution control measures and meteorological factors. Tendency of detailed chemical components in $PM_{2.5}$, including OC, EC, secondary inorganic aerosol (SIA), typical elements (Si, Ca, Fe, Mn, Cu, Pb and K) and soil dust, were also investigated. The air pollution control effect during the past decade was also analyzed at the same time.

MATERIAL AND DATA

Sampling Site

Beijing is located on the northern edge of the North China Plain, surrounded by the Yanshan Mountains in the west, north and northeast. This kind of topography is disadvantageous for air pollutant dispersion. The $PM_{2.5}$ sampling was conducted in different seasons (i.e., spring, summer, autumn and winter) from 2011 to 2015 at the urban atmospheric environment monitoring superstation (39.96°N, 116.36°E) on the Science Building of Beijing Normal University (BNU). The height of sampling site was about 35m from ground. The sampling site was located in commercial and residential areas, and the surrounding traffic was intensive. As a result, it could well reflect the pollution characteristics of urban Beijing. The $PM_{2.5}$ samples were simultaneously collected on quartz fiber filters (Whatman Inc., Maidstone, UK) and Whatmans 41 filters (Whatman Inc., Maidstone, UK) using samplers with flow rate of 100 L min^{-1}, respectively. The sampling was on a 24-h basis. The quartz fiber and Whatmans 41 filters were used for the OC/EC and elements/ions analysis, respectively. The samples collected were put in the polyethylene plastic bags right after sampling and reserved in a refrigerator. All the filters were weighed before and after sampling by electronic balance (Sartorius TB-215D), with precision of 0.01mg, under constant temperature (20 ± 5°C) and relative humidity (40 ± 2%). One or two months per season were selected for $PM_{2.5}$ sampling. A total number of 646 samples were collected, including 144 in spring, 147 in summer, 167 in autumn and 188 in winter, respectively.

Measurement Method

The carbonaceous components (OC and EC) were determined by Optical Carbon Analyzer Model (DRL Model 2001A, Desert research institute, Reno). The precision and detection limits of our experiment were from the blank filter data. The detection limits were 0.82 μg C cm^{-3} for TOC and 0.20 μg C cm^{-3} for TEC, and the precision of the determination of concentration was below 5%. The backup filter was not used in the observation of this study, the same as most literatures referred (except He *et al.*, 2001; Yang *et al.*, 2005; Yang *et al.*, 2011a). The semi-volatile compound loss on the first filters may contribute to the uncertainty of measurement. Samples were tested by Improve programmed temperature rising method. Ions including NO_3^-, SO_4^{2-} and NH_4^+ were analyzed by ion chromatograph (IC, Metrohm 861 Advanced Compact IC). The relative standard deviations were less than 5% (Yang *et al*., 2016b). The limits of detection were less than 0.04 mg L^{-1} for anions and 0.006 mg L^{-1} for cations, respectively. Twenty-two elements (Na, Mg, Al, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Cd, Sb, Ce, Eu and Pb) were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, 7500a, Thermo). Closed vessel digestion method was used to pretreatment for the samples before typical elements analysis. The relative standard deviations were generally below 5%. In addition, all the procedures were strictly quality-controlled and avoid possible contamination of the samples.

Data Sources

In order to analyze the long-term trend of $PM_{2.5}$ and its chemical composition, a comprehensive investigation on

the published literatures related to $PM_{2.5}$ observation (2000–2014) of Beijing was conducted. This study focused on the $PM_{2.5}$ change trend of urban Beijing, and as a result, the selected monitoring sites were all set in the urban area (Fig. 1). In addition, the observation periods should reflect the long-term (one year) $PM_{2.5}$ characteristics of Beijing. Teflon, Whatman® 41 and quartz fiber filters were mainly used for collecting $PM_{2.5}$ samples. The filters were weighed under temperature around 20°C and relative humidity around 35–45%. Elements, inorganic ions and carbonaceous components were mainly analyzed by ICP-MS/ICP-AAS/XRF, Ion Chromatography and Thermal/Optical Carbon Analyzer, respectively. In order to reduce the uncertainty of data and improve the reliability of $PM_{2.5}$ change trend, data from literatures sampling at rural Beijing and in special periods were removed, because the purpose of this manuscript was to investigate the annual average $PM_{2.5}$ concentration change trend in the urban Beijing. Concentrations of other atmospheric pollutions (PM_{10}, SO_2 and NO_2) were mainly obtained from the published data of Beijing Municipal Environmental Protection Bureau (BMEPB, 2015). The annual pollutant emissions were collected from Beijing Municipal Environmental Protection Bureau (BMEPB, 2015) and Zhou (2012b). The meteorology parameters during 2000–2015 were obtained from China Meteorological Data Sharing Service System.

RESULES AND DISCUSION

$PM_{2.5}$ Concentration and Chemical Composition

The annual average $PM_{2.5}$ concentration from 2000 to 2015 in urban Beijing was shown in Fig. 2(a). It could be found that the annual average $PM_{2.5}$ concentration showed a generally decrease trend during the past decade, with an annual average change rate of –1.6 μg m^{-3} $year^{-1}$ (R = 0.53). During the same period, the annual mean PM_{10}, SO_2 and NO_2 concentrations decreased at a rate of 4.3 μg m^{-3} $year^{-1}$, 3.8 μg m^{-3} $year^{-1}$ and 1.7 μg m^{-3} $year^{-1}$, respectively (Fig. 3). The above air pollutants change tendency were mainly affected by the air pollution control measures implemented during the past decade. Since 2000, Beijing had implemented 16 phases' air pollution control measures, mainly including the controlling of industry, motor vehicle, coal combustion and fugitive dust pollution. The generally decrease trends of PM_{10}, $PM_{2.5}$, SO_2 and NO_2 indicated that the control measures implemented during the past decade had significant effects on the air quality improvement of Beijing. The occurrence of lower $PM_{2.5}$ concentration in 2008 (86.2 μg m^{-3}) also showed that strict measures taken before and during the Beijing Olympic Games played a prominent role in mitigating $PM_{2.5}$ pollution. The correlation between annual average $PM_{2.5}$ concentrations and $PM_{2.5}$ emissions was further analyzed, as shown in Fig. 2(b). The analysis results showed the change of annual $PM_{2.5}$ emission influenced the $PM_{2.5}$ concentrations to a great extent, the correlation coefficient between annual average $PM_{2.5}$ concentration and emission was 0.79. The meteorological conditions from 2000 to 2015 were also analyzed. A negative correlation between annual average $PM_{2.5}$ concentrations and wind speeds was found (R = 0.53) (Fig. 2(c)). And the frequency of southerly wind (transport large amount pollutants from southern Hebei to Beijing) and northerly wind (bring clean airs from the north region to Beijing) also have good correlation with $PM_{2.5}$ concentrations, with correlation coefficient of 0.62 and –0.61, respectively (Fig. S2). This indicated that wind was also an important factor affecting the $PM_{2.5}$ change tendency. While other meteorological conditions (e.g., temperature and relative humidity) didn't have obvious impact on $PM_{2.5}$ tendency from an inter-annual perspective (Fig. S2).

The proportion of main detailed chemical components in

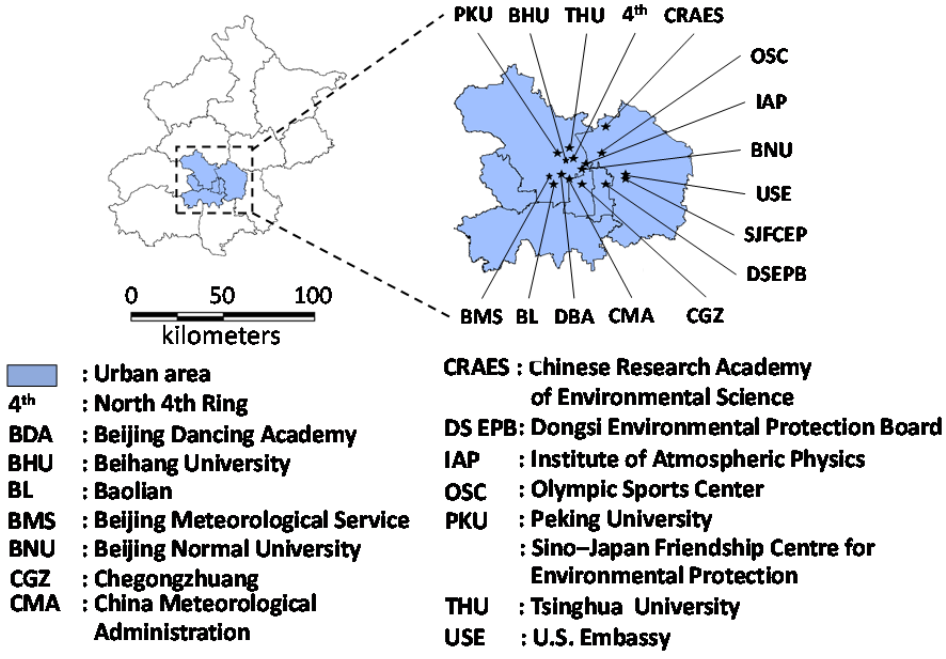

Fig. 1. Location of monitoring sites in this and referred literatures.

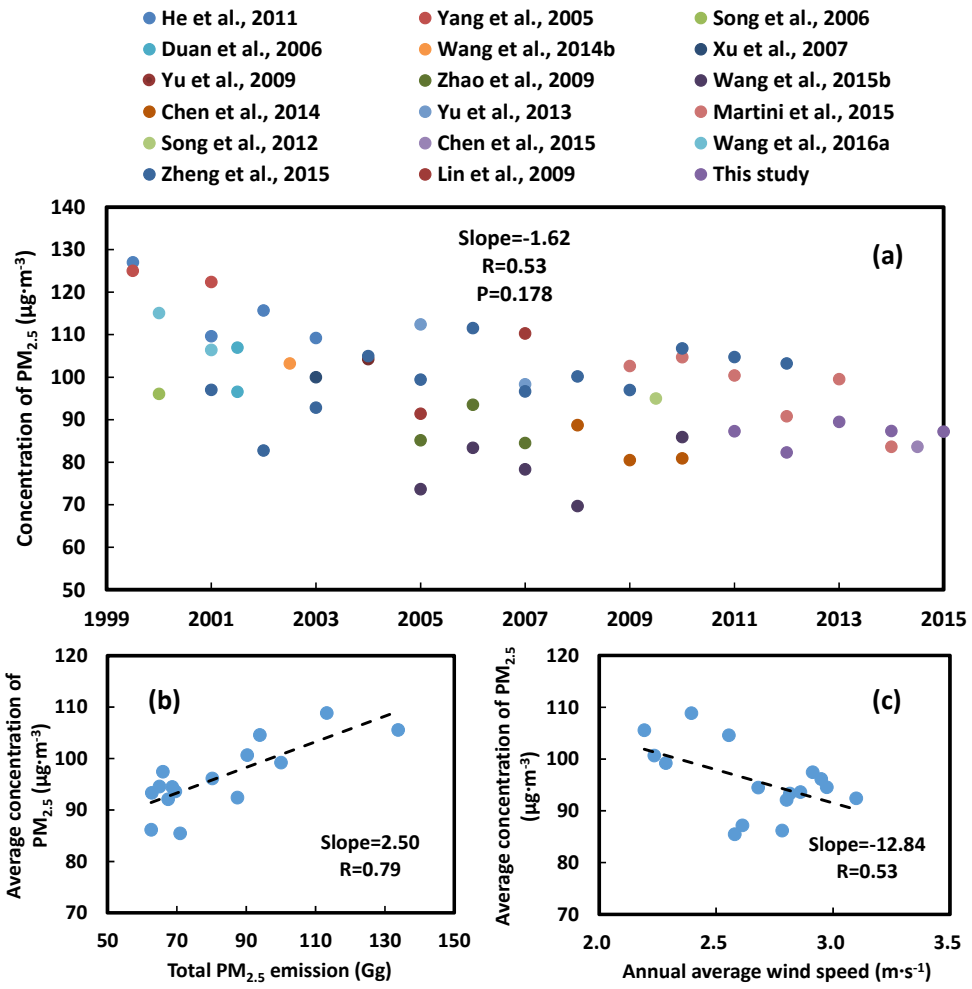

Fig. 2. Annual average $PM_{2.5}$ concentration changes from 2000 to 2015 (a) and correlation with total $PM_{2.5}$ emission (2000–2014) (b) and wind speed (2000–2015) (c).

$PM_{2.5}$ was calculated based on the average values obtained by observation and literature investigation, as shown in Fig. 4. It was illustrated that organic matter (OM) (OM = 1.6 × OC) (Xing *et al.*, 2013) contributed the first mass fraction to $PM_{2.5}$, but the percentage decreased generally from 2000 to 2015, the annual average change ratio (AACR) was –1.1%. The secondary largest contributor was SO_4^{2-} during 2000–2013 (9.4%–20.0%), while NO_3^- has become the second main component (17.2%–18.8%) since 2014. The proportion of soil dust decreased slightly from 12.2% in 2000 to 8.5% in 2015, with an AACR of –0.2% $year^{-1}$. It could also be found from Fig. 4 that the proportion of carbonaceous components (OM + EC) decreased from 2000 (42.6%) to 2015 (26.8%). Although there was secondary organic aerosol (SOA) in the OM, however, considering the slight upward trend of SOA in recent years (detailed analysis could be found in *Carbonaceous components* section), the change tendency of OM + EC indicated that the percentage of primary carbonaceous components (i.e., primary OM and EC) had a downward trend from 2000 to 2015. While at the same time, the percentage of secondary inorganic aerosol (SO_4^{2-}, NO_3^- and NH_4^+) increased at a rate of 0.7% $year^{-1}$. The major component of $PM_{2.5}$ had changed from carbonaceous component to SIA since 2009. It could be summarized that the proportion of primary components (primary OM, EC and soil dust) decreased but the percentage of secondary components (SIA) increased generally during 2000–2015.

Seasonal Variation

The seasonal variation of $PM_{2.5}$ concentration was also analyzed based on the average values of each season in every year, as shown in Fig. 5. The characteristic of seasonal average $PM_{2.5}$ concentration during 2000–2015 in Beijing was winter (114.1 μg m^{-3}) > autumn (93.4 μg m^{-3}) > spring (87.9 μg m^{-3}) > summer (87.0 μg m^{-3}). For the most polluted season, the average $PM_{2.5}$ concentration during 2000–2015 in winter showed four obvious change stages: 2000–2008, 2008–2010, 2010–2012 and 2013–2015. At the first stage (2000–2008), in order to ensure the air quality during the 2008 Green Olympic Games, Beijing implemented a series of strict control strategies, in which coal combustion was an important source to be controlled, such as replacing coal-fired heating with electric heating, forbidding raw coal bulk burning and implementing more stringent air pollutant emission standard for coal-burning boiler. These strict

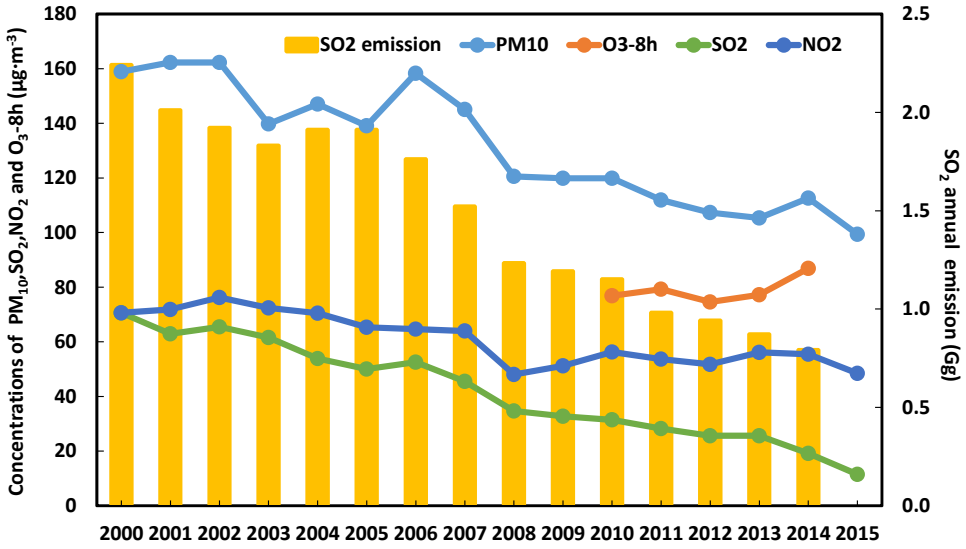

Fig. 3. Annual average concentration of other main pollutants in Beijing.

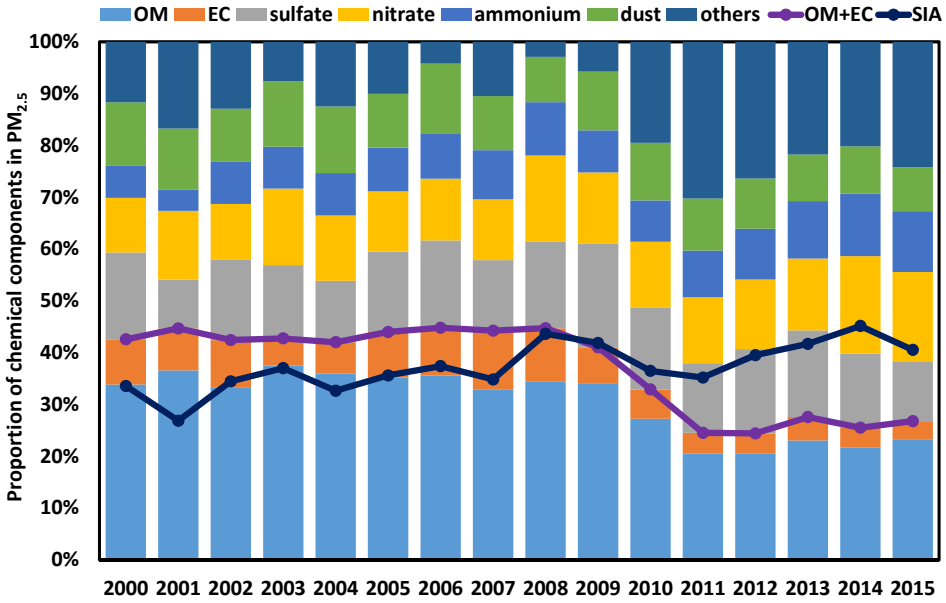

Fig. 4. The proportion of chemical components in $PM_{2.5}$ from 2000 to 2015.

control measures contributed significantly to the reduction of $PM_{2.5}$ concentration in winter (–6.8 μg m^{-3} $year^{-1}$). At the second stage (2008–2010), the concentration of $PM_{2.5}$ had an increment of 32.4% after the Olympic Games. To suppress this rising trend, new measures such as the substitution of clean energy in coal-fired boiler, reconstruction of low surface sources and no burning coal in the urban districts of Beijing were proposed. After implementing these measures, the $PM_{2.5}$ concentration decreased 21.7% from 2010 to 2013. However, because the average wind speed in winter decreased from 2.9 m s^{-1} in 2010–2012 to 2.6 m s^{-1} in 2013–2015, the $PM_{2.5}$ concentration was at a high level in the winter of 2013–2015, the average value was 114.2 μg m^{-3}. General downward trend was found in other three seasons. The annual average $PM_{2.5}$ concentration change rate was 1.0 μg m^{-3} $year^{-1}$, 1.6 μg m^{-3} $year^{-1}$ and 1.3 μg m^{-3} $year^{-1}$ for spring, summer and autumn, respectively.

Carbonaceous Components

The carbonaceous components concentrations during 2000–2015 were illustrated in Figs. 6(a) and Fig. 7(a). It could be found that the concentration of OC had almost no change from 2000 to 2006, with an average concentration around 23.0 μg m^{-3}. From 2006 to 2015, the annual average concentration of OC decreased obviously with a rate of 1.0 μg m^{-3} $year^{-1}$. OC came from two sources: primary OC emission and transformation from VOCs to OC. Motor vehicles were important contributor to VOCs and primary OC emission in the urban Beijing (Cheng *et al.*, 2013a; Zhang *et al.*, 2015a). As a result, the correlation between annual OC concentrations and vehicular OC/VOCs emissions was further analyzed. A good correlation was found between

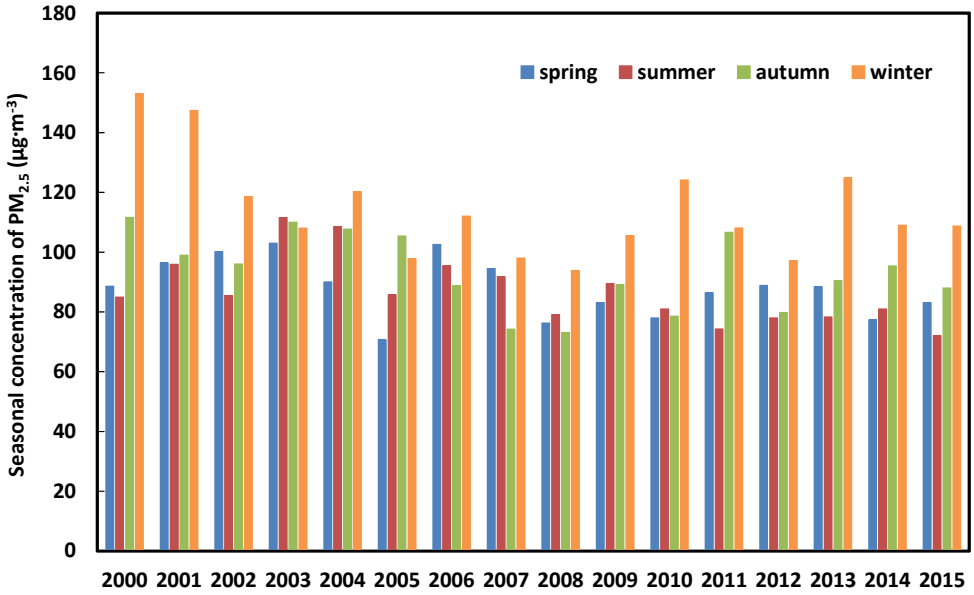

Fig. 5. Seasonal average $PM_{2.5}$ concentration changes from 2000 to 2015.

annual average OC concentrations with vehicular OC emissions (R = 0.95) and with vehicular VOCs emissions (R = 0.90) (Figs. 6(b)–6(c)). Some vehicular emission control measures, such as implementation of increasing strict vehicle emission standards (from state I to state V), improvement of fuel oil quality and development of public transportation, were gradually implemented since 1999. Those mitigated the vehicular OC and VOCs emissions significantly, and as a result reduced the atmospheric OC concentrations in recent years. As a major primary source of OC, coal combustion was severely controlled under a series of measures implemented in Beijing. The correlation coefficient between coal consumption and the annual average concentration of OC reached 0.90 during 2006–2014 (Fig. S1). The similar inter-annual variation as coal consumption showed these measures could also effectively reduce the atmospheric OC concentrations. Biomass burning was another important OC source, especially in autumn and winter (Ji *et al.*, 2016; Tao *et al.*, 2016). Significant correlation between OC and K (tracer for biomass burning, (Li *et al.*, 2014)) (R = 0.88) suggested the control of biomass burning also contributed to the OC concentration variation in urban Beijing (Fig. 6).

The annual average concentration of EC showed a fluctuant trend: it had almost no change before 2003, increased by 1.5 μg m^{-3} $year^{-1}$ from 2003 to 2007, and decreased by 1.0 μg m^{-3} $year^{-1}$ after 2007 (Fig. 7(a)). Incomplete combustion of fossil fuel and biomass combustion were important EC emission sources (Zhang *et al.*, 2015b). As shown in Fig. 7(c), EC had mitigatory correlation (R = 0.64) with the consumption of main fossil fuels (coal and oil) in Beijing, and a downtrend of EC could be found obviously following the replacement of fossil fuel with clean energy (e.g., natural gas and electric) since 2007. The source apportionment result obtained from positive matrix factorization (PMF) analysis showed that about 50% of the OC and EC in urban Beijing were associated with biomass burning processes and the major source of biomass burning aerosol in Beijing was suggested to be the combustion of crop residuals (Cheng *et al.*, 2013b). There were three phases' policies to control crop residuals burning since 1999 (Fig. 7(b)). (1) Straw burning was prohibited in limited areas (e.g., airport, arterial traffic, the people's government and densely populated area) during 1999–2007 (P1). During this period, the straw burning had not been strictly limited and high correlation was found between annual EC concentration and crop yield (R=0.84), these indicated biomass burning was an important contributor source to the atmospheric EC. (2) Comprehensive prohibition in central city and serious straw burning province since 2008 (P2). During this period, although the crop yield was much higher than P1, the EC concentration decreased by 49% due to the strict control of biomass combustion. (3) Putting forward the policy support and incentive mechanism to promote straw comprehensive utilization (e.g., used as fertilizer, feed and industrial raw materials) since 2013 (P3). The encouragement policy in this period further reduced EC concentration of 1.3 μg m^{-3} from 2013 to 2015 on the basis of strict control of straw burning. Further analysis found that the annual EC and K concentrations had almost the same change tendency (R = 0.94) in 2009–2015 (Fig. 7(c)), this indicated open biomass burning control in P2 and P3 was an important factor resulting in the EC's change from another perspective.

The value of OC/EC could indicate the source of carbonaceous aerosols, with high value (> 2) meaning the formation of secondary organic aerosol (SOA) (Tian *et al.*, 2015). The OC/EC ratio ranged from 1.80 to 4.47 in this study, indicating the existence of SOA pollution in Beijing. The continuous rising of OC/EC from 2.07 in 2008 to 4.16 in 2015 illustrated the secondary carbonaceous components pollution is becoming serious. The secondary organic carbon (SOC) in 2011–2015 was further estimated using the EC tracer method (Appendix 1). The ratio of SOC/OC showed a generally continuous growth from 2011 (29%) to 2015 (35%) (Fig. 6(a)), supporting the conclusion above.

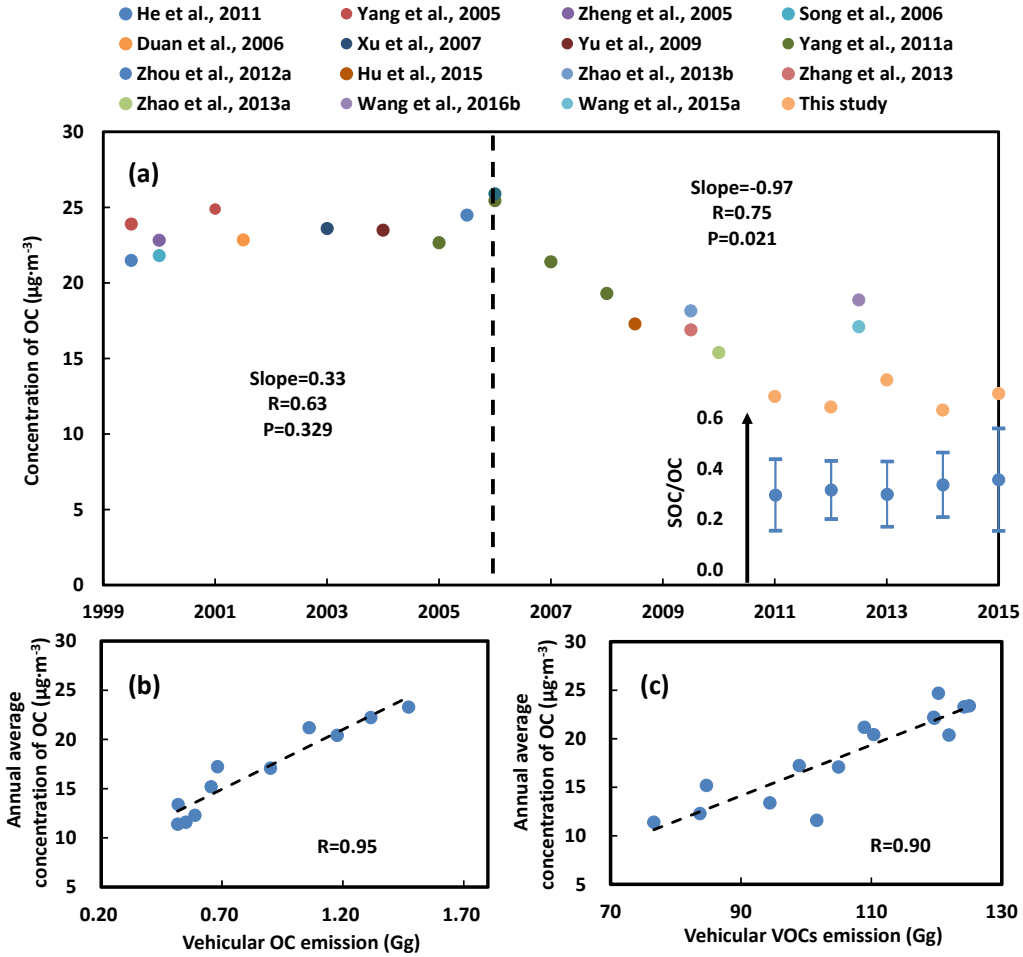

Fig. 6. Annual average OC concentration changes from 2000 to 2015 (a) and correlation with vehicular OC (2004–2014) (b) and VOCs emission (2000–2014) (c).

Secondary Inorganic Aerosol (SIA) Components

Secondary inorganic aerosol (SIA) of $PM_{2.5}$ refereed to the sum of SO_4^{2-}, NO_3^- and NH_4^+. Although the $PM_{2.5}$ concentration had a decrease from 2000 to 2015, the concentration of SIA had a slightly growth at a rate of 0.2 μg m^{-3} $year^{-1}$. From 2000 to 2013, the annual average concentration of SO_4^{2-} decreased by 0.1 μg m^{-3} $year^{-1}$; during 2013 to 2015, SO_4^{2-} showed an obvious decrease from 15.8 μg m^{-3} to 10.1 μg m^{-3} (Fig. 8(a)). The annual average concentrations of NO_3^- and NH_4^+ both presented a general upward trend, fluctuated before 2010, but increased steadily at a rate of 0.8 μg m^{-3} $year^{-1}$ and 0.6 μg m^{-3} $year^{-1}$ after 2010, respectively (Figs. 8(b)–(c)). Sulfate and nitrate were mainly from the oxidation of SO_2 and NO_x. As shown in Figs. 3 and Fig. 8(a), an interesting phenomenon could be found: after more than ten years' mitigation, the emission and concentration of SO_2 had decreased significantly at rates of 10.6 Gg $year^{-1}$ (NBSC, 2001–2015) and 3.6 μg m^{-3} $year^{-1}$, respectively, however, the SO_4^{2-} had no obvious change. Similar phenomenon could also be found from Figs. 3 and Fig. 8(b): the concentration of NO_2 decreased at a rate of 2.4 μg m^{-3} $year^{-1}$ in 2000–2008, and had an fluctuate with an average of 52.6 μg m^{-3} in 2008–2015, but the NO_3^- increased from 11.1 μg m^{-3} in 2000 to 15.0 μg m^{-3} in 2015. The formation of SIA was an oxidation process. O_3 was an important antioxidants, its rising could enhance the atmospheric oxidation and facilitate the precursors (e.g., SO_2 and NO_x) to transform to secondary particles (e.g., SO_4^{2-} and NO_3^-). Tang *et al.* (2009) collected O_3 data at six sites in downtown Beijing and found an increasing tendency at a rate of 1.1 ± 0.5 ppbv yr^{-1} during 2001–2006. Wang *et al.* (2012) also found the photochemistry reaction within the troposphere significantly contributed to an increase trend of tropospheric ozone from 2002 to 2010 in Beijing. The average concentration of O_3-8h in urban Beijing was also collected and found an increase trend at a rate of 1.8 μg m^{-3} $year^{-1}$ during 2010–2014 (Fig. 3) (http://www.bjmemc.com.cn/). The keep growth of O_3 concentration might be the factor that contributed to the increasing trend of NO_3^- and NH_4^+. The slight decrease of SO_4^{2-} may be attributed to both the enhancement of the atmospheric oxidation and the obvious reduction of SO_2 emission. Thus special attention should be paid to the atmospheric oxidation in order to mitigate the $PM_{2.5}$ pollution effectively. The sulfur oxidation rate (SOR) and nitrogen oxidation ratio (NOR) were further calculated to assess the degree of sulfate and nitrate formation. SOR and NOR were estimated by the annual average concentrations of sulfate, nitrate and

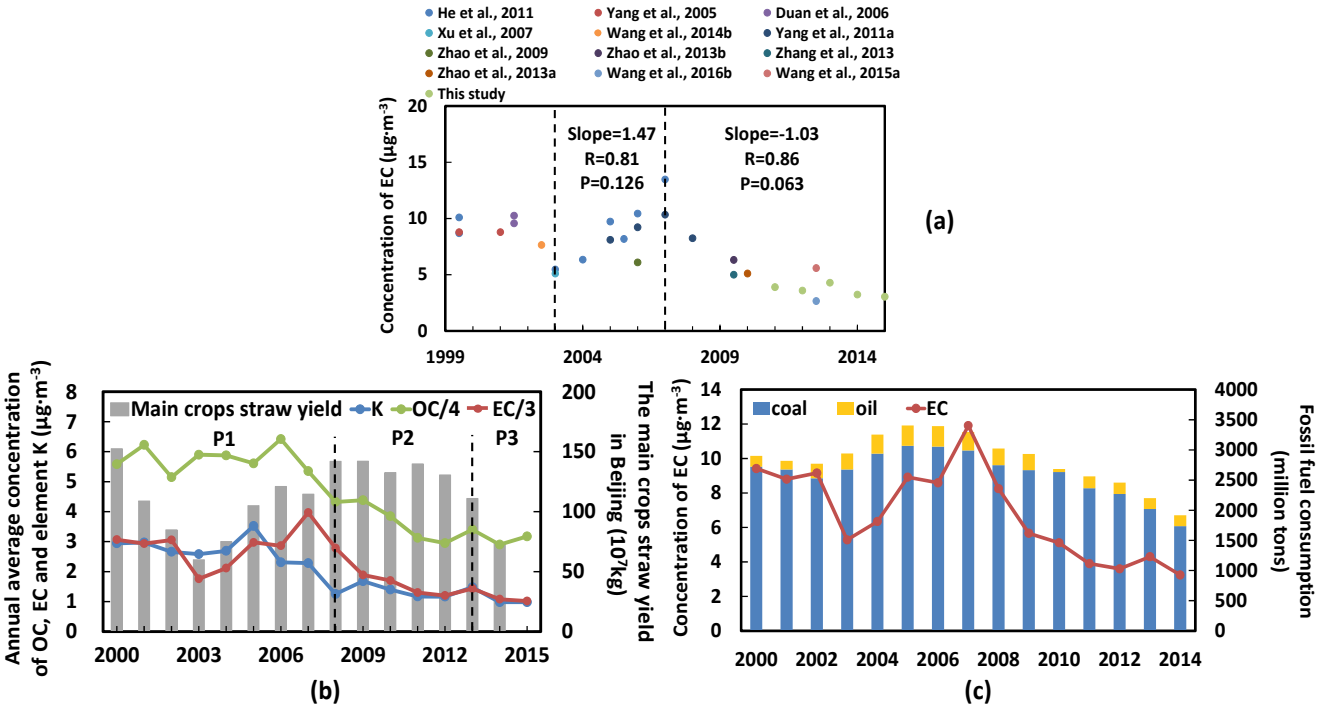

Fig. 7. Annual average EC concentration changes (a), correlation between OC, EC with K and the main corps straw yield (b) and correlation between EC with fossil fuel consumption.

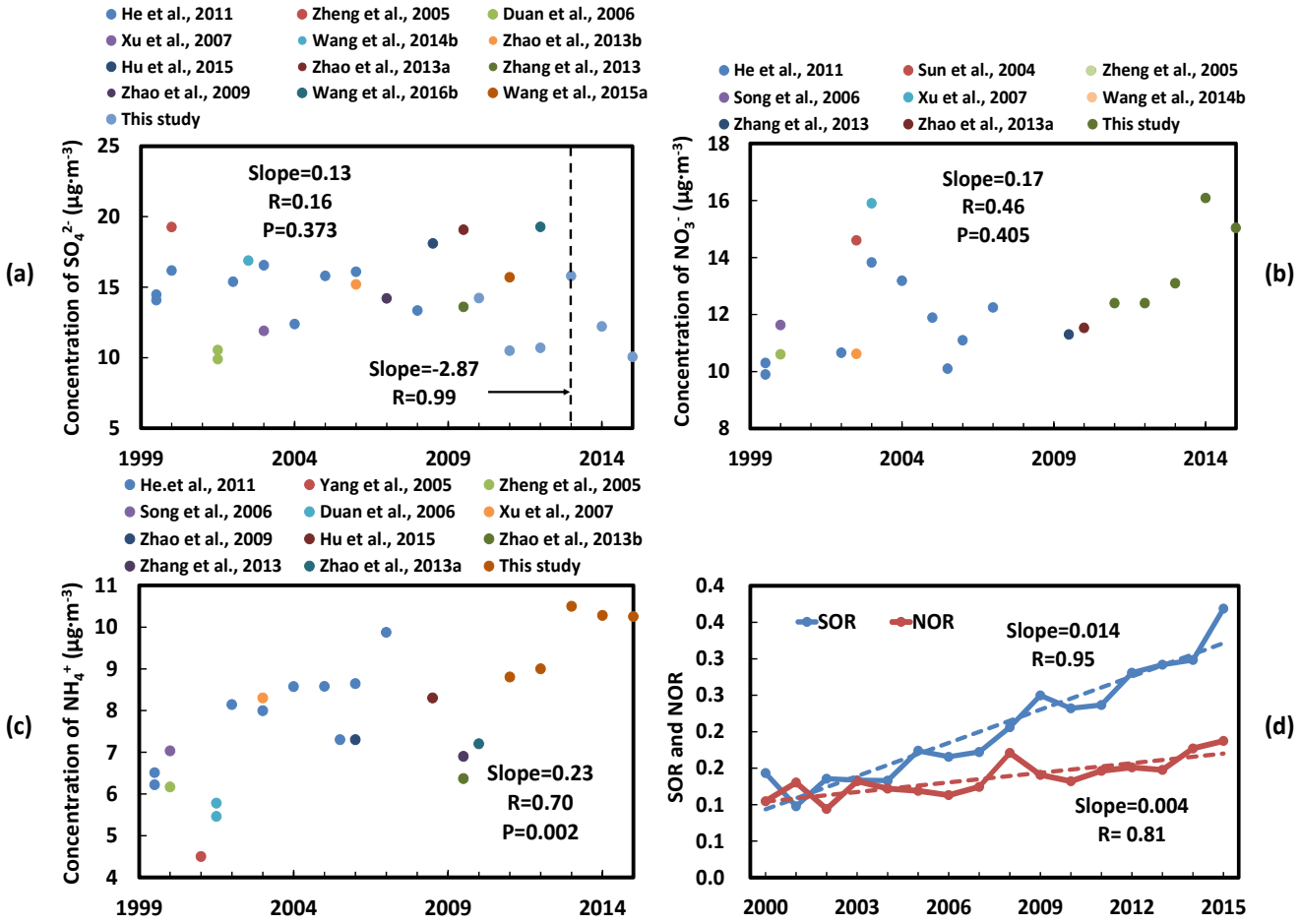

Fig. 8. Concentration of SO_4^{2-} (a), NO_3^- (b), NH_4^+ (c) and conversion rate of sulfate and nitrate (d).

their precursors (SO_2, NO_2) following formula (Liu *et al.*, 2015a):

$$SOR = nSO_4^{2-} / (nSO_4^{2-} + nSO_2) \quad (1)$$

$$NOR = nNO_3^{-} / (nNO_3^{-} + nNO_2) \text{ (n refers to the molar concentration)} \quad (2)$$

The SOR and NOR generally showed an upward trend during 2000–2015, with increase rates of 0.014 $year^{-1}$ and 0.004 $year^{-1}$, respectively (Fig. 8(d)). The enhancement of atmospheric oxidation could facilitate the transformation of precursors (SO_2 and NO_x) to secondary particles. The upward trend of SO_4^{2-} and NO_3^{-} in Beijing might also be contributed from external transport. However, the $PM_{2.5}$ concentration in surrounding areas of Beijing (e.g., Hebei and Tianjin) also had been reduced in recent years (MEPC, 2014). In addition, SOR was suggested to be closely correlated with RH in a short period (Sun *et al.*, 2014; Hu *et al.*, 2016), while a weak correlation was found (R = 0.017) between SOR and RH from an inter-annual perspective (Fig. S3). As a result, enhancement of atmospheric oxidation might be the major factor affecting the SIA changes in Beijing. Considering the deceasing trend of primary $PM_{2.5}$ components (e.g., OM and EC) and the increasing trend of total $PM_{2.5}$ in recent years, control of SIA pollution becomes a crucial problem in order to reduce the $PM_{2.5}$ concentration effectively.

Soil dust and Typical Elements

Typical elements in $PM_{2.5}$ could represent the variation of different pollution sources to some extent. Change tendency of seven typical elements' annual average concentrations were shown in (Fig. 9(a)). In addition, the concentration of soil dust in $PM_{2.5}$ was also calculated by the following formula (Wang *et al.*, 2015a):

$$C_{soil\ dust} = 2.20C_{Al} + 2.49C_{Si} + 1.63C_{Ca} + 2.42C_{Fe} + 1.94C_{Ti} \quad (3)$$

The average concentrations of these typical elements and soil dust all showed a downward trend generally during the past decade. Silicon (Si) and calcium (Ca) could be identified as road and construction dust (Thurston *et al.*, 2011). Concentrations of soil dust, Si and Ca decreased from 2000 to 2008 and increased slightly in 2008–2012. This was consistent with the trend of road dust emission (Fan *et al.*, 2013). The growth (20% $year^{-1}$) of construction areas during 2008 to 2012 was the reason led to the rise of Si, Ca and soil dust (Fig. 9(b)). However, although the construction area was still high in 2013–2014 (BSIN, 2015), the implementation of strict emission control measures, such as the building material pile and the rare land must be covered and transport truck should be cleaned by water before leaving the construction site, reduced the construction emissions and lowered concentrations of Si, Ca and soil dust. Ferrum (Fe), manganese (Mn) and cuprum (Cu) had always been found together from minerals of industrial productions and processes, and as a result could characterize industry emissions (Thurston *et al.*, 2011). The decline of these three elements was benefit from the strict controlling of industry emission control measures in Beijing. The increasingly stringent restriction on industrials' emission concentration, moving out or closing large numbers of heavy polluting industries such as chemical industry, steel industry and coal-fired power plants, mitigated industrial emissions to a great extent (BMEPB, 2015) (http://www.bjepb.gov.cn/). Plumbum (Pb) could be recommended as marker of motor vehicle emissions (Chen *et al.*, 2014). As shown in Fig. 9(c), the concentration of Pb had a good relationship (R = 0.77) with vehicular $PM_{2.5}$ emission in 2000–2014 (Lang *et al.*, 2014). The downward trend of Pb (0.014 μg m^{-3} $year^{-1}$) indicated the motor vehicle control measures implemented in Beijing, such as continuous improvement of motor vehicle emission standards, elimination of high emission vehicles and limiting foreign vehicles with high emissions into Beijing, were effective for primary particle mitigation. Kalium (K) could represent biomass emissions (Li *et al.*, 2014). It was mainly emitted from the straw burning. The straw production was related to grain yield. It could be found from Fig. 7 (b) that, before 2008, the annual K concentration had similar change tendency with grain yield. However, although the grain yield in Beijing still had a high level from 2008–2014 (BSIN, 2009–2015), the average K concentration of decreased from 2.7 μg m^{-3} in 2000–2007 to 1.3 μg m^{-3} in 2008–2014. Prohibition of open straw burning and promotion of straws comprehensive utilization (e.g., straw returning to soil) in Beijing and its surrounding areas should be the main reason reducing the K concentration in recent years.

Comparison with Other Studies

A comparison analysis between this and previous studies was further conducted, as shown in Fig. 10. Before the year of 2007, the tendency of this study was similar with the other four studies. During 2007–2008, the decrease trend of this study was different from Chen *et al.* (2016) and Zheng *et al.* (2015). The uptrend during 2007–2008 appeared in these two studies were abnormal because strict emission control measures were implemented before 2008 in order to achieve the air quality target of Green-Olympic. From 2008 to 2009, the rise of the $PM_{2.5}$ concentration during 2008–2009 in this study was different from the other four literatures. The concentration (102.6 μg m^{-3}) referred from Martini *et al.* (2015) lead to the high value in 2009 of this study. Martini *et al.* (2015) monitored $PM_{2.5}$ concentration by energy decay of beta rays method. The $PM_{2.5}$ concentration obtained by this method was usually at least 15% higher than the concentration collected by gravimetric method (Zheng *et al.*, 2015). As for the change trend in 2011–2012, different trends were found in this study. As shown in Fig. 2(a), the data from Zheng *et al.* (2012) (103.2 μg m^{-3}, nearly 20 μg m^{-3} higher than the results in other studies) increased the average concentration during 2011–2012. The $PM_{2.5}$ trend of this study exhibited similar inter-annual variation as Lv *et al.* (2016) during 2004–2012. This may because Lv *et al.* (2016) contained some same reference (Duan *et al.*, 2006; Zhao *et al.*, 2009; Yu *et al.*, 2013; Chen *et al.*, 2014) with this study.

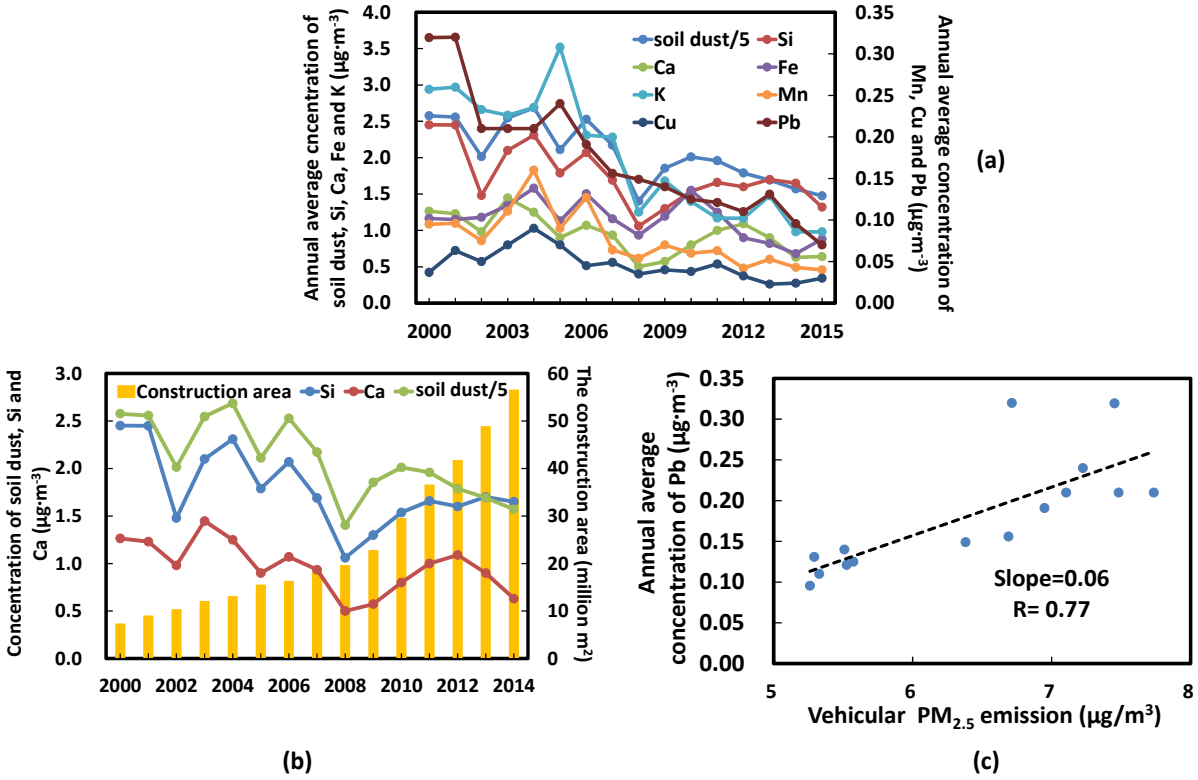

Fig. 9. Annual concentration changes of soil dust and typical elements in $PM_{2.5}$ (a) and comparison with construction area (b) and vehicular $PM_{2.5}$ emission (c).

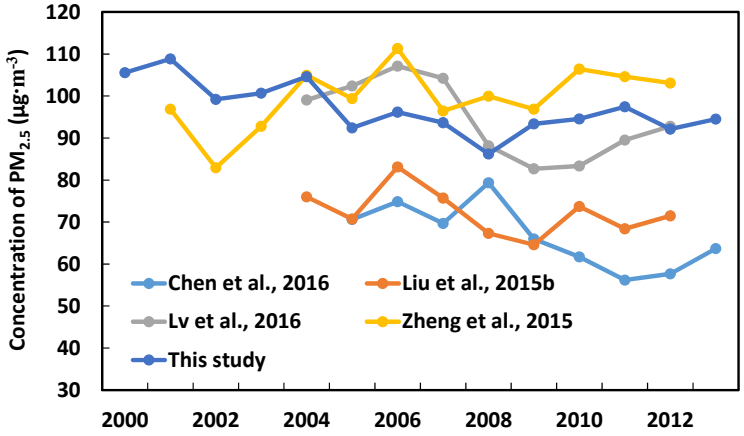

Fig. 10. The comparison of annual average $PM_{2.5}$ concentrations between this and previous studies.

As for the value of $PM_{2.5}$ concentration, different measurement methods and sampling sites would produce different results. The annual average $PM_{2.5}$ concentrations in this paper were generally higher than Liu *et al.* (2015b) and Chen *et al.* (2016)'s study, but lower than Zheng *et al.* (2015)'s work. As for Zheng *et al.* (2015), the concentration of $PM_{2.5}$ was relatively higher than other studies because they used beta rays method to monitor $PM_{2.5}$ concentrations. Liu *et al.* (2015b) measured $PM_{2.5}$ by tapered element oscillating microbalances (TEOM). The ambient temperatures (50°C) led to the loss of semi volatile compounds and up to 25% lower mass concentration was found as compared with gravimetric filter measurements (Liu *et al.*, 2015b). Chen *et al.* (2016) located the sampling site in the campus of the China University of Geosciences (CUG). GCU was surrounded by many universities but very few emission sources, the relatively clean environment made the monitoring concentration lower than other studies.

CONCLUSIONS

$PM_{2.5}$ is the major air pollutant in most areas of China. The $PM_{2.5}$ pollution in the capital – Beijing is typical and has attracted wide concerns from scientists, government, media and the public. Long-term trend analysis of $PM_{2.5}$ could help to assess the effect of historical emission control

measures and provide experience and scientific support for the following mitigation strategy making in Beijing and other areas of China. Based on intensive observation from 2011 to 2015 and comprehensive investigation of $PM_{2.5}$ and its chemical components, the long-term trend of $PM_{2.5}$ and its chemical composition in Beijing was analyzed connected with integrated driving factors.

1. The annual average $PM_{2.5}$ concentration in Beijing generally decreased from 2000 to 2015 with an annual average change rate of –1.5 μg m^{-3} $year^{-1}$. The proportion of carbonaceous components decreased from 2000 to 2015, while the percentage of secondary inorganic aerosol (SIA) increased and became the major component of $PM_{2.5}$ since 2009. $PM_{2.5}$ change trend indicated the implementation of emission control measures had significant effects on the air quality improvement of Beijing, while special attention should also be paid to the atmospheric oxidation in order to further mitigate the $PM_{2.5}$ pollution.
2. The characteristic of mean $PM_{2.5}$ concentration during 2000–2015 in Beijing was winter > autumn > spring > summer. The average $PM_{2.5}$ concentration in winter showed four obvious change stages in winter. Remarkable decrease of $PM_{2.5}$ concentration in winter indicated the coal combustion control measures obtained significant effect on $PM_{2.5}$ pollution mitigation, while adverse weather conditions could also cause serious pollution. The average $PM_{2.5}$ of other three seasons showed a general downward trend.
3. The concentration of OC has almost no change from 2000 to 2006, but decreased from 2007 to 2015. Coal combustion and motor vehicle emission control measures together contributed to the reduction of OC. The concentration of EC showed a fluctuate trend: almost no change before 2003, increased from 2003 to 2007, and decreased after 2007. This may be caused by the replacement of fossil fuel with clean energy and strict control of biomass combustion. The continuous rising of OC/EC and SOC/OC from 2011 to 2015 illustrated the secondary carbonaceous pollution is becoming serious.
4. The concentration of SO_4^{2-} increased slightly from 2000 to 2013, but decreased obviously from 2013 to 2015. The concentrations of NO_3^- and NH_4^+ both presented a general upward trend under the decrease of source emission and atmospheric concentration of precursors (SO_2 and NO_2). The generally upward trend of SOR and NOR illustrated the enhancement of the atmospheric oxidation contributed to the change trend of SIA. Control of SIA pollution became a crucial problem in reducing the $PM_{2.5}$ concentration effectively.
5. Soil dust and typical elements in $PM_{2.5}$ could represent the variation of different pollution sources to some extent. The concentrations of typical elements (Si, Ca, Fe, Mn, Cu and K) and soil dust decreased generally during the past decade. This reflected the mitigation measures on the reduction of source emissions (building construction, industry, motor vehicle and biomass burning) were effective for mitigating primary $PM_{2.5}$ pollution in Beijing.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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