Air pollution in mega cities in China—A review

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Review

Air pollution in mega cities in China

Chak K. Chan[a,*], Xiaohong Yao[a,b]

[a]*Department of Chemical Engineering, Hong Kong University of Science and Technology, Clear Water Bay, Hong Kong, China*
[b]*Key Laboratory of Marine Environmental Science and Ecology, Ministry of Education, Ocean University of China, 238 Songling Road, Qingdao 266100, PR China*

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Abstract

Due to its rapidly expanding economic and industrial developments, China is currently considered to be the engine of the world's economic growth. China's economic growth has been accompanied by an expansion of the urban area population and the emergence of a number of mega cities since the 1990. This expansion has resulted in tremendous increases in energy consumption, emissions of air pollutants and the number of poor air quality days in mega cities and their immediate vicinities. Air pollution has become one of the top environmental concerns in China. Currently, Beijing, Shanghai, and the Pearl River Delta region including Guangzhou, Shenzhen and Hong Kong, and their immediate vicinities are the most economically vibrant regions in China. They accounted for about 20% of the total GDP in China in 2005. These are also areas where many air pollution studies have been conducted, especially over the last 6 years. Based on these previous studies, this review presents the current state of understanding of the air pollution problems in China's mega cities and identifies the immediate challenges to understanding and controlling air pollution in these densely populated areas.

Keywords: Air quality; O_3; $PM_{2.5}$; PM_{10}; Dust storm; Beijing; Shanghai; Hong Kong; Guangzhou

Contents

*Corresponding author. Tel.: +852 2358 7124; fax: +852 2358 0054.
E-mail address: keckchan@ust.hk (C.K. Chan).

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1. Introduction

China has undergone very rapid economic growth since the economic reforms began in 1978. In the last three decades, the annual growth rate of the gross domestic product (GDP) in China was about 10% (China Statistical Yearbook 2005, 2006). The GDP in 2005 reached 18,000 billion *RMB yuan*, equivalent to 2200 billion US dollars. China is now considered to be the engine of world's economic growth. China's economic growth has resulted in an increase in energy consumption, air pollution and associated health effects (HEI International Scientific Oversight Committee, 2004). In 2005, the total energy consumption was equivalent to 2.2 billion tons of coal, which was about three times more than that in 1978 (China Energy Statistical Yearbook 2005, 2006). Coal accounts for more than 70% of the total energy consumption, and emissions from coal combustion are the major anthropogenic contributors to air pollution in China. The emission of SO_2, a major air pollutant from coal combustion, was 2.55×10^7 tons in 2005. SO_2 concentrations exceeded the Chinese Grade-II standards in 22% of the country's cities and caused acid rain problems in 38% of the cities (Report of the State of the Environment in China, 2005). Since 2002, a new economic boom led by the real-estate sector in cities in China has revived heavy industry, particularly for the production of construction

materials, such as steel, cement, aluminum, etc. A decreasing trend of the SO_2 emissions ended in 2002 in China. SO_2 emissions in 2005 exceeded that in 1997 by 12%.

With an increasing number of air pollution episodes and low visibility days reported by the media, much attention has been paid to reducing air pollutant emissions and to improving air quality across cities, municipalities, and provinces in China. For example, a series of laws, regulations, and standards such as the Law of the People's Republic of China on the Prevention and Control of Atmospheric Pollution, the National Ambient Air Quality Standards (GB3095-1996) and the Emission Standards of Air Pollutants for Thermal Power Plants (GB13223-2003) have been formulated and promulgated. Since 2000, a daily air pollution index (API) and air quality levels are available in many cities. The percentage of cities with concentrations of air pollutants below the Chinese Grade-II standards increased from 33.1% in 1999 to 51.9% in 2005, while the percentage of cities with pollutant concentrations exceeding the Chinese Grade-III standards decreased from 40.5% in 1999 to 10.6% in 2005 (Report of the State of the Environment in China, 2005). Air quality in most Chinese cities has improved despite the rapid growth of the economy. However, particulate concentrations such as $PM_{2.5}$ (PM—particulate matter) in most Chinese cities are still far above the World Health Organization Air Quality Guidelines of $10\,\mu g\,m^{-3}$ (annual average) and $25\,\mu g\,m^{-3}$ (24 h average) (WHO, 2005). For example, He et al. (2001) and Ye et al. (2003) reported that $PM_{2.5}$ concentrations in Beijing and Shanghai, the two largest cities in China, were about 10 times and six times the WHO guideline values, respectively. Table 1 summarizes the National Ambient Air Quality Standards (GB3095-1996 in China) and the WHO Air Quality Guidelines (WHO, 2005).

1.1. Urbanization and mega cities in China

Since 1978 in China, increasing numbers of workers have migrated from the countryside to the urban areas, particularly to large and developed cities for better job opportunities. From 1980 to 2005, the urban population in China increased from 19.6 to 40.5%. The number of cities increased to over 660, and more than 170 cities had over 1 million permanent residents (not including the migrant population) in 2004. These metropolises generated 65.5% of the GDP, but at the expense of the environment (China Statistical Yearbook 2005, 2006). Mega cities, which are conventionally defined as cities with populations over 10,000,000, emerged in the 1990s in China and city clusters were developed in the proximity of the mega cities during the last decade (Urban Statistical Yearbook of China 2005, 2006). This urbanization trend is likely to continue in the future (Liu and Diamond, 2005; Shao et al., 2006).

Table 1
Concentration limits for some pollutants in the Chinese National Ambient Air Quality Standards (CNAAQS) and the World Health Organization Air Quality Guidelines ($mg\,m^{-3}$)

Pollutants	Averaging time	CNAAQS			WHOAQG guidelines
		Grade-I	Grade-II	Grade-III	
SO_2	Daily	0.05	0.15	0.25	/[a]
	Annual	0.02	0.06	0.1	0.02
NO_2	Daily	0.08	0.08	0.12	/
	Annual	0.04	0.04	0.08	0.04
CO	Daily	4	4	6	/
O_3	Hourly	0.12	0.16	0.2	0.1[b]
PM_{10}	Daily	0.05	0.15	0.25	0.05
	Annual	0.04	0.1	0.15	0.02
$PM_{2.5}$	Daily	/	/	/	0.025
	Annual	/	/	/	0.01

[a]Not available.
[b]8-h average.

When considering both permanent residents and migrant residents, many cities in China, such as Beijing, Shanghai, Guangzhou, Shenzhen, Xi'an, Shenyang, Chongqing, Nanjing, Wuhan, and Tianjin are mega cities. Their locations are shown in Fig. 1. These mega cities are characterized by various economic modes due to geographical, historical, and political factors. In this review, we focus on Beijing, Shanghai and the Pearl River Delta (PRD) region, which includes cities like Hong Kong, Guangzhou and Shenzhen. Beijing is the capital and the center of politics, economics and culture in China. Heavy industries are gradually being replaced by other less polluting industries such as financial and other service industries, and high-technology manufacturing (Beijing Statistical Yearbook 1997–2005, 1998–2006). Shanghai is the economic center of eastern China and its industrial structure includes heavy industries, such as steel, petrol, and automobile production, along with export processing and financial and other service industries (Shanghai Statistical Yearbook 1997–2005, 1998–2006). The PRD region, including Guangzhou, Shenzhen and Hong Kong, is characterized by export processing industries and financial and other service industries (Guangdong Statistical Yearbook 2005, 2006). The mega cities of Beijing, Shanghai and the PRD region, and their vicinity city clusters accounted for about 20% of the total GDP in China in 2005. Air quality management in these mega cities has tremendous implications on the future development of other Chinese mega cities.

Air pollution problems in mega cities and their immediate vicinities will continue to be one of the top environmental concerns in the next decade in China. In Beijing, Shanghai and PRD, networks of the air quality monitoring stations have been well established during the last two decades. Hundreds of research reports on air pollution have been published and the numbers of such reports are growing rapidly, e.g., the growth of publications on atmospheric aerosols is shown in Fig. 2. It is timely to conduct a critical review on the current state of

Fig. 1. Map of China and the location of major mega cities (Inset–map of the Pearl River Delta region).

understanding of air pollution problems in these regions. There are some studies on air pollution in other mega cities in China but the number of studies on each city is rather small compared to those of the three mega cities we focus in this paper. Thus, this review focuses on Beijing, Shanghai, PRD and their immediate vicinities. Since the PRD is a well-linked region consisting of a few major cities because of urbanization and efficient transportation, we review air pollution in the PRD as a whole.

1.2. Data collection and review objectives

While this review mainly focuses on publications about air pollution in China in international peer-reviewed journals, environmental reports by the Chinese central and local governments (often in Chinese) are also used to examine the long-term trends of air pollution. In most of these reports, except those from Hong Kong, the concentrations of air pollutants are not reported. Instead, only the numbers of hours or days of air pollutant concentrations exceeding the National Ambient Air Quality Standards (GB3095-1996 in China) are available. It should be noted that real-time data from air quality monitoring stations established by local governments are not available to public. Although hundreds of research papers on air pollution in different mega cities are available in journals written in Chinese, we focus on internationally peer-reviewed publications in this review. Most important results and conclusions reported in these research papers in Chinese have also been published in internationally peer-reviewed journals.

The focus of research papers on air quality in Beijing, Shanghai, and the PRD often varies, reflecting the different nature or emphasis of air pollution problems in these locations. For example, in Beijing, PM_{10} is usually the major air pollutant reported and improving visibility is one of the mandates of the local government. The amount of studies on atmospheric particles, including total suspended particles (TSPs), PM_{10}, $PM_{2.5}$ and ultrafine particles (<100 nm) overshadows the number of studies on gaseous pollutants such as NO_2 and O_3. In Hong Kong, publications on both particulates and gaseous pollutants are equally abundant. On the other hand, in Guangzhou, most studies focus on gas-phase volatile organic species and particulate organic species. The availability of the findings determines the focus of this review in different mega cities.

A number of reviews on urban air pollution in China or in various cities before 2003 are available (He et al., 2002; Tang, 2004; Molina and Molina, 2004; Shao et al., 2006). Since 2003, there has been explosive growth on the number of publications on air pollution in mega cities in China (Fig. 2) and a new scenario of air pollution is emerging due to the over consumption of coal. This review builds upon the earlier reviews and includes new knowledge and insights acquired in more recent publications as well

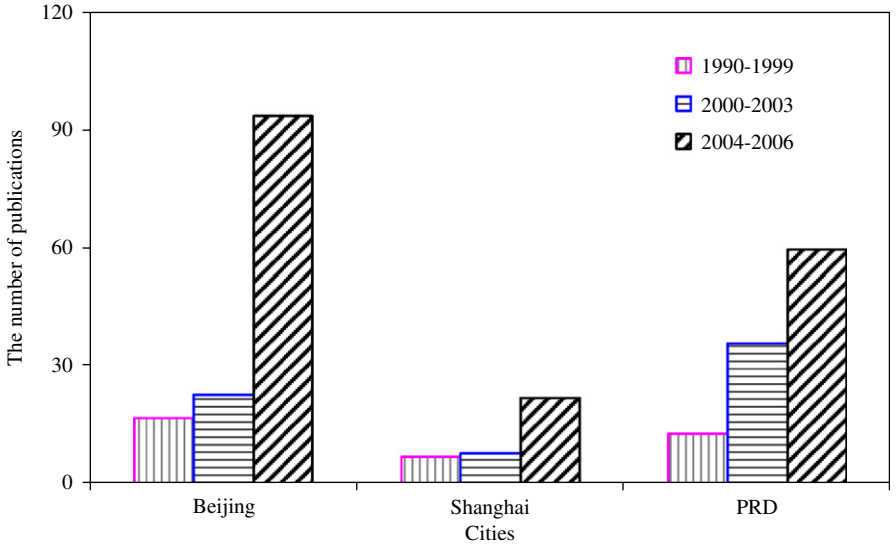

Fig. 2. Rapid growth in the number of publications on atmospheric aerosols only in Beijing, Shanghai and the PRD. Data are from the Science Citation Index since 1990.

as new observations on air pollution in China. While acknowledging that health impacts, biomass burning, high background concentrations of sulfate (SO_4^{2-}) aerosol and continental outflows of particulate pollutants from China are important topics, this review focuses on the long-term trends of major air pollutants, the key results from air pollution studies to date, and the immediate challenges in understanding and controlling air pollution in these mega cities. The paper is organized in the order of discussions of air pollution in Beijing, Shanghai and the PRD region. The length and focus of the discussion for each city reflect the abundance of information available in the literature.

This review is not intended to be exhaustive but to cover the major representative findings in air quality studies in the three mega cities. Furthermore, the information included in the review is limited to the literature published before September 2006. We apologize to those whose works are not included in this review. Because of the large number of authors sharing the same last names, we include their first initials in some citations in this paper for clarity and the convenience of readers.

2. Air pollution in Beijing

Firstly, we give a general description of the topography and meteorology of Beijing and present the spatial and temporal variations of the gaseous pollutants there, followed by a description of the emission inventories of these pollutants developed over the last decade, especially since 2000. The contributions of various sources to the ground level concentrations of air pollutants and ozone (O_3) formation are discussed. Secondly, we focus on atmospheric PM. Emissions inventories, source apportionments, temporal and spatial variations of mass concentrations, chemical compositions, secondary formation of particulate species, regional transport, and radiative properties are summarized.

2.1. Topography and meteorology

Beijing is the capital of China and is located at 39°56′N and 116°20′E on the northwest border of the Great North China Plain. Beijing is surrounded by the Yanshan Mountain in the west, north and northeast and is connected to the Great North China Plain in the south. Beijing covers an area of 16,807.8 km^2, including 16 districts and two counties. Beijing will host the 2008 Olympic Games.

Beijing is in a warm temperate zone and has a typical continental monsoon climate with four distinct seasons. During 2002–2005 (Beijing Statistical Yearbook 1997–2005, 1998–2006), the lowest monthly average temperature (T) was −4 °C in January and the highest monthly average T was 26 °C in July. Domestic heating in Beijing usually starts in mid-November and ends in the following March and it is the major source for SO_2 in the winter season (He et al., 2001; Hao et al., 2005; Wang Y. et al., 2005a; Xu X.D. et al., 2005a). The monthly average relative humidity (RH) is generally less than 60%, except in July, August and September; and the highest monthly average RH is 77% in August. Gao et al. (2005) reported that the high T and RH in August in Beijing were due to subtropical anticyclones that suppressed the uplifting of the lower-troposphere. High T and RH conditions in the summer favor the transformation of air pollutants, resulting in some episodes with high concentrations of secondary pollutants (He et al., 2001; Song et al., 2002; Yao et al., 2003a; Xu X.D. et al., 2003a, b, 2005a, b; Shao et al., 2006). In addition, the annual average number of foggy days (with visibility less than 1 km) in Beijing was 15 days during 1999–2004 (Beijing Environmental Bulletin, 1994–2005). Light fog meteorological conditions (with visibility between 1 and 10 km; also called "mist" in China) occurred more frequently and the annual average number of light foggy days was 180 days during 1999–2004. Foggy conditions promote atmospheric chemical reactions (Ondov and Wexler, 1998; Seinfeld and Pandis, 1998; Jacob, 2000). Hydrophilic aerosols can be cloud condensation nuclei, leading to the formation of fogs (Seinfeld and Pandis, 1998).

The prevailing wind in Beijing is from the north and the northwest, particularly, in the winter and the spring, respectively. The concentrations of air pollutants generally decrease with increasing wind speed (He et al., 2001). However, the strong northwestern wind carries dust from the Gobi desert to Beijing in the spring and leads to low visibility and high PM_{10} concentrations (Fan et al., 1996; Zhang and Iwasaka, 1999; Zhuang et al., 2001; Mori et al., 2003; Sun et al., 2004a; Wang Y. et al., 2005b; Zhang R. et al., 2005).

Since 1990, the population in Beijing has been growing at a rate of about 2% annually. In 2004, Beijing had a population of 15.4 million (Beijing Statistical Yearbook 2005, 2006). The increase in the population has stimulated real-estate development

and accelerated urbanization. The urban area has continuously expanded to the suburbs and the urbanization has led to a difference in land surface temperatures between the urban and suburban areas of 4–6 °C and a decrease in RH in the urban areas (Zhang J.H. et al., 2005; Liu X.-M. et al., 2006). In the last decade, many new tall buildings have been built, hindering the dispersion of air pollutants (Hao et al., 2000; Liu H. et al., 2005). The wide-spread construction activities have also exacerbated Beijing's air pollution problems (He et al., 2001; Yang et al., 2005a; Hao et al., 2005). The growth of the population and GDP (Beijing Statistical Yearbook 2005, 2006) has increased the demand for vehicular transport. The number of vehicles has increased from 1.39 million in 1999 to 2.65 million in 2005, as shown in Fig. 3. Beijing has recently adopted the Euro-III standards to reduce vehicle emissions of air pollutants.

2.2. Gaseous pollutants

2.2.1. Annual and spatial variations

Since 1999, NO_2 and PM_{10} replaced NO_x and TSP, respectively, and are reported in the Beijing Environmental Bulletin. As shown in Fig. 4, the annual average of SO_2 and CO concentrations gradually decreased from $120\,\mu g\,m^{-3}$ and $3.3\,mg\,m^{-3}$ in 1998 to $50\,\mu g\,m^{-3}$ and $2.0\,mg\,m^{-3}$ in 2005. However, the annual average NO_2 concentration has stayed at a level of about $70\,\mu g\,m^{-3} \pm 10\%$ and almost remained constant. The high SO_2 concentration in the winter is mainly due to domestic heating (Wang Y. et al., 2005a; Duan et al., 2006). For example, Wang Y. et al. (2005a) reported that the average SO_2 concentration was 163.7 and $14.8\,\mu g\,m^{-3}$ in the winter and in the summer in 2001–2003 in Beijing, respectively. The CO concentration was at a safe level while the annual average NO_2 concentration exceeded the Grade-II standards.

The number of hours and days with O_3 concentrations exceeding the Grade-II standards decreased during the period of 1999–2002 in Beijing. The lowest numbers of hours and days occurred in 2002 and the number of hours slightly increased after 2002. Since 1998, 12 phases of air pollution control measures have taken place and dozens of measures have been implemented in planning for the Beijing 2008 Olympic Games (http://www.bjepb.gov.cn/bjhb/). For example, to control industrial pollution, high emissions plants were relocated out of Beijing, cleaner production techniques were utilized, and a total industrial emissions control measure was implemented. Dozens of new standards, including the recent Euro-III standards in 2006, were promulgated and implemented to reduce vehicular emissions. Several actions were taken to reduce fugitive dust emissions. The decrease in SO_2, CO, and the number of hours of O_3 exceeding the standards, in general, can be attributed to these actions. Although the vehicular population increased by about 15% per year in Beijing, the CO

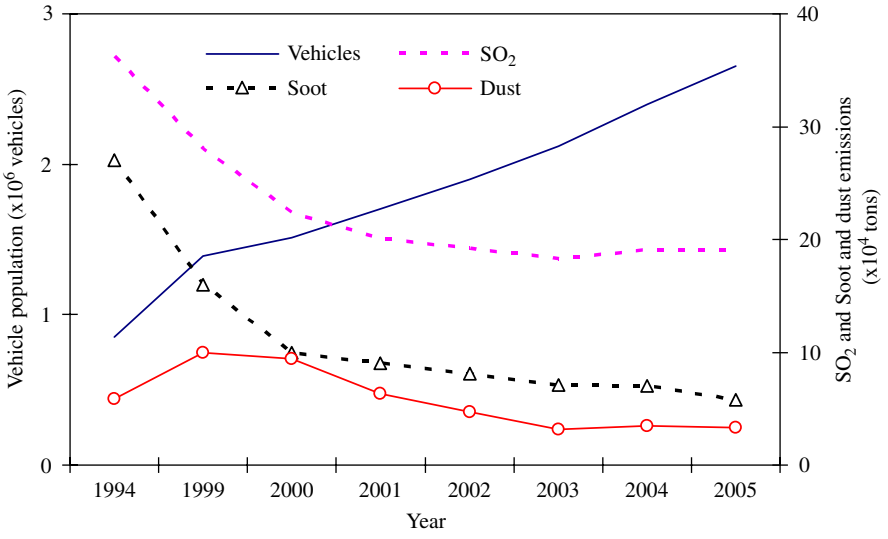

Fig. 3. Annual variations in the vehicular population and SO_2 and soot emissions in Beijing. Data are from the Beijing Environmental Bulletin 1994, 1998–2005 (http://www.bjepb.gov.cn/bjhb/tabid/69/MoreModuleID/445/MoreTabID/66/Default.aspx).

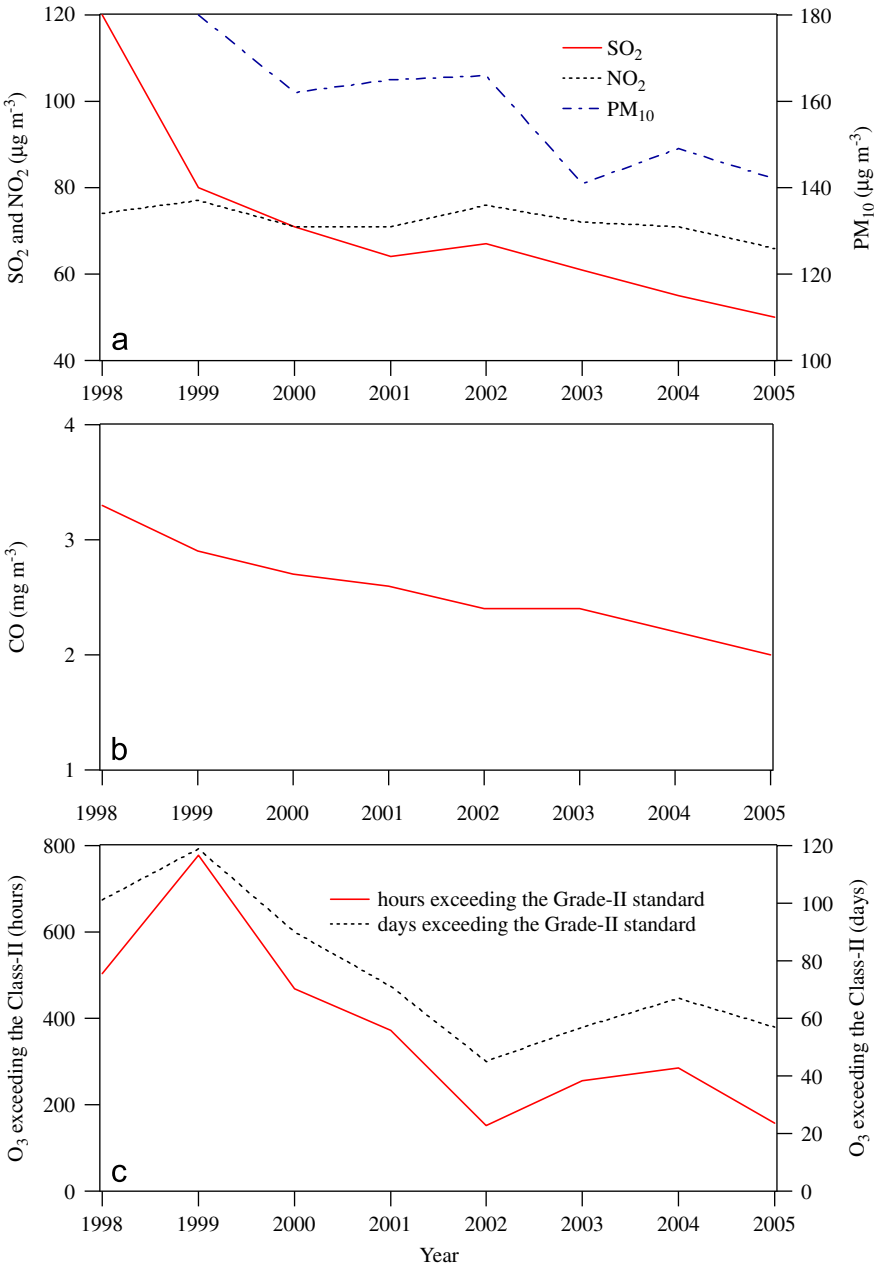

Fig. 4. Annual variations in selected air pollutants in Beijing. PM10 data in 1998 are not available. Data are from the Beijing Environmental Bulletin, 1998–2005 (http://www.bjepb.gov.cn/bjhb/tabid/69/MoreModuleID/445/MoreTabID/66/Default.aspx).

concentrations and the number hours of O_3 exceeding the standards decreased, and the NO_2 concentration remained constant. These trends suggest that vehicular emissions have not increased due to effective control measures. Based on the above analysis, NO_2 and O_3 are the major gaseous pollutions in Beijing.

2.2.2. *Emissions*

The local emissions sources for air pollution include power plants, domestic heating, and industrial, vehicular, and biogenic sources. The Beijing Municipality has developed an emissions inventory of air pollutants (Hao et al., 2005), which is referred to as the "local emissions inventory" in this paper.

The emissions factors in the local emissions inventory are generally the same as those used by Kato and Akimoto (1992), which may not sufficiently reflect the current emissions characteristics of those anthropogenic sources. For example, new technologies, such as desulfurization, low-NO_x combustion, and cleaner production in major industrial facilities, have been implemented recently to reduce air pollutant emissions in Beijing (Hao and Wang, 2005). The emissions factors of the air pollutants from power plants and industrial sources have probably changed significantly. Moreover, the sources and quality of coal have varied due to the shortage of coal supply during the last 5 years in China. In general, the emissions factors of various point and area sources in China have not been well developed. These uncertainties in the emissions factors could severely affect the estimated emissions inventory of air pollutants. Recently, vehicular emissions factors of CO, NO_x and HC (hydrocarbon) in China were investigated by Fu et al. (2001), Wang S.-X. et al. (2001), and Wang Q.-D. et al. (2005).

As reported in the Beijing Environmental Bulletin, SO_2 emissions decreased from 3.62×10^5 tons in 1994 to 1.91×10^5 tons in 2005 as shown in Fig. 3. The local emissions inventory shows that the three major contributors of SO_2 are power plants, domestic heating, and industrial sources; accounting for 49%, 26% and 24% of the total emissions, respectively, in 1999 (Hao et al., 2005). When no external transport of SO_2 to Beijing was considered, Hao et al. (2005) estimated that 39% and 36% of the ground SO_2 concentrations in Beijing were, respectively, from domestic heating and industrial area sources using the ISCST3 model with the local emissions inventory. The power plant and industrial point emissions contributed only 8% and 4% to the ground SO_2 concentrations, respectively, due to their emissions at high elevations (>60 m). More than 20% of the ground SO_2 concentration in Beijing was estimated to be transported from external sources (Yan P. et al., 2005; Xu J.M. et al., 2005).

NO_x ($NO_2 + NO$) emissions in 1995 in Beijing were 2.37×10^5 tons and there was no significant increase in the total NO_x emissions in 1998 (2.39×10^5 tons) (Hao et al., 2000). In Fig. 4, the constant NO_2 concentrations in Beijing from 1998 to 2005 suggest that there is no significant increase in NO_x emissions in Beijing. Hao et al. (2005) estimated that the emissions from vehicles, power plants, and industries in 1999 accounted for 35%, 27% and 26% in the total local NO_x emissions, respectively. They found that 74% of the ground NO_x was due to vehicular emissions while power plants and industrial sources only contributed 2% and 13%, respectively. Using Models-3/CMAQ with the local emissions inventory, Xu J.M. et al. (2005) predicted ground-level NO_2 concentrations that agree well the measured values.

Volatile organic compounds (VOC) play a key role in the photochemical reactions that form O_3 in the atmosphere. The average mass concentration of total VOCs in 2002–2003 at six sites in Beijing, including urban, suburban and rural sites, was $132.6 \pm 52.2\,\mu g\,m^{-3}$, with alkanes, aromatics, and alkenes accounting for 35%, 22%, and 17%, respectively (Liu Y. et al., 2005a). Hao et al. (2005) estimated that the VOC emissions were 2.4×10^5 tons in 1999 and that on-road vehicles accounted for 46% of the total anthropogenic VOC emissions, followed by 23% from solvent and oil distribution and 30% from power plants and industrial and domestic sources. Using the chemical mass balance (CMB) model with the measured ambient VOC concentrations in 2002–2003 and the measured source profiles, Liu Y. et al. (2005a) estimated that vehicular exhaust was the major contributor of VOC (on average 57.7%), followed by painting operations, gasoline vapor, and liquefied petroleum gas (LPG) at 12.4%, 11.3%, and 5.8%, respectively. Lu et al. (2006) obtained similar source apportions using the CMB model and their measurements in 2002 in Beijing. In addition, Wang Z. et al. (2003) developed a biogenic VOC inventory in Beijing and they estimated total annual biogenic VOC emissions to be 1.6×10^4 tons C. The biogenic VOC appears to play a minor role in the total VOC emissions.

NH_3 is an important gas in the atmosphere. It can react with gaseous HCl and HNO_3 to form particulate NH_4Cl and NH_4NO_3 and neutralize sulfuric acid aerosols. Yao et al. (2003a) measured NH_3 concentrations in Beijing. They varied from 4.6 to $42.4\,\mu g\,m^{-3}$ with the highest concentration detected in July, possibly due to thermal decomposition of NH_4NO_3 at high temperatures. Wang W.X. et al. (1997) and Peng et al. (2000) estimated that livestock emissions and fertilizers accounted for 34–54% and 18–43% of NH_3 emissions in Beijing, respectively.

Recently, Klimont et al. (2002) and Streets et al. (2003) developed an emissions inventory that

includes a database for Beijing, based on the TRACE-P experiment. Hereafter, we refer to this emissions inventory as the "TRACE-P emissions inventory". The SO_2 emissions in Beijing from the TRACE-P emissions inventory are approximately 30% higher while the NO_x, CO and VOC emissions are 30–100% lower than those from the local emissions inventory. The differences between the two emissions inventories have not been discussed in the literature. Streets et al. (2006) recently revised the TRACE-P CO emissions inventory and increased the total CO emission in 2001 by 36%. In addition, Kim et al. (2006) used Models-3/CMAQ to evaluate the accuracy of NH_3 emissions in East Asia including Beijing, based on the TRACE-P emissions inventory. They proposed that NH_3 emissions are likely to be overestimated by 1.2–3.8 times. They recommended a 20–75% reduction in NH_3 emissions in the inventory.

2.2.3. Ozone formation

As discussed earlier, NO_2 and O_3 are two major gaseous pollutants and NO_2 is mainly from local on-road vehicle emissions in Beijing. Strengthening the local NO_x emissions regulations from on-road vehicles would alleviate ambient NO_2 pollution. However, the control of O_3 is a complicated problem due to the nature of the non-linear formation of O_3. In this paper, O_3 episodes are events with hourly O_3 concentrations larger than $240\,\mu g\,m^{-3}$ (the Grade-II standards), if not specified otherwise. O_3 episodes are frequently observed during the summer in Beijing (Song et al., 2002; Shao et al., 2006). For example, the maximum hourly O_3 concentration in 2005 reached $424\,\mu g\,m^{-3}$, which was 2.7 times the Grade-II standards and also higher than the maximum concentration of $384\,\mu g\,m^{-3}$ in 1998 (http://www.bjepb.gov.cn). Zheng X. et al. (2005a) observed O_3 episodes in a rural area (the Ming Tombs) in September 2001 when the air mass was transported from Beijing's urban areas to the site. In addition, the hours of O_3 concentrations exceeding the Grade-II standards unexpectedly increased after 2002. NO_x-VOC photochemical reactions are commonly believed to generate O_3 in the atmosphere. Shao et al. (2005) studied the reactivity of VOC in Beijing. They found that alkenes contribute 75% of the atmospheric VOC reactivity; and C_4 and C_5 alkenes are particularly important, although alkenes only account for 15% of VOC by mass. They also found that alkenes are mainly from vehicular emissions and solvents. Since biogenic VOC only accounts for a small faction in the VOC emissions inventory, biogenic VOC plays only a minor role in O_3 formation in Beijing (Shao et al., 2000; Wang and Li, 2002).

Using Models-3/CMAQ with the TRACE-P emissions inventory, Streets et al. (2007) found that the maximum hourly O_3 concentrations in July 2001 in Beijing were more than 90 ppb. To investigate the regional and local contribution to the ground-level O_3, Streets et al. (2007) run Models-3/CMAQ with all the local man-made emissions removed and the predicted O_3 concentrations were 60–90 ppb. They estimated that the regional contribution to the ground-level O_3 could be 30–70%. They also suggested that VOC emissions were underestimated based on the VOC/NO_x ratio. The typical VOC/NO_x ratio was six in Beijing while the typical value in the US is 10–15.

Shao et al. (2006) proposed that strong interactions between atmospheric oxidants and particulate species could be present in Beijing. These interactions are non-linear (Seinfeld and Pandis, 1998) and complicated mathematical models are needed to validate the interactions (Jacob, 2000). Xu J. et al. (2006) studied the influence of four heterogeneous reactions of NO_2, NO_3, N_2O_5 and HO_2 with particles in O_3 formation in June 2000 in Beijing, using the CMAQ-MADRID (Community Multiscale Air Quality-Model of Aerosol Dynamics, Reaction, Ionization and Dissolution) model with the local emissions inventory. They found that the formation of O_3 was VOC-limited in urban areas and the inclusion of the four heterogeneous reactions improved the agreement between the predicted and measured O_3. They also found that heterogeneous NO_2 reactions could increase the O_3 peak by 67 ppb and heterogeneous HO_2 reactions could lower the O_3 peak by 17 ppb. Heterogeneous NO_3 and N_2O_5 reactions appear to be unimportant. In the analysis, the major uncertainties, such as the heterogeneous reaction coefficients used in the simulations, the deliquescence state of the aerosols, and the emission inventories were not discussed. Using the CMAQ-MADRID model, Xu and Zhang (2006) found that O_3 was mainly generated at elevations of 200–800 m and the increase in the ground-level O_3 was due to the transport of O_3 from aloft. Wang and Li (2002) and Tang (2004) suggested that about 40% regional contribution of O_3 came from the cities and provinces southern and southeastern of Beijing. In addition, air pollutants

emitted in Beijing can be transported and affect O_3 formation in downwind areas such as the Shandong Province (Guttikunda et al., 2005).

2.3. Particulate pollutants

PM_{10} is reported to be the major air pollutant on about 90% of the days in the last 7 years in Beijing (Beijing Environmental Bulletin, 1999–2005). On 30% of the days in each year, the daily average PM_{10} concentration exceeded the Grade-II standards. We next examine the emissions inventory, the temporal and spatial variations of abundance, the chemical composition, the radiative properties, the source apportionments of atmospheric particles, and the role of the formation of secondary species and regional transport of PM_{10} in Beijing.

2.3.1. Emissions

As shown in Fig. 3, the total emissions from combustion soot particles decreased from 2.7×10^5 tons in 1994 to 5.8×10^4 tons in 2005 while the industrial dust emissions from non-combustion processes increased from 5.9×10^4 tons in 1994 to 10.0×10^4 tons in 1999. Subsequently, industrial dust emissions decreased to 3.3×10^4 tons in 2005 (Beijing Environmental Bulletin, 1994–2005). Hao et al. (2005) estimated that the local PM_{10} primary sources including fugitive dust, dust from industrial sources, soot particles emitted from on-road vehicles and others accounted for 49%, 28%, 8% and 15% of the total emissions in 1999, respectively. From 1999 to 2005, soot particles and non-combustion industrial dust emissions decreased by about 60% as shown in Fig. 3. Secondary formation and regional transport are important sources of atmospheric particles in Beijing and they will be discussed later.

2.3.2. Annual and seasonal variations of PM_{10} and $PM_{2.5}$ concentrations

As listed in Table 1, PM_{10} is a criteria pollutant in the National Ambient Air Pollutant Standards, but $PM_{2.5}$ is not. Thus, routine measurements at the local government air quality monitoring stations include PM_{10} but not $PM_{2.5}$. Although TSP is still in the current standard, but Environment Bulletin in China does not report TSP after 2001. TSP is particularly affected when dust storms hit Beijing. We return to TSP when we discuss dust storm events. The annual average of PM_{10} decreased from $180\,\mu g\,m^{-3}$ in 1999 to $142\,\mu g\,m^{-3}$ in 2005 in Beijing as shown in Fig. 4. The annual average of PM_{10} was almost constant from 2003 to 2005, and was about 40% higher than the Chinese Grade-II standards and seven times the latest WHO Air Quality Guidelines (WHO, 2005). In 2005, the PM_{10} concentration in urban areas and the southern suburban areas was almost spatially uniform but the PM_{10} concentration in the northern suburban areas was about 30% lower than the PM_{10} concentration in urban areas (Beijing Environmental Bulletin, 2005). Zhang Y.H. et al. (2004) also reported that the daily average PM_{10} concentrations were sometimes higher than $100\,\mu g\,m^{-3}$ at the rural sites of Ming Tomb and Yongledian in Beijing in 2000. The high background concentrations suggest the importance of regional sources of PM_{10} in Beijing.

$PM_{2.5}$ measurements have been widely reported in Beijing in the last 10 years. He et al. (2001) made the first comprehensive $PM_{2.5}$ measurements in Beijing and reported that the annual average of $PM_{2.5}$ was $115\,\mu g\,m^{-3}$ at Chegongzhung (an urban site) from September 1999 to September 2000. The annual average was $96.5\,\mu g\,m^{-3}$ at the same site from August 2001 to September 2002 and the decrease was attributed to the air pollution control measures carried out in Beijing since 1998 (Duan et al., 2006). On the basis of measurements on selected days in four seasons, Zheng M. et al. (2005) and Wang J. et al. (2004) reported annual averages of $PM_{2.5}$ in 2000 and in 2001 of $101.4\,\mu g\,m^{-3}$ and $109.6\,\mu g\,m^{-3}$, respectively. Wang Y. et al. (2005a) reported that the average of $PM_{2.5}$ from 2001–2003 was $154.3\,\mu g\,m^{-3}$, significantly higher than the values reported by the other studies mentioned above. All of these reported annual averages of PM_{10} and $PM_{2.5}$ concentrations are seven times or more greater than the WHO Air Quality Guidelines.

There are relatively small seasonal variations of $PM_{2.5}$ and PM_{10} in Beijing; the highest and lowest seasonal average concentrations of $PM_{2.5}$ and PM_{10} were observed in the winter and in the spring, respectively (He et al., 2001; Duan et al., 2006). The highest seasonal average concentrations were less than twice the lowest average values. Although the low temperature in the winter limited secondary formation of particles, more frequent occurrences of the stagnant weather conditions caused the accumulation of atmospheric particles and high concentration episodes (He et al., 2001; http://www.bjepb.gov.cn/bjhb/tabid/68/InfoID/8898/Default.aspx). In addition, primary particle emissions are expected to

be higher in the winter due to domestic heating practices. While there are numerous $PM_{2.5}$ and PM_{10} measurements on selected days in different seasons (e.g., Wang J. et al., 2004; Wang Y. et al., 2005a; Zheng M. et al., 2005; Xu X.D. et al., 2005c; Yu J. et al., 2006), He et al. (2001) and Duan et al. (2006) provided the longest database for seasonal trend analysis.

Since different sizes of atmospheric particles probably originate from different sources, the $PM_{2.5}/PM_{10}$ ratio has been used for identifying their sources in Beijing (Sun et al., 2004b, 2006a; Chan et al., 2005; Xie et al., 2005; Yu J. et al., 2006). The reported ratios varied spatially and temporally, ranging from 0.25 to 0.88. Despite these variations, high ratios (e.g., larger than 0.6) are generally ascribed to secondary particulate formation of species such as nitrate (NO_3^-), SO_4^{2-} and ammonium (NH_4^+) and organics, while low ratios are ascribed to the overwhelming contribution from fugitive dust or sand dust from long-distance transport (Xie et al., 2005; Duan et al., 2006; Sun et al., 2006a). It should be pointed out that the uncertainties in particle mass concentration measurements can also affect the reported ratios (He et al., 2001), although this issue was not discussed in most studies.

2.3.3. Chemical compositions of $PM_{2.5}$ and PM_{10}

$PM_{2.5}$ mass and chemical compositions in Beijing have been widely studied (He et al., 2001; Sun et al., 2004b; Zhang Y.H. et al., 2004; Chan et al., 2005; Zheng M. et al., 2005; Duan et al., 2005, 2006; Wang Y. et al., 2005a; Guinot et al., 2006; Yu J. et al., 2006). In these studies, the carbonaceous species (the sum of organic carbon and elemental carbon), the secondary species (the sum of SO_4^{2-}, NO_3^- and NH_4^+), and crustal species (Al, Si, Ca, Mg and Fe) accounted for 27–42%, 22–54%, and 11–16% of the $PM_{2.5}$ mass, respectively. There are fewer simultaneous measurements of the three types of species in PM_{10} in Beijing. As will be discussed later, some particulate organics are secondary pollutants, but they are classified under carbonaceous particles for the source apportionment discussion here. A number of studies have reported metal compositions for source apportionments. In this section, we focus on the discussion of the major species that contribute to PM mass. Readers interested in the metal compositions can refer to the references cited for source apportionment analysis.

Using the CMB model and incorporating organic molecular markers, Zheng M. et al. (2005) estimated that secondary sources (the sum of SO_4^{2-}, NO_3^-, and NH_4^+), dust, coal combustion, diesel and gasoline exhaust, and biomass aerosols accounted for 33%, 20%, 7%, 7%, and 6%, respectively, in $PM_{2.5}$. Coal combustion, diesel and gasoline exhaust, and biomass aerosols are the three major contributors to the carbonaceous species. Similar results were obtained by Song et al. (2006a), who used the Positive Matrix Factorization (PMF) model with the same dataset used by Zheng M. et al. (2005).

Using CMB and the ISCST3 model, Hao et al. (2005) found that the two methods yielded similar results for PM_{10} in Beijing in 1999, and the estimated contributions of fugitive dust, coal burning, secondary PM_{10} and vehicular exhaust were found to be 37–56%, 17–25%, 16–20% and 6–22%, respectively. It appears that the contribution of fugitive dust to the PM_{10} mass was almost the same as the sum of other anthropogenic contributions. Song et al. (2006b) used the CALPUFF model to estimate the PM_{10} concentration from 1 January 2000 to 29 February 2000 in Beijing. They reported that road dust only accounted for 13% of the PM_{10} mass. The difference in the estimated apportionment between Hao et al. (2005) and Song et al. (2006b) might be due to temporal variations; however, other possibilities cannot be excluded. There are a number of other studies on particulate chemical compositions in Beijing in the last two decades without source apportionment analysis (Winchester and Bi, 1984; Dod et al., 1986; Ning et al., 1996; Wei et al., 1999; Zhang and Friedlander, 2000; Mukai et al., 2001; Cao et al., 2002). In addition, size distributions of chemical compositions (Lun et al., 2003; Yao et al., 2003a, b; Dillner et al., 2006) were also reported in Beijing. We will return to size distribution data when we discuss secondary formation of particulate species.

Overall, the carbonaceous species and secondary species accounted for more than 60% of the $PM_{2.5}$ mass and about 50% in PM_{10} mass in Beijing.

2.3.4. Carbonaceous species in $PM_{2.5}$ and PM_{10}

As discussed above, carbonaceous species are important particulate components in Beijing. The reported total carbon concentration (TOC = organic carbon (OC) + elemental carbon (EC)) varied from 10.8 to 51.9 $\mu g\,m^{-3}$ (He et al., 2001, 2004; Sun et al., 2004b; Chan et al., 2005; Zheng M. et al., 2005; Duan et al., 2005, 2006; Guinot et al., 2006;

Yu J. et al., 2006). The reported OC/EC ratio was generally between two and three but it can also be larger than six in some cases. These ratios are relatively low compared to those in the US and Europe (Turpin and Huntzicker, 1995; Castro et al., 1999), probably due to the high contribution of EC from coal-combustion in China (Zheng M. et al., 2005). Chow et al. (2005) suggested that the chemical analytical methods used can result in significant differences in the ratio, although the TOC concentration is generally consistent. Variations in the TOC concentration in Beijing may be mainly attributed to spatial and temporal variations.

The secondary organic species (SOC) concentration can be estimated by assuming that the primary OC/EC ratio is relatively constant and that the minimum OC/EC ratio in particles is the primary OC/EC ratio (Turpin and Huntzicker, 1995; Castro et al., 1999). Using the above estimation, the SOC/OC ratio was estimated to be between 30% and 70% in Beijing (Sun et al., 2004b; Chan et al., 2005; Zheng M. et al., 2005; Duan et al., 2006; Yu J. et al., 2006). In those studies, high SOC/OC ratios were generally found in the summer and were ascribed to photochemical formation of SOC at high temperatures. In addition, Chan et al. (2005) reported that the OC/EC ratio and the SOC/OC ratio were 2.9 and 0.3, respectively, and the TOC in PM_{10} was $32\,\mu g\,m^{-3}$ in August 2003 in Beijing.

Biomass burning during the harvest season can be an important source of carbonaceous species in Beijing. For example, Duan et al. (2004) and Zheng X. et al. (2005b) reported that about 50% of the organic carbon came from biomass burning during some severe pollution episodes in Beijing. Using ^{14}C analysis, Yang et al. (2005b) estimated that the contribution of modern carbon to the TOC in $PM_{2.5}$ was 33–48% in Beijing, with higher fractions in the harvest seasons and that modern carbon mainly originated from biomass burning and kitchen emissions.

The total resolved solvent extractable organic compounds accounted for about 5–10% of the organic matter in Beijing (Xu D. et al, 2005; Zheng M. et al., 2005; Hou et al., 2006). The concentrations of solvent-extractable organic matter mainly consisted of fatty acids and showed distinct seasonal variations with the highest concentrations in the winter and the lowest concentrations in the summer (Feng et al., 2005, 2006a).

2.3.5. Formation of secondary particulate species

Gas condensation occurring in the source plumes and the ambient air, heterogeneous reactions on aerosol surfaces or in aerosol droplets, and in-cloud processes have been proposed to be the three main pathways of secondary particulate species formation in Beijing (Zhang and Iwasaka, 1999; Lun et al., 2003; Shi Z. et al., 2003, 2005; Yao et al., 2002a, 2003a, b; Liu X. et al., 2005; Liu Y. et al., 2005b; Matsuki et al., 2005; Wang Y. et al., 2005a, 2006a; Yu J.Z. et al., 2005; Li L. et al., 2006; Sun et al., 2006a). Several approaches have been used to examine the importance of secondary formation of particulate SO_4^{2-} in Beijing as summarized below:

(1) Wang Y. et al. (2005a), Sun et al. (2006a) and Wang Y. et al. (2006a) used the molar sulfur oxidation ratio, defined as $SOR = [SO_4^{2-}]/([SO_4^{2-}]+[SO_2])$, to determine the extent of secondary formation of particulate SO_4^{2-} in Beijing. They found high (substantially larger than 0.1) SOR in the summer and the fall and low (smaller than 0.1) SOR in the winter and the spring and attributed SO_4^{2-} to secondary SO_4^{2-} and primary sources, respectively. However, long range transport of SO_2 and SO_4^{2-} can also influence SOR and complicate the analysis. Additional correlation analysis of SO_2 with SO_4^{2-} would be useful (Yao et al., 2002a).
(2) Zhang and Iwasaka (1999), Shi Z. et al. (2003, 2005), and Matsuki et al. (2005) used single particle morphology to identify secondary formation of SO_4^{2-}. The modification of the morphology of SO_4^{2-} particles, used as an indictor of secondary formation of SO_4^{2-}, was sometimes detected in atmospheric particles in Beijing (Shi Z. et al., 2003; Matsuki et al., 2005) but sometimes was not (Zhang and Iwasaka, 1999; Shi Z. et al., 2005).
(3) Li L. et al. (2006) determined the heterogeneous oxidation rate of SO_2 by O_3 on the surface of calcium carbonate ($CaCO_3$) and proposed that the oxidation process can be important, particularly when Beijing was hit by dust storms. However, they did not analyze field data to support their laboratory studies.
(4) Yao et al. (2003a) found that SO_4^{2-} had a dominant mode at 0.7 μm and a minor mode at 6 μm in the summer in Beijing. The dominant mode at 0.7 μm is the typical size of the droplet mode formed by in-cloud processes (Ondov and Wexler, 1998; Seinfeld and Pandis, 1998).

The minor mode at 6 μm is similar to the mode of Ca^{2+} and Mg^{2+}, suggesting that heterogeneous reactions between ambient SO_2 and crustal species such as $CaCO_3$ can be a major pathway of SO_4^{2-} formation in supermicron particles. Yao et al. (2003a) reported that the size distribution of SO_4^{2-} in the spring exhibited a dominant mode at 0.45 μm and a minor mode at 4 μm due to dry weather, which was also observed in the spring in Beijing by Dillner et al. (2006). Yao et al. (2003a) attributed the dominant 0.45 μm mode to gas condensation in source plumes and the minor mode to heterogeneous reactions between SO_2 and crustal species such as $CaCO_3$. Yu J.Z. et al. (2005) found a good correlation between SO_4^{2-} and oxalate in $PM_{2.5}$ in Beijing and proposed that both of them could be formed by in-cloud processes.

NH_4NO_3 is a semi-volatile species and its concentration is not only affected by formation processes, but also by the gas-aerosol equilibrium. The size distribution of NO_3^- was sometimes similar to SO_4^{2-} with the dominant mode in the submicron particle size but it sometimes exhibited a dominant mode in the supermicron particle size in Beijing (Lun et al., 2003; Yao et al., 2003a; Dillner et al., 2006), depending on T and RH. The dominant mode of NO_3^- in the supermicron particles is believed to be due to the reactions of HNO_3 with $CaCO_3$ because Ca^{2+} was the dominant cation in supermicron particles in Beijing (Yao et al., 2003a).

NH_4^+ was mainly associated with NO_3^- and SO_4^{2-} in particles in Beijing and the ($[NO_3^-]+[SO_4^{2-}]$) to $[NH_4^+]$ equivalence ratio was about 1.2 (Yao et al., 2002a; Lun et al., 2003; Yao et al., 2003a; Wang Y. et al., 2005a; Duan et al., 2006), suggesting that SO_4^{2-} aerosols can be completely neutralized. The size distribution of NH_4^{+-} exhibited a uni-modal in submicron particle size (Yao et al., 2003a). However, no denuders and back-up filters were used in these studies. Sampling artifacts of NO_3^- and NH_4^+ could be important (Pathak et al., 2004a, b; Pathak and Chan et al., 2005). Real-time gas and particulate measurements can alleviate the artifacts (Hu et al., 2002). The strong acidity of PM was not measured.

Some secondary particulate organic species such as dicarboxylic acids and methane sulfonic acid (MSA) were also studied in Beijing (Yao et al., 2002a, 2003b; Hui et al., 2004; Huang X.F. et al., 2005; Yu J.Z. et al., 2005). In these studies, oxalate is the dominant ion of dicarboxylic acids and is generally believed to be formed by in-cloud processes. However, the sum of these species only accounts for less than 10% of SO_4^{2-} in atmospheric particles and likely plays a minor role in the atmosphere.

2.3.6. Regional transport of atmospheric particles

Regional transport of atmospheric particles has been widely studied in Beijing during the last decade. Dust storms are one of the major regional sources in the spring. The most severe dust storm in the last two decades was on 20 March 2002, when the maximum concentration of TSP was about $10\,mg\,m^{-3}$ and the $PM_{2.5}$ concentration was more than $1\,mg\,m^{-3}$ (Sugimoto et al., 2003; Guo J. et al., 2004; Shi Z. et al., 2005; Zhang R. et al., 2005). There are five routes for dust storms to reach Beijing: (a) the Northeastern Sand Land Path; (b) the Northern Mongolia Path; (c) the Northern Desert Path; (d) the Western Desert Path; and (e) the Turning Path, reaching Beijing from the south (Wang A.P. et al., 1996; Zhang and Iwasaka, 1999; Mori et al., 2003; Zhang X.Y. et al., 2003; Wang Y.Q. et al., 2004; Wang Y. et al., 2005b; Sun et al., 2005; Zhang R. et al., 2005). Based on chemical composition analyses (Fan et al., 1996; Zhou et al., 1996; Zhang and Iwasaka, 1999; He et al., 2001; Wang Y. et al., 2005b; Sun et al., 2005), dust storms can be further classified to be Ca-enriched, when the air mass comes from the west and passes through the Loess plateau of China (Western Desert Path), and Ca-deficient in the other cases. When dust storms hit Beijing, crustal species, such as Si, Al, Ca, Fe, etc., from the transport overwhelm the TSP and $PM_{2.5}$ (Cheng et al., 2005; Sun et al., 2005; Xie et al., 2005; Zhang R. et al., 2005).

Other pollutants carried by storm particles and NO_3^- and SO_4^{2-} formation on dust particles were generally believed to be negligible (Zhou et al., 1996; Zhang and Iwasaka, 1999; Zhang X.Y. et al., 2003; Shi Z. et al., 2005; Yuan et al., 2004; Zhang D. et al., 2005). However, some measurements suggested that NO_3^- and SO_4^{2-} formation on dust particles were significant when dust storms hit Beijing (Zhuang et al., 2001; Cheng et al., 2005; Mori et al., 2003; Sun et al., 2005). Analysis of the mechanism of the development of dust storms is needed to reconcile these contradictory results. Guo J. et al. (2004) proposed that the development of dust storms in

Beijing includes four stages, i.e., the accumulation of the pollutants, the clearing out of the pollutants, the addition of pure dust, and the clearing out of the dust. Due to the high wind speed on top of the mixing layer, a dust storm first reaches Beijing at high elevations while the particles at the ground level are mainly "local" accumulation due to the low surface wind speeds (Zhang R. et al., 2005). Compared to that at high elevation, there is a 1–2 h delay before a dust storm arrives at the ground level (Zhang R. et al., 2005). Pollutant particles are rapidly removed when the dust storm arrives at the ground level and persists (Guo J. et al., 2004). Similar dust storm developing processes have also been reported in Qingdao, downwind of Beijing (Guo Z. et al., 2004; Yang C. et al., 2005; Zhang D. et al., 2005) and a number of locations outside China (e.g., Usher et al., 2003; Krueger et al., 2004). When samples were collected over the duration from the first stage to the third stage, the collected aerosol particles could include both local aerosol particles and pure dust particles. On the other hand, the aerosol samples collected during the third stage, which usually lasts from several hours to dozens of hours, mainly include just pure dust particles. Samples from different stages of a dust storm have different aerosol chemical compositions.

Regional transport of atmospheric particles to Beijing on days without dust storms was also modeled in the literature. Streets et al. (2007) used Models-3/CMAQ to estimate that 34% (on average) of $PM_{2.5}$ came from sources outside of Beijing, including Tianjin Municipality, Hebei province, and Shandong province. Chen D.S. et al. (2007) used the same model to show that the mean monthly PM_{10} contributions from these surrounding areas to Beijing were 23.4% in January, 37.9% in April, 40.0% in July, and 37.4% in October, with an annual average of 34.7% in 2002. However, they did not consider the regional contribution from Shangdong province. They estimated the maximum regional contribution to be 80–90% in January when Beijing encountered heavy PM_{10} pollution (the maximum daily average concentration was greater than $400\,\mu g\,m^{-3}$). It is interesting to note that Chen D.S. et al. (2007) and the Beijing Municipal Environmental Protection Bureau (http://www.bjepb.gov.cn/bjhb/tabid/69/MoreModuleID/445/MoreTabID/66/Default.aspx) do not agree on the heavy PM_{10} pollution in January. The latter asserted that the January episodes were caused mainly by local sources. Overall, these modeling results suggest that more than 20% of $PM_{2.5}$ and PM_{10} was from regional transport.

Regional transport can be important sources of SO_4^{2-} aerosols in Beijing. Comparable $PM_{2.5}$ SO_4^{2-} concentrations have been reported in Tsinghua University (a suburb site) and Chegongzhuang (an urban site) and in Ming Tomb and Yongledian (rural sites), Peking University (a suburb site) and Dongsi (an urban site) (He et al., 2001; Zhang Y.H. et al., 2004). In addition, spatially uniform distributions of SO_4^{2-} concentrations over 1000 km in several areas in China are frequently observed from the space as reported in http://www.nrlmry.navy.mil/flambe-bin/aerosol/display_directory_aer2?DIR=/web/aerosol/public_html/globaer/ops_01/mongolia/.

Although crustal species such as Al, Si, Ca, Mg and Fe only accounted for about 10% of the total mass of $PM_{2.5}$ in Beijing (He et al., 2001; Sun et al., 2004b), these species account for about 30% of the total mass of PM_{10} (Sun et al., 2004b). These crustal species can come from several sources such as combustion sources, non-combustion industrial sources, and fugitive dust. The local contributions of these sources to PM_{10} have been discussed earlier in relation to the local emissions inventory. The regional transport of crustal species to Beijing during the periods without dust storm is unclear. Ca^{2+} and Mg^{2+} are mainly found in the super-micron particles (Yao et al., 2003a).

2.3.7. Others

Atmospheric aerosols affect visibility and climate through the scattering and absorption of solar radiation. Haze (with visibility less than 10 km and under $RH < 80\%$) is generally believed to be related to atmospheric aerosols. From 2001 to 2004, the recorded days with haze were 45, 43, 54 and 19 days in Beijing, respectively. The occurrence of hazy days was usually associated with low wind speed ($<2\,m\,s^{-1}$), stagnant weather conditions and sometimes foggy weather conditions in Beijing (Xie et al., 2005; Sun et al., 2006a; Wang Y. et al., 2006a). Xie et al. (2005) observed high $PM_{2.5}/PM_{10}$ ratios ($>70\%$) and high ratios of the sum of SO_4^{2-}, NO_3^-, and NH_4^+ to the total $PM_{2.5}$ mass ($>50\%$) during hazy days. Wang Y. et al. (2006a) observed that the ratio of the sum of SO_4^{2-}, NO_3^-, and NH_4^+ to the total $PM_{2.5}$ mass increased to 46% during hazy days in Beijing. These studies suggest that hazy days in Beijing were mainly because of pronounced secondary species formation (Chen et al., 2003; Kang et al., 2004).

Bergin et al. (2001) found that the low visibility (less than 5 km) in Beijing during some episodes was related to the high light extinction coefficients. About 80% of light scattering at 530 nm was due to submicron particles, which are mainly composed by carbonaceous and secondary species. Song et al. (2003) concluded that visibility in Beijing in 1999–2000 was negatively correlated to the $PM_{2.5}$ mass concentration. The correlation between the aerosol optical thickness (AOT) and visibility in Beijing has also been reported by several researchers. Qiu and Yang (2000) investigated the 0.75 μm AOT and visibility from 1980 to 1994. The AOT increased by a factor of about two in 15 years and was negatively correlated with visibility. Shi X. et al. (2005) also found that the amount of low-cloud coverage and the number of foggy days in Beijing increased with increasing AOT while the hours of solar radiation decreased with increasing AOT. Recently, Xia et al. (2006) found the highest AOT in June (1.25) and the lowest in January (0.41) during 2001–2004 in Beijing. An increase in AOT could greatly affect the regional climate. Gu et al. (2006) found that the increase in AOT in China results in a noticeable increase in precipitation in the southern part of China in July due to the strengthening of the Hadley circulation.

Recently, ultrafine particles have attracted much attention due to their potential health impact and their role as cloud condensation nuclei (Biswas and Wu, 2005). Wehner et al. (2004) observed that nucleation events formed ultrafine particles on 25 days out of 45 days while the particle surface area dropped to a critical value of 100–2000 $\mu m^2 cm^{-3}$ when clear air came from the north of Beijing. During such nucleation events, newly formed particle concentrations were more than 100,000 cm^{-3} and the growth rate was ~1 $nm\,h^{-1}$. Nucleation events in Beijing were also reported by Yu J. et al. (2005) and Guinot et al. (2006). Particulate F, Pb, and Hg in Beijing were investigated by Mukai et al. (2001), Feng et al. (2003), and Sun et al. (2006b) and Wang Z. et al. (2006). These pollutant concentrations were at safe levels. The European Commission recently proposed a directive for limiting toxic metals. The mean annual concentration proposed is 6 $ng\,m^{-3}$ for As, 20 $ng\,m^{-3}$ for Ni and 5 $ng\,m^{-3}$ for Cd in PM_{10} (European Directive 2004/107/CE, 2003). The World Health Organization (WHO) proposed an annual average concentration of 150 $ng\,m^{-3}$ for Mn in PM_{10} (WHO, 2000). Limited measurements in Beijing suggest that the concentrations of these metals are mostly at safe levels (Sun et al., 2004b), compared to the above proposed annual averages.

3. Air pollution in Shanghai

3.1. Topography and meteorology

Shanghai is located at 31°14′N and 121°29′E, and is in the center of China's coastline from north to south as shown in Fig. 1. Shanghai lies on the edge of the broad flat alluvial plain of the Yangtze River Delta with a few mountains in the southwest, and Shanghai is linked to the Jiangsu and Zhejiang Provinces in the west, the East China Sea in the east, and the Hangzhou Bay in the south. Shanghai has an area of 6340 km^2 covering 18 districts and one county. Shanghai is strong in export processing and finance as well as heavy industries with major companies such as the Baoshan Iron and Steel Group Corporation and the Shanghai Petrochemical Complex (Shanghai Statistical Yearbook 1997–2005, 1998–2006). Shanghai will host the 2010 World Expo.

Shanghai has a subtropical monsoon climate with four distinct seasons (Shanghai Statistical Yearbook 1997–2005, 1998–2006). In 2005, the lowest monthly average temperature was 4 °C in January and the highest monthly average temperature was 29 °C in July. There were 112 rainy days with a total precipitation of 1255 mm. The frequent rain removes air pollutants effectively. This is one of reasons for the PM_{10} concentration in Shanghai being less than 50% of that in Beijing, although the local soot emissions and SO_2 and NO_x emissions in Shanghai were 2–3 times those in Beijing during 2003–2005 (Beijing Environmental Bulletin, 1994–2005; Shanghai Environmental Bulletin, 1994–2005). Shanghai is humid and the monthly average RH is higher than 75% (Xu et al., 1997; Yao et al., 2002a; Zhang Y. et al., 2006). Xu et al. (1997) reported that the monthly average total cloud coverage varied from 54% to 82% while the monthly average low-cloud coverage varied from 30% to 40% during 1959–1985. Low clouds have been proposed to play an important role in atmospheric reactions in Hong Kong (Zhuang et al., 1999a). In-cloud formation and transformation of air pollutants could be important in Shanghai.

The prevailing wind direction in Shanghai varies from northwest to northeast in the winter and varies from southeast to southwest in the summer, with an annual average wind speed of 3.1 $m\,s^{-1}$

(Wang Y. et al., 2006b). Feng et al. (2006b) reported that air parcels arriving in Shanghai in the summer were mainly from the East China Sea and carried clean air. On the other hand, air parcels arriving in Shanghai in the winter were from the northeast (Yellow Sea) to northwest (inland) directions and carried polluted air from Jiangsu province.

Since 1995, Shanghai has experienced a negative natural growth in the permanent population at a rate of about −2% annually. However, the migrant population has increased with an annual growth rate of about 8%. In 2005, Shanghai had a permanent population of 13.6 million, and about 5.8 million migrants (Shanghai Statistical Yearbook 2005, 2006). The close to 10% annual growth rate of GDP in the last decade has attracted migrants and accelerated the expansion of the urban areas, leading to a decrease in the cultivated land area from 3.6×10^5 hectares in 1978 to 2.4×10^5 ha in 2005. The total lengths of roads and highways have increased from 3050 to 8110 km and 36 to 560 km, respectively, from 1990 to 2005, while the number of vehicles has increased from 0.58 million in 1998 to 2.22 million in 2005, as shown in Fig. 5. Shanghai adopted the Euro-II standards to reduce vehicular emissions of air pollutants in 2003. Since the 1990s, several projects and measures, such as controlling total SO_2 emissions, the Trans-Century Green Project, and the establishment of non-coal-burning areas have been implemented to improve air quality in Shanghai. As a result, more than 70% of the days in each year had air pollutant concentrations below the Grade-II standards in Shanghai during 1999–2005. The percentage gradually increased to 88.2% in 2005 (Shanghai Environmental Bulletin, 1994–2005).

3.2. Gaseous pollutants

3.2.1. Annual and spatial variations

As shown in Fig. 6, the maximum annual average SO_2 concentration in Shanghai was 69 $\mu g\,m^{-3}$ in 1997 and it gradually decreased to 35 $\mu g\,m^{-3}$ in 2002. The increase in the SO_2 concentrations after 2002 was related to the greater local emissions of SO_2 based on the positive correlation between the ambient SO_2 concentration and the SO_2 emission ($r^2 = 0.59$) as shown in Fig. 6c. The annual average SO_2 concentrations in most years were below the Grade-II standards (60 $\mu g\,m^{-3}$). Only 6% of the days in 2005 had SO_2 as the major pollutant (Shanghai Environmental Bulletin, 2005). From 1996 to 2000, the annual average NO_x concentration varied slightly from 89 to 105 $\mu g\,m^{-3}$. After 2000, the NO_2 concentration was available in the Shanghai Environmental Bulletin. From 2001 to 2005, the annual average NO_2 concentration was 60 $\mu g\,m^{-3}$ ±5% and exceeded the Grade-II standards. Since 2000, the number of vehicles increased by more than 10% annually (Shanghai Academy of Environmental Sciences, 2005). The near constant NO_2 concentration during 2001–2005 suggested an effective control of vehicular emissions of NO_x. Based on the Ambient National Air Quality Standards in China, the SO_2 concentration in Shanghai was at a safe level, but this was not true for NO_2. The CO and O_3 concentrations were not reported in the Shanghai Environmental Bulletin. Ye et al. (2003) reported weekly average CO concentration of 1.7–6.6 $mg\,m^{-3}$ during 1999–2000. There were 13 weeks (28% of the weeks) when the weekly average CO concentration exceeded the Grade-II standard (daily average higher than 4 $mg\,m^{-3}$) and one week (2%) when the weekly average CO concentration exceeded the Grade-III standard (daily average higher than 6 $mg\,m^{-3}$). Higher CO concentrations occurred from January to March and in May while lower concentrations occurred from June to July and from October to December.

Wang H.X. et al. (2005) and Wang H.X. et al. (2006) measured the O_3 concentration at a rural site (Sheshan) in Shanghai from May 1999 to May 2000. The highest and the lowest monthly average O_3 concentrations occurred in May (about 60 ppb) and in January (about 40 ppb), respectively, and the second lowest average was detected in August. Xu et al. (1997) and Xu et al. (1999) measured the O_3 concentrations on selected days in four seasons during 1990–1994, and reported that the maximum and minimum hourly averages of O_3 concentrations occurred in the winter and the summer, respectively. They reported that the seasonal trend of the O_3 concentrations differed from that reported by Wang H.X. et al. (2005) and Wang H.X. et al. (2006), which could be due to different sampling years. Recently, Zhang Y.H. et al. (2006) reported an overall 8-h O_3 average (from 10:00 to 18:00) of 63.3 $\mu g\,m^{-3}$ and a maximum 8-h average of 251.3 $\mu g\,m^{-3}$ in 2001–2004. It is clear that some O_3 episodes have occurred in Shanghai. However, details of these episodes, such as the frequency, the weather conditions, and the temporal and spatial

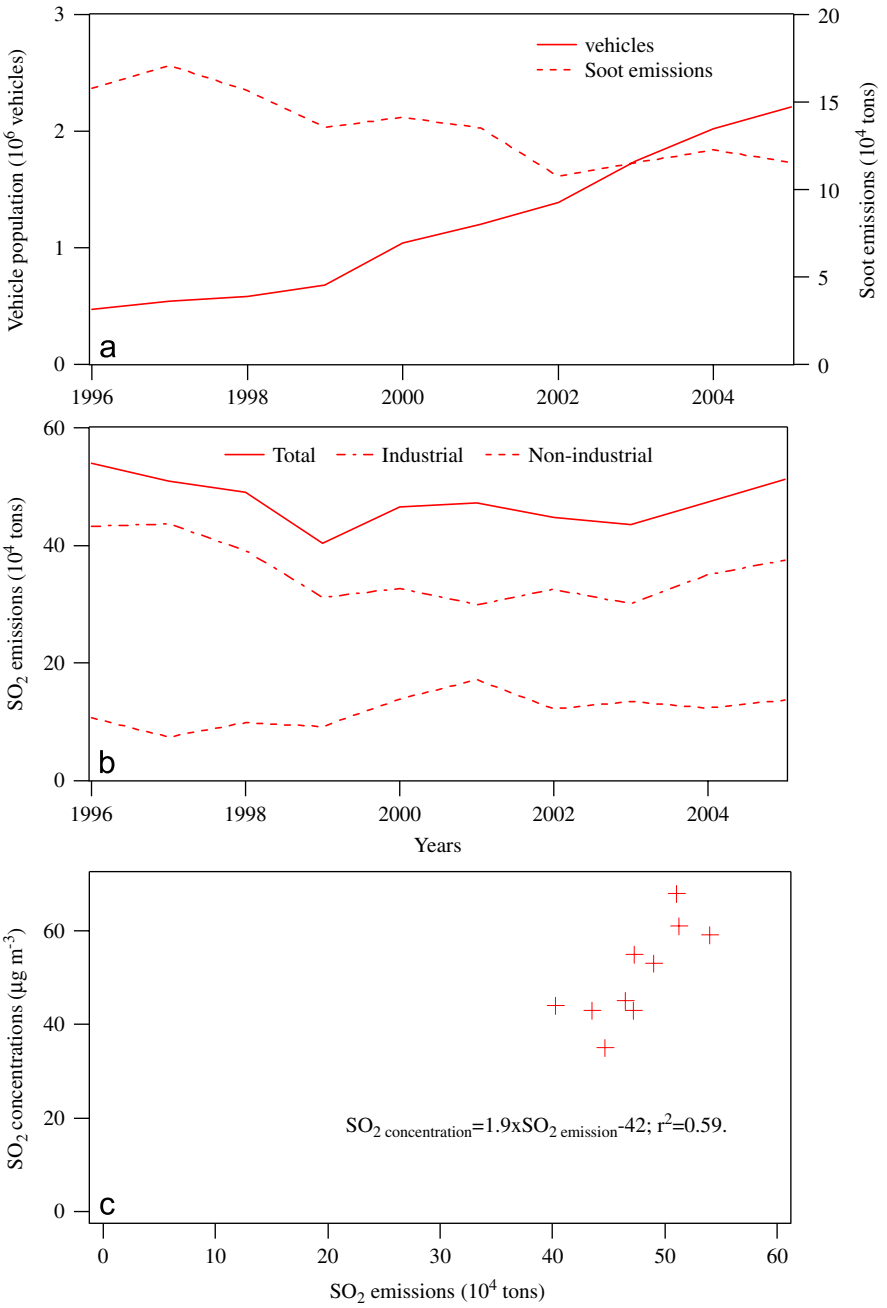

Fig. 5. Annual variations in the vehicular population and SO_2 and soot emissions in Shanghai. Data are from the Shanghai Environmental Bulletin, 1996–2005.

variations of O_3 and other related gases, are not available.

3.2.2. Emissions

As shown in Fig. 5b, the total SO_2 emissions ranged from 4.0×10^5 to 5.4×10^5 tons during 1996–2005 in Shanghai and industrial SO_2 accounted for more than 70% of the emissions. There was no obvious decrease in the SO_2 emissions in the last decade. On the other hand, there has been more than a 40% increase in the non-industrial SO_2 emissions since 2000.

The NO_x emissions increased from 2.8×10^5 tons in 1990 to 4.8×10^5 tons in 1996 and these emissions

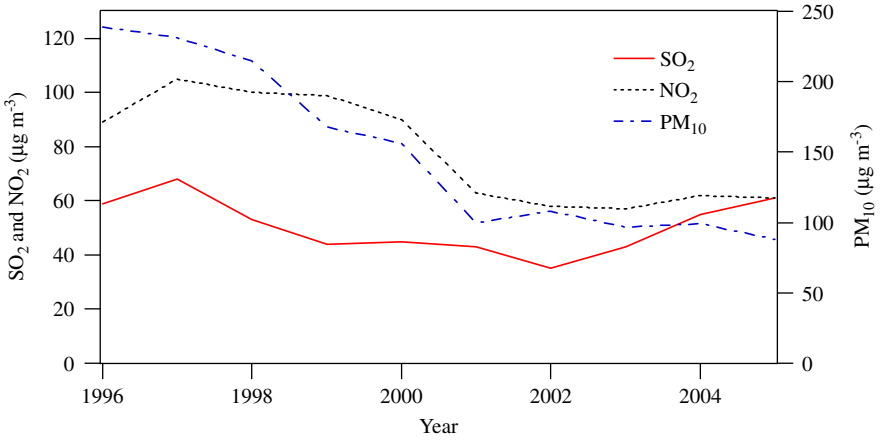

Fig. 6. Annual variations in selected air pollutants concentrations in Shanghai. NO_2 and PM_{10} were not measured before 2001 in Shanghai and NO_x and TSP were used instead of NO_2 and PM_{10} during 1996–2000. Data are collected from the Shanghai Environmental Bulletin, 1996–2005.

remained almost constant between 1996 and 1998 (Hao et al., 2002). Chen C.H. et al. (2006) reported that the NO_x emissions in 2000 were 4.0×10^5 tons, of which there were 1.7×10^5 tons from power plants, 1.0×10^5 tons from industries, 0.9×10^5 tons from transportation and 0.4×10^5 tons from other sources. Incorporating various energy policies, they predicted that NO_x emissions would be 20% higher in 2005 than in 2000 and would continuously increase till 2020, when the NO_x emissions would reach over 6.0×10^5 tons. Cheng et al. (2005) estimated that the annual NO_x, CO, VOC and CO_2 emissions from on-road vehicles were 6.6×10^4, 4.2×10^5, 6.4×10^4 and 7.5×10^6 tons in 2004, respectively.

3.2.3. Ozone formation

Xu and Zhu (1994), Xu et al. (1997) and Xu et al. (1999) studied O_3 formation during 1990–1994 in Shanghai. They found that high O_3 concentrations were associated with the occurrence of high-pressure synoptic systems. They also reported that high O_3 concentrations normally occurred under light wind conditions due to the local photochemical formation of O_3 and were occasionally detected under strong wind conditions attributable to regional transport to Shanghai. In addition, they found that O_3 concentrations increased rapidly on foggy days. Zhao C. et al. (2004) also reported that O_3 episodes in August 1999 in rural areas were mainly due to strong local air pollutant emissions. Tie et al. (2006) developed a global chemical transport model with satellite data to characterize O_3 pollution in eastern China including Shanghai between January 1997 and December 1997. They reported that the daily average O_3 concentration in the summer in eastern China including Shanghai was between 40 and 50 ppb, which was almost double that in the winter. The O_3 concentration and the hydrocarbon concentration in eastern China were 40% less than and only one-fourth of that in the eastern US, respectively. Furthermore, oxidation of CO and hydrocarbon contributed to 54% and 46% of the total O_3 production in eastern China and to 37% and 63% in the eastern US. Tie et al.'s (2006) sensitivity analysis shows that the formation of surface O_3 in eastern China is VOC-limited. Geng et al. (2007) found that the low wind speed caused VOC to accumulate in Shanghai, finally leading to high O_3 concentrations with a maximum value of 80 ppb. On the other hand, the high wind speed facilitated the dispersion of VOC and the resulting maximum value was lower than 30 ppb. They also suggested that aromatics contributed to 70% of O_3 production in Shanghai. A factor that may explain the different percentages of VOC contributions to O_3 formation as reported by Tie et al. (2006) and Geng et al. (2007) is the difference in the air pollutant emissions in 1997 and 2005. In addition, Talbot et al. (2003) and Guttikunda et al. (2005) proposed that the emitted gaseous air pollutants and the O_3 formed in Shanghai could be transported to the western Pacific.

3.3. Particulate pollutants

3.3.1. Emissions

Primary particulate pollutants are important sources of atmospheric particles in Shanghai. As shown in

Fig. 5c, the total emissions of combustion soot particles decreased from 1.58×10^5 tons in 1996 to 1.15×10^5 tons in 2005 in Shanghai. The total emissions of non-combustion particles were only about 15% of the total emissions of soot particles during 2001–2003 (Shanghai Environment Bulletin, 1994–2005). Chen C.H. et al. (2006) reported that the total PM_{10} emissions were 1.24×10^5 tons in 2000 in Shanghai. They predicted PM_{10} emissions of $1.24–1.52 \times 10^5$ tons in 2005 and $1.6–2.43 \times 10^5$ tons in 2020. The lowest emissions of PM_{10} in 2005 predicted by Chen C.H. et al. (2006) were almost the same as the value reported in the Shanghai Environment Bulletin, 2006, assuming that coal consumption is controlled at the level of 4.5×10^7 tons per year.

3.3.2. Annual and spatial variations of TSP, PM_{10} and $PM_{2.5}$

The TSP concentration decreased from $256\,\mu g\,m^{-3}$ in 1996 to $156\,\mu g\,m^{-3}$ in 2000 as shown in Fig. 6 (Shanghai Environmental Bulletin, 1996–2000). Wang Y. et al. (2006b) measured the TSP concentration at two urban sites in Shanghai on selected days from September 2003 to January 2005, and the average TSP concentration was $231\,\mu g\,m^{-3}$, which was 48% larger than that in 2000. From 2001 to 2005, PM_{10}, instead of TSP, was reported to be the major air pollutant on more than 80% of the days in the last 5 years in the Shanghai Environmental Bulletin, 2001–2005. During 2003–2005, the annual average of PM_{10} was below the Grade-II standard. Moreover, Ye et al. (2003) conducted a year-long $PM_{2.5}$ measurement in Shanghai from March 1999 to March 2000 and reported annual averages of $PM_{2.5}$ concentrations of 57.9 and $61.4\,\mu g\,m^{-3}$ at two urban sites. Wang Y. et al. (2006b) measured $PM_{2.5}$ at two urban sites different from those used in Ye et al. (2003) on selected days from September 2003 to January 2005 and the average of the $PM_{2.5}$ concentration was $94.6\,\mu g\,m^{-3}$ and was about 50% higher than that reported by Ye et al. (2003). However, the average PM_{10} concentration during 2003–2005 was only $94.7\,\mu g\,m^{-3}$ (Shanghai Environment Bulletin, 2003–2005). Wang Y. et al. (2006b) also reported that the annual average of the $PM_{2.5}$/TSP mass ratio was 0.41. Zhang Y. et al. (2006) measured the $PM_{2.5}$ and PM_{10} concentrations at nineteen representative urban and suburban sites in Shanghai from December 2002 to January 2003 and reported a $PM_{2.5}/PM_{10}$ mass ratio from 0.32 to 0.85 with an average of 0.6.

3.3.3. Chemical compositions of TSP, PM_{10} and $PM_{2.5}$

Ye et al. (2003) reported that carbonaceous species, secondary species (the sum of SO_4^{2-}, NO_3^-, and NH_4^+), and crustal species accounted for 41.4%, 41.6%, and 9.6% of the $PM_{2.5}$ mass in Shanghai, respectively. Carbonaceous and secondary species are the major contributors to the $PM_{2.5}$ mass and 73% of the carbonaceous species were organic. They also found that the concentrations of different chemical species were almost the same at the two urban sites (about 5 km away from each other). Simultaneous measurements of the carbonaceous species, secondary species and crustal species in atmospheric particles, such as $PM_{2.5}$, PM_{10} and TSP, in Shanghai were not reported in other studies. However, in those studies, the concentrations of ionic species or elements were presented. For example, Wang Y. et al. (2006b) reported that the sum of ionic species including SO_4^{2-}, NO_3^-, Cl^-, F^-, PO_4^{3-}, $HCOO^-$, CH_3COO^-, NO_2^-, MSA, $C_2O_4^{2-}$, NH_4^+, Ca^{2+}, Na^+, K^+, and Mg^{2+} accounted for 26% and 32% of the TSP and $PM_{2.5}$ mass, respectively. The annual average of the SO_4^{2-} and NH_4^+ concentrations in $PM_{2.5}$ were 10.39 and $3.78\,\mu g\,m^{-3}$, respectively. These annual average values are about 50% lower than those reported by Ye et al. (2003), but Ye et al. (2003) and Wang Y. et al. (2006b) reported similar annual averages of NO_3^- concentrations of 6.5 and $6.2\,\mu g\,m^{-3}$, respectively. Based on the comparison of SO_4^{2-}, NO_3^- and NH_4^+ ions in $PM_{2.5}$ measured by Ye et al. (2003) and Wang Y. et al. (2006b), SO_4^{2-} and NH_4^+ concentrations in $PM_{2.5}$ in Shanghai decreased by about 50% during 2003–2005 compared to 1999–2000. On the other hand, the SO_2 concentration in Shanghai during 2003–2005 was about 20% higher than that during 1999–2000. It is not clear why there was an increase in the SO_2 concentration but a simultaneous decrease in the SO_4^{2-} and NH_4^+ concentration in $PM_{2.5}$ during 2003–2005.

OC accounted for about 30% of the total mass of $PM_{2.5}$ in Shanghai (Ye et al., 2003). CMB analysis of $PM_{2.5}$ using extractable organic compounds as tracers shows that about 50% of the organic carbon was from engine exhaust and about 15% was from coal burning (Feng et al., 2006b). The contributions of kitchen emissions and biomass burning to organic carbon in $PM_{2.5}$ were estimated to be 7–9% and 1–5%, respectively (Feng et al., 2006b). In addition, Wang J. et al. (2000), Mukai et al. (2001), Li X. et al. (2005), and Xiu et al. (2005) studied Pb and mercury in Shanghai, and their concentrations were

determined to be at safe levels. The annual averages of As, Cd and Ni concentrations in PM_{10} in Shanghai are not available. Zheng J. et al. (2004) made short-term measurements from November 2001 to January 2002 at seven sites in Shanghai and reported the mean concentrations of As, Cd, Ni and Mn were 42.1, 10.9, 13.9 and 186 ng m^{-3}, respectively. Shu et al. (2001) investigated magnetic properties of atmospheric particles in Shanghai.

3.3.4. Formation of secondary particulate species

Several studies have discussed the formation of secondary particulate species in Shanghai. Wang Y. et al. (2006b) ascribed the SO_4^{2-} formation in Shanghai mainly to heterogeneous reactions based on the SOR (SOR = $[SO_4^{2-}]/([SO_4^{2-}]+[SO_2])$) larger than 0.1. The shortcoming of this approach has been mentioned earlier. Yao et al. (2002a) proposed that local SO_2 emissions were an important source of SO_4^{2-} in $PM_{2.5}$ in Shanghai because the SO_4^{2-} concentration was correlated with the SO_2 concentration ($r = 0.66$). They also found a relatively stable SO_4^{2-}/SO_2 mass ratio over a large range of temperatures, suggesting that gas-phase oxidation of SO_2 played a minor role in the formation of SO_4^{2-}. The sum of SO_4^{2-} and NO_3^- was highly correlated with NH_4^+ ($r = 0.96$) and SO_4^{2-} was incompletely neutralized due to insufficient NH_4^+. Tong et al. (2001) reported high Fe^{3+} concentrations (18–27 μg m^{-3}) in Shanghai and proposed that heterogeneous formation of SO_4^{2-} via Fe^{3+} catalyst reactions could be important in Shanghai.

Xiu et al. (2004) investigated the size distribution of water-soluble ions in Shanghai in 2000. They found that SO_4^{2-} exhibited a bimodal size distribution with a dominant mode at 0.3–0.7 μm and a minor mode at 5 μm. NO_3^- also exhibited a bimodal size distribution but the dominant mode could occur at 5 μm. These size distributions are similar to those observed in Hong Kong, another coastal city to be discussed later (Zhuang et al., 1999a, b). Xiu et al. (2004) found that SO_4^{2-} aerosols in Shanghai were almost completely neutralized, contrary to results reported by Yao et al. (2002a) and Wang Y. et al. (2006b).

4. Air pollution in the Pearl River Delta (PRD) region

4.1. Topography and meteorology

Fig. 1 shows that the PRD region is situated at 21°17′–23°56′N and 111°59′–115°25′E and is connected to the South China Sea. The PRD region covers 4.17×10^4 km^2. Most of the PRD region is located in Guangdong Province (People's Government of Guangdong Province, 2004) while Hong Kong and Macao are governed by the Government of the Hong Kong Special Administrative Region (HKSAR) and the Government of the Macao Special Administrative Region, respectively. The PRD region mainly consists of floodplains, except in Hong Kong, where more than 50% of the land is mountainous area. The PRD region has a subtropical marine monsoon climate. The rainfall is about 1800 mm per year, but it varies from year to year (Guangdong Statistical Yearbook 1997–2005, 1998–2006; http://www.weather.gov.hk/wxinfo/pastwx). Similar to the case in Shanghai, rainfall is a major pathway to remove air pollutants in the PRD region (Wai and Tanner, 2005a, b). The annual variation of rainfall can greatly affect air quality in the whole PRD region as will be discussed later. RH and cloud coverage are high in the PRD region, e.g., the average RH and cloud coverage in Hong Kong in 2005 were 79% and 71%, respectively (http://www.weather.gov.hk/wxinfo/pastwx). High RH and cloud coverage have been proposed to promote secondary formation of particulate species in Hong Kong (Zhuang et al., 1999a, b; Yao et al., 2002b, 2003c; Yu J.Z. et al., 2004a, 2005; Huang et al., 2006), and the same can be said of the whole PRD region. The number of foggy days (with visibility less than 1 km) has gradually increased in the PRD region. For example, the number in Shenzhen was 25 days in the 1960s, 54 days in the 1970s, 79 days in the 1980s, 177 days in the 1990s and 162 days in 2006. Fog processing is expected to play an important role in the formation and transformation of air pollutants. However, there are very few studies on the relationship between fog processes and air pollution in the PRD region (Wu et al., 2006).

The prevailing wind in the PRD region is from the northeast in the winter and from the southeast, south and southwest in the summer. The annual wind speed in the coastal cities, e.g., about 3 m s^{-1} in Shenzhen and Hong Kong, is higher than that in the inland cities, e.g., about 1.5 m s^{-1} in Guangzhou and Foshan (Guangdong Statistical Yearbook 1997–2005, 1998–2006; http://www.weather.gov.hk/wxinfo/pastwx; http://www.weather.gov.hk/contente.htm). The lower wind speed in the inland cities in the PRD region favors the accumulation of air pollutants and exacerbates air pollution

problems (Environmental Protection Plan in the Pearl Delta River Region, 2004).

4.2. Urbanization

The PRD region had a permanent population of 49 million and a large number of migrants in 2004 (Guangdong Statistical Yearbook 1997–2005, 1998–2006). Three mega cities, i.e., Guangzhou, Shenzhen and Hong Kong are situated at the PRD region and they are concentrated in a territory about 150 km in length and 100 km in width. The PRD region, especially the areas between the three mega cities, has undergone rapid urbanization and many smaller towns (e.g., Dongguan) with populations of about one million each have been created in the last two decades. The air quality in those cities and towns has been deteriorating in the last decade. SO_2 and NO_2 concentrations in Dongguan in 2002 exceeded those in Guangzhou and Shenzhen (Environmental Protection Plan in the Pearl Delta River Region, 2004). When the PRD region experiences stagnant synoptic conditions, air pollutants emitted from the local sources in these mega cities, cities and towns accumulate, leading to unique regional air pollution problems across the whole PRD region (Hagler et al., 2006). When regional air pollution occurs, several hot spots with high AOT values can be found together with regions with low AOT values at the borders between cities and/or towns (Wu et al., 2005; Li C.C. et al., 2005).

The PRD region is one of the most developed areas in China and its rapid economical growth has resulted in an increase in air pollutant emissions (Environmental Protection Plan in the Pearl Delta River Region, 2004). The per capita GDP in the PRD region excluding Hong Kong (HK) and Macao grew at a rate of about 15% annually and was more than US$5000 in 2006 (Guangdong Statistical Yearbook, 2006). The increase in air pollutant emissions and urbanization in the PRD region have caused non-compliance with the Grade-II standards in urban areas as well as regional air pollution problems such as increases in the number of O_3 episodes in suburban and rural areas, visibility deterioration, and an increase in the frequency of acid rain as will be discussed later. It should be noted that HK has its own air quality standards while other PRD areas discussed in this paper follow the National Ambient Air Quality Standards shown in Table 1.

4.3. Gaseous pollutants

4.3.1. Annual and spatial variations

As shown in Fig. 7, the annual average SO_2 concentration in Guangzhou increased from 45 in 1999 to 77 $\mu g\,m^{-3}$ in 2005. The annual averages of SO_2 in Shenzhen, at Tsuen Wan in Hong Kong (urban area) and at Tung Chung in Hong Kong were lower but similar concentration levels, from 18 to 27 $\mu g\,m^{-3}$ during 1999–2005. The annual averages in 2000–2001 were 10–40% higher than the averages during 2002–2005 in Shenzhen while the annual average gradually increased during 1999–2005 in Hong Kong. Using the STEM-2K1 chemical transport model, Wang X.M. et al. (2005) estimated that 32.9% of SO_2 concentrations across the whole PRD region including Hong Kong was due to the power plant emissions. Wang S.-L. et al. (2005) estimated that only about 40% of SO_2 concentration in Guangzhou and Shenzhen in 2002 was from local emissions.

The annual average SO_2 concentration at Tap Mun, a background site in Hong Kong, has gradually increased from 9 $\mu g\,m^{-3}$ in 1999 to 14 $\mu g\,m^{-3}$ in 2005. Some studies have suggested that the regional transport of SO_2 was mainly responsible for the episodes with high SO_2 concentrations and high SO_2/NO_y or SO_2/NO_x ratios in Hong Kong (So and Wang, 2003; Wang T. et al., 2003a, b). However, since power plant plumes in Hong Kong can be transported to Tap Mun (Yao et al., 2005), the increase in SO_2 at Tap Mun is possibly from a combination of regional transport and increasing contributions from local power plants.

During 1999–2005, the annual average NO_2 concentration in Guangzhou was 68 $\mu g\,m^{-3}$ $\pm$10%, followed by 64 $\mu g\,m^{-3}$ $\pm$5% at Tsuen Wan in Hong Kong, 54 $\mu g\,m^{-3}$ $\pm$30% in Shenzhen, 46 $\mu g\,m^{-3}$ $\pm$30% at Tung Chung in Hong Kong and 13 $\mu g\,m^{-3}$ $\pm$20% at Tap Mun in Hong Kong. Large spatial variations of NO_2 in the PRD region suggest that NO_2 is mainly from the local sources. The vehicular population in Guangzhou and Shenzhen has increased by about 10% per year over the last decade (Guangzhou Statistical Yearbook 1998–2005, 1999–2006; Shenzhen Statistical Yearbook 1998–2005, 1999–2006). The rapid development of high-rise buildings in these two mega cities hindered the dispersion of street-level air pollutants (Chan et al., 1995; Xie et al., 2003). Since the annual average NO_2 concentrations exceeded the Grade-II standard in Guangzhou and Shenzhen, tighter

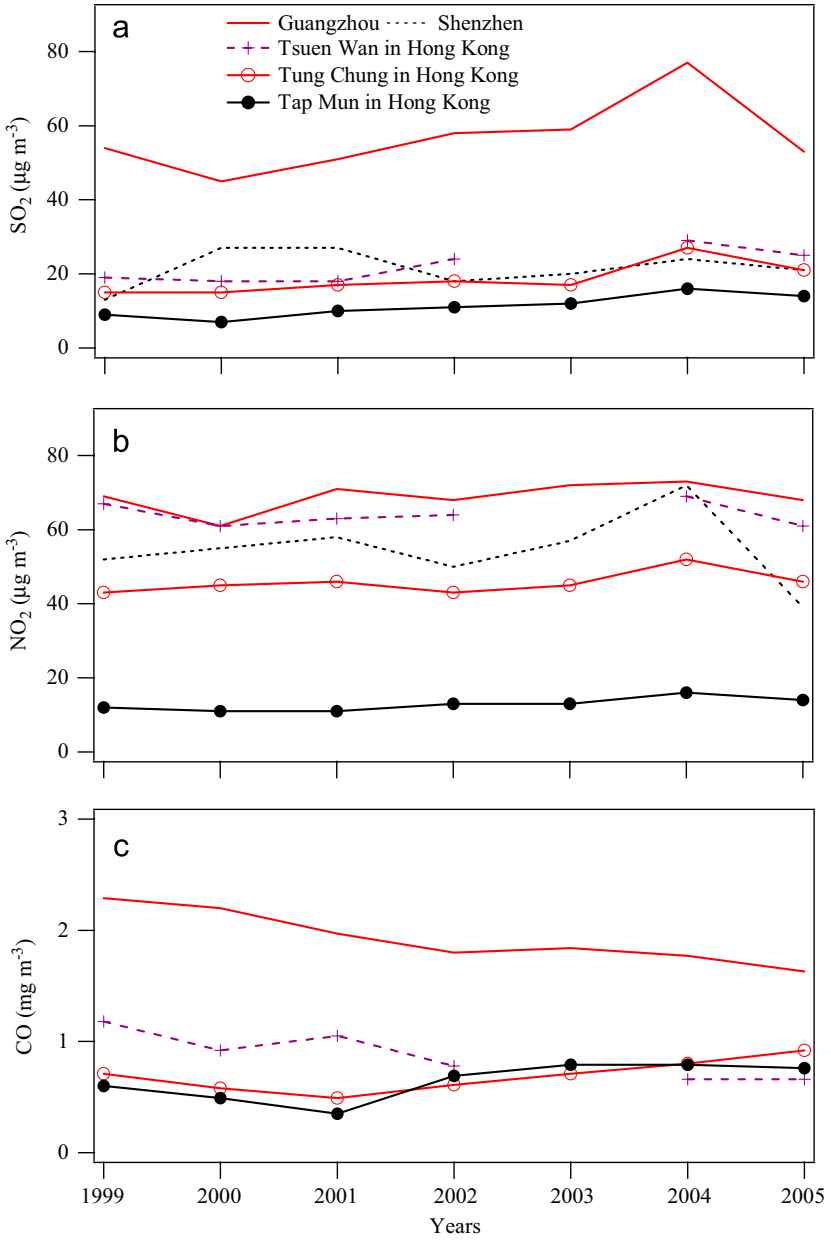

Fig. 7. (a) SO_2, (b) NO_2 and (c) CO concentrations in the PRD region during 1999–2005. Data are from the Guangzhou Environmental Bulletin, Shenzhen Environmental Bulletin, and Air Quality in Hong Kong.

control on NO_x emissions is needed. The same can be said for the control of NO_2 pollution in urban areas and new towns in Hong Kong.

The CO concentration in Guangzhou decreased from 2.3 mg m^{-3} in 1999 to 1.6 mg m^{-3} in 2005. A similar decrease at much lower concentrations also occurred at Tsuen Wan, but not at other sites in Hong Kong. However, the CO concentration is clearly within the safe level in these mega cities.

O_3 concentrations were not reported in the Guangzhou Environmental Bulletin (1994–2005) and the Shenzhen Environmental Bulletin. In Hong Kong, as shown in Fig. 8a, there was no increase in the annual averages of O_3 and ($NO_2 + O_3$) concentrations in the three sites (an urban site, a new town site, and a rural site), except in 2004, when HK had very low rainfall and other pollutants also showed significant increases in concentrations. In addition,

the annual averages of (NO_2+O_3) concentrations at the three sites in Hong Kong were close to each other, suggesting that (NO_2+O_3) is mainly from regional sources. However, the conclusion derived for the annual average (NO_2+O_3) concentration may not be applicable to O_3 episodes, which occasionally occurred in the PRD region when there was stagnant weather. For example, Yao et al. (2005) reported large spatial variations of (NO_2+O_3) concentrations in some O_3 episodes in Hong Kong attributable to the influence of the local power plant plumes in the ground-level vicinity. O_3 episodes with spatially uniform (NO_2+O_3) concentrations are likely due to regional contributions.

4.3.2. *Ozone formation*

O_3 pollution has drawn much attention in Hong Kong in the last decade (Kok et al., 1997; Chan et al., 1998; Wang T. et al., 1998, 2001a, b, 2003a, b, 2005; Lam et al., 2001; Lee Y.C. et al., 2002; Liu and Chan, 2002; So and Wang, 2003; Ding et al., 2004; Huang J.P. et al., 2005; Lam et al., 2005; Yao et al., 2005; Zhang J. et al., 2006). However, O_3 pollution in other PRD areas was much less reported until recently (Shao et al., 2004; Wang X.M. et al., 2005; Deng et al., 2006). In Hong Kong, the high O_3 concentrations usually occurred in the fall and the spring (Chan et al., 1998; Wang T. et al., 1998, 2003a; Air Quality in Hong Kong, 1999–2005). For example, Lee Y. C. et al. (2002) reported that there were only six O_3 episode days in the summer during 1994–1999 in Hong Kong, and the maximum O_3 concentration for the period was 334 $\mu g\,m^{-3}$ recorded in August 1999. However, Huang J.P. et al. (2005) reported that the number of O_3 episode days was 7 in 1999 and 2000, 13 in 2001, and 18 in 2002 and 2003. The number increased to 37 days in 2004 (Air Quality in Hong Kong, 2004). These episodes were classified into three synoptic patterns, i.e., cyclonic, anticyclonic, and trough (Chan et al., 1998; Chan and Chan, 2000; Huang J.P. et al., 2005). The O_3 episodes related to tropical cyclones, continental anticyclones and the low-pressure troughs accounted for 62%, 21% and 17% of the total episodes, respectively, in 1999–2003 (Huang J.P. et al., 2005).

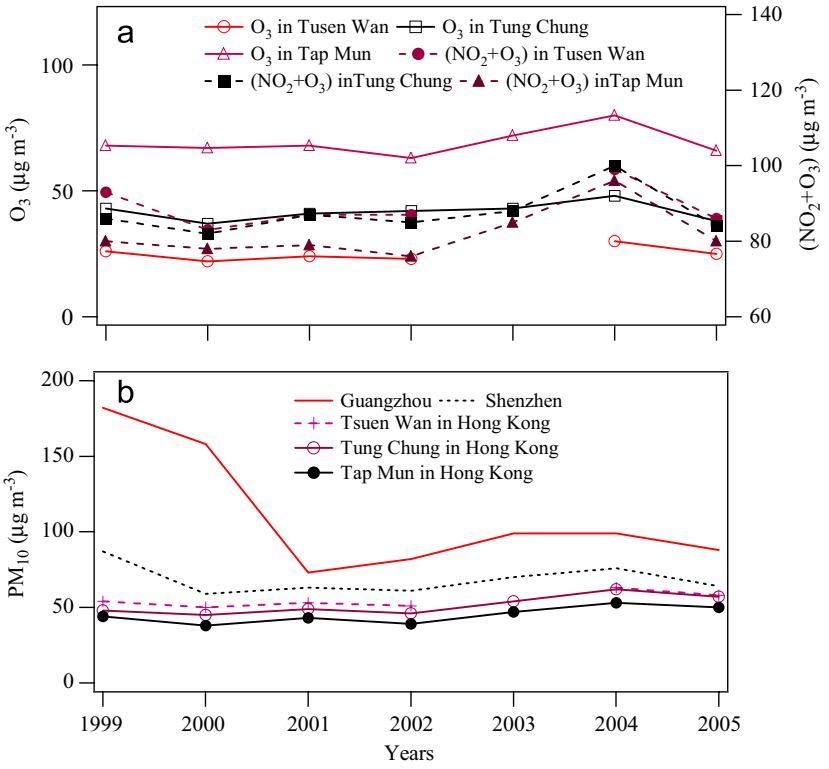

Fig. 8. (a) O_3, (NO_2+O_3) and PM_{10} concentrations in the PRD region during 1999–2005. Data are from the Guangzhou Environmental Bulletin, Shenzhen Environmental Bulletin, and Air Quality in Hong Kong and (b) TSP concentrations in 1999 and 2000 were used for Guangzhou.

The photochemical formation of O_3 in Hong Kong was generally believed to be VOC-limited (Huang J.P. et al., 2005; Lam et al., 2005; Zhang J. et al., 2006). Zhang J. et al. (2006) proposed that anthropogenic VOC, mainly the reactive aromatics, dominated the reactivity of VOC and HONO plays a critical role in the O_3 formation in Hong Kong.

The local versus regional contributions to O_3 episodes in Hong Kong have been widely discussed and the results are conflicting so far (Chan et al., 1998; Wang T. et al., 1998, 2001a, b, 2003a, b; Chan and Chan, 2000; Lam et al., 2001; Lee Y.C. et al., 2002; Liu and Chan, 2002; Ding et al., 2004; Huang J.P. et al., 2005; Lam et al., 2005; Zhang J. et al., 2006). For example, Lee Y.C. et al. (2002) and Liu and Chan (2002) reported that the emissions from on-road vehicles and power plants in Hong Kong were mainly responsible for the O_3 episodes in Hong Kong during 1994–1999, and they also found several convergence zones in Hong Kong created under the calm synoptic conditions. Kok et al. (1997) and Yao et al. (2005) observed high O_3 concentrations (larger than $240\,\mu g\,m^{-3}$) in the power plant plumes at high elevations and the ground-level vicinity downwind of the power plants in Hong Kong, respectively. On the other hand, Ding et al. (2004), Lam et al. (2005) and Huang J.P. et al. (2005) concluded that O_3 episodes during 1999–2003 were mainly due to the regional contributions from outside of Hong Kong. In addition, Zhang J. et al. (2006) reported that about half of O_3 episodes in Hong Kong was mainly caused by the regional transport and the other half was mainly due to the local emissions of air pollutants. The $(NO_2 + O_3)$ concentrations are not discussed in most studies, except by Yao et al. (2005).

The limited studies on O_3 pollution in the PRD region outside of Hong Kong show that, in a rural area of Guangzhou (Xinken) during April to December in 2000, 13.4% of the days had O_3 episodes (Environmental Protection Plan in the Pearl River Delta Region, 2004). The same report also shows that the maximum hourly O_3 concentration in Guangzhou, Foshan, Zhongshan, and Shenzhen in 2000 varied from 300 to $370\,\mu g\,m^{-3}$. In addition, Wang X.M. et al. (2005) reported that the transport sources only contributed 17.8% of the O_3 concentration for the whole region in March 2001. They also found that the O_3 formation in urban areas was VOC-limited and that in the non-urban area was NO_x-limited. Deng et al. (2006) reported that high aerosol concentrations can reduce surface actinic flux up to 70–80% in the PRD region, thus greatly inhibiting the O_3 formation in the PRD region. Heterogeneous reactions between O_3 and its precursors with aerosol particles can inhibit O_3 formation as reported in the literature (Jacob, 2000; Tie et al., 2003). There is no study on the inhibiting mechanism in the PRD region.

Moreover, there are dozens of studies on the precursors of O_3, e.g., CO, VOC and NO_2, and other chemical species related to photochemical formation of O_3 in the PRD region (e.g., Kok et al., 1997; Chung et al., 1999; Lam et al., 1999; Lind and Kok, 1999; Cheng and Lam, 2000; Sin et al., 2001; Chan C.Y. et al., 2002; Chan L.Y. et al., 2002; Ho et al., 2002, 2004; Lee, S.C. et al., 2002; Wang X.M. et al., 2002; Chan et al., 2003, 2004, 2006; Lee and Hills, 2003; Xie et al., 2003; Guo H. et al., 2004; Zhao L. et al., 2004; Chan and Ning, 2005; Richter et al., 2005; Cheng et al., 2006a; Guo et al., 2006; Simpson et al., 2006). In these studies, the characteristics, source apportionments of these gaseous species at various sites were presented and emission factors of these gaseous species from on-road vehicles were estimated.

4.3.3. Emissions

The annual SO_2 emissions in Guangzhou exhibit a decreasing trend during 2000–2005 and it was 14.9×10^4 tons in 2005. In Shenzhen, they jumped from 2.90×10^4 tons in 1999 to 3.84×10^4 tons in 2000 and the emissions slightly increased by about 10% during 2000–2005. In Hong Kong, they slightly increased from 5.65×10^4 tons in 1999 to 6.76×10^4 tons in 2002. From 2003, they increased by about 40% and reached 9.48×10^4 tons in 2004, due to the increased emissions from public utilities.

The annual emissions of NO_x, CO, and VOC are not reported in Guangzhou and Shenzhen regularly. In Hong Kong, the annual NO_x emissions varied from 8.65 to 10.2×10^4 tons during 1999–2005 (http://www.epd.gov.hk/epd/english/environmentinhk/air/data/emission_inve.html). The high NO_x emissions in 2003 (10.2×10^4 tons) were the result of increasing NO_x emissions from public electricity generation, which accounted for about 50% of the total NO_x emissions in Hong Kong. NO_x emissions from road transport and marine traffic accounted for about 30% of the total emissions. During 1999–2005, the decrease in the emissions of NO_x from road transport was almost the same as the increase in NO_x emissions from marine traffic.

In Hong Kong, more than 90% of CO emissions was from road transport. The emissions decreased from 11×10^4 tons in 1999 to 7.58×10^4 tons in 2005. About 80% non-methane VOC (NMVOC) emissions was from consumer products and paint VOC and more than 10% was from road transport. The total emissions of NMVOC gradually decreased from 4.76×10^4 tons in 1999 to 4.02×10^4 tons in 2005.

The SO_2 emissions in the overall PRD region including Guangzhou, Shenzhen, Zhuhai, Dongguan, Zhongshan, Foshan, Jiangmen, Huizhou and Zhaoqing, but not Hong Kong and Macao, were 51.9×10^4 tons in 1997 and they accounted for 75% of the total SO_2 emissions in Guangdong province (http://www.epd.gov.hk/epd/english/environmentinhk/air/studyrpts/study_pearl.html). Power generation, mineral extraction, and manufacturing contributed to 41.4%, 17.2%, and 10.7% to the total SO_2 emissions, respectively. The NO_x emissions were 45.0×10^4 tons in 1997, and motor vehicle and power generation contributed 31.3% and 30.6%, respectively. The VOC emissions were 41.1×10^4

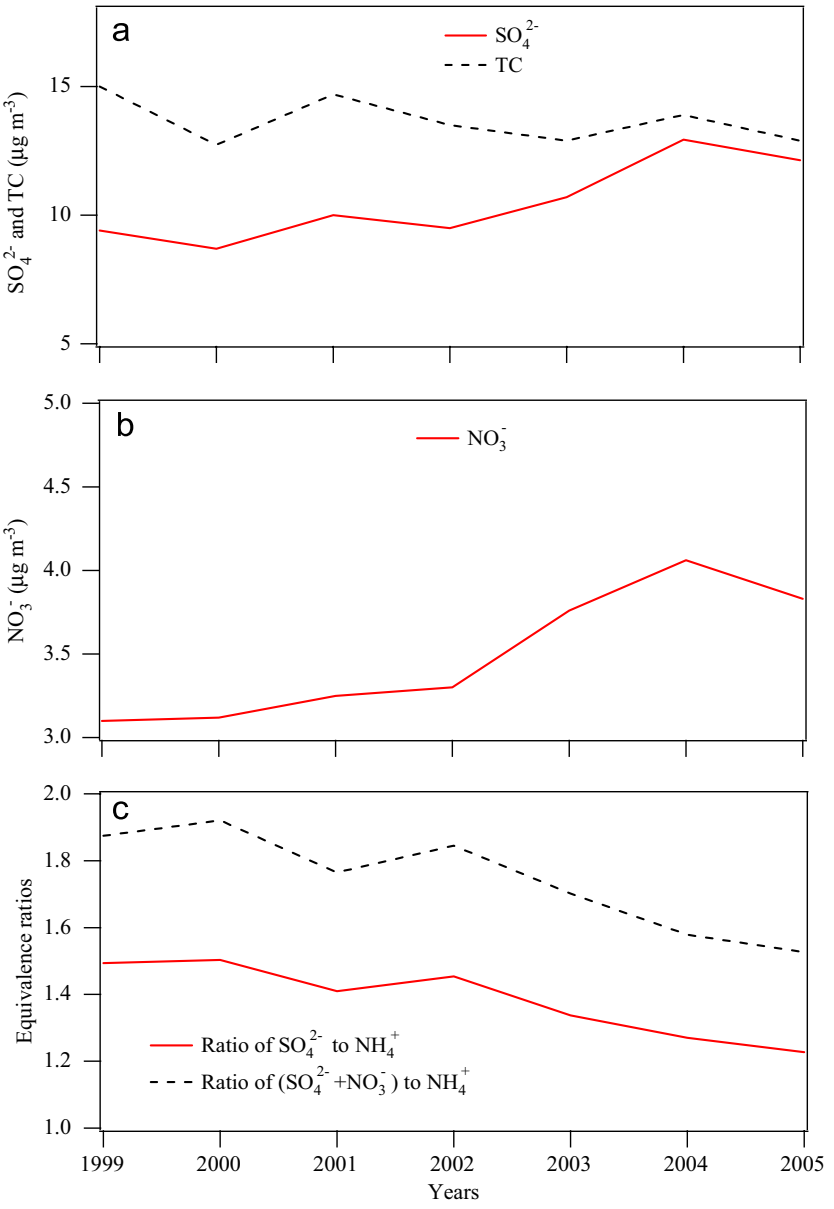

Fig. 9. Selected chemical compositions in PM_{10} in Hong Kong during 1999–2005. Data are from Air Quality in Hong Kong.

tons in 1997. Motor vehicles and paint were two major contributors, which accounted for 58.5% and 13.8% of the total VOC emissions, respectively.

4.4. Particulate pollutants

4.4.1. Chemical compositions of PM_{10} and $PM_{2.5}$

As shown in Fig. 8b, the annual average PM_{10} concentration in Guangzhou ranged from 73 to 99 $\mu g\,m^{-3}$ while the annual average in Shenzhen ranged from 59 to 76 $\mu g\,m^{-3}$ after 2000. In the two cities, the annual averages were below the Grade-II standard without a significant increase during 1999–2005. However, the annual average PM_{10} concentrations in Hong Kong during 2003–2005 were about 20% higher than those before 2003. Moreover, some PM_{10} episodes (with daily average PM_{10} concentrations higher than 180 $\mu g\,m^{-3}$) were occasionally observed in Hong Kong and were associated with anticyclonic and cyclonic synoptic systems over South China (Lee and Hills, 2003). The anticyclonic and cyclonic synoptic systems usually resulted in stagnant air conditions over the PRD region as discussed earlier.

The chemical compositions in PM_{10} are generally not available in Guangzhou and Shenzhen. The annual averages of chemical compositions in PM_{10} in Hong Kong are reported in annual reports on Air Quality in Hong Kong and some are shown in Fig. 9. The average of all sampling sites was used since the chemical compositions at each site in Hong Kong were measured every sixth day. In PM_{10}, SO_4^{2-} increased by about 20% during 2004–2005 and NO_3^- increased by about 30% during 2003–2005. The contribution of the sum of SO_4^{2-}, NO_3^-, and NH_4^+ to the total PM_{10} mass gradually increased from 25% in 1999 to 33% in 2005. However, the total organic contribution to PM_{10} exhibited a slight decrease from 25% in 1999 to 22% in 2005. Adding an estimated amount of about 4 $\mu g\,m^{-3}$ of EC in PM_{10} during 1998–2001 (Yu J.Z. et al., 2004b), the sum of OC, EC, SO_4^{2-}, NO_3^- and NH_4^+ accounted for about 60% of the total mass of PM_{10} in Hong Kong.

It is interesting that the equivalence ratios of $[SO_4^{2-}]$ to $[NH_4^+]$ and $([SO_4^{2-}]+[NO_3^-])$ to $[NH_4^+]$ in Hong Kong were larger than 1.0 as shown in Fig. 9, suggesting that SO_4^{2-} aerosols are acidic. However, the ratios of $[SO_4^{2-}]$ to $[NH_4^+]$ and $([SO_4^{2-}]+[NO_3^-])$ to $[NH_4^+]$ gradually decreased during 1999–2005 and were 1.2 and 1.5 in 2005, respectively. It appears that SO_4^{2-} aerosols in Hong Kong are becoming less acidic. Sampling artifacts of particulate NO_3^-, chloride and NH_4^+ could be significant in Hong Kong and can affect the measured aerosol acidity (Pathak et al., 2003, 2004a, b; Pathak and Chan, 2005). There are no corrections for sampling artifacts in the PM_{10} measurements in the Hong Kong Environmental Protection Department (HKEPD) dataset.

The chemical compositions in PM_{10} collected at the HKEPD stations have been widely used to study their sources (Qin et al., 1997; Lee et al., 1999; Cao et al., 2003a; Pathak et al., 2003; Wai and Tanner, 2004, 2005a, b; Yuan et al., 2006a). In these studies, four major sources, i.e., secondary aerosols, aged and fresh sea-salt, crustal aerosol and vehicular exhaust, were identified and the sum of these sources accounted for more than 60% of the mass of PM_{10}. The estimated regional contribution to PM_{10} collected at HKEPD stations varied from about 40% to more than 60% in various studies due to the different approaches used. Some short-term measurements of PM_{10} in Hong Kong were also reported (e.g., Ho et al., 2003, 2006a, b) and the chemical compositions were in the range of the PM_{10} database collected by HKEPD. Unlike in Beijing, crustal species only accounted for less than 10% of the PM_{10} mass in Hong Kong (Ho et al., 2006a, b).

Several short-term $PM_{2.5}$ measurements in Hong Kong have been reported (Ho et al., 2003, 2006a, b; Pathak et al., 2004a, b; Cheng et al., 2006a, b; Lee et al., 2006; Wang G. et al., 2006). HKEPD launched a more long-term campaign to collect 24-h $PM_{2.5}$ samples every sixth day from 6 November 2000 to 26 October 2001 at three sites including a roadside site in Mong Kok, an urban site in Tseun Wan, and a rural site in Hok Tsui (Louie et al., 2005a, b). In the HKEPD's $PM_{2.5}$ database, the annual average $PM_{2.5}$ concentrations on the roadside and at urban and rural sites were 58.1, 34.1 and 23.7 $\mu g\,m^{-3}$, respectively. At the roadside and urban sites, carbonaceous aerosols accounted for 52–75% of the $PM_{2.5}$ mass, followed by "ammonium sulfate" (assuming complete neutralization of SO_4^{2-} aerosols) accounting for 23–37% of the $PM_{2.5}$ mass. At the background site, ammonium sulfate was the largest contributor to the $PM_{2.5}$ mass, accounting for 51% of the $PM_{2.5}$ mass, while the carbonaceous aerosols only accounted for 32% of the $PM_{2.5}$ mass. Louie et al. (2005a, b) also found that ammonium sulfate

and crustal concentrations showed more uniform spatial distributions than did carbonaceous aerosols, suggesting a larger regional contribution to the former species and a smaller regional contribution to the latter. The chemical compositions in the short-term $PM_{2.5}$ measurements in Hong Kong were generally within the range of HKEPD's $PM_{2.5}$ database.

In the PRD region, Hagler et al. (2006) made measurements of $PM_{2.5}$ at seven sites across Guangzhou, Shenzhen, and Hong Kong during October and December 2002 and March and June 2003. The average $PM_{2.5}$ concentrations ranged from 37 to 71 $\mu g\,m^{-3}$ in Guangzhou and Shenzhen and from 29 to 34 $\mu g\,m^{-3}$ in Hong Kong. In Guangzhou and Shenzhen, carbonaceous aerosols and inorganic secondary aerosols (the sum of SO_4^{2-}, NH_4^+, and NO_3^-) accounted for 37–41% and 33–37% of the $PM_{2.5}$ mass, respectively. In Hong Kong, they reported that the contribution of carbonaceous aerosols and secondary inorganic aerosol to the $PM_{2.5}$ mass were 27–33% and 37–42%, respectively. They found that $PM_{2.5}$ SO_4^{2-} concentrations are usually spatially uniform in the PRD. They also proposed that sources outside of the PRD region contribute a significant fraction of overall $PM_{2.5}$ in Guangzhou, Shenzhen and Hong Kong, although they did not quantify the contributions. The regional SO_4^{2-} distributions over the PRD are also frequently observed from the space as shown in http://www.nrlmry.navy.mil/flambe-bin/aerosol/display_directory_aer2?DIR=/web/aerosol/public_html/globaer/ops_01/mongolia/ (http://www.nrlmry.navy.mil).

4.4.2. *Carbonaceous species in $PM_{2.5}$ and PM_{10}*

Since carbonaceous and secondary inorganic aerosols are the two major contributors to PM_{10} and $PM_{2.5}$ in the PRD region, the primary sources of carbonaceous aerosols and secondary formation of organic and inorganic species were widely investigated in the PRD region (Zheng M. et al., 1997, 2000; Fang et al., 1999; Zhuang et al., 1999a, b, Yao et al., 2002b, 2003c; Cao et al., 2003b, 2004; Ho et al., 2004, 2006b; Yu J.Z. et al., 2004a, b; Louie et al., 2005a, b; Feng et al., 2006a; Hagler et al., 2006). Cao et al. (2003b, 2004) reported that about 80% of the OC in PM_{10} was found in $PM_{2.5}$ in Guangzhou. They also reported that the concentrations of OC and EC in $PM_{2.5}$ in Guangzhou were 22.6 and 8.3 $\mu g\,m^{-3}$ in the winter of 2002, and 15.8 and 5.9 $\mu g\,m^{-3}$ in the summer of 2002, respectively. Feng et al. (2006a) reported a similar seasonal trend of OC and EC concentrations with $\pm 30\%$ of those reported by Cao et al. (2003b, 2004).

In Hong Kong, the annual averages of OC and EC were 8.67 and 4.64 $\mu g\,m^{-3}$ in PM_{10} during 1998–2001, without an obvious increase (Yu J.Z. et al., 2004b). Louie et al. (2005a, b) reported OC and EC values of 8.18 and 5.25 $\mu g\,m^{-3}$, respectively, for November 2000 to October 2001 in $PM_{2.5}$. Cao et al. (2003b, 2004) reported similar results in 2002, within 10% of Louie et al.'s findings (2005a, b). In addition, Zheng M. et al. (2006) estimated that diesel engine exhaust contributed $57 \pm 13\%$ at urban sites and $25 \pm 2\%$ at the rural site to the total OC concentrations in $PM_{2.5}$. They reported that primary sources can explain 49%, 79%, and 94% of the total OC concentrations in $PM_{2.5}$ at the rural, the urban, and the roadside sites, respectively, and they attributed the unexplained OC concentrations to the secondary formation of organics. Cao et al. (2004) and Yuan et al. (2006b) estimated that secondary organic carbon constituted about 50% of the total OC in $PM_{2.5}$ in Hong Kong. Yu J.Z. et al. (2004b) reported that 27–46% of the total carbon at a coastal site in Hong Kong was water-soluble organic compound (WSOC), while 23–28% of the total carbon (TC) in Guangzhou was WSOC (Feng et al., 2006a). Moreover, the EC concentrations at urban sites were more than 70% higher than those at the background site in Hong Kong, suggesting the predominant local contribution of EC at urban sites (Cao et al., 2003b, 2004; Louie et al., 2005a, b; Hagler et al., 2006).

Detailed analyses of the compositions of organic matter have been conducted mainly in Hong Kong and Guangzhou (Zheng M. et al., 1997, 2000; Fang et al., 1999; Bi et al., 2002, 2003, 2005; Louie and Sin, 2003; Sin et al., 2005; Chan et al., 2006; Feng et al., 2006a; Tan et al., 2006; Li J. et al., 2006; Liu G. et al., 2006). In those studies, solvent extractable organic compounds identified and quantified in the particulates included *n*-alkanes, polycyclic aromatic hydrocarbons, *n*-fatty acids, *n*-alkanols and dicarboxylic acids but they only accounted for less than 10% of the total organic matter. Source apportionment analyses were conducted in some studies.

Overall, the concentration of organic carbon was 2–3 times that of EC. The ratio of the secondary OC to the total OC varied from less than 10% to more than 50% in the PRD, depending on the location.

4.4.3. Formation of secondary particulate species

Gas-phase photochemical reactions followed by gas-to-particle condensation have been suggested to be one of important routes for the formation of secondary particulate species in Hong Kong (Sequeira, 2002; Ho et al., 2006b). On the other hand, many studies suggest that in-cloud processes play an important role in the formation of secondary particulate species in Hong Kong (Zhuang et al., 1999a, b; Yao et al., 2002b, 2003c, 2004; Yu J.Z. et al., 2004b, 2005; Huang X.F. et al., 2006). In these studies, SO_4^{2-}, dicarboxylic acids and sometimes some WSOC were found to be mostly dominant at 0.7 μm and to have a minor mode at the supermicron particle size. The 0.7 μm mode was the typical size of the droplet mode formed by in-cloud processes as discussed earlier. Yao et al. (2004) and Yu J.Z. et al. (2004b, 2005) reported good correlations between SO_4^{2-} and dicarboxylic acids or between SO_4^{2-} and WSOC, attributable to in-cloud formation of these species.

The aerosol *in-situ* pH in submicron particles in Hong Kong is estimated to be lower than two (Pathak and Chan, 2005; Yao et al., 2006a, b). The low pH would substantially lower the dissolved S(IV) concentration and limit heterogeneous formation of SO_4^{2-} in submicron particles. However, the low aerosol pH in submicron particles might promote the formation of secondary organic species as proposed in the literature (Jang et al., 2002, 2003). In the PRD region outside of Hong Kong, Hagler et al. (2006) suggested that $PM_{2.5}$ was acidic, favoring the acid-catalyzed secondary formation of organic aerosols. Heterogeneous SO_4^{2-} formation in supermicron particles can be significant due to the presence of sea-salt aerosols and $CaCO_3$ aerosols in Hong Kong (Zhuang et al., 1999b). Heterogeneous formation of secondary species in supermicron particles in Shenzhen also occurs (Yao et al., 2003c; Huang X.F. et al., 2006).

NO_3^- is governed by the gas-aerosol equilibrium. Due to the low *in-situ* aerosol pH in submicron particles in Hong Kong, NO_3^- is mainly found in the supermicron particle size (Zhuang et al., 1999b; Yao et al., 2003c; Pathak et al., 2003, 2004a, b; Pathak and Chan, 2005). In Shenzhen, NO_3^- sometimes dominated in the supermicron particle size and sometimes dominated in submicron particle size, depending on the extent of neutralization of SO_4^{2-} aerosols (Yao et al., 2003c; Huang X.F. et al., 2006). NH_4^+ is mainly associated with SO_4^{2-} and NO_3^- and dominated in the submicron particles in the three mega cities (Zhuang et al., 1999a, b; Ho et al., 2004, 2006b; Yao et al., 2003c; Cheung et al., 2005; Louie et al., 2005a, b; Hagler et al., 2006; Huang X.F. et al., 2006).

The annual averages of As, Cd, Ni and Mn concentrations in PM_{10} were below the safety limits in Hong Kong (Lee et al., 1999) but the annual averages of As, Cd and Ni concentrations in PM_{10} in other areas in the PRD are not available. A short-term study reported that As in PM_{10} had daily average concentrations of over 20 ng m^{-3} (Wei et al., 1999). More studies are needed to evaluate the exposure risks to these toxic metals in the PRD.

4.5. Other air pollution problems

4.5.1. Visibility deterioration

In Guangzhou, Wu et al. (2006) found that the number of hazy days (with visibility less than 10 km and under RH < 80%) decreased from about 240 in 1997 to 65 in 2001 and then gradually increased to 144 days in 2004. As reported in the Shenzhen Environmental Bulletin, the number of hazy days increased from only 6 days per year in the 1980s to up to 74 days per year in the 1990s and has currently reached 164 days in 2006. Wu et al. (2005) found that heavy hazy days, when the visibility was less than 2 km, were associated with AOT larger than 1.0 due to high concentrations of aerosols.

In Hong Kong, the number of hours with visibility below 8 km (and under RH < 80%) is regularly reported. Fig. 10 shows the annual variation in the hours at the Hong Kong Observatory and Hong Kong International Airport, which are about 25 km apart. The number of hours steadily increased in the 1980s and 1990s but dramatically increased after 2002. It is interesting to note that the long-term trend of the hours of low visibility in Hong Kong is very different from that of the number of hazy days in Guangzhou where the largest number occurred in 1997 (not shown). Sequeira and Lai (1998) and Lee and Sequeira (2001, 2002) suggested that the SO_4^{2-}, NO_3^-, and NH_4^+ in aerosol particles are the main species degrading the visibility in Hong Kong. Using the multi-linear regression equations developed for the IMPROVE project (Malm et al., 1994), Cheung et al. (2005) estimated that, under northeasterly and northerly winds, particulate organic matter and ammonium sulfate contributed about 40% and 45% to light extinction. The increase in SO_4^{2-}, NO_3^-, and NH_4^+ concentrations is likely one of

important factors for visibility deterioration in Hong Kong during 2003–2006. Organic matters can also be important contributors to visibility deterioration, although the organic carbon concentration in PM_{10} decreased in 1999–2005 in Hong Kong (http://www.epd-asg.gov.hk/english/report/aqr.php).

Moreover, the numbers of hours of low visibility at the Hong Kong International Airport (suburban area) were much higher than those at the Hong Kong Observatory (urban area), especially during 2004–2006. Huang J.P. et al. (2005) and Lam et al. (2005) proposed that strong sea–land breezes had created a convergent zone of air pollutants engulfing the Hong Kong International Airport. Simultaneous measurements of the chemical compositions of atmospheric particles, not yet available, are needed to understand the difference in visibility between the two locations.

Overall, visibility has rapidly deteriorated since 2003 over the whole PRD region. The increase of SO_4^{2-}, NO_3^-, and NH_4^+ concentrations played an important role in the visibility deterioration in Hong Kong, and other factors may also contribute to the visibility deterioration. Since the continuous observations of chemical compositions of PM_{10} in the PRD region during the last 5 years outside Hong Kong are not available, it is unclear whether visibility deterioration outside Hong Kong can be attributed to the same causes as in Hong Kong.

4.5.2. Acid rain

The percentage of occurrence of acid rain in Guangzhou varied from 63% to 91% of rain days in 1999–2005 but the annual average pH of acid rain was in the narrow range of 4.4–4.8. Liu et al. (2006) reported that the $[SO_4^{2-}]$ to $[NO_3^-]$ equivalence ratio in acid rain gradually decreased from 8.6 in 1985–1990 to 3.0 in 2003–2004 in Guangzhou. These ratios of larger than unity suggest that acid rain was still mainly caused by the oxidation of SO_2. In Shenzhen, the percentage gradually increased from 33% in 1999 to 81% in 2005, and the annual average pH gradually decreased from 5.1 in 1999 to 4.6 in 2005. In Hong Kong, the pH of rain was almost constant at 4.6 and the equivalence ratio was between 2 and 3 during 1990–2005, without any obvious trend (Sequeira and Peart, 1995; Ayers and Yeung, 1996; Tanner et al., 1996; Tanner, 1999; Wai et al., 2005; Air Quality in Hong Kong, 1999–2005).

4.5.3. Others

There are some but limited studies on ultrafine particles and bioaerosols in Hong Kong (Wong et al., 2003; Lee et al., 2004; Lau et al., 2006; Yao et al., 2005, 2006a, b, c). However, much more knowledge is needed to understand their atmospheric behaviors and potential health and environmental impacts.

In Guangzhou and Shenzhen, dozens of measures were taken to reduce the air pollutant emissions in the last decade (Guangzhou Environmental Bulletin and Shenzhen Environmental Bulletin, 1999–2005). The Environmental Protection Plan in the PRD region was developed in 2004. In Hong Kong, the Government of HKSAR gives a high priority to controlling both street-level air pollution and regional smog. A joint monitoring network over

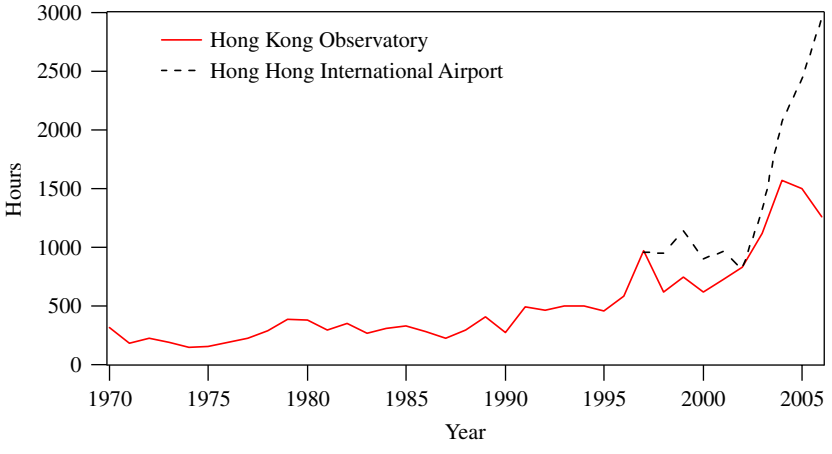

Fig. 10. The hours of visibility lower than 8 km in Hong Kong (The data source is from Hong Kong Observatory at http://www.weather.gov.hk/cis/statistic/hko_redvis_statistic_e.htm).

the PRD region was launched in 2005 for a close surveillance of the regional air quality. An implementation framework for the Emissions Trading Pilot Scheme for Thermal Power Plants in the PRD Region were completed and presented to prospective participating power plants in the PRD region. These measures were effective in reducing some air pollutant emissions, e.g., CO from on-road vehicles, and primary particles from industrial sources in the PRD region. However, they failed to reduce the SO_2 and NO_x emissions since 2003. Tighter controls of industrial SO_2 and NO_x emissions are needed to improve air quality.

5. Air pollution in other mega cities in China

There have been some studies of air pollution in other mega cities in China. Querol et al. (2006) reported the annual average SO_2, NO_2 and PM_{10} concentrations in Wuhan of 38, 60 and 156 $\mu g\,m^{-3}$, respectively, at an urban site (Hankou), and 73, 44 and 197 $\mu g\,m^{-3}$ at an industrial (Changqian) site. Mineral sources from cement, steel manufacture, smelting and fugitive dust were found to contribute to 34% of the PM_{10} mass at the industrial site, followed by a 20% contribution from a coal-fired power plant and a 16% contribution from the anthropogenic regional background. At the urban site, the mixed coal combustion source, the anthropogenic regional background and traffic emissions accounted for 31%, 28% and 16% of the total PM_{10} mass. Querol et al. (2006) also found high SO_4^{2-} concentrations (21 $\mu g\,m^{-3}$, average of 6 samples) in PM_{10} at a rural site in Wuhan, suggesting the influence of strong regional sources. Wei et al. (1999) reported a spatially uniform distribution of S in PM_{10} in Wuhan. While the annual average Ni concentration is at safe levels, the annual averages of As, Cd and Mn concentrations are generally higher than the EU and WHO limits due to industrial and other urban emissions (Querol et al., 2006). In particular, the high annual average concentration (about 70 $ng\,m^{-3}$) of As in Wuhan warrants immediate attention.

Short-term measurements of PM_{10} and $PM_{2.5}$ at an urban site (Huang-pi Jie) and a suburban site (Mo Shan) in Wuhan on selected days in four seasons during 1995 and 1996 yielded an average PM_{10} mass at the urban site of about 120 $\mu g\,m^{-3}$, which was 30% larger than that at the suburban site (Wei et al., 1999). $PM_{2.5}$ accounted for about 60% of the mass of PM_{10}. Waldman et al. (1991) have reported 2-week PM_{10} and $PM_{2.5}$ measurements made at a residential site in Wuhan in 1988. The average mass of PM_{10} and $PM_{2.5}$ was 225 and 139 $\mu g\,m^{-3}$. The difference between the PM_{10} and $PM_{2.5}$ masses in these studies was probably due to the fact that the measurements were made at different times and different sites. Some short-term measurements of atmospheric particles in other mega cities such as Nanjing, Guiyang, Chongqing and Qingdao have also been reported (Wang, G. et al., 2002, 2003; Wei et al., 1999; Xiao and Liu, 2004; Zhang D. et al., 2005; Wang G. et al, 2006). The information provided in these reports is not sufficient for a critical review of air pollution in these cities.

6. Immediate challenges of air pollution in mega cities

Although much effort has been devoted to alleviating air pollution in China's mega cities, challenges remain.

(1) The number of vehicles has increased by about 10% per year in these mega cities. Although the annual average NO_2 concentrations have remained almost constant and CO concentrations have decreased, probably due to improved control of vehicle emissions, the annual average of NO_2 concentrations in these mega cities still exceed the Grade-II standard. Further control of vehicle emissions is needed.
(2) Dozens of air quality monitoring stations have been established to measure concentrations of air pollutants in these mega cities in real time. These data, not yet available in the public domain, are very important for answering key questions such as to the form, long range transport, and source apportionment of air pollutants in these cities. In addition, various chemical species are likely to be unevenly distributed in different sizes of particles due to their specific sources or formation pathways. Measurements relating chemical species distributions to particle size are currently very limited in the mega cities, but are badly needed.
(3) High levels of both primary and secondary air pollutants are frequently detected in these cities, and strong interactions between atmospheric particles and oxidants are suspected (Shao et al., 2006; Xu J. et al., 2006). Due to the non-linear

nature of these interactions, parameterization of the air quality models for these cities need comprehensive sensitivity analysis.

(4) While the influence of dust storms has been often discussed, other regional contributions to air pollution are generally unclear due to uncertainties about emissions levels. This limits the utility of inventory-based modeling results. Comprehensive comparison between their predictions and the measured values is needed to evaluate the accuracy of emission source inventories.

(5) PM_{10} and $PM_{2.5}$ are the major air pollutants in the mega cities most of the time. The accumulation of these particles sometimes causes high pollution episodes and low visibility. The quantitative relationship between the chemical compositions of such particles and their size is, in general, not well established. The role of biomass burning needs to be addressed.

(6) SO_4^{2-} aerosols are often assumed to be completely neutralized in source apportionment and visibility budget analyses. However, hygroscopic growth of SO_4^{2-} aerosols strongly depends on the extent of neutralization, and it affects visibility indirectly. Artifact-controlled measurements, e.g., using denuders and back filters or real-time measurements, are important to accurately estimate the acidity of SO_4^{2-} aerosols and their contribution to visibility deterioration. However, such data, especially long-term data, are very limited. Assuming continuous improvement in the control of SO_2 emissions in the future, denuder/back filters measurements of NO_3^- and NH_4^+, almost now non-existent except for a few studies in Hong Kong, are urgently needed. In addition, NH_3, an important pollutant contributing to particulate SO_4^{2-} and NO_3^- formation and stabilization, should be routinely measured.

(7) The number of foggy days and light foggy days has substantially increased in the mega cities over the last two decades due to increasing air pollution. The liquid water content in fog droplets is expected to be much higher than that in the atmospheric aerosols present during fogless weather, and fog can promote the transformation of air pollutants. There have been too few studies of the relationship between fog and light fog processes and other forms of air pollution. In-cloud processes have been found to play an important role in SO_4^{2-} formation in Beijing and Hong Kong. The role of in-cloud formation of SO_4^{2-} (and secondary organic compounds) in the mega cities should be estimated quantitatively through 3-D modeling.

In addition to these issues, which are applicable to all of China's mega cities, a few specific challenges relate specifically to the three cities reviewed in this paper.

In Beijing, dozens of air pollution control measures have been taken since 1998.Why, then, has the number of the days when the O_3 concentration exceeds the Grade-II standards increased? Why have PM_{10} concentrations not decreased since 2002? These two questions constitute major challenges for air pollution control in Beijing in the future. Answering these two questions requires long-term data collection and more studies on the formation of secondary pollutants, in addition to updating the city's emissions inventory.

In Shanghai, only the frequency of days when air pollutant concentrations exceeded the Grade-II standard is reported in the Shanghai Environmental Bulletin. Much less was known about the actual concentrations of the pollutants, local and transport contributions, and the meteorological conditions associated with the episodes. The few studies, which have been done leave large uncertainties about O_3 formation mechanisms in Shanghai. Although PM_{10} is the major pollutant in Shanghai on more than 80% of days, the chemical composition of the particles has scarcely been reported. Current understanding of air pollution in Shanghai is very limited and not sufficient for proactive pollution control.

In the PRD region, most studies have suggested that O_3 formation is VOC-limited. On the other hand, Wang X.M. et al. (2005) have suggested that O_3 formation outside the urban areas of the PRD is NO_x-limited. Whether the formation of O_3 is VOC-limited or NO_x-limited is very important to formulating a policy to reduce O_3 episodes. In addition, anthropogenic VOC emissions have decreased in Hong Kong, while O_3 episodes have increased during the 1999–2005 period. Outside Hong Kong, anthropogenic VOC emissions are thought mainly to originate from vehicular emissions (about 60%). The CO concentrations in Guangzhou have decreased due to effective vehicular emission control measures. It is unknown whether the total vehicular VOC emissions have decreased or not.

7. Conclusions

Over the last few years there has been a rapid growth in studies and publications about air pollution in China's mega cities, particularly about Beijing, Shanghai and the PRD region, which includes Guangzhou, Shenzhen and Hong Kong. Economic growth and urbanization has resulted in drastic increases in energy consumption, which in turn have caused a large amount of additional pollutant emissions. On 10–30% of days air pollutants exceed the Grade-II standard in the mega cities and their immediate vicinities. Much attention has been paid to reducing emissions, particularly vehicle emissions. Although the number of vehicles has increased by about 10% per year in these cities, NO_2 and CO concentrations have not increased due to effective control measures. SO_2 emissions were successfully controlled in Beijing, but not in Shanghai or the PRD. O_3 episodes are frequent, and the number of episodes is increasing. VOC measurements are in general scarce and are not adequate for understanding O_3 formation.

Particulate pollution is still severe, and it is the major air pollution problem in the mega cities. The sources, formation, and regional transport of air pollutants have been widely studied, and knowledge about these processes has improved over the last decade. Most studies have focused on mass concentration and the chemistry (ions, metals, EC/OC) of the TSP, PM_{10} and $PM_{2.5}$ components. Size distribution studies using cascade impactors are rare, as are real-time size and composition distribution measurements.

There has been a significant improvement in emissions inventories, especially for gaseous pollutants, although they should be updated more often. Accounts of the emissions of primary pollutants are in general satisfactory, but descriptions of the formation of secondary pollutants, including O_3, SO_4^{2-}, NO_3^-, and secondary organic compounds, need improvement. Sampling artifacts in the measurements should be examined, and cross comparisons of related studies in the literature are encouraged.

China's mega cities suffer from very high NO_3^- concentrations, and gaseous and particulate ammonia and particulate NO_3^- should be routinely monitored, preferably through real-time measurements with protocols to minimize sampling artifacts. Finally, there is a paucity of laboratory studies to complement the modeling efforts and field measurements in China. A more balanced effort in capacity building by atmospheric researchers is called for, including fundamental laboratory studies of the physical and chemical behavior of atmospheric aerosols as well as instrumentation development.

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