

A Review of Atmospheric Chemistry Research in China: Photochemical Smog, Haze Pollution, and Gas-Aerosol Interactions

MA Jianzhong^{*1} (马建中), XU Xiaobin¹ (徐晓斌), ZHAO Chunsheng² (赵春生), and YAN Peng¹ (颜鹏)

¹Key Laboratory for Atmospheric Chemistry, Chinese Academy of Meteorological Sciences, Beijing 100081

²Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing 100871

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ABSTRACT

In this paper we present a review of atmospheric chemistry research in China over the period 2006–2010, focusing on tropospheric ozone, aerosol chemistry, and the interactions between trace gases and aerosols in the polluted areas of China. Over the past decade, China has suffered severe photochemical smog and haze pollution, especially in North China, the Yangtze River Delta, and the Pearl River Delta. Much scientific work on atmospheric chemistry and physics has been done to address this large-scale, complex environmental problem. Intensive field experiments, satellite data analyses, and model simulations have shown that air pollution is significantly changing the chemical and physical characters of the natural atmosphere over these parts of China. In addition to strong emissions of primary pollutants, photochemical and heterogeneous reactions play key roles in the formation of complex pollution. More in-depth research is recommended to reveal the formation mechanism of photochemical smog and haze pollution and their climatic effects at the urban, regional, and global scales.

Key words: ozone, aerosols, photochemical smog, haze pollution, gas-aerosol interactions

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1. Introduction

Atmospheric chemistry is a scientific discipline that studies the chemical composition of the natural atmosphere and the related physico-chemical processes: the way gases, liquids, and solids in the atmosphere interact with each other and with the Earth's surface and associated ecosystems, and how human activities may be changing the chemical and physical characteristics of the atmosphere (e.g., Seinfeld and Pandis, 1998; Warneck, 2000). Atmospheric chemistry is an interdisciplinary that can be considered a sub-branch of both atmospheric sciences and environmental sciences. A number of critical environmental issues are associated with a changing atmosphere, from photochemical smog, acidic deposition, toxic air pollutants, and stratospheric ozone depletion, which were well-known by the end of the last century, to haze pollution and

global climate change, to which considerable scientific attention has been given since the beginning of this century (UNEP and C4, 2002; IPCC, 2007). Much of the anthropogenic impact on the atmosphere is associated with our increasing use of fossil fuels and biomass burning as an energy source. To understand and hopefully solve these problems, a great deal of research has been/is being done around the world, and correspondingly the field of atmospheric chemistry has become one of the most rapidly developing and active subjects in Earth sciences over the past several decades.

As the world's largest and fastest developing country in economy, China also faces greater environmental challenges. In China, coal burning supplies ~70% of the energy consumed, and the carbonaceous aerosols and sulfur dioxide (SO₂) from the associated exhausts constitute the primary pollutants in most areas (Yearbook, 2007). While the SO₂ mixing ratio remains

^{*}Corresponding author: MA Jianzhong, mjz@cams.cma.gov.cn

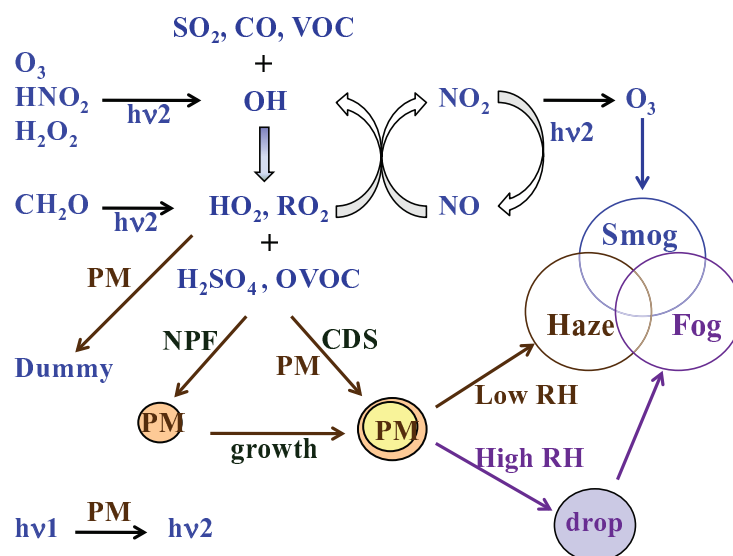


Fig. 1. The most important chemical processes for the formation of photochemical smog and haze-fog. PM refers to aerosol particles, NPF stands for new particle formation, and CDS the condensation of trace gases on aerosol surfaces. The change of “h ν 1” to “h ν 2” indicates the effect of aerosols on the actinic flux and thus J -values.

at high levels, the concentrations of nitrogen oxides ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$) have increased significantly in many areas. Whereas air quality in cities and acid rain remains the main environmental issue (Shao et al., 2006; Tang et al., 2010), regional pollution related to photochemical smog and haze-fog has been recognized as a severe environmental problem in China (Chan and Yao, 2008; Zhang et al., 2008b; Ma et al., 2010a). The most seriously polluted regions are North China (Huabei in Chinese), including the Beijing and Tianjin megacities, the Yangtze River Delta region (YRD), including the Shanghai megacity, and the Pearl River Delta region (PRD), including Guangzhou and Hong Kong megacities. Such air pollution not only has adverse effects on human health and the ecosystems but also may have large impacts on weather and climate at urban and regional scales.

In this paper, we present a review of atmospheric chemistry research in China (mainland) over the last 4 years, from 2006 to 2010. Our foci are photochemistry, aerosol chemistry, and heterogeneous chemistry in the troposphere. The contents of the paper include tropospheric ozone (section 2), aerosol chemistry (section 3), and the interactions between trace gases and aerosols (section 4). Conclusions and remarks are presented in section 5.

2. Tropospheric ozone

Tropospheric ozone is produced from the oxidation of hydrocarbons and carbon monoxide (CO) in the

presence of NO_x and sunlight (Fig. 1). The oxidation of these ozone gaseous precursors is mainly initiated by hydroxyl radicals (OH), formed primarily through the photolysis of O₃, nitrous acid (HNO₂), and hydrogen peroxide (H₂O₂). Atmospheric ozone production in polluted areas involves different stages, from being VOC-sensitive near the sources in urban centers to being NO_x -sensitive further downwind in rural areas. Photochemical smog, characterized by high O₃ and other oxidant levels, has become a major concern for air quality from the urban to regional scale in China.

2.1 Urban ozone pollution

2.1.1 Seasonal variation and trend analyses

Measurements of ozone and its precursors from June 2006 to June 2007 in urban Shanghai showed a spring maximum of ozone, winter peaks of NO_x and CO, and no apparent seasonal variation of volatile organic compounds (VOCs) (Ran et al., 2009). Ground measurements of tropospheric NO₂ vertical column densities (VCDs) by DOAS (Different Optical Absorption Spectrometer) in Shanghai showed that the satellite observations failed to reproduce the high NO₂ amounts over the polluted experimental site (Chen et al., 2009b). To further validate the satellite measurements, extension of ground-based zenith-sky DOAS measurements is required to cover areas with different pollution levels within the whole satellite footprint (Chen et al., 2009b). The seasonal variation pattern

of tropospheric NO₂ VCD measured using the GOME (Global Ozone Monitoring Experiment) satellite was in good agreement with that of surface NO₂ concentration in Beijing, with a minimum occurring in summer and a maximum in winter (Jiang et al., 2006).

Table 1 summarizes the levels of ozone and its precursors measured in Beijing during summertimes of different years. Observations at six urban sites in Beijing showed that, over the 2001–2006 period, the average concentrations of NO_x and O₃ for July–September were 49.2±5.9 ppbv and 26.6±2.8 ppbv, respectively. While the NO_x concentration decreased linearly at a rate of 3.9±0.5 ppbv yr⁻¹ after 2002, ozone concentration increased at a rate of 1.1±0.5 ppbv yr⁻¹ during 2001–2006 (Tang et al., 2009a). In contrast, analyses of satellite data from GOME and SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) showed that tropospheric NO₂ VCDs over all the Chinese megacities, including Beijing, increased significantly during this period (Zhang et al., 2007a).

2.1.2 Photochemical processes

Observational analyses and model simulations revealed that the formation of ozone is limited by volatile organic compounds (VOCs) in the urban areas of Beijing, being more sensitive to NO_x levels in the suburban and more remote areas (Wang et al., 2009h; Xu et al., 2008a). Ambient concentrations of non-methane hydrocarbons (NMHCs) and oxygenated VOCs (OVOCs) and their contributions to ozone formation in Beijing were investigated in numerous studies (e.g., Song et al., 2007; Liu et al., 2009d; Shao et al., 2009; Yuan et al., 2010). OVOCs accounted for 37%–54% of the VOC (NMHC+OVOC) mixing ratio, and the main reactive VOC compounds were aldehydes and alkenes (Shao et al., 2009). Measurements of methacrolein (MACR) and methyl vinyl ketone (MVK), the two specific products from isoprene oxidation in the atmosphere, were performed in Beijing (Xie et al., 2008; Pang et al., 2009), and the active isoprene was estimated to account for 6%–30% of the photochemically produced ozone (Pang et al., 2009). In addition to SO₂, NO_x, and O₃, the vertical distributions of VOCs were measured in the lower boundary layer of Beijing, providing valuable data for studying the effects of meteorological and photochemical processes on urban ozone (Mao et al., 2008; Meng et al., 2008; Sun et al., 2010c).

Similarly, both model and measurement results have indicated that ozone formation is clearly under VOC-sensitive regime in the city of Shanghai, and aromatics play a dominant role in ozone production, in spite of a lower contribution to the total VOC con-

centration (Geng et al., 2007; Geng et al., 2008; Ran et al., 2009). Interestingly, the ozone “weekend effect” was found at the Xujiahui district of Shanghai, with ozone concentrations being higher on weekends than on weekdays (Tang et al., 2008). The ozone weekend effect was also observed at an urban site in the Jinan city of eastern China (Shan et al., 2008). Studies showed that biomass burning could have large impacts on air quality in the cities, e.g., Beijing and Guangzhou (Wang et al., 2007a; Li et al., 2008c). The enhanced aerosol concentrations due to the Southeast Asia biomass burning have been argued to result in a reduction of UV intensity, and thus decrease the formation of ozone in Guangzhou (Deng et al., 2008b).

Changes in the concentrations of HONO, HCHO, O₃, and NO₂ were observed for an autumn period when a strong cold front passed through Beijing (An et al., 2009). Nighttime mean HONO concentration and daily-mean HCHO and NO₂ concentrations ranged from 0.95 ppbv to 4.9 ppbv, from 3.6 ppbv to 9.8 ppbv, and from 9.5 ppbv to 49.3 ppbv, respectively (An et al., 2009). The contributions from the photolysis of HONO, HCHO, and O₃ to the overall daily HO_x production were estimated to range 56%–83%, 11%–36%, and 6%–9%, respectively (An et al., 2009). An observational study of HNO₂ and NO₂ was also performed in Shanghai during a autumn–winter period (Hao et al., 2006). Mean hourly concentrations of HNO₂ and NO₂ during this period were 1.1 ppbv and 24 ppbv, respectively (Hao et al., 2006). The temporal behavior of HONO and NO₂ at an urban site of Guangzhou during summertime was investigated, and high HONO mixing ratios (up to 2 ppb) were observed during daytime (Qin et al., 2009). A nocturnal maximum concentration of ~8.4 ppbv was detected at night, and the HONO to NO₂ concentration ratio ranged from 0.03 to 0.37, significantly higher than previously reported values (Qin et al., 2009). Heterogeneous reactions of NO₂ on aerosol surface could be a key source of HONO, and the effect of relative humidity may also play a role (Hao et al., 2006; An et al., 2009; Qin et al., 2009).

2.1.3 Emission restriction effect studies

Long-range transport may have large impacts on the distribution of ozone and its precursors over urban areas (An et al., 2007; Chen et al., 2009d; Wang et al., 2009h). The ozone pollution in Beijing showed a significant spatial distribution with strong regional contribution (Wang et al., 2009h). The effects of pollutant emission restrictions implemented during the Beijing Olympic Games 2008 on air quality in Beijing and surrounding areas have been investigated in numerous studies. Sharp reductions in the concentrations of ozone precursors and aerosols with comparison to no

emission control periods were observed at the urban sites (Liu et al., 2009a; Wang et al., 2009e; Wang and Xie, 2009; Zhang et al., 2009; Xin et al., 2010), at the rural sites (Wang et al., 2009i; Zhang et al., 2009; Xin et al., 2010), and by satellite (Yu et al., 2010). The reduction in daily air pollutant emissions in Beijing were quantified, with 47% and 57% reduction of NO_x and NMVOC in August than in June 2008 (Wang et al., 2010b). Daytime ozone concentrations were observed to decrease by ~ 15 ppbv in maximum in the surrounding areas, including a rural site far beyond the Beijing urban area (Wang et al., 2009i; Xin et al., 2010). In the urban Beijing, the reduction of ozone was smaller (Zhang et al., 2009; Xin et al., 2010), and an increase in ozone was even reported for some control stages (Wang and Xie, 2009; Zhang et al., 2009). It should be noted that different diurnal variation patterns of urban street ozone for summer time in Beijing were reported (Chen et al., 2009c; Wang and Xie, 2009).

2.2 Ozone pollution in rural areas

2.2.1 Seasonal variation and trend analyses

The seasonal variation of ozone in eastern China is influenced by both Asian summer monsoon and regional photochemical pollution (Wang et al., 2006a; Lin et al., 2008; Xu et al., 2008b; Lin et al., 2009; Meng et al., 2009). Table 2 summarizes the levels of ozone and other trace gases observed in non-urban areas of Huabei, the YRD, and the PRD. At Shangdianzi, a regional Global Atmosphere Watch (GAW) station in Huabei, the monthly mean values of both ozone daily average and ozone daily peak occurred in June and September (Lin et al., 2008; Meng et al., 2009). Interestingly, while the NO_x levels were still higher in spring than in summer, ozone showed the smallest diurnal variation in spring due to its largest concentrations at night (Meng et al., 2009). This indicates that the long-range transport of ozone, probably from the stratosphere, makes a great contribution to the background ozone in Huabei during springtime.

The highest mixing ratios of ozone were observed in May in the YRD (Wang et al., 2006a; Xu et al., 2008b). Long-term observations of surface ozone at Lin'an, a regional GAW station in the YRD, during the periods between August 1991 and July 2006 showed an increasing trend of daytime values and a decreasing trend of nighttime values (Xu et al., 2008b). Such a phenomenon was most likely caused by the increase of NO_x concentration in the YRD, which tends to enhance the ozone production during daytime and the ozone titration at night (Xu et al., 2008b). There are also distinct seasonal and diurnal cycles in ground-level ozone across the PRD region. Lower ozone concentrations were generally observed in summer, while

high O_3 levels were typically found in autumn (Zheng et al., 2010). Long-term measurement of surface ozone has been made at a coastal site in Hong Kong, which is strongly influenced by the outflow of Asian continental air during winter and the inflow of maritime air from the subtropics in summer. An increasing trend of $0.58 \text{ ppbv yr}^{-1}$ was obtained based on the data for 1994–2007, and this significant trend is most likely due to the increased emissions of ozone precursors in the upwind coastal regions of mainland China (Wang et al., 2009f). Long-term trends of yearly and seasonal averages of tropospheric ozone over the whole country and some regions of China were analyzed, based on the tropospheric ozone residue (TOR) data retrieved from satellite measurements (Xu and Lin, 2010). Significant increasing trends of TOR were found over the North China Plain for all seasons except winter, with a maximum rate of $1.10 \text{ DU per decade}$ for summer. There were significant correlations between TOR and SOI (Southern Oscillation Index) for some Chinese regions but not for the North China Plain, suggesting that the observed increasing trend of TOR over the North China Plain may not be linked with the changes in atmospheric circulations (Xu and Lin, 2010).

2.2.2 Episodes of regional ozone pollution

Aircraft measurements in the planetary boundary layer (PBL) of the YRD in autumn revealed a large difference in the concentrations of ozone and its precursors between suburban and rural regions (Geng et al., 2009). In contrast to previous studies, ozone chemical formation in the YRD was suggested to be located in a strongly VOC-limited regime (Geng et al., 2009). Aircraft measurements showed severe ozone pollution over the PRD in autumn, and during some episodes NO_x and ozone concentrations reached 160 ppbv and 100 ppbv, respectively (Wang et al., 2008c). A high pollution level at the top of the PBL was found under certain meteorological conditions (Wang et al., 2008c).

The ocean–land circulation has an important role in regional ozone formation and distribution over the PRD (Jiang et al., 2008; Xu et al., 2008b). During an autumn period, intensive field measurements were conducted simultaneously at the two non-urban sites within the inland PRD region and in Hong Kong, respectively (Guo et al., 2009). Although higher NO_x concentrations were observed at the Hong Kong site (45 ± 2 ppbv) than at the PRD site (31 ± 2 ppbv), ozone diurnal variations showed higher nighttime levels at the Hong Kong site (Guo et al., 2009), indicating the influence of long-range transport on the background ozone level in western Hong Kong. High levels of ozone and other pollutants in Hong Kong could be caused by regional transport from the upwind PRD area, and

Table 1. Summary of ozone and its precursors' concentrations (ppbv) observed in Beijing during summertime.

Sources	Description	O _{3,max}	O ₃	O _x	CO	NO _x	SO ₂	NMHC	HCHO	CH ₃ CHO
Tang et al., 2009a	Six IAP sites, Jul–Sept 2001	84 ^a	27	60		48				
Tang et al., 2009a	Six IAP sites, Jul–Sept 2002	83 ^a	22	61		60				
Tang et al., 2009a	Six IAP sites, Jul–Sept 2003	85 ^a	28	63		53				
Tang et al., 2009a	Six IAP sites, Jul–Sept 2004	84 ^a	25	59		51				
Tang et al., 2009a	Six IAP sites, Jul–Sept 2005	85 ^a	31	63		45				
Tang et al., 2009a	Six IAP sites, Jul–Sept 2006	83 ^a	28	58		40				
Xin et al., 2010	Five IAP sites, Jul–Sept 2008	62 ^b	37	53		18	3			
Chen et al., 2009c	Street site, August 2006		39	72	3360	49	20			
Chen et al., 2009c	CRAES site, August 2006		36	66	1410	39	13			
Zhang et al., 2009	CAMS site, August 2006	154 ^c			1400	25	16			
Zhang et al., 2009	CAMS site, August 2008	118 ^c			647	16	4			
Pang and Mu, 2006	RCEES site, Jun–Aug 2005								14.6	7.1
Shao et al., 2009	PKU site, August 2004							28		
Shao et al., 2009	PKU site, August 2005							36		
Shao et al., 2009	PKU site, August 2006							31	9.2	3.9

Note: Superscripts a for daily maximum, b for 8-h maximum, and c for hourly maximum.

it has been estimated that the influence of regional transport could contribute 50%–90% of surface ozone during nighttime and 40%–70% during daytime (Wang et al., 2006b).

A typical feature of the urban and regional photochemical pollution was observed during the Regional Integrated Experiments on Air Quality over the Pearl River Delta 2004 (PRIDE-PRD2004; Zhang et al., 2008b). The average O₃ concentration was ~73 ppbv northeast of the PRD (upwind), and it increased to ~87 ppbv in the southern area of the PRD (downwind). For the total oxidant O_x (O₃+NO₂), an even more clearly increasing trend was found: ~79 ppbv in the upwind area, ~92 ppbv in the urban area, and ~116 ppbv in the downwind area (Zhang et al., 2008b, c). The monthly mean values of O₃ and O_x at the two stations of Huabei [Shangdianzi, (Lin et al., 2008) and Guocheng (Lin et al., 2009)], which are located north-east and southwest of Beijing, respectively, did not exhibit a clear up-to-down wind trend.

2.2.3 VOC activity and ozone production efficiency

Few measurements of VOCs have been reported for non-urban areas (Tang et al., 2007; Geng et al., 2009; Guo et al., 2009; Liu et al., 2008e; Mao et al., 2009). The concentrations of VOCs in the YRD are relatively small (with a mean value of 6 ppbv) at higher altitudes of the PBL: 52% from alkanes, 16% from alkenes, 17% from aromatics, 3% from ketones, and 5% from other VOCs, respectively (Geng et al., 2009). The aromatics were estimated to contribute the most to ozone production in the YRD near Shanghai (Geng et al., 2009). Alkanes constituted the largest percentage (>40%) in mixing ratios of the quantified VOCs at six sites in the PRD, with an exception of one major industrial site, where aromatics dominated (~52%; Liu et al., 2008e). Whereas alkenes play a predominant role in the VOC reactivity for the urban area, reactive aromatics may also make a great contribution to ozone formation in rural areas of the PRD (Liu et al., 2008e). The effects of biogenic VOCs were also shown to play an important role in the PRD region (Wei et al., 2007).

Table 2. Summary of ozone and its precursors observed in non-urban areas of Huabei, the YRD, and the PRD.

Sources	Description	O ₃	NO _x	CO	SO ₂	NMHC	Suppl.
Meng et al., 2009	Huabei, rural, annual	31	13	742	8	—	O _x : 43
Lin et al., 2009	Huabei, rural, annual	29	29	1520	17	13.7 ^b	O _x : 47
Geng et al., 2009	YRD, suburban, PBL, autumn	35	23	4500	18	4.6	Carbonyl: 0.83
Geng et al., 2009	YRD, rural, PBL, autumn, daytime	55	5	3500	4	4.6	Carbonyl: 0.83
Guo et al., 2009	PRD, suburban, autumn	32	45	570	9	26	Carbonyl: 26
Guo et al., 2009	PRD, suburban, autumn, daytime	35	48	600	10	26	Carbonyl: 26
Guo et al., 2009	PRD, rural, autumn	40	31	1050	32	39	Carbonyl: 43
Guo et al., 2009	PRD, rural, autumn, daytime	59	27	1120	32	39	Carbonyl: 43
Zhang et al., 2008b ^a	PRD, rural, autumn	54	10	730	5	39	
Wang et al., 2008c	PRD, rural, PBL, autumn, daytime	53	32	—	28	—	

Note: ^a Only the data for one rural site are taken into account. The data for NMHC was estimated from the work of Liu et al. (2008e). ^b The data for NMHC was estimated from the work of Shao et al. (2009).

Ozone production efficiency (OPE) is defined as the number of molecules of O₃ formed per NO_x removed from atmospheric ozone-forming oxidation cycles [i.e., P(O₃)/P(NO_x)] (Seinfeld and Pandis, 1998). The ozone production efficiency was estimated to be generally 2–8 in PRD (Wang et al., 2010c), with higher values (10–15) during severe ozone pollution episodes (Sun et al., 2010b). Although the production of ozone in the entire PRD region is highly likely to be VOC limited (Zhang et al., 2008b), one study argued that the production of ozone is possibly controlled by NO_x in the northern or northeastern rural areas of the PRD (Zheng et al., 2010). Based on measurements at the Shangdianzi Regional Background Station in Huabei during March to October 2008, OPE was calculated and its daily value varied in the range of 0.2–21.1, with an average of 4.9±3.6 (Ge et al., 2010).

2.3 Tropospheric ozone over a large scale of eastern China

2.3.1 Model evaluations and satellite data applications

Regional chemical transport models were compared with an extensive set of observations, including the TRACE-P experiment, to evaluate the models' abilities in simulating ozone and relevant species in the troposphere of East Asia (Zhang et al., 2006a,b; Han et al., 2008a; Gao et al., 2009b; Wang et al., 2010e). Almost all models tended to underestimate NO_x levels, due to large uncertainties in either emission estimates or complex chemical mechanism represented (Han et al., 2008a). The model skills for ozone vary largely with region and season, and large differences were found among models for Southeast Asia, where the observations are sparse (Han et al., 2008a).

Satellite data have been widely used to investigate

the chemical characteristics of air pollution in China (Ma et al., 2006; Tie et al., 2006; Li et al., 2007c; Peng et al., 2007; Zhang et al., 2007a; Shi and Zhang, 2008; Shi and Zhang, 2008b). These include aerosol optical depths from the Moderate Resolution Imaging Spectroradiometer (MODIS), tropospheric NO₂ vertical column densities from GOME and SCIAMACHY, tropospheric ozone residue, and tropospheric CO vertical column densities from the Measurements of Pollutants in the Troposphere (MOPITT). The geophysical distribution, seasonal variation, and historical trend of tropospheric NO₂ vertical column densities over China were analyzed on the basis of measurements from the satellite instruments GOME and SCIAMACHY for the period 1997–2006 (Zhang et al., 2007a). A high growth rate of tropospheric NO₂ over eastern China, especially over the industrial areas with a fast economical growth, such as YRD and PRD, was found (Zhang et al., 2007a). Tropospheric NO₂ vertical column densities over China simulated by regional models were compared with GOME and SCIAMACHY satellite data to evaluate the uncertainties in emission inventories and the accuracy of satellite data (Ma et al., 2006; Shi and Zhang, 2008b). The model tended to underestimate the tropospheric NO₂ column density in remote and rural areas of China with respect to the GOME measurements (Ma et al., 2006). The NO₂ column over the eastern China calculated by a global model, MOZART-2, is fairly consistent with the GOME measurements (Zhao et al., 2006). However, MOZART-2 underestimates tropospheric CO column in China with comparison to MOPITT CO, due to the uncertainties of emissions (Peng et al., 2007; Zhao et al., 2006). New CO emission data were obtained by the CO budget analysis using MOZART model, constrained by MOPITT data (Lin et al., 2007b).

2.3.2 Ozone production and transport

The observations at the three mountain sites (Mount Taishan, Huashan Mountain, and Huangshan Mountain) showed a striking ozone pattern of two sharp peaks in May–June and September–October in central east China (Li et al., 2007a). This seasonal pattern is the same as those observed at the ground regional stations of eastern China (Lin et al., 2008; Xu et al., 2008b; Lin et al., 2009; Meng et al., 2009). The calculated net photochemical production of ozone reached 31.8 ppbv d⁻¹, 15.1 ppbv d⁻¹, and 11.4 ppbv d⁻¹ at Mount Taishan, Huashan Mountain, and Huangshan Mountain, respectively (Li et al., 2007a). Model simulations showed that biogenic emissions have remarkable impacts on surface ozone in eastern China, and the increase of ozone concentration generally reaches up to 10–30 ppbv in the VOC-limited areas (Wang et al., 2008a). Measurements of NMHCs were concurrently carried out at a few remote and rural sites in background atmospheres of eastern, northern, southwestern, and southern China, providing valuable information for investigating the sources of air pollution and their impacts on ozone in different regions (Tang et al., 2007, 2009b).

Regional emissions, photochemistry, and monsoon activities control the regional distribution and seasonal variation of ozone and CO over China and their export to the North Pacific, as simulated by the regional models (Li et al., 2008a; Wang et al., 2008a; Gao et al., 2009b) and by a nested-grid global model (GEOS-Chem; Chen et al., 2009a), respectively. A CO-tagging method was implemented in GEOS-Chem to investigate the formation and dispersion of CO pollution over Beijing, and the results suggest that regional emissions make significant contributions to elevated CO levels over Beijing on polluted days (Chen et al., 2009a). An O₃-tagging module was used in a regional model to study the source of near-ground ozone at Mt. Tai in central eastern China (CEC), and the results indicate the importance of the regional-scale transport in the redistribution of ozone that are chemically produced both inside CEC and in the southern part of CEC (Li et al., 2008a).

3. Aerosol chemistry

Aerosols have been recognized as one of the major pollutants that dramatically degrade air quality in China. Atmospheric aerosols have two sources categories, primarily emissions (e.g., from natural dust, anthropogenic fossil combustion and biomass burning) and gas-to-particle conversion (e.g., by the condensation of gaseous sulfuric acid and low volatile oxygen-contained organic compounds) (Fig. 1). Over the past

few decades, haze-clouds or haze-fog has become a widespread weather phenomenon in the polluted regions of China, due to very high loads of aerosol particles, especially those with high hygroscopicity, which are determined by the chemical composition of aerosols in the atmosphere.

3.1 Inorganic ions

The hygroscopicity of aerosols is mainly determined by the composition and amount of inorganic salts they contain. Table 3 summarizes the PM_{2.5} mass concentration and major ionic composition observed in different regions of China. The mass concentration of PM_{2.5} is generally higher in the northern areas, especially during springtime, due to the dust effect. For example, in Beijing mineral dust contributes 52% in spring, 16% in summer, 17% in autumn, and 22% in winter to the total mass in PM_{2.5}, and it comprises up to 90% in dust storms (Han et al., 2007). Although Shanghai is weakly influenced by dust storms, the high mass concentration and low ionic content in spring might be caused by the dust intrusion from outside Shanghai, probably related to dust storms in northwestern China (Wang et al., 2006d). Sulfate is the major water-soluble component for all regions, and sulfate, nitrate, and ammonium are the predominant water-soluble components in cities (Table 3). Interestingly, higher NO₃⁻ than SO₄²⁻ was observed in Beijing during an autumn period (Song et al., 2006) and in Guangzhou during a winter period (Tan et al., 2009b), most probably due to strong photochemical activities and the environment conditions that favor the formation of particle nitrate occurring during the experiments. Whereas the maximum of SO₄²⁻ contents occurs in summer in Beijing, it happens in winter in Shanghai (Table 3), indicating that the formation of SO₄²⁻ is controlled by both its gas precursor (SO₂) and the photochemical oxidation capacity (OH). PM1 observed by an Aerodyne Aerosol Mass Spectrometer (AMS) during summertime in Beijing was composed of 25% sulfate, 22% nitrate, 16% ammonium, and 35% of organic aerosol (OA) species (Sun et al., 2010a).

3.2 Organic aerosols

3.2.1 Species identification and quantification

Organic carbon aerosols (OC) contributed 30%–35% to PM_{2.5}, and the secondary OC accounted for 40% of OC in Chinese urban environments (Cao et al., 2007). The ratio of OC to element carbon (EC) was on the order of 3:1 for the urban locations, but it could reach as high as 6:1 at the rural sites (Zhang et al., 2008a). This indicates that more secondary organic aerosols (SOA) form in the regional background than in urban atmospheres. According to a model

Table 3. (a) Summary of PM_{2.5} mass concentration and major ionic composition observed in the Beijing area. (b) Summary of PM_{2.5} mass concentration and major ionic composition observed in Tongliao (TL), Ürümqi (UR), Xi'an (XN), and Shanghai (SH). (c) Summary of PM_{2.5} mass concentration and major ionic composition observed in Guangzhou, South China Sea (SC-Sea), and the four forests, i.e., Changbai Mountain Nature Reserve (CB), Dongping National Forest Park in Chongming Island (CM), Dinghu Mountain Nature Reserve (DH), and Jianfengling Nature Reserve in Hainan Island (HN).

Sources	Description	PM _{2.5} mass ($\mu\text{g m}^{-3}$)	SO ₄ ²⁻ (%)	NO ₃ ⁻ (%)	NH ₄ ⁺ (%)	Ca ²⁺ (%)	Na ²⁺ (%)
(a)							
Song et al., 2006	Urban, spring	134	18	17	9.1	—	—
Song et al., 2006	Urban, summer	99	29	5.6	7.5	—	—
Song et al., 2006	Urban, autumn	106	14	17	6.8	—	—
Song et al., 2006	Urban, winter	61	14	8.2	5.6	—	—
Song et al., 2006	Urban, annual	96	19	12	7.3	—	—
Han et al., 2007	Urban, dust	536	1.5	0.3	0.3	—	—
Han et al., 2007	Urban, spring	162	9.1	6.0	3.3	—	—
Han et al., 2007	Urban, summer	79.6	28	19	9.5	—	—
Han et al., 2007	Urban, autumn	107	12	11	6.2	—	—
Han et al., 2007	Urban, winter	198	15	8.5	6.5	—	—
Wang et al., 2009d	Urban, summer	~102	31	20	~20	2.6	—
Wang et al., 2009d	Urban, winter	~102	27	17	~12	2.5	—
Sun et al., 2006	Urban, winter, haze-fogs	220	9.7	6.3	4.5	0.9	0.1
Sun et al., 2006	Urban, winter, haze-fogs	37	4.6	2.5	3.8	4.1	0.6
Wang et al., 2006c	Urban, dust	409	3.8	1.2	0.99	1.7	0.35
Wang et al., 2006c	Urban, haze	179	22	14	10	1.0	0.34
Wang et al., 2006c	Urban, clear	107	10	8.6	4.7	2.8	0.52
Yan et al., 2008	Rural, summer, polluted	143	41	11	24	—	—
Yan et al., 2008	Rural, summer, clean	25	23	9.8	11	—	—
(b)							
Shen et al., 2007	TL, spring, dust	255	4.2	1.6	0.65	2.4	0.3
Shen et al., 2007	TL, spring, no-dust	104	4.9	1.4	0.82	2.0	0.8
Shen et al., 2007	TL, spring, pollution	118	15	6.7	4.4	1.8	1.0
Li et al., 2008b	UR annual	189	26	5.0	13	1.1	1.2
Shen et al., 2009	XN, haze	351	27	16	8.2	0.8	—
Shen et al., 2009	XN, dust	138	8.4	2.2	1.0	3.1	—
Shen et al., 2009	XN, normal	130	21	9.2	5.8	1.0	—
Wang et al., 2006d	SH, spring	135	8.6	6.7	3.0	1.1	0.4
Wang et al., 2006d	SH, summer	72	7.6	3.6	3.4	2.2	0.7
Wang et al., 2006d	SH, autumn	96	9.0	3.8	3.7	0.8	0.4
Wang et al., 2006d	SH, winter	76	17	11	5.8	1.8	0.7
Wang et al., 2006d	SH, annual	95	11	6.6	4.0	1.3	0.6
Fu et al., 2008	SH, winter, normal	56	17	12	13	2.8	0.1
Fu et al., 2008	SH, winter, haze	311	25	17	11	0.8	0.6
(c)							
Lai et al., 2007	GZ, summer	111	14	7.8	2.6	—	4.1
Lai et al., 2007	GZ, winter	82	21	5.0	2.4	—	4.5
Hu et al., 2008	GZ, autumn	—	24	7.2	9.2	—	—
Tan et al., 2009b	GZ, winter, normal	95	8.7	4.9	3.6	2.5	0.87
Tan et al., 2009b	GZ, winter, haze	236	14	20	13	2.6	1.3
Zhang et al., 2007b	SC-Sea, winter	28	44	20	22	0.86	0.71
Zhang et al., 2007b	SC-Sea, spring	10	43	21	18	2.2	3.1
Li et al., 2010b	CB, summer	39	33	0.85	10	0.39	0.18
Li et al., 2010b	CM, summer	89	26	12	12	0.43	2.0
Li et al., 2010b	DH, summer	30	34	1.5	10	0.92	1.7
Li et al., 2010b	HN, winter	18	12	7.2	3.1	1.2	3.4

study, SOA concentration was high in the forest areas of southern and northeastern China, with a contribution of SOA to OC by $\sim 50\%$, and it was low in the broad areas north of the Yangtze River (Han et al., 2008b).

More than 100 individual organic compounds were identified and quantified in the $\text{PM}_{2.5}$ samples of Beijing (Huang et al., 2006). Table 4 summarizes the major compound classes, including n-alkanes, polycyclic aromatic hydrocarbons (PAHs), fatty acids, dicarboxylic acids, hopanes, and n-alkanols measured in several studies. Note that measurements of these compounds in larger particles, i.e., PM_{10} or TSP (e.g., Wang et al., 2009c; Zhao et al., 2009; Zhou et al., 2009), are not taken into account. Ambient concentrations of n-alkanes, PAHs, and hopanes in the urban air were 1–2 orders of magnitude higher than those in mountain aerosols and 2–3 orders of magnitude higher than those in marine samples (Wang et al., 2009a). Size distributions of these organic compounds, including water-soluble OC (WSOC), were observed; this data provided valuable information for investigating the sources of these OC and their evolution during transport from the source to background regions (Wang et al., 2009a, b). High biogenic OC tracers were found in the forests of northeastern China and southern China when OC was analyzed in aerosols, including isoprene oxidation products, α -pinene oxidation products, malic acids, sugar alcohols, and sugars that

were sampled from four ecological (boreal-temperate, temperate, sub-tropical, and tropical) regions (Wang et al., 2008d).

The fraction of WSOC generally increases from its pollution source to regional background atmospheres due to aging processes. The fraction of WSOC in total carbonaceous aerosols was found to be greater than $\sim 40\%$ at Changdao Island, a rural site in the transport path of Asian continental outflow (Feng et al., 2007). Even so, a large fraction of WSOC was observed from $\text{PM}_{2.5}$ in Beijing in some cases (Liu et al., 2008a). Food cooking can be a significant source of OC in urban areas (Huang et al., 2006; Zhao et al., 2007). The dominant homologue from Chinese cooking styles was found to be fatty acids, which constitutes 73%–85% of the quantified compounds (Zhao et al., 2007). In addition to the ion chromatography (IC) and gas chromatography (GC) mass spectrometry, new analytical technique, such as the desorption electrospray ionization mass spectrometry (DESI-MS) and single-particle aerosol time-of-flight mass spectrometry (ATOFMS), were applied to analyze organic acids (e.g., oxalic acid) in atmospheric aerosols (Li et al., 2009a; Yang et al., 2009). The averaged concentrations of formic, acetic, and oxalic acids in $\text{PM}_{2.5}$ were reported to be 110 ng m^{-3} , 78 ng m^{-3} , and 353 ng m^{-3} in Beijing, respectively (Wang et al., 2007b).

3.2.2 Formation mechanism studies

Analyses of oxalate-containing particles sampled

Table 4. (a) Summary of the classes of organic compounds (ng m^{-3}) identified from $\text{PM}_{2.5}$ in Beijing (BJ) and from $\text{PM}_{2.1}$ in Baoji (urban), Mt. Tai (MT), Okinawa Island (OI) Guangzhou (GZ). (b) Summary of the classes of organic compounds (ng m^{-3}) identified from $\text{PM}_{2.5}$ in Changdao Island (CI).

Sources	Description	n-Alkanes	PAHs	Fatty acids	Dicarboxylic acids	Hopanes	Total
(a)							
Huang et al., 2006	BJ, spring	52	19	26	20	4	359
Huang et al., 2006	BJ, summer	39	9	370	27	2	452
Huang et al., 2006	BJ, autumn	183	65	483	42	9	792
Huang et al., 2006	BJ, winter	477	283	466	16	16	1283
Huang et al., 2006	BJ, annual	163	79	385	27	7	669
He et al., 2006	BJ, summer	51	16	314	52	5	502
He et al., 2006	BJ, autumn	333	119	512	58	14	1471
He et al., 2006	BJ, winter	375	259	363	27	14	1403
Wang et al., 2009a	Urban, spring	377	153	–	–	16	–
Wang et al., 2009a	Urban, winter	1461	500	–	–	57	–
Wang et al., 2009a	MT, summer	30	8	–	–	0.1	–
(b)							
Wang et al., 2009a	MT, winter	100	49	–	–	1	–
Wang et al., 2009a	OI, spring	5	1	–	–	–	–
Feng et al., 2007	CI, spring	90	29	172	–	2.2	28
Feng et al., 2007	CI, summer	26	7	102	–	2.4	23
Feng et al., 2007	CI, autumn	55	21	131	–	2.1	20
Feng et al., 2007	CI, winter	214	123	291	–	4.6	29

in Shanghai suggest that both in-cloud processing and heterogeneous reactions occurring in hydrated/deliquesced aerosols play important roles in oxalic acid formation (Yang et al., 2009). A formation mechanism of organic salts by amines reaction with aldehydes and/or ketones in the presence of an acid was proposed to explain high molecular weight nitrogen-containing organic salts observed in urban Shanghai aerosols (Wang et al., 2010d). The results of laboratory and field experiments showed that, in addition to H_2O_2 , organic hydro-peroxides potentially play a role in the formation of WSOC via aqueous-phase reactions (Chen et al., 2008; Hua et al., 2008). The presence of $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , Na_2SiO_3 , and CaCl_2 aerosols can enhance SOA generation and increase SOA yields (Hao et al., 2007; Lu et al., 2009).

3.3 Aerosol hygroscopic properties

3.3.1 Geometric hygroscopic growth factor

The long-term trends of fog events and visibility in different regions of China were shown to be correlated with urbanization and industrial activities, and the impact of aerosol pollution has been considered to have a key role, especially in the general degradation of visibility in megacities (Deng et al., 2008a; Shi et al., 2008a; Chang et al., 2009a; Niu et al., 2010). Aerosol chemistry determines the hygroscopicity of aerosols, and the latter influences the size distribution and optical property of aerosols as a function of relative humidity (RH). The aerosol geometric (diameter) hygroscopic growth factor (GF_D), defined as the ratio of the diameter of a particle at a known elevated RH below saturation to that when nominally dry (McFiggans et al., 2006), was observed below the clouds by aircraft measurements during springtime over Huabei (Ma et al., 2010a). In this region GF_D was 1.2–1.6 at RH = 90% in the fine mode, which is typical for polluted continental environments (McFiggans et al., 2006), and GF_D was ~ 2.2 in the lower coarse mode (Ma et al., 2010a). This enhanced hygroscopicity of the coarse aerosols was probably caused by the added fraction of highly soluble inorganic acids to the mineral dust (Ma et al., 2010a). The resulting large, hygroscopic mixed dust–pollution particles are much more effective cloud condensation nuclei (CCN) than either the large insoluble desert dust or the small soluble sulfate particles (Ma et al., 2010a). The hygroscopic behavior of mineral dust was investigated by laboratory experiments (Ma et al., 2010b). It was shown that the hygroscopic behavior of $\text{Ca}(\text{NO}_3)_2/\text{CaCO}_3$ particles is identical to that of pure $\text{Ca}(\text{NO}_3)_2$ particles (Liu et al., 2008g).

3.3.2 Optical hygroscopic growth factor

The aerosol optical (scattering coefficient) hygroscopic growth factor $f(\text{RH})$, defined as the ratio of the

light-scattering coefficient of a bulk of aerosol particles at a specific wavelength (e.g., 550 nm) under wet conditions (e.g., RH=80% or 90%) to that under dry conditions (40% RH or lower), was also investigated by ground measurements (e.g., Pan et al., 2009; Yan et al., 2009; Liu et al., 2010). The average hygroscopic growth factor at an RH of 80%, $f(\text{RH}=80\%\pm 1\%)$, was $\sim 1.26\pm 0.15$ at CAMS (an urban site of Beijing) and 1.24 ± 0.11 at SDZ (a rural site of Beijing) in wintertime (Yan et al., 2009). The mean and standard deviation of $f(\text{RH}=80\%)$ observed during summertime in Beijing was 1.63 ± 0.19 (Liu et al., 2009b). The values of $f(\text{RH}=80\%)$ measured at a rural site in the Beijing-Tianjin-Tangshan region of Huabei during springtime were 1.2 ± 0.02 under dust storm conditions, 1.31 ± 0.03 under clean conditions, and 1.57 ± 0.02 during pollution periods (Pan et al., 2009). Aerosol hygroscopicity had been considered to be generally depressed with the increasing ratio of OC to ammonium sulfate in particle mass. However, an high value of $f(\text{RH})=2.21$ and high OC/ammonium sulfate ratio was observed, reflecting the special physic-chemical character of organic matter and its complex interaction with other compounds in this region (Pan et al., 2009). The means and standard deviations of $f(\text{RH}=80\%)$ observed during summertime in Gangzhou were 2.04 ± 0.28 , 2.29 ± 0.28 , and 2.68 ± 0.59 for urban aerosols, mixed aerosols, and marine aerosols, respectively. The scattering coefficient of aerosols at a rural site of the PRD was estimated to increase by factors of 1.54 and 2.31 when RH increases from 30% to 80% and from 30% to 90%, respectively (Cheng et al., 2008). Whereas the hygroscopic properties of the observed aerosols were shown to have a negative correlation with the total carbon (TC) mass fraction (Liu et al., 2008d), the major chemical components for visibility degradation in Guangzhou were proposed to be TC and sulfate on normal days and nitrate and TC on hazy days, respectively (Tan et al., 2009a). The organic coatings on individual inorganic particles may influence their surface hygroscopicity and optical properties (Li and Shao, 2010). Water dialysis of individual particles has indicated that the organic inclusions/aggregations in the K- and S-rich particles are insoluble in water but that OC from the coatings of individual particles is soluble (Li and Shao, 2010).

4. Interactions between gases and aerosols

In addition to O_3 , photochemical pollution is generally associated with high levels of fine particles. In fact, photochemical smog and haze pollution generally take place simultaneously. Through a series of photochemical processes, primary trace gases (e.g., SO_2

and VOCs) are oxidized to lower volatile species (e.g., H_2SO_4 and OVOCs), the latter being more favorable for the formation of aerosols (Fig. 1). On the other hand, huge amount of aerosols influence trace gases and radicals by changing the photodissociation rates of trace gases and by providing large areas for heterogeneous reactions of trace gases and radicals on aerosol surfaces.

4.1 New particle formation

New particle formation (NPF) events were observed in different regions of China, including Beijing (Shi et al., 2007; Wu et al., 2007; Wu et al., 2008; Yue et al., 2010), Huabei (Wang et al., 2008b; Ma et al., 2010a), the YRD (Gao et al., 2009a), the PRD (Gong et al., 2008; Liu et al., 2008c), and marine areas (Lin et al., 2007a). Even though NPF is typically indicated by the presence of particles with a diameter of ~ 2 nm, different definitions of NPF have been used, based on particle number concentrations measured at larger diameters. For example, in a surface-based observational study for urban Beijing, a burst in the nucleation mode particles was considered to represent newly formed particles if the event duration exceeded 2.5 h and the number concentration of 3–10 nm particles was higher than 10^4 cm^{-3} . In an aircraft observational study, the occurrence of NPF was assumed if the measured concentration of 5–10 nm particles exceeds 10^4 cm^{-3} (Ma et al., 2010a). These NPF events were observed in urban Beijing with a frequency of 10%–70%, based on 1-year measurements from March 2004 to February 2005, with a higher NPF frequency and larger formation rates in spring but a lower NPF frequency and higher growth rates in summer (Wu et al., 2007; Yue et al., 2010). NPF events have been thought to usually occur in conjunction with high-speed wind from the north, low relative humidity (less than 45%), and intense solar radiation in urban Beijing (Wu et al., 2007; Yue et al., 2010). However, widespread NPF events over the entire polluted area of North China were observed by aircraft measurements at higher altitudes (1–3 km) near the clouds, where relative humidity was rather high (Ma et al., 2010a).

Atmospheric aerosol nucleation has been shown to be strongly influenced by sulfuric acid (Yue et al., 2010). NPF events may involve sulfuric acid, ammonia, and oxalic acid, and organic compounds are an important contributor to the growth of the freshly nucleated particles (Yue et al., 2009, 2010). A low condensational sink (corresponding to a low particle surface-area concentration) might be one of the key factors for the occurrence of NPF events in Beijing (Wu et al., 2007) or Huabei (Wang et al., 2008b). However, no consistent connection between number concentra-

tions of nucleation mode particles and surface concentrations has been determined for all NPF events in the PRD (Liu et al., 2008c). In spite of a vast pre-existing particle surface area, intensive NPF could occur under very high H_2SO_4 concentrations driven by high levels of its gaseous precursors SO_2 and OH over the polluted Huabei region (Ma et al., 2010a). Below the clouds, the growth rate (GR) of nucleation mode particles caused by the condensation of H_2SO_4 vapor was estimated to be $5\text{--}21 \text{ nm h}^{-1}$ (Ma et al., 2010a), much higher than the range of $0.1\text{--}11 \text{ nm h}^{-1}$ inferred from ground-based measurements in Beijing (Wu et al., 2007; Yue et al., 2010). The latter referred to an overall nucleation rate by a range of compounds, including organics. High values of GR ($2\text{--}20 \text{ nm h}^{-1}$) have also been reported for the NPF events observed at a rural coastal site in the PRD (Liu et al., 2008c).

4.2 Heterogeneous reactions

4.2.1 Gas condensation on particles

In addition to NPF, the condensation of gases (mostly low volatile acids) on pre-existing particles is an important pathway of gas-to-particle conversion. The latter does not produce new particles in a sense of number, but it can change the size and hygroscopicity of particles significantly (Ma et al., 2010a). Acidic gases (HCl , HONO , HNO_3 , and SO_2), ammonia (NH_3), and water-soluble ions in $\text{PM}_{2.5}$ and PM_{10} were measured during an autumn period in the PRD and a summer period in Beijing (Hu et al., 2008; Wu et al., 2009). These data are valuable for investigating the gas-to-particle process in the atmosphere. Sulfuric acid concentrations were observed in a range of $4.5 \times 10^6 \text{ cm}^{-3}$ to $1.1 \times 10^7 \text{ cm}^{-3}$ in summer 2008 (Yue et al., 2010). When sulfate comprised the major fraction of the ultrafine particles, the condensation and neutralization of sulfuric acid together with coagulation contributed greatly to the growth of these particles (Yue et al., 2010). The mass concentration of organic compounds was significantly higher than that of sulfate (Yue et al., 2010). AMS measurements showed that particles smaller than 200 nm were predominately organic in general, while the accumulation-mode particles with diameters peaking at ~ 600 nm were dominated by sulfate and nitrate in Beijing (Sun et al., 2010a). Whereas the contribution of organic compounds to particle growth has been considered larger than that of sulfate (Yue et al., 2010), these ultrafine particles in urban environments were suggested to be primarily associated with local combustion emissions (Sun et al., 2010a). The mixed inorganic and organic species measured in the larger accumulation mode in urban Beijing might indicate that aging processes of these particulates occur in regional scales (Sun et al.,

2010a).

The condensation mode of sulfate, which is formed by the “gas-phase oxidation and condensation” process, was in the size range of 0.32–0.56 μm , and the droplet mode of sulfate, which is formed by the “condensation and liquid-phase oxidation” process, was in the size range of 0.56–1.0 μm or 1.0–1.8 μm , as measured at a rural site (i.e., Xinken) of the PRD during summertime (Liu et al., 2008b). Nitrate was distributed evenly over fine and coarse particles, and the coarse mode nitrate may have been formed by the reaction of HNO_3 with sea-salt or soil components (Liu et al., 2008b). A case study by single-particle mass spectrometry during summertime in Shanghai indicated a poor correlation between the signals of ammonium and nitrate and thus excluded the possibility of NH_4NO_3 as a major form of particulate nitrate (Wang et al., 2009g). The evolution of sulfate size distributions was observed on a time scale of several hours at a rural site (i.e., Back Garden) of the PRD during summertime (Xiao et al., 2009). The growth of sulfate in the condensation mode was mainly due to the oxidation of SO_2 ($\sim 3\text{--}21.2$ ppbv) by OH ($\sim 3.3 \times 10^6\text{--}1.7 \times 10^7$ cm^{-3}) in the gas phase, and oxygenated OC might also form as a result of the gas-phase oxidation of VOC precursors (Xiao et al., 2009).

4.2.2 Surface chemical mechanism

Laboratory experiment results showed that, in the presence of O_3 , SO_2 could be oxidized to sulfate on the surface of CaCO_3 particles, indicating that the heterogeneous reaction might be an important pathway for sulfate formation in the atmosphere (Li et al., 2006). Dust storm aerosols can interact with pollution gases and pollution aerosols during transport to Beijing, and this mechanism could be important during non-dust days (Yuan et al., 2006). Analysis of individual particles sampled during haze pollution and dust episodes in Beijing have shown that a majority of collected mineral particles are covered by visible coatings of nitrates in haze samples (Li and Shao, 2009). These nitrate coatings are strongly correlated with the presence of alkaline mineral components (e.g., calcite and dolomite), which are involved in atmospheric heterogeneous reactions with two or more acidic gases (e.g., SO_2 , NO_2 , HCl , and HNO_3) (Li and Shao, 2009). The results of another laboratory study suggest that the heterogeneous reaction of NO_2 on CaCO_3 particle is unable to compete with that of HNO_3 in the atmosphere (Li et al., 2010a). The effect of water on the heterogeneous reactions of carbonyl sulfide (OCS) on the surface of $\alpha\text{-Al}_2\text{O}_3$ and MgO has been studied in laboratory experiments; the basic, thick water layer formed on the basic component of mineral dusts may

be the primary contributor to the heterogeneous hydrolysis of OCS in the troposphere (Liu et al., 2009c).

4.2.3 Ozone chemistry effect

In addition to ozone and formaldehyde, nitrous acid (HONO) is considered to be an important OH source in the troposphere, especially in the early morning. High concentrations of HONO with a maximum of 5 ppbv were observed at both urban and rural sites of China, and even during daytime HONO remained at ~ 1 ppbv, making a dominated contribution to the overall daily HO production by photolysis (Su et al., 2008; An et al., 2009). Such strong sources of HONO cannot be explained by current gas-phase chemical mechanisms; they were attributed to heterogeneous reactions of NO_2 on ground and aerosol surfaces (Su et al., 2008).

The impact of dust on tropospheric photochemistry over Beijing megacity was evaluated using a box model (Zhu et al., 2010) and a three-dimensional regional chemical transport model (Xu et al., 2006). The simulation results improved when the heterogeneous reactions were included; the O_3 and HONO model results were close to the observations (Xu et al., 2006). The uncertainty in uptake coefficients of self-removal reactions results in the largest uncertainty in the impacts on trace gases, and the heterogeneous removal of NO_2 was found to be particularly important, because it results in significant levels of uncertainty not only for itself but also for OH and HO_2 . (Zhu et al., 2010). The uptake coefficients of several trace gases (e.g., NO_2 , SO_2 , acetone, carbonyl sulfide (OCS), and ethyl iodide) on mineral dust (e.g., calcite and mineral oxides) or black carbon surfaces were studied (Li et al., 2006, 2007b, 2010a; Jie et al., 2008; Liu et al., 2008f, 2009c; Yin et al., 2008; Cui et al., 2009).

5. Conclusions and remarks

In this paper we have provided a review of atmospheric chemistry research in China from 2006 to 2010, focusing on tropospheric ozone, aerosol chemistry, and the interactions between trace gases and aerosols over the polluted areas of China. China has concurrently suffered photochemical smog and haze pollution over the past decade. These environmental issues present a great challenge and a great opportunity for the atmospheric chemistry community, especially Chinese scientists, to greatly advance the science of atmospheric chemistry. A majority of the publications reviewed here have focused on air pollution and atmospheric chemistry related to North China (Huabei), the Yangtze River Delta (YRD), and the Pearl River Delta (PRD), especially including the

Beijing, Shanghai, and Guangzhou megacities.

Satellite data on tropospheric vertical column densities of trace gases (e.g., NO_2 and CO) have been intensively used in atmospheric chemistry research in China. The analyses have shown that the satellite can observe the regional distribution, seasonal variation, and historical trends of these air pollutants over China. Satellite data analyses together with model simulations can be an efficient method to investigate the chemical characteristics of the atmosphere in a regional scale. Notably, disagreement remains regarding the reported trend of NO_2 between satellite data and ground measurements for some regions (e.g., Beijing), and more specifically designed ground measurements (e.g., passive DOAS observations) are needed to validate the satellite data that include the "spatial averaging effect". While satellite products, provided outside of China (e.g., North America and Europe), have been applied widely, original research on the retrieval of atmospheric composition from satellite measurements is recommended for pollution studies of China.

Long-term ground observations of ozone and its precursors are still sparse in China. Analyses based on limited data showed the increasing trends of ozone at both urban (Beijing) and regional (YRD) sites with large uncertainties. Whereas the former has been associated with decreasing NO_x , the latter has been attributed to increasing NO_x . The formation of ozone has been shown to be limited by VOCs in urban areas (e.g., Beijing, Shanghai, and Guangzhou). However, the dominating controlling factors for ozone formation in polluted rural areas (e.g., Huabei, the YRD, and the PRD) remain unclear, although some studies have declared that these regions are also VOC limited. While the emission restrictions are very efficient in reducing ozone precursors and aerosol mass, the effect on ozone levels has shown to be quite complex. More comprehensive field studies are needed to quantify the effects of anthropogenic and biogenic VOCs on urban, suburban, and rural ozone of different polluted regions of China. In addition to ozone and its precursors, measurements of active intermediate products (e.g., HCHO , H_2O_2 , MAR, and MVK) and hydroxyl/peroxy radicals (e.g., HO , HO_2 , and CH_3O_2), in combination with chemical model simulations, are suggested to fully understand the atmospheric oxidation capacity and the ozone formation mechanism in different regions.

Several studies have shown that aerosols may influence ozone chemistry by changing the photolysis rates and by providing the surface for heterogeneous reactions. High concentrations of HONO, which provides an important OH source, have been observed in the urban areas even during daytime. Although the for-

mation of such huge amounts of HONO is attributed to heterogeneous reactions of NO_2 on aerosol surfaces, the detail chemical mechanism has not been fully understood yet. Although several laboratory studies of heterogeneous reactions have been reported, further work focusing on typical multi-component aerosols sampled in the natural atmosphere over China is recommended.

The NPF events have been observed in different regions of China. Concurrent measurements of sulfuric acid have shown that atmospheric nucleation is strongly influenced by sulfuric acid. In spite of a vast pre-existing particle surface area, intensive NPF events can occur under very high H_2SO_4 concentrations driven by high levels of its gaseous precursors SO_2 and OH over the polluted region. Field measurements and laboratory work have shown that heterogeneous reactions might provide an important pathway for sulfate and nitrate formation in the atmosphere. Coating by inorganic pollution acids has been shown to be very efficient in changing the hygroscopicity of natural dust aerosols. Further studies are needed to determine the role of these acids together with ammonia in the formation and growth of particles.

Large fractions of OC have been observed in different sizes of particles, including the fine and ultra-fine particles. Although more than 100 individual organic compounds have been identified and quantified in the particles, there is still large deficiency in the mass sum of identified OC compounds compared to the total OC mass. Whereas lower carbon-number compounds in the gas phase are generally investigated for ozone chemistry, many higher carbon-number compounds have been identified in the particle phase. The body of knowledge in organic chemistry regarding both gaseous and particulate phases is highly incomplete. Detection of organic compounds in aerosols, the conversion way of organic compounds from the gas phase to the particle phase, and the effect of organic compounds on the physical characters of aerosols, are recommended for more intensive study.

In situ measurements in the different regions of China have shown that photochemistry has large impacts on the formation and hygroscopicity of aerosols. Observations on the ground and by aircraft have shown larger aerosol hygroscopic growth factor in the polluted atmosphere than in the clean atmosphere. While inorganic salts play a dominant role in the enhancement of aerosol hygroscopicity, the effect of organic compounds is quite unclear. Model studies have shown that air pollution can change the chemical and physical characters of the natural atmosphere significantly, can exert strong radiative forcing, and thus can result in regional climate changes in China and East Asia (Chang

et al., 2009b; Li et al., 2009b; Liao et al., 2009; Liu et al., 2009e; Wang et al., 2010a; Zhang et al., 2010). Notably, these models generally represent an ideal case, with chemical and physical parameterizations derived from measurements taken in other regions of the world. The interactions among atmospheric chemistry, aerosols, and clouds are intricate, and models that include fully coupled atmospheric chemistry–aerosol–cloud–radiation processes are needed. Simulations constrained by comprehensive measurements are recommended to study photochemical smog, haze–clouds or haze–fog and their effects on climate change.

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