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Idir Bouarar
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Air Pollution in Eastern Asia: An Integrated Perspective

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Air Pollution in Eastern Asia: An Integrated Perspective

Springer

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Preface

Air pollution has a considerable impact on society in different regions of the world. According to the World Health Organization (WHO), more than three million individuals die prematurely each year from diseases produced or exacerbated by outdoor air pollution: heart failures (40 %), strokes (40 %), pulmonary diseases (11 %), and lung cancers (6 %). These diseases are not limited to the elderlies. Other adverse effects include crop and forest damage, reduction in visibility, enhanced acidic precipitation, eutrophication of water resources, and damages to buildings including historical monuments. By interacting with solar and terrestrial radiation and affecting cloud microphysics, air pollutants and specifically aerosol particles also contribute directly or indirectly to climate change. In recent years, the importance of feedbacks between climate and atmospheric chemistry has been the subject of considerable attention by the scientific community.

The development of economic systems based on industrial and commercial activities as well as services has been at the source of rapid urban growth in the past decades. At the beginning of the nineteenth century, only 3 % of the population lived in urban areas, and in 1950, only 80 cities had a population exceeding one million. Since the end of the twentieth century, half of the world's population lives in urban areas and almost 500 cities host more than one million inhabitants. As of 2015, 35 cities were considered to be megacities (https://en.wikipedia.org/wiki/Megacity), which are large urban areas with a population exceeding ten million. In China, the largest cities are Shanghai, Guangzhou, Beijing, Shenzhen, Wuhan, and Chengdu. The fast development of the economy in Asia has not been without detrimental environmental consequences: the level of air pollution has increased dramatically in the last decades, specifically in the corridor extending from Beijing to Shanghai. Eastern China is indeed constituted of a multitude of closely located cities, which represent substantial sources of primary and secondary air pollutants. At the same time, the dense population in these urban areas is severely affected by high levels of fine particles and oxidants. Air pollution must be regarded as a vast regional problem rather than the addition of several localized urban-scale disturbances.

Until a few years ago, surface measurements of atmospheric pollutants in China were sparse and, in fact, not easily available. The adoption of ambient air quality standards and the development of a national air reporting system have considerably improved the situation. Today, a large number of measurements are available and are very useful not only to monitor the daily variations or long-term trends in air quality, but also to analyze the effectiveness of the measures taken to reduce the sources of pollutants. Further, such data have become the basis for the initialization and the evaluation of air quality forecast models that have been developed and are now used operationally. Figure 1, which reproduces the result of an analysis performed by Rohde and Muller (2015) in eastern China and is based on measurements made at more than 1,500 monitoring stations, provides a quantitative estimate of the magnitude of air pollution. If one considers the fine particles with a radius smaller than 2.5 μm ($PM_{2.5}$) that easily penetrate deep in the respiratory system, 38 % of the population living in this part of China is subject to unhealthy

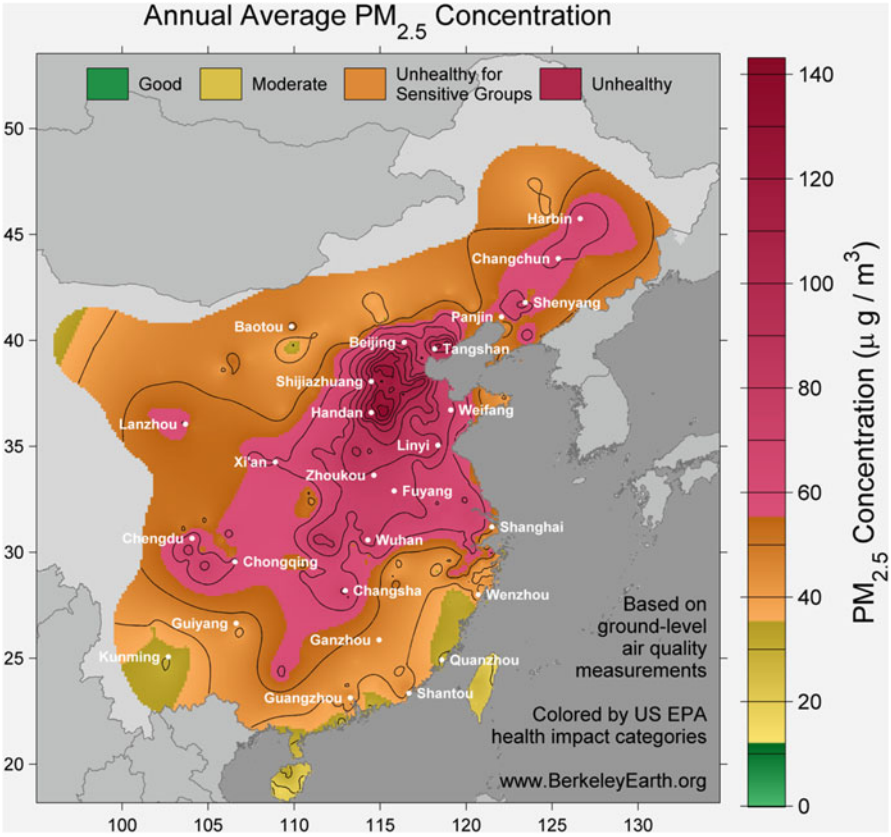

Fig. 1 Annual average pollutant concentration of $PM_{2.5}$ in eastern China in 2014 as determined by Berkeley Earth on the basis of surface measurements made at monitoring stations in China (Reproduced from http://berkeleyearth.org/)

conditions with average $PM_{2.5}$ concentrations exceeding 55 μg m^{-3} (red area on the figure). An additional 45 % of the population lives in areas labeled "unhealthy for sensitive groups" (orange area on the figure) with average $PM_{2.5}$ concentrations higher than 35 μg m^{-3}. The situation is less acute for PM_{10} particles (particles whose radius is less than 10 μm) and even less severe in the case of ozone, even though intermittent episodes of high ozone concentrations are observed during summertime. Concentrations of $PM_{2.5}$ higher than 500 μg m^{-3}, reaching in some rare occasions values of 600–1,000 μg m^{-3}, have been reported in urban areas during stable meteorological conditions in winter (Zheng et al. 2015). A challenge for the scientific community is to investigate the fundamental chemical, physical, and meteorological processes that produce and sustain elevated concentrations of secondary pollutants in eastern Asia. This question has motivated an interdisciplinary group of atmospheric scientists specialized in atmospheric chemistry and microphysics to contribute to two workshops supported by the International Space Science Institute (ISSI), one held in Beijing, China, and the second one in Bern, Switzerland. The presentations made during several intense days of discussions have been assembled in this volume.

The book is presenting an extended view on questions related to air pollution in Asia and specifically in China. It has been written by a group of experts from different continents for a broad audience involving scientists, educators and their students, environmental managers, policy-makers, as well as leaders in public administration and private corporations. It is organized around six distinct parts. The first five chapters in the first part of the volume offer a general perspective on issues related to air pollution including persistent haze events in eastern and southern Asia. The second part presents an overview of air pollution sources (i.e., anthropogenic and biomass burning sources). The third part analyzes in situ observations of chemical species in China, while the fourth part focuses on space observations of gas-phase and aerosol species. The modeling aspects are treated in the fifth part of the volume, which includes a presentation of several air quality forecast systems and an assessment of the role of urbanization on air pollution levels. Finally, the effects of air pollution on health and crop productivity in China are discussed in the last part of the book.

Even though the task is daunting, the problem of air quality can be solved. Experience acquired in other urban areas such as London and Los Angeles (Parrish et al. 2016) shows that decisive action can be taken to reduce the emissions of primary air pollutants and of the precursors of secondary pollutants. China has already taken important steps toward an effective mitigation of air pollution. The objective of this volume is to provide fundamental elements that will help decision-makers design effective science-informed policies that will lead to long-term improvements of air quality and to successfully manage short-term air pollution

episodes that substantially affect the quality of life of the people and strongly impact the economy.

Acknowledgments The editors of this volume would like to thank the International Space Science Institute in Bern, Switzerland and in Beijing, China for their support of the two workshops that led to this book. They also thank the contributors to this volume and the reviewers of the different chapters. On behalf of several contributors, the editors gratefully acknowledge the support from the European Community's Seventh Framework Programme (FP7) under the PANDA project grant agreement n° 606719. Their gratitude goes also to Springer Publishing Company and specifically to Petra van Steenbergen who has greatly facilitated the preparation of this book.

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Part I
General Perspective

Chapter 1
Overview of Persistent Haze Events in China

Renyi Zhang, Pengfei Tian, Yemeng Ji, Yun Lin, Jianfei Peng, Bowen Pan, Yuan Wang, Gehui Wang, Guohui Li, Weigang Wang, Fang Zhang, Xidan Feng, Lian Duan, Jiaxi Hu, Wilmarie Marrero-Ortiz, Jeremiah Secrest, and Min Hu

Abstract As the world's second largest economy, China has experienced severe haze pollution, with fine particulate matter (PM) reaching unprecedentedly high levels across many cities. In addition to the profound impacts on human health and ecosystems, fine PM interacts directly and indirectly with the Earth's radiation budget, influencing weather and climate. An understanding of the PM formation

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mechanism is critical in the development of efficient mitigation policies to minimize the local, regional, and global impacts. The formation mechanisms leading to severe haze episodes with exceedingly high $PM_{2.5}$ levels in China remain uncertain, and the abundance and chemical constituents of $PM_{2.5}$ depend on the complex

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interplay between meteorology, emission sources, and atmospheric chemical processes. This paper discusses the various aspects relevant to persistent haze in China, including the fundamental chemistry and meteorological conditions. The similarity and difference in PM formation between Beijing and other world regions are illustrated. Future research needs relevant to persistent haze formation in China are discussed.

Keywords Haze in China • Particulate matter • PM sources and formation • Aerosol precursors

1.1 Introduction

China has experienced persistent air pollution because of its fast-developing economy and urbanization (Zhang et al. 2015). The recent severe haze events with unprecedentedly high PM levels in China have attracted large public attention due to significantly reduced visibility and negative health effects. For example, as one of the most populous cities in the world with a population of over 21 million, Beijing is prone to develop stagnant conditions, because of surrounding mountains to the north of the city (Guo et al. 2014). In 2013 the annual level of $PM_{2.5}$ (particles with the aerodynamic diameter smaller than 2.5 μm) in Beijing was 89 μg m^{-3}, with the daily maximum $PM_{2.5}$ value reaching 568 μg m^{-3}; there were more half of the days when the $PM_{2.5}$ concentration exceeded 300 μg m^{-3}. Haze in China occurs in all seasons; the occurrence of haze episodes is more frequent and severe in wintertime, because of increased pollutant emissions from coal combustion for house heating and unfavorable meteorological conditions. The Chinese government has implemented major measures to improve air quality since the 1990s, by relocating power plants and industrial factories away from the city, replacing coal by clean energy such as electricity and natural gas, and promulgating stricter traffic emission standards (Xinhua 2012). Primary PM emissions from industry have been significantly reduced, but secondary fine PM has remained elevated, because of the presence of high levels of gaseous precursors from urban transportation and regional industrial facilities (Guo et al. 2014).

Fine PM profoundly impacts human health, visibility, the ecosystem, weather, and climate (IPCC 2013), and these PM effects are largely dependent of the aerosol properties, including the number concentration, size, and chemical composition. PM is emitted directly into (primary) or formed in the atmosphere through gas-to-particle conversion (secondary) (Fig. 1.1) (Zhang et al. 2012a; Seinfeld and Pandis 2006). Also, primary and secondary PM undergoes chemical and physical transformations and is subjected to cloud processing and transport in the atmosphere. The mechanisms leading to fine PM formation remain highly uncertain (Guo et al. 2014; Zhang et al. 2012a, 2013; Wang et al. 2010; Seinfeld and Pandis 2006; Khalisov et al. 2013), particularly for those processes related to the PM origin and growth. For example, the chemical constituents of organic carbon (OC),

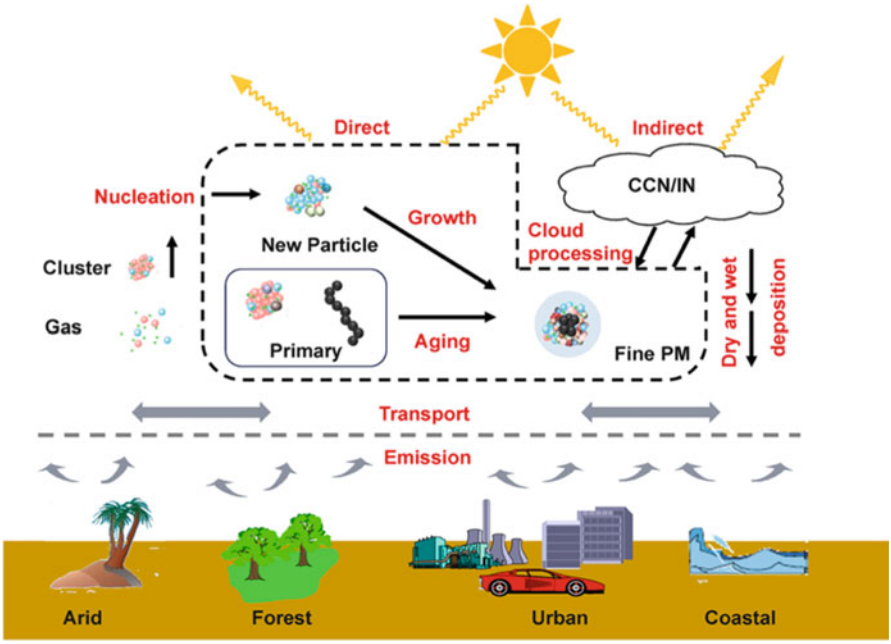

Fig. 1.1 Schematic representation of the formation, growth, and processing of atmospheric aerosols (After Zhang et al. 2015)

sulfate, nitrate, ammonium, trace metals, and elemental carbon (EC) have been commonly identified in fine PM, reflecting the diverse primary and secondary sources from the traffic, industry, biosphere, and other anthropogenic activities, along with regional components from biomass burning and agricultural activities (Guo et al. 2014; He et al. 2011; Zhang et al. 2008a, b). Since there exist a large number of natural and anthropogenic emission sources for PM, source apportionments are difficult and the currently available methodologies are rather inaccurate and often produce conflicting results (Zhang et al. 2008a, b, 2015; Guo et al. 2014; He et al. 2011).

The abundance and chemical constituents of $PM_{2.5}$ vary considerably in China, depending on complex interplay between meteorology, pollution sources, and atmospheric chemical processes (Zhang et al. 2013; Wang et al. 2011a, b, 2014; Wu et al. 2007; Suh et al. 2003; Guo et al. 2012, 2013). For example, on the basis of ambient measurements and receptor model analysis, the contribution to the annual mean $PM_{2.5}$ in Beijing has been suggested to be mainly from industrial pollution and secondary inorganic aerosol formation, but negligibly from traffic emissions (Zhang et al. 2013). In addition, meteorological conditions may govern regional and long-range transport of air pollutants (Dickerson et al. 2007; Guo et al. 2014).

Fine PM has been unequivocally associated with adverse human health impacts (NRC 2004). The human health effects of fine PM range from aggravating allergies,

the development of serious chronic diseases, to premature death (Gauderman et al. 2004; Künzli et al. 2005; Pope and Dockery 2006; Correia et al. 2013; Matus et al. 2012). Both epidemiological and toxicological studies have indicated that smaller particles are more closely linked with adverse health outcomes than larger ones (Schlesinger et al. 2006). Although ultrafine particles (less than 100 nm) contribute negligibly to the total ambient PM mass, they are typically present in high number concentrations under urban environments and have a higher probability than larger particles to deposit in the pulmonary region after inhalation. Long-term exposure to fine PM has been associated with a 6% increase in the risk of premature death (Pope and Dockery 2006; COMEAP 2006). In China, more than 20 million cases of respiratory illnesses were reported in 2007 (Millman et al. 2008).

Aerosols also exhibit a broad range of impacts on the atmosphere, directly by interfering with the solar radiative transfer and indirectly by influencing cloud formation (IPCC 2013; NRC 2004). Specifically, those particles modify the lifetime and albedo of clouds, precipitation, and lightning (Fan et al. 2007a, 2008; Wang et al. 2011a, b; Orville et al. 2001; Williams et al. 1991), modulate photochemistry (Li et al. 2005; Tie et al. 2003, 2005), promote multiphase chemistry (Seinfeld and Pandis 2006; Fu et al. 2008; Zhao et al. 2005), degrade local, regional, and global air quality (Lin et al. 2014; Li 1998, 2010), and ultimately impact the Earth energy budget (IPCC 2013). By serving as cloud condensation nuclei (CCN) or ice nuclei (IN), aerosols influence the macro- and micro- properties of clouds (Li et al. 2008a, b, 2009) and regulate the cloud lifetimes and the precipitation efficiency (Rosenfeld et al. 2008; Tao et al. 2012; Wang et al. 2013a, b; Koren et al. 2005; Fan et al. 2007b; Yuan et al. 2008). Atmospheric measurements and modeling studies have revealed invigorated convective clouds resulting from elevated aerosol levels over urban regions (Rosenfeld et al. 2008; Koren et al. 2005). Also, the aerosol effects on urban precipitation have been shown to increase the rainfall rate under clean conditions but decrease the rainfall rate under polluted conditions, suggesting a plausible distinction of the aerosol effects on precipitation between developed and developing countries (Li et al. 2008a, b). Presently, the estimates of the cloud adjustment by aerosols range from 0.06 to 1.33 W m^{-2} in the global radiative forcing budget on the top of the atmosphere, representing the largest uncertainty in climate projections of anthropogenic activities (IPCC 2013). In addition, long-range transport of urban and regional PM from Asia has been implicated in climatically altered mid-latitude cyclones over the Pacific Ocean (Li et al. 2008a, b; Zhang et al. 2007; Wang et al. 2014). There has been a growing interest in the plausible interaction between regional and Asia Monsoon (Fig. 1.2) (Wu et al. 2016).

The aerosol-cloud interaction corresponds to one of the most poorly represented processes in atmospheric models, because of largely varying aerosol properties under diverse environmental conditions. For example, the CCN activation efficiency of aerosols to form cloud droplets is dependent of hygroscopicity, which is related to the particle size and chemical compositions (Petters and Kreidenweis 2007; Ma et al. 2013; Zhang et al. 2014). In addition, atmospheric aging of primary and secondary PM considerably modifies the hygroscopicity, because of increased

Fig. 1.2 Haze, weather, and climate interaction (After Wu et al. 2016)

coating by inorganic and organic species from the secondary formation processes (Zhang et al. 2008a, b). A change in the particle hygroscopicity also impacts the atmospheric lifetime of aerosols, since wet deposition represents one of the key removal processes for aerosols. On the other hand, cloud-processing of gaseous species has been suggested to account for an important fraction of the tropospheric aerosol loading (Seinfeld and Pandis 2006; Fu et al. 2008).

The optical effects of aerosols also impact visibility and air quality. In addition to scattering light, certain aerosol types, such as black carbon (BC), mineral particles, and brown carbon, are light absorbing (Wang et al. 2013a, b; Laskin et al. 2015). Enhanced light absorption and scattering by aerosols stabilize the atmosphere, resulting in a negative feedback on air quality and inhibition of cloud formation (Barbaro et al. 2013). Furthermore, the direct and indirect effects of aerosols on clouds often operate in the opposite directions (Fan et al. 2008).

The deficiencies in our understanding of fine PM formation hinder the developments of predictive atmospheric models to simulate pollution episodes and efficient mediation policies to minimize its local, regional, to global impacts. This paper reviews the various aspects relevant to persistent haze formation in China, including the fundamental chemistry and meteorological conditions. The similarities and differences in fine PM between China and other world regions are compared and discussed. Finally, the concluding remarks and future research needs relevant to persistent haze events in China are presented.

1.2 Emissions of Primary PM and PM Precursors

Most of primary PM in China is emitted from combustion sources, such as highway vehicles, industries, residential fuel combustion, and etc. Road dust, including paved road dust and construction dust, are also important in urban areas. In Beijing, residential fuel combustion dominates the local primary PM emissions (Zhao et al. 2009). Guo et al. (2013) summarized the source apportionment results from 2000 to 2008 in Beijing: secondary formation represents the major fine PM source, except in the winter heating period when the primary PM contribution is comparable to the secondary formation. Traffic emissions represent the most important primary PM sources in Beijing in the early part of the decade; however, several control measures of vehicle emissions have been implemented since the 2008 Olympic Games, resulting in a decreased total vehicle contribution. Coal combustion also significantly contributes to the ambient fine PM, particularly during the winter. Cooking emission contributes to up to 10% of fine PM (Wang et al. 2009). The contribution of biomass burning is highly seasonal and may be important during the harvest seasons (i.e., spring and fall).

Since VOCs, NO_x, SO_2, and NH_3 represent the major precursors for secondary fine PM formation, an accurate representation of their emission sources is critical for air quality modeling (Zhang et al. 2003). For VOC emissions, major anthropogenic sources include gasoline and diesel vehicles, solvent utilization, and industries, in addition to emissions from the biosphere and biomass mass burning. For NO_x emissions, diesel and gasoline vehicles and engines, industries, and coal combustion are the common sources in urban areas. In Beijing, NO_x emission from power plants, industries and motor vehicles is estimated to account for 40%, 20% and 20% of the particle-phase nitrate (Zhang et al. 2012b). Since there is little local emission of NO_x from power plants in Beijing, most of NO_x is likely due to local transportation and regional transport from upwind source areas (Ying et al. 2014). SO_2 in urban regions is mainly from fuel combustion. In Beijing, SO_2 from regional coal combustion dominates the emissions. In urban areas with heavy traffic, NH_3 emissions from catalyst-equipped motor vehicles can significantly contribute to NH_3 concentrations (Ying and Kleeman 2006; Zhang and Ying 2010).

For air quality modeling in China, the most widely used emission inventories are the Transport and Chemical Evolution over the Pacific (TRACE-P) (Streets et al. 2003) and the Intercontinental Chemical Transport Experiment (INTEX-B) inventories (Zhang et al. 2009). In addition, the regional emission inventory in Asia (REAS) (Ohara et al. 2007) and its successor REAS2 (Kurokawa et al. 2013) have also been used in air quality modeling studies. Recently, detailed emission inventories for the entire China have been complied and employed in a number of modeling studies (Mijling et al. 2013; Stavrakou et al. 2013).

1.3 Secondary PM Formation Processes

1.3.1 New Particle Formation

Aerosol nucleation has been measured under diverse environmental conditions, and globally new particle formation (NPF) accounts for about 50% of the aerosol number production in the troposphere (Zhang et al. 2012a; Zhang 2010). NPF events have been frequently observed in many urban areas of China (Peng et al. 2014). The efficient occurrence of NPF in urban regions may be explainable because of the presence of high levels of aerosol nucleation precursors, such as sulfur dioxide, ammonia, amines, and anthropogenic VOCs (Guo et al. 2014; Zhang et al. 2008a, b, 2012a, b; He et al. 2011).

NPF events occur frequently in Beijing, according to several field campaigns, such as the Campaign of Atmospheric Research in Beijing and Surrounding Areas (CAREBejing) in the summer of 2008 (Guo et al. 2014; Wu et al. 2007; Yue et al. 2010; Wang et al. 2011b, 2015). An annual statistics of NPF measurements from March 2004 to February 2005 shows that NPF events occur during all seasons in Beijing, with the frequency of 50%, 20%, 35%, and 45% for the spring, summer, fall, and winter, respectively (Wu et al. 2007). The nucleation rates at 1.5 nm ($J_{1.5}$) and 3 nm (J_3) in NPF events observed in Beijing range from several to 100 particles cm^{-3} s^{-1} (Wu et al. 2007; Yue et al. 2010; Wang et al. 2011b, 2015), comparable with the values observed in other cities (Kulmala et al. 2004). Gaseous sulfuric acid is shown to play an important role in NPF in Beijing (Yue et al. 2010; Wang et al. 2011a, b). For instance, Yue et al. (2010) showed that the average formation rates during the CAREBeijing-2008 campaign linearly correlate with the sulfuric acid concentration with a high correlation coefficient ($R^2 = 0.85$). In addition, Wang et al. (2015) suggested that organic compounds participate in the nucleation process in Beijing, since the nucleation rate exhibits a good correlation with the concentrations of sulfuric acid and organic vapors.

1.3.2 Evolutions in the Particle Properties During Haze Events

Typically, the measured $PM_{2.5}$ properties in China exhibit a periodic cycle of 4–7 days (Fig. 1.3) (Guo et al. 2014). For example, the particle mass concentration in Beijing is less than several tens of μg m^{-3} (clean) in the beginning of each cycle and reaches within 2–4 days several hundreds of μg m^{-3} (polluted). A higher number concentration of smaller particles exists during the clean period, and a slightly lower particle number concentration of larger particles exists during the polluted period. During a pollution episode, an average daily particle mass growth of 50–110 μg m^{-3} typically coincides with a daily increase of 40–65 nm in the mean diameter. The total particle number concentration is more than 200,000 cm^{-3}

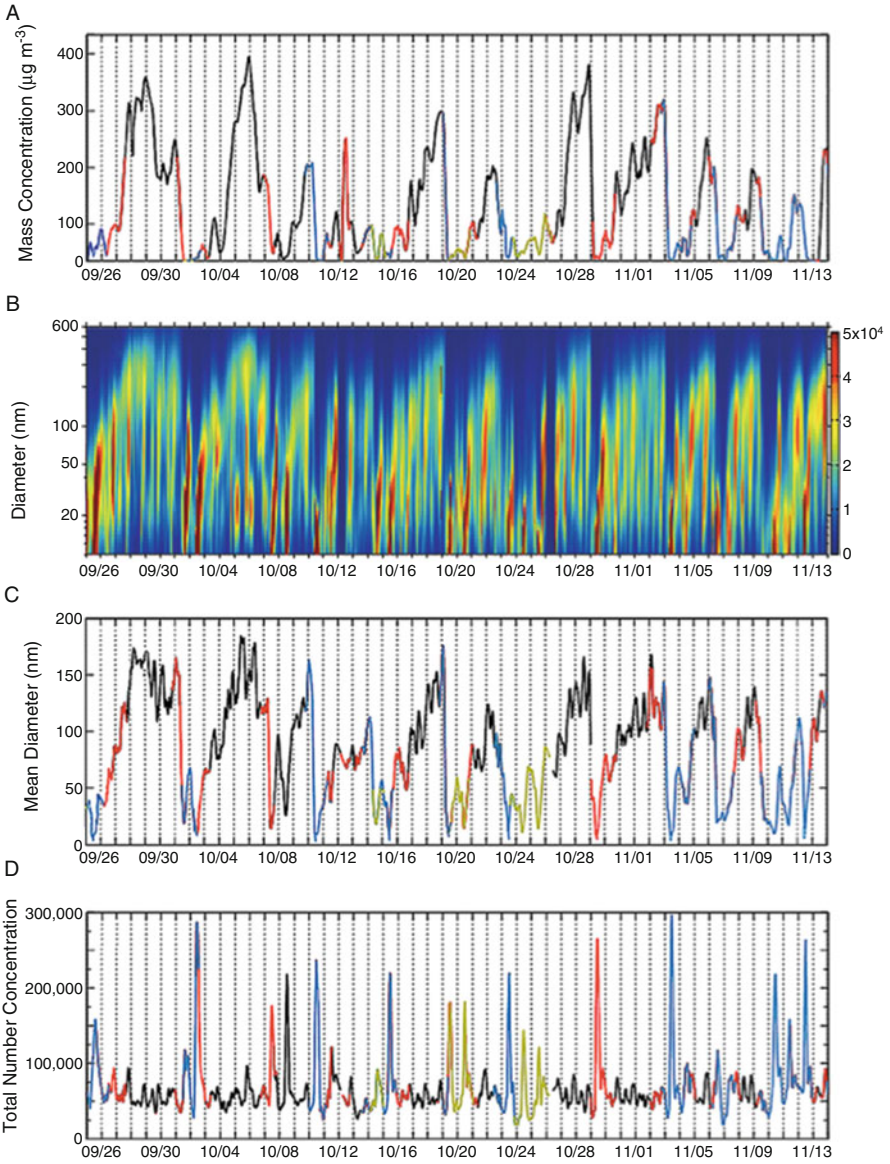

Fig. 1.3 Periodic $PM_{2.5}$ cycles in Beijing. Temporal evolutions of $PM_{2.5}$ mass concentration (**a**), number size distribution (**b**), mean diameter (**c**), and total number concentration (**d**) during the $PM_{2.5}$ events from 25 September through 14 November 2013 (After Guo et al. 2014)

during the clean period, and decreases slightly and remains at about 50,000 cm^{-3} throughout the polluted period. The periodic cycles of severe haze episodes in Beijing are largely driven by meteorological conditions; stagnation typically develops with weak southerly wind from polluted industrial source regions (Guo

et al. 2014). The average particle mass concentrations of 35 and 114 μg m^{-3} during the clean and polluted periods correspond closely to the northerly and southerly wind conditions, respectively.

The development of PM episodes in Beijing is characterized by two distinct aerosol formation processes, i.e., nucleation and growth (Fig. 1.4) (Guo et al. 2014). Nucleation consistently occurs prior to a polluted period, producing a high number concentration of nanoparticles under clean conditions. The PM growth process is reflected by the particle mass concentration exceeding several hundreds of μg m^{-3} within 2–4 days, which is accompanied by a continuous size growth from the nucleation mode particles. Figure 1.4 shows very large particle mass increases of 270 μg m^{-3} (on Sept. 27, 2013) and 210 μg m^{-3} (on Oct. 4, 2013) during the daytime (6:00 a.m. to 6:00 p.m.), when the particle mean size increases by 60–70 nm. During the clean period, there exist high concentrations of ultrafine particles, but those particles contribute negligibly to the particle mass concentration; the severe pollution episodes in Beijing are attributable to the presence of numerous large particles.

The PM chemical composition in Beijing consists dominantly of organics (44%) and nitrate (22%), followed by sulfate (17%), ammonium (15%), and chloride (2%) (Fig. 1.5) (Guo et al. 2014). Measurements show continuously increasing mass concentrations of organics, sulfate, and nitrate during the transition and polluted periods, correlating closely with the evolutions of the $PM_{2.5}$ mass concentration and mean particle size (Fig. 1.5). The organic mass fraction dominates in the clean period and decreases slightly from the clean to polluted periods. In contrast, the contributions of sulfate and nitrate to the particle mass concentration increase slightly during the pollution episodes. The primary organic aerosol mass fraction decreases during the episodes, indicating small contributions of primary particle emissions to the pollution development (Guo et al. 2014).

The gaseous aerosol precursors (i.e., VOCs, NO_x, and SO_2) are high during the pollution episodes in Beijing (Fig. 1.6). For example, the SO_2 and NO_x peak concentrations are over 40 and 200 ppb, respectively, and the aromatic hydrocarbons (xylenes and toluene) represent the most abundant types of VOCs, with the xylene peak concentration of more than 10 ppb. The photochemical oxidation and/or multi-phase reactions of VOCs, SO_2, and NO_x lead to formation of less or non-volatile species (Zhao et al. 2006; He et al. 2014), contributing to the aerosol organic, sulfate, and nitrate constituents, respectively. Furthermore, the contributions from primary emissions and regional transport of particles to the formation of severe haze episodes in Beijing have been demonstrated to be small, suggesting that regulatory controls of gaseous emissions for VOCs and NO_x from local transportation and SO_2 from regional industrial sources represent the key steps to reduce the urban PM level in Beijing (Guo et al. 2014).

The density and hygroscopicity measurements also reveal an organic dominant composition in Beijing (Fig. 1.7). The peak effective density of 1.1 g cm^{-3} during the clean day is indicative of PM dominated by organics. The increased hygroscopicity and effective density from clean to polluted periods reveal the formation of an internal mixture of secondary organic and inorganic species, with increasing

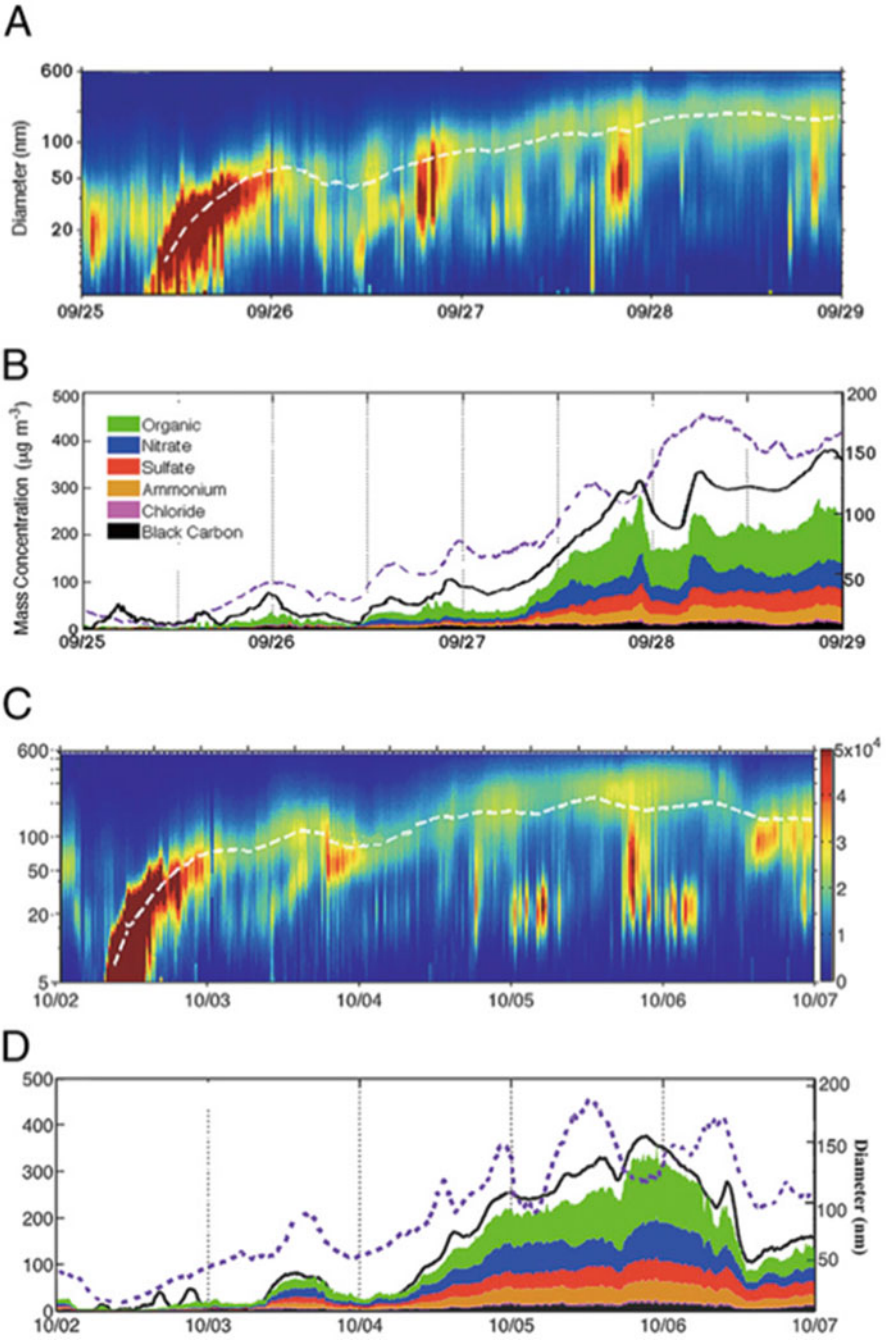

Fig. 1.4 Aerosol nucleation and growth during the PM2.5 episodes on 25–29 September and 2–7 October. (**a**, **c**) Temporal evolutions of particle number size distribution and mean diameter (*white dashed curve*) on 25–29 September (**a**) and 2–7 October (**c**). (**b**, **d**) PM2.5 mass concentration (*black solid line*), mean diameter (*purple dashed line*), and PM1 (particulate matter smaller than 1.0 μm) chemical composition on 25–29 September (**b**) and 2–7 October (**d**) (After Guo et al. 2014)

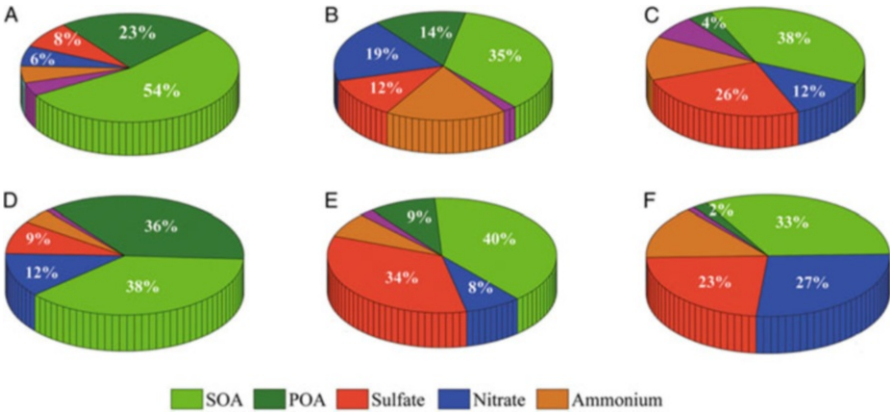

Fig. 1.5 Particle chemical compositions during the clean, transition, and polluted periods for the 25–29 September and 2–7 October episodes. (**a**–**c**) Chemical compositions for 80-nm (**a**), 100-nm (**b**), and 240-nm (**c**) particles measured by the AMS at 1500 h on 25 September, 1200 h on 27 September, and 1800 h on 28 September, respectively (After Guo et al. 2014)

contributions from sulfate and nitrate (Guo et al. 2014). The average SSA (at the wavelengths of 470 and 500 nm) in Beijing is near 0.87, indicating the presence of moderately strong absorbing aerosol and higher than those in the southern regions in China (Lee et al. 2007). The differences in SSA in urban regions may cause totally opposite aerosol radiative effects and lead to distinct feedback processes on PM formation and removal (Yang et al. 2013, Yang and Li 2014).

The efficient aerosol nucleation and growth in Beijing are clearly distinct from those in other urban cities. For example, the variations in the PM properties (i.e., particle number, size, and mass concentration) in Houston, LA, and Mexico City typically exhibit a clear diurnal characteristic. In contrast, the aerosol properties in Beijing exhibit continuous evolutions from the nucleation mode particles over an extended period of 2–4 days, yielding numerous large particles during the pollution episodes. The considerably more efficient aerosol nucleation and growth processes in Beijing are explained by much higher concentrations of aerosol precursor gases, i.e., anthropogenic VOCs (aromatics), NO_x, and SO_2 than those in the other three cities (i.e., Fig. 1.6). On the other hand, the particle compositions in Beijing exhibit a general similarity to those commonly measured in many global urban areas, consistent with the chemical constituents that are dominated by secondary aerosol formation (Fig. 1.8). For example, the PM mass fractions of organic, sulfate, and nitrate of 44%, 17%, and 22% in Beijing are nearly identical to those of 44%, 17%, and 24% in Los Angeles, respectively, likely reflecting the chemical constituents dominantly from traffic emissions (i.e., VOCs and NO_x). Table 1.1 summarizes the air pollutant characteristics between Beijing and other world cities.

Most recently, it has been shown that severe haze formation involves a coupling between atmospheric chemical processes and the height of the atmospheric planetary boundary (PBL): rapid aging of BC particles in China considerably enhances

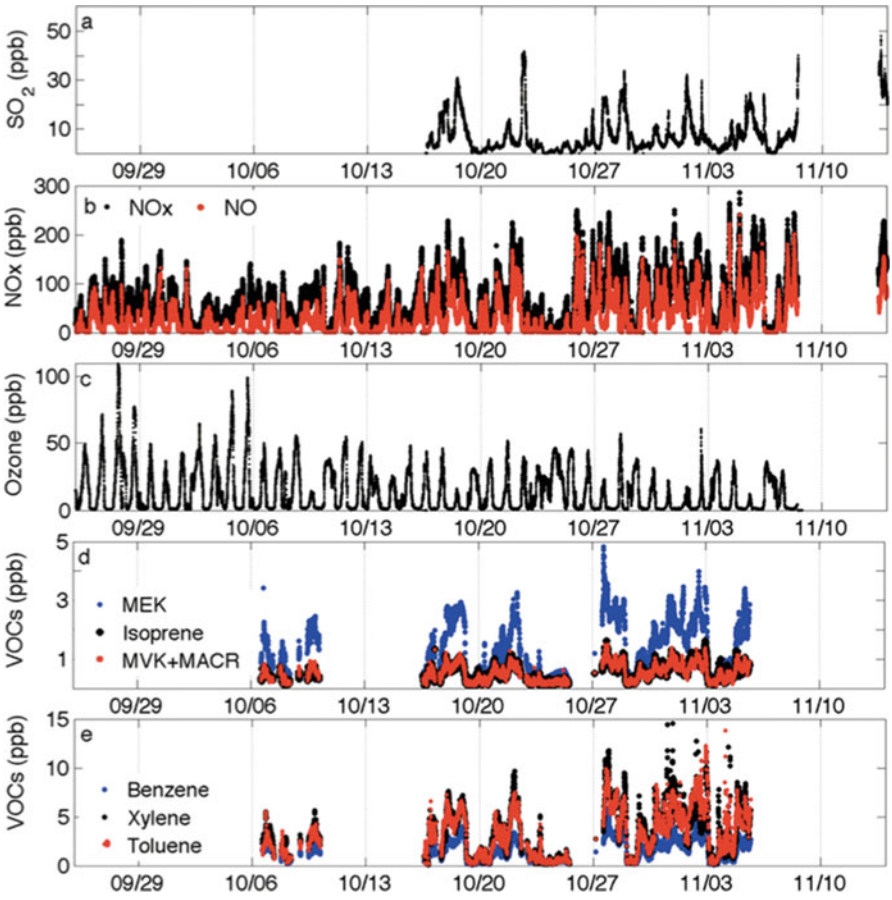

Fig. 1.6 Measurements of gaseous species. (**a**–**c**) Concentrations of SO2 (**a**), NOx (**b**), and O3 (**c**) measured between 25 September and 13 November. (**d**, **e**) Biogenic species, i.e., isoprene, methacrolein (MACR), methyl vinyl ketone (MVK), and methyl ethyl ketone (MEK) (**d**) and aromatic hydrocarbons, i.e., xylene, benzene, and toluene (**e**) (After Guo et al. 2014)

light absorption and contributes to stability and pollutant accumulation in China (Peng et al. 2016). Furthermore, there exists an anti-correlation between the photochemical activity and aqueous chemistry during the severe haze evolution (i.e., from the clean, transition, to polluted periods) in China, the sulfur to sulfate conversion plays a central role in facilitating aqueous production of the major secondary constituents (Wang et al. 2016). Clearly, the multi-sources and high-emissions of the PM precursors are the main reason for persistent haze occurrence in China (Zhang et al. 2015; Guo et al. 2014).

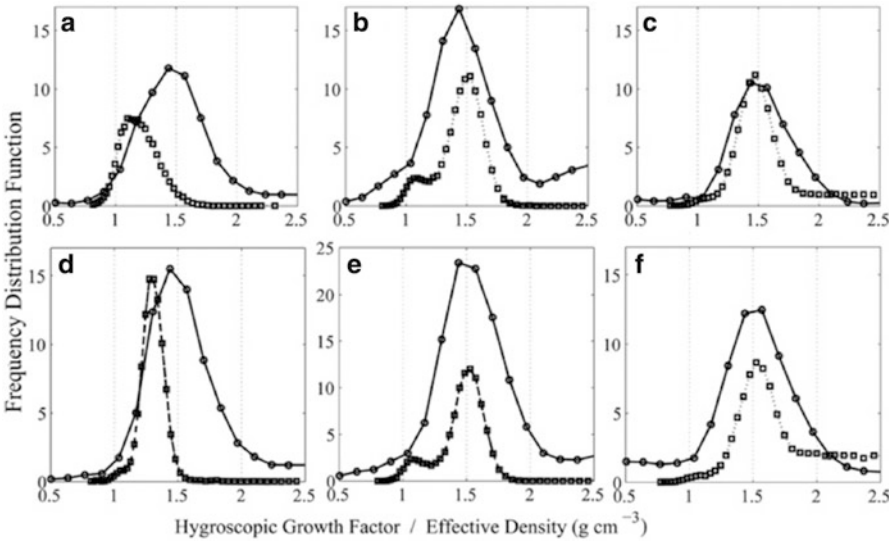

Fig. 1.7 Particle hygroscopicity and density during the clean, transition, and polluted periods for the 25–29 September and 2–7 October episodes. (**a**–**c**) Effective density (*solid line, circles*) and hygroscopicity (*dashed line, squares*) for 46-nm (**a**), 97-nm (**b**), and 240-nm (**c**) particles at 1500 h on 25 September, 1200 h on 27 September, and 1800 h on 28 September, respectively. **d**, **f** Effective density (*solid line, circles*) and hygroscopicity (*dashed line, squares*) for 46-nm (**d**), 97-nm (**e**), and 240-nm (**f**) particles measured at 1500 h on 2 October, 1200 h on 4 October, and 1200 h on 5 October, respectively (After Guo et al. 2014)

Table 1.1 Summary of air pollutant characteristics in Houston, Los Angeles, Mexico City, and Beijing (After Zhang et al. 2015)

	Houston, Texas	Los Angeles, California	Mexico City, Mexico	Beijing, China
Population	6.2 million	18 million	21.2 million	21.2 million
City (Metropolitan) area	1625 (10,062) km^2	1302 (87,940) km^2	2072 (3540) km^2	3820 (16,801) km^2
Number of vehicles	3.3 million	6.1 million	3.5 million	5.4 million
Primary pollutants	NO_x, VOCs (light olefins)	NO_x, VOCs (aromatics)	NO_x, VOCs (Alkanes, aromatics)	NO_x, SO_2, VOCs (aromatics)
Secondary pollutants	Ozone	Ozone, PM	Ozone, PM	PM
Average annual SO_2	0.35 ppb	0.37 ppb	2.50 ppb	7.8 ppb
Average annual $PM_{2.5}$ concentration	12 μg m^{-3}	18 μg m^{-3}	27 μg m^{-3}	102 μg m^{-3}
Average measured Particle concentration	10,700 cm^{-3}	20,000 cm^{-3}	21,000 cm^{-3}	72,900 cm^{-3}
Highest measured PM concentration	40,000 cm^{-3}	50,000 cm^{-3}	40,000 cm^{-3}	250,000 cm^{-3}
Dominant PM Composition	Organics (35%) and sulfate (30%)	Organics (44%) and nitrate (24%)	Organics (66%) and sulfate (14%)	Organics (44%) and nitrate (22%)

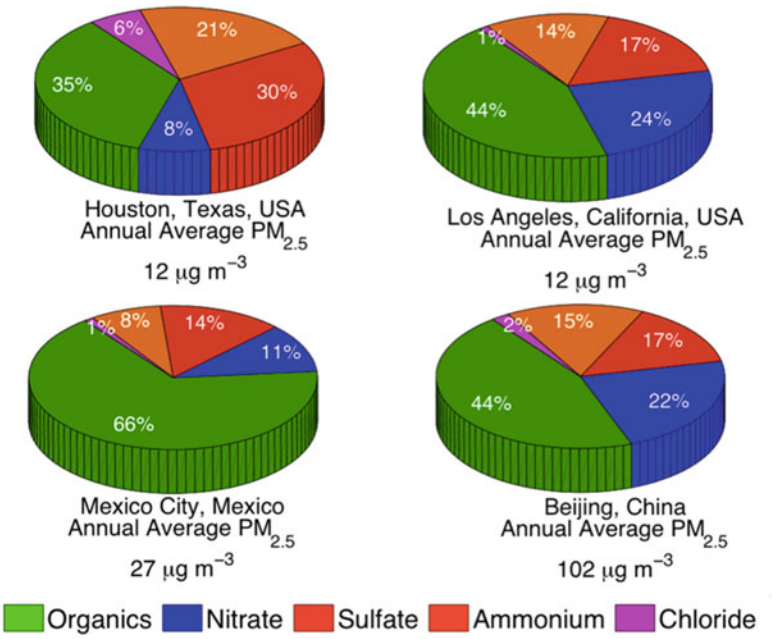

Fig. 1.8 Non-refractory compositions of particulate matter determined by an aerosol mass spectrometer in Houston, Los Angeles, Mexico City, and Beijing (After Zhang et al. 2015)

1.4 Challenges and Future Research Needs

The formation of persistent haze in China exhibits distinct characteristics in the particle properties, dependent of the emission sources, formation mechanisms, removal, and meteorological conditions. The secondary PM formation in China is most prominent, characterized by two aerosol formation mechanisms, i.e., nucleation and growth. Typically, there exist clear diurnal variations in the PM number, size, and mass concentration in other regions, reflecting the interplay between primary emissions, new particle formation, photochemical growth, removal, and the PBL variation. In contrast, haze events in China typically exhibit a periodic cycle of 4–7 days (Fig. 1.3). Aerosol nucleation consistently precedes a polluted period in Beijing, producing a high number concentration of nano-sized particles under clean conditions, and accumulation of the particle mass concentration exceeding several hundred micrograms per cubic meter is accompanied by a continuous size growth from the nucleation-mode particles over multiple days to yield numerous large particles (Fig. 1.3).

The efficient aerosol nucleation and growth in China are attributable to highly elevated concentrations of gaseous aerosol precursors, most noticeably anthropogenic VOCs, NO_x, and SO_2 emitted from local transportation and regional industrial activities (Fig. 1.6). For example, during pollution episodes the peak SO_2 and

NO_x concentrations exceed 40 and 200 ppb, respectively, and the peak xylene and toluene concentrations exceed 10 ppb in Beijing (Guo et al. 2014). These concentrations are significantly higher than those observed in the other urban regions worldwide.

The combination of the enormously efficient aerosol nucleation and growth over an extended period leading to the severe $PM_{2.5}$ episodes in Beijing is uniquely different from those typically observed in the urban regions of other countries and pristine environments worldwide. The efficient secondary formation in China is also reflected by the much higher annual $PM_{2.5}$ mass concentration (102 μg m^{-3}) than those in Houston (12 μg m^{-3}), Los Angeles (12 μg m^{-3}), and Mexico City (27 μg m^{-3}) (Fig. 1.8). On the other hand, the efficient secondary aerosol formation processes identified in China may also be characteristic of those in other urban cities of the developing world (such as India and other Asian developing countries), because of rapidly growing economy and fast urbanization leading to lower standards but higher rates for air pollutant emissions.

The currently available atmospheric chemical mechanisms in the gas and aqueous phases have yet to quantitatively account for the rapid accumulations of the PM chemical constituents under polluted conditions in China (i.e., Fig. 1.3), particularly for the formations of secondary organic matter, sulfate, and nitrate (Wang et al. 2013a, b). It is plausible that there exist synergetic effects among the various organic and inorganic compounds (i.e., organics, sulfate, nitrate, basic species, etc.) to enhance the particle growth (Guo et al. 2014). For example, the presence of basic species (i.e., ammonia and amines) may not only considerably enhance sulfate and nitrate formation, but also promote secondary organic matter production under polluted environments (Qiu and Zhang 2013).

The chemical mechanisms for the formation of secondary organic matter may also be distinct in China, leading to not only different PM production rates, but also different product types and aerosol properties. For example, while gas-particle partitioning of organic matter is linearly dependent of the gaseous reactant concentrations, oligomerization for hydration of small α-dicarbonyls and polymerization for aldol condensation of large aldehydes correspond to second- or higher-order reactions with respect to the organics to form high-molecular-weight species in the particle-phase. The latters are expected to occur more efficiently with elevated gaseous concentrations under polluted conditions. In addition, while the gas-phase oxidation leading to formation of semi- and low volatility and subsequent gas-particle partitioning increase particle hygroscopicity, oligomerization/polymerization generally leads to decreased hygroscopicity (Guo et al. 2014). Furthermore, organic matter formed from oligomerization/polymerization may exhibit enhanced light absorption in the ultraviolet and visible ranges (Wang et al. 2013a, b). Presently, very few of the multi-phase reactions have been incorporated into atmospheric models to assess their roles in the formation, growth, transformation, properties, and impacts of urban fine PM (Xu et al. 2014).

To improve the understanding on the formation of urban fine PM, future laboratory kinetic and mechanistic measurements of multi-phase chemistry are needed, particularly for those leading to secondary organic matter, sulfate, and

nitrate formations under atmospherically relevant conditions, i.e., RH, temperature, and S concentrations. Laboratory experiments are also needed to quantify the different aerosol properties (i.e., hygroscopicity and optical properties) formed from the multi-phase reactions involving organic and inorganic species. Those experimental results on atmospheric multi-phase chemistry may assist in not only atmospheric modeling, but also interpretation and identification in field measurements for urban fine PM.

Further atmospheric measurements are needed to monitor simultaneously the gaseous aerosol precursors and a comprehensive set of aerosol properties (i.e., particle number, size, chemical composition, morphology, light scattering and absorption, and hygroscopicity, etc.). Atmospheric field measurements are crucial to providing the information on the temporal and spatial distributions of gaseous concentrations and PM properties under diverse urban environments. To achieve the highest level of chemical speciation, the development of more advanced analytical techniques is required, including identification and quantification of the diverse gaseous aerosol precursors (i.e., carbonyls, sulfuric acid, nitric acid, organic acids, basic species, etc.) present in ambient air at ppb or lower levels and detailed chemical composition characterization of aerosols from the molecular cluster ($<$1 nm), sub-nm, to sub-μm size ranges. Simultaneous measurements of the particle density, hygroscopicity, volatility, and optical properties can further assist in identification of the particle chemical compositions, since those particle properties are distinct for different PM types (Guo et al. 2014; Levy et al. 2013, 2014). Furthermore, improved physically based parameterizations of aerosol nucleation and growth developed on the basis of and validated against laboratory and field studies are required for incorporation into atmospheric models.

The human health effects of high concentrations of ultrafine particles produced from NPF under urban environments need to be carefully evaluated in China. For example, the PM episodes in Beijing typically consist of the clean and polluted periods, characterized by exceedingly high concentrations of ultrafine particles ($>10^5$ particles cm^{-3} for particles smaller than 100 nm) and numerous large particles (about 5×10^4 for particles larger than 150 nm), respectively. While many epidemiological studies have emphasized the correlations of the various health syndromes with the $PM_{2.5}$ levels (NRC 2004; Pope and Dockery 2006), little is known on the potential health outcomes of highly elevated concentrations of ultrafine particles produced from NPF under clean conditions.

The regional and climatic impacts of urban fine PM need to be further assessed to quantify their direct and indirect radiative forcing. Considering the profound societal implications of urban fine PM on human health, ecosystem, weather, and climate, it is imperative that sound science is employed to develop effective regulatory polices to mediate the local, regional, and global impacts of urban fine PM. The large urban centers/megacities may also represent the ideal locations to best achieve the co-benefits in simultaneously controlling air pollution and mitigating climate change (Zhu et al. 2012). Furthermore, it is essential that knowledge is transferred from well to less researched urban areas and the experience in