

## • Original Paper •

**Growth Rates of Fine Aerosol Particles at a Site near Beijing in June 2013**Chuanfeng ZHAO<sup>\*1,2</sup>, Yanan LI<sup>1</sup>, Fang ZHANG<sup>1</sup>, Yele SUN<sup>3</sup>, and Pucai WANG<sup>3</sup><sup>1</sup>*State Key Laboratory of Earth Surface Processes and Resource Ecology, and College of Global Change and Earth System Science, Beijing Normal University, Beijing 100875, China*<sup>2</sup>*Joint Center for Global Change Studies, Beijing 100875, China*<sup>3</sup>*Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100875, China*

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## ABSTRACT

Growth of fine aerosol particles is investigated during the Aerosol–CCN–Cloud Closure Experiment campaign in June 2013 at an urban site near Beijing. Analyses show a high frequency (~ 50%) of fine aerosol particle growth events, and show that the growth rates range from 2.1 to 6.5 nm h<sup>-1</sup> with a mean value of ~ 5.1 nm h<sup>-1</sup>. A review of previous studies indicates that at least four mechanisms can affect the growth of fine aerosol particles: vapor condensation, intramodal coagulation, extramodal coagulation, and multi-phase chemical reaction. At the initial stage of fine aerosol particle growth, condensational growth usually plays a major role and coagulation efficiency generally increases with particle sizes. An overview of previous studies shows higher growth rates over megacity, urban and boreal forest regions than over rural and oceanic regions. This is most likely due to the higher condensational vapor, which can cause strong condensational growth of fine aerosol particles. Associated with these multiple factors of influence, there are large uncertainties for the aerosol particle growth rates, even at the same location.

**Key words:** growth rate, fine aerosol particle, Xianghe**Citation:** Zhao, C. F., Y. N. Li, F. Zhang, Y. L. Sun, and P. C. Wang, 2018: Growth rates of fine aerosol particles at a site near Beijing in June 2013. *Adv. Atmos. Sci.*, **35**(2), 209–217, <https://doi.org/10.1007/s00376-017-7069-3>.**1. Introduction**

Aerosol is ubiquitous in Earth's atmosphere and causes considerable impacts to society, including changes in climate, atmospheric chemistry and human health (IPCC, 2007). Fine aerosol particles have strong negative effects on atmospheric visibility and human health by entering the respiratory, circulatory and nervous systems (Harrison and Yin, 2000; Kreyling et al., 2004). The climate impact of aerosol is one of the largest uncertainties in current climate model simulations (IPCC, 2007), and atmospheric particle formation has been identified as one of the most important aerosol processes that should be explicitly treated in next-generation climate models (Ghan and Schwartz, 2007). Many studies have focused on both the formation and growth of fine aerosol particles (Kulmala et al., 2004a, 2012; Spracklen et al., 2006, 2010; Yu et al., 2010).

While aerosol particle formation and growth events can be identified based on the evolution of size distributions and particle number concentrations following the definition of Kulmala et al. (2004a), their quantitative observation requires measurements of aerosol particle size distributions down to

sizes as small as 3 nm in diameter (McMurry, 2000). Actually, even 3 nm is not small enough to study the aerosol formation process, which is mainly the nucleation process (Kulmala et al., 2004b). Therefore, using aerosol measurements from differential mobility particle sizer or scanning mobility particle sizer (SMPS) systems with a cutoff size of 3 nm or even larger, is not suitable for the detection of nucleation and the initial steps of particle growth (Kulmala et al., 2004b). In this paper, we only examine the growth of atmospheric fine aerosol with a size range of 10–100 nm without considering the formation process.

The growth of atmospheric fine aerosol particles is a frequent global phenomenon, and one of the major sources for accumulation-mode (0.1–2.0 μm) aerosol particles in both clean and polluted atmospheres. When fine aerosol particles grow large enough to serve as cloud condensation nuclei (CCN), they modify Earth's radiation budget by reflecting solar radiation directly or indirectly, e.g. via cloud albedo (Twomey, 1974; Garrett and Zhao, 2006; Lubin and Vogelmann, 2006; Spracklen et al., 2008; Zhao et al., 2012), cloud lifetime (Albrecht, 1989), or cloud invigoration (Rosenfeld et al., 2008; Li et al., 2011). Thus, it is important to study the growth of fine aerosol particles.

In recent times, Beijing has continuously experienced many heavy haze events almost every year, and vehicle emis-

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sions through secondary aerosol formation have become one of the major sources of atmospheric pollution in city. It is therefore important for us to know the frequency of fine aerosol particle growth events and understand the growth rate (GR) of fine aerosol particles.

In this study, we examine the GRs of fine aerosol particles near Beijing based on 10 fine aerosol particle growth events found during a field experiment in June 2013. We then try to understand the different GRs over several locations found by various studies.

## 2. Field experiment

A comprehensive intensive observation period field experiment concerning aerosol and clouds, called the Aerosol–CCN–Cloud Closure Experiment (AC3E), was carried out during 1–30 June 2013 at Xianghe (39.80°N, 116.96°E; 35 m above sea level) in Hebei Province, China, located about 60 km southeast of Beijing. Figure 1 shows the location of the site. It lies in a plain area surrounded by agricultural land, densely occupied residences, and light industry. Situated close to the local downtown area with a population of 50 000 and between two megacities (Beijing and Tianjin), the site experiences frequent pollution plumes deriving from urban, rural or mixed origin.

AC3E provided a series of observations such as aerosol particle size distribution, mass distribution, chemical composition, cloud condensation nuclei and meteorological status. The present study mainly uses the measurements of aerosol particle size distribution and particle chemical composition. Aerosol particle size distribution (10–500 nm) is measured with TSI's SMPS 3082. Before entering the instrument, the air flow is dried with a silica gel diffusion dryer to an average relative humidity of < 5%. The measurements of non-refractory submicron (40 nm to 1  $\mu$ m) aerosol species including organics, sulfate, nitrate, ammonium and

chloride are obtained with an Aerodyne Aerosol Chemical Speciation Monitor (Sun et al., 2012). Detailed descriptions of these instruments can be obtained from the instrument manuals and corresponding references (e.g. Sun et al., 2012). CCN data, which are obtained at supersaturations of 0.2%, 0.5% and 0.8% using a Droplet Measurement Technologies continuous-flow CCN counter (Lance et al., 2006), are also used in this study to examine the impact of fine aerosol particle growth.

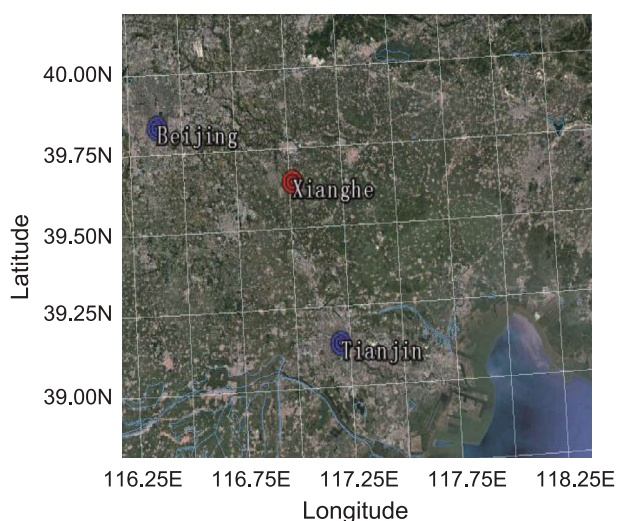
The meteorological conditions were recorded during the campaign. For most cases, the weather was hot and wet, with an average temperature of 23.6°C and an average ambient relative humidity of 72.3%.

## 3. Theory and method

### 3.1. Fine aerosol particle growth

Although not the focus of our study, we begin by providing a brief summary of the mechanisms for fine aerosol particle formation. As shown in Kulmala et al. (2000), several nucleation mechanisms have been proposed to explain fine aerosol particle production, along with meteorological-related nucleation enhancement processes such as turbulent fluctuations, waves and mixing (Easter and Peters, 1994; Nilsson and Kulmala, 1998). Two fine aerosol particle formation theories—binary nucleation theory (water and sulfuric acid) (Doyle, 1961; Raes et al., 1992; Kulmala et al., 1998) and ternary nucleation theory (sulphuric acid–ammonia–water) (Coffman and Hegg, 1995; Korhonen et al., 1999)—have indicated the importance of sulfuric acid and ammonia to the formation of fine aerosol particles.

Our focus is the growth of fine aerosol particles. As shown in Table 1, several mechanisms for fine aerosol particle growth have been proposed by Kulmala et al. (2004b). The study indicated that the first, third and fourth mechanisms shown in Table 1 do not require additional vapors other than those participating in the nucleation processes (which are the major mechanisms for fine aerosol particle formation), whereas the other two mechanisms do. In general, condensational growth associated with mechanisms 1–3 is more significant when concentrations of condensable vapors are higher, and the efficiency of these three mechanisms should decrease with growth time and then particle sizes due to the consumption of condensable vapors; self-coagulation efficiency increases with sizes during the aerosol growing stage; and multi-phase chemical reactions are favored by an acidic environment. Recently, Yue et al. (2010) indicated that fine aerosol particle growth process is mainly caused by three mechanisms: intramodal coagulation, extramodal coagulation with larger pre-existing particles, and vapor condensation. Different from Kulmala et al. (2004b), Yue et al. (2010) indicated negative effects of extramodal coagulation for the growth of fine aerosol particles: the growing aerosol particles can be scavenged or removed by pre-existing larger particles. We should note that many studies (e.g., Kulmala et al., 2005; Kuang et al., 2012) show the primary mechanism for the



**Fig. 1.** Location of the site at Xianghe, where we carried out the AC3E campaign field measurements.

**Table 1.** Mechanisms listed in Kulmala et al. (2004b) for fine aerosol growth.

No.	Mechanism	Characteristics
1	Condensation of nucleating vapors	Growth rate increases rapidly and then stays relatively constant
2	Growth by other vapors	Soluble vapors can participate in the growth of fine aerosol particles after activation (nano-Kohler mechanism) and insoluble vapors after heterogeneous nucleation. After these initial steps, multicomponent condensation of organic and inorganic vapors will continue the growth.
3	Charge-enhanced condensation	This effect decreases rapidly with increasing size.
4	Self-coagulation	Self-coagulation effect increases with aerosol sizes.
5	Multi-phase chemical reactions	These effects are favored by an acidic environment.

growth of fine aerosol particles is the condensation of sulfuric acid vapor and low-volatility organic vapors. In summary, the growth of fine aerosol particles should be strongly associated with the condensation of sulfuric acid vapor and low-volatility organic vapors, the concentration of pre-existing large size aerosol, the concentration of fine aerosol particles, and favorable meteorological conditions.

**3.2. Method for growth rate calculation**

Fine aerosol particle growth events are identified in this study based on the evolution of aerosol particle size distributions following the definition of Kulmala et al. (2004a). Specifically, an obvious growth trend in particle size distributions can be found during fine aerosol particle growth events.

Following the expression in Heintzenberg (1994), GR is defined as the growth rate of fine aerosol particles at mean diameter  $D_m$  within a time period  $\Delta t$ :

$$GR = \frac{\Delta D_m}{\Delta t} \tag{1}$$

Note that the mean diameter  $D_m$  is a mean geometric diame-

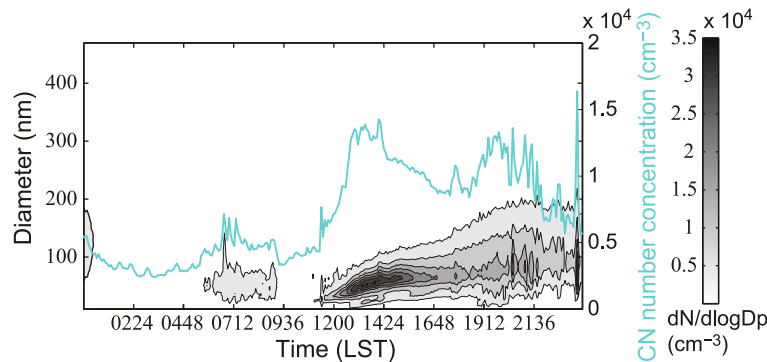
ter of a log-normal ultrafine aerosol particle mode, which has been fitted to the number size distribution. GR can also be expressed as (Kulmala et al., 1998)

$$\frac{dD_p}{dt} = \frac{4m_v\beta_M DC}{\rho D_p} \tag{2}$$

where  $D_p$  is the particle radius,  $m_v$  is the molecular mass of condensable vapor,  $D$  is the diffusion coefficient,  $C$  is the vapor concentration,  $\rho$  is the particle density, and  $\beta_M$  is the transitional correction factor for the mass flux. Equation (2) shows that GR should be related to condensable vapor, particle size and particle concentration. As indicated earlier, both condensation and coagulation play important roles for fine aerosol particle growth.

Figure 2 shows the temporal variation of aerosol particle size distribution and total aerosol number concentration in the size range from 10 nm to 500 nm on 17 June 2013. Based on the identification method described above, a fine aerosol particle growth event occurs on this day. The aerosol number concentration shows a sharp increase in the initial stage (1100–1400 LST) of this growth event due to the conversion of fine aerosol from sizes below 10 nm to above 10 nm. Considering the two facts that the aerosol number concentration does not change much in the initial stage (such as 10–50 nm) and there are generally heavy emissions of  $NO_x$  and  $SO_2$  gases from strong traffic pollution and burning coal in this region, the fast growth of fine aerosol particles in the initial stage should be associated with condensational growth, as shown in Eq. (2). In the later stage, the aerosol number concentration decreases gradually, which should be due to intramodal and extramodal coagulations. Interestingly, there is a jump in aerosol number concentration between 1900 and 2100 LST, which should be due to the aerosol particles from other sources such as biomass burning. For the measured size range between 10 and 500 nm, there is a clear increasing trend in aerosol particle sizes with time during 1100–2200 LST. It is highly likely that new aerosol particle formation occurs at times before 1100 LST, such as 0900–1100 LST, which is consistent with the findings of many other studies (e.g., Wu et al., 2007; Zhang et al., 2011).

Figure 3 illustrates the calculation of GR using Eq. (1)



**Fig. 2.** An example of a fine aerosol particle growth event that occurred on 17 June 2013.



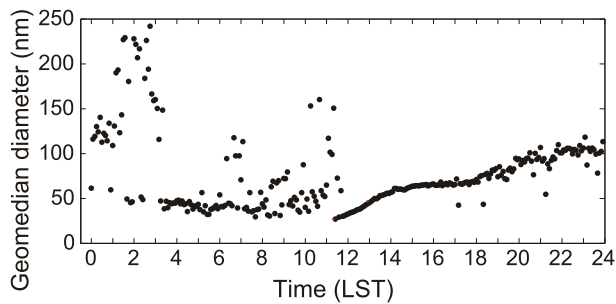


Fig. 3. An example of the calculation of GR on 17 June 2013.

for fine aerosol particles measured at Xianghe on 17 June 2013. Using the time series data of aerosol particle size distributions,  $\Delta D_m$  and  $\Delta t$  can be easily estimated. The fine aerosol particle GR is slightly larger for the period 1100–1430 LST than for 1430–1800 LST, which could be associated with the decreasing condensation efficiency and increasing extramodal coagulation efficiency while the intramodal coagulation efficiency also increases. After 1800 LST, the GR becomes a little larger again. Roughly estimated, the mean particle size increases from 25 nm to 100 nm from 1100 to 2230 LST, corresponding to a mean GR of  $6.5 \text{ nm h}^{-1}$ .

Large uncertainties in the estimations of GR could exist. As indicated by Kulmala et al. (2004a), the main problem for GR calculation is to distinguish between fine mode and pre-existing large aerosol particles. The GR is defined as the slope of the linear fitting line between aerosol particle mean size and time. However, different from that shown in Shi and Qian (2003), the mean sizes of fine aerosol particles usually do not show a perfect positive linear relation with time because of two issues. One is the existence of large sized background aerosol particles, and the other is the fluctuation of the particle size distributions. Both make it difficult to identify

the size classes that belong to the fine aerosol particle growth events. Unless it is very clear, we need to make a good guess based on our knowledge. Sometimes, it is even difficult to give an accurate estimate for the start and end points of fine aerosol particle growth events, which usually also affects the calculation of the fine aerosol particle GR. Considering these factors of influence, uncertainties in determined GRs are also examined in this study. For example, the uncertainty for the determined GR values in Fig. 3 is estimated as  $\pm 0.8 \text{ nm h}^{-1}$ .

The observed particle size distributions can be classified into three modes: “nucleation mode”, with size  $D_m \leq 25 \text{ nm}$ ; “Aitken mode”, with a size range of 25–100 nm; and “accumulation mode”, with size range of 100–1000 nm. The nucleation mode and Aitken mode aerosol particle GRs are generally different. Considering the aerosol size range measured here is between 10 and 500 nm, the average GRs of fine aerosol particles in the size range of 10–100 nm are examined with Eq. (1) in this study.

## 4. Results and discussion

### 4.1. Fine aerosol particle growth at Xianghe

Figure 4 shows the temporal variation of measured aerosol size spectra between 10 and 500 nm during the AC3E campaign period of 9–25 June 2013, except 18 June when an instrument error occurred. While not always obvious, fine aerosol particle growth events occur on days 9, 10, 11, 12, 13, 17, 19, 20, 21 and 23. The frequency of days that fine aerosol particle growth events occur is around 50%. Assuming these fine aerosol particles are formed locally, the occurrence frequency of fine aerosol growth events is much larger than that found by Wu et al. (2007) and Shen et al. (2011), which show about 20% and 12% respectively in summer. Note that Wu et al. (2007) and Shen et al. (2011) used SMPS measure-

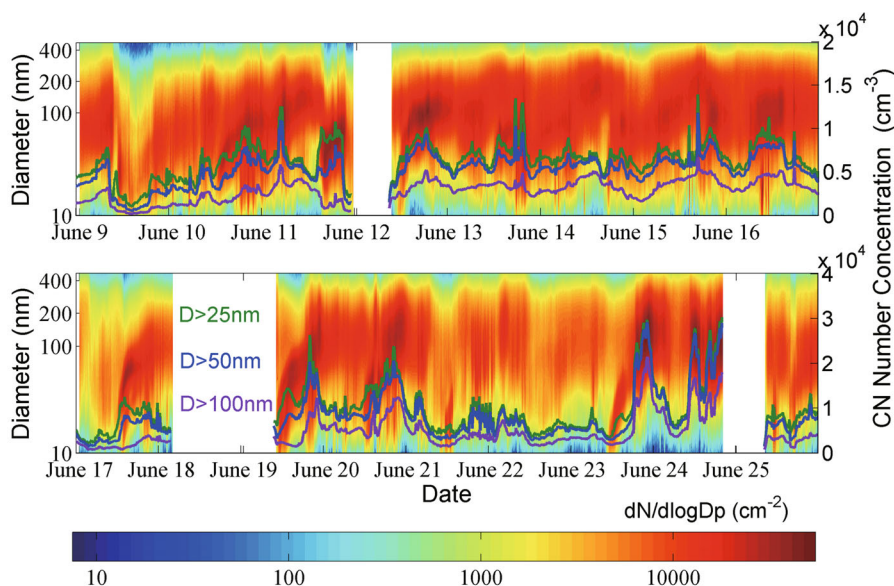


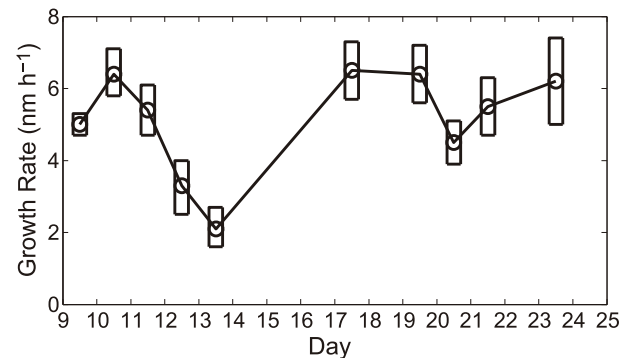
Fig. 4. Temporal variation of particle number size distribution between 10 nm and 500 nm and aerosol number concentration with sizes larger than 25 nm (green line), 50 nm (blue line), and 100 nm (purple line), for the AC3E campaign period of 9–25 June 2013.

ments with a lower size limit of 3 nm, and what they determined were frequencies of new particle formation events that occurred in years other than 2013. Also, Xianghe is a little farther away from central Beijing. Consistent with most studies (e.g., Kulmala et al., 2004a; Wu et al., 2007), fine aerosol particle growth events often occur on clean and sunny days, and the particles can grow large enough as accumulation mode aerosol in several hours or 1–2 days. Most of these fine aerosol particle growth events observed here typically begin around 0900–1200 LST, which is consistent with Zhang et al. (2011) and Wu et al. (2007).

Figure 4 shows the temporal variation of aerosol number concentration with sizes larger than 25 nm, 50 nm and 100 nm separately, which exactly illustrates this point. Aerosol with sizes larger than 100 nm (accumulation-mode aerosol) can be treated as pre-existing large sized background aerosol in the initial stage of a fine aerosol particle growth event, which is generally minimal in concentration during the day of the growth event. Thus, we can use the daily minimum aerosol concentration in the accumulation mode to estimate the relative impact caused by extramodal coagulation on the mean growth rate of fine aerosol particles in an event. Unfortunately, there is no clear relationship between the daily minimum accumulation-mode aerosol concentrations and the GRs of fine aerosol particles, as shown in Fig. 4. This may imply a deficiency of extramodal coagulation.

For all fine aerosol particle growth event days during the AC3E campaign, the GRs are calculated and shown in Fig. 5. The GR values range from 2.1 to 6.5  $\text{nm h}^{-1}$ , with an average value around 5.1  $\text{nm h}^{-1}$ . These values are roughly consistent with the findings from previous studies in the Beijing area (Wu et al., 2007; Yue et al., 2010; Zhang et al., 2011), which show averaged GRs of about 3–5  $\text{nm h}^{-1}$ . However, as indicated in Fig. 4, large uncertainties exist for determined fine aerosol particle GRs at each event, which is usually about 0.5–1  $\text{nm h}^{-1}$ .

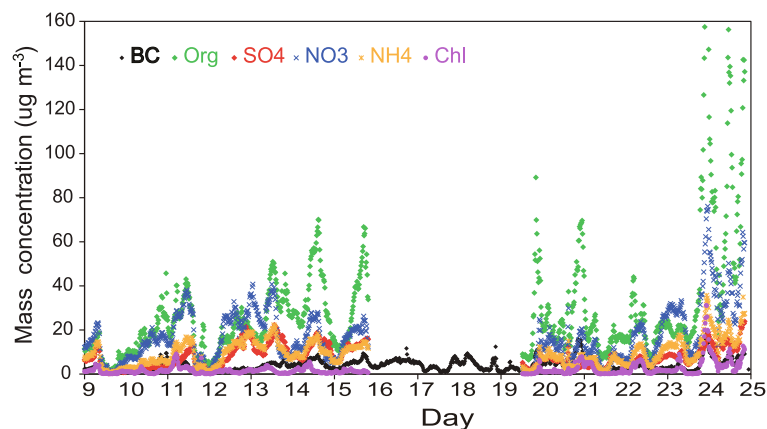
Figure 6 shows that the dominant aerosol chemical compositions are organics and nitrate, with relatively smaller amounts of ammonium and sulfate, which is slightly different



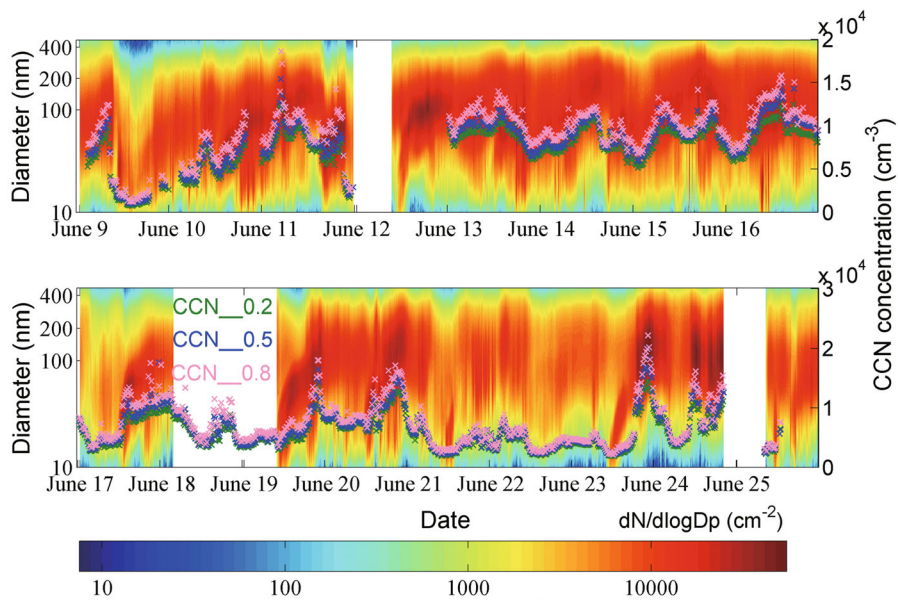
**Fig. 5.** Growth rates of fine aerosol particles for observed events during the AC3E campaign between 9 and 25 June 2013. The circles represent the mean values and the bars represent the ranges.

from the findings of Zhang et al. (2011) in which the amount of sulfate was more than that of nitrate. Note that the chemical composition from the Aerosol Chemical Speciation Monitor (ACSM) in Fig. 6 is for aerosol particles with sizes between 40 nm and 1  $\mu\text{m}$ . Here, we simply assume that the particles with sizes between 10 and 500 nm measured by SMPS have the same chemical composition as obtained by ACSM. The  $\text{NO}_x$  and  $\text{SO}_2$  gases are emitted mainly from strong traffic pollution and burning coal (Zhu et al., 2016), which serve as precursors of fine aerosol particles and provide an acidic environment that can cause fast growth of fine aerosol particles through vapor condensation. As shown in Zhu et al. (2016, Figs. 2 and 3), both observation and model simulation results for a short period during the observation window show high concentrations of  $\text{NO}_x$  and  $\text{SO}_2$ , at roughly 400 ppb and 25 ppb, respectively. These help make the growth of fine aerosol particles faster. Also, the acidic environment strengthens the multi-phase chemical reactions such that fine aerosol particles can grow faster.

One important point regarding the growth of fine aerosol particles is that large aerosols play important indirect radiative roles by serving as CCN. Figure 7 shows the temporal



**Fig. 6.** Temporal variation of aerosol chemical composition measured during the AC3E campaign between 9 and 25 in June 2013.



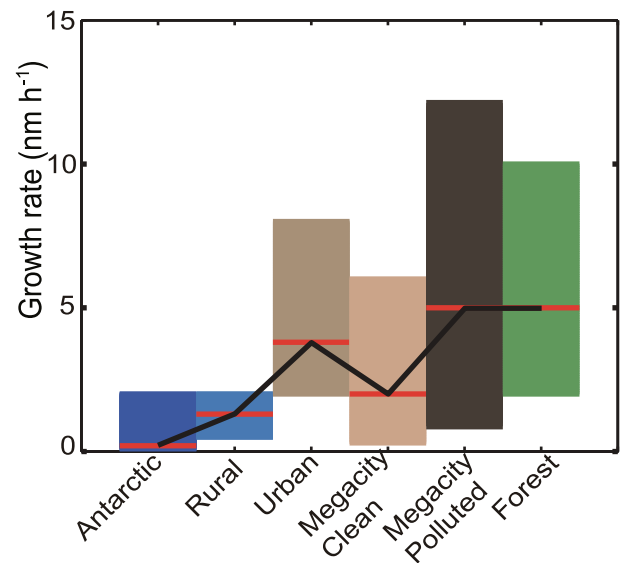
**Fig. 7.** Temporal variation of CCN concentration at supersaturations of 0.2%, 0.5% and 0.8% during the AC3E campaign between 9 and 25 June 2013.

variation of CCN during the AC3E campaign. For almost every fine aerosol growth event, the concentration of CCN is lowest in the initial stage, and quickly increases with the growth of the fine aerosol particles. When the aerosol particles grow large enough, the intramodal and extramodal coagulations make the CCN number concentration decrease. From Fig. 7, we can also identify the main growth trends as found in Fig. 4: a significant increase in CCN on the days when fine aerosol particles grow. When the environment is suitable for cloud formation, increased CCN will have strong impacts on both cloud microphysical properties and radiation budgets.

#### 4.2. Spatial variation of growth rates

By combining various findings on the GRs of fine aerosol particles at different locations, we can examine the spatial variation of GRs. Table 2 lists the fine aerosol particle GRs found by various studies over six different types of locations: clean Antarctic region, slightly polluted rural areas, polluted urban areas, relatively clean (or lightly polluted) megacities, polluted megacities, and boreal forest. The reference studies, locations, and growth rates obtained are also listed in the table. Note that there are strong seasonal variations for fine aerosol particle GRs found by many of these previous studies. Higher GRs of fine aerosol particles are found during summer than in winter, which is potentially associated with the higher precipitable water vapor concentration in summer. Based on the studies listed in Table 2, we provide a rough estimate of the mean GR of fine aerosol particles over each location. These are: 0.2, 1.3, 3.8, 5.0, 2.0 and 5.0  $\text{nm h}^{-1}$ , for the Antarctic, rural, urban, polluted megacity, relatively clean megacity, and boreal forest, respectively. Note that these estimates are very rough and large uncertainties could exist.

Figure 8 shows the variation in mean GRs over the six locations indicated in Table 2. The bars represent the potential



**Fig. 8.** Variation of fine aerosol particle growth rates with location [clean Antarctic, clean rural, urban, and megacities (divided into clean and polluted), and forest]. The results are from different studies shown in Table 1. The bars represent the most likely ranges and the red lines indicate the mean values of GRs for the corresponding location types.

ranges of fine aerosol particle GRs and the red lines indicate the estimated mean values of GRs for the corresponding location types. In general, the fine aerosol particle GRs have a large variability, even in the same location type dominated by similar aerosol types. This suggests significant influences from other environmental factors such as meteorological conditions and pre-existing background aerosol pollution. These results presented in section 3 imply that one dominant mechanism for the variation of GRs with location is vapor condensation. Both fine aerosol particle GRs and condensable

**Table 2.** Growth Rates (GRs) of fine aerosol particles reported by various studies at different locations. The GR estimates for different types of regions in summer are also shown in the table. Note SP\*, S\*, F\* and W\* indicate spring, summer, fall and winter, respectively. The results from references with an asterisk are from Table 2 in Kulmala et al. (2004a).

Location Type	Location	Reference	GR	Mean GR estimates in S*
Ocean	Antarctic	Park et al. (2004)* Gras (1993)*	0.13 0.1	0.2 (0.1–2)
Rural or remote continental locations	Idaho Hill, Colorado	Weber et al. (1997)	0.5–2	1.3 (0.5–2)
Forest or farmland	Idaho Hill, Colorado	Eisele and McMurry (1997)	1–2	5.0 (2–10)
	Industrialized agricultural area	Birmili et al. (2003)	0.5–8 (S*: 2–8; W*: 0.5–2.5)	
	Boreal forest in southern Finland	Makela et al. (1999)*	2–3	
	Boreal forest in southern Finland	Makela et al. (2000)*	1–17 (S*: 4–10; W*: 1–2.5)	
Urban areas	Boreal forest in southern Finland	Kulmala et al. (1998)*	3–20	3.8 (2–8)
	Central Europe	Neusüss et al. (2002)*	3.5 ( $\pm 1.0$ )	
	St. Louis	Shi and Qian (2003)*	0.5–9 (S*: 3.5–8; W*: 0.5–2.5)	
Megacities	North China Plain	Shen et al. (2011)	SP*: 3.0; S*: 4.2; F*: 3.1; W*: 3.7	Clean: 2.0 (0.3–6); Polluted: 5.0 (1.0–12)
	Western Yangtze River Delta	Herrmann et al. (2013)	6–7	
	Beijing (summer)	Zhang et al. (2011)	3.2	
	Beijing	Wu et al. (2007)	Clean: $\sim 1.8 (\pm 2.0)$ Polluted: $\sim 4.4 (\pm 3.2)$	
	Beijing	Yue et al. (2010)	2004S*: 0.3–12.3 2005S*: 5–11 2006S*: 1.2–5.6 2013S*: $\sim 4.3 (1.7–6.5)$	
	Xianghe (close to Beijing)	This study		

vapor concentration are larger in urban and polluted megacity regions compared with Antarctic and rural regions. For relatively clean megacities, the fine aerosol particle GRs lie between those of urban and rural regions. Due to the release of volatile organic compounds from boreal regions, the mean fine aerosol particle GR over boreal forest is also large—almost the same as that over polluted megacities. In addition, multi-phase chemical reactions are generally larger in the urban, megacity and boreal regions.

## 5. Summary and discussion

The growth of fine aerosol particles is a frequent phenomenon in Earth's atmosphere and plays an important role for local environments and global climate change. Based on short-term aerosol observations during the AC3E campaign, the present study shows a high frequency ( $\sim 50\%$ ) of fine aerosol particle growth events at Xianghe in summer. The GRs of fine aerosol particles during the AC3E campaign range from 2.1 to 6.5  $\text{nm h}^{-1}$ , with a mean value of  $\sim 5.1 \text{ nm h}^{-1}$ . The most likely contribution to the GRs of fine aerosol particles from four factors of influence are discussed in this study, including vapor condensation, intramodal coagulation, extramodal coagulation and multi-phase chemical reactions. For Xianghe, with its heavy releases of organic, nitrate and sulfate materials (Zhu et al., 2016), vapor condensation should play a major role for the growth of fine aerosol

in the initial stage, making GRs high. Considering the several mechanisms for fine aerosol particle growth proposed by Kulmala et al. (2004b) and Yue et al. (2010), in the following stages, the combined impacts of extramodal and intramodal coagulations should contribute to the growth of fine aerosol particles, along with condensation growth. Due to the existence of pre-existing large size aerosol particles and the fluctuations of aerosol size distributions (such as on 19, 20 and 21 June in Fig. 4), large uncertainties could exist for our calculated GRs of fine aerosol.

A review of previous findings about the GRs of fine aerosol particles shows higher GR values over megacity, urban and boreal forest regions compared with rural or oceanic regions, most likely caused by the more significant vapor condensation effects. The heavy releases of organic, nitrate and sulfate materials in urban and megacity regions, and the heavy releases of volatile organic compounds from boreal forest regions, make the condensational vapor in megacity, urban and boreal forest regions much greater than in rural or oceanic regions. In short, GRs of fine aerosol particles are influenced by multiple factors including condensational vapor, pre-existing large aerosol particles, and various other environmental factors, causing observed values to vary within a broad range and with large uncertainties, even over the same location.

This study uses theories and results from previous studies to explain our observational findings for the fast GRs (2.1–



6.5 nm h<sup>-1</sup>) of fine aerosol particles found at Xianghe near Beijing, which include both the condensational growth and coalescence growth of fine aerosol particles. Whilst carried out in some of the studies cited here, model simulations or lab investigations have not been carried out to further evaluate our explanation, which would be a valuable approach to take in the future. Moreover, this study calculates the GRs of fine aerosol particles without considering their dissipation through dry deposition, while precipitation scavenging or wet deposition has been excluded. The dry deposition may have slightly increased the calculated GRs since it is easier for smaller particles to be deposited at the growth size range between 10 and 500 nm (Zhang et al., 2001). In other words, the GRs of fine aerosol particles could be slightly smaller in the study area.

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## REFERENCES

- Albrecht, B., 1989: Aerosols, cloud microphysics and fractional cloudiness. *Science*, **245**, 1227–1230, <http://dx.doi.org/10.1126/science.245.4923.1227>.
- Birmili, W., H. Berresheim, C. Plass-Dülmer, T. Elste, S. Gilge, A. Wiedensohler, and U. Uhrner, 2003: The Hohenpeissenberg aerosol formation experiment (HAFEX): A long-term study including size-resolved aerosol, H<sub>2</sub>SO<sub>4</sub>, OH, and monoterpenes measurements. *Atmos. Chem. Phys.*, **3**, 361–376, <https://doi.org/10.5194/acp-3-361-2003>.
- Coffman, D. J., and D. A. Hegg, 1995: A preliminary study of the effect of ammonia on particle nucleation in the marine boundary layer. *J. Geophys. Res.*, **100**, 7147–7160, <https://doi.org/10.1029/94JD03253>.
- Doyle, G. J., 1961: Self-nucleation in the sulfuric acid-water system. *The Journal of Chemical Physics*, **35**, 795–799, <https://doi.org/10.1063/1.1701218>.
- Easter, R. C., and L. K. Peters, 1994: Binary homogeneous nucleation: Temperature and relative humidity fluctuations, non-linearity, and aspects of new particle production in the atmosphere. *J. Appl. Meteor.*, **33**, 775–784, [https://doi.org/10.1175/1520-0450\(1994\)033<0775:BHNTAR>2.0.CO;2](https://doi.org/10.1175/1520-0450(1994)033<0775:BHNTAR>2.0.CO;2).
- Eisele, F. L., and P. H. McMurry, 1997: Recent progress in understanding particle nucleation and growth. *Philos. Trans. Roy. Soc. B*, **352**, 191–201, <https://doi.org/10.1098/rstb.1997.0014>.
- Garrett, T. J., and C. F. Zhao, 2006: Increased Arctic cloud long-wave emissivity associated with pollution from mid-latitudes. *Nature*, **440**(7085), 787–789, <https://doi.org/10.1038/nature04636>.
- Ghan, S. J., and S. E. Schwartz, 2007: Aerosol properties and processes: A path from field and laboratory measurements to global climate models. *Bull. Amer. Meteor. Soc.*, **88**, 1059–1083, <https://doi.org/10.1175/BAMS-88-7-1059>.
- Gras, J. L., 1993: Condensation nucleus size distribution at mawson, Antarctica: seasonal cycle. *Atmos. Environ. A*, **27**(9), 1417–1425, [https://doi.org/10.1016/0960-1686\(93\)90127-K](https://doi.org/10.1016/0960-1686(93)90127-K).
- Harrison, R. M., and J. X. Yin, 2000: Particulate matter in the atmosphere: Which particle properties are important for its effects on health? *Science of the Total Environment*, **249**(1–3), 85–101, [https://doi.org/10.1016/S0048-9697\(99\)00513-6](https://doi.org/10.1016/S0048-9697(99)00513-6).
- Heintzenberg, J., 1994: Properties of the log-normal particle size distribution. *Aerosol Science and Technology*, **21**, 46–48, <https://doi.org/10.1080/02786829408959695>.
- Herrmann, E., and Coauthors, 2013: New particle formation in the western Yangtze River Delta: First data from SORPES-station. *Atmos. Chem. Phys. Discuss.*, **13**, 1455–1488, <https://doi.org/10.5194/acpd-13-1455-2013>.
- IPCC, 2007: *Climate Change 2007: The Physical Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Solomon et al., Eds., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 996 pp.
- Korhonen, P., M. Kulmala, A. Laaksonen, Y. Viisanen, R. McGraw, and J. H. Seinfeld, 1999: Ternary nucleation of H<sub>2</sub>SO<sub>4</sub>, NH<sub>3</sub>, and H<sub>2</sub>O in the atmosphere. *J. Geophys. Res.*, **104**, 26 349–26 353, <https://doi.org/10.1029/1999JD900784>.
- Kreyling, W. G., M. Semmler, and W. Möller, 2004: Dosimetry and toxicology of ultrafine particles. *Journal of Aerosol Medicine*, **17**(2), 140–152, <https://doi.org/10.1089/0894268041457147>.
- Kuang, C., M. Chen, J. Zhao, J. Smith, P. H. McMurry, and J. Wang, 2012: Size and time-resolved growth rate measurements of 1 to 5 nm freshly formed atmospheric nuclei. *Atmos. Chem. Phys.*, **12**, 3573–3589, <https://doi.org/10.5194/acp-12-3573-2012>.
- Kulmala, M., A. Laaksonen, and L. Pirjola, 1998: Parameterizations for sulfuric acid/water nucleation rates. *J. Geophys. Res.*, **103**, 8301–8307, <https://doi.org/10.1029/97JD03718>.
- Kulmala, M., L. Pirjola, and J. M. Mäkelä, 2000: Stable sulphate clusters as a source of new atmospheric particles. *Nature*, **404**, 66–69, <https://doi.org/10.1038/35003550>.
- Kulmala, M., H. Vehkamäki, T. Petäjä, M. Dal Maso, A. Lauri, V.-M. Kerminen, W. Birmili, and P. H. McMurry, 2004a: Formation and growth rates of ultrafine atmospheric particles: A review of observations. *Journal of Aerosol Science*, **35**, 143–176, <https://doi.org/10.1016/j.jaerosci.2003.10.003>.
- Kulmala, M., and Coauthors, 2004b: Initial steps of aerosol growth. *Atmos. Chem. Phys.*, **4**, 2553–2560, <https://doi.org/10.5194/acp-4-2553-2004>.
- Kulmala, M., T. Petäjä, P. Mönkkönen, I. K. Koponen, M. Dal Maso, P. P. Aalto, K. E. J. Lehtinen, and V.-M. Kerminen, 2005: On the growth of nucleation mode particles: Source rates of condensable vapor in polluted and clean environments. *Atmos. Chem. Phys.*, **5**, 409–416, <https://doi.org/10.5194/acp-5-409-2005>.
- Kulmala, M., and Coauthors, 2012: Measurement of the nucleation of atmospheric aerosol particles. *Nature Protocols*, **7**, 1651–1667, <https://doi.org/10.1038/nprot.2012.091>.
- Lance, S., A. Nenes, J. Medina, and J. N. Smith, 2006: Mapping the operation of the DMT continuous flow CCN counter. *Aerosol Science and Technology*, **40**, 242–254, <https://doi.org/10.1080/02786820500543290>.
- Li, Z. Q., F. Niu, J. W. Fan, Y. G. Liu, D. Rosenfeld, and Y. N.



- Ding, 2011: Long-term impacts of aerosols on the vertical development of clouds and precipitation. *Nature Geoscience*, **4**, 888–894, <https://doi.org/10.1038/NGEO1313>.
- Lubin, D., and A. M. Vogelmann, 2006: A climatologically significant aerosol longwave indirect effect in the Arctic. *Nature*, **439**, 453–456, <https://doi.org/10.1038/nature04449>.
- Makela, J. M., I. K. Koponen, P. Aalto, and M. Kulmala, 1999: One-year data of submicron size modes of tropospheric background aerosol in southern Finland. *J. Aero. Sci.*, **31**, 595–611, [https://doi.org/10.1016/S0021-8502\(99\)00545-5](https://doi.org/10.1016/S0021-8502(99)00545-5).
- Makela, J. M., M. Dal Maso, L. Pirjola, P. Keronen, L. Laakso, M. Kulmala, and A. Laaksonen, 2000: Characteristics of the atmospheric particle formation events observed at a boreal forest site in southern Finland. *Boreal Environ. Res.*, **5**, 299–313, ISSN 1239-6095.
- McMurry, P. H., 2000: A review of atmospheric aerosol measurements. *Atmos. Environ.*, **34**, 1959–1999, [https://doi.org/10.1016/S1352-2310\(99\)00455-0](https://doi.org/10.1016/S1352-2310(99)00455-0).
- Neustüss, C., and Coauthors, 2002: Characterization and parameterization of atmospheric particle number, mass, and chemical size distributions in central Europe during LACE-98 MINT. *J. Geophys. Res.*, **107**(D21), 8127, <https://doi.org/10.1029/2001JD000514>.
- Nilsson, E. D., and M. Kulmala, 1998: The potential for atmospheric mixing processes to enhance the binary nucleation rate. *J. Geophys. Res.*, **103**, 1381–1389, <https://doi.org/10.1029/97JD02629>.
- Park, J., H. Sakurai, K. Vollmers, and P. H. McMurry, 2004: Aerosol size distributions measured at the South Pole during ISCAT. *Atmos. Environ.*, **38** (32), 5493–5500, <https://doi.org/10.1016/j.atmosenv.2002.12.001>.
- Raes, F., A. Saltelli, and R. Van Dingenen, 1992: Modelling formation and growth of H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O aerosols: Uncertainty analysis and experimental evaluation. *Journal of Aerosol Science*, **23**, 759–771, [https://doi.org/10.1016/0021-8502\(92\)90042-T](https://doi.org/10.1016/0021-8502(92)90042-T).
- Rosenfeld, D., U. Lohmann, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M. O. Andreae, 2008: Flood or drought: How do aerosols affect precipitation? *Science*, **321**, 1309–1313, <https://doi.org/10.1126/science.1160606>.
- Shen, X. J., and Coauthors, 2011: First long-term study of particle number size distributions and new particle formation events of regional aerosol in the North China Plain. *Atmos. Chem. Phys.*, **11**, 1565–1580, <https://doi.org/10.5194/acp-11-1565-2011>.
- Shi, J. P., and Y. Qian, 2003: Continuous measurements of 3 nm to 10 μm aerosol size distributions in St. Louis, M.S. Thesis, Department of Mechanical Engineering, University of Minnesota, Minneapolis, MN 55455.
- Spracklen, D. V., K. S. Carslaw, M. Kulmala, V.-M. Kerminen, G. W. Mann, and S.-L. Sihto, 2006: The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales. *Atmos. Chem. Phys.*, **6**, 5631–5648, <https://doi.org/10.5194/acp-6-5631-2006>.
- Spracklen, D. V., and Coauthors, 2008: Contribution of particle formation to global cloud condensation nuclei concentrations. *Geophys. Res. Lett.*, **35**, L06808, <https://doi.org/10.1029/2007GL033038>.
- Spracklen, D. V., and Coauthors, 2010: Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation. *Atmos. Chem. Phys.*, **10**, 4775–4793, <https://doi.org/10.5194/acp-10-4775-2010>.
- Sun, Y., Z. F. Wang, H. B. Dong, T. Tang, J. Li, X. L. Pan, P. Chen, and J. T. Jayne, 2012: Characterization of summer organic and inorganic aerosols in Beijing, China with an Aerosol Chemical Speciation Monitor. *Atmos. Environ.*, **51**, 250–259, <https://doi.org/10.1016/j.atmosenv.2012.01.013>.
- Twomey, S., 1974: Pollution and the planetary albedo. *Atmos. Environ.*, **8**, 1251–1256, [https://doi.org/10.1016/0004-6981\(74\)90004-3](https://doi.org/10.1016/0004-6981(74)90004-3).
- Weber, R. J., J. J. Marti, P. H. McMurry, F. L. Eisele, D. J. Tanner, and A. Jefferson, 1997: Measurements of new particle formation and ultrafine particle growth rates at a clean continental site. *J. Geophys. Res.*, **102**, 4375–4385, <https://doi.org/10.1029/96JD03656>.
- Wu, Z. J., and Coauthors, 2007: New particle formation in Beijing, China: Statistical analysis of a 1-year data set. *J. Geophys. Res.*, **112**(D9), D09209, <https://doi.org/10.1029/2006JD007406>.
- Yu, F. Q., and Coauthors, 2010: Spatial distributions of particle number concentrations in the global troposphere: Simulations, observations, and implications for nucleation mechanism. *J. Geophys. Res.*, **115**, D17205, <https://doi.org/10.1029/2009JD013473>.
- Yue, D. L., and Coauthors, 2010: The roles of sulfuric acid in new particle formation and growth in the mega-city of Beijing. *Atmos. Chem. Phys.*, **10**, 4953–4960, <https://doi.org/10.5194/acp-10-4953-2010>.
- Zhang, L. M., S. L. Gong, J. Padro, and L. Barrie, 2001: A size-segregated particle dry deposition scheme for an atmospheric aerosol module. *Atmos. Environ.*, **35**(3), 549–560, [https://doi.org/10.1016/S1352-2310\(00\)00326-5](https://doi.org/10.1016/S1352-2310(00)00326-5).
- Zhang, Y. M., X. Y. Zhang, J. Y. Sun, W. L. Lin, S. L. Gong, X. J. Shen, and S. Yang, 2011: Characterization of new particle and secondary aerosol formation during summertime in Beijing, China. *Tellus B*, **63**, 382–394, <https://doi.org/10.1111/j.1600-0889.2011.00533.x>.
- Zhao, C. F., S. A. Klein, S. C. Xie, X. H. Liu, J. S. Boyle, and Y. Y. Zhang, 2012: Aerosol first indirect effects on non-precipitating low-level liquid cloud properties as simulated by CAM5 at ARM sites. *Geophys. Res. Lett.*, **39**, L08806, <https://doi.org/10.1029/2012GL051213>.
- Zhu, Y., and Coauthors, 2016: Distribution and sources of air pollutants in the North China plain based on on-road mobile measurements. *Atmos. Chem. Phys.*, **16**, 12 551–12 565, <https://doi.org/10.5194/acp-16-12551-2016>.