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Variations of Cloud Condensation Nuclei (CCN) and aerosol activity during fog-haze episode: a case study from Shanghai

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Abstract

Measurements of Cloud condensation nuclei (CCN), condensation nuclei (CN) and aerosol chemical composition were performed simultaneously at an urban site of Shanghai from 6 to 9 November 2010. The variations of CCN number concentration (N_{CCN}) and aerosol activity (activated aerosol fraction, N_{CCN}/N_{CN}) were examined during a fog-haze co-occurring event. Anthropogenic pollutants emitted from vehicles and unfavorable meteorological conditions such as low planetary boundary layer (PBL) height exerted a great influence on CN and black carbon (BC) loadings. N_{CCN} at 0.2 % supersaturation (SS) mostly fell in the range of 994 to 6268 cm^{-3}, and the corresponding N_{CCN}/N_{CN} varied between 0.09 and 0.57. N_{CCN} and N_{CCN}/N_{CN} usually were higher in hazy days due to increased aerosol concentration in the accumulation mode (100–500 nm), and lower in foggy-hazy and clear days. BC mass concentration posed a strong positive effect on N_{CCN} in foggy-hazy and hazy days, whereas it poorly correlated with N_{CCN} in clear days. N_{CCN}/N_{CN} was weakly related with BC both in foggy-hazy/hazy and clear days. By using a simplified particle hygroscopicity (κ), the calculated critical dry size (CDS) of activated aerosol did not exceed 130 nm at 0.2 % SS in spite of diverse aerosol chemical compositions. The predicted N_{CCN} at 0.2 % SS was very successful compared with the observed N_{CCN} in clear days ($R^2 = 0.96$) and foggy-hazy/hazy days ($R^2 = 0.91$). In addition, their corresponding ratios of predicted to observed N_{CCN} were on average 0.95 and 0.92, respectively. More organic matter is possibly responsible for this closure difference between foggy-hazy/hazy and clear days. These results reveal that the particulate pollutant burden exerts a significant impact on N_{CCN}, especially N_{CCN}/N_{CN} promotes effectively during the polluted periods.

1 Introduction

Cloud Condensation Nuclei (CCN), which constitutes an important fraction of atmospheric aerosol, can influence the microphysical and radiative properties and lifetime

of cloud indirectly and consequently impact the hydrological cycle (IPCC, 2007). The elevated CCN loadings (N_{CCN}) tend to reduce cloud droplet size and then suppress precipitation in shallow and short-lived clouds (Lohmann and Feichter, 2005). But it can promote great convective overturning and enhance precipitation in deep convec-
5 tive clouds (Rosenfeld et al., 2008). Numerous aerosol properties, including particle size distribution, chemical composition and mixing state, are closely linked with the ability of particles to take up water vapor, i.e. the ability to act as CCN (Baumgard-ner et al., 2003; Kuwata and Kondo, 2008; Cubison et al., 2008). To date, the current assessment of aerosol indirect effects induced by increasing anthropogenic aerosols
10 remains poorly understood, and this brings a big uncertainty in fully picturing climate change (Andreae et al., 2005; IPCC, 2007).

Owing to advanced instrument development, the aerosol-cloud interaction and its im-pact on climate have attracted increasing attention in the last decades. Many ground-based measurements on CCN have been performed in diverse environments, describ-
15 ing a global map of CCN distribution in the surface atmosphere (Baumgardner et al., 2003; Yum et al., 2004, 2005; Reade et al., 2006; Juranyi et al., 2010; Leng et al., 2013). In urban environments, the new particle formation and growth, and haze pol-lution were observed recently as having a significant impact on N_{CCN} (Ritesh et al., 2007; Kuang et al., 2009). In recent years, CCN studies have raised the relative im-
20 portance of several influence factors controlling aerosol CCN activity, of which size has been announced as the major factor in determining the CCN activation of aerosol par-ticles (Dusek et al., 2006; Anttila and Kerminen, 2007; Hudson, 2007; Quinn et al., 2008; Jimenez et al., 2009; Leng et al., 2013). However, how chemical composition especially organic compounds to link with aerosol activity and then CCN has not been
25 fully understood. In fact, up to 90 % of the aerosol population has been formed by car-bonaceous substances, and among them 10–70 % is water-soluble (Moffet et al., 2008; Stone et al., 2008). Particularly, various externally or internally-mixed particulate com-ponents comprised in urban air mass can significantly affect the CCN-sized spectra of atmospheric particles (Svenningsson et al., 2006; Reade et al., 2006; Kuwata et al.,

2007). This has posed a major challenge to study aerosol composition and predict CCN activity (Hagler et al., 2007; Hings et al., 2008; Henning et al., 2010).

Due to rapid industrialization in Asia for decades, anthropogenic particles and relevant precursor emissions have increased significantly, and numerous studies have indicated that the increasing anthropogenic aerosol loading has significantly changed cloud microphysical and radiative properties (Streets et al., 2000, 2008; Shao et al., 2006; Wang et al., 2006; Qian et al., 2006; Rosenfeld et al., 2007; Matsui et al., 2010; Zhang et al., 2013). In China, studies on CCN have been done widely such as at polluted sites located in Yufa (Wiedensohler et al., 2009), Beijing (Yue et al., 2011), Shouxian (Liu et al., 2011) and Shanghai (Leng et al., 2013), and suburban sites in Guangzhou (Rose et al., 2010, 2011) and Wuqing (Deng et al., 2011). To our knowledge, little attention has been paid on the impacts of fog or haze on CCN and activated aerosol particles. The increases of haze occurrences are evident in the eastern and southwestern cities in China (Che et al., 2009). Shanghai is a huge metropolis in China, and the occurrence intensity of foggy and hazy days on annual time scale has been increasing gradually especially in winter (Tie and Cao, 2009), which is deeply affected by fine particle pollution enhancement and possibly linked with particle hygroscopicity (Ye et al., 2011).

This study presents continuous measurements of CCN and aerosol during a fog-haze episode from 6 to 9 November 2010 in Shanghai. The aim is to provide insights on CCN and aerosol activity variations under fog-haze co-occurring conditions. The instrumentation and data used in the study are described in Sect. 2. The aerosol physical and chemical properties are introduced in Sect. 3. Section 4 presents the evolution of CCN and aerosol activity. The relationship between aerosol and CCN is discussed in Sect. 5. Conclusions from the study are given in Sect. 6.

2 Methods

2.1 Observational site

The instruments for CCN and aerosol measurements were mounted roughly 20 m above ground on the roof of a building in the campus of Fudan University in Shanghai (31°18′ N, 121°29′ E). The site is surrounded by populated residential and commercial areas, as well as urban streets. The East China Sea is roughly 40 km east of the site, and the prevailing wind directions are southeasterly in summer and northeasterly in winter. Local time (LT) hereafter employed in this study is 8 h ahead of UTC.

2.2 Measurement and instrumentation

N_{CCN} was measured using a continuous flow and single column CCN counter (model CCN-100, Droplet Measurement Technologies, USA), in which an optical particle counter (OPC, 0.75–10 μm) is employed to detect activated cloud droplets (Roberts and Nenes, 2005; Lance et al., 2006). The instrument was housed in an air-conditioned weather-proof container with temperature maintaining at 20 °C. The ambient aerosol airflow passed through a dryer (active carbon) to lower relative humidity below 30 % before entering the instrument (Leng et al., 2013). The CCN counter was calibrated using ammonium sulfate before the study, as did calibrations for temperature gradient, flow, pressure and OPC to maintain stable SS according to the DMT operation manual. In order to ensure accurately counting, zero checks were performed before and after the campaign and regularly every two months. The effective water vapor supersaturation (SS) changed alternately at 0.2 % interval within 0.2–1.0 %. Although the CCN counter can operate well under conditions of particles only in a few thousand number per cubic centimeter and corrections must need for larger concentrations ($> 5000\,\mathrm{cm}^{-3}$) (Lathem and Nenes, 2011), we still used the measured N_{CCN} directly at 0.2 % SS in this study since it seldom reached the upper limit.

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A high-resolution wide-range particle spectrometer (WPS-1000 XP, MSP) was employed to observe particle size distributions in the size range of 10 nm–10 µm. The calibration and operating methodology of WPS has been described elsewhere (Zhang et al., 2010). In addition, we have compared the aerosol size spectra measured by WPS with those measured in parallel by a calibrated scanning mobility particle sizer (SMPS, TSI 3080) with higher accuracy in the size range of 20–800 nm, including size-resolved particle concentrations and peak sizes, and a strong correlation between them was derived with correlation coefficient $R^2 > 0.95$ (Leng et al., 2013). The result confirms the reliability of WPS measurements for successfully characterizing the number concentration and size distribution of condensation nuclei (CN).

Planetary boundary layer (PBL) height and aerosol vertical extinction profile were measured using a set of micro pulse lidar (MPL) system (MPL-4B-532) with pulse energy 6–10 µJ and pulse repetition frequency 2500 Hz. The MPL is an eye safe, compact and autonomous instrument, and an effective tool used widely in the world to provide available high spatial (30 m) and temporal resolution information of aerosol vertical distributions (Menut et al., 1999; Cohn and Angevine, 2000; Brooks, 2003).

An online Aethalometer (AE-31, Magee Scientific Co., Berkeley, California, USA) was employed to measure black carbon (BC) at a 5 min time resolution. Based on the strong absorbtivity of BC to light at near infrared wavelengths (Hansen et al., 1984; Weingartner et al., 2003), BC concentration is determined using the measured light attenuation at 880 nm and the appropriate value of specific attenuation cross section proportional to BC mass (Petzold et al., 1997). The attenuation can be obtained by calculating the difference between light transmission through the particle-laden sample spot and the particle-free reference spot in the filter (Cheng et al., 2006; Dumka et al., 2010). The operation, calibration and maintenance of AE-31 have been described in detail by Cheng et al. (2010).

An online analyzer for Monitoring Aerosols and Gases (MARGA, ADI 2080, the Netherlands) was employed to measure the concentration of major inorganic water-soluble ions (e.g. Na+, K^+, Mg^+, Ca^+, SO_4^{2-}, Cl^-, NO_3^- and NH_4^+) in ambient aerosol

particles. Instructions for the methods of sampling, operation and internal calibration have been described in detail elsewhere (Du et al., 2011). Moreover, the mass concentrations of particulate matter (PM) with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$), meteorological factors and atmospheric visibility were measured by a con-

5 tinuous PM ambient monitor (FH62C14, Thermo), an automatic weather monitoring system (HydroMet^{TM}, Vaisala) and a automatic visibility monitor, respectively.

2.3 Air mass backward trajectory

The HYSPLIT-4 model developed by the Air Resources Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA), USA (Draxler et al., 2003),

10 was employed to compute 24 h air mass backward trajectories ending at 500 m height and starting at 0:00 LT and 12:00 LT for each day. By doing so, we can identify aerosols from different source regions and analyze their effects on aerosol activity to compile a full view of the relation between fog-haze event and N_{CCN}. According to these calculated trajectories plotted in Fig. 1, aerosol was classified into two categories: (1)

15 maritime aerosol transported by air masses from marine areas on 6 November 2010 carrying dominant oceanic particles, (2) continental aerosol in air mass traveling a long distance over inland areas on 7, 8 and 9 November 2010 and carrying more anthropogenic particles (e.g. BC). Exactly, the maritime air mass originated from the China Eastern Sea, traveled northwesterly slow-moving across the Hangzhou Bay and finally

20 arrived in Shanghai on 6 November. Then the air mass changed its direction to southeasterly at around 8.00 a.m. on 7 November, and originated from northern inland areas and traveled across the North China Plain (NCP) and the eastern region of China. The continental sources contained increasing industrial and agricultural emissions (e.g. biomass burning) due to long-term rapid economy growth and large population in the

25 last few decades. We hope to better understand the impact of aerosols with or without anthropogenic particulate pollutants on CCN in this study by comparing these two categories.

3 Results

3.1 Overview of the fog-haze event

Haze is traditionally defined as an atmospheric phenomenon that the sky clarity is obscured by dust, smoke and other dry particles, and atmospheric visibility and relative humidity (RH) are usually less than 10 km and 80 % over one haze episode (Fu et al., 2008). The high frequency of haze or hazy days is observed in winter, especially in the urban environments of northern China (Sun et al., 2006). During the haze event, the enhancement of particulate pollutants may greatly affect aerosol activity and N_{CCN}. The study performed in the Indo-Gangetic plain shows that winter haze exerts a significant impact on the fog and low-cloud formation (Gautam et al., 2007). Fog can be viewed as a lower-atmospheric near-surface cloud, and plays an important role in processing aerosol particles and trace gases (Gultepe et al., 2007; Biswas et al., 2008). On one hand, physically similar to cloud droplet, fog droplet also forms by water vapor condensing on dry aerosol particle under supersaturated conditions. On the other hand, generally formed in the shallow boundary layer containing local emissions, urban fog traps more pollutants than cloud at high altitudes (Fisak et al., 2002; Herckes et al., 2007). The fog or foggy day is defined as a weather with patterns of low visibility (< 10 km) and higher RH (> 90 %). When 80 % $<$ RH $<$ 90 %, the weather was referred as a complex of haze and fog co-occurring (e.g. foggy-hazy) in the present study. Figures 2 and 3 show a 4 day time series of pressure, atmospheric visibility, RH, temperature, wind speed and direction, and PBL height from 6 to 9 November 2010. In fact, since RH seldom reached up to 90 %, thus the period focused in the present study were characterized as hazy and foggy-hazy days. The haze pollution lasting at least 4 h has been identified as one haze event by an earlier study in Shanghai, where authors paid attention to the formation of haze pollution (Du et al., 2011).

As shown in Figs. 2–7, the 4 day period was classified into three parts: a hazy episode (marked in black open boxes) from 22:00 to 23:00 LT on 6 November and 10:00 LT on 7 November to 13:00 LT on 8 November, a foggy-hazy episode (marked

in red open boxes) from 23:00 LT on 6 November to 10:00 LT on 7 November, and the rest for clear days. Statistics for meteorological conditions is listed in Table 1. During the hazy and foggy-hazy days, the average atmospheric visibility was about 4.44 km and 2.33 km, respectively, much lower than 15.4 km in the clear days. The winds from the east and the south brought clean maritime aerosol during the clear days, however, the winds from the north and the west brought polluted anthropogenic aerosol during the hazy and foggy-hazy episodes. The particulate and gaseous matters, including pollutants (e.g. BC) emitted from agricultural biomass burning were transported along the air mass pathways (Fig. 1), led to a significant reduction of aerosol extinction coefficient (Fig. 3). In addition, the PBL height downed to below 500 m and further suppressed the dilution of pollutants.

3.2 Physical and chemical properties of aerosol

In order to visually identify aerosol evolution, particles in the size range of 10 nm to 10 µm were categorized into 7 sub-size bins: 10–20 nm (nucleation mode), 20–50 nm and 50–100 nm (Aitken mode), 100–200 nm, 200–500 nm and 0.5–1 µm (accumulation mode), and 1–10 µm (coarse mode) (Fig. 4). The similar classification has been done in the measurements at the same site by Zhang et al. (2010). In this study, the integrated particle size-resolved number concentrations (N_{CN}) exhibited a regular diurnal cycle, with two peaks (9000–16 000 cm^{-3}) almost within the traffic rush hours. The mean N_{CN} exhibited no obvious difference between the foggy-hazy (8367 cm^{-3}) and clear (8956 cm^{-3}) days, but it showed a higher value (10 500 cm^{-3}) in the hazy days, revealing a larger loading of particulate pollutants.

In general, the 20–100 nm (Aitken mode) particles are mostly dominant in all size particles probably due to local traffic emissions and meteorological conditions (Ferin e al., 1990). The temporal variation trend of Aitken mode was similar to N_{CN}. It was interesting that the particles of 100–500 nm (accumulation mode) dominated in N_{CN} in the hazy days with peak concentrations higher than 7500 cm^{-3}, almost twice as

much as the clear days ($4000\,\mathrm{cm}^{-3}$). However, the foggy-hazy days are comparable to the clear days, showing a mostly unchanged evolution of the fractions of individual size bin to total particles and N_{CN}. Figure 5 shows the size distributions of aerosol surface and length describing particle morphology, which were calculated from the number size distribution by assuming a spherical particle (Gao et al., 2009). It was clear that the particles in accumulation mode dominated aerosol length and surface area during the hazy days, providing a larger space for heterogeneous chemical reactions of atmospheric precursors possibly to enhance particle aging.

Figure 6 shows the temporal variations of eight major inorganic water soluble ions in aerosol particles and four gaseous pollutants sampled during this study period. Measurements for SO_4^{2-}, Cl^- and NO_3^- were unavailable from 10:00 LT on 7 November to 08:00 LT on 8 November. Substantially, the average concentration of aerosol total water soluble ions (TWSI) in the hazy days ($54.52\,\mu\mathrm{g\,m}^{-3}$) was comparable to the foggy-hazy days ($50.37\,\mu\mathrm{g\,m}^{-3}$), and roughly 2 times that of the clear days ($26.22\,\mu\mathrm{g\,m}^{-3}$). For the percentage of individual ions in TWSI, NH_4^+ and K^+ were relatively higher by a factor of 1.8 in the hazy and foggy-hazy days than in the clear days. Despite the lack of SO_4^{2-} and NO_3^- partly during the hazy days, we can still conjecture their promotion on the basis of their gaseous precursor evolution of SO_2 and NO_2.

Gaseous pollutants are released into the atmosphere from natural and anthropogenic emissions. Among them, SO_2 is known as one of the most important gaseous pollutants and a precursor responsible for acid rain. Also, it can participate in the formation of new particles through converting into gaseous H_2SO_4, which is the most common nucleation species due to its low vapor pressure at typical atmospheric temperature (Zhang et al., 2006b; Urone et al., 1968). Secondary aerosols produced from the formation of new particles contribute more to the global aerosol burden than primary aerosols and are important sources of CCN (Merikanto et al., 2009; Yu et al., 2008). Recent studies have shown the enhanced solubility of SO_2 due to its reaction in fog droplets during a severe fog measured in the North China Plain, and this finding has provided important support for better understanding of the acidity in clouds (Zhang

et al., 2013). NO_2 mainly comes from vehicle traffic emissions in urban areas (Wang et al., 2006). Nitrogen oxides (NO, NO_2, N_2O_5) undergo heterogeneous reactions with aerosol particles (e.g. sea salt or dust) during they are transported in the atmosphere (Elizabeth et al., 2006). Thus, high gaseous pollutant content can result in larger CN loadings and subsequently more CCN particles in the atmosphere. On the whole, the loading of these precursor gases in the foggy-hazy and hazy days exceeded that in the clear days, specifically NO_2 by a factor of 2 and SO_2 by a factor of 1.5. Moreover, SO_2 and NO_2 concentrations reached their peaks around 0:00 LT on 8 November corresponding to the highest levels of CCN and aerosol activity, implying their potential effects on CCN production, which will be discussed in the next section.

3.3   ccn concentration and aerosol activity

3.3.1   CCN and aerosol activity

Figure 7 presents the temporal variations of N_{CCN} and activated aerosol fraction (N_{CCN}/N_{CN}) at SS 0.2 %, N_{CN}, and BC during the campaign. Totally, N_{CN} fell in the range of 4270–15 771 cm^{-3} and averaged at 9344 cm^{-3}, and N_{CCN} varied between 994 cm^{-3} and 6268 cm^{-3} and averaged at 2929 cm^{-3}. High N_{CCN}/N_{CN} (0.41) and N_{CCN} (4362 cm^{-3}) were observed during the hazy days, followed by the foggy-hazy (0.29, 2377 cm^{-3}) and clear (0.28, 2432 cm^{-3}) days (Table 2). The temporal variation of N_{CCN}/N_{CN} and N_{CCN} was closely related with aerosol particle size spectra and chemical composition such as accumulation mode (100–500 nm) and water soluble ion content (Figs. 4 and 6). Figure 8 gives the temporal variations of number concentrations of larger aerosol particles (e.g. particles larger than 80 nm and 100 nm) and their corresponding ratios with N_{CCN} at SS 0.2 %. The larger aerosol particles showed significant increase during the hazy days and varied strongly correlated with N_{CCN}. More fractions of particles larger than 80 nm were activated into CCN during the hazy days (86 %) and foggy-hazy days (84 %) than that during the clear days (76 %).

Although in different SS conditions, N_{CCN} was measured at other urban or urban-like environments such as the west coast of Tasmania ($32\,cm^{-3}$) and the west coast of Korea ($5292\,cm^{-3}$) at SS 1.0 % (Yum et al., 2004, 2005), and Mexico city ($3000\,cm^{-3}$), Ireland ($208–346\,cm^{-3}$) and Vienna ($820\,cm^{-3}$) at SS 0.5 % (Baumgardner et al., 2003; Reade et al., 2006; Burkart et al., 2011). An even larger N_{CCN} ($6000\,cm^{-3}$) was measured at SS 0.17 % in Beijing (Deng et al., 2011). The average N_{CCN}/N_{CN} of this study (0.32) was higher than that measured in Vienna (0.13 at SS 0.5 %, CN 13–929 nm) and Finland (0.1–0.3 at SS 0.2 %, CN 3–1000 nm). The increased N_{CCN}/N_{CN} was derived at larger SS in urban environments such as Shanghai (0.47 at SS 0.8 %, CN 10–10 000 nm) and Korea (0.64 at SS 1.0 %, CN 10–500 nm) (Yum et al., 2005; Burkart et al., 2011; Sihto et al., 2011; Leng et al., 2013).

As expected, N_{CN} behaved in diurnal cycle with an apparent pattern of bi-modal distribution, and N_{CCN} showed a similar temporal variation (Fig. 7). N_{CN} and BC usually peaked, and reached their highest values of $15\,000\,cm^{-3}$ and $35\,\mu g\,m^{-3}$ during the rush hours (i.e. 07:00–09:00 and 16:00–19:00 LT), indicating that the anthropogenic pollutants emitted from vehicles contributes to a large part of CN and BC loadings. In addition, the favorable meteorological conditions such as low wind speed and planetary boundary layer (PBL) height (500 m) also posed a great influence on CN and BC loadings (Fig. 3).

In a broad view, N_{CCN} showed a sharp increase starting at 00:00 LT on 8 November, and rose from $994\,cm^{-3}$ to $6268\,cm^{-3}$ within less than 10 h. Similar to N_{CCN}, BC also rose from $10\,\mu g\,m^{-3}$ to $35\,\mu g\,m^{-3}$ during the same period. N_{CN} was consistent with N_{CCN}, and they varied almost synchronously. However, N_{CCN}/N_{CN} changed in one step mostly opposite to N_{CCN} and N_{CN} (Fig. 7). The possible reason for this contradictory tendency of N_{CN} enhancement vs. N_{CCN}/N_{CN} reduction is that the unactivated nanoparticles, which burst partly from primary emissions of vehicles and/or partly from secondary particles due to the chemical reactions of atmospheric gaseous precursors (Fig. 5) (Du et al., 2011), contributes relatively larger to N_{CN} other than N_{CCN}.

3.3.2 Black carbon and CCN

As a part of hydrophobic aerosols, pure BC particles acquire hydrophilic coatings as they age in the atmosphere, and then the aged BC becomes sufficiently hydrophilic and serves as CCN for cloud condensation formation (Ritesh et al., 2007). On the other hand, BC particles can release sensible heat by effectively absorbing solar radiation, thereby increasing the critical supersaturation of CCN and preventing aerosol to act as CCN (Conant et al., 2002). Biomass burning emits a large amount of trace gases and carboneous particles into the atmosphere, and leads to changes in climate and precipitation, as well as aquatic and terrestrial ecosystem (Andreae et al., 2004). The wild fires contribute a significant fraction of global CCN burden (Pierce et al., 2007; Andreae et al., 2009). Large quantities of active agricultural fire sites were detected from satellites over China on 7 November 2010 (Fig. 1), whereas no obvious wild biomass burning activities were observed during the rest days. Based on the calculated 24 h air mass backward trajectories, the air mass that passed right through the agricultural fire regions in the Jiangsu and Anhui provinces on 7 November reached the sampling site in the next day, bringing large quantities of aged BC particles after a long range transport. This resulted in a severe increase of particle mass concentration and a significant reduction of aerosol extinction coefficient on 7 and 8 November (Fig. 3). As discussed in Sect. 3.2, NO_2 and SO_2 concentrations increased synchronously during the whole period (Fig. 6), and they would undergo heterogeneous reactions on the surface of BC particles to change particle microphysical and chemical properties, making BC particles sufficiently hydrophilic to act as CCN (Ritesh et al., 2007).

Relationship analyses between N_{CCN}, N_{CCN}/N_{CN} and BC were calculated using hourly-averaged data, and the correlation coefficients (R^2) are listed in Fig. 9. Surprisingly, BC strongly correlated with N_{CCN} ($R^2 = 0.85$) in the foggy-hazy and hazy days, whereas they showed a poor linear relationship ($R^2 = 0.25$) in the clear days. The possible reason is BC particle aging by heterogeneous reactions with gaseous pollutants (e.g. NO_2 and SO_2) to be activated CCN during pollutant atmospheric trans-

port (Ritesh et al., 2007). In addition, so many studies have proposed that the aged BC is efficient CCN (Dusek et al., 2006; Anttila and Kerminen, 2007; Hudson, 2007). However, N_{CCN}/N_{CN} was poorly related with BC for both foggy-hazy/hazy and clear days (R^2 = 0.43 and 0.07, respectively), indicating that BC maybe a relatively more important contributor to unactivated particles especially in nanoscale sizes (e.g. traffic emission) than activated CCN.

3.4 Relationship of aerosol and CCN

Although aerosol size distributions were measured only in the size range of 10–10 000 nm, they were still used to predict N_{CCN} according to Köhler theory (Köhler et al., 1936). Toward this end, the particle hygroscopicity "kappa" (κ) was used in the closure calculation. According to Petters and Kreidenweis (2007), the κ parameter for one multicomponent particle can be obtained through weighting each component κ_i by their volume fractions in the mixture,

$$\kappa = \sum_i \varepsilon_i \kappa_i \tag{1}$$

where ε_i is the volume fraction of chemical compounds in particles, and κ_i is the effective κ of individual chemical composition.

Assuming aerosol particles are completely internal-mixed, a simplified κ was calculated using water soluble inorganic ions (organic matter data is unavailable). Aerosol particle compositions were classified into three categories (Petters and Kreidenweis., 2007; Wiedensohler et al., 2009), and κ_i and densities for each component are shown in Table 3, in which "others" is defined as "$PM_{2.5}$-BC-inorganic ions". The critical dry size (CDS) of particle to be activated as CCN at one SS can hence be determined by

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the following equation:

$$S(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)} \exp\left(\frac{4\sigma_{s/a}M_\omega}{RT\rho_\omega D}\right) \qquad (2)$$

where ρ_ω is the density of water, M_ω is the molecular weight of water, $\sigma_{s/a}$ is the surface
tension of the solution/air interface, R is the universal gas constant, κ is the hygroscop-
icity parameter, T is temperature, D is the diameter of the droplet and $S(D)$ is the
critical dry size under a given SS. Detailed information for the derivation of Eq. (2) can
be found in Petters and Kreidenweis (2007). Equation (2) applies over the entire range
of humidity and solution hygroscopicity and can be utilized to predict the conditions
of cloud droplet activation. The critical SS for a selected dry size of particle is deter-
mined from the maximum of the curve for Eq. (2). Computed for $\sigma_{s/a} = 0.072\,\mathrm{J\,m}^{-2}$ and
$T = 298.15\,\mathrm{K}$, the calculated CDS varied between 60 nm and 130 nm and averaged at
102 nm. Particularly, the hourly-averaged CDS during the foggy-hazy/hazy days was
slightly lower (96 nm) than during the clear days (105 nm). So far, the comparable or
relatively higher CDS has also been found in diverse regions and for various aerosol
types, despite of different calculation models and SS. For example, the fresh aerosol
particles emitted by an aircraft internal combustion engine have a CDS range of 146–
301 nm at SS 0.7 %, depending on varying operating conditions (Hitzenberger et al.,
2003). Furutani et al. (2008) investigated three types of aerosol masses along the
southern coast of California, and the CDS was estimated at 110 nm at SS 0.6 % for
fresh ship exhaust, 70–110 nm for fresh anthropogenic aerosols and roughly 50 nm
for aged anthropogenic and clean maritime aerosols. In Vienna, the CDS has a wide
gap between 69 nm and 368 nm, and averaged at 169 nm (Burkart et al., 2011). Quinn
et al. (2008) observed the CDS in a narrow range of 70–90 nm for maritime aerosols in
the Gulf of Mexico, and a moderate range of 90–170 nm in the ship channels of Hous-
ton with high marine traffic densities close to industrial and anthropogenic sources.

The CCN population can be effectively viewed as a subset of measured aerosol size
distributions since the operating range includes the majority of atmospheric particles

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(10–10 000 nm). Therefore, the predicted N_{CCN} can be calculated through integrating particles upward in size from the bottom CDS to the upper boundary.

The prediction for CCN is generally success (Fig. 10). It is worth note that the predicted N_{CCN} at SS 0.2 % was more correlated with the observed N_{CCN} in the clear days ($R^2 = 0.96$) than the foggy-hazy/hazy days ($R^2 = 0.91$), and the corresponding ratios of predicted to observed N_{CCN} were 0.95 and 0.92, respectively. The achieved closure calculation suggested that water soluble inorganic ions played a major role in contributing the κ value. In fact, 83.8 % of the κ was expressed by $SO_4^{2-} + NO_3^- + NH_4^+$ in total (in another study by our group, not published yet), with their individual contribution to be 39.8 %, 31.7 % and 12.3 %, respectively. On the other hand, the mean ratio of predicted to observed N_{CCN} never reached up to 1, suggesting that organic matter would play a second role and make up the rest of κ.

4   Conclusions and discussion

A continuous 4 day data obtained at an urban site of Shanghai over a fog-haze event from 6 to 9 November 2012 was analyzed for CCN and aerosol. Overall, meteorological conditions such as wind speed, wind direction and temperature exerted a great influence on CN and BC loadings. Human activity is an essential factor to control emissions of aerosol and CCN in urban environments. N_{CCN}/N_{CN} and N_{CCN} usually were higher in the hazy days due to increased aerosols in the accumulation mode, and lower in the foggy-hazy and clear days. Of special interest, the low N_{CCN}/N_{CN}, N_{CN} and N_{CCN} during the foggy-hazy days can plausibly explain in three aspects: (1) the limited data input introduces some uncertainties, (2) the possible physical effects such as boundary layer evolution, transportation and atmospheric dilution are not considered, (3) the plausible emergence of fog droplets and particles leads to the reduction of aerosol number concentration.

BC was correlated well with N_{CCN} in the foggy-hazy and hazy days but the clear days, as not N_{CCN}/N_{CN} did. More BC is aged during the foggy-hazy/hazy days, hence more

CCN is activated (Dusek et al., 2006; Anttila and Kerminen, 2007; Hudson., 2007). However, there exists a different perspective. For example, BC has been found to significantly suppress cloud formation in the Indo-Gangetic plain (Ritesh et al., 2007). Pure BC particles are hydrophobic and can release heat by absorbing solar radiation, hence they would increase the critical SS of aerosol to act as CCN and further suppress the tendency of CCN to become cloud droplets. However, aged BC particles are sufficiently hydrophilic by acquiring hydrophilic coatings in the atmosphere, and become CCN and favor aerosol indirect forcing (Conant et al., 2002; Ritesh et al., 2007). In this study, BC particles moved a long-distance from inland and aged during the transporting process, thereby it favors CCN formation.

By using a simplified κ parameter, the critical dry size never exceeded 130 nm. In spite of the absence of organic matter, the CCN closure calculation was still achieved, suggesting that aerosol major water soluble ions contribute to effective κ. The predicted N_{CCN} was close to the observed during the clear days than the foggy-hazy/hazy days having more organic matter. In summary, water soluble inorganic ions constituted the majority of particle hygroscopicity (κ) estimation, while organic matter made up the rest. It is noted that organic matter is essential to build the exact CCN prediction models.

This paper mainly explored how N_{CCN}, N_{CN} and N_{CCN}/N_{CN} vary under a fog-haze co-occurring condition, as well as the major influential factors to these activities. The results revealed that the particulate pollutant burden exerts a significant impact on N_{CCN}, especially N_{CCN}/N_{CN} is effectively promoted during the polluted periods (e.g. haze). Importantly, the fog-haze transformation is highly complicated involving numerous changes of aerosol in physical and chemical properties, which remains poorly understood. There presents the results of only a case, so more efforts are needed for highlighting the comprehensive effects of fog and haze on CCN in urban environments.

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Table 1. Statistics of meteorological parameters in different weather conditions.

	Clear day	Foggy-hazy day	Hazy day	All
Temperature (°C)	14.39	14.59	16.53	15.02
Wind direction (deg)	157.22	191.36	260.59	191.27
Wind speed ($m\,s^{-1}$)	1.95	1.28	2.27	1.94
Pressure (hPa)	1021.92	1019.19	1019.45	1020.83
RH (%)	58.13	84.93	58.33	61.95
Visibility (km)	15.41	2.33	4.44	10.42
PBL (km)	1.2	0.58	0.62	0.93
Extinction coefficient	0.32	0.27	0.76	0.62

Table 2. Statistics of CCN, CN, CCN/CN and BC in different weather conditions.

	Clear day	Foggy-hazy day	Hazy day	All
CCN range (cm^{-3})	994–5096	1677–2947	2088–6268	994–6268
CCN average (cm^{-3})	2432	2377	4362	2929
CN range (cm^{-3})	4270–15 168	4815–13 922	6033–15 771	4270–15 771
CN average (cm^{-3})	8956	8367	10 500	9344
CCN/CN range	0.09–0.48	0.18–0.40	0.25–0.57	0.09–0.57
CCN/CN average	0.28	0.29	0.41	0.32
BC range ($\mu g\,m^{-3}$)	4.51–20.40	6.7–14.7	8.3–35.2	4.51–35.20
BC average ($\mu g\,m^{-3}$)	8.57	9.58	21.26	12.24

Table 3. Effective hygroscopicity parameters (κ_i), and densities of the three category compositions in fine particles.

Species	Data source	κ_i	Density (g cm^{-3})
Sulfate and Nitrate	$SO_4^{2-} + NO_3^- + NH_4^+$	0.6	1.7
Sodium chloride and marine aerosols	$Na^+ + Cl^-$	1.0	2.2
Insoluble compounds	BC	0	1.0
	others	0	2.0
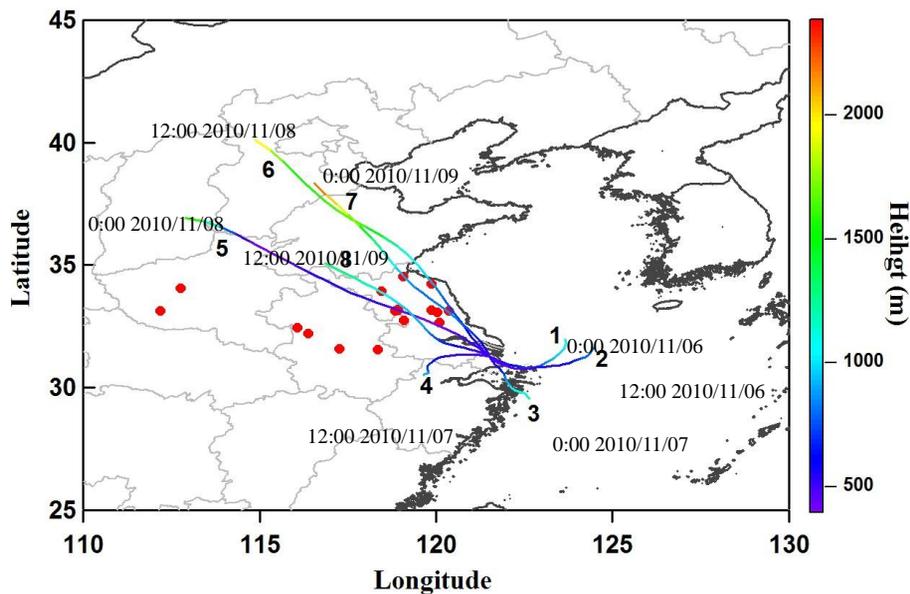
Figure 1. Agricultural fire scattering areas and air mass transport pathways across these regions. All red spots represent biomass burning sites on 7 November measured from MODIS satellite. Starting time (LT) is labeled in the figure.
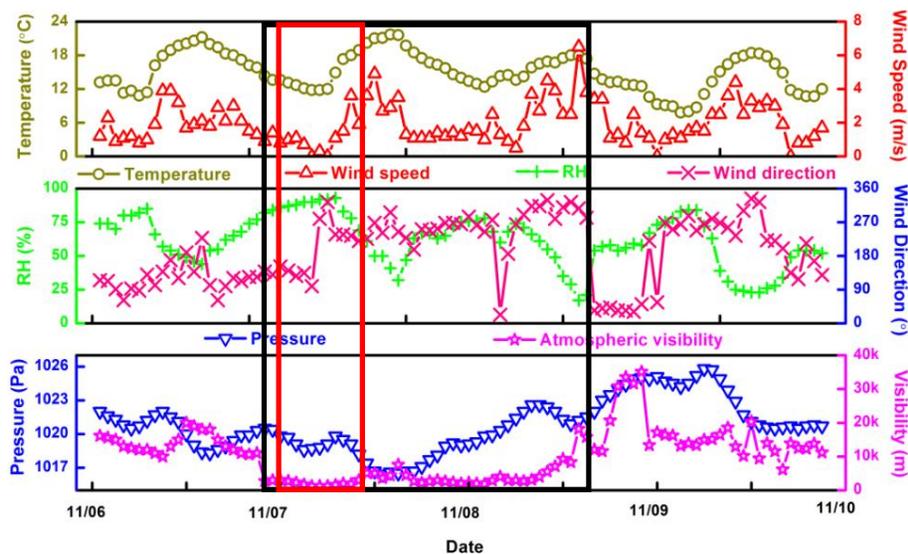
Figure 2. Temporal variations of temperature, wind speed and direction, RH, pressure and atmospheric visibility.
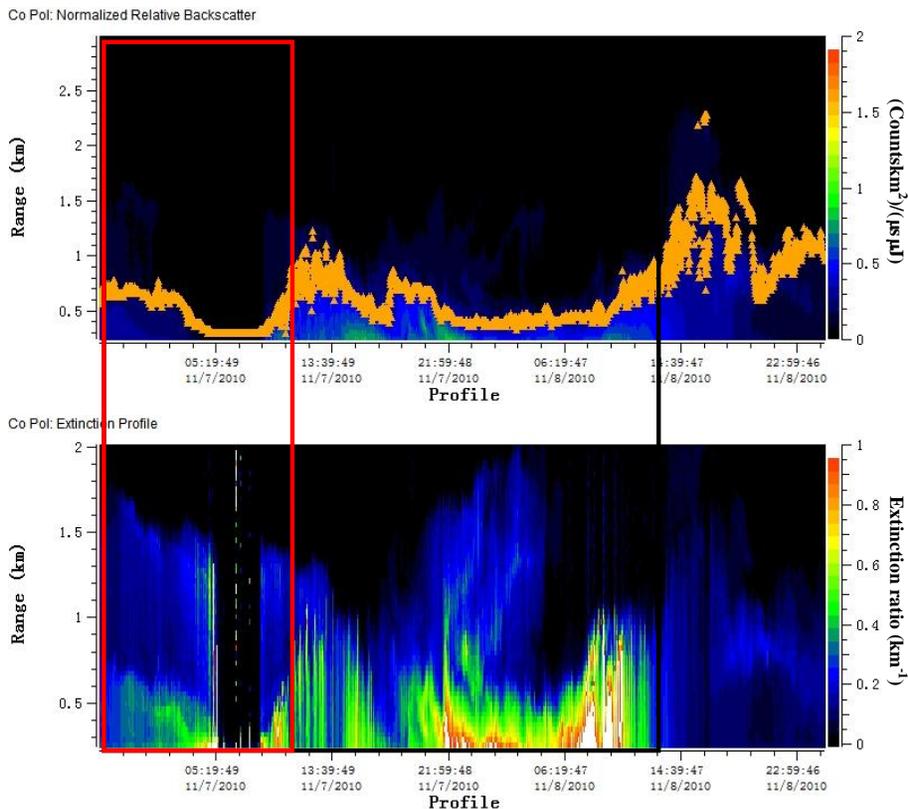
Figure 3. Temporal variations of PBL and vertical extinction coefficient measured by MPL lidar.
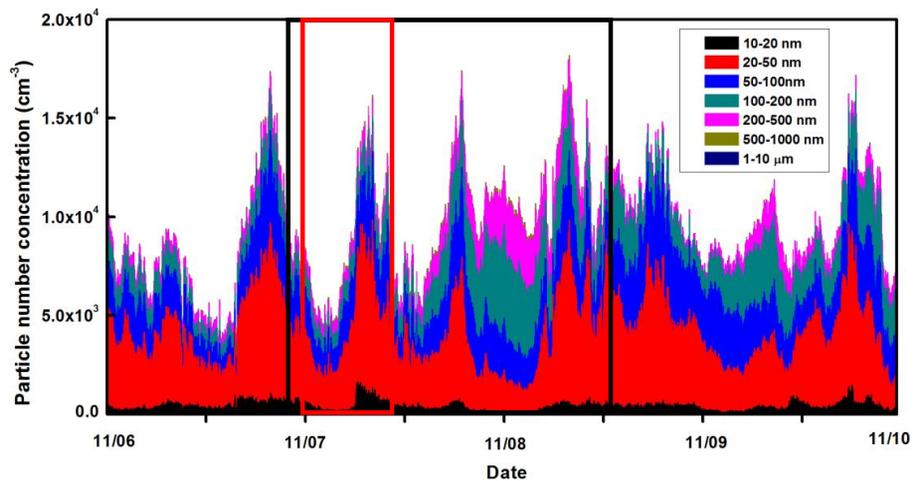
Figure 4. Hourly mean particle number concentrations of different sub-size bins.
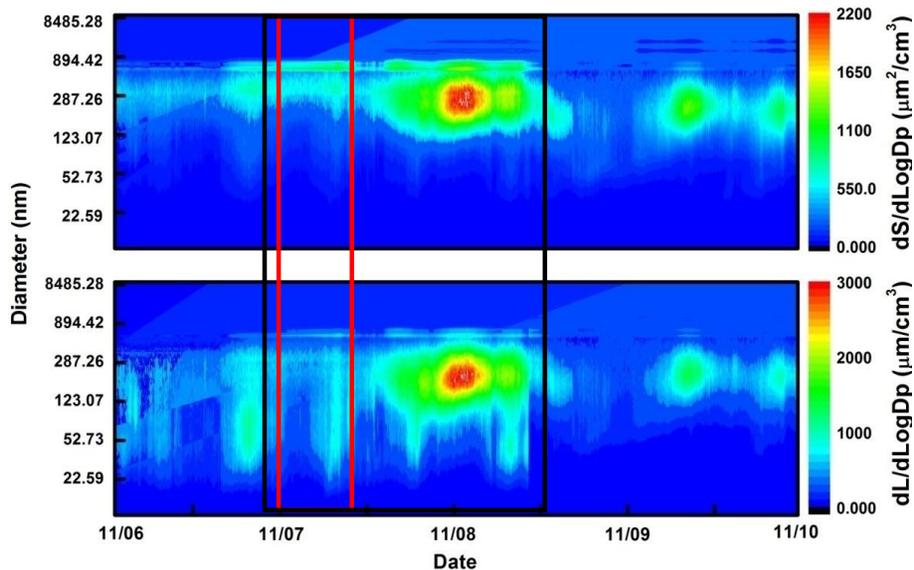
Figure 5. Contour plots of surface area size distribution.
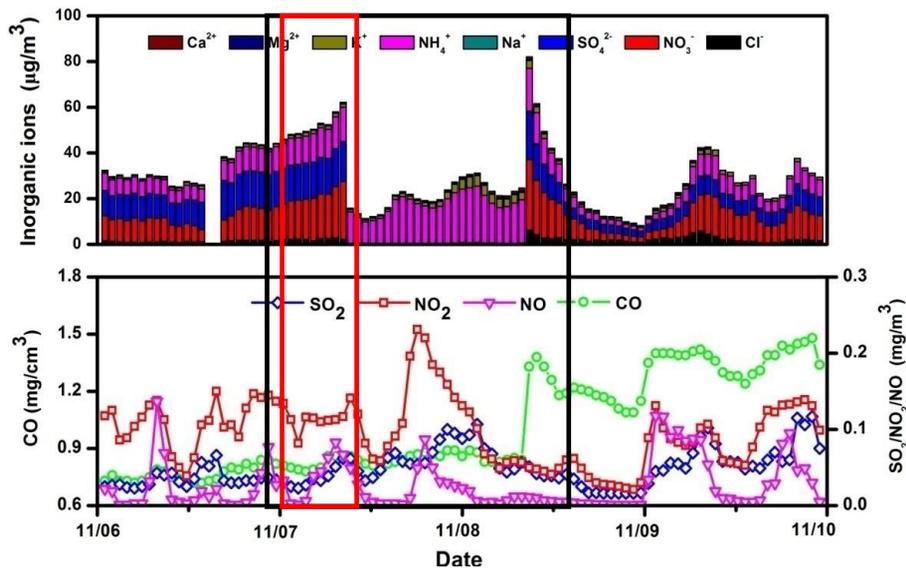
Figure 6. Temporal variations of particle water soluble ion composition and trace gases.
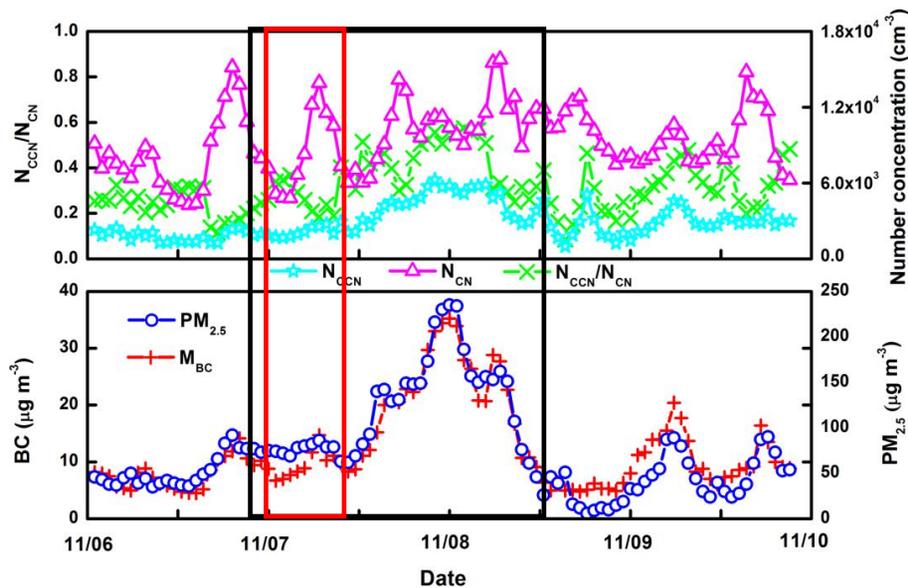
Figure 7. Temporal variations of N_{CN}, N_{CCN} at 0.2 % SS, BC, $PM_{2.5}$ and N_{CCN}/N_{CN}.
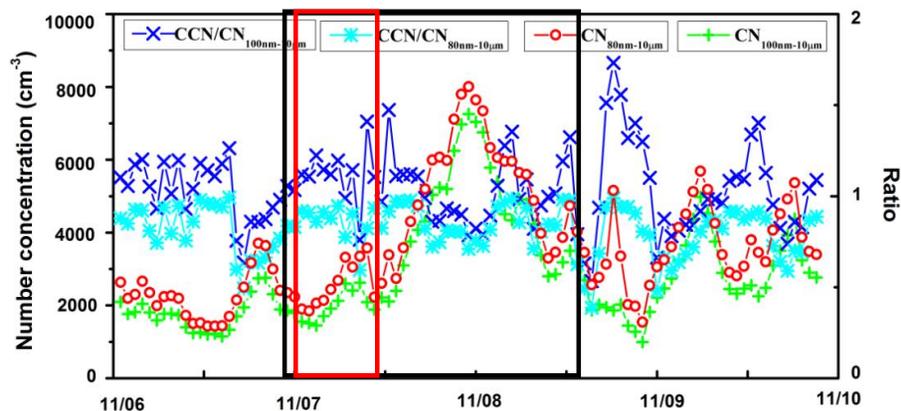
Figure 8. Temporal variations of $CN_{100\,nm-10\,\mu m}$, $CN_{80\,nm-10\,\mu m}$, $CCN/CN_{100\,nm-10\,\mu m}$ at 0.2 % SS and $CCN/CN_{80\,nm-10\,\mu m}$ at 0.2 % SS.

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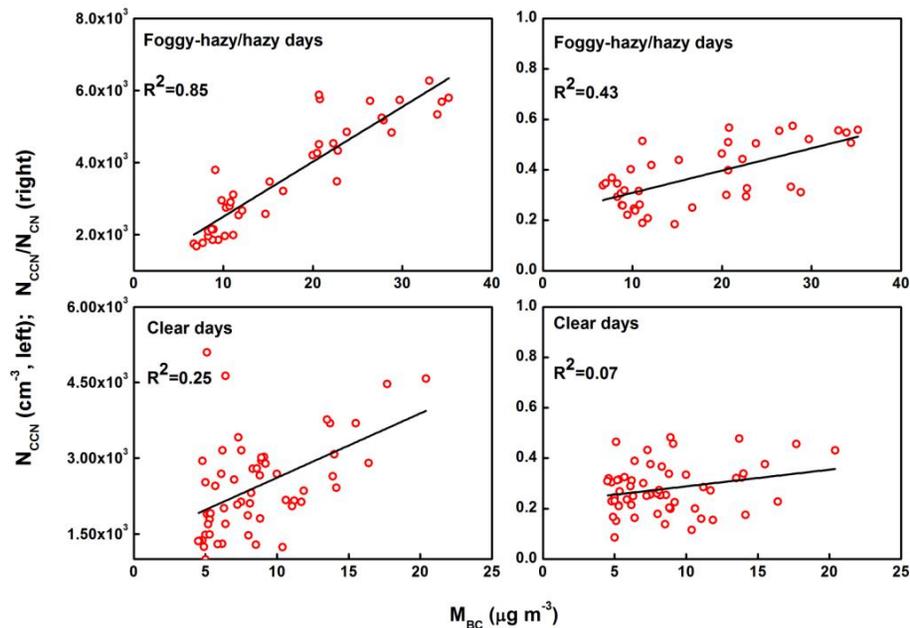
Figure 9. Correlations of BC mass concentration (M_{BC}) to N_{CCN} and N_{CCN}/N_{CN} (0.2 % SS).
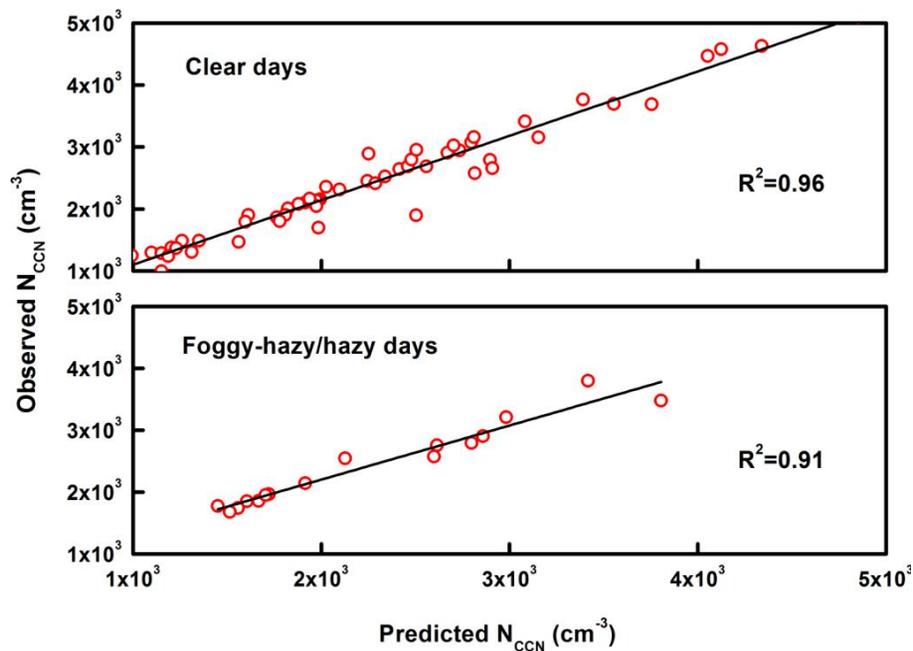
Figure 10. Correlations of observed and predicted N_{CCN} (0.2 % SS) in the clear foggy-hazy/hazy days.

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