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A comparative study of cloud condensation nuclei measured between non-heating and heating periods at a suburb site of Qingdao in the North China

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HIGHLIGHTS

- NPF and regional pollution events greatly influence CN and sequent CCN.
- More organics in particles during a heating period lowered CCN activity.
- CCN activity of grown new particles sometimes was constant.

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ABSTRACT

In this paper, we studied concentrations of cloud condensation nuclei (CCN) and CCN activity measured at a suburb site of Qingdao in the North China during a non-heating period from 19 May to 3 June 2013 and a heating period from 6 November to 6 December 2013. Concentrations of CCN during the non-heating period were 3.1 ± 1.9 (mean ± standard deviation), 4.9 ± 3.4, 5.6 ± 3.9, 6.1 ± 4.3 and 6.5 ± 4.6 in unit of $\times 10^3$ cm^{-3} at supersaturation (SS) of 0.2%, 0.4%, 0.6%, 0.8% and 1.0%, respectively. The corresponding CCN activities of atmospheric particles were 0.28 ± 0.17, 0.43 ± 0.24, 0.48 ± 0.26, 0.52 ± 0.27 and 0.54 ± 0.28, respectively. Concentrations of CCN during the heating period were 3.1 ± 1.3, 6.4 ± 2.3, 8.5 ± 2.9, 9.6 ± 3.5 and 10 ± 3.9 in unit of $\times 10^3$ cm^{-3} at SS of 0.2%, 0.4%, 0.6%, 0.8% and 1.0%, respectively. The corresponding CCN activities were 0.11 ± 0.06, 0.24 ± 0.11, 0.31 ± 0.14, 0.35 ± 0.15 and 0.37 ± 0.15, respectively. At SS ≥ 0.4%, concentrations of CCN were significantly larger during the heating period than during the non-heating period with 95% confidence because of larger particle number concentrations associated. However, CCN activities were significantly lower during the heating period than during the non-heating period at each SS. The lower CCN activities during the heating period were analyzed in terms of origins, median mobility diameter and possible chemical composition of atmospheric particles. However, when new particle formation events during the heating and the non-heating periods were considered alone, CCN activities of grown new particles at the same size range sometimes appeared to be constant regardless of the heating or non-heating periods.

1. Introduction

Cloud Condensation Nuclei (CCN) activated from atmospheric aerosol particles (condensation nuclei, CN) play a key role in the formation of clouds and precipitation, and thus influence the hydrological cycle and the climate in various scales. It has been reported that the droplets in a cloud can get increased in concentration and decreased in size with the CCN concentration rising. This could lead to less precipitation in shallow and short-lived clouds and more precipitation in deep convective clouds (Rosenfeld et al., 2008). Not all aerosol particles can activate as CCN. CCN activity of aerosol particles reflects the ability of particles that enable the condensation of water vapor and formation of cloud droplets at a given level of water vapor supersaturation (SS). To understand the aerosol–cloud–precipitation relationship, it is important to determine CCN activity of aerosol particles under

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different atmospheric conditions or from various sources (IPCC, 2007; Andreae and Rosenfeld, 2008). Previous studies show that three factors, i.e., aerosol size distribution in number concentration, chemical composition and mixing state, determine CCN activity of aerosol particles and variability of CCN concentrations (Dusek et al., 2006; Hudson, 2007; Quinn et al., 2008; Ervens et al., 2010; Rose et al., 2010, 2011; Wang et al., 2010; Gunthe et al., 2011; Meng et al., 2014). These factors are related to not only aerosol origins, but also aerosol aging processing in the atmosphere. Aerosol origins vary a lot in different atmospheric environments, so does aerosol aging processing (Chan and Yao, 2008). More studies are essential in those areas where concentrations of CCN and CCN activities of aerosol particles are unknown.

Deng et al. (2014) recently studied regional precipitation variability from the North Pacific Ocean to inland of China during 1979–2012 and they found that temporal precipitation variability in most of oceanic and coastal areas can be explained well by thermodynamic and dynamic components associated with the global warming, except the Yellow Sea and the coastal areas within 250 km from the Yellow Sea. The exceptional areas suffer severe air pollution (Fang et al., 2009; Gao et al., 2011; Feng et al., 2012; Zhu et al., 2014) and are situated downwind of air pollutant outflow from city clusters in the North China. Deng et al. (2014) also found a significant correlation between cloud characteristic parameters and aerosol optical depth over the areas. However, the observations on concentrations of CCN and CCN activities of aerosol particles in the areas were non-existed and the knowledge gap limits the understanding of the aerosol–cloud–precipitation relationship.

In this work, we measured concentrations of CCN (N_{ccn}) in May–June and November–December 2013 at a suburb site, ~7 km away from the coastline of the Yellow Sea, downwind of air pollutant outflow in the North China. We studied variability of N_{ccn} in terms of the contribution from primary aerosol particles such as fresh traffic-emitted particles and fresh biomass burning aerosol particles and from aged particles, e.g., in aged SO_2-poor and SO_2-rich industrial plumes, in moderate and severe regional pollution events and in new particle formation (NPF) events. In addition, we investigated CCN activities of aerosol particles from different sources, in moderate and severe pollution events and in NPF events with varying atmospheric SS, particularly the relationship between CCN activity and the growth of new particles.

2. Experimental

Two campaigns were performed during the periods from 19 May to 3 June and from 6 November to 6 December 2013 in the campus of Ocean University of China (OUC) (36°09′37″N, 120°29′44″E). The sampling site is located at a suburb area in the northeast of Qingdao City (Fig. 1) and is downwind of city clusters in the North China (Gao et al., 2011). Several cities at 200–500 km away from the sampling site in the west and northwest directions suffer from severe air pollution and recently rank as ones of the most polluted cities in China. The second sampling period covered the lower-temperature heating period on 6–14 November and the regular heating period since 15 November, but there was no space-heating during the first sampling period. The mean value of $PM_{2.5}$ in mass concentration over the whole Qingdao City was 85 ± 60 μg m^{-3} during the second sampling period, but the concentration was only 45 ± 26 μg m^{-3} during the first sampling period (http://aqicn.org/city/qingdao/). A moderate regional pollution event was observed on 21–22 May when the hourly average of $PM_{2.5}$ in mass concentration mostly exceeded 100 μg m^{-3}. A heavy regional pollution event (heavy haze) occurred on 3–5 December 2013 when the hourly average of $PM_{2.5}$ in mass concentration mostly exceeded 200 μg m^{-3}.

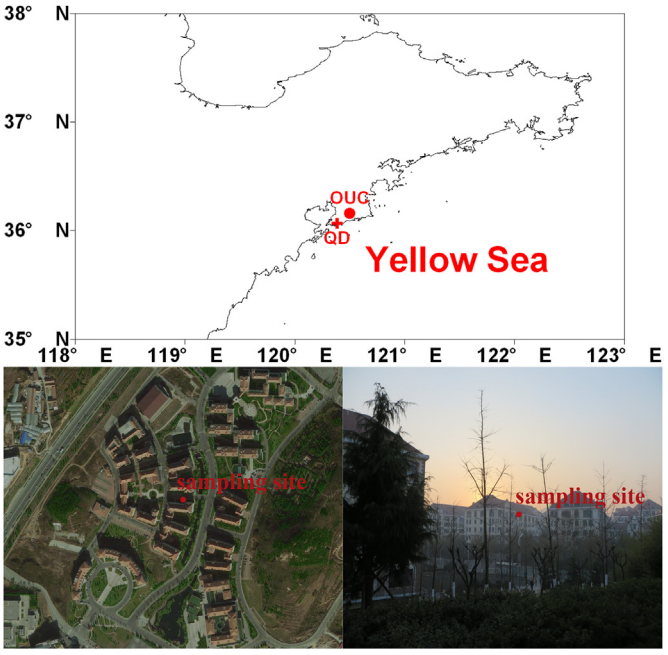

Fig. 1. Maps of the sampling site.

The instruments were placed in an air-conditioned room on the fifth floor of an academic building. The conductive tubes (TSI, US) were used for sampling; the sample flow passed through a diffusion dryer filled with silica gel (TSI, US) and then split into different instruments (Zhu et al., 2014). The length from the tube inlet to each instrument was approximately 1.5 m. In May–June 2013, a Fast Mobility Particle Sizer (FMPS, TSI Model 3091), a Condensation Particle Counter (CPC, TSI Model 3775), a Droplet Measurement Technologies continuous flow CCN counter (CCNC, DMT Model 100) were used to measure the particle number concentration and the bulk CCN concentration. The FMPS measured atmospheric particles over the range of 5.6–560 nm in 32 channels in 1-s time resolution and the data were corrected using the approach proposed by Zimmerman et al. (2015). The CPC measured particle size down to 4 nm and was operated in 2-s time resolution with an inlet flow of 1.5 L min^{-1}. The total number concentration of CN (N_{cn}) used in this study was the particle number concentration measured by the CPC. The DMT-CCNC was operated at a total flow rate of 0.45 L min^{-1} with a sheath to aerosol flow ratio of 10 and we calibrated it using size-selected ammonium sulfate particles for effective SS (Rose et al., 2008). The CCNC was set to step through five different SS of 0.2%, 0.4%, 0.6%, 0.8% and 1.0% based on the temperature gradient (ΔT) varying from 3 K to 16 K of the flow column. The number concentration of CCN (N_{ccn}) could be collected at each supersaturation period which lasted 20 min, but the first 5 min data were discarded because of establishment of supersaturation equilibrium. A SO_2 Analyzer (Thermo Model 43i), a NO_x Analyzer (Thermo Model 42i), an O_3 Analyzer (Thermo Model 49i) were also operated in five-minute time resolution and a meteorological monitoring system was used in one-minute time resolution to obtain the real-time observational data of gases and meteorological parameters. The hourly average of gases in mixing ratio was used for discussion in this study if not specified.

In November–December 2013, a Nanoscan SMPS Nanoparticle Sizer (TSI Model 3910) was used instead of the FMPS and the FMPS was used for simultaneously measuring particle number concentration over the Yellow Sea and Bohai Sea (Liu et al., 2014). Other instruments still operated like in May–June. The SMPS operated at