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Comparison of aerosol and cloud condensation nuclei between wet and dry seasons in Guangzhou, southern China

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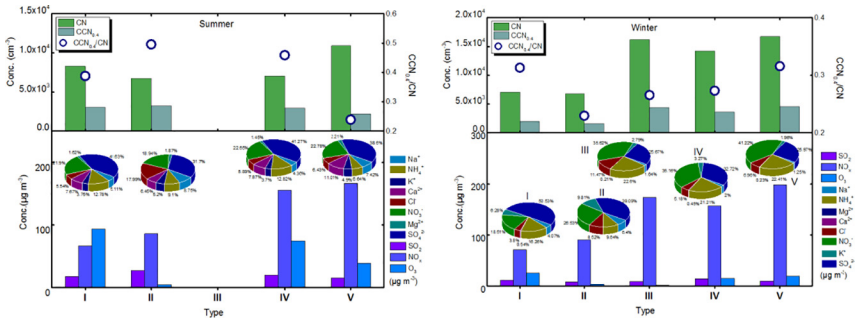
HIGHLIGHTS

- CCN, CN and CCN/CN are of strong seasonality. N_{CCN} and N_{CCN}/N_{CN} are mostly higher in summer than in winter, but N_{CN} is not.
- Air mass type and pollution sources had significant effect on CN loading as well as CCN concentration.
- Anthropogenic emissions and pollutant aging along transportation matter a lot in changing aerosol CCN activity.
- The pollution influences CN and aerosol CCN activation by different ways based on pollution conditions in two seasons.

GRAPHICAL ABSTRACT



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ABSTRACT

Cloud condensation nuclei (CCN), condensation nuclei (CN) and aerosol chemical composition were measured simultaneously at an urban site of Guangzhou from July to August 2015 and in January 2016, and the seasonal variations of aerosol activated fractions (N_{CCN}/N_{CN}) as well as their relevant influence factors were further studied accordingly. N_{CN} is generally higher in winter (dry season), whereas N_{CCN} and N_{CCN}/N_{CN} are mostly higher in summer (wet season) instead. In particular, N_{CCN} and N_{CCN}/N_{CN} are much lower at smaller supersaturation levels ($SS < 0.2$) in winter. In spite of similar diurnal variations for N_{CCN} and N_{CN}, N_{CCN}/N_{CN} indicates an opposite tendency, relatively lower at midday, dusk and before midnight. Other than the size of particles as well as their chemical composition, some other factors, such as mass, gas precursors, pollutant transportation, meteorological conditions, etc., also contribute to the variations of N_{CCN} and N_{CCN}/N_{CN}. Particles from the local source or local-oceanic combination source cast influence on CN and CCN significantly, while the pollutants originating from and crossing over distant polluted areas contribute largely to CCN/CN. N_{CN} and N_{CCN} are relatively higher under pollution-free conditions in summertime and polluted conditions in wintertime, but N_{CCN}/N_{CN} is just the opposite. On various polluted conditions, aerosol CCN activities are greatly discrepant between summer and

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winter, especially during mist or heavy haze periods. The results imply that anthropogenic pollutants exert critical impacts on aerosol CCN activation.

1. Introduction

Atmospheric aerosol is ubiquitous throughout the globe, and greatly affects climate and earth radiation balance through changing light scattering and absorption directly and affecting clouds and precipitation processes indirectly (Twomey, 1974; Lohmann and Feichter, 2005). For climate change prediction, one of the largest uncertainties arises from the impacts of primary and secondary aerosols on clouds and radiative forcing (IPCC, 2013). As a subset of aerosol, CCN not only matters in the formation of clouds and precipitation, but also affects atmospheric chemistry and physics (Pruppacher et al., 1997; Seinfeld and Pandis, 2006; Heintzenberg and Charlson, 2009; Pöschl et al., 2009), and even induces essential changes in meteorological models at all scales (Huang et al., 2007). Owing to the substantial increase of anthropogenic emissions of particles and gaseous precursors, aerosol-cloud interaction has changed cloud microphysical and radiative properties to more extent, which emphasizes the crucial function of CCN on predicting regional and global climate changes (Richter et al., 2005; Shao et al., 2006; Zhao et al., 2006; Rosenfeld et al., 2007; Li et al., 2007; Deng et al., 2008). On account of high spatiotemporal variability and complex transformation in the atmosphere, it is of great importance to acquire more information about CCN and aerosol at various regions.

Currently, the field measurements in different environments have pictured a map of global CCN distribution (Delene and Deshler, 2001; Baumgardner et al., 2003; Yum et al., 2005; Detwiler et al., 2010; Jurányi et al., 2010; Leng et al., 2016), and also explored the influence of the size, chemical composition, mixing state and even partial pressure of water vapor on aerosol CCN activation (Pruppacher et al., 1997; Baumgardner et al., 2003; Yum et al., 2005; Rose et al., 2008; Kuang et al., 2009; Gunthe et al., 2009; Sihto et al., 2011). Several investigations have shown that the particle size is more important than chemical composition to determine aerosol activation, despite a considerable change of CCN at low SS caused by chemical composition variation (Dusek et al., 2006; Hudson, 2007; Kuwata et al., 2008; Kuwata and Kondo, 2008; Kammermann et al., 2010; Rose et al., 2008). However, Cubison et al. (2008) and Mochida et al. (2006) argued that detailed chemical composition and mixing state should be paid more attention to in terms of aerosol CCN activity. The response of aerosol activation towards the mixing state of the particles indicates the potential contribution of major anthropogenic pollution sources to CCN group (Che et al., 2016; Wang et al., 2010). In fact, aerosols are mainly produced by primary emissions and secondary formation, influenced by local and regional pollutant sources and weather conditions (Zhang et al., 2010; Du et al., 2011; Cheng et al., 2008; Fu et al., 2008). To better understand CCN, more efforts are needed to focus on the relationship between precursors, CN and cloud droplet (CD) all over the world.

For pollutions, existent emissions and unfavorable atmospheric convection often cause huge particle loading and visibility (Vis.) impairment at the surface and thus threat human health seriously. In recent years, the complex air pollutions have emerged in China, like haze in winter and high concentration of ozone in summer, which have already received widespread attention from both scientists and policy-makers (Xu et al., 2011; Gao et al., 2007). The accumulating particles and unfavorable meteorological conditions (i.e. planetary boundary layer (PBL)) always deteriorate situation, and as a result, causing a serious and long-lasting polluted incident regionally (Leng et al., 2016). Numerous measurements have achieved meaningful CCN data at polluted places in China such as Beijing, Tianjin, Wuqing, Yufa, Shanghai, Hong Kong, Shouxian and suburban of Guangzhou (Yue et al., 2011; Zhang et al., 2012; Deng et al., 2011; Wiedensohler et al., 2009; Meng et al., 2014; Liu et al., 2011; Leng et al., 2014, 2016; Rose et al., 2010). Nonetheless, to date, there are still few studies of CCN measurement performed at one urban site of southern China.

Guangzhou, located in the Pearl River Delta (PRD) of southeastern China, is one of the mega cities undergoing rapid economic growth. Dominated by the Asian monsoon system, Guangzhou is mainly affluent with clean air masses from southwestern sea areas in wet season (summer) and polluted air masses from northern inland areas in dry season (winter) (Zhang et al., 2013). Extreme and continuing pollution accidents have been reduced due to improved air quality in recent years, however, haze event still happens yet (http://www.gdep.gov.cn/). Recognizing the important role of CCN on changing climate and precipitation, it is imperative to explore the relationship between CCN and pollution in this urban area, since few attentions had been paid on before.

This paper exhibits continuous online measurements of CCN and aerosol chemical composition during wet and dry seasons at an urban site of Guangzhou in 2015. It is aimed to characterize general aerosol CCN activity and relevant influence factors in the typical urban place located in southern China, and compare their seasonality to give insights into the discrepancy of aerosol CCN activation under different polluted conditions.

2. Instrumentation and observation

The instruments of CCN and aerosol measurements were fixed on the roof of a 50-meter-high building at the monitoring station of South China Institute of Environmental Science (SCIES), Ministry of Environmental Protection in Guangzhou, China (23.07°N, 113.21°E). The site is located in one densely-populated urban district without obvious industrial emission sources surrounded (Tao et al., 2014). The prevailing wind directions are southeasterly in summer and northeasterly in winter.

CCN number concentration was measured by using a continuous 500 cm^3/min-flow stream wise thermal gradient CCN counter (CCN-100, Droplet Measurement Technologies, USA) at five discrete SS levels. The principle and operation of this type of CCN counter is described in details elsewhere (Roberts and Nenes, 2005; Lance et al., 2006). The CCN counter was calibrated for airflow, pressure, temperature gradient and optical particle counter (OPC) after using standard ammonium sulfate to ensure stable SS (Leng et al., 2014) and reliable data before the measurement and periodically during the subsequent monitoring. A dryer (Nafion tube) was also applied to reduce the inhaled air relative humidity (RH) to below 20% before entering the counter.

A combination of Scanning Mobility Particle Sizer (SMPS, TSI 3080) and Aerodynamics Particle Sizer (APS) was employed to measure particle number concentrations within the range of 13 nm–20 μm. The AIM software of TSI company is employed to track the variation of size distributions together with applying multiple charge and diffusion correction. The SMPS, consisting of differential mobility analysis (DMA) and condensation particle counter (CPC), can count size-resolved particles of 13–800 nm with a high accuracy, while APS measures particles in 350 nm–20 μm. In order to extend the number size distribution to Stokes equivalent diameters by combining SMPS and APS measured range, the following equation was utilized by premising